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PRELIMINARY EVALUATION OF ALTERNATIVE WASTE
FORM SOLIDIFICATION PROCESSES
VOLUME I: IDENTIFICATION OF THE PROCESSES

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SUMMARY

This document contains preconceptual design data on 11 processes for the solidification and isolation of nuclear high-level liquid wastes (HLLW). The processes are:

- in-can glass melting (ICGM) process
- joule-heated glass melting (JHGM) process
- glass-ceramic (GC) process
- marbles-in-lead (MIL) matrix process
- supercalcine pellets-in-metal (SCPIM) matrix process
- pyrolytic-carbon coated pellets-in-metal (PCCPIM) matrix process
- supercalcine hot-isostatic-pressing (SCHIP) process
- SYNROC hot-isostatic-pressing (SYNROC HIP) process
- titanate process
- concrete process
- cermet process.

For the purposes of this study, it was assumed that each of the solidification processes is capable of handling similar amounts of HLLW generated in a production-sized fuel reprocessing plant. It was also assumed that each of the processes would be enclosed in a shielded canyon or cells within a waste facility located at the fuel reprocessing plant. Finally, it was assumed that all of the processes would be subject to the same set of regulations, codes and standards.

Each of the solidification processes converts waste into forms that may be acceptable for geological disposal. Each process begins with the receipt of HLLW from the fuel reprocessing plant. In this study, it was assumed that the original composition of the HLLW would be the same for each process. The process ends when the different waste forms are enclosed in canisters or containers that are acceptable for interim storage.

Overviews of each of the 11 processes and the bases used for their identification are presented in the first part of this report. Each process, including its equipment and its requirements, is covered in more detail in Appendices A through K. Pertinent information on the current state of the art and the research and development required for the implementation of each process are also noted in the appendices.

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1.0 INTRODUCTION

Nuclear HLLW is isolated when spent fuel from a nuclear reactor is processed. In this process, the spent fuel is disassembled, the fuel rods are opened and the fuel is dissolved in acid. Then various elements are extracted from the acid solution, leaving behind a liquid that is rich in nuclear fission products. This liquid contains the majority of the radioactivity initially contained in the spent nuclear fuel; thus, this liquid is the focal point of efforts to prove that nuclear waste can be safely stored and isolated from humans and their environment.

It is commonly believed that solid forms of nuclear waste, particularly those that exhibit low leaching rates in ground waters, are best suited for long-term disposal. The High-Level Waste Immobilization Program conducted by Pacific Northwest Laboratory (PNL) for the Department of Energy (DOE) has as its objective the development of processes for converting HLLW into solid forms that have been demonstrated to be physically, chemically and radioactively stable. A major portion of this program has involved in-depth studies on low-melting (1000° to 1150°C) borosilicate glasses. Today, the technology for producing monolithic, borosilicate-glass waste forms has been developed to the level where it could be used to process nuclear waste.

To insure that other options for encapsulating nuclear fission products from spent fuel were considered, a significant part of the High-Level Waste Immobilization Program was devoted to investigating waste forms other than monolithic glass. Other waste forms could have advantages over glass because they might have more favorable physical characteristics, because they could provide additional layers of protection and/or because they might have attributes that are more compatible with a wide variety of wastes or geological disposal sites. For example, a crystalline waste form may be more desirable because of its more favorable physical properties. Synthetic mineral waste forms are another example. Some synthetic mineral compounds are thermally stable and might be more resistant to leaching than glass. Examples of this type of waste form are supercalcine (McCarthy 1977) and SYNROC (Ringwood 1978).

Developing waste forms that will provide multiple layers of protection has been one of the primary emphases at PNL. Such waste forms would be compatible with the multibarrier concept illustrated in Figure 1. The inner core would consist of an inert solid, such as supercalcine or a glass marble. The inner core could then be coated with leach- and oxidation-resistant materials, cast in a metal matrix and placed in a metal canister. The resulting composite waste form would exhibit improved thermal stability and mechanical strength, and the added layers would greatly improve leach resistance. However, each additional process step would add to the cost of making the product, increase the complexity of the process, and tend to decrease plant reliability.

The basic goal in the United States is to develop waste forms that will strictly limit the amount of radioactivity that reaches the biosphere. The properties of the waste forms will significantly contribute to the success of such a goal. Due to some of the inherent characteristics of the monolithic, glass waste form, such as internal stresses and heterogeneity, there

FIGURE 1. The Multibarrier Concept for Isolating High-Level Waste

are concerns as to whether such a large glass form is a suitable waste form. Therefore, other waste forms that may have more desirable characteristics are being considered.

In pursuing the nuclear waste management goal, the United States should produce an acceptable waste form product for each of several specific wastes by a feasible, simple and cost effective process. This optimization of these waste forms and their processes is very important to the continued use of nuclear energy in the world. Thus, assessments of alternative waste forms and solidification processes need to be continued.

Pacific Northwest Laboratory's Alternative Waste Forms Program, which was established in fiscal year 1979 to further assess waste form products and processes, places specific emphasis on the development of glass-ceramic, metal-matrix and coated-particle waste forms. This document identifies the process design ramifications of 11 waste form processes. The document will be used as a basis for subsequent evaluations and comparisons of the feasibility of establishing viable production processes in the remote mode. The results of this effort will be published as Volume II of this document. A follow-up document will also be published on the characterization of selected alternative waste forms. It will identify bases for selecting methods of immobilizing nuclear wastes that have promise and should be developed further.

2.0 OVERVIEW OF ALTERNATIVE WASTE FORM SOLIDIFICATION PROCESSES

In this section, each of the 11 solidification processes presented in this study are summarized briefly. The following information is covered:

- how the process works
- major processing concerns
- state of process development
- expected waste form characteristics.

The first three items above are summaries of the information presented in Appendices A through K in this document. Information on the fourth item came from a study of the properties of alternative waste forms (Rusin, Gray and Wald 1979). In general, the properties of alternative waste forms are compared to those of glass monoliths.

2.1 IN-CAN GLASS MELTING PROCESS

In the ICGM process (see Appendix A), HLLW is dried to a fine powder in a spray calciner. The powder, or calcine, is mixed with crushed glass frit, and the mixture falls continuously into a heated canister where it melts to form a glass monolith. Temperatures as high as 1050°C are used in this process. Pressurized systems are not used. Major processing concerns are:

- assuring the proper ratio of calcine to glass frit and the proper mixing of the two during the melting operation
- preventing the calcine/frit mixture from accumulating in the connecting section between the spray calciner and the canister, while maintaining melting temperatures in the canister
- minimizing dust spills when switching canisters.

This process is in an advanced state of development at PNL. It has been demonstrated with radioactive waste on an engineering scale. Full-scale, nonradioactive equipment has been operated extensively. The ICGM process was selected for potential use at the fuel reprocessing plants at Barnwell, South Carolina and West Valley, New York and is considered the reference process for this study. At the time (1976) the ICGM process was selected for these reprocessing plants, it was considered to be the state-of-the-art technology. The ICGM process is also the back-up process for a proposed immobilization plant at Savannah River.

Some of the characteristics of the monolithic, glass waste form are given below:

- Its defined oxide waste-loading is 30 wt%.
- Its density is approximately 3.0 kg/L.
- Its resistance to leaching is excellent due to the high chemical inertness of the glass and its low surface-to-volume ratio.
- Its thermal stability is somewhat uncertain above approximately 500°C. It has been suggested that glass may not be thermodynamically stable during its toxic lifetime; it may

devitrify and the resulting changes may adversely affect the durability of the glass. (The leach rate usually increases by a factor of about 2 and never more than by a factor of 10.) In addition, the temperature of the glass must be maintained well below the temperature at which it was formed ($\sim 1050^{\circ}\text{C}$) to prevent high-density crystals from settling and concentrating.

- Its radiation stability is excellent. Only small changes in density will occur, and radioactively induced cracking will be minimal.
- Its resistance to damage on impact is excellent. Impact tests have shown little damage to full-sized waste forms.
- Its amenability to assuring product quality is poor. However, quality may be inferred through analyses of processing data, such as the weights, volumes, temperatures and flow rates of the materials being processed.
- It is difficult to recycle. Although the glass that does not meet required processing standards can be melted, drained from its canister and recycled, significant quantities of glass will be left behind in the canister. This residual glass may require the use of strong chemicals to remove it from the canister. However, dissolving the glass in chemicals may result in dissolving much, if not all, of the canister to which the glass has adhered, thereby increasing the complexity of recycling.

2.2 JOULE-HEATED GLASS MELTING PROCESS

In the JHGM process (see Appendix B), HLLW is calcined and mixed with glass frit. The mixture falls into a joule-heated glass melter, where it melts due to the heat generated by the resistance of molten glass to the flow of an electrical current. Molten glass accumulates in the melter and is periodically poured into canisters. Temperatures up to 1200°C are used in this process. Pressurized systems are not used. Major processing concerns are:

- preventing and/or accommodating solids that settle and accumulate at the bottom of the melter
- controlling volatility when operating in the idling mode.

The JHGM process is in an advanced state of development at PNL and is also currently being developed at Savannah River Laboratory (SRL). Full-scale, nonradioactive equipment has been extensively demonstrated, although radioactive operations have not yet been conducted. This process has been selected by SRL as the basic process for solidifying HLLW at the Savannah River Project.

The expected characteristics of the waste form produced by the JHGM process are similar to but more homogeneous than those of the ICGM waste form due to the mixing that takes place in the melter. Its amenability to quality assurance may be superior to ICGM if the glass can be sampled as it is being poured into a canister.

2.3 GLASS-CERAMIC PROCESS

The GC process (see Appendix C) is based on a celsian GC waste form developed at the Hahn-Meitner Institute of Germany. In the GC process, glass is produced by a method identical to that used in the JHGM process, except that the canisters are smaller in diameter and may contain fins for improving heat transfer. In addition, the glass-filled canisters are heated in a controlled-temperature cycle to first nucleate a fine dispersion of crystals in the glass and then to maximize the final crystalline content of the waste form. Temperatures up to 1350°C are used in this process. Pressurized systems are not used. Major processing concerns are:

- preventing and/or accommodating solids that settle and accumulate in the bottom of the melter
- preventing crystalline sludge formation
- controlling volatility during idling periods
- controlling melt composition and canister temperature gradients to yield acceptable GC quality.

The GC process is fairly well developed, although full-scale waste forms have not been produced. While a full-scale system for producing glass has been extensively demonstrated, the higher melting temperatures required by the GC process may require a modified melter design. The sensitivity of the GC waste form to variations in composition and temperature also needs further study.

Some of the expected characteristics of the GC waste form are listed below:

- Its defined oxide waste-loading is 30 wt%.
- Its density is approximately 3.0 kg/L.
- Its resistance to leaching is similar to that of glass. However, if improperly formed, its resistance to leaching may be inferior to glass.
- Its thermal stability is superior to that of glass, since it remains solid at temperatures at which glass is molten.
- Its radiation stability is excellent and similar to that of glass.
- Its amenability to quality assurance is similar to the JHGM waste form. Samples of glass being poured into a canister may be collected, but these samples will require heat treating to simulate the GC waste form.
- It is more difficult to recycle than glass monoliths. The high temperatures required to melt the GC may preclude completely draining the canister since the metal canister could soften or melt. Once removed from the canister, the GC may need to be converted to a fine powder or solution to ensure that the crystals are soluble when being recycled in the glass melter.

2.4 MARBLES-IN-LEAD MATRIX PROCESS

In the MIL matrix process (see Appendix D), glass is produced by a method identical to that used in the JHGM process. However, instead of pouring glass into canisters, the glass is poured into a marble-making device. Marbles produced by this device are fed into a canister and are cast in lead. Temperatures in this process are as high as 1250°C. Pressurized systems are not used. Major processing concerns are:

- preventing and/or accommodating solids that settle and accumulate at the bottom of the melter
- designing a reliable marble-making device that can be operated and maintained remotely
- operating and maintaining a number of devices for handling process solids.

The MIL matrix process has been developed on a nonradioactive, engineering scale at PNL. A modification of this process, named the PAMELA process, has been developed in Belgium. Research in metal casting has also been conducted at Argonne National Laboratory and at Idaho National Engineering Laboratory. A process to produce glass pellets from transuranic wastes is under development at Rocky Flats.

Some of the expected characteristics of the MIL waste form are given below:

- Its defined oxide waste-loading in the glass phase is 30 wt%. Its overall waste-loading is 4 wt% and 11 vol%.
- Its glass density is approximately 3.0 kg/L. Its overall density is approximately 7.7 kg/L.
- Its resistance to leaching is somewhat superior to that of glass monoliths. The lead annulus between the canister and the basket containing the glass marbles provides an additional barrier to leaching. The relative surface areas of glass marbles and glass monoliths are similar since glass monoliths can crack after casting.
- Its thermal stability may be somewhat superior to that of glass monoliths. The presence of the lead improves heat transfer, which reduces the temperature of the glass in the middle of the canister. The lead may also act as a heat sink in accidents involving fire, which minimizes the temperature that the glass might attain. The reactivity between molten lead and glass has been found to be low.
- Its radiation stability is similar to that of glass monoliths.
- Its resistance to damage on impact is superior to that of glass monoliths due to the shock-absorbing nature of the lead.
- Its amenability to quality assurance is superior to that of glass monoliths since marbles can easily be sampled. Assuring the quality of lead castings may depend upon analyses of processing data as in the ICGM process.
- Marbles are easier to recycle than glass. However, the presence of lead in the final waste form may seriously complicate recycling because the lead may be difficult to

separate from the marbles. Any lead residue on the marbles may adversely affect the melting operation and the quality of the recycled glass.

2.5 SUPERCALCINE PELLETS-IN-METAL MATRIX PROCESS

"Supercalcine" is a silicate-based waste form developed at Penn State University. In the SCPIM process (see Appendix E), a mixture of HLLW and supercalcine-forming additives are dried to a fine powder in a spray calciner. The powder is then formed into spherical pellets using a disc pelletizer. These pellets are sintered in a furnace to densify the pellets and to form the desired mineral phases. The sintered pellets are then cast in lead as described in the MIL process. Temperatures of up to 1250°C are used in this process. Pressurized systems are not used. Major processing concerns are:

- determining the proper quantity of supercalcine-forming additives
- containing dust and volatile species
- operating and maintaining a number of devices for handling process solids
- recycling solid scraps from the process.

The SCPIM process has undergone development at PNL. At PNL supercalcine powders have been produced and pelletized in full-scale equipment. Pellets have been sintered in laboratory-scale furnaces and have been cast in lead in small-diameter pipe sections. At Idaho National Engineering Laboratory, a modified version of this process has been fairly well developed on a nonradioactive level.

Some of the expected characteristics of the full-sized waste form produced by the SCPIM process are:

- Its defined oxide waste-loading in the supercalcine-pellet phase is 78 wt%. Its overall waste-loading is 8 wt% and 18 vol%.
- Its density is approximately 3.8 kg/L in the supercalcine phase. Its overall density is approximately 8.9 kg/L.
- Its resistance to leaching is superior to that of glass monoliths due to the lead barrier. The long-term leach resistance of supercalcine should be comparable to that of glass.
- Its thermal stability is superior to that of glass. The supercalcine remains solid at temperatures at which glass is molten. At high temperatures, supercalcine retains volatile species better than glass. Low reaction rates between molten lead and supercalcine have been found.
- Its radiation stability is uncertain. The long-term effects of transmutation and metamictization need to be assessed.
- Its resistance to damage on impact is superior to that of glass monoliths due to the shock-absorbing nature of the lead.

- Its amenability to quality assurance is superior to that of glass monoliths since supercalcine pellets can be readily sampled and tested. Quality assurance of the lead castings may depend on the analyses of processing data as in the ICGM process.
- Its amenability to recycling is superior to that of glass monoliths when in the loose pellet form. However, recycling sintered supercalcine is more difficult than recycling glass marbles because supercalcine requires more grinding and blending.

2.6 PYROLYTIC-CARBON COATED PELLETS-IN-METAL MATRIX PROCESS

In the PCCPIM matrix process (see Appendix F), sintered supercalcine pellets are produced as in the SCPIM process. Each pellet is then coated with a thin coat of pyrolytic carbon. The coated pellets are then cast in lead. Temperatures of up to 1250°C are used in this process. Pressurized systems are not used. Major processing concerns are:

- those already stated under the SCPIM process
- the use of explosive gas mixtures in the coating process.

The PCCPIM process has undergone limited development. As previously stated, nonradioactive supercalcine powder has been produced and pelletized in full-scale equipment. Pyrolytic-carbon coatings have been applied at Battelle, Columbus Laboratories using laboratory-scale equipment. A low-temperature, pyrolytic-carbon coating process has also been developed on a laboratory scale in Austria. The technology that has been developed for coating high-temperature, graphite-reactor (HTGR) fuel pellets is also applicable.

Some of the expected characteristics of the full-sized waste form produced in the PCCPIM matrix process are:

- Its defined oxide waste-loading is 78 wt% in the supercalcine-pellet phase. Its overall waste-loading is 7 wt% and 15 vol%.
- Its density is approximately 3.8 kg/L in the supercalcine phase. Its overall density is 8.9 kg/L.
- Its resistance to leaching is superior to that of glass waste forms due to the protective pyrolytic-carbon coating and the lead barrier. Its long-term leach resistance is expected to be superior to that of glass.
- Its thermal stability is superior to that of glass. The supercalcine will remain a solid until approximately 1300°C. Tests have indicated very low, if any, reactivity between supercalcine and molten lead. The potential for reactions between the carbon coating and the supercalcine has not been determined.
- Its radiation stability is uncertain. The effects of transmutation and metamictization need to be determined.
- Its resistance to damage on impact is superior to that of glass monoliths due to the shock-absorbing nature of the lead.

- Its amenability to quality assurance is superior to that of glass monoliths where only pellets are concerned. However, quality assurance of the coating may be very difficult, and the quality assurance of the lead casting may depend on analyses of processing data as in the ICGM process.
- Its amenability to recycling may be superior to that of glass monoliths when in the loose pellet form. However, recycling sintered supercalcine may be more difficult than recycling glass marbles because supercalcine requires more grinding and blending. Pyrolytic-coated pellets may also require oxidation before recycling.

2.7 SUPERCALCINE HOT-ISOSTATIC PRESSING PROCESS

In the SCHIP process (see Appendix G), supercalcine powder is produced as in the SCPIM process. The powder is added to a canister where it is densified by vibration. The canister is heated to denitrate and sinter the supercalcine powder into a semidense monolith. The canister is then filled with crushed glass and is placed in a hot-isostatic press (HIP). The HIP is evacuated of air and is gradually heated to melt the glass. When the glass has melted, the HIP is pressurized and heated further. Under high temperature and pressure, the glass acts as a hydraulic fluid which evenly compacts the supercalcine. This finalizes the formation of the desired mineral phases and compacts the supercalcine to near its theoretical density. The maximum temperature used in this process is 1150°C. Pressures range up to 100 MPa (15,000 psi). Major processing concerns are:

- establishing and optimizing the time/temperature/pressure requirements for hot-isostatic pressing
- controlling cesium and ruthenium volatility during sintering
- identifying a glass with the desired properties for use in the HIP
- designing and maintaining a safety system for the highly pressurized HIP
- operating and maintaining the HIP in a remote cell.

The development of the SCHIP process is at the conceptual stage. Nonradioactive, supercalcine powders have been sintered on a laboratory-scale and have been hot-isostatically pressed inside small-diameter tubes. Hot-isostatic presses of the size required for the SCHIP process are being used in nonradioactive industries. Glass has not been used as a hydraulic fluid to hot-isostatically press supercalcine, although it has been used to press other ceramics.

Some of the expected characteristics of the full-sized, SCHIP waste form are listed below:

- Its defined oxide waste-loading is 78 wt% in the supercalcine phase. Its overall waste-loading is 13 wt%.
- The density of the supercalcine is 4.9 kg/L.
- Its resistance to leaching is approximately equal to that of glass in the short term. It may be superior to that of glass in the long term.

- Its thermal stability is superior to that of glass since it is a stable solid at temperatures at which glass is molten. High-temperature tests have indicated that supercalcine retains cesium better than glass.
- Its radiation stability is uncertain. The possibility of cracking due to swelling caused by isotopic transmutation needs to be determined. The effects of metamictization are unknown.
- Its resistance to damage on impact is better than that of glass monoliths but less than that of the metal matrix forms.
- Its amenability to quality assurance is approximately equivalent to that of glass monoliths produced in the ICGM process. As in the ICGM process, quality may be inferred through analyses of processing data. Samples may be obtained as the canister is being filled, but they must be further treated to be representative of the SCHIP waste form.
- Its amenability to recycling may be somewhat superior to that of glass because it can be reworked at higher pressures and temperatures and for longer periods in the HIP. However, the presence of melted glass will seriously complicate matters because the waste to be recycled must be crushed and blended with additives prior to resintering and repressing.
- Reactions between supercalcine and glass need to be evaluated.

2.8 SYNROC HOT-ISOSTATIC PRESSING PROCESS

"SYNROC" is the name of a titanate-based, ceramic waste form developed in Australia. In the SYNROC HIP process (see Appendix H), HLLW is converted to a dry powder in a spray calciner. The dry powder is blended with crushed SYNROC additives that are sized to maximize packed density. The blended materials are then added to a canister, vibrated, sintered and hot-isostatically pressed as in the SCHIP process. The maximum temperature used in the process is 1350°C. Pressures range up to 100 MPa (15,000 psi). Major processing concerns are the same as for the SCHIP process.

As in the SCHIP process, the SYNROC HIP process is only in the conceptual stage of development. SYNROC powders have been produced and pressed using laboratory-scale equipment. Glass has not been tested as a hydraulic fluid. However, all of the major pieces of equipment required in this process have been used in nonradioactive applications.

Some of the expected characteristics of the full-sized, SYNROC HIP waste form produced by the process selected for this study are listed below:

- Its overall waste loading is 30 wt%.
- The SYNROC density is 4.4 kg/L.
- Its resistance to leaching is expected to be superior to that of glass, especially under hydrothermal conditions. This needs to be verified.

- Its thermal stability is superior to that of glass since it is a stable solid at temperatures at which glass is molten.
- Its radiation stability is uncertain. The possibility of cracking due to swelling caused by isotopic transmutation needs to be assessed. The susceptibility of SYNROC to metamictization is unknown. The effects of transmutation and metamictization on SYNROC should be less than on supercalcline, which has a higher waste loading.
- Its resistance to damage on impact may be superior to that of glass monoliths but less than that of the metal matrix waste forms. However, this conclusion is based on limited tests showing resistance to damage on impact of hot-pressed supercalcline over glass monoliths by a factor of two.
- Its amenability to quality assurance is approximately equivalent to that of glass monoliths produced in the ICGM process. As in the ICGM process, quality may be inferred through analyses of processing data. Samples may be obtained as the canister is being filled, but they must be further treated to be representative of the SYNROC HIP waste form.
- Its amenability to recycling may be somewhat superior to that of glass monoliths since it can be reworked at higher pressures and temperatures and for longer periods in the HIP. The presence of melted glass will seriously complicate matters because the waste to be recycled must be crushed and blended with additives prior to resintering and repressing.
- Reactions between SYNROC and glass need to be evaluated.

2.9 TITANATE PROCESS

This process (see Appendix I) is based on a titanate-ceramic waste form developed at Sandia Laboratories. In this process HLLW, calcium hydroxide and granular titanate additives are mixed together in a large, heated vessel. The water is boiled away and the remaining contents are further heated to remove nitrates and to densify the materials. A charge of the resulting material is added to a die in a uniaxial hot press, where it is pressed at high temperatures to a dense monolith. The die containing the pressed monolith is removed from the press and cooled. Later the titanate monolith is removed from the die and placed in a canister. Temperatures up to 1100°C are used in this process. Pressures up to 6.8 MPa (1000 psi) are used. Major processing concerns are:

- accomplishing high enough evaporation rates in the evaporation/calclination vessels
- preventing buildups in the vessels and transfer lines
- controlling the spread of radioactive dust
- accommodating the various mechanical requirements of pressing, separating the pressed monolith from the die, and handling the process solids.

The titanate process is at the conceptual stage of development. Small samples of waste forms have been produced at Sandia Laboratories, and radioactive tests have been conducted at

Oak Ridge National Laboratory (ORNL). Uniaxial hot presses of the size required for this process are commercially available, although considerable modification will be required to make the equipment remotely operable and maintainable.

Some of the expected characteristics of the full-sized titanate waste form are listed below:

- Its defined oxide waste-loading is 25 wt%.
- Its density is 4 kg/L.
- Its resistance to leaching may be superior to that of glass. Long-term leaching tests under radioactive conditions have not been conducted.
- Its thermal stability is superior to that of glass due to the high temperatures under which it is formed.
- Its radiation stability is uncertain.
- Its resistance to damage on impact may be superior to that of glass. However, this conclusion is based on a limited impact test that showed hot-pressed supercalcline to be better than glass monoliths.
- Its amenability to quality assurance is superior to that of glass monoliths. Since the titanate waste form is not produced in its canister, the entire waste form may readily undergo tests for its resistance to leaching and to damage on impact.
- Its amenability to recycling may be superior to that of glass monoliths because the titanate waste form is separate from its canister. The waste form can also be reinserted into a die and hot-pressed at elevated temperatures and pressures.

2.10 CONCRETE PROCESS

The concrete process (see Appendix J) is based largely on the FUETAP (formed under elevated temperatures and pressure) concrete concept developed at ORNL. In this process, HLLW is dried to a fine powder in a spray calciner. A batch of powder is combined with water and concrete-forming additives in a mixer. The mixed batch of concrete is then cast in a canister and placed in an autoclave. The temperature and pressure are raised in the autoclave to prevent the concrete from boiling and to accelerate the hardening of the concrete. When the concrete has hardened, the canister is removed from the autoclave and placed in air storage for an extended period of time to allow excess water to escape. Then the canister is sealed. Temperatures up to 800°C are used in this process. Pressures up to 4 MPa (600 psi) are used. Major processing concerns are:

- controlling the rate at which the concrete is heated to avoid boiling before hardening and to avoid excessive centerline temperatures after drying
- minimizing and cleaning concrete buildups in the mixer and off-gas lines
- establishing time requirements for the elimination of excess water from the concrete.

The concrete process is in a preliminary stage of development. Considerable research on concrete as a waste form has been conducted at ORNL and at SRL. Full-sized mixers and autoclaves have not been demonstrated in this process, although the equipment is well developed in nonradioactive processes.

Some of the expected characteristics of the full-sized, concrete waste form are listed below:

- Its defined oxide waste-loading is 19 wt%.
- Its density is 1.6 kg/L.
- Its resistance to leaching is considerably lower than that of glass due to its high level of porosity and the low leach resistance of concrete materials.
- Its thermal stability is inferior to that of glass due to the release of water from hydration at temperatures below 500°C.
- Its radiation stability is inferior to that of glass due to radiolysis of water and residual nitrates. However, the concrete mixture catalyzes the recombination of radiolysis products, which may mitigate some concerns about pressurized canisters.
- Its resistance to damage on impact is expected to be somewhat inferior to that of glass.
- Its amenability to quality assurance is similar to that of glass produced in the JHGM process. It is somewhat superior to ICGM waste forms in that representative samples can be collected as the canister is being filled.
- Its amenability to recycling may be somewhat superior to that of glass monoliths due to the fact that concrete is more soluble in nitric acid than glass.

2.11 CERMET PROCESS

The cermet process (see Appendix K) is based on concepts developed at ORNL. Cermet (ceramic metal) is a composite material containing fine ceramic particles dispersed in a leach-resistant, metallic phase. In the cermet process, HLLW and cermet-forming additives are dissolved in molten urea. Then the solution is dried to a fine powder in a spray calciner. The dried powder is mixed with a small amount of water in a pin-mixer agglomerator to yield small, dense pellets. These pellets are fed to an extruder, where they are converted to cylinders that in turn are fed to a reduction/sintering kiln. In the kiln, the combination of heat and a hydrogen atmosphere causes the reduction of certain oxides to the metallic state and the overall densification of the cylinder. The resulting cermet monolith is inserted into a canister, which is subsequently sealed. Temperatures up to 1050°C are used in this process. Pressures of approximately 30 MPa (4,500) psi are required in the extruder. Major processing concerns are:

- treating high volumes of off gas
- operating and maintaining a number of devices for handling solids
- establishing the requirements for the reliable reduction and sintering of cermet cylinders
- controlling processing steps requiring hydrogen gas.

The cermet process is at a conceptual state of development. Small, radioactive, cermet waste forms have been produced at ORNL using laboratory-scale equipment. Nonradioactive testing has also been limited to laboratory-scale equipment. Some of the technology used in the production of commercial cermets may apply to this process.

Some of the expected characteristics of the full-sized, cermet waste form are listed below:

- Its defined oxide waste-loading is 30 wt%.
- Its density is 6.5 kg/L.
- Its resistance to leaching may be comparable to that of glass over the short term; its long-term leach resistance is thought to be inferior.
- Its thermal stability may be superior to that of glass. It is a solid at temperatures at which glass is molten. Its thermal conductivity is superior to that of glass, which will result in more even temperatures throughout the waste form. The long-term resistance of the metal phase to thermodynamic changes is not known.
- Its radiation stability is uncertain. The ductile nature of the metallic phase may permit radiation-induced density changes to occur without cracking the metal matrix. However, the radiolysis of ground waters in the repository may lead to accelerated oxidation of the metallic phase.
- Its resistance to damage on impact is expected to be superior to that of glass monoliths due to the shock-absorbing nature of the metallic phase.
- Its amenability to quality assurance is superior to that of glass monoliths. Because the waste form is produced outside its canister, the waste form may be tested for resistance to leaching, surface defects, etc.
- It may be easier to recycle than glass monoliths if reworking in the reduction/sintering kiln is successful. When reworking is not successful, dissolving the waste form for recycling to the spray calciner may be very difficult.

3.0 OVERALL PROCESS BASES

3.1 PROCESS CAPACITY

The waste facility will solidify all of the HLLW produced in a fuel reprocessing plant operating at a rate of 2000 MTHM/yr. There are 567 L of HLLW/MTHM. The waste facility will operate for 300 d/yr and will process HLLW at a rate of 3800 L/d. The facility will be designed to operate at 1.5 times the nominal operating capacity, or 5700 L/d, as a contingency for unexpected problems.

3.2 HLLW COMPOSITION

The reference HLLW is an aqueous waste produced by the solvent extraction processing (first cycle) of spent nuclear fuels to recover uranium and plutonium. The spent fuel that is being used as the basis for the solidification processes and equipment design has the following characteristics:

- type--pressurized water reactor, uranium enriched
- enrichment = 3.3% ^{235}U
- burnup = 33,000 MWd/MTHM
- power level = 30 MW/MTHM
- neutron flux = $2.92\text{E}13 \text{ n/cm}^2\text{-s}$
- time out of reactor = 5 yr.

The HLLW contains over 99% of the fission products and transplutonium elements contained in the spent fuel; thus, the HLLW contains most of the radioactivity associated with reprocessing spent fuel. In addition, the HLLW contains 0.5% each of the plutonium, uranium and iodine that was present in the spent fuel, and it contains an insignificant fraction of the krypton, xenon and carbon 14. The HLLW also contains inert materials, which can vary significantly in composition (Energy Research and Development Administration 1976). However, by comparison with other proposed HLLW compositions, the reference HLLW is low in inert materials. The one exception is gadolinium, which is added as a neutron absorber for criticality safety during the fuel-dissolving step.

Virtually all of the constituents of the HLLW are in solution in nitric acid with the exception of small quantities of refractory, heavy-metal oxides and metallic particles of rhodium, palladium and zirconium. A small quantity of tributyl phosphate (TBP), entrained during the solvent extraction process, may also be present as an immiscible liquid. Tributyl phosphate reaction products may also exist in solution. The constituents of the HLLW are given in Table 1. The design bases of the HLLW are listed below:

- $\text{HNO}_3 = 2\text{M}$
- specific gravity = 1.17 g/cm^3
- heat loading = 3.46 W/L
- oxide waste-loading = 92.6 g/L
- nominal operating rate = 158 L/h
- design capacity rate = 237 L/h

TABLE 1. Constituents of HLLW

Constituents	Quantity, g/L
H ⁺ (a)	2.0
NO ₃ (a)	199.0
Inert metals	19.38 (total)
Na(a)	0.15
Fe	2.00
Ni	0.13
Cr	0.33
Gd	15.90
p(a)	0.87
Activation products	0.5
Fission product metals(a)	44.7 (total)
I	0.002
Cs	4.3
Rb	0.59
Ru	3.76
Te	0.99
Others	35.06
Actinide metals	10.5 (total)
U	8.43
Pu	0.11
Others	1.96

(a) Possible volatile species.

3.3 INTERIM STORAGE, TRANSPORTATION AND DISPOSAL

Following the solidification of HLLW, the waste (which is contained in storage canisters) will be transferred to a storage facility located onsite. The storage facility may be comprised of water basins, air-cooled vaults or concrete surface-silos. Storage conditions will be controlled to prevent the deterioration of the waste form. However, both deterioration and canister failure are expected, and provisions will be made for installing overpacks on any or all canisters before shipping. Overpacks will meet the performance criteria of primary canisters. Spent fuel and solidified HLLW may be stored in the same facility.

Canisters delivered to the storage facility will meet the acceptance criteria of the storage facility as well as those for shipping and disposal in a federal repository. Acceptance criteria will include the following:

- The waste canister will be a maximum of 0.60 m in OD and 3 m in length.
- The waste canister will be constructed of 304L stainless steel or a similar material.

- The canister wall must be thick enough to withstand the conditions during filling and subsequent stresses during handling.
- Except for the concrete process, the canister will be seal-welded before being placed in the storage facility.
- The sealed canister will exhibit a leak rate of less than 10^{-5} atm/cm²/s. The leak rate will be determined by a helium leak-test.
- Defective welds will be repaired by welding over the defective weld, welding a new lid over the defectively welded lid, or by overpacking.
- The canister will be checked for surface irregularities such as cracking, bulging, warpage and oxidation. Overpacking will be required when significant surface irregularities are found.
- Contamination on the surface of the canister will not exceed the limits for removable contamination that are given in Table 2.
- The canister will generate no more than 3.06 kW when in the storage facility or at the time of filling. Most of the immobilization processes are capable of handling higher heat loadings than this. Considering that the fission product heat decays rapidly in the first few years after the spent fuel is discharged from a reactor, the waste canisters could contain more than 3 kW of heat if they were filled prior to five years.
- The radiation levels of the canister must not exceed 10^5 rem/h when measured one foot from the canister mid-point.

Following a period of storage at the site of the fuel-reprocessing-plant waste facility, the canister will be transported to a federal repository. The federal repository will provide

TABLE 2. Limits for Removable Contamination^(a)

Contamination	Maximum Permissible Level for Any 300-cm ² Surface Area	
	Ci/cm ²	dpm/cm ²
Natural or depleted uranium and natural thorium:		
Beta-Gamma	10 ⁻³	2200
Alpha	10 ⁻⁴	200
All other beta-gamma- emitting radionuclides	10 ⁻⁴	200
All other alpha-emitting radionuclides	10 ⁻⁵	22

(a) 49 CFR 173.397

facilities for the disposal of waste canisters and, perhaps, for the interim storage of the waste canister prior to final disposal. The cumulative interim storage period may be up to 50 yr.

The canisters will be shipped in shipping casks similar to those used for shipping spent fuel. The cask/canister waste system will not release radioactive materials during accidents involving fire, impact or immersion. If a cask/canister waste system cannot be designed to withstand these accidents, the fuel-reprocessing-plant waste facility and the federal repository will be located at the same site, or special transportation methods will be used to prevent these accidents.

It is assumed that the repository will be located in rock salt excavations deep beneath the earth's surface. Canisters disposed of at the federal repository may generate between 0.5 kW and 2.0 kW depending on the time of emplacement. The disposed canister (and its over-pack) will be contained by a protective barrier(s), such as a concrete vessel. Together, the protective barrier and all other insulating media, including air gaps between the repository rock and the canister wall, will have a thermal insulation value equivalent to approximately 0.15 m of crushed rock at 90°C. If the canister contains about 0.5 kW, the effect will be a maximum temperature of approximately 100°C at the wall of a 0.60-m-dia canister. This is a conservative value, but may be appropriate for first repositories or until operating experience is obtained.

3.4 REGULATIONS, CODES AND STANDARDS

The waste facility will be licensed by the Nuclear Regulatory Commission (NRC). Therefore, the design will conform to applicable regulatory guides and criteria issued by NRC for nuclear reactors and fuel reprocessing plants. The facility and equipment will also be designed in accordance with federal, state and local codes, regulations and standards.

3.4.1 Siting

The waste facility will be an integral part of a nuclear fuel reprocessing plant. Therefore, the facility will conform to the criteria of the fuel-reprocessing-plant site; the waste facility should not require additional siting criteria since it doesn't possess additional features not associated with a fuel reprocessing plant. Economic considerations and the efficiency of integrating the waste facility with other plant operations will determine the location of the facility within the plant.

3.4.2 Environmental Protection

To provide a facility with minimal impact on the environment, the waste facility will be built and operated according to all applicable environmental protection standards and guidelines. The facility will be designed to contain any design-based accidental release of radioactive materials within the confines of the buildings and grounds at or below federally

established limits. In addition, the facility will be designed to withstand the effects of significant earthquakes, floods, winds, fires, explosions, missiles, etc. without compromising building containment or safety functions.

3.4.3 Safeguards

The waste facility will be designed, operated and protected in a manner that minimizes opportunity for theft, sabotage and other subversive activities.

3.4.4 Criticality

Due to the low concentration of fissile material present in the HLLW, a criticality is an unlikely event. However, in processes where the concentration of fissile material may be possible, the equipment will be designed and/or controls instituted to preclude the likelihood of a criticality. A nuclear-materials accountability system will be used to monitor and control fissile material within the facility.

3.4.5 Contamination Control

The facility will be designed to confine radioactive contamination within processing vessels such as tanks, pipes and hoods. However, it is inevitable that spills from processing vessels will occur and that contamination will spread to other surfaces in the process cells. Surface contamination from such spills will be controlled to minimize the possibility of contamination spreading to areas that are not designed for higher levels of radioactivity.

The confinement of contamination in processing vessels and cells will be accomplished by several methods:

- Areas of higher contamination potential will be isolated from areas of lower contamination potential.
- Ventilation air will flow from areas of lower contamination potential to areas of higher contamination potential.
- Potentially contaminable areas will be designed to either contain any spilled solutions or solids or to direct the flow of such spills to areas of higher contamination potential.

3.4.6 Radiological Protection

The facility will be designed and operated so that personnel radiation exposure will be controlled to levels "as low as reasonably achievable." This will be accomplished by the use of radiation shielding and by monitoring and limiting personnel radiation exposure.

Effluents released from the facility to the environs will be monitored for radionuclide content. The facility will be capable of ceasing or containing the flow of effluents under all normal conditions and all design-based accident conditions.

3.4.7 Process Safety Features

Process protection systems will be used to assure that acceptable operating design limits are not exceeded and to detect potentially hazardous conditions and malfunctions. Should a potentially hazardous situation be detected, hazard-suppression systems will be automatically

activated where practical. Protection systems will be reliable, easily maintainable and testable in place. The systems will have sufficient redundancy to maintain their function in the event of testing or the failure of a single component. Protection systems will be fail-safe.

3.5 USEFUL LIFE OF WASTE FACILITY

The waste facility will be designed and constructed according to the following design considerations so that it will have a useful operation life of 30 yr.

- Every component will be designed for maintenance by taking into consideration its remote or non-remote operating environment and design-based accident scenarios.
- Every component used will be designed, procured, fabricated and operated in accordance with quality assurance requirements (DOE 1974).
- The durability of the material to be used will be known, and allowances will be made for radiation damage, corrosion and wear.

3.6 DECOMMISSIONING OF FACILITY AND EQUIPMENT

In selecting and designing a waste immobilization process, consideration will be given to decommissioning the facility and equipment. The processing systems will be suitably leak tight over the lifetime of the facility to prevent excessive accumulations of high-level materials outside the processing systems. Accumulations of high-level materials will be removable by remote means to reduce the accumulations to levels permitting the final disassembly and disposal of the equipment in the waste facility. All equipment in the waste facility at the time of decommissioning will be decontaminated and/or converted to waste forms whose properties suit the hazard level of the waste involved.

3.7 BASES FOR WASTE FACILITY OPERATIONS AND EQUIPMENT

The process areas in the waste facility and their associated functions are listed in Table 3. The nonprocess areas within the facility (not listed in Table 3) include access aisles, change rooms, bathrooms, an emergency aid room, a health-physics station, analytical laboratories, and various offices for support groups such as quality assurance, radiation monitoring and process engineering. All other services required in the operation of the waste facility, for example utilities and materials, will be located within or provided by the fuel reprocessing plant.

This study focuses on the design of operations that will take place in the process cell(s), for example waste solidification and off-gas treatment. Therefore, this section deals primarily with the bases for the operations within the process cell(s), the process cell(s) itself, and the equipment used in the process cell operations. Support groups and services are discussed briefly in sections 3.8 and 3.9.

TABLE 3. Waste Facility Process Areas and Associated Functions

<u>Process Area</u>	<u>Function</u>
Process cell(s)	Provides for the receipt, solidification and packaging of HLLW, inspection of canisters, repairing or reprocessing of waste products and canisters, process off-gas treatment, cell-air filtration, and the handling of cell waste.
Cell ventilation tunnel	Routes air from the waste facility to the fuel-reprocessing-plant air protection system.
Hot pipe trench	Provides for the routing of radioactive solutions between the process cell and other process areas.
Control and monitoring areas	Contains means of controlling and monitoring the process.
Maintenance areas and stations	Provide for equipment access and repairs.
Warm service-gallery	Provides access to the hot pipe trench, some instrument converters and transmitters, samplers for waste solutions and service piping.
Cell service-gallery	Contains cold equipment and services and controls for cell operations.
Aqueous make-up area	Contains chemical storage and delivery systems to the process cell(s).

3.7.1 Process Cell Operations

The major process cell operations are solidifying HLLW, packaging the solidified waste and pretreating process off gas. Together, these operations comprise the scope of the preconceptual process designs described in this study and will be referred to collectively as the "solidification process."

The solidification process will yield a waste form product that is resistant to leaching, resistant to damage on impact, and thermally and radioactively stable. Product quality will be assured by sampling and analyzing the product and/or by maintaining process variables at

levels that have been shown through testing to assure high product quality. Rigid controls will be required for those processes that are not amenable to direct product sampling and analysis.

The solidification process will be integrated with other processes in the fuel reprocessing plant. For example, HLLW from the fuel reprocessing plant will be delivered to storage tanks in the waste facility. The HLLW from the fuel reprocessing plant will be sampled and chemically adjusted to ensure that the HLLW meets the criteria of the solidification process. Likewise, gases discharged from the off-gas system must meet the criteria for gases entering the air protection system in the fuel reprocessing plant. Secondary solid and liquid wastes routed to the incineration and cementation facilities must also meet the acceptance criteria for their respective processes. Finally, the waste canisters must meet the criteria for interim storage, shipping and disposal.

The solidification process will be controlled through comprehensive sampling and analysis and through a reliable instrument system. The data received from sample analyses and instrument readouts will be used to ensure that material can be transferred from one phase of the process to the next and from facility to facility, while meeting the required acceptance criteria of each succeeding area. These data will also be used for controlling the physical and chemical changes occurring within a process. Finally, these data will provide the bases for a computerized process-control system that can be manually overridden only by qualified individuals.

Operations in the process cell(s) will be maintained continuously for periods as long as practical. Short operating periods will be minimized because they tend to produce large quantities of scrap. During extended shutdowns, HLLW will be removed from processes where leaving it could compromise product quality and/or equipment performance. Such waste will be removed by completing process steps prior to shutdown or by flushing, vacuuming or otherwise transferring the waste to other locations.

3.7.2 Process Cell Requirements

All operations within the process cell(s) will be performed remotely. Since remotely controlling operations in a nuclear facility is a very specialized and demanding activity, the design of the facility and the process cell(s) will provide for continuous operations. Process cell operations will be performed and/or monitored from a central control room. However, control may be selectively exercised from local control consoles which will be supplemented by viewing windows, closed circuit televisions or periscopes. The control consoles will also have instrument systems for detecting equipment malfunctions and fluctuations in the properties of the HLLW. Routine adjustments and the rerouting of process materials will be done from the consoles. Other adjustments or malfunctions may be corrected through the use of manipulators or other devices.

The process cell(s) will be designed and constructed according to the highest level of structural classification to confine radioactive material during or following any design-based

accident. The cell walls will be constructed of ordinary or high-density reinforced concrete. The walls will be thick enough to reduce the radiation dose rate at the outer wall to 1 mrem/h. The cells will be lined with 304L stainless steel sheet to aid in cell decontamination and decommissioning and to protect the cell walls from corrosive chemicals and vapors. All structural steel in the cells, including crane rails, fittings, anchorage and embedded metal items, will be made from 304L stainless steel or 304 stainless steel-clad carbon steel.

Equipment mounted on process cell walls will be mounted on ears, yokes or racks. Equipment on the floor of the cell will be mounted on guide racks or dunnages. Manipulators and cranes will move on rails built into the cell walls. Wall penetrations to adjacent operating galleries will have in-service piping and electrical and instrument connections. The placement of facilities and equipment will depend on how equipment is handled. The width and height of the process cell(s) will be determined by the size of the equipment and remote-maintenance and remote-servicing requirements.

Small, remote-maintenance station(s) will be located in each process cell for minor and routine maintenance. A viewing window and remotely operable master-slave manipulators will be located at each station. In addition, television cameras will be strategically located to provide coverage for operating, inspecting and repairing equipment. Floor and wall space will be provided in each process cell(s) for the storage of tools and other maintenance equipment. Space will also be provided between the process equipment in the cell for handling equipment and for moving items.

3.7.3 Process Cell General Equipment Requirements

Remote operations require that all process equipment and equipment for handling process materials either be automated or be mechanized and remotely operable. Therefore, process cell equipment will be designed for simplicity, reliability and easy maintenance. Moving parts that require frequent maintenance will be used as little as possible. The use of master-slave and electro-mechanical manipulators for the remote operation of equipment will also be kept to a minimum. Finally, process cell equipment will be made from materials that are resistant to corrosion, radiation damage and processing conditions.

The process equipment will be designed and built in modules. Each module will be designed for remote installation and removal using a crane hoist and/or manipulators. Equipment modules that are expected to fail within the lifetime of the facility will be remotely replaceable without removing other major equipment. Modules that will require frequent maintenance will be located in shielded niches in the cell walls, where they will be accessible. Modules will be mounted on racks, rails and other supporting devices. The dimensions of the modules will be strictly controlled. Alignment dowels, studs and fixed mounting trunions will be used to ensure exact positioning of equipment and components.

Auxiliary equipment that will require contact maintenance, such as feeders, welders and mass spectrometers, will be located outside the cell if possible. Air locks, anti-backflow devices and block valves will be provided between the in-cell and out-of-cell equipment to ensure safe operations.

Hanford-type and other remote piping and electrical connectors will be used for service and process connections. Hanford-type, remote tank-flanges may also be used to install pumps, agitators and filter assemblies. "V"-flanges and other quick-connective devices may also be used.

Other general equipment requirements are listed below:

- The amount of equipment used in the solidification process will be kept to a minimum.
- There will be a low incidence of failure of the solidification process.
- Equipment will be used that can be remotely changed quickly and easily.
- There will be provisions for disconnecting, reconnecting, moving, reinstalling and adjusting equipment and components.
- Quick-disconnect power, gas and hydraulic devices will be used.
- There will be provisions for decontaminating the equipment and for the removal of any process material.
- Oil-based hydraulic systems or components will be used as little as possible.
- There will be considerable redundancy in facility service, maintenance and transfer equipment to reduce plant downtime and radiation exposure.

3.7.4 Major Processing Equipment Requirements

Processing equipment surfaces normally exposed to HLLW will be isolated from the cell environment to the extent practical. This will usually be accomplished by housing or otherwise containing the surfaces exposed to HLLW. The contained areas will be maintained at pressures lower than those maintained in the cell by connecting the contained areas to the ventilation system or the off-gas system. Process equipment that may be exposed to internal pressures exceeding the capacity of the ventilation system or the off-gas system will be fitted with devices that permit the safe release of pressure to the cell. This release of pressure will not result in pressurizing the cell relative to areas of lower controlled activity and will not damage the cell or other equipment.

Processing equipment will be designed to minimize the buildup of material in the equipment. Equipment that will be prone to the internal buildup of material will be fitted with sensors to monitor buildups. Provisions will be made for in-situ internal decontamination (e.g., by vacuuming or flushing) of process equipment prone to failure or to material buildups that could require disassembling and expose highly contaminated surfaces to the cell environment. Tanks and hoppers whose functions are to accumulate HLLW will be designed for natural or forced cooling.

Process equipment that may be exposed to internal fire and/or explosive gas mixtures will be fitted with monitoring instruments and suppression devices such as inert-gas purges and water spray-equipment.

3.7.5 Instrument Systems

Instrument lines will be designed to facilitate calibration work and cleaning of plugged lines. Out-of-cell transmitters that could potentially become contaminated will be protected by equipment that will prevent the backflow of radioactive materials. Instrument systems will be standardized, and maintenance stations will be provided with devices for making instrument repairs.

3.7.6 Piping Systems

All processing services and routings between process equipment will be designed for remote installation and removal. Lines between the gallery areas and the process vessels will normally be sloped to drain to the process vessels. Piping systems will be designed to prevent radioactive solutions from siphoning or backing up into areas of lower radioactivity and to minimize holdup of material. The capability to clean out piping systems in situ will be provided.

3.7.7 Sampling Equipment

It will be necessary to sample materials for process control and quality assurance. The required samples will be collected at remote sampling stations, where simple analyses such as calorimetry and gamma spectrometry may be performed. Samples for more complex analyses will be decontaminated at the sampling stations and transferred to the analytical laboratory via a pneumatic-tube transfer system.

3.7.8 Manipulators and Cranes

The major handling equipment in the process cell(s) will include bridge crane(s), bridge-mounted, electro-servo or electro-mechanical manipulators (EMM) and/or wall-traversing EMM, and master-slave manipulators. Manipulators will be used to perform all remote functions that cannot be adequately performed using impact wrenches and other tools suspended from the bridge cranes. Programmed manipulators may be used to perform functions that are repeated often. Electro-mechanical and programmable manipulators will be preferred over master-slave manipulators.

3.8 SUPPORT GROUPS AND FUNCTIONS

- A quality assurance program will be established for the design, construction and operation of the waste facility in accordance with DOE Manual, Richland Operations Office, Appendix 0820 (DOE 1974).
- Facility protection against fire and subversive activities will be provided. Automatic fire-suppression systems will be used. A fire department will also be located onsite to provide back-up fire protection.

The waste facility will also be designed and operated in such a way as to discourage subversive activities. A nuclear-materials accountability program will be established to control the inventory and movement of radioactive materials. A plant security force will provide back-up protection.

- An analytical laboratory will perform analyses on process materials in the number and to the degree of accuracy required to support all operations.
- Radiation monitoring procedures will be established to monitor and control personnel radiation exposure and releases of radioactivity inside and outside the plant.
- A process-engineering group will monitor process data and will provide adequate staffing to resolve technical problems.
- An operations group will be responsible for operating the process within licensing and production requirements.
- A maintenance team will be responsible for repairing equipment at a rate commensurate with production goals.

3.9 SUPPORT SERVICES

3.9.1 Utilities and Materials

Utilities such as cooling water, process water, steam, electricity, instrument and ventilation air, and emergency utilities will be provided by the fuel reprocessing plant. (Emergency utilities will be provided to ensure containment of radioactivity and general safety during periods in which normal utilities are not operating). Heating, ventilation and air conditioning utilities will also be located in the fuel reprocessing plant. Special utilities such as argon and high-pressure air will be located in the waste facility. Process materials such as canisters, helium, welding equipment, spare equipment and chemicals may be stored in the fuel-reprocessing-plant warehouse. Chemicals will be prepared in the aqueous make-up area of the waste facility. Utilities and process materials will be delivered to contaminated areas using equipment (and procedures) that will prevent the backflow of radioactive contamination. Such equipment will include check valves, continuous purges, funnel breaks and air locks.

3.9.2 Decontamination

Decontamination services will be provided to reduce surface contamination of waste canisters and equipment that has failed to levels permitting their further handling and/or ultimate disposal. Within the decontamination cell(s), a variety of decontamination methods such as flushing, leaching, steam cleaning, brushing, ultrasonic cleaning and electro polishing may be used. All effluents from the decontamination process will be compatible with either the HLLW solidification process or a cementation process for immobilizing intermediate- and low-level wastes. Access from the decontamination cells to the processing cells and maintenance areas will be provided.

3.9.3 Maintenance

The waste facility will employ both remote- and contact-maintenance techniques to maintain the process equipment and support systems. A well equipped maintenance facility will provide for the remote repair and check-out of contaminated equipment and for contact maintenance on clean or decontaminated equipment. The maintenance facility will be supplemented by remote-

maintenance stations located in each major cell, where a piece of equipment could be sent for minor repairs or for the replacement of modules. Provisions to decontaminate and perform contact maintenance on maintenance equipment, such as bridge cranes and manipulators, will be provided.

Most defective equipment in the processing cells and other remote areas will be replaced with spare units. The defective equipment will be disconnected and either moved to a decontamination cell for cleaning and then to a maintenance area for contact maintenance, or moved directly to a maintenance station for remote maintenance. If the unit can be repaired, it will be repaired and either reinstalled or stored for future use. If not repairable, the unit may undergo further decontamination and dismantling before it is compacted, placed in a container and disposed of. All new and replacement components will be checked dimensionally and operationally in mock-up test stations before being placed in service.

3.9.4 Secondary Waste Processing

Secondary radioactive wastes will be generated in the processing of HLLW. These wastes will include solids, liquids and gases. Facilities for handling these wastes, such as the incineration and cementation facilities, will also treat similar wastes generated by the fuel reprocessing plant; therefore, these facilities are not considered part of the waste facility in this study.

Miscellaneous process scrap will normally be dissolved, or crushed to a fine powder, and recycled through the HLLW solidification process. If the high-level scrap is not compatible with the HLLW solidification process, it will be diluted or otherwise treated to conform to cementation facility processing specifications.

Solid wastes that are noncombustible include failed processing equipment, tools, light bulbs, wire ends and miscellaneous waste. Most noncombustible solid wastes will be decontaminated by methods such as flushing, leaching, steam cleaning, brush scrubbing or ultrasonic cleaning. By-products generated in the decontamination process may be solidified in the cementation facility. Following decontamination, solid noncombustible wastes will normally be reduced in volume by methods such as dismantling, cutting, shredding or ram compacting. These wastes will then be packed in drums, canisters and boxes and disposed of.

Noncombustible liquid wastes include decontamination solutions, condensates and scrubber solutions. All high-level noncombustible liquid waste will be concentrated and transferred to a HLLW holding tank for recycling if the waste is compatible with the particular HLLW solidification process. If not, the waste will be diluted or otherwise treated to conform to cementation facility processing specifications. Intermediate- or low-level, noncombustible liquid waste will normally be concentrated and solidified in the cementation facility.

Combustible solid wastes, such as cloth, paper, wood, plastics and resins, and combustible liquid wastes, such as solvents and oils, will be separated into low-level and intermediate-level streams and burned in two separate incineration processes. Ash from the intermediate-level-waste incinerator will normally be solidified in the cementation facility.

Gaseous wastes include gases from the HLLW drying and heating processes and gases from the vessel and cell ventilation systems. The decontamination of gases from the HLLW drying and heating processes is treated in this report as part of the overall HLLW solidification process. Gases from the ventilation systems will normally be passed through high-efficiency particulate air (HEPA) filters before being discharged to the air protection system. Spent HEPA filters will be handled as solid waste.

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APPENDIX A

IN-CAN GLASS MELTING PROCESS

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APPENDIX A IN-CAN GLASS MELTING PROCESS

A.1 INTRODUCTION

The ICGM process is basically a two-step treatment of the HLLW (Larson and Bonner 1976). The first step is to dry the HLLW to form a calcine. A glass frit that yields a durable borosilicate glass is added to the calcine as it is formed. The mixture is then melted into a molten-glass solution that is approximately 30 wt% waste. This melting process takes place in the actual storage canister.

The ICGM process has several desirable attributes. First, it is basically a continuous process involving few steps. Each canister, of course, is handled on a batch basis. Second, either the process materials are in chemical solution, or gravity can be used to convey process materials from one process step to another. Finally, the ICGM process requires few remotely operated mechanical devices. The ICGM process for making borosilicate waste glass has undergone substantial development and has been demonstrated with actual HLLW. In this document, the ICGM process is the reference process.

A.2 PROCESS DESCRIPTION

The ICGM process is depicted in Figure A-1. A HLLW feed system stores the HLLW and assures a homogeneous feed to a spray calciner. The spray calciner atomizes the HLLW feed inside an externally heated barrel-furnace, where the HLLW is dried and essentially converted to an oxide form. The steam, atomizing air and NO_x gases are vented to an off-gas system through a sintered-metal filter box attached to the spray calciner. A gravimetric frit-feeder feeds glass frit, in the proper ratio, into the bottom funnel of the spray calciner, where it mixes with the calcine and falls through a connecting section to a canister inside an in-can melting furnace, or in-can melter (ICM). The connecting section leads to two canisters. When one canister is full, the calcine/frit mixture is diverted to the second canister, thereby permitting continuous operation.

Because of the continuous operation, very little calcine builds up in the spray calciner. The calcine that does build up on the walls of the spray calciner barrel is periodically vibrated loose. Calcine that is deposited on the sintered-metal filters is blown off by compressed-air pulses. The loosened calcine falls into the canister. Both the spray calciner and the in-can melting processes have been shown to be tolerant of a wide range of operating parameters in tests conducted at PNL.

When a canister is full, it undergoes what is called "fining" during which melting of the calcine and glass frit goes to completion at a temperature of approximately 1050°C . After the calcine and frit have completely melted, a cooldown period is initiated. Once the canister has recovered sufficient tensile strength ($\sim 700^\circ\text{C}$), the canister is removed and transferred to a forced-air cooling rack. There, it is cooled further to prevent crystals from forming in the glass. The canister is then transferred to a lid-closure station, where a helium source is placed inside the canister and a lid is remotely welded to the canister. The closure weld and the canister are then tested for helium leaks. Provisions are made for overpacking the canister if a leak is detected. Finally, the exterior of the canister is decontaminated and the canister is transported to storage.

The off-gas system consists of a serial arrangement of units that process gases and particulates. The system removes particulates, removes volatile fission-products, condenses steam, and recovers some of the NO_x gases. The decontaminated air is fed to the fuel reprocessing plant's air protection system.

The ICGM process has been designated the reference process in this study because the majority of the other processes also require a HLLW feed system, a spray calciner, an off-gas system and canister treatment such as closure welding, leak checking and decontamination.

FIGURE A-1. Flow Diagram for ICGM Process

A.3 BASES AND ASSUMPTIONS

Some of the criteria, bases and assumptions for the ICGM process are noted in Section 3, Overall Process Bases. Other applicable items are noted below.

A.3.1 PROCESS BASES

The constituents of the HLLW and the history of the fuel from which it is derived are given in Section 3. Figure A-2 is a flowsheet of the mass balances for the ICGM process. The major constituents of the waste glass are given in Table A-1.

Table A-2 gives the important characteristics of the ICGM process materials and the waste glass product. Glass has proven to be a particularly good waste form because it is relatively easy to make, and when glass is a solution it dissolves a wide range of radionuclides. In addition, the glass frit that is used in the ICGM process has been formulated to exhibit durability and stability, and it has a relatively low melting temperature of 1050°C. A potential disadvantage of glass is its tendency to devitrify into a more water-soluble crystalline form under certain oxidation states during melting and under certain temperature conditions during cooldown. Further work is being done to identify operating parameters that can be controlled to prevent devitrification.

The waste facility will solidify the HLLW produced in a fuel reprocessing plant operating at a rate of 2000 MTU/yr. The design factor is 1.5 times the nominal operating capacity. The plant is designed to solidify 5700 L of HLLW/d into 1750 kg of glass. This requires a glass melting rate of 73 kg/h and the filling of a canister every 3 h, 45 min. Glass can be melted at this rate in an ICM, but the entire cycle which includes canister preheating, glass fining and discharging of off gas, and switching canisters is judged to require at least 6 h. Therefore, it was assumed that the process equipment will be constrained to two equal-sized process units each consisting of a spray calciner and two in-can melting furnaces; this assumption allows 7.5 h to fill and process a canister.

The requirements for storing waste canisters in a salt repository will restrict the decay heat of a waste canister at the time of emplacement. As discussed in the Overall Process Bases Section, the waste canisters are assumed to be filled with waste from five-year-old fuel and then stored for an interim period of up to 50 yr. Therefore, ICGM canisters will be filled with 273 kg (91 L) of waste glass that produce 3.06 kW of heat at the time of filling.

A.3.2 CANISTER

Some general requirements for waste canisters are given in the Overall Process Bases Section. Additional general design criteria are given below:

- The canister must be compatible with a remotely operated waste facility and it will be handled by a single device.

FIGURE A-2. Mass Balance Flowsheet for ICGM Process^(h)

(a) metals as NO₃ or PO₄ salts

(b) L/MTHM

(c) fission products

(d) actinides

(e) activation products

(f) 2M nitric acid

(g) g mol/MTHM

(h) based on nominal operating capacity of 3800 L/d of HLLW

TABLE A-1. Major Constituents of the Waste Glass

<u>Oxide</u>	<u>Quantity, wt%</u>
Na ₂ O	9.0
Fe ₂ O ₃	0.93
NiO	0.06
Cr ₂ O ₃	0.16
Gd ₂ O ₃	5.9
FP _x O _y	18.0
Act ₂ O _g	0.7
UO ₃	3.3
PuO ₂	0.04
AP _r O _t	0.23
P ₂ O ₅	0.65
SiO ₂	38.0
B ₂ O ₃	11.0
TiO ₂	4.2
Al ₂ O ₃	1.0
K ₂ O	5.2
CaO	2.1

TABLE A-2. Characteristics of Process Materials and Waste Glass Product

<u>Characteristics</u>	<u>Process Materials</u>			<u>Waste Glass Product</u>
	<u>HLLW</u>	<u>Calcine</u>	<u>Frit</u>	
Specific gravity at 4°C	1.17	1.2	1.6	3.0
Temperature, °C	40	500	Ambient	800
Heat loading, W/L	3.46	44.9	---	33.7
L/MTHM	567	43.7	76.6	58.3
Nominal operating rate, L/d	3800	292	510	389

- The canister must be compatible with an overpack. An overpack may be used if the canister does not meet quality specifications or if added protection is necessary during disposal.
- The canister must be capable of withstanding the conditions in the furnace during the melting operation. Specifically, the canister must maintain integrity at the glass melting temperature of 1050°C, with a design margin of 50°C to accommodate a high-temperature condition in the furnace.
- The canister must provide the necessary protection and containment of the waste glass during normal and accident conditions; any operations involving the canister must not adversely affect the waste glass, including 50 yr of interim storage and transport to a repository.

Specific design features of waste canisters that should be considered are size, material and canister wall thickness. The size of the canister will be determined by the .5 kW heat loading permitted at the repository. Therefore, based on a 30 wt% waste loading with a glass density of 3 g/cm^2 and the assumptions regarding reprocessing and storage given in the Overall Process Bases Section, the amount of glass in a canister will be limited to 91 L (273 kg). However, the canister must also have a free space of approximately 30 cm (11.8 in.) at the top to accommodate some pile-up and foaming. In addition, the canister will have eight internal fins to facilitate heat transfer that will take up 5.6% (7.2 L) of the canister volume. Thus, assuming a 30.5 cm (12 in.) dia, the length of the canister will be 176 cm (69 in.) plus a 10-cm (4 in.) flange/handling device, which will make the overall length 186 cm (6.1 ft). Figure A-3 is a cutaway of the canister showing the internal fin assembly and the approximate configuration.

The canister wall thickness is determined by taking into consideration the lifting load at elevated temperatures, internal corrosion, external oxidation, creep, spallation, tensile stress during cooling, ruggedness factors and canister material. These are discussed in more detail below.

The canister will be cooled from 1050°C to 700°C over a two-hour period in order to restore strength to the canister wall before lifting the canister. For handling and impact stresses, the rules of the cask designer's guide (Shapperd 1968) have served as a preliminary basis for canister design. These rules provide a safety factor of 3.0 on yield for a static 1.0-g lifting load. The ruggedness factor is protection against canister rupture in the event of dropping or impact during movement. These criteria have not been well developed as of this time. However, it is anticipated that the ruggedness factor may result in a wall thickness that represents an order-of-magnitude safety factor for the other wall thickness design criteria already described.

At the temperatures of the ICGM process, both corrosion and creep deformation must be considered. Data on internal corrosion caused by the glass and external oxidation from the furnace environment are both covered by Slate and Maness (1978). The creep strength of candidate canister materials and design methods for the creep regime are addressed by Simonen and Slate (1977) and VanEcho and Simonen (1978).

During cooling below the glass transition temperature, the walls of the canister are placed in a state of tensile stress. This is due to the fact that the glass contracts less as it cools than does the steel canister. Residual stresses in the canister have been measured up to the yield strength of 235 MPa (35,000 psi) under some conditions. Tests indicate that rapid water quenching can redistribute the stresses and possibly reduce the canister stresses to much lower levels. However, such quenching would conflict with the desire to minimize

FIGURE A-3. Cutaway View of ICGM Process Canister

thermal stress cracking in the glass. Therefore, cooling will be carried out with forced air in such a way as to optimize canister stress redistribution while minimizing glass cracking. Since water basin storage is specified in this process, the canister material should be either stainless steel or a nickel-chromium-iron type alloy. If stainless steels are used, care must be exercised so that the combined effects of alloy composition, material sensitization, residual stress and water chemistry do not lead to stress-corrosion cracking during interim water storage.

For the preconceptual design phase the canister wall thickness is specified as 64 ^{mm} (1/4 in.) and the material as stainless steel from the 304 series. This material costs about 60% less than a nickel-chromium-iron type alloy. However, one advantage of the latter alloy is that it does not undergo significant external oxidation and would probably not require the inert argon atmosphere within the retort. Also, the nickel-chromium-iron type alloy is not as prone to sensitization.

A.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

Nonradioactive solid wastes, such as general office and shop wastes, will be handled by facilities in the fuel reprocessing plant. Radioactive liquid effluents will be recycled within the waste facility as HLLW or intermediate-level liquid waste (ILLW). It is also assumed that radioactive process scrap will be recycled and incorporated into the ICGM process. Other nonradioactive and radioactive wastes that will require processing are listed in Table A-3. There will be systems in the fuel reprocessing plant to treat, reduce the volume, and package the liquid and solid wastes generated in the ICGM process.

TABLE A-3. Secondary Nonradioactive and Radioactive Wastes That Will Require Processing

<u>Secondary Wastes</u>	<u>Estimate of Volume of Waste per Unit Time</u>
Nonradioactive	
Liquids	
Cooling water	500 L/min
Process water	50 L/min
Sanitary waste	10 L/min
Gases	
Ventilation flow	500 scmm
NO _x	10 L/min
Radioactive	
Off-Gas system and ventilation flows	5 Ci/yr
Solids	
Trash (paper, plastics, cloth and glass)	200 m ³ /yr (20 Ci/yr)
Failed equipment ^(a)	50 m ³ /yr ^(b)

(a) The volume of failed equipment is a function of the complexity of the process.

(b) Partially compacted (10⁴ Ci/yr if well decontaminated).

A.3.4 UTILITIES AND MATERIALS

Estimates of the normal utilities and materials needed for the ICGM process are given in Tables A-4 and A-5, respectively. Any emergency utilities that might be required, such as electricity or cooling water, are not included, nor are maintenance and replacement materials. Only chemicals used for periodic cleanup are included.

TABLE A-4. Estimate of Utilities Needed for the ICGM Process

<u>Utility</u>	<u>Required Capacity</u>	<u>Average Use Rate</u>
Electricity	1000 kWA	600 kWA
Water	1800 L/min	800 L/min
Air	750 scmm	500 scmm
Steam	15 kg/min	2 kg/min

TABLE A-5. Estimate of Materials Needed for the ICGM Process

<u>Material</u>	<u>Amount Needed per Year</u>
Frit	246,000 kg
Canisters	1,282
Helium sources	1,282
Oxygen	19,240 scm
Nitric acid (12.2M)	12,000 L
Caustic (19M)	8,000 L
Argon	1,500 scm
Detergent	1,000 kg

A.3.5 INTERIM STORAGE

There will be storage facilities within the fuel reprocessing plant for the required interim storage of the waste canisters before they are shipped to a federal repository. Waste canisters from the ICGM process will be stored under water, which will be both the shielding and cooling medium.

A.4 FACILITY AND EQUIPMENT REQUIREMENTS

The design requirements for a waste facility are generally similar for all the solidification processes and are, therefore, presented in Section 3, Overall Design Bases. However, an attempt has been made to determine the size of the ICGM process cell so that the size and complexity of the process cells for each solidification process can be compared. A preliminary layout of the process cell and supporting operations and maintenance cells for the ICGM process is shown in Figure A-4. To estimate the size of the process cell, the volume of each piece of major equipment used in the ICGM process was estimated. A list of major processing equipment and their volume requirements is given in Table A-6. The volume estimate for each piece of equipment was obtained by determining the size of the actual piece of equipment and adding approximately a one-half meter clearance around the equipment to allow for protuberances, support brackets and service connections. The estimate also includes space above the canister processing stations equivalent to one canister height. Finally, the layout in Figure A-4 allows for the space required for remote operation and maintenance, equipment replacement and decontamination of process equipment. These space allowances were added on the basis of engineering judgment.

FIGURE A-4. In-Can Glass Melting Process Cell and Equipment Arrangement

TABLE A-6. Process Cell Volume Requirements for Major Processing Equipment

<u>Equipment</u>	<u>Dimensions (width x depth x height), m</u>	<u>Volume, m³</u>
HLLW batch tank A	2.5 x 2.5 x 4	25
HLLW batch tank B	2.5 x 2.5 x 4	25
HLLW feed tank	1.5 x 1.5 x 3	7
HLLW piping manifold	2 x 3 x 3	18
HLLW sample analysis equipment	2 x 3 x 3	18
Spray calciner 1	3.5 x 3 x 4	42
Spray calciner 2	3.5 x 3 x 4	42
ICM furnace A	2 x 5 x 4	40
ICM furnace B	2 x 5 x 4	40
ICM furnace C	2 x 5 x 4	40
ICM furnace D	2 x 5 x 4	40
Forced-air cooling rack	2 x 3 x 4	24
Lid-closure station	2 x 2 x 4	16
Helium leak-detection station	2 x 2 x 4	16
Decontamination station	2 x 4 x 5	40
Canister rack	2 x 3 x 4	24
Storage transfer	2 x 2 x 3	<u>12</u>
TOTAL		469

A.5 STATE OF PROCESS DEVELOPMENT

The ICGM process involves mechanical and chemical operations that must be performed remotely by equipment that will require remote maintenance and remote removal in a production plant. In addition, some of the required process steps have not been performed on a basis comparable to that for a full-sized production plant.

The spray calcination equipment is highly developed. Spray calciners were tested extensively in a nonradioactive pilot plant and in design verification tests before the Waste Solidification Engineering Prototype (WSEP) runs were started using radioactive waste. In the cold, pilot-plant studies, over 6000 L of simulated waste were calcined in about 440 run h. The WSEP program was designed to yield information needed for technical, economic and safety evaluations of the spray calciner. About 8600 L of high-level aqueous radioactive waste containing 25-M Ci were successfully solidified in 13 demonstration runs over a three-year period (1966 to 1970) with a total run time of about 580 h. In the WSEP program, the remote operation and maintenance of the calciner, feed system and effluent treatment system were demonstrated using an overhead crane and master-slave manipulators.

The Waste Fixation Program (WFP) began at PNL in 1972. As part of this program, the heated-wall spray calciner is currently being prepared in connection with a full-scale ICM as a total waste solidification system. Recent development has resolved questions concerning atomizing nozzle wear and blockage, interior-wall scale accumulation and spray calciner capacity. To date over 1072 h of operation with the pilot-scale spray calciner and 1525 h of operation with the full-scale spray calciner have been logged processing simulated high- and intermediate-level wastes.

The Nuclear Waste Vittrification Project (NWVP) was initiated in 1976 to demonstrate the solidification and packaging of actual HLLW (from light-water reactor (LWR) fuels) as borosilicate glass in canisters. The project was completed in 1979 after two canisters of actual waste were prepared. The spray calciner used for this project processed approximately 1236 L of actual HLLW in 87 h of operating time.

The state of development of in-can melting is also well advanced. It was one of the vittrification processes demonstrated in the WSEP program using radioactive waste and it was the vittrification process used in the more recent NWVP program to produce two canisters of borosilicate glass containing actual radioactive waste. In addition, at PNL the pilot-scale and the full-scale ICMs have been operated more than 900 h at glass melting temperatures using simulated waste glasses. This time is essentially only the time required to melt the glass and does not include preheating and cooldown time. The difference between ICM and spray calciner operating times is due to the fact that spray calciners were used to produce calcine for other types of vittrification melters being developed.

The knowledge of the off-gas-system process requirements for the ICGM process has been developed to an advanced level by various ongoing programs. However, scaling the system up to handle larger volumes and incorporating remote features will require additional developmental efforts.

A large amount of work has been performed on developing the waste storage canister and the necessary systems for processing it. Two basic designs for canisters have been developed and analyzed. A report by Simonen and Slate (1979) provides a detailed description of the analyses that have been performed. The technology is available to design a canister and to develop the necessary processing equipment for a particular solidification process or plant. Some nonradioactive demonstration work would be required before an actual remote facility is built.

The filling of about 30 full-scale canisters using the ICGM process has been demonstrated. The dia of these canisters ranged between 12 in. and 28 in., and the materials used were 304L and Inconel 601[®]. In addition, about 70 canisters developed for or filled by the JHGM process have been evaluated and tested. Other developmental studies made on canisters and canisters of waste or simulated waste include:

- the application of various cooling cycles and the evaluation of the effects on the glass
- remote welding and inspection of the weld
- placement of canisters in water storage for long-term evaluation of corrosion
- establishing a data base for canister design.

[®] Tradename of International Nickel Co.

A.6 UNIT OPERATIONS

The ICGM process can be divided into five major unit operations: the HLLW feed system, the spray calciner, the in-can melting furnace (or ICM), canister treatment and the off-gas system. In this section, each of these unit operations is discussed in detail, including the equipment that is used and areas needing further research and development. Drawings and data sheets on individual pieces of equipment discussed in this section can be found at the back of Appendix A in Section A.8.

A.6.1 HLLW FEED SYSTEM

Due to the concern and need for limiting the possibility of contamination spreading from different systems, the majority of the HLLW feed system will be located in a separate cell from the one housing the major processing equipment. The HLLW feed system must cool, continuously agitate, ventilate and transfer the HLLW. Specific requirements for the HLLW feed system are listed in Table A-10 in Section A.8. The following equipment is part of the HLLW feed system:

- HLLW storage tanks
- HLLW feed tank
- agitators (both mechanical and hydraulic)
- transfer and feed pumps
- cooling coils and jackets
- tank ventilation systems
- sampling system
- recirculation system
- instrument system

Two equal-sized storage or batch tanks each having the capacity to hold a two-day feed supply of HLLW (11,400 L) are required. This allows feeding from one tank while sampling and necessary chemical composition treatments are carried out on the other tank. There is also a smaller feed tank. Thus, HLLW can be transferred in batches from either storage tank to the feed tank. Information concerning fissile radionuclide control must be factored into the design of the system to preclude any chance of criticality from sludge buildup and to protect against inadvertent transfer of solutions containing fissile concentrations that could approach criticality.

An agitation system is necessary to keep all solids suspended in the HLLW. It includes a recirculation feed line that circulates the HLLW towards the spray calciner where a smaller, single line takes the HLLW to the spray calciner nozzle. The recirculation line is sized to supply sufficient volume to prevent sludge deposition in the piping. Provisions for water, steam and air flushing are required for the recirculation line. Tank agitation for all three tanks is a combination of mechanical agitation by motor-driven paddles through the tank top and high-volume recirculation lines with replaceable jets that can be withdrawn and replaced

through the top of the tanks. Since HLLW is an abrasive solution that will cause wear in the high-volume recirculation lines, maintenance and replacement ease is a major design requirement.

The HLLW produces a maximum of 3.46 W/L of decay heat. In addition, pump and agitation energy is added to the solution. The temperature should be maintained at less than 40°C to prevent excessive evaporation. The cooling coils should be able to handle the projected heat load; however, the tank cooling jackets should provide enough cooling independently of the coils to prevent the HLLW from boiling in the event the cooling coils are unavailable. Since there is a potential for leakage of the HLLW into the cooling water in the event of damage to the coils or tank jacket, the cooling water should be isolated from the ultimate heat sink by a heat exchanger. In addition, the closed-circuit, cooling-water system should have redundant radiation alarms to detect any leakage.

A redundant tank ventilation system is required to remove the hydrogen and oxygen resulting from radiolytic decomposition as well as any volatile radionuclides and NO_x given off by the solution.

The HLLW sampling system is used to measure the specific gravity, heat content, molarity and radioactivity of the solution and concentrations of certain key elements. This data is used to determine the proper ratio of frit to calcine necessary to produce high-quality borosilicate glass. Accurate characterization of the HLLW may be even more important when the final waste form is to be a ceramic, since there may be less margin allowed in the process parameters than there is in the case of the glass waste form. The sampling system permits the retrieval of liter-quantity samples inside an analytical chemistry cell, where more detailed analysis of the HLLW can be conducted. The instrument system for the HLLW feed system must measure the following:

- temperature of 1) all tanks, 2) closed-circuit cooling water and 3) ultimate heat sink
- level of 1) all tanks and 2) flush water
- flow rates on the feed line
- pressure on the feed pump and the feed and recirculation lines
- valve position on all remotely actuated valves
- electrical current on all motors
- alarms on 1) high tank level, 2) low tank level, 3) radiation in sump, 4) radiation in closed-circuit cooling water, 5) criticality, 6) high temperature and 7) low tank vacuum
- interlocks as required by final design.

The primary considerations in operating the HLLW system are feeding a homogeneous solution of known characteristics, avoiding sludge buildup in the tanks and lines, and avoiding spills and equipment damage due to improper operating procedures. The uninterrupted feeding of a well characterized homogeneous solution is achievable if an efficient sampling system is developed

and if the agitation system can prevent a buildup of sludge and can screen out solids that may plug the small-diameter nozzle feed line. For the preconceptual design stage, the intertank transfers are to be made manually by the operator. As always there is the potential for human error, but an adequately designed alarm system will alert the operator to out-of-limit process parameters and will serve as a backup to prevent human error. Process time-sequence studies will have to insure that plant processes that demand operator action cannot occur simultaneously, or that in the event of simultaneous operator requirements there are sufficient operators on shift to prevent overfilling or valve misalignment.

Since sludge buildups, spills and equipment damages are likely to occur as a result of human error and since the tank piping is relatively complex to provide redundancy and flexibility in transferring and sampling HLLW, thorough operating procedures will be required. For example, flushing and blowing out the transfer lines and keeping flush water recirculating in an empty tank will be procedurally controlled to minimize the opportunity for the formation of hard deposits.

Overdilution of the HLLW during operation of the system is permissible since the spray calciner will be conservatively rated and will be capable of calcining a more dilute HLLW. Overconcentration of the HLLW is a less likely event, but it can be corrected by diluting it if necessary.

The amount of redundancy required in the HLLW subsystems will be determined in part by taking the preconceptual design and performing a failure-mode analysis on the HLLW system. Research and development on system reliability may indicate the need for more interlocks in valve and pumps and the need for a programmed, feed-transfer control system. A sampling system that can give continuous readings also needs to be researched and developed.

The overall HLLW feed system is shown in Figure A-10 in Section A.8. The feed tank pump circulates HLLW in a loop at approximately 15,000 L/h, and from this loop a line with a capacity of 4000 L/h feeds the spray calciner. The feed line is a nominally small-diameter line to keep the fluid velocity high enough to prevent sedimentation. This small line and the orifice at the magnetic flowmeter and at the ceramic nozzle tip are subject to plugging if the HLLW system is allowed to generate sludge that is not filtered out by an appropriately sized strainer. Means of flushing out this section of feed line should be provided in the event of blockage or process shutdown. The flowmeter should be accurate to within 2% since inaccuracies in the HLLW feed rate and the frit feed rate will combine to produce an off-standard product.

A.6.2 SPRAY CALCINER SYSTEM

Calcination is one of the most effective means of transforming an aqueous waste slurry of nitrates into a dry oxide form. It is the first step for many of the waste form solidification processes. There are several calcination devices (see Section A.7, Alternative Processes and Equipment). The spray calciner has been selected for the ICGM process because of its simple construction and operation as compared to the other calcination devices. Minimum buildups of radioactive materials are a major advantage of the spray calciner.

As discussed in the preceding section on bases and assumptions, the ICGM process employs two spray calciners, each feeding two ICMs. Each of the spray calciner systems includes the following subsystems:

- atomizing nozzle and HLLW flowmeter
- spray calciner furnace and barrel
- sintered-metal filters
- seal pot
- frit delivery system.

The connecting section between the spray calciner and the ICM is treated as part of the ICM system. Also included is the frit delivery system.

During spray calciner operation, HLLW is metered and pumped to the spray calciner at a pressure of 1 to 2 atm. The waste is pneumatically atomized in an internal-mix nozzle and is sprayed into the top center of a cylindrical calcination chamber, as shown in Figure A-11 in Section A.8. The atomized liquid waste in the form of 70- μ m-dia droplets is sequentially evaporated, dried and calcined as it falls through the calcination chamber.

The nozzle is an internal-mix type with an alumina insert for the discharge port. The alumina insert is more wear resistant than a steel-tipped nozzle, although it is more susceptible to thermal shock. A cross-sectional view of the nozzle is shown in Figure A-12 (see Section A.8). Each nozzle is optimized for a particular operating range; in particular, the nozzle tip opening is sized to give maximum atomization with minimum atomizing air.

Automatic control of the HLLW feed rate has been demonstrated by two methods. One method is to use the back-pressure effect of the atomizing air to correct small variations in the liquid feed rate that are measured by the magnetic flowmeter. This type of control system needs to be restricted so that a blockage of flow does not allow the feedback control system to signal for complete reduction in the atomizing air. Otherwise, the blockage may clear suddenly, and without atomization a stream of the HLLW would be directed down into the ICM. The second automatic control system that has been demonstrated uses a control valve in the HLLW recirculation line.

The calciner chamber walls are heated to about 800°C by an external, multizone resistance furnace. Inside the chamber enough heat is transferred to the droplets by a combination of radiation and convection to produce calcine containing typically less than 0.5% moisture and less than 5% residual nitrate. Drying of the tiny droplets occurs in a fraction of a second, thus minimizing volatilization of radionuclides. Depositions on heat-transfer surfaces inside the calciner chamber are prevented by the periodic operation of side-mounted vibrators.

On passing through the chamber, water, vapor and atomizing air entrain 15% to 50% of the calcine particles, but more than 99.9% of these particulates are de-entrained by porous stainless steel filters. The calcine collected on the filters is periodically removed by a pulse of blowback air. The calcine that is removed from the filters falls into the calciner delivery funnel and is discharged along with the calcine that falls directly from the calcination chamber.

Several spray calciners have been operated and maintained in a radioactive environment. Therefore, considerable remote engineering technology exists. The atomizing nozzle, vibrators, furnace and sintered-metal filters, which are most prone to failure, should be readily removable. Complicating the remote maintenance of the items are the thermocouple leads, the air lines to the vibrators and filter blowback nozzles, and the electrical leads to the furnace, insulation and support brackets. The effect of considerable vibration must be taken into account when designing the furnace insulation and the support brackets. Metal fatigue should be analyzed because of the superimposition of vibrational loadings, which may work-harden the chamber and supports. Thermal cycling, which may lead to creep and stress cracking, should also be analyzed. Finally, there is a real need to be able to quickly identify and locate air inleakage. A system to adequately do this in a remote environment has not been demonstrated.

The spray calciner furnace, which heats the spray calciner chamber, can be either an induction or resistance type. For this preconceptual design, the furnace is specified to be a four-zone, resistance type. The division of the furnace into zones at different vertical levels is in recognition of the different heat loadings resulting from the convective air currents in the furnace shell and the spray pattern inside the barrel.

A preliminary drawing of the instrument system for the spray calciner is shown in Figure A-13 in Section A.8. The temperature measurement points of the spray calciner furnace and barrel are not all shown. A reasonable estimate would be eight temperature measurement points (some redundant) for each furnace zone and an additional eight points to monitor the nozzle, filters and delivery funnel regions.

Table A-11 (see Section A.8) is the equipment data sheet for the spray calciner system. In some cases, specific equipment is referenced. These examples are to aid in estimating costs, but are not intended to restrict design options.

A schematic of the glass frit feed system is shown in Figure A-14 in Section A.8. Since each calciner nominally requires 25.5 kg/h (612 kg/d) of glass frit, there must be some type of continuous air-lock mechanism to permit the continuous addition of frit into the cell. In this preconceptual design study, the frit feed mechanism is considered to be a gravimetric feeder. Auger/vibrator mechanisms have also been demonstrated, but are not recommended. In both cases, the frit feed mechanism has been identified as a high-maintenance item.

A.6.3 IN-CAN MELTING FURNACE

The in-can melting furnace is the central component of the ICGM process. This is because the glass frit and the calcined HLLW fall directly from the spray calciner into a canister inside a furnace where the mixture is melted to form the waste glass. The primary advantages of this process are continuous operation and a minimum number of process steps involving liquid or powdered waste forms.

As discussed in Section A.3, Bases and Assumptions, it is necessary to have four ICMs. Each ICM system includes the following subsystems:

- canister retort

- furnace
- spray calciner to canister connection
- furnace positioning mechanism
- canister weight and level indicator
- ICM instrument system.

The general arrangement of the ICM is shown in Figure A-15 in Section A.8. Table A-12 (also Section A.8) is the equipment data sheet for the ICM. More detailed information on ICM operations and research and development needed are given below for each subsystem.

The ICM furnace can be of the induction or resistance type. The resistance type is the more fully developed type. The furnace assembly is movable; it is presumably mounted on rails. Therefore, all of the services and instruments must connect through flexible leads. The furnace has several horizontal heating zones. Each of the horizontal zones is independently controlled by temperature feedback from a thermocouple probe into that zone of the furnace. This method of control has been demonstrated and found to be effective. The presence of a nickel-chromium-iron alloy retort within the furnace, to guard against contamination in the event of canister failure, has a small effect on temperature control in that it tends to smear out the temperature profile within the furnace. However, the control is still sufficient to respond to the need for more power when the glass level reaches that zone. Early research and development studies sought a method of cooling lower zones as they were filled with glass, primarily as a way to prevent overheating due to self-heating of the waste. However, zone cooling has not proved to be satisfactory when no self-heating is present. Therefore, the furnace is operated at a more or less constant temperature. The top and bottom furnace zones require more power because of end heat losses. The zone containing the glass melting level also requires more power to provide the necessary sensible heat and the heat of fusion.

The retort within the furnace is not a firm requirement of this process. However, a retort makes it easier to keep the canister atmosphere inert, which helps prevent oxidation and spallation of stainless steel canisters. A retort would also serve as a barrier to the spread of contamination in the event a canister should fail and breach. A retort would need to be designed to support and position the canisters to facilitate coupling to the spray calciner. In addition, after the canister is filled it must be cooled in situ to 700°C or lower to permit handling. Research and development work would be required to determine whether this can be accomplished with cooling air on the outside of the retort or whether the cooling air would have to be used inside the retort, thereby sweeping out the inert gas. A load cell that measures just the retort and canister weight, or in some cases the total furnace weight, is part of the ICM instrument system. The uncertainty about canister weight, because of forces transmitted through the connecting section, is a problem requiring further research and development.

The connecting section between the canister in the furnace and the spray calciner is probably the most mechanically complex component of the ICGM process. Figure A-16 in Section A.8 shows the general arrangement of the connecting section. The diverter valve obviously determines which canister is receiving the calcine/frit mixture. The cone valves serve two primary

purposes. They seal off the delivery tube to prevent contamination release and air inleakage during the canister changeout, and they tend to distribute the calcine/frit mixture evenly around the edges of the canister rather than let it pile up in the center. The cone valve and the lower portion of the connecting section have a heat-loading problem and must be cooled with water. Air cooling didn't work. If these surfaces are allowed to go much beyond 400°C, the glass frit will begin to melt and stick to these surfaces, ultimately causing blockages.

The connecting section has several design features that require further research and development. The connecting section flange must form a tight seal (some air inleakage permitted) with the canister flange, but the connection must be performed remotely and requires a system for alignment and quick connection. The cone valve extends down into the top of the canister and must be retracted and closed before the canister can be disconnected and the furnace moved. With this design it is possible for the stem of the valve passing through the delivery pipe to become jammed with material, which would prevent the canister from disconnecting. Research and development are required to come up with alternative designs to the cone valve or to establish backup procedures for freeing the connecting section. In addition, current prototype models of the connecting section use metal bellows to allow the connecting flange to be raised 4 to 6 in. in order to clear the top edge of the furnace when it is moved. This is due to the fact that the canister sits down in the retort chamber of the furnace. A movable, insulated collar fits over the top of the furnace and the retort to prevent excessive heat loss and to seal in the inert-gas atmosphere of the retort. Therefore, furnace movement and canister uncoupling as they are currently envisioned may be the most mechanically complex operations in the ICGM process. Controlling the position of the furnace is part of the system used to move the furnace and change the canister. The system for moving the furnace and changing the canister is described in Section A.6.4, Canister Closure, Decontamination and Transport System. As currently envisioned, a system of instrumented stops and guide pins would be incorporated into the design to insure accurate alignment.

Figure A-17 in Section A.8 is a preliminary drawing of the instrument system and the primary services required for the ICM. The general role of thermocouples in the furnace control feedback loop has already been discussed. There will also be several temperature monitoring points. The need for a system capable of tracking the weight of the glass fill has already been discussed. A prototype of a gamma-emission detection system was tested during canister filling with radioactive glass in the NWVP program and promises to give an accurate level detection reading.

A canister filling and changeout sequence involves the steps and approximate times given below:

- new canister is preheated as furnace is brought back up to temperature--2 h
- new canister is filled--7.5 h
- canister is held at temperature to assure complete melting and release of gas bubbles (called fining)--2 h

- canister is cooled enough to permit lifting from furnace--2 h
- furnace is moved and canister is switched--1.5 h.

These times are the design values, which reflect a fill rate 50% greater than the nominal operating rate. Therefore, during nominal operations all of these times will increase by 50%. The assumed repository requirements on the maximum decay heat allowed per canister restrict the amount of glass in a canister to 91 L; the canister is 30.5 cm in dia. These small canisters must be switched much more frequently than the larger canisters that have been used for nonradioactive, prototype research and development work. Therefore, it will be necessary to demonstrate that these preheating, fining, cooling and canister changeout rates can be achieved.

Some of the operating problems that have been encountered in ICM operation are bridging of frit/calcline at the top of the canister, plugging of the connecting section and foaming of the molten glass inside the canister. The foaming is a problem, particularly if it occurs when the canister is nearly full. Experience with ICM prototypes has shown that controlling the feed composition and operating within the melting capabilities of the furnace can greatly reduce the probability of these abnormal operating occurrences.

A.6.4 CANISTER CLOSURE, DECONTAMINATION AND TRANSPORT SYSTEM

The ICGM process is designed to produce a canister of waste glass every 3 h and 45 min. The following operations are required as part of the canister handling system:

- bring seven canisters into process cell
- place new canister in an ICM
- move ICM under spray calciner
- move ICM back to loading position
- move filled canister to forced-air cooling station
- move cooled canister to closure station
- move closed canister to leak-detection station
- move closed canister to decontamination station
- move decontaminated canister to storage rack
- move decontaminated canisters to interim storage.

For this design, a conservative approach has been taken in that all of the individual canister operating stations are separate. In a production facility design, it may be possible to combine the lid-welding and leak-detection stations. Combining stations is desirable in that it reduces the number of times a canister must be moved and reduces the size and complexity of the process cell. The undesirable effect of combining stations is the concentration of relatively complex equipment in a small area.

The canister handling system consists of overhead cranes for moving the canisters between operating stations and a track for moving the ICM furnace from a loading position to a filling position. Various types of manipulators are used for operation of the canister stations and for equipment maintenance. The canister transport sequence is shown in Figure A-5. The design

FIGURE A-5. Canister Transport Sequence

production rate of 6.4 canisters/d requires that one canister (but not the same one) be moved between all stations approximately every 3.75 h. If one crane is used to make all 6 of the transfers, a transfer must be made every 1/2 h. This assumes that the second crane can bring in the fresh canisters and remove the filled canisters.

To determine the number of canister racks required at each operating station, it is necessary to estimate the time that each operation (forced-air cooling, closure lid welding, etc.) will require and to allow extra time for potential difficulty in carrying out each operation. The maximum residence times shown in Figure A-5 were determined using the following equation:

$$\text{maximum canister residence time} = \frac{\text{number of canister racks at station}}{7 \text{ canisters processed/d}} \times 24 \text{ h} - 1 \text{ h of canister transfer time}$$

The subtraction of an hour transfer time results in a conservative estimate of the maximum residence time for each canister since in actual practice the transfer time should be shorter.

Once all of the operations in the ICGM process are well defined, it will be possible to establish a crane schedule to insure that there are no conflicts between steps in the canister transport sequence that require a crane and the other operations that require a crane, such as operating the canister/spray calciner connecting section or possibly placing the welding machine atop a canister. Detailed crane requirements are not available at this time since the equipment layout at each station will be a function of the facility layout. The crane must also be available for maintenance work. An auxiliary crane will provide some flexibility, but simultaneous operation of both cranes will be limited by the potential for interference. For example, cranes on separate rails cannot pass if the top crane is loaded.

The first station out of the furnace is the forced-air cooling rack. The canister is cooled to at least 750⁰F in the furnace before being lifted. Once in the forced-air cooling rack the cooling continues for 15 h, by which time the waste glass and canister have rejected the heat added due to glass formation and are essentially at a steady-state temperature determined by the internal heat generation and the forced-air cooling. The data on the forced-air cooling rack are given in Table A-13 in Section A.8.

While in the forced-air cooling rack, the canister has a twist-lock transport lid put in place. When the canister is moved to the closure-welding station, the lid is removed, the welding surface is ground and the twist-lock closure with the seal plate is installed and welded by the gas-tungsten arc (GTA) method of welding. The GTA welding method does not require the use of a filler metal. The surfaces must be clean and the fit-up must leave no gaps between the canister and the seal plate. Gas-tungsten arc welding has been used at PNL to weld actual radioactive canisters. A prototype of a compact GTA, remote welder has been developed at PNL.

The GTA welder, shown in Figure A-6, makes autogenous fillet welds that can range from 1/8 to 1/4 in. thick. Tight tolerances are maintained on the lid dimensions to maintain the appropriate gap between the welding tip and the lid. This eliminates the need for automatic tracking or voltage controls, which greatly simplifies the design. The welder is positioned from a hole in the center of the lid so the only fit-up requirement is to minimize the gap between the lid and the canister top. Cooling of the welding torch is done passively by the cover gas and a small, finned heat pipe. Very few parts on the welder require maintenance. However, those parts that do require routine maintenance are easily replaced. If some major damage to the welder occurs, it can be easily replaced. The total cost for the in-cell portion of the welder is quite low, making replacement a reasonable alternative.

Several special features were included on the welder to improve its remote operation. No organic parts were used except for one O-ring on a cable and the motor insulation. Both parts are easily replaceable. Only the torch rotates so the supply cables do not twist. The welding current supply and ground paths are part of the structure. The welding current passes through gold plating on the support arm to the torch, and the ground contact is made through the bottom of the welder to the lid. No additional connections are required.

FIGURE A-6. Photograph of the Prototype GTA Welder for Canister Closure

Operation of this welder is quite simple. The welder is removed from a storage location and is set on top of the canister lid. No special canister handling is necessary. The lid is then tack-welded in four locations, and the automatic welding sequence is started. The welding head rotates around the lid making the weld and then automatically slows to a stop. It is expected that with further development a simple procedure can be followed to make high-quality, reproducible welds. Figure A-7 is a schematic of a closure weld. Table A-14 in Section A.8 is the equipment data sheet for the canister closure-welding station.

After the closure welding has been performed, the canister is moved to a rack where leak detection is performed. Two leak-detection methods that can be used in a remotely operated, highly radioactive cell are helium detection and ultrasonic examination. For the canister to meet its design goals and to perform properly in subsequent operations, the closure weld must

FIGURE A-7. Schematic of a Canister Closure Weld

be leak tight. A "leak" is defined as any imperfection in the canister that allows gases, liquids or solids to enter or leave the canister. Because nothing is absolutely leak tight, the amount or size of a leak must be quantified. The definition of a leak will be dependent upon environmental conditions. For this report, a leak will be measured in atmospheric cubic centimeters per second ($\text{atm cm}^3/\text{s}$) at room temperature.

Criteria for allowable leakage from packages containing nuclear materials are contained in a number of documents, such as the Code of Federal Regulations and the American National Standards Institute's (ANSI) guide (ANSI 1974). However, waste canisters are not specifically mentioned. Leakage of materials from inside the canister is of greatest concern because the glass is radioactive. Canisters contain only waste glass and some nonradioactive gases, primarily helium. The ANSI (ANSI 1974) states that it is conservative to assume that solids, powders or slurries leak as water. Hence, a criteria of water tightness would seem to suffice. The major concern of material leaking from outside into the canister is water, since water is an important part of several processing steps and is the interim storage cooling and shielding medium. Studies indicate that a helium leak rate of $10^{-4} \text{ atm cm}^3/\text{s}$ would be a suitable value for water tightness. For this preconceptual design study, a target of $10^{-5} \text{ atm cm}^3/\text{s}$ is specified (see Section 3, Overall Design Bases). It should be possible to detect leaks within the range of 10^{-6} to $10^{-5} \text{ atm cm}^3/\text{s}$. Under ideal situations up to $10^{-12} \text{ atm cm}^3/\text{s}$ have been detected, but the piping, fillers, cold traps and test cells required for hot-cell operations will limit the detection range to the former range, which is well within the $10^{-4} \text{ atm cm}^3/\text{s}$ criteria for water tightness.

Figure A-8 is a conceptualized drawing of the helium leak-detection system. The helium source is placed in the canister just before lid-closure welding. It has not been determined at this time whether the source should be a small, pressurized cylinder with a timed release or a helium-saturated, sintered-metal block. Table A-15 is Section A.8 is the equipment data sheet for the leak-detection station.

The closure weld can also be inspected by ultrasonic techniques. There are currently no regulations requiring this, but it may prove necessary to assure leak tightness over the remainder of the canister service life. Ultrasonic inspection detects internal defects in the weld that could increase in size as a result of cyclic loads on the lid. Ultrasonic tests have been routinely used for inspection of in-service nuclear components.

The twist-lock closure was especially designed for ultrasonic inspection. Analyses have been conducted on the structural integrity of the closure weld to determine the defect size that ultrasonic inspection must detect. The results of these analyses are reported by Simonen, Griffiths and Thurgood (1978). In general, the analyses showed that a defect of half the thickness of the weld or larger must be detected. This is well within the range of most ultrasonic equipment. Figure A-9 is a schematic of the ultrasonic weld-inspection process.

Following leak detection, the canister is decontaminated. The purpose of this decontamination stage is to reduce the contaminant load in the storage pool. However, there are also strict limits for permissible contamination when shipping the canister to the repository or to an interim repository. Therefore, the canister will be decontaminated again, if necessary, before the canister is shipped offsite.

The decontamination station is envisioned as a rack capable of holding two canisters. Some of the options for decontamination include soaking in a chemical cleaning solution, cleaning by high-pressure water and steam sprays, and scrubbing with brushes in conjunction with spray cleaning and electro polishing. Electro polishing (Allen et al. 1978) is a process where

FIGURE A-8. Conceptualized Schematic of the Helium Leak-Detection System

FIGURE A-9. Schematic of the Ultrasonic Weld-Inspection Process

the canister serves as the anode and the ionic solution tank serves as the cathode in an electrolytic cell. The passage of current results in the anodic dissolution of the metal surface and, under proper operating conditions, the smoothing of the surface. The surface material is removed uniformly, and there is no preferential attack of grain boundaries or other microstructural features. Table A-16 in Section A.8 is the equipment data sheet for the decontamination station. Figure A-18 in Section A.8 is a conceptualized decontamination station employing steam spray and powered, mechanical scrubbers.

In the final step of the canister handling system, the decontaminated waste glass canisters are transported to the storage pool. The transport system proposed for this preconceptual design is a flooded tunnel leading to the storage pool. A crane would place the canister in a tracked-sled rack that transfers it to the storage pool, similar to the way spent fuel elements in a reactor are transported. The proposed transport system has some special design considerations. The process cell is lined with stainless steel to contain contamination and to facilitate decontamination flushing. The entrance to the transport canal must be protected from possible contamination during normal operation and during decontamination periods, since contamination could spread to the storage pool. In addition, the design must include provisions for replacing and maintaining the tunnel transport system since this type of mechanism requires relatively high maintenance.

A.6.5 OFF-GAS SYSTEM

The off-gas system for the ICGM process consists of two identical systems each of which serves a spray calciner. One of these systems is shown schematically in Figure A-19 in Section A.8 (Hanson and Kaser 1976). The off-gas system condenses the steam generated during calcination and filters out any particulates or volatile fission products that may pass through the sintered-metal filters on the spray calciner. The first stage in the system is a venturi scrubber. It condenses the bulk of the steam and scrubs many of the particulates from the off

gas. While the venturi scrubber does not have a large decontamination factor (DF) for particulates and volatiles, it can scrub large quantities of particulate matter from the off-gas stream without becoming clogged, as might be the case with a condenser or a packed-bed scrubber. Thus, the venturi scrubber serves as a backup in case there is a rupture of one of the sintered-metal filters. A knockout pot that collects any aerosols entrained in the off gas is also an integral part of the venturi scrubber.

The off gas, consisting primarily of noncondensable gas and residual particulates and volatile fission products, then passes to the tube side of the tube-and-shell condenser which effectively removes particulates and some volatiles. After the condenser stage the off gas passes to an NO_x recovery unit. This step reduces the NO_x emissions from the plant discharge stack and recovers HNO_3 . The NO_x absorption step is a bubble cap tower that uses oxygen to oxidize the NO to NO_2 . A demister is required downstream to recover entrained HNO_3 mist.

Following the NO_x absorber and associated condenser, the off gas is heated and passed through a ferric-oxide bed to absorb the volatile fission product ruthenium tetra-oxide. A HEPA filter is placed after the ruthenium absorber bed to guard against the entrainment of any particulates from the bed.

The last volatile product that must be removed is a small amount of iodine that is carried by the HLLW and is volatilized in the spray calciner. The off gas is dried by heating and then passes through a solid absorber bed such as silver zeolite. After the iodine adsorption step the off gas is passed through two HEPA filters to trap any loose adsorber-bed particles and to increase the DF. The off gas then proceeds to the air protection system of the fuel reprocessing plant.

Two condensate evaporators are incorporated into the off-gas system to recycle the radio-nuclides that are carried by the condensate back into the HLLW going to the spray calciner. These evaporators also recycle part of the nitric acid back to the fuel reprocessing plant.

At some point in the off-gas system, a vacuum-control valve is required. The vacuum in the spray calciner controls the valve and the valve adjusts for fluctuations in the air and liquid feed rate in the spray calciner.

While it is anticipated that the two off-gas systems can be interconnected, further research and development are required to identify the degree of redundancy required in the various components for reliable operation and for continual operation during maintenance of the off-gas system.

The flow rates of the gaseous constituents in the off gas are listed in Table A-7. As can be derived from the table, the percentages of air, steam and NO_x in the off gas are 27, 69 and 4, respectively. The steam, which is condensed in the venturi scrubber, the knockout pot and the first condenser, is drained to the condensate tank. The liquid in the condensate tank is concentrated and is re-injected with the HLLW into the spray calciner. The scrubbing solution used to recover more of the NO_x in the bubble cap tower goes to the second

evaporator. The condensate from the second evaporator is periodically pumped to the acid fractionator system in the fuel reprocessing plant.

The flow rates of the radionuclides in the off gas are given in Table A-8. A conservative set of DF values for each stage and for each type of radionuclide are given in Table A-9. Each type of decontamination stage and the expected DF ranges are discussed by Christian and Pence (1977).

The function and design of each stage in the off-gas system are given in the equipment data sheet (see Table A-17, Section A.8). Additional developmental efforts are required to design an off-gas system for a full-scale, radioactive process.

TABLE A-7. Off-Gas Flow Rates--Gases

Gaseous(a) Effluent	Flow Rate Entering Off-Gas System(b), L/min	Flow Rate Exiting Off-Gas Systems(d), L/min
Atomizing plus inleakage air(c)	1134	1134
NO _x	72	36
HNO ₃	118	Negligible
Steam	2924	Negligible
TOTAL	4248	1170

(a) From the spray calciner at 350°C, 0.97 atm.

(b) Flow rates are the combined flow rates of each off-gas system to reflect process totals. These are nominal operating flow rates; design flow rates are 1.5 times these.

(c) 934 L/min atomizing plus 200 L/min air inleakage assumed.

(d) At standard temperature and pressure.

TABLE A-8. Off-Gas Flow Rates--Radionuclides

Radionuclide Effluents	Flow Rate Entering Off-Gas System(a)		Flow Rate Exiting Off-Gas System	
	g/d	Ci/d	g/d	Ci/d
Particulates(b)	312	2.1E3	6.2E-13	4.2E-12
Cs(c)	12	6.6E2	2.4E-14	1.3E-12
Ru	144	1.2E3	1.4E-10	1.2E-9
I(d)	36	5.0E-3	2.8E-4	3.9E-8

(a) Total for both off-gas systems; nominal operating rates; design rate is 1.5 times these values.

(b) Includes all fission products, actinides, activation products and nonradioactive metal additions (except Cs, Ru and I contributions have been subtracted).

(c) Assumed volatility of 0.07%.

(d) 2% of I in fuel is assumed to be present in HLLW; 100% of this amount enters the off-gas system.

TABLE A-9. Decontamination Factors Expected in the Off-Gas System^(a)

Stage of System	Particulates ^(b)	Ruthenium	Iodide
Spray calciner with sintered-metal filters	1,000	100	1
Venturi scrubber with knockout box	50	10	1
Condenser	1,000	1,000	1
NO _x scrubber	10	10	1
Demister	100	1	1
Ruthenium adsorber	1	1,000	1
HEPA filter (1 stage)	100	10	1
Iodine adsorber	1	10	1,000
HEPA filter ^(c) (3 stages)	<u>100,000</u>	<u>100</u>	<u>4</u>
TOTAL	5.0×10^{17}	1×10^{14}	4,000

(a) $DF = \frac{\text{activity into component}}{\text{activity out of component}}$.

(b) Includes condensed particulates of volatiles such as Cs and Te.

(c) Includes one roughing stage with DF = 10 and 2 HEPA filters with DF = 100 each.

A.7 ALTERNATIVE PROCESSES AND EQUIPMENT

Alternative methods of producing a calcine form of HLLW are the rotary-kiln calciner used in France and the fluidized-bed calciner used in the United States. The rotary-kiln calciner was developed as part of the French Atelier Vitrification Marcoule (AVM) facility for the conversion of HLLW to a phosphate-glass waste form. The rotary-kiln calciner consists of an inductively heated, metal cylinder that is slightly inclined. HLLW is slowly added to the raised end of the cylinder which continuously rotates around its axis. A suspended bar that extends the length of the cylinder helps to remove dried calcine from the wall of the cylinder. The dried calcine exits through the lower end of the cylinder. The calcine feed system in this process is attractive in that no pumps are required. All solutions are moved by the air-lifting method.

The rotary-kiln calciner has undergone extensive radioactive testing and is now considered a fully developed process. The process will be used in the HLLW waste processing plant under construction at Marcoule, France, and may also be used in a similar capacity in Germany. This process has the advantages of small size and significantly reduced off-gas handling requirements. Disadvantages include the possibility of high seal maintenance and the uncertainties in scaling up this equipment to meet full-scale production plant requirements.

The fluidized-bed calciner has been developed and operated on a production basis with HLLW at Idaho National Engineering Laboratory. In this process HLLW, kerosene and oxygen are sprayed into a fluidized bed of calcine. The calcine is fluidized by the addition of heated air through a perforated plate at the bottom of the calciner. The combustion of the kerosene supplies heat which dries and denitrates the HLLW, yielding more calcine. The calcine is either entrained in the off-gas stream where it is later separated, or if sufficiently heavy, remains behind as part of the fluidized bed of calcine. This process has the advantage of small size. Disadvantages include the use of potentially explosive material, the formation of clinkers in the bed, and greater process complexity.

An approach under development in Great Britain that eliminates the need for a calciner is the rising-level glass process, or the FINGAL Process. In this process, HLLW and glass-forming additives are added directly to a heated canister. Evaporation, denitration and melting all take place in the canister. The biggest advantage of the process is its simplicity. Disadvantages include product quality uncertainties, long processing times which greatly increases equipment requirements, and serious off-gas problems.

Other glass melting approaches are a joule-heated melter that is fed a mixture of calcine and glass frit, a joule-heated melter that is fed a mixture of HLLW and glass powder, and the AVM facility melting processes.

A.8 EQUIPMENT DATA SHEETS AND DRAWINGS

A glossary of symbols and abbreviations used in the figures is given below:

rack or panel mounted		needle valve
locally or in-line mounted		reverse flow check valve
two measured variables		restricting flow orifice
flow element		jet mixer
motor		process service lines
diaphragm-operated valve		pneumatic instrument lines
solenoid-operated valve		primary process lines
ball valve		electronic wires or hidden lines

A&OA	access & operating area	FR	flow recorder
AM	ammeter	FRP	fuel reprocessing plant
AR	argon	FT	flow transmitter
AS	air supply	FY	converter
ATM	atmosphere	HLA	high-level alarm
CFM	cubic feet per minute	Δ HWA	high weight rate change alarm
CI	circuit interrupter	IC	ionization changer
CM	chemical make-up	IS	current switch
CSTR	canister	IY	relay
CR MAINT	crane maintenance	JR	electrical power recorder
CS	cooling supply	JS	electrical power switch
CW	cooling water	KJC	timer
DOV	diaphragm-operated valve	KWH	kilowatt hours
IAS	instrument air supply	LC	load cell
DPA	differential-pressure alarm	LI	level indicator
DPI	differential-pressure indicator	LLA	liquid-level alarm
DPR	differential-pressure recorder	LLI	liquid-level indicator
DPT	differential-pressure transmitter	LLR	liquid-level recorder
DR	density or specific gravity recorder	Δ LWA	liquid weight rate change alarm
EL	elevation	M	motor
E/M	electrical mechanical	OA	operating area
ETM	elapsed time meter	OG	operating gallery
FA	flow alarm	OGS	off-gas system
FC	flow controller	OR	oxygen recorder
FE	flow sensing element	P	pressure
FI	flow indicator		

PCV	pressure control valve	TE	temperature element
PD	positive-displacement blower	TGA	tungsten gas arc
PE	pressure sensing element	TI	temperature indicator
PGM	programmer	TR	temperature recorder
PI	pressure indicator	TRC	temperature recorder controller
PR	pressure recorder	TT	temperature transmitter
WR	weight recorder	TYP	typical
Δ PR	change in pressure recorder	VC	valve controller
PS	pressure switch	VIB	vibrator
RC	ratio controller	VM	volt meter
REF	reference pressure header	VSD	variable-speed drive
RPM	revolutions per minute	VSM	variable-speed motor
RR	radiation recorder	W<	welding & leak test
RW	raw water (unsanitary)	Δ WA	weight rate change alarm
S	solenoid	WC	weight controller
SE	sensing element	Δ WC	weight rate change controller
SG	sample or service gallery	Δ WI	weight rate change indicator
SPC	supercalcine	$\Sigma\Delta$ WI	weight total and rate change indicator
SPL	sample	Δ WR	weight rate change recorder
SS	steam supply	WS	weight supply
S/S	stainless steel	XFR	transfer room
SUM	summer		
SW	selector switch		
TA	temperature alarm		
TC	temperature controller		

TABLE A-10. Equipment Data Sheet for HLLW Feed System

Function

Receive, store, analyze, mix and feed HLLW to spray calciner.

Process Interfaces

HLLW supply from fuel reprocessing plant; spray calciner system.

Design Parameters

304 L stainless steel; storage tanks (2) = 2.0 m dia x 4.5 m high; feed tank = 1.5 m dia x 1.6 m high; inter-tank transfer system with capability of transferring HLLW to and from all tanks.

Operating Conditions

Temperature = 40°C; pressure = 30 in. H₂O vacuum to ambient; feed tank capacity = 600 L; inter-tank transfer capacity = 2000 L/h.

Features

Storage tanks: closed-circuit coil to remove 30 kW @ 40°C; cooling jacket to remove 30 kW @ 95°C; mechanical agitator and air sparger ring.

Feed Tank: closed-circuit coil to remove 20 kW @ 40°C; cooling jacket to remove 20 kW @ 95°C; mechanical agitation and recirculation transfer eductors.

Services or Auxiliaries

Electrical power; compressed air; cooling water; steam; service water.

Space Requirements

Storage tanks are each 3 m by 3 m by 10 m high; feed tank is 2.0 m by 2.0 m by 4 m high

Maintenance Methods

Remote cranes and hoists

Remarks

FIGURE A-10. High-Level Liquid Waste Feed System to Spray Calciner

FIGURE A-11. Schematic of the Spray Calcination System

FIGURE A-12. Cross-Sectional View of HLLW Atomizing Nozzle

FIGURE A-13. Schematic of Instrument System for Spray Calciner System

TABLE A-11. Equipment Data Sheet for the Spray Calciner

Function

Atomize, dry and calcine HLLW.

Process Interfaces

HLLW feed system; off-gas system; ICM system.

Design Parameters and Features

Calciner chamber: type--right cylinder; dimensions = 73 cm in dia by 244 cm high (cylindrical portion only); heat transfer area = 5.6 m²; material = nickel-chromium-iron alloy; wall thickness = 0.64 cm; bottom cone has 60° (from horizontal plane) sloped sides.

Furnace: type--480-V, 3-phase, electrical resistance; number of annular zones = 4; control--silicon-controlled rectifier; maximum power = 140 kW at 35 kW/zone; normal operating temperature = 800°C; maximum operating temperature = 1000°C.

Atomizing nozzle: type--pneumatic, internal mix, such as Model 72 by Spray Systems Co.; nozzle tip--304 stainless steel with a 96% dense alumina insert; atomizing air--facility compressed air; air-to-liquid ratio = 300 to 1 volumetric; liquid control--magnetic flowmeter feedback to valve in HLLW recirculation line.

Filters: type--sintered-powdered-metal cylinders; total filter area = 7.5 m²; mean pore size = 65 μ; material is 316 stainless steel; number of filter cylinders = 34; cylinder dimensions are 7 cm in OD, 91 cm long and 0.16 cm thick.

Filter blowback system: type--venturi; blowback air pressure = 4.1 to 6.8 atm gauge; blowback nozzle size = 0.38 to 0.46 cm; percent of filters cleaned simultaneously = 20; blowback cycle = 8 min; blowback duration = 0.25 s.

Vibrators: type--pneumatic dry lubricant, such as Model 1300 AC Vibramight by Cleveland Vibratory Co. or equivalent; motive air pressure = 2.7 atm gauge; location--two 180° apart on side of calciner chamber, and one on cone directly beneath nozzle.

Glass frit feed system (2): components are frit bin, proportional feeder and air-lock valves; feed rate = 0 to 30 kg/h; nominal rate = 17 kg/h; bin size = 612 kg of frit (255 L/day); proportional feeder--potential candidate is a gravimetric unit containing continuous weight belt; air locks--air locks must prevent significant air inleakage and prevent migration of contamination out of cell.

Operating Conditions

Capacity = 79 L/h; chamber temperature = 700°C; filter temperature = 350°C; pressure = 0.97 atm abs.

Services or Auxiliaries

Electrical = 480-V, 3-phase and 120-V, 1-phase; compressed air = 80 psi; flushing water = 50 psi.

Space Requirements

3.5 m by 3.5 m by 4 m high

Maintenance Methods

Remote except for proportional frit feeder

Remarks

FIGURE A-14. Continuous Glass Frit Feeder

FIGURE A-15. General Arrangement of ICM System

TABLE A-12. Equipment Data Sheet for the ICM System

Function

Vitrify calcined HLLW as a glass within a metal storage canister.

Process Interfaces

Spray calciner system; canister transport system.

Design Parameters

Capacity = 0 to 75 kg glass/h; temperature = 0° to 1200°C; pressure = 0.75 to 1.5 atm abs.

Operating Conditions

Capacity = 25 kg glass/h; temperature = 1050°C; pressure = 0.97 atm abs.

Features

Retort: dimensions are 38 cm in ID by 196 cm long by 0.64 cm thick; material--nickel-chromium-iron alloy.

Furnace: type--resistance heating with SiC glow bars, 480-V, ac, 3-phase; number of heated zones = 5, each approximately 40 cm long; size of heated chamber is 70 cm x 70 cm by 200 cm high; overall size is 170 cm x 170 cm by 220 cm high; power required = 30 kW/zone or 150 kW total; cooling required = 35 kW; maximum operating temperature = 1200°C; type of control is cross-over, triggered silicon-controlled rectifiers.

Calciner-To-Canister connection: material--stainless steel or nickel-chromium-iron alloy where combination of high temperatures and high strain loads are encountered; cooling required at cone valve and connection piping immediately above canister; maximum temperature allowed = 400°C.

Glass Level Detectors: types--load cell records retort and canister weight; gamma radiation detector located outside of cell views canister through collimators cast into cell wall.

Services or Auxiliaries

Electrical power; compressed air; electromechanical manipulator; cooling water.

Space Requirements

Each furnace will require a 2-m-wide by 5-m-deep by 4-m-high space for access and movement to loading position.

Maintenance Methods

Remote

Remarks

Each in-can melting furnace will be mounted on tracks and it will be moved out approximately 2 m to allow for canister changeout. Therefore, all power, cooling air and instruments must be on flexible leads.

FIGURE A-16. Schematic of the Spray Calciner to ICM Connecting Section

FIGURE A-17. Preliminary Instrumentation Requirements for ICM System

TABLE A-13. Equipment Data Sheet for the Forced-Air Cooling Rack

Function

To provide controlled cooling for canister filled with waste glass.

Process Interfaces

Part of the canister transport, closure and decontamination sequence.

Design Parameters

Number of cooling stations = 6; type of cooling is 1000 L/min forced air per station (6000 L/min total); type of structure is rack with baffled shrouds as cooling units; material--stainless steel 304; instrumentation includes 1) rotameters on air supply and 2) thermocouples monitoring air exit temperature.

Operating Conditions

Temperature = 0° to 800°C.

Services or Auxiliaries

Compressed air.

Space Requirements

2 m by 3 m horizontal by 4 m high

Maintenance Methods

Remote

Remarks

TABLE A-14. Equipment Data Sheet for the Canister Closure Station

Function

To contain and cool the canister during weld preparation and lid closure.

Process Interfaces

Part of the canister transport, closure and decontamination sequence.

Design Parameters and Features

Type is 2-station cooling rack with 200 L/min forced-air cooling; material--stainless steel 304.

- An air motor powered grinding jig, to be designed, will be used for weld preparation.
- A GTA welding unit (uses no filler material) will be placed on the canister lid and an electric motor will drive the weld electrode around the lid-to-canister seal. This unit has been demonstrated.

Operating Conditions

Temperature = 0° to 800°C.

Services or Auxiliaries

Electrical; compressed air; television; controlled lighting; electromechanical manipulator; and scrap vacuum collector.

Space Requirements

2 m by 2 m horizontal by 4.0 m high

Maintenance Methods

The rack will be maintained remotely. The weld preparation jig and the GTA welding unit will be decontaminated and repaired in the hot shop.

Remarks

TABLE A-15. Equipment Data Sheet for the Canister Leak-Detection Station

Function

To demonstrate the integrity of the canister lid-closure weld by a helium leak check.

Process Interfaces

Part of the canister transport, closure and decontamination sequence.

Design Parameters and Features

Cooling rack: type--an open structure will support two stations, each consisting of a baffled shroud with a 200 L/min cooling air flow; material--stainless steel.

Vacuum hood: A vacuum hood connected to an out-of-cell mass spectrometer will be placed over the closure lid and sealed on the canister shoulder. It will be evacuated from out of the cell for the helium leak-detection process.

Mass spectrometer: commercial equipment.

Operating Conditions

Temperature = 0° to 300°C.

Services or Auxiliaries

Compressed air; electro-mechanical manipulator.

Space Requirements

2 m by 2 m by 4 m high

Maintenance Methods

Rack maintenance is remote; mass spectrometer is contact maintenance.

Remarks

It may be possible to combine this station with the lid-closure-weld station.

TABLE A-16. Equipment Data Sheet for the Canister Decontamination Station

Function

To provide rough decontamination of the canister prior to transferring it to the onsite, interim storage pool.

Process Interfaces

Part of the canister transport, closure and decontamination sequence.

Design Parameters and Features

Method--rotary steel brushes scrub scale and decontamination from canister; type--an open rack structure containing two rectangular tanks, where power scrubbing with decontamination solution is carried out and a storage tank where decontamination solution is filtered and cooled; material--stainless steel 304; size of decontamination tanks = 50 cm x 50 cm x 2 m high; storage tank cooling--closed-cycle, in-tank cooling coil of 6 kW capacity; brush motors are 2 hp air operated; pump--air-operated diaphragm with 300 L/h capacity; filters--disposable screen plus resin exchange; storage tank dimensions = 1 m in dia by 1.5 m high.

Operating Conditions

Temperature = 0^o to 90^oC.

Services or Auxiliaries

Compressed air and closed-cycle cooling water.

Space Requirements

2 m by 4 m horizontal x 5 m high

Maintenance Methods

Remote

Remarks

FIGURE A-18. Conceptual Design of a Steam and Mechanical Scrubber Decontamination Station

FIGURE A-19. General Layout Diagram of the Off-Gas System

TABLE A-17. Equipment Data Sheet for the Off-Gas System

Function

Condense steam and remove radionuclides, nitric acid and NO_x from the off gas.

Process Interfaces

Spray calciner; reprocessing plant air protection system (The two off-gas systems are capable of being interconnected.).

Design Parameters and Features

Venturi Scrubber: spray nozzle operating feed rate = 90 L/min and design rate = 135 L/min; calciner operating off-gas flow rate = 4814 L/min and design rate = 7220 L/min; H_2O + HNO_3 condensate operating rate = 80 L/h and design rate = 120 L/h; operating heat load = 68 kW and design = 102 kW; pressure = 0; gas/liquid separator--integral knockout pot; material--stainless steel 304L (possibly titanium for corrosion resistance).

Condenser: type is shell-tube, single pass, baffled floating head with gas on tube side; material--stainless steel 304L; gas flow = 0 to 8000 L/min (overrated to compensate for loss of venturi efficiency); heat load = 30 kW.

NO_x Absorber: type is bubble cap tower; material--stainless steel 304L; gas flow = 600 L/min operating and 900 L/min design; oxygen flow = 44 L/min operating and 66 L/min design.

Demister: type--baffles, bubble caps and metal mesh in enclosure; gas flow = 570 L/min operating and 850 L/min design; material--stainless steel 304L; pressure drop = 2 in. H_2O operating and 4 in. H_2O design.

Ruthenium Adsorber: type--ferric oxide particle bed (18 to 25 mesh); preheater = 6 kW operating and 8 kW design; gas temperature = 350°C ; bed size = 160 L for 3 months operation; bed capacity = 19.4 kg Ru; gas flow = 1200 L/min operating and 1800 L/min design.

Iodine Adsorber: type--silver zeolite particle bed (12-14 mesh); gas flow = 920 L/min operating and 1380 L/min design; temperature = 0° to 200°C ; material--stainless steel 304L; bed size = 20 L for 3 months operation; bed capacity = 1.6 kg I.

HEPA filter: type--case containing 1 roughing filter plus 2 HEPA filters; capacity = 820 L/min operating and 1230 L/min design; temperature = 0° to 150°C ; dimensions are 35 cm by 35 cm by 15 cm for each filter and overall assembly is 1.5 m.

Operating Conditions (at exit from spray calciner)

HNO_3 flow = $8.1\text{E}3$ L/h; H_2O flow = $2.0\text{E}5$ L/h; air flow = $7.8\text{E}4$ L/h; NO_x flow = $4.9\text{E}3$ L/h; temperature = 350°C .

Services or Auxiliaries

Space Requirements

Maintenance Methods

Remarks

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APPENDIX B

JOULE-HEATED GLASS MELTING PROCESS

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APPENDIX B

JOULE-HEATED GLASS MELTING PROCESS

B.1 INTRODUCTION

The JHGM process produces a monolithic, glass waste form similar to that produced by the ICGM process. The JHGM process facility will have the capability to 1) dry the HLLW to form a calcine powder, 2) incorporate glass frit, 3) melt the calcine and frit mixture, 4) fill canisters with the molten glass, 5) cool them, 6) seal-weld the canisters, 7) check canister integrity, and 8) decontaminate canisters and transport them to an interim storage area. The JHGM process permits continuous glass melting, higher temperatures for better glass product formation and lower canister temperatures as compared to the ICGM process.

B.2 PROCESS DESCRIPTION

The JHGM process flow diagram is shown in Figure B-1. The flow diagram is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. The process material mass balances given in the reference ICGM process (see Figure A-2) are applicable to the JHGM process, since borosilicate-glass monoliths are the waste products of both processes.

As covered in the ICGM process, HLLW (its composition is specified in Table A-2 and in the Overall Design Bases Section) is received from the fuel reprocessing plant and is temporarily stored in the HLLW feed system. Here it can be analyzed and any necessary treatment done before transferring it to the spray calciner.

The JHGM process requires only one spray calciner, which is similar to those used in the reference process but is larger and has a greater capacity. In the JHGM process, the calcined waste is mixed with glass frit in the lower section of the spray calciner and the calcine/frit mixture falls by gravity into the joule-heated melter, where it forms a cold cap of material on top of a reservoir of molten waste glass. The spray calciner operates continuously since the molten glass can rise in the melter while canisters are being changed.

The molten glass is drawn up from the bottom of the melter into a heated discharge trough within the melter. The molten glass is poured into canisters similar to those used in the ICGM process. Since the glass has already been formed in the melter, canister filling can be carried out much more rapidly than in the ICGM process and the JHGM canisters are not exposed to temperatures as high as those in the ICGM process. The system to handle and process the JHGM canisters is essentially the same as for the reference ICGM process. The off-gas system for the JHGM process is also basically the same as that for the ICGM process, except that it has a larger capacity. It may be either a single or dual system depending on operating efficiencies.

FIGURE B-1. Joule-Heated Glass Melting Process Flow Diagram

B.3 BASES AND ASSUMPTIONS

Most of the bases and assumptions for the JHGM process are covered in Section 3, Overall Process Bases and in Appendix A. Other process criteria applicable to the JHGM process are included in this section.

B.3.1 PROCESS BASES

As noted above, the JHGM process solidifies the HLLW produced in a fuel reprocessing plant operating at a rate of 2000 MTHM/yr. Design capabilities are 1.5 times the nominal operating values. With a waste loading of 30 wt% and a glass density of 3.0 kg/L, the process is designed to solidify 5700 L of HLLW/d into 1750 kg of glass containing 33.7 W/L of heat. This requires a design glass-melting rate of 73 kg/h and a nominal melting rate of 49 kg/h. At the design rate a canister is filled every 3 h and 45 min.

B.3.2 CANISTER

The canisters for the JHGM process are very similar to those described in Appendix A for the ICGM process. The main difference between the ICGM and JHGM processes is that in the JHGM process the melting is not carried out in the canister; therefore, the operating temperature is lower and the material requirements for the canister are reduced. As discussed in the Overall Process Bases Section, the canister loading is restricted to 3.06 kW for 5-yr-old waste products. Therefore, the canister size must be 30.5 cm in dia by 185 cm long (12 in. in dia by 6.1 ft long). It is assumed that the canister will be made from a 0.64-cm-thick stainless steel alloy. Internal fins are not required in the JHGM canister as they are in the ICGM canister.

B.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTES

Table B-1 contains an estimation of the effluents, waste and scrap generated in the JHGM process. These estimates are similar to, or slightly more than, the estimates given in Appendix A (see Table A-3) for the ICGM process. One major difference is the limited lifetime of the joule-heated melter. It is assumed that after a two-year lifetime, the entire melter-box refractory lining and residual waste glass will have to be cut up, compacted and disposed of. While other types of furnaces may also require periodic replacement, the unique problem with the joule-heated melter is the fairly significant amount of residual radioactivity due to the glass trapped on the refractory lining. It is assumed that the fuel reprocessing plant will have provisions for treating and packaging all types of wastes. It is also assumed that all of the process scrap containing HLLW will be recycled and incorporated into the JHGM process.

TABLE B-1. Secondary Radioactive and Nonradioactive Wastes
That Will Require Processing

<u>Secondary Wastes</u>	<u>Estimate of Volume of Waste per Unit Time</u>
Nonradioactive	
Liquids	
Cooling water	700 L/min
Process water	50 L/min
Sanitary waste	10 L/min
Gases	
Ventilation flow	500 scmm
NO _x	<10 L/min
Radioactive	
Off-gas system and ventilation flows	<5 Ci/yr
Solids	
Trash (paper, plastics, cloth, and glass)	200 m ³ /yr (<20 Ci/yr)
Failed equipment	70 m ³ /yr partially compacted (10 ⁵ Ci/yr if well decontaminated)
Ruthenium bed material	1600 kg/yr (3.6 x 10 ⁵ Ci/yr)
Iodine bed material	60 kg/yr (1.5 Ci/yr)
HEPA filters	200 kg/yr (1 Ci/yr)

B.3.4 UTILITIES AND MATERIALS

It is assumed that the utilities and materials required for the JHGM process are the same as those listed in Appendix A for the ICGM process (see Tables A-4 and A-5).

B.4 FACILITY AND EQUIPMENT REQUIREMENTS

A layout of the JHGM process equipment is shown in Figure B-2. The process cells are contained in a reinforced concrete structure that provides ample open space and areas for equipment access and replacement. All of the ancillary areas of the JHGM process are identical to those of the ICGM process. The overall requirements for the waste facility and the support groups and services are covered in Section 3, Overall Process Bases. The limitations and demands imposed by the requirements of remote operation and remote maintenance of all in-cell equipment are of prime importance.

The major processing equipment in the process cell and the approximate volume required for each piece of equipment are listed in Table B-2. To determine volume requirements a piece of equipment was represented as a rectangular parallelepiped that has one-half meter on all sides to allow for protubences and service connections. For the canister treatment racks, the vertical height was extended by an amount equivalent to the canister overall length. Other space was allowed for operations, maintenance, equipment replacement and decontamination.

FIGURE B-2. Joule-Heated Glass Melting Process Cell and Equipment Arrangement

TABLE B-2. Process Cell Volume Requirements
for Major Processing Equipment

<u>Equipment</u>	<u>Dimensions (width x depth x height), m</u>	<u>Volume, m³</u>
HLLW System		
Batch tank A	2.5 x 2.5 x 4	25
Batch tank B	2.5 x 2.5 x 4	25
Feed tank	1.5 x 1.5 x 3	7
Piping manifold	2.0 x 3.0 x 3	18
Sample analysis Equipment	2.0 x 3.0 x 3	18
Spray calciner	3.5 x 3.0 x 5	58
Joule-heated glass melter	2.5 x 3.0 x 5	38
Canister processing system		
Forced-air cooling rack	2.0 x 3.0 x 4	24
Lid-closure station	2.0 x 2.0 x 4	16
Helium leak-detection station	2.0 x 2.0 x 4	16
Decontamination station	2.0 x 4.0 x 5	40
Unfilled canister rack	2.0 x 3.0 x 4	24
Storage pool canal	2.0 x 2.0 x 3	<u>12</u>
TOTAL		321

B.5 STATE OF PROCESS DEVELOPMENT

The JHGM process requires the use of chemical and mechanical operations that will be performed remotely by equipment that will require remote maintenance and remote removal in a production facility. In addition, some of the required process steps have not been performed on a full-sized production scale.

As noted in Appendix A, considerable research, development and tests have been performed at PNL on spray calcination, and its state of development is well advanced. As a result spray calcination equipment can easily be applied to the JHGM process.

Joule-heated melters have been widely applied to the industrial melting of glass. In addition, nuclear-scale, joule-heated melters have been installed and operated in various research and development programs at PNL. Approximately 46,000 kg of glass have been produced and poured from melters at PNL and SRL. However, significant concerns regarding foaming of the molten glass, reboiling of the glass, operating the melter remotely, maintaining the multi-electrodes, sludge buildups on the bottom of the melter, pouring the glass remotely, and melter lifetime need to be resolved.

The state of development of the off-gas system, the canisters and the processing of canisters is covered in Appendix A.

B.6 UNIT OPERATIONS

Some of the JHGM process unit operations are the same as the ICGM process unit operations. Information on the function and operation of processing equipment, and areas requiring additional research and development are presented below. Drawings and data sheets on systems and equipment discussed in this section can be found at the back of Appendix B in Section B.8.

B.6.1 HLLW FEED SYSTEM

The requirements for and the operation and function of the HLLW feed system for the JHGM process are identical to those described in detail in Appendix A for the ICGM process. The concerns and the required research and development identified for the reference ICGM process are also applicable to the JHGM process.

B.6.2 SPRAY CALCINER SYSTEM

The function, construction and operation of the spray calcination system for the JHGM process is the same as that described in Appendix A for the ICGM process. However, only one spray calciner is required, and its specifications are given in Table B-3 (see Section B.8). The research and development efforts required for the ICGM process spray calciners are also applicable to the JHGM process.

B.6.3 JOULE-HEATED GLASS MELTER

The joule-heated melter is the central component of this glass-producing process. This type of melter is commonly used in the production of commercial glassware. It is based on the principle that molten glass is a conducting solution because of its ionic composition. Therefore, it is possible to pass an alternating current through electrodes at either end of a refractory tank to directly generate heat within the glass solution. The incoming mixture of calcine and glass frit falls into a mound of solids, called a cold cap, that floats on top of the molten glass. The melting of this cold cap releases gas, which is drawn back up through the spray calciner and the sintered-metal filters into the off-gas system.

Experience has shown that the best way to discharge the molten glass from the ceramic melter is by drawing glass from near the bottom of the tank into an uprising discharge chute. The glass can be drawn off by tilting the whole melter or by feeding air through a platinum tube into the riser section, forming an air lift. The air lift works on the principle that air bubbles in the molten glass cause the bulk glass in the riser section to have a lower density than the glass in the main tank. The difference in pressure causes an outpouring of molten glass into the discharge chute. Some of the advantages of taking molten glass from the bottom of the tank are 1) there is less chance of pouring some of the unmelted solid materials that float on the top of the molten glass, 2) glass that is poured has had more time to come to equilibrium composition, and 3) the tendency of any of the heavier glass phases to settle on the bottom is counteracted by continually drawing the molten glass from the bottom of the melter.

The joule-heated melter also includes the following subsystems:

- spray calciner to melter connection
- melter electrical system
- melter shell and refractory inserts
- canister filling system
- canister weight and glass level indication
- melter instrument system.

The general arrangement of a joule-heated melter is shown in Figure B-3 in Section B.8. Additional data on the equipment is given in Table B-4 (also Section B.8). Data on the operation of the melter and areas needing additional research and development are given below.

B.6.3.1 Spray Calciner to Melter Connection

The connecting section between the spray calciner and the joule-heated melter is a piping section with provisions for thermal expansion and, possibly, some cooling. The cooling may be required to prevent the frit and calcine mixture from melting and sticking to the connecting section near the melter entrance and eventually forming a plug. Some research and development has been carried out to develop a differential-pressure monitor to monitor the pressure drop of the off gas being drawn up through the connecting section as a means of measuring calcine and frit buildup. However, variations in inleakage have made this means of detecting blockage unsuccessful.

B.6.3.2 Melter Electrical System

The electrical system includes two main electrodes (as shown in the glass tank in Figure B-3, a plasma torch system used for start-up, and resistance heaters in the glass discharge section. The start-up torch protrudes into the air space of the melter lid and is only used to melt the initial glass-forming charge in the melter tank. Once the glass is molten and becomes ionic, the alternating current from the main electrodes takes over. The start-up system is removed prior to operation.

The main electrodes are made from a nickel-chromium-iron alloy (for example Inconel 690) and are air-cooled to minimize corrosion caused by the glass. Additional research and development is needed to determine the optimum tradeoff between the electrode corrosion rate and the disadvantage of electrode cooling. Excessive electrode cooling is self-defeating because more power must be added to the glass tank, which raises the current density of the electrodes and increases the electrode temperature and the corrosion rate. The electrical, air and thermocouple connections to the electrodes are made through Conax[®] connectors to electrically insulate the electrodes from the melter tank and service connections. The connections are also packed with a moldable refractory to minimize air inleakage through the melter shell.

There are several other areas in which additional research and development are required. To date, an automatic power control system based on temperature, as measured at the electrode,

[®] Trademark of Conax Corporation.

has not proven satisfactory. Some success has been achieved by using the glass resistance as a control parameter. However, one difficulty is that glass resistance is also very dependent on glass composition. If thermocouples permanently installed in the bottom of the glass tank could be proven reliable for the life of the melter, it might be possible to control power input directly from the glass temperature. Any control system must maintain the electrode temperature below an as yet undetermined temperature to avoid excessive corrosion.

B.6.3.3 Melter Shell and Refractory Inserts

The melter shell is a stainless steel box with bottom and side cooling jackets. Its purpose is to provide structure for the internal refractory system, serve as a cool outer boundary, contain minimum air leakage, and freeze and stop any glass leakage through the refractories. The top of the melter must be insulated, stainless steel panels bolted on to permit the construction of the refractory glass tank. The melter shell also holds the majority of the instruments, cooling and cabling supports. Finally, it contains the necessary fittings and thermowells for instruments that measure temperatures.

There are two types of refractory inserts. One type is the contact refractory brick that contains the glass; it must have a very high temperature rating and must be highly resistant to corrosion from the glass. The K-3 Monofrax[®] has been used successfully as the glass contact refractory in developmental melters. However, this refractory brick has a relatively high thermal conductivity. The K-3 Monofrax has also been used in the discharge chute riser section and for the discharge trough. The second type of refractory is a less corrosion resistant but more insulative refractory, such as Alumafrax[®] castable, which is used in the outer layers of the refractory system. The two types of refractories are cut to form an overlapping structure wherever possible. A compressible material, such as Fiberfrax[®], is used between the outer refractory and the melter box to absorb the refractory growth as the melter is heated.

B.6.3.4 Canister Filling System

The melter tank is designed with a sloping bottom that leads to a low point at the discharge chute. This design permits the molten glass and sludges to be air-lifted from the lowest point of the sloped bottom. The melter design shown in Figure B-3 incorporates two discharge sumps. Each sump is near one of the main electrodes, and each sump itself contains an electrode to facilitate discharging sludge accumulations. Figure B-4 in section B.8 illustrates the design of the air-lift discharge system. An Inconel or platinum tube positioned in the glass discharge riser section injects a low volume of air bubbles into the molten glass. The lower density air and glass mixture is then forced up to the level of the discharge trough where it overflows to the discharge face and drops into the canister. Glass flow is stopped by cutting off the flow of air into the air-lift system. The discharge trough and discharge face are heated by electrical-resistance heaters to maintain these areas at the desired glass-pouring temperature. In addition to the normal discharge requirement, the melter must be drained at the end of its life or in the event that a malfunction in the electrodes or power

[®]Trademark of Carborundum, Inc. (K-3 Monofrax, Alumafrax, Fiberfrax).

supplies cannot be repaired in time (approximately 6 to 12 h) to prevent the tank of glass from solidifying. The air-lift system may be capable of draining the tank, but this capability has not been demonstrated to date. Therefore, an air-cooled freeze valve is incorporated in the bottom of the melter. The freeze valve contains a resistance heater to facilitate melting the glass plug. An emergency dump canister is located beneath the freeze valve.

B.6.3.5 Canister Weight and Glass Level Indication

As a canister is filled with molten glass, the net weight and level of the glass are constantly monitored. Load cells can be used to indicate the weight, and the level can be determined using a gamma detection device similar to that used in the ICGM process. The glass weight and level indications provide the means of controlling the amount of glass in canisters and may also be used to infer glass quality.

B.6.3.6 Melter Instrument System

The degree to which the joule-heated melter should be instrumented is still an area requiring research and development. The developmental melters have been highly instrumented, typically having 30 thermocouples, 12 cooling fluid flowmeters, glass tank specific gravity indicators and plenum pressure indicators. In addition, there are several view ports to observe the glass tank and discharge area. The advantages of having an extensive instrument system must be balanced against the maintenance requirements and maintenance access restrictions created by the instruments. The instruments proposed for the melter system are listed in Table B-4 (Section B-8). The list reflects a conservative approach in that it calls for essentially the same number of instruments as required for the developmental melters.

B.6.4 CANISTER CLOSURE, DECONTAMINATION AND TRANSPORT SYSTEM

The connecting section for the JHGM process may require a different canister opening flange than the flange proposed for the ICGM canister described in Appendix A. However, the closure weld testing and performance criteria are the same for both processes; thus, the canister closure, decontamination and transport system is essentially the same as that described in Appendix A for the ICGM process.

B.6.5 OFF-GAS SYSTEM

The joule-heated melter and attached canister are connected to the off-gas system via the spray calciner. The off-gas system for the JHGM process is basically the same as that described for the ICGM process in Appendix A, except that the JHGM off-gas system has a higher capacity.

Two identical, parallel off-gas systems were identified for the ICGM system because of the use of two spray calciners. For operating efficiency, parallel systems may be required for the JHGM process. Additional data on the JHGM off-gas system is given in Table B-5 in Section B.8.

During any prolonged periods when the JHGM process is shut down, the sintered-metal filters in the spray calciner could become clogged with volatiles from the tank of molten glass.

Apparently the volatiles are captured on the calcine during calciner operating periods and do not penetrate into the pores of the sintered-metal filters. However, during calciner idling times the protective calcine layer on the filters is greatly reduced and the volatiles condense and accumulate on the filters. Research and development is required to identify and to resolve this area of concern. In addition, the needed research and development identified in Appendix A for the ICGM off-gas system are applicable to the JHGM process.

B.7 ALTERNATIVE PROCESSES AND EQUIPMENT

An alternative to the JHGM process is the pot melter used in the French AVM process. This process has a somewhat limited melting capacity; thus, two or more melting pots may be required to meet production requirements. The permissible melting temperature is also lower (approximately 1050⁰ versus 1200⁰C for the glass melter), which may lead to a lower quality glass. Finally, the expected lifetime of the pot melter is expected to be low, which could result in higher maintenance and disposal costs.

B.8 EQUIPMENT DATA SHEETS AND DRAWINGS

The reader should refer to Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

TABLE B-3. Equipment Data Sheet for the Spray Calciner System

Function

Atomize, dry and calcine HLLW.

Process Interfaces

HLLW feed system; off-gas system; joule-heated melter.

Design Parameters and Features:

Calciner chamber: type--right cylinder; dimensions = 91 cm in dia by 300 cm high (cylindrical portion only); heat transfer area = 8.7 m^2 ; material--nickel-chromium-iron alloy; wall thickness = 0.64 cm; bottom cone has 60° sloped sides.

Furnace: type--480-V, 3-phase electrical resistance; number of annular zones = 4; control--silicon-controlled rectifier; maximum power = 280 kW at 70 kW/zone; normal operating temperature = 800°C ; maximum operating temperature = 1000°C .

Atomizing nozzle: type--pneumatic, internal mix, such as Model 72 by Spray Systems Co.; nozzle tip is 304 stainless steel with a 96% alumina insert; atomizing air--facility compressed air; air-to-liquid ratio = 300 to 1 volumetric; liquid control--magnetic flowmeter feedback to valve in HLLW recirculation line.

Filters: type--sintered powdered metal cylinders; total filter area = 13.9 m^2 ; mean pore size = 65μ ; material--316 stainless steel; number of filter cylinders = 68; cylinder dimensions = 7 cm in OD, 91 cm long and 0.16 cm thick.

Filter blowback system: type--venturi; blowback air pressure = 4.1 to 6.8 atm gauge; blowback nozzle size = 0.38 to 0.46 cm; percent of filters cleaned simultaneously = 20; blowback cycle = 8 min; blowback duration = 0.25 s.

Vibrators: type--pneumatic dry lubricant such as Model 1300 AC Vibramight by Cleveland Vibrator Co. or equivalent; motive air pressure = 2.7 atm gauge; location--two 180° apart on side of calciner chamber and one on cone.

Glass frit feed system (2): components are frit bin, proportional feeder and air-lock valves; feed rate = 0 to 55 kg/h; nominal feed rate = 34 kg/h; bin size = 1225 kg of frit (510 L/d); proportional feeder--potential candidate is a gravimetric unit with a continuous weight belt; air locks--air locks must prevent significant air leakage and prevent migration of contamination out of cell.

Operating Conditions

Capacity = 158 L/h; chamber temperature = 700°C ; filter temperature = 350°C ; pressure = 0.97 atm abs.

Services or Auxiliaries:

480-V, 3-phase electrical; 120-V, single-phase electrical; 80-psi compressed air; 50-psi flushing water.

Space Requirements

3.5 m by 3.0 m horizontal by 5 m vertical (includes $\sim 0.5 \text{ m}$ access on all sides).

Maintenance Methods

Remote except for proportional frit feeder.

Remarks

Figure B-3. Joule-Heated Ceramic Melter

TABLE B-4. Equipment Data Sheet for the Joule-Heated Glass Melter

Function

Continuously melts calcined HLLW and supplemental glass frit into a borosilicate glass.

Process Interfaces

Spray calciner system; canister transport system.

Design Parameters

Capacity = 75 kg glass/h; temperature = 900⁰ to 1350⁰C; pressure = 0.75 to 1.5 atm abs.

Operating Conditions

Capacity = 50 kg glass/h; temperature = 1250⁰C; pressure = approximately 0.97 atm abs.

Features

Spray calciner to melter connection: dimensions--15-cm dia pipe, approximately 1.5 m long; materials--stainless steel 304L; temperature = 400⁰C; services required are cooling air and instrument air.

- The connecting section accommodates thermal expansion and minimizes flow area discontinuities that may cause buildup and blockage of the granular/powder feed mixture.
- It may require integral vibrators to prevent blockage.
- It must be readily disconnected by remote means.

Main electrodes: size--approximately 60 cm tall by 60 cm wide by 15 cm thick with a 10 cm by 15-cm "foot"; material--chromium-nickel-iron alloy; operating temperature = 1050⁰C with operational upset limits of 1150⁰C for short duration; features--internally air-cooled, L-shaped with small foot at bottom of electrode, face temperatures monitored by thermocouples and electrically insulated from melter shell; service required is sufficient cooling air at ≤ 1 atm gauge to meet operating temperature requirement.

Sump electrodes: size--15-cm-dia cylinder by 10 cm high; material--chromium-nickel-iron alloy; operating temperature = 1050⁰C with operational upset limits of 1150⁰C for short duration; features are internally air-cooled; temperature monitored by thermocouple and electrically insulated from melter liner; service required is sufficient cooling air at ≤ 1 atm gauge to meet operating temperature requirement.

Discharge resistant heaters: size--four 1-kW silicon-carbide rod heaters and two 1-kW, silicon-carbide U-rod heaters; temperatures = 0⁰ to 1400⁰C.

Plasma torch start-up system: capacity = two 20-kW output torches; features--commercial plasma torches modified to make them adaptable to remote connection to the melter for initial start-up and restarts.

Power supply: type--SCR power control, feeding isolation transformer; capacity = 0 to 100 VAC (peak), 0 to 1500 amps.

Melter shell: size = 1.6 m by 2.0 m horizontal by 1.8 m high (approximate outside dimensions); material--stainless steel 304L double wall with baffled air and water cooling; features--provide frame for refractory lining, serves as air-inleakage barrier, provides cooled barrier to glass leakage through refractory inserts, and serves as connection bulkhead for melter service connections.

Refractory inserts: type--K-3 as the glass corrosion-resistant, melter-cavity liner; Alfax-66 castable for superior insulative properties in back of the K-3 lining, and LDS[®] moldable for thermal insulation and thermal expansion properties; cavity size = 1 m by 0.6 m horizontal by 0.6 m deep (additional cavity clearance in melter lid); features--K-3[®] is used for all glass contact surfaces; insulation is cut and inter-layered to minimize glass leakage from the cavity zone.

Canister filling system: type--air-lift principle employing platinum tubes routed through the discharge riser tunnels to each sump; capacity = 0 to 100 kg/h.

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TABLE B-4. (contd)

Canister weight and glass level indication: weight indication type--load cell on the canister support platform; level indication type--gamma-radiation profile scanner built into canister support platform.

Melter instrument system:

- Temperature is measured on melter shell coolant circuits, melter freeze valve, melter lid cavity, glass discharge trough cavity, glass discharge face cavity, electrodes (profile), and glass (profile).
- Flow of melter shell coolants and electrodes coolant is measured.
- Power is measured for all electrodes and resistance heaters.

Services or Auxiliaries

Electrical power (440-V AC, 3-phase and 120-V AC 1-phase); compressed air; instrument air; cooling water; electro-mechanical manipulator.

Space Requirements

The melter, including an approximately 0.5 m allowance for protubences, is approximately 2.5 m by 3.0 m long by 3.0 m high. The canister must fit below the melter. Therefore, allowing room for the canister plus an emergency container suitable for dumping the molten glass adds an additional 2 m to the actual dimensions. The overall size requirements are then 2.5 m by 3.0 m long by 5.0 m high.

Maintenance Methods

Must be capable of remote operation and capable of maintenance while operating in an "idling mode."

Remarks

Figure B-4. Air-Lift Glass Discharge System

TABLE B-5 Equipment Data Sheet for the Off-Gas System

Function

Condense steam and remove radionuclides, nitric acid and NO_x from the off gas of the vitrification process equipment.

Process Interfaces

Spray calciner; reprocessing plant air protection system.

Design Parameters and Features

Venturi scrubber: spray nozzle operating feed rate = 90 L/min operating and 135 L/min design; calciner operating off-gas flow rate = 9,627 L/min and design rate = 14,440 L/min; H₂O + HNO₃ condensate operating rate = 160 L/h and design rate = 240 L/h; operating heat load = 68 kW and design = 102 kW; pressure = 0; gas/liquid separator--integral knockout pot; material = stainless steel 304L (possibly titanium for corrosion resistance).

Condenser: type is shell tube, single pass, baffled floating head with gas on tube side; material--stainless steel 304L; gas flow = 0 to 15,000 L/min (overrated to compensate for loss of venturi efficiency); and heat load = 30 kW.

NO_x absorber: type is bubble cap tower; material = stainless steel 304L; gas flow = 1200 L/min operating, 1800 L/min design; oxygen flow = 44 L/min operating, 66 L/min design.

Demister: type--baffles, bubble caps, and metal mesh in enclosure; gas flow = 570 L/min operating, 850 L/min design; material--stainless steel 304L; and pressure drop = 2 in. H₂O operating, 4 in. H₂O design.

Ruthenium adsorber: type--feric oxide particle bed (18 to 25 mesh); preheater = 6 kW operating; 8 kW design; gas temperature = 350°C; bed size = 160 L for 3 months operation; bed capacity = 19.4 kg Ru; gas flow = 2400 L/min operating, 3600 L/min design.

Iodine adsorber: type--silver zeolite particle bed (12 to 14 mesh); gas flow = 1840 L/min operating, 2760 L/min design; temperature = 0° to 200°C; materials--stainless steel 304L; bed size = 20 L for 3 months operation; bed capacity = 1.6 kg I.

HEPA filter: type = case containing 1 rough filter plus 2 HEPA filters; capacity = 1644 L/min operating; 2468 L/min design; temperature = 0°C to 150°C; dimensions = 35 cm x 35 cm x 15 cm for each filter; overall assembly 1.5 m.

Operating Conditions (at exit from spray calciner)

HNO₃ flow = 234 L/min; H₂O flow = 6843 L/min; air flow = 2408 L/min; NO_x flow = 142 L/min; temperature = 350°C; pressure = 0.97 atm.

Services or Auxiliaries

Space Requirements

Maintenance Methods

Remarks

APPENDIX C

GLASS-CERAMIC PROCESS

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APPENDIX C

GLASS-CERAMIC PROCESS

C.1 INTRODUCTION

Glass-ceramic waste forms may be a desirable alternative to the borosilicate-glass monoliths produced by the reference ICGM process and the JHGM process described in Appendices A and B, respectively. The interest in GC waste forms is due, in part, to some of the characteristics of the borosilicate-glass waste form. The principal concern about the monolithic glass form is the possibility that it may devitrify, either during the initial cooling operation or during the loss of cooling in interim storage or transit or in the geologic repository. The devitrified glass may be up to an order-of-magnitude more water soluble than the original glass, although this level of solubility is still very low. It is also possible that the devitrified glass may be more subject to microstructural cracking, which could increase the exposed surface area of the glass should the canister barrier ever become breached. The probability of this glass degradation scenario occurring may be small and actually mean little in terms of waste migration in the geologic formation, but it has contributed to an interest in developing waste forms that would not have these possible disadvantages.

Glass-ceramic waste forms are being considered because they should be thermally and mechanically more stable than glass waste forms. A GC, as defined in a patent by S. D. Shookey (1960), is a myriad of fine-grained crystals relatively uniform in size, randomly oriented and homogeneously dispersed in a glass matrix; the crystals comprise the major portion of the body. The term "fusion melt" refers to a crystalline glass, but the crystals can be large and nonuniformly distributed. Therefore, all GC are fusion-melt forms, but the converse is not true. A GC waste form should have the following characteristics:

- high waste-loading capability (≥ 20 wt%)
- leach-resistant crystalline form
- leach-resistant residual glass phase
- high mechanical and thermal shock resistance
- low thermal expansion coefficient
- high thermal conductivity
- high softening point.

The major steps in the GC process are: 1) calcining the HLLW to a powder, 2) adjusting the glass/frit composition to produce a parent glass that will form a crystalline phase that meets the definition of a GC, 3) melting the parent glass and the calcine, 4) pouring, forming and cooling the glass, 5) heat treating the glass to nucleate the fine, homogeneous crystalline structure, 6) further heat treating to maximize the glass transformation to crystalline form, and 7) processing and handling the canisters.

C.2 PROCESS DESCRIPTION

The flow diagram for the GC process is shown in Figure C-1. It is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. Design capabilities are 1.5 times the normal operating values. As covered under the ICGM process, HLLW (its composition is given in Appendix A and in the Overall Design Bases Section) is received from the fuel reprocessing plant and stored in the HLLW feed system. Here it can be analyzed and treated, if necessary, before transferring it to the spray calciner.

The GC process requires only one spray calciner, which is similar to those of the reference process but is larger and has a greater capacity. The calcined waste is mixed with glass frit in the lower section of the spray calciner and the calcine/frit mixture falls by gravity into the joule-heated melter where it forms a cold cap of material on top of a reservoir of molten waste glass. The spray calciner operates continuously since the molten glass can rise in the melter while canisters are being changed.

The molten glass is drawn up from the bottom of the melter into a heated discharge trough within the melter. The molten glass is poured into canisters of the same volume but approximately twice the length of those used in the ICGM process. Since the glass has already been formed in the melter, canister filling can be carried out much more rapidly than in the ICGM process.

After the canisters are switched, the filled canister is put in a heat treatment unit where it can be cooled rapidly and stabilized at an average temperature of approximately $650^{\circ}\text{C} \pm 50^{\circ}\text{C}$ for 4 h. The heat treatment unit employs zone cooling to prevent axial temperature gradients. During this heat treatment phase, the glass forms the fine structure of crystal nucleation sites. The heat treatment unit then brings the canister up to a temperature of approximately $850^{\circ}\text{C} \pm 50^{\circ}\text{C}$ for 6 h, during which time the crystal growth approaches an asymptotic value and the glass is converted to GC. From this point on the canister containing the GC product follows the same canister processing steps described in Appendix A. It is seal-welded, leak-tested, decontaminated and then put into interim storage.

The off-gas system for the GC process is basically the same as for the ICGM process. However, it will have a larger capacity; it may be either a single or dual system depending on operating efficiencies.

FIGURE C-1. Glass-Ceramic Process Flow Diagram

C.3 BASES AND ASSUMPTIONS

Most of the bases and assumptions for the GC process are covered in the Overall Process Bases Section and in Appendix A on the ICGM process. Other pertinent items are noted here.

The GC process is at a relatively early stage of development. Laboratory-scale work is still underway on the best GC formulation, and pilot-scale work is just beginning (De, Luksheiter, Lutze, Malow and Scheimer 1976; De, Luksheiter, Lutze, Malow, Schiemer and Tymochowicz 1976; Oguino et al. 1979; McElroy et al. 1979). Therefore, the process preconceptual design presented here is heavily based on assumptions and on engineering judgment.

C.3.1 PROCESS BASES

Depending on the composition of the glass, crystals can form as the glass cools, or if the glass is held at an elevated temperature long enough, molecules can rearrange themselves into crystalline form. These methods have been used for years in making basalt products in the electrical, chemical and construction industries. Attempts in the early 1900's to improve the crystalline form of basalt products were only partially successful, in that the crystals formed were still coarse and unevenly distributed. In the past two decades, a better understanding of 1) the role of nucleating agents, 2) required oxidation states, and 3) heat control requirements of the nucleation and crystallization stages has resulted in the development of GC satisfying Shookey's (1960) definition of GC. Corningware® is a good example of a GC, in that it is 95% to 98% crystalline. It is because of its uniformly distributed, fine-grained crystalline form that GC has thermal and mechanical shock resistance superior to borosilicate-glass forms.

Glass-ceramic waste forms are usually named after the dominant crystalline phase present. Therefore, waste forms, such as h-celsian, diopside, perovskite and euryptite contain these crystals as the dominant phase. However, several of the crystalline phases will be present in any one form, but in different ratios. A requirement of GC waste forms is that the radionuclides of the calcined HLLW be bound up in the crystalline phases as part of the crystalline structure, or be captured within the crystalline lattice. In addition, the residual glass phase, which will also contain some of the radionuclides, must be durable when in a nuclear waste form. Much of the laboratory work that has been done to date has been with a simulated 20 wt% waste loading. However, in this study it is assumed that a 30 wt% waste loading is achievable.

A GC in which the residual glass is enriched with silica and alumina is desirable since it means a more leach-resistant residual glass. The GC compositions contain higher percentages of alumina than does the borosilicate-glass composition (10 to 24 wt% for GC as compared to 1 wt% for a typical borosilicate glass). A high borate (≥ 10 wt%) concentrations is undesirable because it impedes nucleation and leads to coarse crystal growth (De et al. 1976a). Glass-ceramics contain 1 to 10 wt% borate versus 15 wt% for typical borosilicate glass. As a result

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of the above considerations, the parent glasses for GC normally require a melting temperature of 1250⁰ to 1350⁰C. Due to higher melting temperatures, the joule-heated ceramic melter will require electrodes that are more corrosion resistant than the Inconel alloys proposed for the JHGM process. Tin-oxide electrodes are presently being tested for this temperature range. In addition, since the parent glass of the GC form is especially formulated to enhance crystallization, it may pose an operational problem due to a crystalline sludge forming in the joule-heated melter.

The dominant crystalline phase formed, and therefore the type of GC, is controlled by both the glass/frit formulation and by the heat treatment during the nucleation and crystal growth phases. Since some variation in the composition of the HLLW can be expected in a large facility, the HLLW feed system must be able to accurately analyze the feed solution and add glass formers that may be necessary to achieve the desired parent glass.

The post-melt heat treatments necessary to make the GC are the most significant variation from the JHGM process described in Appendix B. A GC form may eventually be developed that can be produced by controlling the cooldown history of the parent glass after it is poured into the canister (Oguino et al. 1979; De et al. 1976b). However, the present assumption is that molten glass must be poured rapidly into a canister that is continuously being cooled and that will be exposed to controlled heat treatment/cooling cycles. After undergoing the heat treatment process that creates the GC, it is not necessary to maintain the same degree of cooling as is specified for the reference processes. This is because devitrification (uncontrolled crystallization) occurs at temperatures above approximately 650⁰C, and the GC will be thermally stable up to approximately 900⁰C. It should be noted that a thermal analysis has not been performed at this preconceptual design stage to insure that the assumed heat treatments can be conducted within the temperature and time limits specified for the assumed canister size.

Based on work performed at the Hahn-Meitner Institute for Nuclear Research, Berlin, Germany and on bench-scale formulation tests being done at PNL, the h-celsian type GC may emerge as the preferred composition. Both the German work and the current PNL work is based on a 20 wt% loading of the simulated waste in the GC form. Neither process used the reference HLLW composition selected for this study. Therefore, a proven material balance for a 30 wt% waste loading is not available. Table C-1 gives the composition ranges for the parent glass of the h-celsian GC and the composition of the calcine produced from the reference HLLW. Further research and development is required to define exactly the composition of the parent glass and to demonstrate a quality product with a 30 wt% waste loading.

C.3.2 CANISTER

The decay heat loading of the canister is restricted by the same criteria given for the borosilicate-glass processes. Therefore, the heat loading in the canister is limited to 3.06 kW. Assuming a 30 wt% waste loading in the GC, each canister will contain 272 kg (91 L) of GC.

The canisters are constructed of stainless steel to withstand the long-term pool storage before being emplaced in a repository. The canister connection and handling flange must meet

TABLE C-1. Composition of Process Materials

Constituents	Quantity, wt%
Parent Glass	
SiO ₂	32-37
Al ₂ O ₃	10-17
B ₂ O ₃	5-8
CaO	6-10
BaO	18-23
Na ₂ O	2-6
Li ₂ O	0-2
TiO ₂	5-11
ZnO	4-6
Ca ₂ O	0-2
Waste Calcine	
Fission product oxides	59.9
Actinide oxides	2.3
UO ₃	10.9
PuO ₂	0.1
Activation product oxides	0.8
Na ₂ O	0.2
Fe ₂ O ₃	3.1
NiO	0.2
Cr ₂ O ₃	0.5
Gd ₂ O ₃	19.8
P ₂ O ₅	2.2

the same criteria as the canister for the JHGM process. These criteria include 1) ease of canister handling with a quick-connection grapple from an overhead crane, 2) rapid connection to the melter with minimum air inleakage, 3) ease of lid fit-up for remote welding, and 4) a lid closure design suitable for ultrasonic weld-inspection.

As discussed above in Section C.3.1, an analysis of the thermal history of the glass in the canister has not been performed. This analysis needs to be combined with the heat treatment specifications for the particular GC chosen, to ensure that the temperature gradients and times to heatup or cooldown can be accomplished given the assumed canister dimensions. Present analyses indicate that a 21.6-cm-dia canister containing waste with an assumed heat loading of approximately 33 W/L would have an approximately 90°C temperature gradient from the canister centerline to the wall at steady state. While the degree of thermal gradient that can be tolerated during the growing of the fine-grained GC is not known at this time, the canister dimensions are assumed to be 22.9 cm in OD by 310 cm in length. This is essentially doubling the length of the ICGM and JHGM canisters, while halving the volume per unit length.

Cooling fins are assumed to be necessary in the canister to achieve the temperature histories necessary to provide a high-quality GC. Cooling fins in a relatively cold canister can cause glass-pouring problems because of a tendency for the glass to freeze on the cooling fins near the top of the canister. However, this problem can usually be overcome by preheating the canister and pouring the glass rapidly so the incoming glass remelts any glass that tends to freeze on the cooling fins. Rapid pouring also tends to eliminate the voids common in the bottom of a canister. The voids are formed by the glass freezing into a structure resembling coiled rope as the glass enters a cold canister. Figure C-2 is a cutaway view of the proposed canister.

FIGURE C-2. Glass-Ceramic Canister

C.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTES

The handling requirements and volume estimates for effluents, waste and scrap from the GC process are the same as those estimated for the JHGM process described in Appendix B.

C.3.4 UTILITIES AND MATERIALS

The utility and material estimates for the GC process are the same as those listed for the ICGM process in Tables A-4 and A-5 in Appendix A. Since the tables only show the overall requirements for the waste facility, small differences due to individual pieces of equipment are not indicated. In addition, since both processes lead to a similar waste form in the same type of canister, there is little difference in material requirements.

C.3.5 INTERIM STORAGE

It is assumed that the GC canisters will be stored under water in pools in the fuel reprocessing plant before being moved to final storage or disposal. In compliance with the project assumptions given in the Overall Process Bases Section, the canister will be stored in interim storage for a period of approximately 50 yr. After 50 yr the decay heat of the canister will have decreased to 0.5 kW, and the canister can be placed in a geologic repository.

Because the GC waste form is already a crystalline ceramic form, there is not the concern that elevated temperatures may cause product deterioration, such as devitrification. Therefore, there are more options for the type of storage for these canisters, which might open the possibility of using carbon steel instead of stainless steel for the canisters if air storage is sufficient or available.

C.3.6 SHIPPING

The advantages of the GC in terms of storage also apply to shipping. Elevated temperatures could be tolerated during shipping without fear of product degradation, which may be a concern with borosilicate-glass waste forms.

C.3.7 DISPOSAL

The GC waste form can tolerate storage at temperatures that might lead to devitrification and mild degradation of a borosilicate-glass waste form. Therefore, a canister containing GC could theoretically contain a higher decay heat loading than a canister containing glass. However, in practice the repository heat loading criterion is the determining factor.

C.4 FACILITY AND EQUIPMENT REQUIREMENTS

The required GC process cell volume is between that required for the ICGM process and the JHGM process. Due to the length of the canisters the cell height is greater. The process cell is a reinforced concrete structure that contains ample open space and areas for equipment access and replacement. The overall requirements for the waste facility and its support groups and services are covered in the Overall Process Bases Section. The demands and limitations imposed by the requirements of remotely operating and maintaining all in-cell equipment are of major importance.

The layout of the GC processing equipment is shown in Figure C-3 and the major pieces of equipment required are given in Table C-2. The volume for each piece of equipment includes one-half meter on all sides to allow for protuberances and service connections. In addition, the canister processing equipment volume includes the height of a canister above the top of the equipment. The cell layout shows additional space for maintenance, decontamination and equipment replacement.

FIGURE C-3. Cell Arrangement

TABLE C-2. Process Cell Volume Requirements
for Major Processing Equipment

Equipment	Dimensions (width x depth x height), m	Volume, m ³
HLLW system		
Batch tank A	2.5 x 2.5 x 4	25
Batch tank B	2.5 x 2.5 x 4	25
Feed tank	1.5 x 1.5 x 3	7
Piping manifold	2.0 x 3.0 x 3	18
Sample analysis Equipment	2.0 x 3.0 x 3	18
Spray calciner	3.5 x 4.0 x 5	58
Joule-Heated glass melter	2.5 x 3.0 x 6	45
Canister processing system		
GC heat treatment unit	3.0 x 3.5 x 8	84
Forced-air cooling rack	2.0 x 3.0 x 6	36
Lid-closure station	2.0 x 2.0 x 6	24
Helium leak-detection station	2.0 x 2.0 x 6	24
Decontamination station	2.0 x 4.0 x 7	56
Canister rack	2.0 x 3.0 x 6	36
Storage transfer	2.0 x 2.0 x 5	<u>20</u>
TOTAL		476

C.5 STATE OF PROCESS DEVELOPMENT

German researchers at the Hahn-Meitner Institute in Berlin have carried out laboratory- and semitechnical-scale work on the use of GC as a nuclear waste form (De et al. 1976a and b). They have also carried out limited radioactive work on a small scale with the French commissariat at L'Energie Atomique in the Vulcain Cell at Marcoule, France. They have identified four GC that they feel are promising alternatives to borosilicate glass. These are celsian, diopside, eucryptite and perovskite. The principal crystal phases for each of these types are given in Table C-3. The German researchers have performed leach and impact tests and have measured the thermal expansion of the GC. In general, they found that GC have better resistance to damage on impact and lower thermal expansion coefficients, which are desirable features. The leach test results were mixed; some of the phases showed improved leach resistance, while other phases and the residual glass showed poorer leach resistance than borosilicate glass.

The Japanese Power Reactor and Nuclear Fuel Development Corporation (PNC) sponsored the Nippon Electric Glass Company to investigate five GC types (Oguino et al. 1979). The same four GC types referenced in the German work were found to have potential as a GC waste form. The Japanese have established the requirement that the GC be formed during a controlled

TABLE C-3. Principal Crystal Phases in the GC

<u>Glass-Ceramic</u>	<u>Crystal Phases</u>
Celsian	h-celsian ($\text{BaAl}_2\text{Si}_2\text{O}_8$) perovskite (CaTiO_3) BaMoO_4 pollucite ($(\text{Cs,Na})\text{AlSi}_2\text{O}_6$)
Eucryptite	B-eucryptite (LiAlSiO_4) CaMoO_4 $(\text{Ce,Zr})\text{O}_2$ $\text{Ca}_4\text{La}_6(\text{SiO}_4)_6(\text{OH})_2$ Li-Zn-silicates pollucite
Perovskite	perovskite (CaTiO_3) MO-nosean $\text{Na}_8(\text{AlSiO}_4)_6\text{MoO}_4$ $(\text{Ce,Zr})\text{O}_2$ SrMoO_4
Diopside	pollucite ($(\text{Cs,Na})\text{AlSi}_2\text{O}_6$) diopside $\text{CaMg}(\text{SiO}_3)_2$ CaMoO_4 MO-nosean $\text{Na}_8(\text{AlSiO}_4)_6\text{MoO}_4$

cooldown process as opposed to the multistep heat treatment process normally required to form a GC. Therefore, they concentrated their work on a diopside GC that could be formed during a controlled cooldown process. They have cast up to 62 kg of this material in a 30-cm-dia canister, and they report satisfactory GC characteristics. However, from the data reported it appears that the leach resistance of the diopside GC may be poorer than that of borosilicate glass.

Ongoing work at PNL includes laboratory investigations of several GC forms and a high-temperature, joule-heated ceramic melter with tin-oxide electrodes. The melter has produced up to 100 kg of parent glass for GC forms (McElroy et al. 1979). Future work should provide a detailed comparison of the GC waste with the borosilicate-glass waste form.

C.6 UNIT OPERATIONS

The equipment and process operations for the GC process are basically very similar to those for the JHGM process. Variations in the melter design and in the canister processing sequence are described in the following subsections.

C.6.1 HLLW FEED SYSTEM

The construction and function of the HLLW feed system are the same as described in Appendix A for the ICGM process. However, the operation of the system is expected to require more attention, since the GC form is not as tolerant of variations in the batch chemistry as is the borosilicate-glass waste form. This will require more accurate analysis of the HLLW feed and closer control over any additives needed to achieve the correct parent glass for the GC form. Research and development work is required to define the HLLW treatment process necessary to insure uniformity. In addition, the concerns identified for the reference process HLLW feed system also need to be addressed.

C.6.2 SPRAY CALCINER

The number, size, function, construction and operation of the spray calciner system are the same as described in Appendix B for the JHGM process. The research and development efforts noted in Appendix A for spray calciners are also required for the GC process.

C.6.3 JOULE-HEATED GLASS MELTER

The joule-heated glass melter for the GC process must be designed to melt glasses at temperatures of at least 1350°C, which is about 200°C higher than the temperatures used in the JHGM process. The Monfrax K-3 insulation used for the JHGM process can also be used under these high-temperature conditions. However, it is anticipated that the air-cooled Inconel electrodes used in the JHGM process are not suitable for sustained operation at such high temperatures. Therefore, the melter for the GC process is specified as having tin-oxide electrodes. These perform satisfactorily as long as the glass batch is kept in a high-oxidation state.

There are two other aspects of the melter design that will require further investigation for the GC process. One is the potential problem with excessive volatility of some of the radionuclides at the higher temperatures. The second is the potential need to maintain high pouring rates, on the order of 100 kg/h, to eliminate void and blockage formations in the canister. This preconceptual design study assumes that the design presented in Appendix B for the joule-heated glass melter is capable of operating within both of these potential constraints. Thus, the function, general operation, design and configuration of and the research and development work required for this melter are the same as those covered in Appendix B.

C.6.4 CANISTER CLOSURE, DECONTAMINATION AND TRANSPORT SYSTEM

The canister heat treatment necessary to convert the parent glass into the GC form is carried out by four, combination furnace/cooling units. The heat treatment consists of rapid cooling to a crystal nucleation temperature of approximately 650°C, which is maintained for 4 h. This is followed by allowing the internal heat generation to raise the glass temperature to 850°C and maintaining that for 8 h. The heat treatment unit contains baffles to separate the canister heating and cooling into five, 2-ft zones. This feature prevents a significant vertical thermal gradient from developing in the canister. Each zone has redundant canister wall contact thermocouples to aid in heat treatment control. The units are used primarily in the cooling mode, but contain glow-bar heaters in each zone to achieve proper heat treatment. Further thermal analysis may show that the heaters are not required. Table C-4 provides information on these furnace/cooling units.

TABLE C-4. Equipment Data Sheet for the GC Heat Treatment Unit

Function

Provides canister temperature control necessary to turn the parent glass waste form into a GC waste form.

Process Interfaces

Canister transport system.

Design Parameters and Features

Capacity = 4 canisters; material--steel frame and exterior liner; each canister pod is insulated with refractory.

Temperature control: Each canister pod will consist of five horizontal zones separated by baffles to obtain a uniform axial temperature profile in the canister. Each zone will have the capacity to supply 3-kW heating from glow bars or 10-kW cooling with forced air.

Instrumentation: shielded, contact thermocouples capable of measuring the canister wall temperature in each axial zone of the canister pod; power production in each axial zone when in heating mode; heat removal rate in each axial zone when in cooling mode.

Dimensions: Each canister pod will consist of five heating/cooling zones 60 cm in height. The interior dimensions of the rectangular pod are approximately 60 cm by 60 cm by 340 cm. Baffles that separate the axial zones fit closely around the 22.9-cm-dia canister.

Operating Conditions

Temperature = 800°C; pressure is ambient.

Services or Auxiliaries

Electrical power (45 kW of 220 VAC); cooling air (500 scfm).

Space Requirements

Approximate dimensions are 2.5 m by 3.0 m by 4 m high.

Maintenance Methods

Remote maintenance in place is required. The overhead hoist and a remote control, electro-mechanical manipulator is used for carrying out maintenance activities.

Remarks

Temperature control of the canister as it is being filled is also specified as part of the heat treatment process. This is carried out in a baffled, clam-shell canister sleeve that wraps around the canister.

The remaining steps, lid-closure welding, leak inspection, decontamination and transport out of the cell are similar to the steps described in Appendix A for the ICGM process. A melter to canister connection system has not been designed. It must meet the requirements given in Appendix B.

C.6.5 OFF-GAS SYSTEM

The off-gas system for the GC process and the off-gas system requirements are the same as described for the JHGM process in Appendix B. A potential problem requiring further research and development work is the clogging of the sintered-metal filters in the spray calciner by the volatiles from the high-temperature glass used in the GC process.

C.7 ALTERNATIVE PROCESSES AND EQUIPMENT CONSIDERED

The direct liquid-fed, joule-heated melter could be considered as an alternative to the combination of the spray calciner and the joule-heated glass melter, although the feasibility of producing a high-temperature glass with a liquid-fed melter has not been thoroughly investigated.

An alternative to the monolithic form of the GC would be the production of marbles, which would then be subsequently heat-treated. Since the marbles could be expected to fuse together during the heat treatment, it would be necessary to combine the heat treatment with immobilization of the marbles in a metal matrix. This would mean that an alloy that could be poured at the desired nucleation temperature (approximately 650°C) would have to be used and that the canister containing the marbles and the metal would then have to be raised to the crystal growth temperature of approximately 850°C and held there for up to 8 h. This method may provide superior control of the heat treatment as compared to a monolith in a canister, but it raises the process complexity beyond either the GC process or the MIL process.

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APPENDIX D

MARBLES-IN-LEAD MATRIX PROCESS

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APPENDIX D
MARBLES-IN-LEAD MATRIX PROCESS

D.1 INTRODUCTION

Marbles are a glass waste form alternative to the glass monoliths produced in the ICGM and JHGM processes (the ICGM process is also referred to herein as the reference process). In the JHGM process, glass is drained directly into a canister. Product quality verification may be difficult in this process as well as in the ICGM process. Marble production provides separate steps between the melter and the canister to allow for quality assurance and the possibility of recycling through the melter if a below-standard product is produced. In addition, multiple layers of protection can provide an improved waste form. Obviously, metal matrices increase mechanical strength and improve thermal characteristics (Jardine and Steindler 1978). Use of a metal matrix with the waste glass marbles results in a multibarrier product with improved thermal conductivity and lower internal temperatures than the products from the ICGM and JHGM processes. However, the production of the MIL waste form increases operation complexity, cost and risk when compared to the ICGM and JHGM processes.

The MIL facility will have the capability to 1) calcine and melt the HLLW, 2) form small glass marbles, 3) load the marbles into a canister, 4) fill the voids and annulus of a canister with a lead matrix, 5) seal-weld the canister, 6) check its integrity, and 7) decontaminate and transport canisters to an interim storage facility before final disposition.

D.2 PROCESS DESCRIPTION

The MIL process is depicted in the process flow diagram shown in Figure D-1. As covered in the reference process (Appendix A), HLLW (the basic composition is shown in the Overall Process Bases Section) is received in the waste facility and is temporarily stored in the HLLW feed system. Here it can be sampled and necessary adjustments made before transferring it to the spray calciner.

HLLW is transferred from the feed system and introduced into the calciner at a controlled rate through a pneumatic atomizing spray nozzle. The spray calciner decomposes the chemicals in the HLLW into an off gas and a metal oxide calcine that is suitable for making glass. Glass frit (a glass-forming additive) is introduced into the bottom of the calciner at a rate proportional to the HLLW feed rate to the calciner.

The spray calciner for the MIL process has an additional entrance near the bottom that is not provided on the reference calciners. This is for the introduction of crushed, recyclable feed generated downstream in the process. The recycled glass and/or frit-calcine mixture drops from the calciner cone through a remotely made connection to a joule-heated glass melter at a nominal operating rate of 51 kg/h.

As described in the JHGM process (Appendix B), calcine and frit are fed into a melter for melting into glass. Energy is supplied to the melter by passing an alternating current through molten glass between immersed electrodes. Calcine and frit flow from the calciner directly onto the surface of the molten glass. Thus, a relatively cool layer, or cold cap, forms on the molten glass. The cool layer continuously settles and melts into the molten pool. After a given residence time, the melt flows under a weir and up risers into the overflow trough where it is continuously discharged as a thin molten stream.

The molten glass stream is poured through a remotely made connection into an enclosure housing the marble machine. The marble machine is the first step of this process that deviates significantly from the ICGM and JHGM processes. The MIL process lays the stream of molten glass across the top of moving and vibrating molds at a nominal rate of 51 kg/h. As the molds containing glass are moved along the vibrating track, the molten glass is formed into semi-molten spheres, which cool in a few seconds to form marbles. When the molds reach the end of the vibrating track, the glass marbles are removed from the molds onto a vibrating sorter.

The sloping, vibrating sorter, or table, is located in the same enclosure under the exit of the marble machine. Spherical shaped marbles roll off of the sorter in one area and are then fed into the pneumatic transfer system. Slag, glass ribbons, broken marbles and deformed marbles do not roll and hence are vibrated off the sorter in other areas and fall to the bottom of the marble machine enclosure, where they are fed into a mill for crushing and recycling.

FIGURE D-1. Marbles-in-Lead Matrix Process Flow Diagram

The mill is attached to the bottom of the marble machine enclosure. Slag resulting from overpouring, deformed marbles, glass ribbons and dust feeds into the mill by gravity and/or vibration. When enough material is accumulated, the mill is started to reduce the glass material to particle sizes that can be conveyed in the pneumatic system and recycled into the main process stream at the spray calciner. The solid process scrap material is conveyed to a scrap collection hopper located at a sufficient elevation so the material can be control-fed into the base of the spray calciner.

As indicated, the marble machine and sorter are contained in an enclosure to control the flow of contaminated material and the operating environment of the marble machine. Means of preheating the marble machine molds and of assuring movement of scrap material are included in the enclosure.

A closed, negative-pressure pneumatic system with two loops is contemplated to 1) transport acceptable marbles at an operating rate of 49 kg/h to the marble surge hopper and 2) to convey ground, scrap glass particles to a scrap collection hopper at rates up to and equal to that of glass production.

A stainless steel surge hopper is used to collect the marbles being conveyed via the pneumatic system. Glass fines that may have been conveyed with the marbles are separated and carried out the top of the surge hopper via the pneumatic system to the scrap collection hopper. The marbles flow from the surge hopper to the volume hopper via gravity.

When filled to the level of its upper valve, the volume hopper contains slightly less marbles than required to completely fill the inner basket (approximately 260 kg) of the waste canister. Closing the upper slide valve establishes the batch size to be fed to the inner basket. A fill mechanism serves the functions of forming a moveable transition between the volume hopper and the inner basket of the canister and directing and controlling the flow of the marbles into the inner basket. After the inner basket has been filled with the measured volume of marbles, the opening in the top of the inner basket is closed and the canister and its contents are transferred to the station where molten lead is poured into the annulus and voids in the canister.

Before pouring the lead, the canister contents may have to be cooled; then the canister and its contents are heated to an equilibrium temperature at or near the pouring temperature of lead. With venting and by sequencing the heating and pouring, molten lead is poured into the canister and fills all of the remaining voids. After lead filling, the canister is seal-welded and checked. It is then inspected, decontaminated and placed in storage.

D.3 BASES AND ASSUMPTIONS

Many of the criteria and bases for the MIL process are identified in the Overall Process Bases Section. Data pertinent to the HLLW and its waste forms are also presented under the ICGM process (Appendix A). However, there are additional bases and assumptions applicable to this process, and these are noted below.

D.3.1 PROCESS BASES

As previously noted, the flow diagram for the MIL process is shown in Figure D-1, and it is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. Design capabilities are 1.5 times the nominal operating values. At a waste loading of 30 wt% and a glass density of 3.0 kg/L, the process is designed to solidify 5700 L of HLLW per day. This requires a design glass-melting rate of 73 kg/h and a nominal glass-melting rate of 49 kg/h. The bulk glass contains a heat loading of 33.7 W/L. The bulk density of the marbles is 1.72 kg/L in large volumes.

D.3.2 CANISTER

As noted in the Overall Process Bases Section, a major constraint on the MIL process, as well as other processes, is the allowable heat loading of the canister. The canister criteria are:

- centerline temperature $\leq 550^{\circ}\text{C}$
- wall temperature $\leq 100^{\circ}\text{C}$ after 50 yr
- heat loading = 3.06 kW/canister.

These criteria permit the placing of approximately 150 L of waste in the glass marble form in a canister. To utilize a reasonable length-to-diameter ratio for process operations, lead filling and handling, a canister 51 cm in OD and approximately 152 cm tall (20 in. in OD by 5 ft tall) was selected for the MIL process; the canister and inner basket are shown in Figure D-2. The inner basket is positioned inside the canister before the head is welded in place. Other aspects of the canister are covered in Appendix A.

D.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

For this study it is assumed, with one exception, that the effluents from the MIL process are similar in content and volume to the effluents from the reference process (Appendix A). The exception is the effluents associated with the operation of the lead-melting furnace. For simplicity, this furnace is located and operated outside of a radioactive or contaminated zone, so its effluents are similar to those of a lead-melting furnace in a commercial installation.

Except for additional worn equipment due to the fact that the MIL process is more expanded and complex than the reference process, it is assumed that the wastes from the MIL process are of the same order of magnitude in volume and contents as the reference process. It is also assumed that systems and provisions are included in the fuel reprocessing plant to treat,

FIGURE D-2. Canister and Inner Basket

reduce the volume and package both liquid and solid wastes. Finally, in the MIL process, it is assumed that all solidified process scrap that may contain high-level waste is recycled and incorporated into the process.

D.3.4 UTILITIES AND MATERIALS

The estimated utilities and essential materials required are shown in Tables D-1 and D-2, respectively. These estimates are based on the requirements for the reference process with adjustments for additional process steps and anticipated space and volume changes.

TABLE D-1. Estimate of Utilities Required for MIL Process

<u>Utility</u>	<u>Capacity Required</u>	<u>Average Use Rate</u>	<u>Disposition</u>
Electricity	1600 kW	900 kW	NA
Raw water	2000 L/min	600 L/min	Recycled as HLLW or ILLW
Process water	600 L/min	20 L/min	HLLW or ILLW
Sanitary water	40 L/min	20 L/min	Sanitary or chemical sewer-tile field
Air	750 scmm	500 scmm	As appropriate
Steam	25 kg/min	5 kg/min	As appropriate

TABLE D-2. Estimate of Materials Required for MIL Process(a)

<u>Material</u>	<u>Amount Required Per Unit Time</u>
Frit	820 kg/d
Canisters	4.52/d
Caustic (19M)	8,000 L/yr
Nitric acid (12.2M)	12,000 L/yr
Helium sources	5.0/d
Argon	5,000 L/d
Detergent (for example Tide)	1,000 kg/yr
Oxygen	6,000 L/d

(a) Excludes maintenance materials and apparel.

D.4 FACILITY AND EQUIPMENT REQUIREMENTS

The MIL process is contained in a reinforced concrete structure similar to that shown in Figure D-3. The major pieces or equipment systems are listed below:

- HLLW feed system
- calciner
- ceramic melter
- marble maker system
- pneumatic transfer system
- scrap recycling system
- marble hopper and fill system
- metal fill system
- canister handling system
- off-gas system

The overall requirements for the waste facility and its support groups and services are covered in the Overall Process Bases Section. The major factors to be considered in future process evaluations or cost estimates are the requirements for remote operations and for remote maintenance of all in-cell equipment.

FIGURE D-3. Marbles-in-Lead Matrix Waste Facility General Arrangement

D.5 STATE OF PROCESS DEVELOPMENT

The MIL process introduces a number of process steps that have not been performed using wastes or simulated wastes. In addition, a number of the process steps have not been performed on a scale near that required for a production facility. The MIL process also involves a number of involved mechanical/chemical steps that have to be performed remotely using equipment that will require remote maintenance and remote removal when used in a production facility.

Considerable research, development and tests have been performed at PNL on spray calciners and ceramic melters. As a result, these major pieces of equipment and their appurtenants can easily be applied to the MIL process.

The production of acceptable quality, simulated-waste glass marbles has been successfully demonstrated on a laboratory scale (Rusin et al. 1978). Small volumes of simulated waste in the marble form have been encapsulated successfully (Rusin et al. 1978).

Although the performance of glass as a waste form has been the subject of considerable research, additional studies are required on glass-to-metal bonding interactions and chemistry. Studies on the corrosion resistance of matrix metals in geochemical environments are also required. The multibarrier concept of glass marbles in a metal matrix has good potential for further development, because it offers increased ruggedness and inertness. However, there is also some increase in technological complexity. Future development of marbles as a waste form only requires the demonstration of marble production, their encapsulation and the integration of these operations into an overall process.

D.6 UNIT OPERATIONS

The MIL process has been divided into 16 unit operations. Information on the operation of these systems and areas requiring additional research and development are covered in the following subsections. Drawings and data sheets on the equipment and systems discussed in this section can be found at the back of this appendix in Section D.9.

D.6.1 HLLW FEED SYSTEM

The function of and the requirements for the design and operation of the HLLW feed system for the MIL process are the same as for the reference process (Appendix A). The preparation, verification, analysis, start-up, transfer, operation and shutdown considerations for the HLLW feed system are identical or similar to those of the reference process. The concerns and the necessary research and development identified for the reference process are also applicable to the MIL process.

D.6.2 SPRAY CALCINER

The function of the spray calciner for the MIL process is the same as for the reference process spray calciners. The MIL process requires one, large spray calciner of the type developed at PNL, and its start-up, operating and shutdown procedures are basically the same as those for the reference process; it is the size and capacity specified for the JHGM process in Appendix B. The introduction and coordination of the recycled scrap fuel is the major exception to the ICGM and JHGM processes. The research and development efforts noted in the reference process for spray calciners will also be required during the further optimization of this MIL process.

D.6.3 OFF-GAS SYSTEM

The requirements for and the function and operation of the MIL process off-gas system are the same as for the reference ICGM process; it has the same capacity as the JHGM off-gas system. Due to the use of two spray calciners, two parallel and identical off-gas systems are specified for the ICGM process. For operating efficiency, parallel systems may also be appropriate for the MIL process. The research and development specified for the ICGM and JHGM processes are also required for the MIL process off-gas system.

D.6.4 JOULE-HEATED GLASS MELTER

The function of the melter is to continuously convert glass formers and dried nuclear waste, or calcine, into a chemically stable, waste glass product. The MIL process requires one melter the same size and capacity specified for the JHGM process in Appendix B. Its start-up, operating and shutdown procedures and requirements are basically the same as for the JHGM process. The major exception is that the melt discharge must be coordinated with the speed and operation of the marble machine.

A remote melter is currently being developed and tested at PNL. This effort needs to be finalized; the research and development efforts noted in Appendix B for the melter are also required for the optimization of the MIL process.

D.6.5 MARBLE MAKER

One end of the marble machine is located under the melter discharge tube, so when the melter is in the pouring mode, the stream of molten glass is placed across the top of the moving mold cavities. The glass stream separates due to its viscosity and due to the opening and speed of the mold cavities; globs are then formed into spheres by the vibration of the mold. The spheres are cooled to the solid state and to a temperature so that they will not stick to each other or deform when leaving the mold.

The marble machine consists of aluminum molds mounted adjacent to each other on a continuous roller chain. The mold conveyor follows a set path and is powered by a variable- and adjustable-speed drive. During the first portion of its cycle, the mold conveyor travels on sections that can be vibrated pneumatically. There are at least five sections whose vibrational forces and frequencies can be individually varied. Provisions to cool the empty, returning molds are included. The machine also has a tensioning system, a method to detect mold conveyor deviation, a lubrication system and the necessary temperature detectors.

As indicated above, the operation of the marble machine must be coordinated with that of the glass melter. For start-up, the marble machine vibrators are turned on, the conveyor is started, the molds are preheated to entry temperature and the conveying rate is set to correspond to the expected pouring rate or to nominal operating conditions. Just before pouring is started, the mold preheating is terminated and mold cooling is started.

During operations, the pouring rate and conveyor speed must be maintained in the proper relationship. The mold temperature and marble exit temperature must also be kept within prescribed limits. For normal shutdown, the marble machine is operated after pouring is terminated to clear glass from all of the molds. The power, cooling and vibration systems are then stopped.

A marble machine has been built and operated at PNL. To date, in very limited runs, this machine has worked very well and marbles with acceptable physical characteristics have been made from simulated waste glass. This effort needs to be continued to resolve problems and to instigate changes as covered in detail in Section D.7.

A schematic of the marble machine is shown in Figure D-4 (Section D.9), and design and operating conditions are given in Table D-3 (Section D.9). Additional operating requirements are given in Section D.6.7, which covers the marble machine enclosure.

D.6.6 SORTER

The material leaving the marble machine is deposited via gravity on a sloping table that can be vibrated. The slope and vibration can be adjusted so that acceptable, spherical-shaped

marbles that roll exit in one area, while slag, glass ribbons, broken or deformed marbles and other material exit in other areas. The sorter must be started before pouring of the molten glass into the marble machine and continues to operate until all glass is cleared from the machine.

Incorporated with the sorter are means to sense and monitor the temperature and weight of exiting marbles, a method to evaluate shape and a sampling system. The sampling system is used to remove individual marbles from the process stream; then they are sent to an analytical station or laboratory for the checking of diameters, shapes, composition, and chemical and physical characteristics.

One of the positive features of the MIL process is that it affords the opportunity to monitor and analyze the quality of the glass waste product before it is in a form in which corrections or adjustments cannot be made. At or near the exit of the marble machine is a logical place to monitor and sample marbles. A system to remotely monitor the temperature, weight and shape of the marbles needs to be developed. A remote sampling system and an analytical process for checking the necessary attributes such as shape, diameter, composition, and chemical and physical characteristics in a turnaround time compatible with the processing rate also needs to be devised and tested.

A sorter, as shown in Figure D-5 in Section D.9, has been made and used in conjunction with the operation of the marble machine. The design of a similar unit that is compatible with other aspects of the marble making station needs to be developed and tested. The equipment data sheet (see Table D-4 in Section D.9) gives design and operating conditions for the sorter.

D.6.7 ENCLOSURE

The control of the marble machine environment probably requires that it be located in an enclosure. Provisions for the remote operation, monitoring, removal and replacement of the marble machine and the sorter are included. The enclosure also limits the spread of contaminated process material and provides for its control or recycling.

In addition to providing services to and from the marble maker and sorter, the enclosure has a heater to induce heat into the marble machine molds before start-up. The enclosure also provides for remote connections to the glass melter, the pneumatic system and the mill. No special operating procedures are anticipated for the closure itself. Internal conditions require continuous monitoring during operation to assure that processing variables such as temperature and humidity are within set parameters.

An enclosure design that is compatible with the operating requirements of the enclosed equipment and with the remote aspects of operation needs to be developed and tested. The marble machine enclosure is shown in Figure D-6 (see Section D.9), and data on the enclosure are given in Table D-5 (see Section D.9).

D.6.8 MILL

Dry scrap consisting of dust, deformed marbles, glass ribbons or glass slag is fed into a vertical mill or crusher via gravity, vibrators or air slides. The mill is operated on an intermittent or as-needed basis to reduce the scrap to particle sizes that can be conveyed and recycled into the main process stream. During operations, a means of monitoring the flow of material either into or out of the mill is required. A sound detection system may be used to monitor the operation of the mill. The discharged material is fed by gravity into a pneumatic system pickup point.

There are a number of types of commercial mills, crushers or cutters that are capable of processing glass similar to that in the MIL process. An engineering review and, possibly, laboratory tests are required to determine which type of mill is best suited for this application. Tests are also needed to determine what size is required to handle the largest anticipated globs of glass and to reduce these and other scrap to a particle size capable of being conveyed pneumatically. It is expected that these lab-type tests could be conducted in a manufacturer's plant with simulated glass material.

After completion of the lab tests, a mill must be designed for remote operations, maintenance, installation and replacement. This piece of equipment should be made and tested in a cold facility in conjunction with the marble maker and a pneumatic conveying system. Data on the mill are given in Table D-6 (see Section D.9).

D.6.9 PNEUMATIC SYSTEM

The transport of radioactive materials should be via negative pressure. The prime mover is a rotary, positive-displacement type blower, which provides a vacuum to the receiver vessels via blowback filters. Process material is conveyed through short sections of 300-series stainless steel piping. The short sections allow for remote removal of the piping. Pipe connectors limit protuberances at the joints. Process material enters the system through feeder control valves. Exhaust air is removed from the top of the systems' vessels through sintered stainless steel filters that can be blown back with air pulses. Load cells on the receiving hoppers and/or flow indicators in the system are used to monitor system operation and cleanout.

The pneumatic system must be operating when pouring molten glass onto the marble machine is started and until all of the marbles are cleaned out of the marble machine and sorter. When it is necessary or desirable to convey scrap particles to the collection hopper, the marble loop is valved off and the pneumatic system is started before starting the mill.

For a vacuum and/or closed system it is necessary for the process materials to be introduced into the system through air-lock devices. The design of these air locks affects pressure losses and drops as well as process material inertia and acceleration.

A schematic of the pneumatic system is shown in Figure D-7 (see Section D.9); additional data is given in Table D-7 (see Section D.9). Research, development, testing and designing are required for a pneumatic system capable of transporting 1/2-in.-dia marbles made of high-level nuclear wastes. These efforts are described in detail Section D.7.

D.6.10 RECYCLE FEED

The final receiving vessel for the pneumatic system is the scrap collection hopper located in the upper portion of the process cell. This stainless steel, cylindrical-shaped hopper with a capacity of about 500 kg stores the solid, process scrap material. The hopper is equipped with cooling coils, temperature sensors and level or weight devices to monitor and control the contents.

The material collected in the scrap collection hopper is recycled to the spray calciner through a weigh feeder and gravity pipes. The feed system is similar to the glass frit feed system covered in the ICGM process (Appendix A). It is necessary to monitor and measure additions to the spray calciner and coordinate these with the regular frit feed.

D.6.11 MARBLE SURGE HOPPER

A cylindrical-shaped, stainless steel vessel with an approximate capacity of 300 L is the collection point for the marbles from the pneumatic system. The surge hopper is located high in the cell so gravity can be used for the subsequent operations with the marbles. The flow of marbles out of the surge hopper and into the volumetric discharge hopper is controlled by a slide valve that can close through a column or stream of marbles. The surge hopper is equipped with cooling coils, temperature sensors and level or weight devices to monitor and control the contents.

At this point in the process, material flow is converted from a continuous to a batch type operation. If the surge hopper is filled up, the system operations must be stopped. The level and weight of the marbles in the surge hopper and the internal temperature are monitored during operating periods. Provisions are incorporated to maintain the temperature of the contained marbles below a prescribed limit. The marble surge hopper is shown in Figure D-8 (see Section D.9) and other data are given in Table D-8 (see Section D.9).

D.6.12 VOLUME HOPPER

The volume of this stainless steel hopper, including its inlet connection to its upper valve, equals the volume to be filled by marbles in the inner basket of the storage canister. The slide valve between the surge hopper and the volume hopper is normally open until the proper amount of marbles are in the volume hopper, and then it is closed. Like the surge hopper, the volume hopper is equipped with cooling coils, temperature sensors and level or weight sensors. The canister fill mechanism controls the flow of marbles from the volume hopper.

Operating the volume hopper mainly involves proper sequencing of the upper inlet or feed valve, assuring the proper fill volume, drainage through the canister fill mechanism, and assuring that all of the measured volume has drained through and out of the vessel. Temperatures must be monitored anytime process material is present so cooling can be initiated if necessary.

The volume hopper is shown in Figure D-9 (see Section D.9), and supporting data are given in Table D-9 (see Section D.9).

D.6.13 FILL MECHANISM

After an inspected (and accepted) canister with an inner basket is located under the marble fill station, the fill mechanism is inserted. After starting a vacuum system to remove dust and particles from the falling marbles, the marbles are transferred from the volume hopper to the inner basket of the canister. During and after marble filling, inert gas is introduced into the center of the canister via the fill mechanism to cool the marbles if required and to assist in the entrainment of dust into the vacuum system. After completion of the fill, the gas and vacuum systems are valved off and the fill mechanism is retracted from the canister. The canister fill mechanism is shown in Figure D-10 (see Section D.9), and additional data on the fill mechanism are given in Table D-10 (see Section D.9).

The crucial batch operation of the MIL process is the remote filling and monitoring of the inner basket in the canister with the proper amount of marbles on a repetitive basis with no over- or underfills. An engineering design should be made for a remote, production-sized system that includes the complete volume or measuring system and the canister fill mechanism. A full-scale model should be fabricated and tested in a laboratory demonstration. This demonstration would include the complete remote operation and monitoring of the system as well as the verification of the remote maintenance and replacement aspects.

D.6.14 INNER BASKET CLOSURE

After the inner basket has been filled with the proper amount of marbles (approximately 260 kg), the canister is rotated out from under the retracted fill mechanism and is transferred to a station where a device is inserted into the canister to close the opening in the top of the inner basket. To allow for long periods of time between marble filling and lead pouring and to prevent the radioactive decay heat from causing the marbles to soften or melt, provisions to cool the canisters while in this or subsequent stations are provided. Upon retraction of the closure device, the canister is transferred to the next station.

D.6.15 METAL FILL STATION

The canister is lifted into the canister heater and up against the metal fill mechanism. Provisions are included to cool the canister or its contents via an internal gas flow as required and to heat the canister and its contents to within 50°C of the melting point of the metal alloy to be used (300° to 400°C). When this equilibrium temperature has been reached, the cooling gas flow is terminated, the canister heaters are turned off and the valve is opened to start the lead pouring from the furnace. The liquid metal is introduced through the center pipe to the bottom of the canister and back up through the marbles and the outer annulus. If necessary, a vacuum can be induced in the canister through the metal fill mechanism before and during pouring of the lead. The lead is poured continuously until the desired level is reached, and then the valve is closed. During the lead-pouring process, the bottom and possibly the walls of the canister are cooled to control lead shrinkage voids.

After the valve is closed, the canister is lowered approximately one foot. The metal control valve is then re-opened and metal is permitted to reach the new desired level. As the

metal level drops due to the 5% shrinkage, additional lead is added as required to complete the filling of the canister to the selected level. Then the canister is lowered and removed from under the metal fill station and the metal level is checked.

The lead-melting furnace is located out of but adjacent to the process cell in a service area so the molten lead can flow by gravity through a heated entry tube to the metal fill mechanism. With this out-of-cell location the lead furnace, its controls and its operation are basically the same as a commercial furnace. The heating and controlling of the temperature in the entry tube is a special application as is the control of its isolation or block valves. Assuming the lead-melting furnace is located outside of the process cell also reduces the number of in-cell components and the complexity of the process. An engineering and design program is required to assess if this assumption is the best approach and to design the lead-pouring system, especially the in-cell components.

After the design for the lead-pouring system has been completed or identified, in-cell components should be fabricated and installed in a demonstration mode. Thus, the operation of a remote, metal fill mechanism including metal flow control and monitoring could be demonstrated and evaluated. The remote maintenance and replacement aspects could also be verified.

The metal fill station is shown in Figure D-11 (see Section D.9) and data for the station are given in Table D-11 (see Section D.9). Data on the lead-melting furnace is given in Table D-12 (Section D.9).

D.6.16 CANISTER PROCESSING

After the proper metal level is assured and the canister is moved into the weld station (see Appendix A), a helium source is inserted and the lid is seal-welded. Next, the canister weld is helium leak-checked to ensure its integrity. The canister is also visually inspected. Canisters that fail to pass the leak test are rewelded if possible. Sealed canisters are then moved to a decontamination chamber where any loose contamination is removed. From here the canisters are placed in the in-cell storage racks or are transferred out of cell to an interim waste storage area as noted for the reference process.

D.7 RESEARCH AND DEVELOPMENT REQUIRED

As noted in the above sections on station or system components, there is a definite need to demonstrate all of the remote aspects of each unit operation in a cold demonstration mode. This need also applies to the complete MIL process, and it is recommended that following, or in conjunction with, component tests the system be operated using simulated material. Such a demonstration could also be used to verify and demonstrate the cell arrangement and the equipment handling system.

Although testing the system is a very important aspect in any remote nuclear installation, it should not be started until after the requirements of the major stations or components have been fully explored. For the MIL process, additional efforts should be expended first on 1) the marble machine to improve its configuration and to increase its production rate, 2) the establishment of the design requirements for a pneumatic system to convey nuclear wastes in the form of 1/2-in.-dia marbles, 3) a remote monitoring and analytical station for the marbles, 4) the glass-to-metal bonding interactions, and 5) a metal fill station capable of filling the required size canisters.

D.7.1 MARBLE MACHINE

Small quantities of glass marbles have been made using a machine built by PNL that was based on a Corning Glass Works' patented technique (Knapp and Smith 1962). In its limited production to date, this machine has worked very well and has produced marbles with acceptable physical characteristics. Its operation and development should be continued with improvements or changes made in the following areas:

- Overall production rate of the current test set-up is about 35 kg/h. This needs to be increased to at least 75 kg/h.
- Start-up and shutdown techniques need to be tested and operating procedures developed to optimize speed and rate of pour and to eliminate large globs of slag, small and imperfect spheres and glass stringing.
- A closed-loop cooling system for the track and molds needs to be developed before pouring radioactive materials. The use of water sprays directly in the hot molds is one way of reducing the inlet mold temperature to about 130°C (which has been found to be adequate and necessary); however, this method will spread contaminants and should not be used in the radioactive installation.
- The current prototype marble machine is approximately 18 ft long. This length results in a configuration that will be very difficult to enclose and to operate and handle remotely in a production facility.
- The operation of commercial marble making machines requires continued operator surveillance and adjustments. The Corning type machines are not as dependent on operators as

other machines; however, techniques, procedures and systems need to be developed so the marble making operation can be monitored, adjusted and kept in continued operation remotely.

- A production-sized machine designed for remote operations, maintenance, installation and replacement needs to be developed, built and operated in a cold facility, where all of the remote maintenance and operational aspects can be tested before installing such a system in a radioactive facility. The cold facility would include all aspects of the marble making station including the marble maker, sorter, sampler, scrap handler and enclosures.

D.7.2 PNEUMATIC SYSTEM

From the perspective of safety and contamination control, it has been deemed that any pneumatic system conveying nuclear materials should be operated at a negative pressure in relation to its surrounding environment. A literature review shows that there are very few commercial pneumatic systems operating in the negative pressure mode (Skriba, Wu and Yang 1977). Generally, positive-pressure pneumatic systems are preferred due to their greater line-carrying potential and the greater pressure drop available. However, the unacceptability of leakage in a system transporting nuclear materials dictates that they be operated as a negative-pressure system.

A review of government and industrial laboratories that are engaged in the processing of nuclear fuels indicated only a few were either studying or using pneumatic transfer systems, and most of this effort was with powders (Skriba, Wu and Yang 1977). Although there are systems manufacturers supplying the pneumatic industry, most are independent, specialty suppliers of components. Large suppliers are few in number and may have the tendency to concentrate only on large installations. Small systems are usually designed, procured and installed by the customer. Most hardware components are available from more than one supplier.

The literature available on pneumatic transport system designs is quite extensive, and many design equations have been developed to predict system operating performance. Some of these process design equations are based only on theory; few have been empirically verified. Thus, the performance of a transport system cannot be predicted with a high degree of confidence.

The potential for automation of pneumatic transfer systems is very good. Much of the equipment was developed for bulk handling and storage markets. These systems tend to be large and spread out so that automated operation is desirable. However, maintainability and reliability information for nuclear or analogous system services has not been an area of concern.

A number of suppliers have refined their equipment for sanitary service in areas such as food and pharmaceutical operations, whose requirements are similar in some respects to those of the nuclear area. Further modifications will undoubtedly have to be made, but some of the basic designs are compatible with a nuclear application.

A high degree of confidence is more important in a nuclear material process than in most others, because such a system must operate remotely and require low maintenance. In addition, design changes after system installation and shakedown are extremely expensive and difficult.

Additional engineering studies and laboratory-scale tests using 1/2-in.-dia glass marbles are required to establish system design criteria, operating parameters and equipment specifications. This testing could be performed at a manufacturer's test laboratory, as several manufacturers have systems set up to give preinstallation assurance for different or unusual products or requirements. Preliminary testing is required to determine such parameters as:

- open or closed system
- pipe sizes and bend radius
- velocity and pressure drop ranges
- solids-to-gas ratios
- blower type size, speed and capacity
- marble and particle feeder concepts
- component criteria.

A second logical step in the development of a negative-pressure pneumatic system for the transport of nuclear material would be a cold, pilot-scale installation to demonstrate the system and hardware under simulated process conditions. With such a set-up, a test program could be conducted to 1) explore operating regimes within the control ranges of the system, 2) determine optimal operating conditions and parameters and assess wear and life (e.g., bend radius versus wear), 3) optimize design of solid feeder systems thus reducing pressure drop and improving solids inertia and acceleration, 4) finalize the required remote operations and maintenance provisions, and 5) determine provisions required to eliminate operating problems such as jamming.

A major, unknown component of a negative-pressure pneumatic system that conveys 1/2-in.-dia marbles is the air-lock feed valves. The operation of this type system will require air locks to limit inleakage at all feed points. However, the rotary or double-flapper valves normally used for powders etc. may not be applicable for use with 1/2-in.-dia marbles due to jamming. An engineering program is required to find or to develop an air-lock feeder for 1/2-in.-dia glass marbles.

D.7.3 CANISTER HANDLING

The design and development aspects noted under the reference process (Appendix A) for canister handling and closure are also required for the MIL process. The design of remote systems for the moving, holding, heating and cooling the MIL canisters is required in the next phase of process identification. Selected components of the MIL canister handling system should be fabricated and tested to verify their capability and their remote aspects.

D.8 ALTERNATIVE PROCESSES AND EQUIPMENT

Only a very limited time has been available to develop the preconceptual design bases for the MIL process described in this document. As such, there are a number of viable alternatives that have not been considered or that should be explored further in the subsequent steps necessary to finalize a process design. Some such alternatives are noted in this section.

D.8.1 GENERAL

The alternatives noted in the reference process and in Appendix B pertaining to spray calciners and glass melters are also applicable to the MIL process. One such alternative that would reduce cell height and process components is the elimination of the spray calciner as noted by Bonner and McElroy (1978). They state that:

Due to the efficient heat input capabilities of the ceramic melter, feeding of liquid waste without prior calcination is an attractive option. In this process, liquid waste and slurried glass formers flow into the melter covering the melt surface with a boiling liquid layer. Melter off-gas treatment is essentially that of an evaporator. As liquid waste is added, equilibrium is established with solids from the liquid waste precipitating and eventually melting in the molten pool. Two sets of horizontal electrodes, one above the other, are used in the liquid-fed melter. During liquid feeding, power can be increased to the upper electrodes near the surface of the glass melt where most energy is needed to vaporize the liquid. Otherwise, the melter is constructed and functions similar to the calcine-fed melter.

Another alternative that should be evaluated further is the depositing of marbles directly into a canister. If this would produce an acceptable waste form product, the complexity and requirements of the MIL process could be reduced considerably. This alternative may produce a waste form where the marbles might fuse together at least on point of contact. If used, it would require a system to divert marbles into selected canisters and to monitor and measure the amount added to each canister.

D.8.2 CELL ARRANGEMENT

The cell arrangement displayed in Figure D-2 shows all of the remote processing equipment located in one cell or canyon that is quite high and long. Building costs could be reduced if the size of the cell could be reduced; cell height is of particular concern. To limit and control the potential spread of contaminants, it may be desirable to locate the process equipment in two or more cells. This alternative needs to be considered, while at the same time giving adequate attention to the flow of process material and the remote requirements for operations, maintenance and replacement of equipment.

D.8.3 MARBLE MACHINE

As noted in the section on research and development required for the marble machine (Section D.7.1), a more compact marble making machine is required. It is felt this can best be achieved by refining selected manufacturing processes. As noted by Rusin et al. (1978).

Conventional glass marbles are produced by shearing a viscous glass stream and passing the glass or "gob" through a pair of grooved steel rollers. The rollers are

between 0.6 to 1.2 m long and an additional length is required for cooling of the glass marbles. Typical production rates for 1.6-cm-dia marbles are 180 marbles/min per machine. Normally two machines are fed by a single double-gobbing system.

The conventional marble manufacturing process cannot be easily adapted to production of waste glass marbles in a remote facility. The major drawback is that a high level of operator maintenance is required. The shearing or gobbing system produces glass gobs of a wide tolerance and other gobs stick together. Because of these variations, the marbles may get caught in or fall through the rollers. The rollers need operator attention approximately every 15 minutes.

Another requirement with conventional marble production is that the viscosity-temperature relationship must be within a specified range to allow shearing of the glass stream and to allow sufficient time to form the marble. This range is designated the "working range" of the glass and is determined by the slope of the viscosity-versus-temperature curve. For comparative purposes the working range can be defined as the temperature interval between the flow point (10^5 poise) and the softening point ($10^{7.5}$ to $10^{8.0}$ poise) of the glass. It is highly probable that the 100°C working range of the waste glass is much too short for forming by the conventional process.

A rotary machine and glass-streaming techniques used in the PAMELA process that was developed by the German Federal Ministry of Research and Technology have considerable merit and should be considered further as viable alternatives to the current marble making machine concepts for a remotely operated and maintained process installation.

D.8.4 PNEUMATIC SYSTEM

An alternative that may reduce in-cell components but would increase cell height is the use of gravity instead of pneumatics to convey marbles from the marble making station to the surge hopper. This alternative is worthy of future consideration.

D.8.5 MATRIXING

As noted in Section D.6.15, for the purpose of this study, the lead-melting furnace is located outside of the process cell. An alternative would be to locate the furnace in the cell or to bring ladles of molten lead into the contaminated zone. This alternative may be worthy of additional study.

For this study, it is assumed that the metal matrix is lead or a lead alloy. Material availability and costs may preclude these from being viable materials in a production facility. Due to the softening temperature of the waste marbles, only a limited number of metals or alloys may be used; aluminum is a viable alternative that needs to be considered further. Other metals or alloys should also be evaluated.

D.9 DRAWINGS AND DATA SHEETS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

FIGURE D-4. Marble Machine

TABLE D-3. Equipment Data Sheet for the Vibratory Marble Machine

Function

Form molten glass stream into spherical-shaped marbles.

Interfaces

Ceramic melter; material classifier.

Design Parameters

Rate = 75 kg/h; specific gravity = 3.0; remote operations, monitoring, maintenance, removal and replacement.

Operating Conditions

Glass inlet temperature = $>1000^{\circ}\text{C}$; marble outlet temperature = $<500^{\circ}\text{C}$; mold temperature at entry = 115° to 135°C .

Features

Mold preheater; mold vibrators; track cooling; mold cooling; belt drive with speed control, belt tensioning, lubrication system and loose belt detection; temperature sensors.

Services or Auxiliaries

Compressed air; drain; ventilation; instrumentation.

Electrical: 10-kW capacity; average use rate = 2 kW.

Water: 400 L/min capacity; 100 L/min average use rate.

Space Requirements

3 ft wide by 18 ft long by 4 ft high

Maintenance Methods

Remote

Remarks

See drawings: SK1-78-112377, Concept Sketch HLW Glass Marble Making Machine and SK2-240-21078, Glass Marble Machine, Sheets 1-5.

*DIMENSIONS FOR REFERENCE
ONLY, INDICATES A GIVEN
PLANE.*

FIGURE D-5. Sorter

TABLE D-4. Equipment Data Sheet for the Sorter

Function

Separate acceptable marbles from scrap; provide analytical station input.

Interfaces

Marble machine; marble machine enclosure; sample system.

Design Parameters

Rate = 75 kg glass/h; specific gravity = 3.0; remote operations, monitoring, sampling, removal and replacement.

Operating Conditions

Process material inlet temperature = 500°C; ambient temperature = 25° to 135°C.

Features

Variable vibrator; table cooling.

Services or Auxiliaries

Compressed air; water; drain; electrical; instrumentation.

Space Requirements

2 ft wide by 3 ft long by 1 ft high

Maintenance Methods

Remote

Remarks

Similar to Cleveland Model Co. vibrator #50.

FIGURE D-6. Marble Machine Enclosure

TABLE D-5. Equipment Data Sheet for the Marble Machine Enclosure

Function

Provide controlled environment for marble machine; limit spread of process material.

Interfaces

Glass melter; scrap mill; pneumatic system; various services for internal equipment.

Design Parameters

Remote installation and removal; remote installation and removal of internal mechanisms.

Operating Conditions

Ambient temperature 25⁰ to 135⁰C; negative air pressure in reference to cell.

Features

Dust and air tight; provide for remote access to marble machine and classifier; heater for marble machine molds; remote service connection; provisions for marble machine and sorter services; means to clear scrap and dust from interior and into mill (vibrators, air slides).

Services or Auxiliaries

Compressed air; ventilation; water; drain; electrical; instrumentation.

Space Requirements

3 ft wide by 20 ft long by 6 ft high

Maintenance Methods

Remote

Remarks

TABLE D-6. Equipment Data Sheet for the Glass Mill or Crusher

Function

Reduce glass slag, marbles and ribbons to conveyable particle sizes.

Interfaces

Marble machine enclosure; pneumatic system.

Design Parameters

75 kg glass/h; remote operations, removal and replacement; receipt material less than 8 in. ring size; discharge material less than 1/8 in. maximum dimension; process material has specific gravity = 3.0 and 3 Mohs hardness.

Operating Conditions

Internal ambient temperature = 25^o to 135^oC; external ambient temperature = 25^oC; process material <500^oC.

Features

Stainless steel and abrasion resistant material; remote adjustments.

Services or Auxiliaries

Electrical: 5 kW capacity; 1 kW average use rate.
Instrumentation.

Space Requirements

2 ft by 3 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Similar to a Sprout-Waldron Koppers Co. impact mill.

FIGURE D-7. Pneumatic System

TABLE D-7. Equipment Data Sheet for the Pneumatic System

Function

Transport marbles; transport glass particles.

Interfaces

Marble machine enclosure; marble surge hopper; scrap collection hopper; scrap mill.

Design Parameters

Transport 75 kg glass/h; specific gravity = 3.0; particle hardness = 3 Mohs; temperature = 100^o to 500^oC; remote operations; in-cell components remote monitoring; removal and replacement.

- prime mover sized to meet maximum load requirements of system with oversized motor as allowance for additional loads imposed on system
- smallest pipe size(s) capable of carrying design capacity within pressure-drop constraint
- piping run routing to minimize line length and number of turns
- downward or inclined lines for process material to be avoided
- process material to enter through air locks.

Operating Conditions

Maximum inlet vacuum = 15 in. Hg; system pressure drop = 5 psi; solids-to-gas ratio range = 10 to 20; conveying velocity = 2500 to 5000 ft/min; terminal velocity = 3000 to 4000 ft/min.

Features

Positive displacement blower: variable speed; 2000 to 4000 rpm; 80 to 150 acfm @ 10 psia inlet and 16 psia outlet

Pipe: stainless steel; 1 to 2 in. in dia; mill run 2B interior finish; 10 to 20 ft sections; bend radius for particles 2 ft or greater and for marbles 4 ft or greater

Connectors: Cherry Burrell's I Line or similar type; remote flanged joints if required for rigidity

Filters: sintered 300-series stainless steel with short, pulsed air blow back

Line valves: full ported ball valves.

- Air flow with flow deviation control will be provided upstream of the initial process entry point for process transfer run.
- For closed-loop systems, a cooler will be provided to remove the heat added to the air by the compressive action of the blower. The temperature of the air stream at the process entry point will be approximately 100^oF.
- A surge tank will be provided upstream of the process entry points to dampen air flow pulsations.
- Pressure controls will be provided at the line inlet to assure a negative pressure in the transfer line such that 13.7 psia is not exceeded.
- Dry air (dew point 0^oC or lower) will be introduced to the conveying line at a negative pressure of about 13 psia and at an adequate linear flow velocity.
- Differential pressure will be monitored across all filter systems.

Services or Auxiliaries

Compressed air; conditioned air; drain; instrumentation.

Electrical: 10 kW demand; 4 kW average use rate

Water: 100 L/h demand, 40 L/h average use rate

Space Requirements

Maintenance Methods

In-cell components: remote

Remarks

Special design required for this installation.

FIGURE D-8. Marble Surge Hopper

TABLE D-8. Equipment Data Sheet for the Marble Surge Hopper

Function

Provide storage for marbles between continuous and batch operations of process.

Interfaces

Pneumatic transport system; volume hopper.

Design Parameters

Remote operations, removal and replacement; contains up to 300 L of marbles; stainless steel construction.

Operating Conditions

Temperature range = 25⁰ to 450⁰C; pressure range = 10 in. Hg vacuum to 25 psig.

Features

Load cells or level sensors; cooling coils; temperature sensors; pressure indicators.

Services or Auxiliaries

Instrumentation; water; drain; air or electrical.

Space Requirements

Maintenance Methods

Remote

Remarks

FIGURE D-9. Marble Volume Hopper

TABLE D-9. Equipment Data Sheet for the Volume Hopper

Function

Measures and limits batch size of marbles to be inserted into a canister.

Interfaces

Surge hopper; canister fill mechanism.

Design Parameters

Stainless steel construction; remote operations, removal and replacement; limit batch size to that required by a canister design.

Operating Conditions

Temperature range = 25^o to 450^oC; pressure is slightly negative to slightly positive.

Features

Load cells or level sensors; cooling provisions; temperature sensors; inlet valve is operated remotely.

Services or Auxiliaries

Instrumentation; compressed air; electrical; water; drain.

Space Requirements

Maintenance Methods

Remote

Remarks

FIGURE D-10. Canister Fill Mechanism

TABLE D-10. Equipment Data Sheet for the Canister Fill Mechanism

Function

Connects volumetric hopper with canister inner basket; directs and controls flow of marbles.

Interfaces

Volumetric hopper; canister.

Design Parameters

Remote operations, removal and replacement; stainless steel construction.

Operating Conditions

Temperature range = 25° to 450°C.

Features

Mate with canister; vertical movement.

Services or Auxiliaries

Vacuum; inert gas; compressed air or electrical.

Space Requirements

Maintenance Methods

Remote

Remarks

FIGURE D-11. Metal Fill Station

TABLE D-11. Equipment Data Sheet for the Metal Fill Station

Function

Feed molten lead to the canister

Interfaces

Canister; metal furnace

Design Parameters

Remote operation, removal and replacement

Operating Conditions

Temperature range = 25^o to 400^oC; pressure range 0 to 125 psig

Features

Mate with canister

Services or Auxiliaries

Electrical power; compressed air; hydraulics; inert gas; cooling water; drain; instrumentation; vacuum

Space Requirements

Maintenance Methods

Remote

Remarks

TABLE D-12. Equipment Data Sheet for the Lead-Melting Furnace

Function

Transform solid lead into the liquid form

Interfaces

Metal fill mechanism

Design Parameters

Operating Conditions

Temperature range = 0 to 450^oC

Features

Control of pouring rate and temperatures; slag-free pouring.

Services or Auxiliaries

Cooling water; drains; ventilation supply and exhaust; instrumentation
Electrical power: 120 kW demand; 30 kW average use rate

Space Requirements

Maintenance Methods

Contact

Remarks

Similar to an ASEA presspour type furnace

D.10 REFERENCES

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APPENDIX E

SUPERCALCINE PELLETS-IN-METAL MATRIX PROCESS

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APPENDIX E

SUPERCALCINE PELLETS IN METAL MATRIX PROCESS

E.1 INTRODUCTION

Another option for the inner core of a multibarrier waste concept is supercalcine pellets. Supercalcine is a crystalline assemblage of mutually compatible refractory and leach-resistant solid solution phases incorporating HLLW ions (Rusin et al. 1978). Supercalcine by itself is superior to ordinary calcine that is formed without additives. Supercalcine can also be used as the core for a composite, multibarrier waste form (Energy Research and Development Administration 1976). When supercalcine is formed into pellets, the pellets provide an alternative waste form to the glass monoliths produced in the ICGM and JHGM processes. The ICGM process is also referred to herein as the reference process.

As with glass marbles, the production of supercalcine pellets affords an opportunity for quality analysis and recycling of unacceptable product after spray calcining and before filling the canister. In addition, some pellet forms have a lower preparation temperature than glass (Lamb 1978). When incorporated in a metal matrix they produce a multibarrier product with improved thermal conductivity and lower internal temperatures than the product from the reference process. The production of the SCPIM form requires more complex operations than the reference process, which adds to the operating cost and risk. Future development of supercalcine pellets as a waste form will require the demonstration of supercalcine pellet production and encapsulation and the integration of these operations into an overall, remotely operable process.

The SCPIM facility will have the capability to 1) proportion and thoroughly mix supercalcine-forming chemicals into the HLLW, 2) calcine the solution to fix various ions in thermodynamically stable crystalline phases, 3) form the calcine crystals into small pellets that are then heat-treated, 4) load these into a canister, 5) fill the voids and annulus of the canister with lead, 6) seal-weld the canister, 7) check the integrity of the canister, and 8) decontaminate the canister and transport it to an interim storage facility before final disposition.

E.2 PROCESS DESCRIPTION

A flow diagram of the SCPIM process is shown in Figure E-1. As covered under the ICGM process (Appendix A), HLLW is received in the waste facility and is temporarily stored in a specially designed HLLW feed system. The composition of the basic HLLW is similar to that given in the Overall Process Bases Section. The HLLW is modified with selected liquid additives to form the supercalcine feed. The liquid additives, usually including Ca, Sr, Al and Si, are selected according to the waste composition and add from 35 to 45 wt% to the HLLW.

The spray calciner for the SCPIM process is the same as those for the ICGM process except that it has additional capacity, may be operated at a lower temperature and does not have frit feeding provisions. The supercalcine solution is jetted to the spray calciner. The liquid flow is pneumatically atomized at a controlled rate into a heated chamber, where granular crystalline calcine particles and an off gas are produced.

The supercalcine particles are fed through an air lock into a hopper where they are cooled. The particles then flow into a system that feeds the disc-pelletizer at a selected rate. Disc-pelletizing is an agglomeration technique that forms spherical pellets from powder particles (Floyd and Engelleitner 1967). Disc-pelletizing is an optimum method for compacting nuclear waste compositions when followed by sintering (Rusin et al. 1978). In this process the supercalcine particles are consolidated into high-density spheres, or green pellets, by the addition of binders and the rotating action of the inclined surface of the pelletizer.

The pellets from the disc-pelletizer are fed onto a classifier/separator where the fines, the oversized pellets and other wet scrap are separated for recycling back into the process stream. The green pellets within the desired size range are isolated, sampled and fed into the sintering furnace.

In the sintering furnace, the green pellets are brought up to and held at sintering temperatures and are then cooled at a controlled rate to drive off the moisture and other volatiles; thus, smaller, more dense and more durable pellets are formed.

After leaving the furnace, the sintered pellets are fed to another classifier where the pellets can be sampled; the fines and oversized globs are diverted and fed to a mill for crushing and recycling. Pellets of acceptable sizes are fed through an air-lock valve into the pneumatic transfer system.

A closed-circuit, negative-pressure pneumatic system with two loops is contemplated to 1) transport acceptable pellets to the canister surge hopper and 2) to convey scrap particles to the scrap collection hopper.

Canister surge hoppers, volume hoppers and fill mechanisms similar to those identified for the MIL process in Appendix D are used to convey a measured volume of pellets into the inner basket of the waste canister. Provisions are incorporated in the volume hopper to move unacceptable pellets via gravity to the mill for crushing and recycling. The solid scrap from the mill is fed into the pneumatic system through an air-lock valve and is conveyed to the scrap collection hopper where the material can be fed back into the calcine hopper.

NOMINAL OPERATING FLOWS INDICATED

FIGURE E-1. Supercalcine Pellets-in-Metal Matrix Process Flow Diagram

The canister containing pellets is handled and its annulus and voids are filled with molten lead as covered in the MIL process. After being filled with lead the canister is seal-welded, leak-checked, inspected, decontaminated and placed in storage as covered in Appendix A.

E.3 BASES AND ASSUMPTIONS

Most of the bases and criteria for the SCPIM process are noted in the Overall Process Bases Section. Data on the HLLW and its waste forms are also presented under the ICGM process (Appendix A). Other applicable assumptions and bases are identified in the following subsections.

E.3.1 PROCESS BASES

As noted above, the flow diagram for the SCPIM process is shown in Figure E-1. It is based on the solidification of HLLW produced in a fuel reprocessing plant operating at a rate of 2000 MTHM/yr. Design capabilities are 1.5 times the nominal operating values. Additional processing data is given below:

- Supercalcine feed to spray calciner
 - operating rate = 203 L/h
 - design rate = 305 L/h
 - HLLW loading = 78 wt%
 - heat loading = 2.7 W/L
 - oxide content (in HLLW and supercalcine formers) = 92.6 g/L
- Calcine
 - operating rate (oxide) = 18.8 kg/h
 - design rate (oxide) = 28.2 kg/h
 - operating rate (with 9.5% volatiles) = 20.8 kg/h
 - design rate (with 9.5% volatiles) = 31.2 kg/h
- Supercalcine pellets
 - specific gravity of lightly vibrated pellets = 0.78 kg/L
 - specific gravity of sintered pellets = 3.8 kg/L
 - heat loading of individual pellets = 111 W/L
 - bulk heat loading assuming 50% packing density = 55.5 W/L
 - wet scrap design rate = 4 kg/h
 - dry scrap design rate = 2 kg/h.

E.3.2 CANISTER

As noted in the Overall Process Bases Section, a major constraint is the allowable heat loading of the canister. This criterion permits the placing of approximately 55 L of waste in the supercalcine pellet form in a canister. To utilize a reasonable length-to-diameter ratio for process operations, lead filling and handling, a canister 30.5 cm in OD and approximately 183 cm tall (12 in. in OD by 6 ft tall) was selected for the SCPIM process. The canister and inner basket are shown in Figure E-2. The inner basket is positioned inside the canister before the flange is welded in place. Data on the SCPIM canister is also included in Appendix A.

E.7

FIGURE E-2. Canister and Inner Basket

E.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTES

For this study it is assumed that, with one exception, the effluents from the SCPIM process are similar in content and volume to the effluents from the ICGM process (Appendix A). The effluents associated with the operation of the metal-melting furnace are the exception. For process and maintenance simplicity, this furnace is located and operated outside of the process cell; therefore, its effluents are similar to those of a metal-melting furnace in a commercial installation and are not a major factor.

Except for more worn equipment due to the fact that the SCPIM process is more expanded and complex than the reference process, it is assumed that the wastes from the SCPIM process are of the same order of magnitude in volume and content as the wastes from the reference process. It is also assumed that systems and provisions are included in the fuel reprocessing plant to treat, reduce the volume and package both liquid and solid wastes. Finally, in the SCPIM process it is assumed that all solidified process scrap that may contain high-level waste is recycled back into the process.

E.3.4 UTILITIES AND MATERIALS

Estimates of the required utility services and essential materials are shown in Tables E-1 and E-2, respectively. These estimates are based on the requirements for the reference process with adjustments for additional process steps and anticipated space and volume changes.

TABLE E-1. Estimate of Utilities Needed for SCPIM Process

<u>Utility</u>	<u>Capacity Required</u>	<u>Average Use Rate</u>	<u>Disposition</u>
Electricity	1450 kW	850 kW	--
Raw water	1800 L/min	600 L/min	Recycle or environment
Process water	600 L/min	40 L/min	HLLW or ILLW
Sanitary water	40 L/min	20 L/min	Sanitary or chemical sewer-tile field
Air	750 scmm	500 scmm	As appropriate
Steam	25 kg/min	5 kg/min	As appropriate

TABLE E-2. Estimate of Materials Required for SCPIM Process(a)

Material	Amount Required Per Unit Time
AL(NO ₃) 9 H ₂ O	35,500 kg/yr
Ca(NO ₃) ₂	10,000 kg/yr
30% colloidal SiO ₂	62,500 kg/yr
Sr(NO ₃) ₂	5,500 kg/yr
Canisters	6.4/d
Caustic (19M)	10,000 L/yr
Nitric acid (12.2M)	12,000 L/yr
Helium sources	6.5/d
Argon	5,000 to 6,000 L/d
Detergent (for example Tide)	1,000 kg/yr
Ammonia	120,000 kg/yr

(a) Excludes maintenance materials and apparel.

E.4 FACILITY AND EQUIPMENT REQUIREMENTS

The SCPIM process is contained in a reinforced concrete structure similar to that shown in Figure E-3. The major pieces or equipment systems are listed below:

- calciner feed system
- calciner
- disc-pelletizer system
- sintering furnace
- pneumatic transfer system
- scrap recycling system
- marble hopper and fill system
- metal fill system
- canister handling system
- off-gas system.

The overall requirements for the waste facility and its support groups and services are covered in the Overall Process Bases Section. The major factors to be considered in process evaluations or cost estimates are the requirements for remote operations and for remote maintenance of all in-cell equipment.

FIGURE E-3. General Arrangement of SCPIM Waste Facility

E.5 STATE OF PROCESS DEVELOPMENT

The SCPIM process requires a number of process steps that have not been performed using wastes or simulated wastes, or on any basis near that required for a production facility. In addition, this process involves a number of involved mechanical/chemical steps that must be performed remotely with equipment that requires remote maintenance and remote removal when utilized in a production facility.

Considerable research, development and tests have been performed at PNL on spray calcination and spray calciners, and this effort has established a very good and firm basis for this major process component. These data can be applied to the SCPIM process.

Research on supercalcine was first initiated early in the 1970's at Pennsylvania State University. Many laboratory-scale tests and evaluations have been performed over the years on supercalcine (Rusin et al. 1978; McCarthy 1977). Small volumes of simulated waste in the uncoated supercalcine form have been encapsulated successfully (Rusin et al. 1978). Calcine pellets have been made from simulated HLLW at PNL as well as at other laboratories (Lamb and Cole 1979). The multibarrier concept of supercalcine pellets in a metal matrix has good potential for further development, because it offers increased ruggedness and inertness. However, there is also an increase in process and technological complexity. On the other hand, uncoated supercalcine pellets in lead offer enhanced inertness at a much lower level of technological complexity than glazed or chemical-vapor-deposition (CVD) coated supercalcine (Rusin et al. 1978).

Future development of supercalcine pellets as a waste form will require the demonstration of pellet production and encapsulation and the integration of these operations into an overall, remotely operable process. The radiation stability of supercalcine also needs to be determined.

E.6 UNIT OPERATIONS

The components of the SCPIM process have been grouped into 16 unit operations. In this section, information on the operation of each system and areas requiring further research and development are covered. Drawings and data sheets on the equipment and operations discussed in this section can be found at the back of this appendix in Section E.9.

E.6.1 HLLW FEED SYSTEM

The function of and requirements for the design and the operation of the HLLW or calciner feed system are covered in detail under the reference process (Appendix A). The HLLW feed system for the supercalcine process is basically the same as for the reference process except that it includes provisions to 1) introduce and incorporate supercalcine additives with the HLLW, 2) maintain the proper solution, 3) sample and analyze the feed solution, and 4) contain the larger volume that is required.

After the supercalcine additives (which may be solutions of calcium and aluminum nitrate and an aqueous colloidal suspension of silica) have been added to formulate the desired feed, the other preparations (verification, analysis, start-up, transfer, operation and shut-down) for the SCPIM calciner feed are similar to those covered in the reference process. Parameters for the supercalcine HLLW feed system are noted in Table E-3 in Section E.9.

The basic HLLW feed system has been exposed to considerable tests and developmental programs, and its requirements and parameters have been well established. However, considerable efforts are required to identify the requirements and parameters for a particular supercalcine feed system. Some of the specific areas of concern are covered in detail in Section E.7.

E.6.2 SPRAY CALCINER

The function, design and operation of the SCPIM spray calciner are basically the same as for the reference process spray calciner (Appendix A). The SCPIM process does not require a frit feeding system but does require an additional capacity of about 28% over the combined capacity of the two spray calciners in the reference process.

Supercalcine feed is introduced to the heated spray chamber through an air-atomized spray nozzle. The volume of radioactive waste is significantly reduced when converted from a liquid to a powder. The liquid waste is dried and converted to oxides and reaction off gases. Calcine collects at the bottom of the calciner after falling through the chamber or after being removed from the off-gas stream by sintered filters.

The research and development efforts noted in the reference process for the spray calciner are also required for the optimization of the SCPIM process. The spray calciner for the supercalcine process also requires the following efforts to resolve and/or determine concerns and unknowns:

- test a larger volume feed system

- test calciners with supercalcine solutions to determine such items as probability of sintered buildups
- establish and test methods of detecting and removing sintered buildups (water sprays, temperature cycling, vibration, etc.)
- establish operating parameters and limits for supercalcine
- evaluate effect supercalcine additives may have on catalyzing an explosion from tramp impurities, and if effect is positive establish safe alternatives
- evaluate the effect of supercalcine additives on the volatility of species such as cesium and ruthenium and thereby determine off-gas processing requirements.

E.6.3 OFF-GAS SYSTEM

The requirements for and the function and operation of the SCPIM process off-gas system are the same as for the ICGM off-gas system. The ICGM process has an off-gas system for each spray calciner; for operating efficiency, two parallel systems may also be desirable in the SCPIM process, although there is only one spray calciner. The capacity of the SCPIM process off-gas system is about 30% greater than that of the reference process. The research and development efforts specified in Appendix A are also required.

E.6.4 CALCINE HOPPER AND PELLETIZER FEEDER STATION

A remotely operated air lock is located below the spray calciner and above the calcine hopper to minimize air flows through the calcine feeder system. The air-lock valve is self-sealing, has cooling provisions and is operated by a variable-speed drive so the supercalcine particles can normally be fed into the hopper at the same rate in which they are formed.

The operation of the air lock is relatively simple and is coordinated with the operation of the spray calciner and the pelletizer feeder. The cooling provisions can be controlled to protect the valve and to assist in the reduction of the supercalcine temperature. A spray-flush system removes calcine buildup and cleans the valve internally. Design and operating conditions for the air-lock valve are indicated in Table E-4 in Section E.9.

A relatively tall and very narrow, water-cooled stainless steel hopper, with a surge capacity for a couple of hours of spray calciner operation, is the collecting media for the spray calciner product. Here the supercalcine is cooled to below 100°C, and a uniform supply of particles is provided to the pelletizer feeder. If the surge hopper is full, the processes upstream must cease, and when the supply of particles in the surge hopper is depleted, the operation of the pelletizer should be stopped. The operation of the hopper mainly involves monitoring such items as the level, flow and temperature of the contents, and the internal pressure. Table E-5 in Section E.9 is the equipment data sheet for the calcine hopper.

The pelletizer feeder is the third component of this station. It provides a consistent feed to the disc-pelletizer at the selected rate. In addition, the feeder and the feed prevent large volumes of air movement either into or out of the surge hopper. The start-up,

operation and shutdown of the feeder must be coordinated with that of the disc-pelletizer. Feed flow and temperature must be monitored and maintained within established limits. Data on the pelletizer feeder are given in Table E-6 in Section E.9.

Air-lock devices consisting of flapper, star or rotary valves are available commercially and some of these may perform adequately and withstand the operating temperatures of the SCPIM process. However, none of these commercial valves have been designed for remote maintenance and replacement. Research and development are required to design a valve for a radioactive, remotely operable facility. Once it is designed the valve should be operated in conjunction with a spray calciner to verify the valve design. For example, the following should be tested: its operation with calcine particles, provisions for remotely operating and maintaining it, and methods for cleaning or flushing the valve. In addition, the developmental efforts described in Section E.7 are required for the pelletizer feeder.

E.6.5 DISC-PELLETIZER STATION

As indicated above, the supercalcine is fed to the disc-pelletizer by a feeder. The liquid spray is fed into the pelletizer by the system depicted in Figure E-4 (see Section E.9). The liquid binder is responsible for the strength of the green pellets. It fills the pores and binds adjacent particles together either by forming chemical bonds or by capillary action. The binder also acts as a lubricating medium to obtain the densest particle packing configuration possible for that material. When the calcine, or powder, is somewhat soluble in the liquid binder, the strength of the dry pellets is improved; upon drying, the soluble components form interlocking, crystalline networks between adjacent particles.

The pelletizer consists of a rotating, inclined pan into which powder and liquid binder are fed at controlled rates. The powder initially forms small pellets that are shaped and compacted by collisions with other pellets and with the pan. These pellets then grow in a snow-balling fashion as more powder and binder are added. The net result is a dynamic, equilibrium condition where pellets are continuously discharged as more pellets are formed. The size of the pellets is determined by the materials used and pelletizer operating conditions.

The stable operation of the disc-pelletizer to produce acceptable-quality pellets of a desired size is a function of:

- feed powder characteristics
- feed powder-binder interactions
- liquid binder properties
- pelletizer operating parameters.

These variables must be maintained within reasonable limits to maintain process equilibrium. In particular, the inherent sensitivity of a small pelletizer requires precise control of the major parameters: the powder feed rate, the binder feed rate, and the powder-to-binder ratio. Unanticipated changes in any of these will have a direct impact on the size and strength of the green pellets. Table E-7 in Section E.9 lists other attributes of the pelletizer.

The second major component of the pelletizer station is a classifier/seperator where the green pellets are screened to remove and recycle loose powder and oversized pellets. The

pelletizer and classifier/separator system contains dust removal or suppression provisions to maintain process cleanliness with minimal material loss and contamination spread.

As with the calcine hopper-feeder station, nitric acid spray-flush provisions are incorporated into this station. All dust, under- and oversized pellets and products from spraying and flushing drain into a wet scrap vessel where these materials can be recycled to the calciner feed system.

Also incorporated into this station is the means of assessing green pellet quality on a semicontinuous basis. Since the pelletizer station is one part of essentially a continuous process, its overall operation has to be coordinated with others up- and downstream. The limited operating parameters such as binder-to-powder ratio necessitate precise process control and sampling systems.

One of the positive aspects of the SCPIM process is that it affords the opportunity to monitor and analyze the quality of the waste form before it is converted into a form where corrections or adjustments cannot be made. The exit at the pelletizer is one of the logical places in this process to monitor and analyze product quality.

An enclosed and remotely operated and maintained classifier/separator to separate powders and oversized material from suitable pellets and to handle the green pellets without damaging them needs to be designed, developed, fabricated and tested in conjunction with a pelletizer. A system capable of monitoring such items as flow rate, quality, strength, etc. of the pellets also needs to be devised and implemented. This may include a remote sampling system. Finally, the positive or negative impact that the radioactive decay heat may have on design parameters of systems handling green pellets should be assessed.

A disc pelletizer has been operated at PNL on a limited basis with some very positive results. This effort needs to be continued to resolve problems and to determine operating parameters as noted in detail in Section E.7.

Additional data for the classifier/separator and the wet scrap tank are shown in Tables E-8 and E-9 (see Section E.9), respectively.

E.6.6 SINTERING FURNACE

The pellets that are within an acceptable size range are fed from the classifier/separator to a vertical sintering furnace where they may be mixed with a granular, carrier medium to prevent pellet damage during movement through the furnace. The carrier is a free flowing, high-temperature material that is compatible and inert with the supercalcine pellets; alumina (Al_2O_3) may be such a material.

The mixture of pellets and carrier are fed in at the top of the sintering furnace. The downward movement of the mixture through the preheating, sintering and cooling muffles is controlled by the exit feeder or valve at the furnace base. If necessary, an atmospheric gas or gases can be introduced at the base of the furnace with enough pressure to overcome the pressure drop across the moving bed of supercalcine pellets and carrier; thus, there is intimate contact between the pellets and the gas for proper sintering control.

As with other stations in the system, the sintering furnace requires a relatively long period of time to bring it up to operating temperatures and conditions. Exact operating procedures are dependent on the final furnace design, but the following general start-up procedures are required: 1) introduce a flow of atmospheric gas to purge the furnace components, 2) turn power on to the furnace windings and gradually raise temperature (at 100°C/h) to 500° to 600°C , 3) hold at this temperature for 5 to 10 h to bake all moisture from the furnace insulation, 4) raise furnace temperature at 300°C/h to about 100°C above the desired operating temperature, 5) as the furnace temperature approaches 1000°C , start water through the cooling coil and start exit feeder to initiate carrier flow, and 6) when the maximum temperature has been achieved, reduce the controlled temperature to the desired operating temperature and allow it to stabilize with the carrier grain moving through the furnace.

When equilibrium conditions have been achieved in the preheating, sintering and cooling muffles, the furnace is ready to accept the flow of supercalcine pellets. The furnace throughput, the pellet-to-carrier ratio and the operating temperatures all require monitoring and controlling during operation. The furnace operation must be integrated with the pelletizer upstream and the separators and pneumatic systems downstream. Shutdown is basically the reverse of the start-up procedures.

Means of separating carrier from pellets and recycling the carrier to the top of the furnace are integral components of the sintering furnace station. After separation from the carrier, the sintered pellets are fed to another classifier/separator. The sintering furnace is shown in Figure E-5 (see Section E.9), and additional data on the furnace are given in Table E-10 (Section E.9).

A research, development and design program, as outlined in Section E.7, for the vertical sintering furnace is required to assure the availability of a production-scale furnace that can be used in a radioactive installation and that can be maintained or removed remotely.

E.6.7 CLASSIFIER/SEPARATOR

The sintered supercalcine pellets are fed via gravity into a classifier/separator where the fines and the oversized material are separated and fed to a mill via gravity. The pellets within an acceptable size range are fed through an air lock to the pneumatic system. Incorporated in this station are the means to monitor the temperature and weight of the pellets. There is also a sampling system where individual pellets can be removed from the process stream and sent to an analytical station or laboratory to check their shape, composition and chemical or physical characteristics.

The operation of this station must be started before any pellets leave the sintering furnace and must be continued until the pellet flow ceases. It must also be coordinated with the operation of the pneumatic system and the mill. Table E-11 in Section E.9 gives additional design and operating conditions for this classifier/separator. The research and development work identified above for handling and sampling green pellets is also applicable to sintered pellets.

E.6.8 MILL

Any scrap consisting of powders, small or broken pellets and pellets fused together is fed into a vertical mill or crusher via gravity or vibrators. The mill is operated on an intermittent or as-needed basis to reduce the supercalcine scrap material into particle sizes that can be conveyed pneumatically and recycled back into the process stream. During operations, a means of monitoring the flow of material into or out of the mill is required. Data on the mill are noted in Table E-12 in Section E.9. Essentially, the same research, designing and testing outlined for the remote mill in the MIL process in Appendix D are required for a mill in this process.

E.6.9 PNEUMATIC SYSTEM

Transport of radioactive materials should be via negative pressure. As in the MIL process, the prime mover is a rotary, positive-displacement type blower, which provides a vacuum to the receiver vessels via blowback filters. Process material is conveyed through short sections of 300-series stainless steel piping; the short sections permit remote removal of the piping. Pipe connectors limit protuberances at the joints. Process material enters the system through feeder control valves. Exhaust air is removed from the top of the systems' vessels through sintered, stainless steel filters that can be blown back with air pulses. Load cells on the receiving hoppers and/or flow indicators in the system are used to monitor system operation and cleanout.

The pneumatic system must be operating when the sintered pellet separator/classifier is operating and must remain operating until all of the pellets have cleared the separator. When it is necessary or desirable to convey scrap particles to the collection hopper, the pellet loop is valved off and the pneumatic system is started before starting the mill.

For a vacuum and/or closed system, it is necessary for the process material to be introduced into the system through air-lock devices. The design of these air locks affects pressure losses and drops as well as process material inertia and accumulation. The research, development and testing outlined in the MIL process (Appendix D) are also applicable to a pneumatic system to convey supercalcine pellets.

A schematic of a pneumatic system is shown in Figure D-7 (Section D.9 in Appendix D). Additional data for SCPIM pneumatic transfer system is given in Table E-13 in Section E.9.

E.6.10 RECYCLED FEED

The final receiving vessel for the pneumatic system is the recycled scrap collection hopper located in the upper portion of the process cell. Solid process scrap material is stored in this stainless steel hopper. The collected material is eventually recycled back into the process stream at the calcine hopper through a weigh feeder and gravity pipes.

The feed system for recycled scrap is similar to the glass frit feed system covered under the ICGM process (Appendix A). It is necessary to monitor and measure all material flows and coordinate these with the calcine hopper and pelletizer feeder operations.

E.6.11 PELLET SURGE HOPPER

A cylindrical-shaped, stainless steel vessel is the collecting point for the supercalcine pellets from the pneumatic system. The surge hopper is located high in the cell so gravity can be used for subsequent operations with the pellets. The flow of pellets out of the surge hopper and into the volume hopper is controlled by a valve that can close through a column or stream of pellets. The surge hopper is equipped with cooling coils, temperature sensors and level or weight devices to monitor and control the contents.

At this point in the process, the material flow is converted from a continuous to a batch type operation. If the surge hopper is full, operations upstream must be stopped. The level and weight of the pellets in the surge hopper and the internal temperature must be monitored during operating periods. Provisions are incorporated to maintain the temperature of the contained pellets below a prescribed limit, which is below the highest allowable temperature of the supercalcine. Other data on the pellet surge hopper are given in Table E-14 in Section E.9.

E.6.12 VOLUME HOPPER

The volume of this stainless steel hopper, including its inlet connection to the entry valve, equals the volume to be filled by supercalcine pellets in the inner basket of the storage canister. The valve between the surge hopper and volume hopper is normally open until the proper amount of pellets are in the volume hopper. Like the surge hopper, the volume hopper is equipped with cooling coils, temperature sensors and level or weight sensors. The flow of pellets out of the volume hopper is controlled by the canister fill mechanism.

The operation of the volume hopper mainly involves the proper sequencing of the upper inlet or feed valve, assuring of the proper fill volume, drainage through the canister fill mechanism, and assuring that all of and only the measured volume has been drained through and out of the vessel. Temperatures must be monitored anytime process material is in the hopper so that cooling can be initiated if necessary. Supporting data for the volume hopper are given in Table E-15 in Section E.9.

E.6.13 FILL MECHANISM

After an inspected canister with an inner basket is on the canister transfer system and is located under the pellet fill station, the fill mechanism is inserted into the canister. When a vacuum system is started to remove dust and particles from the falling pellets, the transfer of supercalcine pellets from the volume hopper to the inner basket of the canister is begun. During and after filling, inert gas is introduced into the center pipe of the canister via the fill mechanism to cool the pellets if required and to help entrain dust into the vacuum system. After filling is completed, the gas and vacuum systems are valved off and the fill mechanism is retracted from the canister. Additional data on the fill mechanism is given in Table E-16 in Section E.9.

The crucial batch operation in the SCPIM process is the remote filling (and monitoring) of the inner basket in the canister with the proper amount of pellets on a repetitive basis

with no over- or underfills. An engineering design should be made for a remote, production-sized system that includes the complete measuring system and the canister fill mechanism. A full-scale model should then be fabricated and tested in a laboratory demonstration. This demonstration would include the complete, remote operation and monitoring of the system as well as the verification of the remote maintenance and replacement aspects.

E.6.14 INNER BASKET CLOSURE

After the inner basket has been filled with the proper amount of supercalcine pellets, the canister is transferred to a station where a device is inserted into the canister to close the opening in the top of the inner basket. To allow for long periods of time between filling and lead pouring and to prevent the radioactive decay heat from causing the pellets to soften or melt, provisions to cool the canisters while in this or subsequent stations are provided. Upon retraction of the closure device, the canister is transferred to the next station.

E.6.15 METAL FILL STATION

Next, the canister is lifted into the canister heater and up against the metal fill mechanism. Provisions are included to cool (via an internal gas flow) and/or heat (canister reactors) the canister and its contents, as required to maintain a temperature within 50°C of the melting point of the metal alloy to be used (300° to 400°C). When this equilibrium temperature has been reached, the cooling gas flow is terminated, the canister heaters are turned off and the valve is opened to start the metal pouring from the furnace. The liquid metal is introduced through the center pipe to the bottom of the canister and back up through the pellets and the outer annulus. A vacuum can be induced in the canister through the metal fill mechanism, if necessary, before and during pouring of the metal. The metal is poured continuously until the desired level is reached, and then the valve is closed. During the metal pouring process, the bottom and possibly the walls of the canister are cooled to control lead shrinkage voids.

The canister is then lowered approximately 1 ft, the metal-flow control valve is re-opened and the lead is permitted to reach the new desired level. As the lead level drops due to the 5% shrinkage, additional metal is added as required to complete the filling of the canister to the selected level. Then the canister is lowered and removed from under the metal fill station, and the metal level is checked.

The metal-melting furnace is located out of but adjacent to the process cell in a service area, so the molten lead can flow by gravity through a heated entry tube to the metal fill mechanism. With this out-of-cell location, the furnace, its controls and its operation are basically the same as for a commercial furnace. Heating and controlling the temperature in the entry tube is a special application as is the control of its isolation or block valves. Additional data on the metal fill station and the metal-melting furnace are given in Tables E-17 and E-18 (see Section E.9), respectively.

This study assumes that the metal-melting furnace is located outside of the process cell to reduce in-cell components and complexity. However, an engineering and design program is

required to determine if this assumption is the best approach and to design the metal-pouring system, especially the in-cell components. When this design is completed or identified, inter-cell and in-cell components should be fabricated and installed in a demonstration mode. Then the operation of a remote, metal-filling mechanism, including metal-flow control and monitoring, could be demonstrated and evaluated. The remote maintenance and replacement aspects could also be verified.

E.6.16 CANISTER PROCESSING

After the proper metal level is assured, the canister is moved into the weld station, a helium source is inserted and the lid is seal-welded. Next, the canister weld is helium leak-checked to ensure its integrity. The canister is also visually inspected. Canisters that fail to pass the leak test are rewelded, if possible. Sealed canisters are then moved to a decontamination chamber where any loose contamination is removed. From here the canisters are placed in the in-cell storage racks or are transferred out of cell to an interim storage area. These canister treatment steps are covered in more detail in Appendix A.

E.7 RESEARCH AND DEVELOPMENT REQUIRED

There is a definite need to demonstrate all of the remote aspects of each unit operation in a cold mode. This need also applies to the complete SCPIM process, and it is recommended that following or in conjunction with component tests the system be operated with simulated supercalcine material. These demonstrations could also be used to verify the arrangement of the equipment in the cell.

Although testing the overall system is an important aspect in any remote, nuclear installation, it should not be done before the requirements and needs of the unit operations have been identified. For the SCPIM process, additional efforts should be applied first to 1) identifying supercalcine feed system parameters, including sampling and analytical procedures, 2) evaluating supercalcine radiation stability, 3) resolving operational problems with a disc-pelletizer and its feed systems, 4) establishing design and operating parameters for the sintering furnace, 5) identifying design requirements for a pneumatic system to carry nuclear wastes in the supercalcine pellet form, 6) identifying the basis for remote, pellet sampling and monitoring systems, and 7) establishing a system to remotely fill canisters with lead.

E.7.1 HLLW FEED SYSTEM

The basic HLLW feed system has undergone considerable development and evaluation, and its operating and design requirements are well established. Remaining areas of concern are covered in Appendix A. Other areas of concern that must be resolved to assure consistent consolidation of crystalline ceramics for a supercalcine feed system are:

- amounts and kinds of solutions required to formulate supercalcine compositions
- parameters and controls for adding and mixing supercalcine solutions to HLLW (proportionate and redundant metering, agitation and control of temperature, acidity and gelation)
- effects of radiation on supercalcine solutions
- accuracy required for HLLW sampling and analysis
- effects of recycle streams.

E.7.2 CALCINE HOPPER AND FEEDER

The calcine hopper and feeder have been operated at PNL in conjunction with the operation of a disc-pelletizer (Rusin et al. 1978). This experience has indicated the following problems that must be resolved:

- Calcine must be cooled to well below 100°C before it is fed to the disc-pelletizer to avoid steam formation. As a result, a design program should be instigated to determine the amount of heat contained in the supercalcine particles due to calcination and due to

radioactive self-heating. In addition, a design of a remotely operated and maintained production-sized hopper feed arrangement capable of removing this heat without impeding the flow of calcine should be developed and reliability-tested under all operating conditions, including cleanout and maintenance.

- Systems to measure calcine level and flow rate on a continuous basis need to be devised, implemented and tested.
- Calcine must be fed to the disc-pelletizer at a controllable and uniform rate. There are several types of feed systems that may be applicable to this installation. A vibratory-screw type system has been used at PNL with limited success. In conjunction with the first item listed above, long-term tests should be performed to assure that precise control ($\pm 1\%$) of the fine particle feed can be achieved over the range of operating parameters, such as hopper-to-pelletizer pressure differential, calcine temperature differences, hopper feed depth and variations in calcine bulk densities.

E.7.3 DISC-PELLETIZER

In operations at PNL, stable continuous operation of a disc-pelletizer at low feed rates was not possible to achieve (Rusin et al. 1978). Thus, continuing efforts are required to resolve the following concerns or problems:

- designing, fabricating and testing of a remotely operated and maintained disc-pelletizer, whose rate capacity is compatible with that of the supercalcine system
- developing and establishing supercalcine pelletizing parameters
- limiting and controlling dusting during pelletizer operation
- adequate monitoring and controlling of powder feed rate, binder feed rate and powder-to-binder ratio
- establishing binder options and compositions for powder temperature ranges and different operating conditions
- determining impact radioactive particle decay heat may have on uniform operation and pellet quality
- developing method to remove and clean mud and dried buildup from the pelletizer and its component parts
- developing an adequate pellet sampling system and procedure.

Pellets made with just water as a binder may not have the most desirable properties. Additional research and development needs to be performed on various combinations of solid and liquid binders.

E.7.4 SINTERING FURNACE

A small, cylindrical vertical furnace designed to sinter bulk quantities of highly radioactive fuels in a controlled and reducing atmosphere has been developed and is undergoing

proof-of-principle tests at PNL. This furnace has power concentrated in a small area, excellent heat transfer to the product, low heat losses and a high throughput capacity. This ongoing work has indicated that this furnace is the only type with a number of attributes that are either desirable or necessary for remote operations and maintenance; commercially available, horizontal-type furnaces are not applicable to remote maintenance. The horizontal types require a large space, and involved mechanical systems are required to move material through the furnace. Therefore, it appears prudent to proceed with a development, design and test program to establish a remote, vertical furnace capable of sintering supercalcine pellets. This program could take advantage of the efforts performed to date for nuclear fuel pellets and use that prototype furnace for tests such as the following:

- cold tests to determine carrier grain size and composition, pressure drops versus grain size and pellet-to-carrier ratios, carrier-to-pellet ratio needed to protect pellets, and pellet/carrier flow characteristics
- tests of methods to separate carrier from pellets and to recycle carrier
- hot thermal tests to evaluate pellet/carrier compatibility and flow characteristics under sintering conditions, assess and eliminate pellet damage, establish preheat, sintering and cooling rates or times, and determine gas compositions required
- other thermal tests to determine impact of various moisture content of incoming pellets and the operating requirements and impacts if binders other than water are required or used.

Other tests and developmental efforts will be required to evaluate and determine the consequences if elements such as cesium, sodium or ruthenium volatilize during the sintering process and are condensed or deposited. In addition, the tendency for gas flow, opposite to the gravity flow of the pellets, to condense out volatiles on the incoming cold pellets and carrier is an area of concern that must be checked out and resolved if a problem. Finally, carrier and furnace contamination and cleanup needs to be evaluated and resolved.

When the above mentioned tests are completed, a production-sized furnace designed for remote operation, maintenance and replacement should be fabricated and operated in a facility where all aspects of the remote operations and maintenance can be verified before exposing the system to a radioactive environment.

E.7.5 CANISTER FILLING AND HANDLING

The research and test efforts identified under the MIL process (Appendix D) for canister filling and under the reference process (Appendix A) for canister handling and closure will also be required for the SCPIM process. In addition, selected components and aspects of the SCPIM canister handling system should be fabricated and tested to verify their capabilities and remote aspects.

E.8 ALTERNATIVE PROCESSES AND EQUIPMENT

Only a very limited time has been available to develop the preconceptual design bases that are presented in this document for the SCPIM process. As such, there are a number of viable alternatives that have no doubt not been considered or that should be explored further in the subsequent steps of finalizing a process design. Some of the alternatives noted in the reference process may also be applicable to the SCPIM process, particularly those pertaining to the spray calciner. Other alternatives are noted below.

E.8.1 CELL ARRANGEMENT

The cell arrangement displayed in Figure E-3 shows all of the remote processing equipment located in one cell or canyon that is quite high and long. There is a strong possibility of lowering building costs if this size could be reduced; cell height is of particular concern. In addition, to limit and control the potential spread of contaminants, it may be desirable to locate the process equipment in two or more cells. This alternative needs to be considered while at the same time giving adequate attention to the flow of process material and the requirements for the remote operation, maintenance and replacement of equipment.

E.8.2 DISC-PELLETIZER

As covered in Section E.7.3, calcine must be fed to the pelletizer at a controllable and uniform rate. Tests with a vibratory-screw feeder have indicated considerable variances in feed rate. Other feeder types, such as gravimetric and loss-of-weight, were considered but have not been used to date because preliminary evaluations indicate more serious control or remote operational problems with these. Calcine feeder alternatives need to be evaluated further early in the research and development program.

Systems other than a disc-pelletizer that have or could be used to make supercalcine pellets include stirred-bed calciners, fluidized-bed calciners and pellet presses of different kinds or configurations. Early in any additional efforts on the SCPIM process, these alternatives should be considered in terms of their possible application in a remote, production-scale operation.

E.8.3 SINTERING FURNACE

The application of microwave energy to dry pelletized HLLW has been tested (Priebe et al. 1979). Preliminary tests indicate that hard, unbroken pellets of at least some compositions can be produced in 1/10 to 1/3 of the time required in a radiant type furnace. Additional tests and evaluations are planned at Idaho National Engineering Laboratory. These tests should be followed on a continuous basis, and if the advantages appear to outweigh the disadvantages, due consideration should be given to developing this process as an alternative to the vertical sintering furnace.

E.8.4 PNEUMATIC SYSTEMS

An alternative that may reduce in-cell components, but would increase cell height is the use of gravity instead of pneumatics to convey pellets from the sintering furnace station to the surge hopper. This alternative should be evaluated further.

E.8.5 MATRIXING

As noted in Section E.6.15, for the purpose of this study the metal-melting furnace is located outside of the process cell. An alternative would be to locate the furnace in the cell or to bring ladles of molten lead into the contaminated zone. This may be worthy of additional study.

For this study, it is also assumed that the metal matrix is lead or a lead alloy. Material availability and cost may preclude these from being viable materials in a production facility. Due to the softening temperature of the waste pellets, only a limited number of metals or alloys may be applicable. The use of aluminum is a viable alternative that needs to be considered further; other metals or alloys should also be evaluated.

E.9 DRAWINGS AND DATA SHEETS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

TABLE E-3. Equipment Data Sheet for Supercalcine HLLW Feed System

Function

Receive, store and analyze HLLW; prepare calciner feed solution and deliver to spray calciner.

Interfaces

HLLW supply; chemical make-up; spray calciner.

Design Parameters

Remote operation, removal and replacement; stainless steel construction; vigorous agitation systems; handle soda-rich solutions; accurate analysis of solutions; intertank transfer to and from all tanks; two storage tanks; one feed tank.

Operating Conditions

Same as reference process except tank capacities are 125% of reference process.

Features

Same as reference system except more accurate HLLW sampling and analytical systems, supercalcine addition system and thorough mixing systems.

Services or Auxiliaries

Same as reference process.

Space Requirements

Maintenance Methods

Remote

Remarks

TABLE E-4. Equipment Data Sheet for Air-Lock Valve

Function

Isolate hopper and spray calciner environments.

Interfaces

Spray calciner; calcine hopper.

Design Parameters

Self-sealing between entrance and exit or purgable; radiation resistant; remote operation, installation and replacement.

Operating Conditions

Temperature = 150° to 250°C; 10 to 60 rpm; 10 to 60 kg/h of calcine.

Features

Stainless steel construction; variable-speed drive; sealed closed-loop cooling; spray-flush system.

Services or Auxiliaries

Electrical; rpm monitor; temperature sensors; compressed air; nitric acid; water; drain.

Space Requirements

2 ft by 3 ft by 1 ft high

Maintenance Methods

Remote

Remarks

Special design but similar to: The Young Industries Inc. rotary valves; Wm. W. Meyers and Sons, Inc., Roto-Flo air-lock feeder valves; Sprout-Waldron Koppers Co., Inc. modular rotary valves; Joy Manufacturing Co., air-lock or dust valves.

TABLE E-5. Equipment Data Sheet for Calcine Hopper

Function

Provide cooled and uniform feed supply.

Interfaces

Air-lock valve; calcine feeder.

Design Parameters

Stainless steel construction; remote operation, replacement and installation; 2-h surge capacity (2 ft³).

Operating Conditions

Temperature = 75^o to 90^oC; flow rate = 30 kg/h.

Features

Vibrators; closed-loop cooling; spray-flush system.

Services or Auxiliaries

Water; drain; air; electrical; temperature; detectors; weight or fill detectors; pressure sensors; nitric acid.

Space Requirements

6 in. by 24 in. by 48 in. high

Maintenance Methods

Remote

Remarks

Special design for this application and selected feed system.

TABLE E-6. Equipment Data Sheet for Pelletizer Feeder

Function

Provide consistent feed rate to disc-pelletizer.

Interfaces

Calcine hopper; disc-pelletizer.

Design Parameters

Stainless steel construction; radiation resistant; automatic and remote operation; remote installation and replacement; resistant to nitric acid wash-down.

Operating Conditions

Temperature = 75^o to 90^oC; 0 to 100 kg/h calcine;

Calcine Properties: shape--agglomerated semi-spherical particles;

particle density = 2.8 g/cm³ (typically approximately 4 g/cm³);

bulk density = 0.5 to 1.3 g/cm³ (typically 1.2 g/cm³);

particle size = 1 to 10 μ ;

particle hardness is approximately 5 to 7 Mohs.

Features

Variable speed; flow detection; spray-flush system.

Services or Auxiliaries

Electrical; rpm monitor; temperature detectors; nitric acid; compressed air.

Space Requirements

2 ft by 4 ft by 2 ft high

Maintenance Methods

Remote

Remarks

Special application that adapts Vibra Screw Inc. screw feeder; K-TRON Corp. feeder; Acrison, Inc. feeder; Merrick Scale Mfg Co. Flow Star feeder or similar device.

TABLE E-7. Equipment Data Sheet for Pelletizer

Function

To form calcine particles into larger pellets or spheres.

Interfaces

Calcine feeder; separator.

Design Parameters

Enclosed operations; stainless steel construction; automatic and remote operation; remote replacement and removal; design rate = 30 kg/h.

Operating Conditions

Temperature = 35^o to 90^oC; rate = 0 to 50 kg/h;

Pellets: shape is spherical 1/8 to 1/4 in. in dia; density = 0.8 to 1.5 g/cm³; moisture content is approximately 30%.

Features

Variable speed; binder feed; variable angle; variable nozzle locations; spray-flush system; sealed enclosure; rate monitor.

Services or Auxiliaries

Electrical; pressure sensor; temperature sensors; speed and position indicators; nitric acid; compressed air; ventilation supply and exhaust.

Space Requirements

2 ft by 2 ft by 2 ft high

Maintenance Methods

Remote

Remarks

Special adaptation of small, commercial pelletizer such as Ferrotec Inc. 16-in. pelletizer; Sprout Waldron Koppers Co., Inc. Model 400, or smaller models.

TABLE E-8. Equipment Data Sheet for the Green Classifier/Separator

Function

Separate large and undersized material from acceptable spheres.

Interfaces

Disc-pelletizer; enclosure.

Design Parameters

Stainless steel construction; automatic and remote operations; remote replacement and installation.

Operating Conditions

Temperature = 35^o to 90^oC; rate = 0 to 50 kg/h; fines are less than 1/8 in.; oversized particles are greater than 1/4 in.; product is 1/8 to 1/4 in. in dia.

Features

Remote adjustments; spray-flush system.

Services or Auxiliaries

Electrical; compressed air; sample station; nitric acid; instrumentation.

Space Requirements

3 ft by 4 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Special installation that will require a device similar to: SWECO Inc. Model LS24544 separator; Eriez Magnetics Model 361 separator; or Sprout-Waldron Koppers Co., Inc. classification machines.

TABLE E-9. Equipment Data Sheet for the Wet Scrap Tank

Function

Collect wet scrap and spray-flush solutions.

Interfaces

Spray calciner; calcine hopper; disc-pelletizer station; HLLW feed system.

Design Parameters

Stainless steel construction; automatic and remote operations; remote installation and replacement.

Operating Conditions

Temperature = 25^o to 100°C; nitric acid solutions.

Features

Closed-loop cooling; agitation system(s); jet transfer; specific gravity; weight factor; temperature pressure sensors.

Services or Auxiliaries

Steam; air; electrical; water; drain; instrumentation.

Space Requirements

3 ft in dia by 3 ft high

Maintenance Methods

Remote

Remarks

Special design required.

FIGURE E-5. Vertical Sintering Furnace

TABLE E-10. Equipment Data Sheet for the Sintering Furnace

Function

Reduce moisture content and increase density of pellets by solid-state sintering.

Interfaces

Green pellet classifier/separator; sintered classifier/separator.

Design Parameters

Automatic and remote operations; remote replacement and installation; design rate = 28 kg/h.

Feed material: spherical 1/8 in. to 1/4 in. in dia; density approximately 0.8 to 1.5 g/cm³; moisture content is approximately 30%.

Outlet material: 0.100 in. to 0.250 in. in dia; density = 3.0 to 4.0 g/cm³

Operating Conditions

950° to 1250°C for 2 h; heating @ 300°C/h; cooldown @ 400°C/h.

Features

Variable flow rate; temperature range up to 1400°C; off-gas system connection; pellet/carrier separator; carrier conveyor and feeder system; closed-loop cooling; modular construction.

Services or Auxiliaries

Electric power 120 V, 480 V, 45 kW connected, and 80 kW demand; temperature detectors; flow indicators; water supply and drain 25 gpm; instrument system has 1 kW of isolated shielded voltage.

Space Requirements

5 ft in dia by 15 ft high

Maintenance Methods

Remote

Remarks

Special design required.

TABLE E-11. Equipment Data Sheet for the Sintered Classifier/Separator

Function

Separate large or fused hunks of material from standard size pellets; provide sample station interface.

Interfaces

Vertical sintering furnace; pneumatic system; mill.

Design Parameters

Automatic and remote operations; remote installation and replacement.

Operating Conditions

Temperature = 25^o to 100^oC; rate = 0 to 50 kg/h; fines are less than 1/16 in. in dia; oversized particles are greater than 1/4 in.; product is 1/10 in. to 1/4 in. in dia.

Features

Remote adjustment; temperature sensors; pressure sensors; flow indicators.

Services or Auxiliaries

Electrical; sample station; instrumentation.

Space Requirements

3 ft by 4 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Special installation, similar to that of green pellet classifier.

TABLE E-12. Equipment Data Sheet for the Pellet Mill or Crusher

Function

Reduce sintered pellet material to conveyable particle size.

Interfaces

Separator/classifier; pellet hoppers; pneumatic system.

Design Parameters

30 kg of supercalcine pellets/h; remote operations, removal and replacement; material is less than 8 in. ring size; discharge material is less than 1/16 in. maximum dimension; process material specific gravity = 4.0 and hardness = 5 to 7 Mohs.

Operating Conditions

Internal temperature is ambient, 25^o to 150^oC; external temperature is ambient, 25^oC.

Features

Stainless steel and abrasion resistant material; remote attachments.

Services or Auxiliaries

Electrical; instrumentation.

Space Requirements

2 ft by 3 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Special application by adapting commercial devices such as Sprout-Waldron Koppers Co., Inc. crushers or impact mills; Alpine American Corp. hammer or cutting mills; or Mikropul Corp. pulverizing machinery.

TABLE E-13. Equipment Data Sheet for the Pneumatic System

Function

Transport pellets and pellet particles.

Interfaces

Pellet classifier; pellet surge hopper; scrap collection hopper; scrap mill.

Design Parameters

Transport 30 kg pellets/h; specific gravity = 4.0; particle hardness = 5.9 Mohs; temperature = 100^o to 500^oC; remote operations; in-cell components remote monitoring, removal and replacement.

- prime mover sized to meet maximum load requirements of system with oversized motor so allowance for additional loads imposed on system
- smallest pipe size(s) capable of carrying design capacity within pressure drop constraint
- piping run routing to minimize line length and number of turns
- downward or inclined lines for process material to be avoided
- process material to enter through air locks

Operating Conditions

Maximum inlet vacuum = 15 in. Hg; system pressure drop = 5 psi; solids-to-gas ratio range = 10 to 20; conveying velocity = 2500 to 5000 ft/min; terminal velocity = 3000 to 4000 ft/min.

Features

Positive-displacement blower: variable speed; 2000 to 4000 rpm; 50 to 150 acfm @ 10 psia inlet and 16 psia outlet

Pipe: stainless steel; 1 to 2 in. dia; mill run 2B interior finish; 10 to 20 ft sections; bend radius for particles 2 ft or greater, for pellets 4 ft or greater

Connections: Cherry Burrell's I Line or similar types; remote flanged joint if required for rigidity

Filters: sintered 300-series stainless steel with short pulsed air blown back

Line valves: full ported ball valves

- Air flow with flow deviation control will be provided upstream of the initial process entry point for process transfer run.
- For closed-loop systems, a cooler will be provided to remove the heat added to the air by the compressive action of the blower. The temperature of the air stream at the process entry point will be approximately 100^oF.
- A surge tank will be provided upstream of the process entry point to dampen air flow pulsations.
- Pressure controls will be provided at the line inlet to assure a negative pressure in the transfer line such that 13.7 psia is not exceeded.
- Dry air (dew point 0^oC or lower) will be introduced to the conveying line at a negative pressure of about 13 psia and at an adequate linear flow velocity.
- Differential pressure will be monitored across all filter systems.

Services or Auxiliaries

Compressed air; conditioned air; instrumentation; drain;

Electrical: capacity = 10 kW; average use rate = 4 kW

Water: capacity = 100 L/h; average use rate = 40 L/h.

Maintenance Methods

In-cell components--remote.

Remarks

Special design required for this installation.

TABLE E-14. Equipment Data Sheet for the Pellet Surge Hopper

Function

Provide storage for pellets between continuous and batch operations of process.

Interfaces

Pneumatic transport system; volume hopper.

Design Parameters

Remote operations, removal and replacement; contain up to 110 L of pellets; stainless steel construction.

Operating Conditions

Temperature range = 25° to 450°C; pressure range = 10 in. Hg vacuum to 25 psig.

Features

Load cells or level sensors; cooling coils; temperature sensors; pressure indicators.

Services or Auxiliaries

Instrument system; water; drain; air or electrical.

Space Requirements

Maintenance Methods

Remote

Remarks

Design similar to that of the MIL surge hopper.

TABLE E-15. Equipment Data Sheet for the Volume Hopper

Function

Measures and limits batch size to be inserted in a canister.

Interfaces

Surge hopper; canister fill mechanism; recycling mill.

Design Parameters

Stainless steel construction; remote operations, removal and replacement; limit batch size to that required by a canister design (55 L).

Operating Conditions

Temperature range = 25⁰ to 450⁰C; pressure--slightly negative to slightly positive.

Features

Load cells or level sensors; cooling provisions; temperature sensors; inlet valve--remote operation.

Services or Auxiliaries

Instrument system; drain; compressed air; electrical; water.

Space Requirements

Maintenance Methods

Remote

Remarks

Similar to that in MIL process.

TABLE E-16. Equipment Data Sheet for the Canister Fill Mechanism

Function

Connects volume hopper with canister basket; directs and controls flow of marbles.

Interfaces

Volumetric hopper; canister.

Design Parameters

Remote operations; removal replacement; stainless steel construction.

Operating Conditions

Temperature range = 25° to 450°C.

Features

Mate with canister; vertical movement.

Services or Auxiliaries

Vacuum; inert gas; compressed air or electrical.

Space Requirements

Maintenance Methods

Remote

Remarks

Similar to that of MIL process.

TABLE E-17. Equipment Data Sheet for the Metal Fill Station

Function

Supply molten lead to the canister.

Interfaces

Canister; metal-melting furnace.

Design Parameters

Remote operations, removal and replacement.

Operating Conditions

Temperature range = 25° to 400°C; pressure range = 0 to 125 psig.

Features

Mate with canister.

Services or auxiliaries

Electrical power; instrument systems; inert gas; compressed air; cooling water; hydraulics; drain.

Space Requirements

Maintenance Methods

Remote

Remarks

Similar to that in MIL process.

TABLE E-18. Equipment Data Sheet for the Metal-Melting Furnace

Function

Transform solid lead into a liquid form.

Interfaces

Metal fill mechanism.

Design Parameters

Operating Conditions

Temperature = 0° to 450°C.

Features

Control of pouring rate and temperature; slag-free pouring.

Services or Auxiliaries

Electrical power; cooling water; instrument system; drains; ventilation supply and exhaust.

Space Requirements

Maintenance Methods

Contact

Remarks

Identical to furnace in MIL process.

E.10 REFERENCES

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APPENDIX F

PYROLYTIC-CARBON COATED PELLETS-IN-METAL MATRIX PROCESS

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APPENDIX F
PYROLYTIC-CARBON COATED PELLETS-IN-METAL MATRIX PROCESS

F.1 INTRODUCTION

Another option for the inner core of a multibarrier waste concept is supercalcine pellets coated with layers of pyrolytic carbon. Supercalcine is a crystalline assemblage of mutually compatible refractory and leach-resistant solid solution phases incorporating HLLW ions (Rusin et al. 1978). Supercalcine by itself is superior to ordinary calcine that is formed without additives. Supercalcine can also be used as the inner core for a composite, multibarrier waste form (Energy Research and Development Administration 1976). When supercalcine is formed into pellets, the pellets provide an alternative waste form to the glass monoliths produced in the ICGM and JHGM processes. (The ICGM process is also referred to herein as the reference process).

Coatings around the supercalcine pellets provide increased leach and impact resistance as well as increased thermal protection during encapsulation in a metal matrix. Coating the pellets can also provide an oxidation barrier if necessary. The coating is an integral part of the multibarrier concept; it is the last barrier between the inner core, which contains the waste, and the environment.

As with glass marbles, the production of coated supercalcine pellets affords an opportunity for quality analysis and recycling of unacceptable product after the spray calciner and before filling a canister. Several metal, oxide and carbon coatings have been successfully applied to simulated waste particles at General Atomics Company, ORNL, Battelle, Columbus Laboratories (BCL) and PNL (Rusin et al. 1978). When incorporated in a metal matrix, the coated supercalcine pellets produce a multibarrier product with enhanced inertness due to improved thermal stability, mechanical strength and leach resistance over the product from the reference process. Since the technological complexity of a waste fixation process increases with increased inertness or stability, the production of this waste form requires more complex operations than the reference process, which adds to the operating cost and risk.

Future development of coated supercalcine pellets as a waste form will require the demonstration of supercalcine pellet production, the application of a suitable coating, encapsulation of the coated pellets in a metal matrix, and the integration of these operations into an overall remote process.

The PCCPIM process facility will have the capability to 1) receive and store the HLLW, 2) proportion and thoroughly mix supercalcine-forming chemicals into the HLLW, 3) calcine the solution to fix various ions in thermodynamically stable crystalline phases, 4) form the calcine crystals into small pellets that are then heat-treated, 5) coat the individual pellets with carbon, oxides or metals, 6) load these into a canister, 7) fill the voids and annulus of the canister with lead, 8) seal-weld the canister, 9) check the integrity of the canister, and (10) decontaminate and transport the canister to interim storage before final disposition.

F.2 PROCESS DESCRIPTION

A flow diagram of the PCCPIM process is shown in Figure F-1. As covered under the ICGM process (Appendix A), HLLW is received in the waste facility and is temporarily stored in a specially designed HLLW feed system. The composition of the basic HLLW is similar to that given in the Overall Process Bases Section. The HLLW is modified with selected liquid additives to form the supercalcine feed. The liquid additives, usually including Ca, Sr, Al and Si, are selected according to the waste composition and add from 35 to 45 wt% to the HLLW.

The spray calciner for the PCCPIM process is the same as those in the ICGM reference process except that it has additional capacity, may be operated at a lower temperature and does not have frit feeding provisions. The supercalcine solution is jetted to the spray calciner. The liquid flow is pneumatically atomized at a controlled rate into a heated chamber, where granular, crystalline calcine particles and an off gas are produced.

The supercalcine particles are fed through an air lock into a hopper where they are cooled. The particles then flow into a system that feeds the disc-pelletizer at a selected rate. Disc-pelletizing is an agglomeration technique that forms spherical pellets from powder particles (Floyd and Engelleitner 1967). Disc-pelletizing is an optimum method for compacting nuclear waste compositions when followed by sintering (Rusin et al. 1978). In this process the supercalcine particles are consolidated into high-density spheres or green pellets by the addition of binders and the rotating action of the inclined surface of the pelletizer.

The pellets from the disc-pelletizer are fed onto a classifier/separator where the fines, the oversized pellets and other wet scrap are separated for recycling back into the process stream. The green pellets within the desired size range are isolated, sampled and fed into the sintering furnace.

In the sintering furnace, the green pellets are brought up to and held at sintering temperatures and are then cooled at a controlled rate to drive off the moisture and other volatiles; thus, smaller, more dense and more durable pellets are formed.

After leaving the furnace, the sintered pellets are fed to another classifier where the pellets can be sampled; the fines and oversized globs are diverted and fed to a mill for crushing and recycling. Pellets of acceptable sizes are fed into a surge hopper where they can be stored and batch-fed to the pyrolytic coater.

In the coater, the pellets are raised to the desired process temperature and are tumbled in an oxygen-free environment. A carbon-bearing gas, nickel carbonyl, is introduced into the coater, and when heated the gas breaks down and carbon is deposited on the surfaces of the pellets. After a batch of pellets has been processed long enough to achieve a coating of the desired thickness, they are fed through an air-lock valve and into the pneumatic transfer system.

A closed-circuit, negative-pressure pneumatic system with two loops is contemplated to 1) transport the coated pellets to the canister surge hopper and 2) to convey scrap particles to the scrap collection hopper.

FIGURE F-1. Pyrolytic-Coated Pellets-in-Metal Matrix Process Flow Diagram

Canister surge hoppers, volume hoppers and fill mechanisms similar to those identified for the MIL process (Appendix D) are used to convey a measured volume of coated pellets into the inner basket of the waste canister.

Provisions are incorporated into the volume hopper to move unacceptable pellets via gravity to the mill for crushing and recycling. The solid scrap from the mill is fed into the pneumatic system through an air-lock valve and is conveyed to the scrap collection hopper, where the material can be fed back into the calcine hopper.

The canister containing coated pellets is handled and its annulus and voids are filled with molten lead as covered in the MIL process. After being filled with lead, the canister is seal-welded, leak-checked, inspected, decontaminated and transported to a storage area as covered in Appendix A.

F.3 BASES AND ASSUMPTIONS

Most of the bases and criteria for the PCCPIM process are noted in the Overall Process Bases Section. Data on the HLLW and its waste forms are also presented in Appendix A. Other applicable assumptions and bases are identified in the following sections.

F.3.1 PROCESS BASES

As previously noted, the flow diagram for the PCCPIM process is shown in Figure F-1. It is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. Design capabilities are 1.5 times the normal operating values. Additional processing data is given below:

- Supercalcine feed to spray calciner
 - operating rate = 203 L/h
 - design rate = 305 L/h
 - HLLW loading = 78 wt%
 - heat loading = 2.7 W/L
 - oxide content (in HLLW and supercalcine formers) = 92.6 g/L
- Calcine
 - operating rate (oxide) = 18.8 kg/h
 - design rate (oxide) = 28.2 kg/h
 - operating rate (with 9.5% volatiles) = 20.8 kg/h
 - design rate (with 9.5% volatiles) = 31.2 kg/h
- Supercalcine pellets
 - specific gravity of lightly vibrated pellets = 0.78 kg/L
 - specific gravity of sintered pellets = 3.8 kg/L
 - heat loading of individual pellets = 111 W/L
 - bulk heat loading assuming 50% packing density = 55.5 W/L
 - wet scrap design rate = 4 kg/h
 - dry scrap design rate = 2 kg/h

F.3.2 CANISTER

As noted in the Overall Process Bases Section, a major constraint is the allowable heat loading of the canister. This criterion permits the placing of approximately 65 L of waste in the coated supercalcine pellet form in a canister. To utilize a reasonable length-to-diameter ratio for process operations, lead filling and handling, a canister 30.5 cm in OD and approximately 2.1 m tall (12 in. in OD x 82 in. tall) was selected for the PCCPIM process. The canister and inner basket are shown in Figure F-2. The inner basket is positioned inside the canister before the flange is welded in place. Data on the PCCPIM canister are also covered in Appendix A.

FIGURE F-2. Canister and Inner Basket

F.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

For this study it is assumed that with two exceptions the effluents from the PCCPIM process are similar in content and volume to the effluents from the reference process (Appendix A). The exceptions are the effluents associated with the operation of the metal-melting furnace and the effluents emitted from the pyrolytic-carbon coating step. For process and maintenance simplicity, the metal-melting furnace is located and operated outside of the radioactive zone so its effluents are similar to those of a commercial furnace and are not a major factor. The off gas from the coating process requires a separate, special system to handle, treat and recycle elements before they enter the reference off-gas system or the process cell systems.

Except for additional worn equipment due to the fact that the PCCPIM process is more expanded and complex than the reference process, it is assumed the wastes from the PCCPIM process are of the same order of magnitude in volume and content as the wastes from the reference process. It is also assumed that systems and provisions are included in the fuel reprocessing plant to treat, reduce the volume and package both liquid and solid wastes. Finally, in the PCCPIM process it is assumed that all solidified process scrap that may contain high-level waste is recycled back into the process.

F.3.4 UTILITIES AND MATERIALS

Estimates of the required utility services and essential materials are shown in Tables F-1 and F-2, respectively. These estimates are based on the requirements for the reference process with adjustments for additional process steps and anticipated space and volume changes.

TABLE F-1. Estimate of Utilities Needed for PCCPIM Process

<u>Utility</u>	<u>Capacity Required</u>	<u>Average Use Rate</u>	<u>Disposition</u>
Electricity	1500 kW	850 kW	NA
Raw water	1850 L/min	600 L/min	Recycle or environment
Process water	600 L/min	40 L/min	Recycle as HLLW or ILLW
Sanitary water	40 L/min	20 L/min	Sanitary or chemical sewer-tile field
Air	750 scmm	500 scmm	As appropriate
Steam	25 kg/min	5 kg/min	As appropriate

TABLE F-2. Estimate of Materials Needed for PCCPIM Process(a)

<u>Material</u>	<u>Amount Required Per Unit Time</u>
AL(NO ₃) 9 H ₂ O	35,500 kg/yr
Ca(NO ₃) ₂	10,000 kg/yr
30% colloidal SiO ₂	62,500 kg/yr
Sr(NO ₃) ₂	5,500 kg/yr
Canisters	5.5/d
Caustic (19M)	10,000 L/yr
Nitric acid (12.2M)	12,000 L/yr
Helium sources	6.0/d
Argon	6,000 to 7,000 L/d
Detergent (for example Tide)	1,000 kg/yr
Ammonia	120,000 kg/yr
NiCO ₄	300 L/d
C ₂ H ₂	700 L/d
Nitrogen	2,500 L/d
Oxygen	500 L/d

(a) Excludes maintenance materials and apparel.

F.4 FACILITY AND EQUIPMENT REQUIREMENTS

The PCCPIM process is contained in a reinforced concrete structure similar to that shown in Figure F-3. The major pieces or equipment systems are listed below:

- calciner feed system
- calciner
- disc-pelletizer system
- sintering furnace system
- coater system
- pneumatic transfer system
- scrap recycling system
- marble hopper and fill system
- metal fill system
- canister handling system
- off-gas system

The overall requirements for the waste facility and its support groups and services are covered in the Overall Process Bases Section. The major factors to be considered in process evaluations or cost estimates are the requirements for remote operations and for remote maintenance of all in-cell equipment.

FIGURE F-3. General Arrangement of PCCPIM Waste Facility

F.5 STATE OF PROCESS DEVELOPMENT

The PCCPIM process requires a number of process steps that have not been performed using wastes or simulated wastes, or on any basis near that required for a production facility. Coatings are technologically complex, but do offer considerable enhanced inertness if this is required or desired. In addition, this process involves a number of involved mechanical/chemical steps that must be performed remotely using equipment that requires remote maintenance and remote removal when utilized in a production facility.

Considerable research, development and tests have been performed at PNL on spray calcination and spray calciners, and this effort has established a very good and firm bases for this major processing component. This data can be applied to the PCCPIM process.

Research on supercalcine was first initiated early in the 1970's at Pennsylvania State University. Many laboratory-scale tests and evaluations have been performed over the years on supercalcine (Rusin et al. 1978; McCarthy 1977). Calcine pellets have been made from simulated HLLW at PNL as well as at other laboratories (Lamb and Cole 1979).

Pyrolytic-carbon coatings have been thoroughly investigated and demonstrated for HTGR fuel development and production. Chemical-vapor-deposition coating of supercalcine has been demonstrated on materials containing waste (Rusin et al. 1978).

The multibarrier concept of coated supercalcine pellets in a metal matrix is a candidate for further development. It offers increased ruggedness and inertness and it is extremely leach resistant; however, it has the potential of a large increase in process and technological complexity over the reference process. Future development of coated supercalcine pellets as a waste form will require the demonstration of pellet production, the radiation stability of supercalcine, the application of a suitable coating, the encapsulation of the coated pellets in a metal matrix and the integration of these operations into an overall remote process.

F.6 UNIT OPERATIONS

The components of the PCCPIM process have been grouped into 17 unit operations. In this section, information on the operation of each system and areas requiring further research and development are covered. Drawings and data sheets on the equipment and operations discussed in this section can be found at the back of this appendix in Section F.9.

F.6.1 HLLW FEED SYSTEM

The function of and the requirements for the design and operation of the HLLW or calciner feed system are covered in detail under the reference process (Appendix A). The HLLW feed system for the supercalcine process is basically the same as for the reference process except that it includes provisions to 1) introduce and incorporate supercalcine additives with the HLLW, 2) maintain the proper solution, 3) sample and analyze the feed solution, and 4) contain the larger volume that is required.

After the supercalcine additives (which may be solutions of calcium and aluminum nitrate and an aqueous colloidal suspension of silica) have been added to formulate the desired feed, the other preparations (verification, analysis, start-up, transfer, operation and shutdown) for the PCCPIM calciner feed are similar to those covered in the reference process. Parameters for the supercalcine HLLW feed system are noted in Table F-3 in Section F.9.

The basic HLLW feed system has undergone considerable tests and developmental programs, and its requirements and parameters have been well established. However, considerable efforts are required to identify the requirements and parameters for a particular supercalcine feed system. Some of the specific areas of concern are covered in detail in Section F.7.

F.6.2 SPRAY CALCINER

The function, design and operation of the supercalcine spray calciner are basically the same as for the reference spray calciner (Appendix A). This supercalcine process does not require a frit feeding system, but it does require an additional capacity of about 28% over the combined capacity of the two spray calciners in the reference process.

The supercalcine feed is introduced to the heated spray chamber of the calciner through an air-atomized spray nozzle. The volume of radioactive waste is significantly reduced when converted from a liquid to a powder. The liquid waste is dried and converted to oxides and reaction off gases. Calcine collects at the bottom of the calciner after falling through the chamber or after being removed from the off-gas stream by sintered filters.

The research and development efforts noted under the reference process for the spray calciner are also required for the optimization of the PCCPIM process. The spray calciner for the supercalcine process also requires the following efforts to resolve and/or determine concerns and unknowns:

- test a larger volume feed system

- test calciners with supercalcine solutions to determine such items as probability of sintered buildups
- establish and test methods of detecting and removing sintered buildups (water sprays, temperature cycling, vibration, etc.)
- establish operating parameters and limits for supercalcine
- evaluate effect supercalcine additives may have in catalyzing an explosion from tramp impurities, and if effect is positive, establish safe alternatives
- evaluate the effect of supercalcine additives on the volatility of species such as cesium and ruthenium and thereby determine off-gas processing requirements.

F.6.3 OFF-GAS SYSTEM

The requirements for, the function and operation of, and the research and development efforts required for the basic PCCPIM process off-gas system are the same as for the ICGM process, which has an off-gas system for each spray calciner. The PCCPIM off-gas system may also consist of parallel units for efficiency. Its capacity is 40% to 50% greater than the reference process off-gas system.

F.6.4 CALCINE HOPPER AND PELLETIZER FEEDER STATION

A remotely operated air lock is located below the spray calciner and above the calcine hopper to minimize air flows through the calcine feeder system. The air-lock valve is self-sealing, has cooling provisions and is operated by a variable-speed drive so the supercalcine particles can normally be fed into the hopper at the same rate in which they are formed.

The operation of the air lock is relatively simple and is coordinated with the operation of the spray calciner and the pelletizer feeder. The cooling provisions can be controlled to protect the valve and to assist in the reduction of the supercalcine temperature. A spray-flush system removes calcine buildups and cleans the valve internally. Design and operating conditions for the air-lock valve are indicated in Table F-4 in Section F.9.

A relatively tall and very narrow, water-cooled stainless steel hopper, with a surge capacity for a couple of hours of spray calciner operation, is the collecting media for the spray calciner product. Here the supercalcine is cooled to below 100°C, and a uniform supply of particles is provided to the pelletizer feeder. If the surge hopper is full the processes upstream must cease, and when the supply of particles in the surge hopper is depleted the operation of the pelletizer should be stopped. The operation of the hopper mainly involves monitoring such items as the level, flow and temperature of the contents, and the internal pressure. Table F-5 in Section F.9 is the equipment data sheet for the calcine hopper.

The pelletizer feeder is the third component of this station. It provides a consistent feed to the disc-pelletizer at the selected rate. In addition, the feeder and the feed prevent large volumes of air movement either into or out of the surge hopper. The start-up, operation

and shutdown of the feeder must be coordinated with that of the disc-pelletizer. Feed flow and temperature must be monitored and maintained within established limits. Data on the pelletizer feeder are given in Table F-6 in Section F.9.

Air-lock devices consisting of flapper, star or rotary valves are available commercially and some of these may perform adequately and withstand the operating temperatures of the PCCPIM process. However, none of these commercial valves have been designed for remote maintenance and replacement. Research and development are required to design a valve for a radioactive, remotely operable facility. Once it is designed the valve should be operated in conjunction with a spray calciner to verify the valve design. For example, the following should be tested: its operation with calcine particles, provisions for remotely operating and maintaining it, and methods for cleaning or flushing the valve. In addition, the developmental efforts described in Section F.7 are required for the pelletizer feeder.

F.6.5 DISC-PELLETIZER STATION

As indicated above, the supercalcine is fed to the disc-pelletizer by a feeder. A liquid binder is also sprayed into the pelletizer. The liquid binder is responsible for the strength of the green pellets. It fills the pores and binds adjacent particles together either by forming chemical bonds or by capillary action. The binder also acts as a lubricating medium to obtain the densest particle packing configuration possible for that material. When the calcine, or powder, is somewhat soluble in the liquid binder, the strength of the dry pellets is improved; upon drying, the soluble components form interlocking, crystalline networks between adjacent particles.

The pelletizer consists of a rotating, inclined pan into which powder and liquid binder are fed at controlled rates. The powder initially forms small pellets that are shaped and compacted by collisions with other pellets and with the pan. These pellets then grow in a snowballing fashion as more powder and binder are added. The net result is a dynamic, equilibrium condition where pellets are continuously discharged as more pellets are formed. The size of the pellets is determined by the materials used and the pelletizer operating conditions.

The stable operation of the disc-pelletizer to produce acceptable-quality pellets of a desired size is a function of:

- feed powder characteristics
- feed powder-binder interactions
- liquid binder properties
- pelletizer operating parameters.

These variables must be maintained within reasonable limits to maintain process equilibrium. In particular, the inherent sensitivity of a small pelletizer requires precise control of the major parameters: the powder feed rate, the binder feed rate, and the powder-to-binder ratio. Unanticipated changes in any of these will have a direct impact on the size and strength of the green pellets. Table F-7 in Section F.9 lists other attributes of the pelletizer.

The second major component of the pelletizer station is a classifier/separator where the green pellets are screened to remove and recycle loose powder and oversized pellets. The

pelletizer and classifier/separator system contains dust removal or suppression provisions to maintain process cleanliness with minimal material loss and contamination spread.

As with the calcine hopper-feeder station, nitric acid spray-flush provisions are incorporated into this station. All dust, under- and oversized pellets and products from spraying and flushing drain into a wet scrap vessel where these materials can be recycled to the calciner feed system.

Also incorporated into this station is the means of assessing green pellet quality on a semicontinuous basis. Since the pelletizer station is one part of essentially a continuous process, its overall operation has to be coordinated with others up- and downstream. The limited operating parameters such as binder-to-powder ratio necessitate precise process control and sampling systems.

One of the positive aspects of the PCCPIM process is that it affords the opportunity to monitor and analyze the quality of the waste form before it is converted into a form where corrections or adjustments cannot be made. The exit at the pelletizer is one of the logical places in this process to monitor and analyze product quality.

An enclosed and remotely operated and maintained classifier/separator to separate powders and oversized material from suitable pellets and to handle the green pellets without damaging them needs to be designed, developed, fabricated and tested in conjunction with a pelletizer. A system capable of monitoring such items as flow rate, quality, strength, etc. of the pellets also needs to be devised and implemented. This may include a remote sampling system. Finally, the positive or negative impact that the radioactive decay heat may have on design parameters of systems handling green pellets should be assessed.

A disc-pelletizer has been operated at PNL on a limited basis with some very positive results. This effort needs to be continued to resolve problems and to determine operating parameters as noted in detail in Section F.7.

Additional data for the classifier/separator and the wet scrap tank are shown in Tables F-8 and F-9 (see Section F.9), respectively.

F.6.6 SINTERING FURNACE

The pellets that are within an acceptable size range are fed from the classifier/separator to a vertical sintering furnace where they may be mixed with a granular, carrier medium to prevent pellet damage during movement through the furnace. The carrier is a free flowing, high-temperature material that is compatible and inert with the supercalcine pellets; alumina (Al_2O_3) may be such a material.

The mixture of pellets and carrier is fed in at the top of the sintering furnace. The downward movement of the mixture through the preheating, sintering and cooling muffles is controlled by the exit feeder or valve at the furnace base. If necessary, an atmospheric gas or gases can be introduced at the base of the furnace with enough pressure to overcome the pressure drop across the moving bed of supercalcine pellets and carrier; thus, there is intimate contact between the pellets and the gas for proper sintering control.

As with other stations in the system, the sintering furnace requires a relatively long period of time to bring it up to operating temperatures and conditions. Exact operating procedures are dependent on the final furnace design, but the following general start-up procedures are required: 1) introduce a flow of atmospheric gas to purge the furnace components, 2) turn power on to the furnace and gradually raise temperature (at 100°C/h) to 500° to 600°C , 3) hold at this temperature for 5 to 10 h to bake all moisture from the furnace insulation, 4) raise furnace temperature at 300°C/h to about 100°C above the desired operating temperature, 5) as the furnace temperature approaches 1000°C , start water through the cooling coil and start exit feeder to initiate carrier flow, and 6) when the maximum temperature has been achieved, reduce the controlled temperature to the desired operating temperature and allow it to stabilize with the carrier grain moving through the furnace.

When equilibrium conditions have been achieved in the preheating, sintering and cooling muffles, the furnace is ready to accept the flow of supercalcine pellets. The furnace throughput, the pellet-to-carrier ratio and the operating temperatures all require monitoring and controlling during operation. The furnace operation must be integrated with the pelletizer upstream and the separators and downstream systems. Shutdown is basically the reverse of the start-up procedures.

Means of separating carrier from pellets and recycling the carrier to the top of the furnace are integral components of the sintering furnace station. After separation from the carrier, the sintered pellets are fed to another classifier/separator. The sintering furnace is shown in Figure F-4 (see Section F.9) and additional data on the furnace are given in Table F-10 (Section F.9).

A research, development and design program, as outlined in Section F.7, for the vertical sintering furnace is required to assure the availability of a production-scale furnace that can be used in a radioactive installation and that can be maintained or removed remotely.

F.6.7 CLASSIFIER/SEPARATOR

The sintered supercalcine pellets are fed via gravity into a classifier/separator where the fines and the oversized material are separated and fed to a mill via gravity. The pellets within an acceptable size range are fed into a surge hopper, where they are stored and fed in batches to the pyrolytic coater.

Incorporated in this station are the means to monitor the temperature and weight of the pellets. There is also a sampling system where individual pellets can be removed from the process stream and sent to an analytical station or laboratory to check their shape, composition and chemical or physical characteristics.

The operation of this station must be started before any pellets leave the sintering furnace and must be continued until the pellet flow ceases. It must also be coordinated with the operation of the coater system and the mill. Table F-11 in Section F.9 gives additional design and operating conditions for this classifier/separator. The research and development identified above for handling and sampling green pellets are also applicable to sintered pellets.

F.6.8 MILL

Any scrap consisting of powders, small or broken pellets and pellets fused together is fed into a vertical mill or crusher via gravity or vibrators. The mill is operated on an intermittent or as-needed basis to reduce the supercalcine scrap material into particle sizes that can be conveyed pneumatically and recycled back into the process stream. During operations, a means of monitoring the flow of material into or out of the mill is required. Data on the mill is noted in Table F-12 in Section F.9. Essentially, the same research, designing and testing outlined for the remote mill in the MIL process in Appendix D are required for a mill in this process.

F.6.9 COATER STATION

As noted above, the acceptable sintered pellets are continuously fed from the classifier/separator into the coater surge hopper. The hopper is a relatively small, stainless steel vessel with a surge capacity for a couple hours of production. It may require cooling provisions for its contents or valve. The exit from the hopper contains a valve that permits sintered pellets to flow through when open and is an oxygen-tight seal when closed. If the surge hopper is full, the processes upstream must be stopped. Operating the hopper mainly involves monitoring such items as the level, flow and temperature of the contents and the internal pressure. The coater surge hopper is shown in Figure F-5 (see Section F.9) and additional data on the hopper are given in Table F-13 (Section F.9).

The coater is the major component of this station. Measured batches of pellets flow into the coater when the surge hopper exit valve is opened. When the desired batch size is obtained, the hopper is valved-off to effect a gas seal. The coater is then purged of air and oxygen, and if not already operating, the screw auger is started and the heaters turned on. When operating temperatures are reached, carbon-bearing gases are introduced into the bottom of the coater.

The coater basically consists of two concentric, vertically mounted cylinders with an auger in the center. The auger lifts the supercalcine pellets in the center and they fall through the outer annulus until they are picked up and recycled by the auger. Heating coils surround the coater, and when the internal operating temperature is reached, the carbon-bearing gas decomposes and carbon is deposited on the pellets as well as other surfaces. After the desired coating has been achieved, the heaters and auger can be left in operation or may be turned off; the carbon-bearing gases are valved-off and the exit valve leading to the pneumatic system is opened. The exit area of the coater may require some cooling and purge provisions to assure the proper functioning of the exit valve.

Batch sizes and times is a function of the coating thickness desired, equipment size and deposition efficiency of the gas at a given operating temperature. A very dense and quite uniformly thick coating can be applied to the pellets without sharp edges. Coatings of 1 to 2 μ /min are anticipated on the surfaces of pellets with a batch size of 50 kg and a temperature range of 400⁰ to 500⁰C, using nickel carbonyl gas in an environment containing low levels of air or oxygen.

Unfortunately the walls of the coating equipment are also coated with carbon. This is removed on a periodic basis by heating the inside of the coater to about 800°C and purging it with air or oxygen. Burning the coating off the walls may require a period of 3 to 4 h, and it may be required after every three or four batches. The coater is shown in Figure F-6 (see Section F.9) and Table F-14 gives other features of the coater (Section F.9). Considerable research and development are required for the pyrolytic coater and its off-gas system requirements. These are identified in Section F.7.

The off gas from the coater, which consists of nickel carbonyl, nickel dust, carbon dust, CO₂ etc., must be processed to avoid deposits within the off-gas system and to prevent explosive mixtures from building up. There may be some incentives to pass the off gas through a high-temperature chamber into which air or oxygen is purged as an oxidizing agent before the off gas reaches a system of filters. Due to the potential hazards of mixing H₂ and O₂, the attributes of the high-temperature chamber need to be studied. Preliminary flow diagrams are shown in Figure F-7 in Section F.9. A viable alternative is also shown in Figure F-7, where the nickel dust is removed by filtration. Next, stainless steel wool filters are used to further decompose or break down the gases before passing them through absolute filters and into a CO absorber. The coater off-gas system will require full-sized parallel filters so a unit can be isolated and replaced without shutting the process down.

F.6.10 PNEUMATIC SYSTEM

Transport of radioactive materials should be via negative pressure. As in the MIL process, the prime mover is a rotary, positive-displacement type blower, which provides a vacuum to the receiver vessels via blowback filters. Process material is conveyed through short sections of 300-series stainless steel piping; the short sections permit remote removal of the piping. Pipe connectors limit protuberance at the joints. Process material enters the system through feeder control valves. Exhaust air is removed from the top of the systems' vessels through sintered, stainless steel filters that can be blown back with air pulses. Load cells on the receiving hoppers and/or flow indicators in the system are used to monitor system operation and cleanout.

The pneumatic system must be operating prior to any exit of pellets from the coater and must remain operating until all of the pellets have cleared the coater. When it is necessary or desirable to convey scrap particles to the collection hopper, the pellet loop is valved off and the pneumatic system started before starting the mill.

For a vacuum and/or closed system, it is necessary for the process material to be introduced into the system through air-lock devices. The design of these air locks affects pressure losses and drops as well as process material inertia and accumulation. The research, development and tests outlined in the MIL process (Appendix D) are applicable to a pneumatic system to convey coated pellets.

A schematic of a pneumatic system is shown in Figure D-7 (Section D.9 in Appendix D) in the MIL process. Additional data for PCCPIM pneumatic transfer system are given in Table F-15 in Section F.9.

F.6.11 RECYCLED FEED

The final receiving vessel for the pneumatic system is the recycled scrap collection hopper located in the upper portion of the process cell. Solid process scrap material is stored in this stainless steel hopper. The collected material is eventually recycled back into the process stream at the calcine hopper through a weigh feeder and gravity pipes.

The feed system for recycled scrap is similar to the glass frit feed system covered under the ICGM process (Appendix A). It is necessary to monitor and measure all material flows and coordinate these with the calcine hopper and pelletizer feeder.

F.6.12 PELLET SURGE HOPPER

A cylindrical-shaped, stainless steel vessel is the collecting point for the coated supercalcine pellets from the pneumatic system. The surge hopper is located high in the cell so gravity can be used for subsequent operations with the pellets. The flow of pellets out of the surge hopper and into the volume hopper is controlled by a valve that can close through a column or stream of pellets. The surge hopper is equipped with cooling coils, temperature sensors and level or weight devices to monitor and control the contents.

At this point in the process, the material flow is again converted from a continuous to a batch type operation. If the surge hopper is full, operations upstream must be stopped. The level and weight of the pellets in the surge hopper and the internal temperature must be monitored during operating periods. Provisions are incorporated to maintain the temperature of the contained pellets below a prescribed limit, which is below the highest allowable temperature of either the supercalcine core or the coating. Other data on the pellet surge hopper are given in Table F-16 in Section F.9.

F.6.13 VOLUME HOPPER

The volume of this stainless steel hopper, including its inlet connection to the entry valve, equals the volume to be filled by coated pellets in the inner basket of the storage canister. The valve between the surge hopper and volume hopper is normally open until the proper amount of pellets are in the volume hopper. Like the surge hopper, the volume hopper is equipped with cooling coils, temperature sensors and level or weight sensors. The flow of pellets out of the volume hopper is controlled by the canister fill mechanism.

The operation of the volume hopper mainly involves the proper sequencing of the upper inlet or feed valve, assuring the proper fill volume, drainage through the canister fill mechanism, and assuring that all of and only the measured volume has been drained through and out of the vessel. Temperatures must be monitored anytime process material is in the hopper so that cooling can be initiated if necessary. Supporting data for the volume hopper are given in Table F-17 in Section F.9.

F.6.14 FILL MECHANISM

After an inspected canister with an inner basket is on the canister transfer system and is located under the pellet fill station, the fill mechanism is inserted into the canister. When

a vacuum system is started to remove dust and particles from the falling pellets, the transfer of the coated supercalcine pellets from the volume hopper to the inner basket of the canister is begun. During and after filling, inert gas is introduced into the center pipe of the canister via the fill mechanism to cool the pellets if required and to help entrain dust into the vacuum system. After filling is completed, the gas and vacuum systems are valved off, and the fill mechanism is retracted from the canister. Additional data on the fill mechanism are given in Table F-18 in Section F.9.

The crucial batch operation in the PCCPIM process is the remote filling (and monitoring) of the inner basket in the canister with the proper amount of pellets on a repetitive basis with no over- or underfills. An engineering design should be made for a remote, production-sized system that includes the complete measuring system and the canister fill mechanism. A full-scale model should then be fabricated and tested in a laboratory demonstration. This demonstration would include the complete remote operation and monitoring of the system as well as the verification of the remote maintenance and replacement aspects.

F.6.15 INNER BASKET CLOSURE

After the inner basket has been filled with the proper amount of coated supercalcine pellets, the canister is transferred to a station where a device is inserted into the canister to close the opening in the top of the inner basket. To allow for long periods of time between filling and lead pouring and to prevent the radioactive decay heat from causing the pellets to soften or melt, provisions to cool the canisters while in this or subsequent stations are provided. Upon retraction of the closure device, the canister is transferred to the next station.

F.6.16 METAL FILL STATION

Next, the canister is lifted into the canister heater and up against the metal fill mechanism. Provisions are included to cool (via an internal gas flow) and/or heat (canister reactors) the canister and its contents, as required to maintain a temperature within 50°C of the melting point of the metal alloy to be used (300° to 400°C). When this equilibrium temperature has been reached, the cooling gas flow is terminated, the canister heaters are turned off and the valve is opened to start the metal pouring from the furnace. The liquid metal is introduced through the center pipe to the bottom of the canister and back up through the coated pellets and the outer annulus. A vacuum can be induced in the canister through the metal fill mechanism, if necessary, before and during pouring of the metal. The metal is poured continuously until the desired level is reached, and then the valve is closed. During the metal-pouring process, the bottom and possibly the walls of the canister are cooled to control lead shrinkage voids.

The canister is then lowered approximately 1 ft, the metal-flow control valve is re-opened and the lead is permitted to reach the new desired level. As the lead level drops due to the 5% shrinkage, additional metal is added as required to complete the filling of the canister to the selected level. Then the canister is lowered and removed from under the metal fill station and the metal level is checked.

The metal-melting furnace is located out of but adjacent to the process cell in a service area, so the molten lead can flow by gravity through a heated entry tube to the metal fill mechanism. With this out-of-cell location, the furnace, its controls and its operation are basically the same as for a commercial furnace. Heating and controlling the temperature in the entry tube is a special application as is the control of its isolation or block valves. Additional data on the metal fill station and the metal-melting furnace is given in Tables F-19 and F-20 (see Section F.9), respectively.

This study assumes that the metal-melting furnace is located outside of the process cell to reduce in-cell components and complexity. However, an engineering and design program is required to determine if this assumption is the best approach and to design the metal-pouring system, especially the in-cell components. When this design is completed or identified, inter-cell and in-cell components should be fabricated and installed in a demonstration mode. Then the operation of a remote, metal-filling mechanism, including metal-flow control and monitoring, could be demonstrated and evaluated. The remote maintenance and replacement aspects could also be verified.

F.6.17 CANISTER PROCESSING

After the proper metal level is assured, the canister is moved into the weld station, a helium source is inserted and the lid is seal-welded. Next, the canister weld is helium leak-checked to ensure its integrity. The canister is also visually inspected. Canisters that fail to pass the leak test are rewelded, if possible. Sealed canisters are then moved to a decontamination chamber where any loose contamination is removed. From here the canisters are placed in the in-cell storage racks or are transferred out of cell to an interim storage area. These canister treatment steps are covered in more detail in Appendix A.

F.7 RESEARCH AND DEVELOPMENT REQUIRED

There is a definite need to demonstrate all of the remote aspects of each unit operation in a cold mode. This need also applies to the complete PCCPIM process, and it is recommended that following or in conjunction with component tests the system be operated with simulated supercalcine material. These demonstrations could also be used to verify the arrangement of the equipment in the cell.

Although testing the overall system is an important aspect in any remote, nuclear installation, it should not be done before the requirements and needs of the unit operations have been identified. As with the SCPIM process, additional efforts for the PCCPIM process should be applied first to 1) identifying supercalcine feed system parameters, including sampling and analytical procedures, 2) evaluating supercalcine radiation stability, 3) resolving operational problems with a disc-pelletizer and its feed systems, 4) establishing design and operating parameters for the sintering furnace, 5) identifying design requirements for a pneumatic system to carry nuclear wastes in the coated supercalcine pellet form, 6) identifying the basis for remote pellet sampling and monitoring systems, and 7) establishing a system to remotely fill canisters with lead. The design and operating parameters for the pyrolytic-carbon coater and its off-gas system also need to be established and verified.

F.7.1 HLLW FEED SYSTEM

The basic HLLW feed system has undergone considerable development and evaluation, and its operating and design requirements are well established. Remaining areas of concern are covered in Appendix A. Other areas of concern that must be resolved to assure consistent consolidation of crystalline ceramics for a supercalcine feed system are:

- amounts and kinds of solutions required to formulate supercalcine compositions
- parameters and controls for adding and mixing supercalcine solutions to HLLW (proportionate and redundant metering, agitation, and control of temperature, acidity and gelation)
- effects of radiation on supercalcine solutions
- accuracy required for HLLW sampling and analysis
- effects of recycle streams.

F.7.2 CALCINE HOPPER AND FEEDER

The calcine hopper and feeder have been operated at PNL in conjunction with the operation of a disc-pelletizer (Rusin et al. 1978). This experience has indicated the following problems that must be resolved:

- Calcine must be cooled to well below 100°C before it is fed to the disc-pelletizer to avoid steam formation. As a result, a design program should be instigated to determine the amount of heat contained in the supercalcine particles due to calcination and due to radioactive self-heating. In addition, a design of a remotely operated and maintained

production-sized hopper feed arrangement capable of removing this heat without impeding the flow of calcine should be developed and reliability-tested under all operating conditions, including cleanout and maintenance.

- Systems to measure calcine level and flow rate on a continuous basis need to be devised, implemented and tested.
- Calcine must be fed to the disc-pelletizer at a controllable and uniform rate. There are several types of feed systems that may be applicable to this installation. A vibratory-screw type system has been used at PNL with limited success. In conjunction with the first item listed above, long-term tests should be performed to assure that precise control ($\pm 1\%$) of the fine particle feed can be achieved over the range of operating parameters, such as hopper-to-pelletizer pressure differential, calcine temperature differences, hopper feed depth and variations in calcine bulk densities.

F.7.3 DISC-PELLETIZER

In operations at PNL, stable continuous operation of a disc-pelletizer at low feed rates was not possible to achieve (Rusin et al. 1978). Thus, continuing efforts are required to resolve the following concerns or problems:

- designing, fabricating and testing of a remotely operated and maintained disc-pelletizer, whose rate capacity is compatible with that of the supercalcine system
- developing and establishing supercalcine pelletizing parameters
- limiting and controlling dusting during pelletizer operation
- adequate monitoring and controlling of powder feed rate, binder feed rate and powder-to-binder ratio
- establishing binder options and compositions for powder temperature ranges and different operating conditions
- determining impact radioactive particle decay heat may have on uniform operation and pellet quality
- developing method to remove and clean mud and dried buildup from the pelletizer and its component parts
- developing an adequate pellet sampling system and procedure.

Pellets made with just water as a binder may not have the most desirable properties. Additional research and development needs to be performed on various combinations of solid and liquid binders.

F.7.4 SINTERING FURNACE

A small, cylindrical vertical furnace designed to sinter bulk quantities of highly radioactive fuels in a controlled and reducing atmosphere has been developed and is undergoing proof-of-principle tests at PNL. This furnace has power concentrated in a small area, excellent

heat transfer to the product, low heat losses and a high throughput capacity. This on-going work has indicated that this furnace is the only type with a number of attributes that are either desirable or necessary for remote operations and maintenance. Commercially available, horizontal-type furnaces are not applicable to remote maintenance. The horizontal types require a large space, and involved mechanical systems are required to move material through the furnace. Therefore, it appears prudent to proceed with a development, design and test program to establish a remote, vertical furnace capable of sintering supercalcine pellets. This program could take advantage of the efforts performed to date for nuclear fuel pellets and use that prototype furnace for tests such as the following:

- cold tests to determine carrier grain size and composition, pressure drops versus grain size and pellet-to-carrier ratios, carrier-to-pellet ratio needed to protect pellets, and pellet/carrier flow characteristics
- tests of methods to separate carrier from pellets and to recycle carrier
- hot thermal tests to evaluate pellet/carrier compatibility and flow characteristics under sintering conditions, assess and eliminate pellet damage, establish preheat, sintering and cooling rates or times, and determine gas compositions required
- other thermal tests to determine impact of various moisture content of incoming pellets and the operating requirements and impacts if binders other than water are required or used.

Other tests and developmental efforts will be required to evaluate and determine the consequences if elements such as cesium, sodium or ruthenium volatilize during the sintering process and are condensed or deposited. In addition, the tendency for gas flow, opposite to the gravity flow of the pellets, to condense out volatiles on the incoming cold pellets and carrier is an area of concern that must be checked out and resolved if a problem. Finally, carrier and furnace contamination and cleanup needs to be evaluated and resolved.

When the above mentioned tests are completed, a production-sized furnace designed for remote operation, maintenance and replacement should be fabricated and operated in a facility where all aspects of the remote operations and maintenance can be verified before exposing the system to a radioactive environment.

F.7.5 PYROLYTIC COATER

Pyrocarbon coating by CVD is a well investigated technology due to the efforts expended in HTGR fuel development and production. The CVD technology for applying coatings to calcine and supercalcine particles was primarily developed at BCL (Rusin et al. 1978).

In the coated supercalcine processes, the diameter and porosity of the pellet core must be controlled through pelletizing operating parameters. The pellet diameter selection must be based on at least the following considerations: 1) pelletizing efficiency, 2) volume shrinkage during sintering, 3) sintering time, 4) coating ease, 5) temperature gradients within the pellets, and 6) waste loading of coated pellets. Similarly, the desired pellet porosity must be

determined with consideration of: 1) pellet strength, 2) thermal conductivity, 3) coating ease, 4) waste loading, and 5) process limitations. Larger pellets, while increasing waste loading by requiring less fractional coating volume, may increase sintering time and will contain larger temperature gradients within the pellets. Also, larger pellets are handled with greater difficulty in CVD coaters. Pellets with near zero porosity would have maximum strength, thermal conductivity and volumetric waste loading; however, CVD coating requires a certain amount of porosity to allow for any differences in thermal expansion of coatings and cores. A factor that must also be considered is the minimum porosity obtainable through the pelletizing and sintering processes.

Optimization of coating thickness requires evaluation of inertness and economics. Increasing coating thickness improves inertness, but at the same time increases cost and coating time. Two other factors to be considered are changes in thermal conductivity and waste loading with coating thickness; however, inertness and economics should outweigh the latter considerations in determining and optimizing coating thickness.

The following properties of uncoated pellets can be improved by coating (Newman and Kofler 1979):

- mechanical strength--the maximum applicable load increases with pyrolytic-carbon coating
- fines rubbing off--coatings prohibit undesired contamination by rubbed off fines
- porosity--coatings prohibit every penetration into the pellets and prevent sucking by capillary forces in a metal-pellets composite
- temperature resistance--temperature doesn't cause damage with pyrolytic-carbon coating under 1100⁰K in an oxidizing atmosphere
- thermal conductivity--the thin, good conductive layers raise the conductivity values
- suitability of metal embedding--the applied coating acts as an important barrier against leaching across the composite
- leach resistance--leach tests in Soxhlet extractors on pyrolytic-carbon coated granules resulted in negligible weight losses (but if the specimen included damaged or cracked granules the detected quantities of cesium in water after a leaching test approximated the data of uncoated pellets).

As covered above, coatings are technologically complex, but coated pellets of high-level waste do offer enhanced inertness over uncoated pellets. However, considerable research and development are required before this can be considered to be a viable process for a particular supercalcine composition and a remote production facility. The following initial basic research and development efforts are required:

- the development of a laboratory-scale coater with a capacity of approximately 1 L for applying pyrolytic-carbon coating and for demonstrating remote operations
- the continuation of the investigation and development of pyrolytic-carbon coating processes with an emphasis on applying these coatings at temperatures lower than those typically used at present

- an investigation of the handling and processing of the off gas, and the development of a process more adaptable to a radioactive environment and its ramifications.

The above efforts need to focus on correlating coating characteristics, compatibility with substrate and leach resistance with deposition temperature, carbon carrier, metal catalyst, reagent concentrations and reaction times. If it is not feasible to deposit carbon directly on the selected substrate because of 1) different thermal expansion coefficients, 2) agglomeration when coating is applied, 3) poor coating adhesion, and 4) reactions between coating and substrate, then alternatives for applying a barrier and/or crushable layers need to be examined for the substrate composition.

Some experimental efforts have been conducted to explore the lowest suitable temperature for coatings (Newman and Kofler 1979). The initial research and development efforts listed above need to determine whether leach-resistant carbon coatings can be applied to supercalcine pellets by the low-temperature approach. If not, processing parameters using the lowest possible deposition temperature need to be identified.

The laboratory-scale coater, as noted above and identified as having the best potential of fulfilling the program objectives, needs to be designed, fabricated and operated to demonstrate its ability to perform the desired functions. Larger-scale models for full remote operation and maintenance could then be designed based on the results of the laboratory-scale tests. In particular, coater off-gas systems and components, their attributes and design parameters, need to be identified, fabricated and tested to assure the safe and economic operations of the coater station in a radioactive environment.

F.7.6 CANISTER FILLING AND HANDLING

The research and test efforts identified in the MIL process (Appendix D) for canister filling and in the reference process (Appendix A) for canister handling and closing will also be required for the PCCPIM process. In addition, selected components and aspects of the PCCPIM canister handling system should be fabricated and tested to verify their capabilities and remote aspects.

F.8 ALTERNATIVE PROCESSES AND EQUIPMENT

Only a very limited time has been available to develop the preconceptual design bases that are presented in this document for the PCCPIM process. As such, there are a number of viable alternatives that have no doubt not been considered or that should be explored further in the subsequent steps of finalizing a process design. Some of the alternatives noted in the reference process may also be applicable to the PCCPIM process, particularly those pertaining to the spray calciner. Other alternatives are noted below.

F.8.1 CELL ARRANGEMENT

The cell arrangement displayed in Figure F-3 shows all of the remote processing equipment located in one cell or canyon that is quite high and long. There is a strong possibility of lowering building costs if this size could be reduced; cell height is of particular concern. In addition, to limit and control the potential spread of contaminants, it may be desirable to locate the process equipment in two or more cells. This alternative needs to be considered while at the same time giving adequate attention to the flow of process material and the requirements for the remote operation, maintenance and replacement of equipment.

F.8.2 DISC-PELLETIZER

As covered in Section F.7.3, calcine must be fed to the pelletizer at a controllable and uniform rate. Tests with a vibratory-screw feeder have indicated considerable variances in feed rate. Other feeder types, such as gravimetric and loss-of-weight, were considered but have not been used to date because preliminary evaluations indicate more serious control or remote operational problems with these. Calcine feeder alternatives need to be evaluated further early in the research and development program.

Systems other than a disc-pelletizer that have or could be used to make supercalcine pellets include stirred-bed calciners, fluidized-bed calciners and pellet presses of different kinds or configurations. Early in any additional efforts on the PCCPIM process, these alternatives should be considered in terms of their application in a remote, production-scale operation.

F.8.3 SINTERING FURNACE

The application of microwave energy to dry pelletized HLLW has been tested (Priebe et al. 1979). Preliminary tests indicate hard, unbroken pellets of at least some compositions can be produced in 1/10 to 1/3 of the time required in a radiant type furnace. Additional tests and evaluations are planned at Idaho National Engineering Laboratory. These tests should be followed on a continuous basis, and if the advantages appear to outweigh the disadvantages, due consideration should be given to developing this process as an alternative to the vertical sintering furnace.

F.8.4 PYROLYTIC COATER

Pyrolytic-carbon coatings have been applied to simulated wastes by the use of fluidized beds, vibrating beds and drum coaters with good success on a preliminary basis and a small scale. Depending on how these processes adapt to large-scale, remote operations, they may be viable alternatives to the coater concept identified herein. Additional evaluations and laboratory-scale tests using these processes on supercalcine waste pellets should be done if this concept should prove not to be adaptable.

F.8.5 PNEUMATIC SYSTEMS

An alternative that may reduce in-cell components, but would increase cell height is the use of gravity instead of pneumatics to convey pellets from the sintering furnace station to the surge hopper. This alternative should be evaluated further.

F.8.6 MATRIXING

As noted in Section F.6.16, for the purpose of this study the metal-melting furnace is located outside of the process cell. An alternative would be to locate the furnace in the cell or to bring ladles of molten lead into the contaminated zone. This may be worthy of additional study.

For this study, it is also assumed that the metal matrix is lead or a lead alloy. Material availability and cost may preclude these from being viable materials in a production facility. Due to the softening temperature of the waste pellets, only a limited number of metals or alloys may be applicable. The use of aluminum is a viable alternative that needs to be considered further; other metals or alloys should also be evaluated.

F.9 DRAWINGS AND DATA SHEETS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

TABLE F-3. Equipment Data Sheet for Supercalcine HLLW Feed System

Function

Receive, store and analyze HLLW; prepare calciner feed solution and deliver to spray calciner.

Interfaces

HLLW supply; chemical make-up; spray calciner.

Design Parameters

Remote operation; removal and replacement; stainless steel construction; vigorous agitation systems; handle soda-rich solutions; accurate analysis of solutions; intertank transfer to and from all tanks; two storage tanks; one feed tank.

Operating Conditions

Same as reference process except larger tank capacities.

Features

Same as reference system except more accurate HLLW sampling and analytical systems, supercalcine addition system and thorough mixing systems.

Services or Auxiliaries

Same as reference process.

Space Requirements

Maintenance Methods

Remote.

Remarks

TABLE F-4. Equipment Data Sheet for Air-Lock Valve

Function

Isolate hopper and spray calciner environments.

Interfaces

Spray calciner; calcine hopper.

Design Parameters

Self-sealing between entrance and exit or purgable; radiation resistant; remote operation, installation and replacement.

Operating Conditions

Temperature = 150° to 250°C; 10 to 60 rpm; 10 to 60 kg/h of calcine.

Features

Stainless steel construction; variable-speed drive; sealed closed-loop cooling; spray-flush system.

Services or Auxiliaries

Electrical; rpm monitor; temperature sensors; compressed air; nitric acid; water; drain.

Space Requirements

2 ft by 3 ft by 1 ft high.

Maintenance Methods

Remote

Remarks

Special design but similar to: The Young Industries Inc. rotary valves; Wm. W. Meyers and Sons, Inc. Roto-Flo air-lock feeder valves; Sprout-Waldron Koppers Co., Inc. modular rotary valves; or Joy Manufacturing Co. air-lock or dust valves.

TABLE F-5. Equipment Data Sheet for Calcine Hopper

Function

Provide cooled and uniform feed supply.

Interfaces

Air-lock valve; calcine feeder.

Design Parameters

Stainless steel construction; remote operation, replacement and installation; 2-h surge capacity (2 ft³).

Operating Conditions

Temperature = 75^o to 90^oC; flow rate = 30 kg/h.

Features

Vibrators; closed-loop cooling; spray-flush system.

Services or Auxiliaries

Water; drain; air; electrical; temperature; detectors; weight or fill detectors; pressure sensors; nitric acid.

Space Requirements

6 in. by 24 in. by 48 in. high

Maintenance Methods

Remote

Remarks

Special design for this application and selected feed system.

TABLE F-6. Equipment Data Sheet for Pelletizer Feeder

Function

Provide consistent feed rate to disc-pelletizer.

Interfaces

Calcine hopper; disc-pelletizer.

Design Parameters

Stainless steel construction; radiation resistant; automatic and remote operation; remote installation and replacement; resistant to nitric acid wash-down.

Operating Conditions

Temperature = 75° to 90°C; 0 to 100 kg/h calcine;

Calcine Properties: shape--agglomerated semi-spherical particles; particle density = 2.8 g/cm³ (typically approximately 4 g/cm³); bulk density = 0.5 to 1.3 g/cm³ (typically 1.2 g/cm³); particle size = 1 to 10 μ ; particle hardness is approximately 5 to 7 Mohs.

Features

Variable speed; flow detection; spray-flush system.

Services or Auxiliaries

Electrical; rpm monitor; temperature detectors; nitric acid; compressed air.

Space Requirements

2 ft by 4 ft by 2 ft high

Maintenance Methods

Remote

Remarks

Special application that adapts Vibra Screw Inc. screw feeder; K-TRON Corp. feeder; Acrison Inc. feeder; Merrick Scale Mfg Co. Flow Star feeder or similar device.

TABLE F-7. Equipment Data Sheet for Pelletizer

Function

To form calcine particles into larger pellets or spheres.

Interfaces

Calcine feeder; separator.

Design Parameters

Enclosed operations; stainless steel construction; automatic and remote operation; remote replacement and removal; design rate = 30 kg/h.

Operating Conditions

Temperature = 35^o to 90^oC; rate = 0 to 50 kg/h;

Pellets: shape is spherical 1/8 to 1/4 in. in dia; density = 0.8 to 1.5 g/cm³; moisture content is approximately 30%.

Features

Variable speed; binder feed; variable angle; variable nozzle locations; spray-flush system; sealed enclosure; rate monitor.

Services or Auxiliaries

Electrical; pressure sensor; temperature sensors; speed and position indicators; nitric acid; compressed air; ventilation supply and exhaust.

Space Requirements

2 ft by 2 ft by 2 ft high

Maintenance Methods

Remote

Remarks

Special adaptation of small, commercial pelletizer such as Ferrotec Inc. 16-in. pelletizer; Sprout-Waldron Koppers Co., Inc. Model 400, or smaller models.

TABLE F-8. Equipment Data Sheet for the Green Classifier/Separator

Function

Separate large and undersized material from acceptable spheres.

Interfaces

Disc-pelletizer; enclosure.

Design Parameters

Stainless steel construction; automatic and remote operations; remote replacement and installation.

Operating Conditions

Temperature = 35⁰ to 90⁰C; rate = 0 to 50 kg/h; fines are less than 1/8 in.; oversized particles are greater than 1/4 in.; product is 1/8 to 1/4 in. in dia.

Features

Remote adjustments; spray-flush system.

Services or Auxiliaries

Electrical; compressed air; sample station; nitric acid; instrumentation.

Space Requirements

3 ft by 4 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Special installation that will require a device similar to SWECO Inc. Model LS24544 separator; Eriez Magnetics Model 361 separator; or Sprout-Waldron Koppers Co., Inc. classification machines.

TABLE F-9. Equipment Data Sheet for the Wet Scrap Tank

Function

Collect wet scrap and spray-flush solutions.

Interfaces

Spray calciner; calcine hopper; disc-pelletizer station; HLLW feed system.

Design Parameters

Stainless steel construction; automatic and remote operations; remote installation and replacement.

Operating Conditions

Temperature = 25° to 100°C; nitric acid solutions.

Features

Closed-loop cooling; agitation system(s); jet transfer; specific gravity; weight factor; temperature pressure sensors.

Services or Auxiliaries

Steam; air; electrical; water; drain; instrumentation.

Space Requirements

3 ft in dia by 3 ft high.

Maintenance Methods

Remote.

Remarks

Special design required.

FIGURE F-4. Vertical Sintering Furnace

TABLE F-10. Equipment Data Sheet for the Sintering Furnace

Function

Reduce moisture content and increase density of pellets by solid-state sintering.

Interfaces

Green pellet classifier/separator; sintered classifier/separator.

Design Parameters

Automatic and remote operations; remote replacement and installation; design rate = 28 kg/h.

Feed material: spherical 1/8 in. to 1/4 in. in dia; density approximately 0.8 to 1.5 g/cm³; moisture content is approximately 30%.

Outlet material: 0.100 in. to 0.250 in. in dia; density = 3.0 to 4.0 g/cm³

Operating Conditions

950^o to 1250^oC for 2 h; heating @ 300^oC/h; cooldown @ 400^oC/h.

Features

Variable flow rate; temperature range up to 1400^oC; off-gas system connection; pellet/carrier separator; carrier conveyor and feeder system; closed-loop cooling; modular construction.

Services or Auxiliaries

Electric power 120 V, 480 V, 45 kW connected, and 80 kW demand; temperature detectors; flow indicators; water supply and drain 25 gpm; instrument system has 1 kW of isolated shielded voltage.

Space Requirements

5 ft in dia by 15 ft high

Maintenance Methods

Remote

Remarks

Special design required.

TABLE F-11. Equipment Data Sheet for the Sintered Classifier/Separator

Function

Separate large or fused hunks of material from standard size pellets; provide sample station interface.

Interfaces

Vertical sintering furnace; coater system; mill.

Design Parameters

Automatic and remote operations; remote installation and replacement.

Operating Conditions

Temperature = 250 to 1000°C; rate = 0 to 50 kg/h; fines are less than 1/16 in. in dia; oversized particles are greater than 1/4 in.; product is 1/10 in. to 1/4 in. in dia.

Features

Remote adjustment; temperature sensors; pressure sensors; flow indicators.

Services or Auxiliaries

Electrical; sample station; instrumentation.

Space Requirements

3 ft by 4 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Special installation, similar to that of green pellet classifier.

TABLE F-12. Equipment Data Sheet for the Pellet Mill or Crusher

Function

Reduce sintered pellet material to conveyable particle size.

Interfaces

Separator/classifier; pellet hoppers; pneumatic system.

Design Parameters

30 kg of supercalcine pellets/h; remote operations, removal and replacement; material is less than 8 in. ring size; discharge material is less than 1/16 in. maximum dimension; process material specific gravity = 4.0 and hardness = 5 to 7 Mohs.

Operating Conditions

Internal temperature is ambient, 25^o to 150^oC; external temperature is ambient, 25^oC.

Features

Stainless steel and abrasion resistant material; remote attachments.

Services or Auxiliaries

Electrical; instrumentation.

Space Requirements

2 ft by 3 ft by 4 ft high

Maintenance Methods

Remote

Remarks

Special application by adapting commercial devices such as Sprout-Waldron Koppers Co., Inc. crushers or impact mills; Alpine American Corp. hammer or cutting mills; or Mikropul Corp. pulverizing machinery.

FIGURE F-5. Coater Surge Hopper

TABLE F-13. Equipment Data Sheet for the Coater Surge Hopper

Function

Provide surge storage and batch feed to water.

Interfaces

Classifier/separator; coater.

Design Parameters

Remote installation and operation; stainless steel construction; 40-L capacity.

Operating Conditions

25° to 600°C; 0 to 50 kg/h.

Features

Oxygen-tight exit valve or system.

Services or Auxiliaries

Cooling water; drain.

Space Requirements

1-1/2 ft in dia by 4 ft high

Maintenance Methods

Remote

Remarks

Special design.

FIGURE F-6. Coater

TABLE F-14. Equipment Data Sheet for the Pyrolytic Coater

Function

Coat waste pellets with layers of pyrolytic carbon.

Interfaces

Coater surge hopper; pneumatic system; coater off-gas system.

Design Parameters

Automatic and remote operations; remote installation and replacement; feed material pellets are 0.100 to 0.250 in. in dia, 3.0 to 4.0 g/cm³ design rate = 30 kg/h; batch size and rate = 50 kg/h.

Operating Conditions

350° to 600°C (depends on gas); low air or oxygen content and controlled environment; 800° to 900°C for periodic carbon burn-off.

Features

Off-gas treatment system; variable batch size; purge; controlled environment.

Services or Auxiliaries

Electrical 35 kW capacity, 15 kW average use rate; compressed air; N₂; NiCO₄; C₂H₂; argon; water.

Space Requirements

2 ft in dia by 5 ft high

Maintenance Methods

Remote

Remarks

New and special design required.

FIGURE F-7. Coater Off-Gas System Flow Diagrams

TABLE F-15. Equipment Data Sheet for the Pneumatic System

Function

Transport pellets and pellet particles.

Interfaces

Pellet coater; pellet surge hopper; scrap collection hopper; scrap mill.

Design Parameters

Transport 30 kg pellets/h; specific gravity = 4.0; particle hardness = 5.9 Mohs; temperature = 100° to 500°C; remote operations; in-cell components remote monitoring, removal and replacement.

- prime mover sized to meet maximum load requirements of system with oversized motor so allowance for additional loads imposed on system
- smallest pipe size(s) capable of carrying design capacity within pressure drop constraint
- piping run routing to minimize line length and number of turns
- downward or inclined lines for process material to be avoided
- process material to enter through air locks

Operating Conditions

Maximum inlet vacuum = 15 in. Hg; system pressure drop = 5 psi; solids-to-gas ratio range = 10 to 20; conveying velocity = 2500 to 5000 ft/min; terminal velocity = 3000 to 4000 ft/min.

Features

Positive-displacement blower: variable speed; 2000 to 4000 rpm; 50 to 150 acfm @ 10 psia inlet and 16 psia outlet

Pipe: stainless steel; 1 to 2 in. in dia; mill run 2B interior finish; 10 to 20 ft sections; bend radius for particles 2 ft or greater, for pellets 4 ft or greater

Connections: Cherry Burrell's I Line or similar types; remote flanged joint if required for rigidity

Filters: sintered 300-series stainless steel with short pulsed air blow back

Line valves: full ported ball valves

- Air flow with flow deviation control will be provided upstream of the initial process entry point for process transfer run.
- For closed-loop systems, a cooler will be provided to remove the heat added to the air by the compressive action of the blower. The temperature of the air stream at the process entry point will be approximately 100°F.
- A surge tank will be provided upstream of the process entry point to dampen air flow pulsations.
- Pressure controls will be provided at the line inlet to assure a negative pressure in the transfer line such that 13.7 psia is not exceeded.
- Dry air (dew point 0°C or lower) will be introduced to the conveying line at a negative pressure of about 13 psia and at an adequate linear flow velocity.
- Differential pressure will be monitored across all filter systems.

Services or Auxiliaries

Compressed air; conditioned air; instrumentation; drain.

Electrical: capacity = 10 kW; average use rate = 4 kW

Water: capacity = 100 L/h; average use rate = 40 L/h

Maintenance Methods

In-cell components--remote.

Remarks

Special design required for this installation.

TABLE F-16. Equipment Data Sheet for the Pellet Surge Hopper

Function

Provide storage for pellets between continuous and batch operations of process.

Interfaces

Pneumatic transport system; volume hopper.

Design Parameters

Remote operations, removal and replacement; contain up to 110 L of pellets; stainless steel construction.

Operating Conditions

Temperature range = 25^o to 450^oC; pressure range = 10 in. Hg vacuum to 25 psig.

Features

Load cells or level sensors; cooling coils; temperature sensors; pressure indicators.

Services or Auxiliaries

Instrument system; water; drain; air or electrical.

Space Requirements

Maintenance Methods

Remote.

Remarks

Design similar to that of the MIL surge hopper.

TABLE F-17. Equipment Data Sheet for the Volume Hopper

Function

Measures and limits batch size to be inserted in a canister.

Interfaces

Surge hopper; canister fill mechanism; recycling mill.

Design Parameters

Stainless steel construction; remote operations, removal and replacement; limit batch size to that required by a canister design (65 L).

Operating Conditions

Temperature range = 25^o to 450^oC; pressure--slightly negative to slightly positive.

Features

Load cells or level sensors; cooling provisions; temperature sensors; inlet valve--remote operation.

Services or Auxiliaries

Instrument system; drain; compressed air; electrical; water.

Space Requirements

Maintenance Methods

Remote

Remarks

Similar to that in MIL process.

TABLE F-18. Equipment Data Sheet for the Canister Fill Mechanism

Function

Connects volume hopper with canister basket; directs and controls flow of marbles.

Interfaces

Volumetric hopper; canister.

Design Parameters

Remote operations; removal replacement; stainless steel construction.

Operating Conditions

Temperature range = 25^o to 450^oC.

Features

Mate with canister; vertical movement.

Services or Auxiliaries

Vacuum; inert gas; compressed air or electrical.

Space Requirements

Maintenance Methods

Remote

Remarks

Similar to that of MIL process.

TABLE F-19. Equipment Data Sheet for the Metal Fill Station

Function

Supply molten lead to the canister.

Interfaces

Canister; metal-melting furnace.

Design Parameters

Remote operations; removal and replacement.

Operating Conditions

Temperature range = 25⁰ to 400⁰C; pressure range = 0 to 125 psig.

Features

Mate with canister.

Services or auxiliaries

Electrical power; instrument systems; inert gas; compressed air; cooling water; hydraulics; drain.

Space Requirements

Maintenance Methods

Remote.

Remarks

Similar to that in MIL process.

TABLE F-20. Equipment Data Sheet for the Metal-Melting Furnace

Function

Transform solid lead into the liquid form.

Interfaces

Metal fill mechanism.

Design Parameters

Operating Conditions

Temperature = 0⁰ to 450⁰C.

Features

Control of pouring rate and temperature; slag-free pouring.

Services or Auxiliaries

Electrical power; cooling water; instrument system; drains; ventilation supply and exhaust.

Space Requirements

Maintenance Methods

Contact.

Remarks

Identical to furnace in MIL process.

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APPENDIX G

SUPERCALCINE HOT-ISOSTATIC PRESSING PROCESS

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APPENDIX G
SUPERCALCINE HOT-ISOSTATIC PRESSING PROCESS

1.0 INTRODUCTION

Hot-isostatically pressed supercalcine is an alternative waste form using supercalcine (McCarthy 1977) as the ceramic host medium. Supercalcine is produced by adding the oxide of Si and the nitrates of Ca, Al and Sr to the HLLW solution before calcination. These components are added in carefully defined proportions, so that during calcination they will react with the components of radioactive waste to form assemblages of desired crystalline phases having apatite, fluorite, scheelite, pollucite and spinel structures. An important objective is to ensure crystallization of ^{137}Cs (one of the most dangerous and mobile species) in the stable and insoluble mineral pollucite ($\text{CsAlSi}_2\text{O}_6$).

In pelletized supercalcine waste forms, such as those covered in Appendices E and F, supercalcine is formed into pellets and fired at about 1200°C for 1 to 2 h. This results in the formation of a fine crystalline, but moderately porous phase assemblage in which radioactive waste atomic species are distributed according to the principle of chemical equilibrium. These pellets are then either placed in a lead matrix or are coated with layers of pyrolytic carbon to enhance leach and impact resistance and then placed in the lead matrix to improve thermal stability and mechanical strength. However, an inherent disadvantage of the supercalcine pellet waste forms is the relative solubility of the moderately porous host material as compared to that of a dense host material.

In the envisioned SCHIP process, the mixture of oxide additives and the radioactive waste are combined and heat-treated at 1150°C to form large, sintered billets having a porosity of about 20%. The billets are then vacuum-degassed, sealed, and isostatically pressed at about 1150°C and 100 MPa (15,000 psi) to form the desired crystalline grain size, uniformity and dense phase assemblage approaching 100% of that theoretically possible. Leach resistance should be considerably enhanced because of the greatly reduced permeability and surface area. The mechanical strength of the host material should also be increased because of the stress-crack healing capabilities of the hot-isostatic pressing process.

The SCHIP process is more complex than the reference process (Appendix A) and more costly because of the high cost of hot-isostatic pressing equipment. It does have a lower risk of product quality abnormalities, but requires more safeguard, equipment and cell design considerations.

The SCHIP process facilities will have the capability to 1) proportion and thoroughly mix supercalcine-forming chemicals into the HLLW, 2) calcine the solution and form specific assemblages of desired crystalline phases, 3) collect and heat-treat the supercalcine crystals into sintered billets, 4) vacuum-degas, seal and hot-isostatically press the sintered billet to a desired density, crystalline grain size and uniformity, 5) load the hot-isostatically pressed

billet into a canister, 6) close and seal-weld the canister, 7) check the canister seal, 8) decontaminate the canister, and 9) transport the canister to an interim storage area prior to final disposition.

G.2 PROCESS DESCRIPTION

The SCHIP process is depicted in the process flow diagram shown in Figure G-1. As covered in the reference process (Appendix A), HLLW (the basic composition is given in the Overall Design Bases Section) is received at the waste facility and is stored in the HLLW feed system. Here the HLLW is modified with selected liquid additives to form a supercalcine solution. Solution additives, usually including Ca, Sr, Al and Si, are selected according to the HLLW composition and are thoroughly mixed with the HLLW to comprise 22 wt% of the supercalcine solution (78 wt% waste loading). The solution can be sampled and necessary adjustments made before transferring it to the spray calciner feed tank.

The supercalcine solution is continuously fed into the calciner at a controlled operating rate of 203 L/h through a pneumatic atomizing spray nozzle. Crystalline supercalcine particles and an off gas are produced in the heated calciner chamber. The supercalcine particles flow via gravity through a delivery system into a sintering furnace at an operating rate of 21 kg/h.

In the sintering furnace, supercalcine particles accumulate in an alumina container that is sealed to the delivery system. When the container is filled with the desired quantity, the supercalcine flow is diverted to a second, adjacent furnace. The first furnace is then closed off from the delivery system with an insulation plug in the cover and the supercalcine is heated to its sintering temperature.

During sintering, the accumulated supercalcine uniformly shrinks to approximately 19% of its original bulk volume, and it loses 10% of its weight through volatilization. The resulting sintered billet is cylindrical and roughly proportional to the container dimensions.

When the billet has cooled below the radioactive waste volatilization temperature, the furnace is translated radially out from under the delivery system so that the container and sintered billet can be moved through an enclosure hatch with an overhead crane. The container is decontaminated with superheated steam and is placed in a holding furnace where glass beads are added. Glass beads having a dense-packing particle size distribution are metered into the container in sufficient volume to completely submerge the billet when the glass beads are subsequently fused.

After the glass beads have been added, the container is transported with an overhead crane and placed in a HIP. A cover and yoke are then positioned to seal the hot-isostatic pressing vessel. Initially, pressure in the hot-isostatic pressing vessel is reduced to degas the sintered billet and the glass-bead pack. Then the temperature of the alumina container is increased radiantly to fuse the glass beads and hermetically encapsulate the sintered billet. When the glass is completely fused, argon gas is pumped into the vessel and the vessel is pressurized. Under pressure and high temperatures, the fused glass acts as an impermeable, hydrostatic fluid to densify the submerged supercalcine billet to essentially 100% of theoretical.

The HIP is then concurrently depressurized and somewhat cooled for opening and removing the alumina container. The relatively hot container is transferred to an overpack canister with an overhead crane. At this time, crushed hulls may be added to the alumina container above the glass level.

FIGURE G-1. Flow Diagram for SCHIP Process

A helium source is then added to the canister, and its closure is installed and seal-welded. After the weld is leak-tested, the canister is inspected, decontaminated and transported to storage.

G.3 BASES AND ASSUMPTIONS

The preconceptual design of a production process for hot-isostatically pressed supercalcine, requires a number of assumptions in terms of the process bases, canister, effluents, waste, scrap, utilities and materials. These are generally identified in Section 3, Overall Process Bases. Assumptions specific to the SCHIP process are identified in the following subsections.

G.3.1 PROCESS BASES

As indicated previously, the process flow diagram for the SCHIP process is shown in Figure G-1. The diagram is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. Design capacities are 1.5 times the nominal operating values. Data on the HLLW is presented in the Overall Process Bases Section and in Appendix A. Additional processing data is listed below:

- Supercalcine solution feed to the spray calciner
 - HLLW loading = 78 wt%
 - operating rate = 203 L/h
 - design rate = 305 L/h
 - heat loading = 2.7 W/L
 - oxide content (in HLLW and supercalcine formers) = 92.6 g/L
- Calcination output (with volatiles)
 - volatile residue = 10 wt%
 - operating rate = 20.8 kg/h
 - design rate = 31.2 kg/h
 - bulk density = 0.8 kg/L
 - heat loading = 62.2 W/kg
- Sintered billet
 - heat loading = 3060 W
 - bulk charge (with volatiles) = 117 kg (146 L)
 - charge time--
 - operating = 5.6 h
 - design = 3.7 h
 - thermal cycle time = 11.0 h
 - furnaces required = 4
 - sintered density = 3.8 kg/L
 - sintered weight and volume (without volatiles) = 105 kg and 27.6 L
- Hot-isostatic pressing
 - cycle time (including degassing and glass fusion) = 10.6 h
 - HIPs required = 3

glass sealing medium--
density = 2.3 kg/L
bulk charge (beads) = 133 kg (76.9 L)
fused volume = 57.7 L
hot-isostatically pressed supercalcine--
density = 4.9 kg/L
billet weight and volume = 105 kg and 21.4 L

- Overpack canister
operating rate = 1285/yr (4.3/d)
design rate = 1925/yr (6.4/d)
- End product
heat loading = 3060 W
weights--
supercalcine = 105 kg
hulls = 223 kg
glass = 133 kg
alumina container = 125 kg
canister (304L) = 196 kg
TOTAL = 782 kg

G.3.2 CANISTER

Some general requirements for waste canisters are given in the Overall Process Bases Section and are discussed in more detail in Appendix A. Specifics for the SCHIP process canister are noted here. The canister, or end product, is shown in Figure G-2. It consists of a sealed, 304L stainless steel outer shell with appropriate lifting means and an open alumina container that holds the hot-isostatically pressed supercalcine billet completely submerged in glass. The alumina container and glass are for processing purposes only and are not considered a part of the HLLW immobilization system because of their expected thermal cracking during storage.

For supercalcine with a 78 wt% waste loading, the size of the canister is limited by the allowable heat content (3060 W/canister). The alumina container dimensions are selected to contain an equivalent bulk charge of "as-calcined" supercalcine (146 L). A maximum, practical container height, within the 3-m canister height limitation, was selected to minimize the canister diameter for the purpose of minimizing hot-isostatic pressing equipment cost, thermal response times and the centerline temperature of the supercalcine billet during storage. Allowing for 5% overfill capacity, a nominal charge depth of 2.7 m was selected, resulting in an inside container dia of 26.3 cm. The resulting outside dimensions of the canister are expected to be about 32 cm in dia by 3 m high.

Based on the size of the container and on the previously identified process bases, the hot-isostatically pressed supercalcine billet dimensions are about 14 cm in dia by 1.4 m high.

FIGURE G-2. Canister

Approximately 57.7 L of fused glass are required to submerge the billet during hot-isostatic pressing. The void in the alumina container above the glass level may be filled with approximately 223 kg of crushed, fuel rod hulls prior to closing the canister. This represents about 54% of the hulls from which the contained radioactive waste was extracted.

The total weight of the canister, including the optional hulls, is estimated to be 782 kg. The contained supercalcine represents about 13% of this weight. On this basis, about 0.5 MT of total canister weight will be required per MTHM processed.

Based on estimated thermodynamic properties and preliminary calculations, the steady-state centerline temperature of the supercalcine billet is roughly 740°C when the canister wall is held at 100°C . The corresponding maximum temperature of the glass is roughly 595°C , which is about the expected annealing point of the glass medium. If so, the glass should provide adequate support against billet shift, based on the typical 50°C annealing temperature allowance before deformation is noticeable and on the estimated 75°C thermal gradient through the glass.

Should a canister of this design be exposed to 50°C air storage, the centerline temperature may reach 825°C with a corresponding maximum glass temperature of roughly 690°C . Gradient shifting of the billet and the glass may occur if the canister is not upright; however, the glass temperature is well below the expected 820°C softening point and should maintain billet position in an upright canister.

G.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

For this study, it is assumed that the effluents from the SCHIP process are similar in content and are the same order of magnitude in volume as those from the reference process. Except for the additional equipment for this expanded and more complex process that eventually will have to be discarded, it is assumed that the wastes from this process are the same order of magnitude in volume and contents as the reference process. Systems and provisions are assumed to be included in the fuel reprocessing plant to treat, reduce in volume and package both liquid and solid wastes. All solidified process scrap that may contain radioactive waste is recycled and incorporated into the SCHIP process.

G.3.4 UTILITIES AND MATERIALS

Estimates of the utility services and essential materials required for the SCHIP process are given in Tables G-1 and G-2, respectively. These order-of-magnitude estimates are based on the requirements for the reference process with appropriate adjustments for differing process steps and anticipated space and volume changes.

As with the reference process, only the utilities used in the SCHIP process and provided from the fuel reprocessing plant are shown; any emergency utilities required, such as electricity and cooling water, are supplied from the fuel-reprocessing-plant emergency services. Maintenance and replacement materials are also not shown. Only chemicals used for periodic facility decontamination and cleanup are included.

TABLE G-1. Estimate of Utilities Required for SCHIP Process

<u>Utility</u>	<u>Required Capacity</u>	<u>Average Use Rate</u>	<u>Disposition</u>
Electricity	3850 kW	2500 kW	--
Raw water	2800 L/min	1300 L/min	Recycle or environment
Process water	600 L/min	40 L/min	Recycle as HLLW or ILLW
Sanitary water	40 L/min	25 L/min	Sanitary chemical sewer-tile field
Air	1100 scmm	600 scmm	As appropriate
Steam	30 kg/min	6 kg/min	As appropriate

TABLE G-2. Estimate of Materials Required for SCHIP Process^(a)

Material	Amount Needed per Year
Alumina container	1,285 (4.3/d)
Canisters	1,285 (4.3/d)
Glass beads	172 MT (572 kg/d)
Al(NO ₃) 9H ₂ O	35,500 kg
Ca(NO ₃) ₂	62,500 kg
30% colloidal SiO ₂	10,000 kg
Sr(NO ₃) ₂	5,500 kg
Caustic (19M)	8,000 L
Nitric acid (12.2M)	12,000 L
Helium sources	1,285 (4.3/d)
Argon	12,900 scm (43 scm/d)
Ammonia	120,000 kg
Detergent	1,000 kg

(a) Excludes maintenance materials and apparel.

G.4 FACILITY AND EQUIPMENT REQUIREMENTS

The SCHIP process is contained in a reinforced concrete structure similar to that shown in Figure D-3 for the MIL process (Appendix D). However, the number, size and arrangement of the cells differs from that shown in Figure D-3 and needs to be determined after more data is established on the SCHIP process and its equipment. The overall requirements for the waste facility and its support groups and services are covered in the Overall Process Bases Section. The requirements for remote operation and remote maintenance of all in-cell equipment are the major concern when establishing the bases for a process installation. The major pieces of equipment and their approximate space requirements are given in Table G-3.

TABLE G-3. Process Cell Equipment Requirements and Dimensions

<u>Equipment</u>	<u>No. Required</u>	<u>Dimensions (Width x Length x Height), m</u>
HLLW batch tank	2	3 x 3 x 4
HLLW feed tank	1	2 x 2 x 3
Spray calciner	1	3.5 x 4 x 6
Delivery system	1	3 x 3 x 4
Sintering furnace enclosure	1	5 x 5 x 5
Decontamination station	4	2 x 2 x 4
Glass-charging station	2	2 x 2 x 6
Hot-isostatic press	1	4 x 8 x 8
Canister loading station	1	1 x 1 x 6
Canister cooling rack	1	2 x 3 x 6
Lid-closure station	1	2 x 2 x 6
Leak-detection station	1	2 x 2 x 6
Decontamination station	1	2 x 4 x 7
Canister rack	1	2 x 3 x 6
Storage transfer	1	2 x 2 x 5

G.5 STATE OF PROCESS DEVELOPMENT

Although considerable research has been conducted on simulated supercalcine by McCarthy (1977) and colleagues and at PNL, to date only small samples of this material have been hot-isostatically pressed and these samples have not yet been fully evaluated. Major problems have been precompaction of bulk supercalcine powder and container fracturing during hot-isostatic pressing. However, the fact that existing hot-isostatic pressing 1) technology removes porosity, 2) enhances strength and 3) results in desirable crystalline grain size and uniformity in ceramic powder materials points to the potential advantages of applying this process to significantly improve the immobilization of high-level waste in supercalcine.

Very small samples (5 cm^3) of simulated supercalcine have been successfully sintered in an alumina receptacle at PNL. However, material characterizations of bulk powder and of sintered supercalcine have not been performed.

At least one manufacturer of hot-isostatically pressed, consolidated-powder shapes (including ceramic powders) uses a vitreous or glass-like container in their pressing operation. The vitreous containers are claimed to have several advantages over metal cans because they are leak proof and deform uniformly under heat and pressure. However, the use of glass as the hydraulic fluid in the SCHIP process may be unique and is expected to require special properties and criteria.

Considerable research, development and demonstrations have been performed at PNL on spray calciners, solution preparation systems and other accessory systems. Calcine delivery systems operating in conjunction with sealed furnaces (to 1100°C) have also been developed and demonstrated. These accomplishments provide a sound basis for this segment of the SCHIP process.

Hot-isostatic pressing equipment has been developed in nonradioactive applications to exceed the size capacity and process requirements identified for this SCHIP process. Production systems with rapid throughput capability have also been developed. Application of hot-isostatic pressing in a radioactive environment with remote control has been proposed by ASEA (Sweden) and is being researched. At least two manufacturers of hot-isostatic pressing equipment have indicated the practicality of making the hot-isostatic pressing operation remotely operable. A comprehensive analysis of the radiological safety, including possible accidents, has been made in connection with ASEA's research, and the radiation emissions to the external environment are purportedly less than 1/1000 of the permissible standards.

The SCHIP process does require a number of involved process steps that must be operated and maintained remotely. This adaptation on a radioactive basis will be a major undertaking.

G.6 UNIT OPERATIONS

The overall SCHIP process consists of a number of unit operations. The function and operation of each, as they differ from the reference process, are described in greater detail in this section. Schematic drawings and equipment data sheets summarizing the design requirements are included at the back of the appendix in Section G.9. Required research and development work specific to the SCHIP process is also identified.

G.6.1 HLLW FEED SYSTEM

The HLLW feed system for supercalcine is basically the same as for the reference process except that it includes provisions to 1) more precisely sample and analyze the feed solution, 2) introduce and thoroughly mix supercalcine additives with the HLLW, and 3) contain and feed the larger volume that is required. The function of the HLLW feed system and requirements for its design are described in detail in Appendix A.

Precise chemical analyses of the HLLW must be taken to determine the proper proportions of supercalcine-forming additives. Supercalcine additives (Ca, Sr, Al and Si) may be introduced as solutions of strontium, calcium and aluminum nitrates and as an aqueous colloidal suspension of silica. These additives comprise 22 wt% of the final supercalcine waste form. Parameters for the supercalcine HLLW feed system that may differ from the reference process are noted in Table G-4 in Section G.9.

The basic HLLW feed system has undergone considerable development and evaluation, and its operating parameters and design requirements have been well established. Nevertheless, considerable research and development work is required to identify parameters and requirements for a particular supercalcine calciner-feed system. Areas of specific concern are:

- parameters for introducing and mixing supercalcine solutions into the HLLW (proportionate and redundant metering, agitation and the control of temperature, acidity and gelation)
- effects of radiation on supercalcine solutions
- accuracy required for HLLW sampling and analysis
- effects of recycle streams.

G.6.2 SPRAY CALCINER

The function, design and operation of the supercalcine spray calciner is basically the same as for the reference process except for a 28% greater capacity requirement. In addition, the SCHIP process does not require a system for feeding frit. These exceptions are noted in Table G-5 (see Section G.9).

The research and development efforts for the supercalcine spray calciner include those noted for the spray calciner used in the reference process and the following:

- determine the probability of sintered-calcine buildups during calcination of supercalcine solutions

- if buildups occur, develop and evaluate a detection method and a means of removing the buildups (e.g., water sprays, thermal cycling, vibration, etc.)
- evaluate the effect that supercalcine additives may have on catalyzing or suppressing explosive conditions with tramp impurities
- evaluate the effect of supercalcine additives on the volatility of species such as cesium and ruthenium and thus, on off-gas process requirements.

G.6.3 OFF-GAS SYSTEM

The function and operation of and the research and development work required for the SCHIP process off-gas system are the same as those for the reference ICGM process. However, the required capacity is about 30% greater than the reference process off-gas system. In addition, two parallel systems may be required for operating efficiency.

G.6.4 SUPERCALCINE DELIVERY SYSTEM

The continuous output of the spray calciner is directed to one of four, batch sintering furnaces by the supercalcine delivery system. Process off-gas from the other three furnaces concurrently flows upward through the delivery system to the off-gas system.

Figure G-3 in Section G.9 shows the delivery system. It consists of a four-way diverter and four identical delivery tubes, each with an associated mechanism for remote coupling to an alumina container positioned in the sintering furnace. A dust-tight coupling is accomplished by a weighted spout that compresses a seal retained in the container lip. A disc-bellow interfaces with the delivery tube and the spout to seal the system while accommodating thermal expansion and spout decoupling for container exchange. An insulated plug is used to close the spout opening during the sintering cycle and container exchange. The plug allows the venting of off gas, but prevents appreciable losses of residual supercalcine when closed. The spout nests into the furnace cover and is lifted with the cover to permit radial transferring of the furnace for container exchange.

An offset funnel inside the diverter directs the flow of supercalcine to one of the delivery tubes, which is attached and sealed to the alumina container. A rotary actuator automatically signals the funnel when the desired quantity of supercalcine has accumulated in the sintering container. The quantity of supercalcine that has accumulated may be detected by a variety of methods (e.g., radiation-activated level sensor, fill time, accumulated weight, etc.) depending on the consistency of calciner output and on detailed equipment design considerations. Table G-6 in Section G.9 summarizes the requirements of the supercalcine delivery system.

A two-position diverter has been developed and thoroughly evaluated at PNL as a part of the ongoing process demonstrations. Nevertheless, a four-way diverter and supercalcine delivery system must be designed and demonstrated. Developmental considerations are:

- positive control of the supercalcine flow and dusting

- the seal between the spout and the alumina container
- identification of reliable, radiation-resistant actuators and bearings
- prevention of excessive supercalcine accumulations (caused by sintering, condensation of off gases, etc.)
- controlling the spread of residual supercalcine during container exchange.

G.6.5 SINTERING FURNACE

The supercalcine-loaded alumina container is heated by a sintering furnace, which dries and densifies the supercalcine into a rigid, cylindrical billet. The density increases from about 0.8 kg/L to about 3.8 kg/L, or about 78% of theoretical.

Four identical sintering furnaces are arranged to mate-up with the furnace covers described as a part of the supercalcine delivery system. Each furnace is supported on a track and mechanized to translate radially out from under its cover to facilitate alumina container exchange. The neck of the container is shaped to facilitate remote coupling and decoupling with a lifting device, as well as sealing to the delivery system.

At the start of a cycle, an empty container is transported and lowered into an empty sintering furnace. Guides in the furnace center the container. The furnace is remotely translated under its cover; the cover and delivery tube spout are then lowered to engage the furnace and couple to the alumina container. The container is then preheated to about 650°C, and the insulated plug is retracted in preparation for delivery of supercalcine powder.

The alumina container is sized to accumulate 146 L of supercalcine powder. At a nominal calciner output rate of 21 kg/h and a bulk density of 0.8 kg/L, a nominal 5.6 h are required to accumulate this quantity. During this time, the first supercalcine powder in the furnace self-heats to approximately 750°C at the center of the container. The preheat temperature is selected to drive off residual moisture without excessive volatilization of cesium and other radionuclides.

When filled, the supercalcine flow is diverted to an alumina container in another furnace and the insulated plug is inserted to thermally close off the delivery system. The supercalcine charge is then heated to approximately 1150°C in a sintering thermal cycle similar to that shown in Figure G-4 (see Section G.9). Heating and cooling rates are moderated to prevent thermal gradients in the alumina container that could cause fracturing. The length of the sintering cycle and the maximum design calcining rate determine the number of sintering furnaces required. If the sintering cycle is 14.7 h (including a 3.7-h design base fill time), four furnaces are required. At the nominal flow rate of 21 kg/h, 5.6 h would be required to fill the 146-L container, and three furnaces would handle the sintering.

Once the centerline temperature of the billet is below the critical volatilization temperature for cesium, sodium and ruthenium (and others), the delivery system is uncoupled and the furnace is translated outward for container removal. Data on the sintering furnace is given in Table G-7 (see Section G.9) and a furnace is depicted in Figure G-5 (Section G.9).

A very small sample (0.01 L) of simulated, nonradioactive supercalcine powder has been uniformly densified during sintering in an alumina container. The material did not adhere to the container walls. Scale-up experimentation is required to determine the applicability of this approach at volumes 15,000 times that already demonstrated. A major factor is the time required to heat the bulk supercalcine powder, due to the inherently low thermal conductivity of powders. This factor will largely determine the number of furnaces required. Another consideration is the stability of the sintered billet within the alumina container during subsequent transport and charging with glass beads.

Other research and development considerations include:

- characterization of the bulk supercalcine material (thermal conductivity versus temperature; density versus temperature-time; specific heat, etc.)
- effects of internal heat generation on sintering shrinkage and on the heating and cooling rates
- uniformity of densification within a large, sintered billet
- cost effective design of an alumina container for thermal and mechanical durability
- design of a durable furnace and a remotely replaceable heating element to withstand the cleaning and thermal cycles
- development of a reliable, remotely operable furnace translator to withstand radiation and cleaning solutions
- determination of the frequency and amount of cleaning required
- development of a reliable, dust-tight seal between the alumina container and the delivery system
- positioning of the alumina container in the sintering furnace
- determination of the volatilization characteristics of bulk supercalcine and their effects on the quality of the supercalcine billet
- establishment of maximum cooling rates to maintain integrity of the sintered billet and alumina container.

G.6.6 FURNACE ENCLOSURE

A furnace enclosure is required to limit the spread of supercalcine process dust during accumulation, sintering and container exchange and to provide for its recycling. Although a fairly reliable, dust-tight seal is envisioned between the delivery system and the alumina container, experience in delivering the reference process calcine underscores the difficulty of absolute dust control. The sintering of supercalcine before breaking this seal should appreciably decrease airborne dust, as does glass fusion in the reference process. Nevertheless, supercalcine dust contamination of the furnace and alumina container is expected.

The enclosure is compartmentalized to isolate each furnace and prevent cross-contamination while facilitating individual spray-flush cleaning cycles between sintering cycles. The contaminated cleaning solution is recycled to the solution make-up tank. A remotely operable hatch provides access to each compartment for canister exchange. While the hatch is open, an air curtain is established to limit the spread of airborne contaminants outside the enclosure.

The enclosure provides flexible service connections to the furnace and furnace translator. It also includes provisions for the remote operation, monitoring, maintenance, removal and installation of the enclosed components.

Table G-8 (see Section G.9) summarizes the specifications for the enclosure shown in Figure G-6 (Section G.9). A large, developmental design effort is required to demonstrate an enclosure that is compatible with the internal equipment and its remote aspects. As a part of this effort, the effectiveness of an air curtain in preventing the spread of contaminants needs to be determined and demonstrated.

G.6.7 CONTAINER LIFTING DEVICE WITH COVER

A lifting device (carried by an overhead crane) automatically engages the alumina container in the sintering furnace through the opened enclosure hatch. The neck of the container is designed for this purpose. The lifting device has a temporary cover that seals the container opening (using the same captive seal that was used to seal the container to the delivery system). This cover prevents the spread of sintered particulates while moving the container through the air curtain of the furnace enclosure and prevents the entry of steam during subsequent container decontamination. After decontamination, the alumina container is transported to the glass-bead charging station where the cover is automatically removed during disengagement of the lifting device.

A developmental design effort is required to satisfy the lifting device specifications summarized in Table G-9 in Section G.9. Of particular concern is the integration of the lifting device design with the container neck design to provide positive, automatic engagement and disengagement and prevent excessive thermal shock or mechanical stress to the alumina container. The problem of handling broken containers must also be considered.

G.6.8 ALUMINA CONTAINER DECONTAMINATION

After sintering operations, the exterior surfaces of the alumina container are decontaminated to limit the spread of process dust to subsequent glass-bead charging and hot-isostatic pressing operations. The HIPs will most likely be located in a separate cell chamber (from calcination and sintering). The glass-bead charging station may be located in an intermediate chamber that serves as a pass-through air lock.

To reduce processing time, the alumina container is removed from the sintering furnace while still relatively hot. Steam, superheated to the container temperature, is used for decontamination to prevent excessive thermal gradients in the alumina container. Design

specifications for the decontamination station are summarized in Table G-10 (see Section G.9). The station is depicted in Figure G-7 (see Section G.9).

G.6.9 GLASS-BEAD CHARGING STATION

Glass beads are charged into the unfilled areas of the alumina container. When melted, the glass serves as the pressing fluid in the HIP. The glass-bead charging station consists of a holding furnace with provisions for spillage collection, an articulated spout, a metering valve, a thermally controlled delivery tube, air-lock valving, a preheat hopper (located outside the cell) and a spillage recycling pump and hopper.

After decontamination, the container is placed in the holding furnace, and its cover is removed with the lifting device. The furnace is maintained at about 350°C. The articulated spout is automatically positioned to direct the metered flow of glass beads into the container. The beads are preheated to about 350°C to avoid thermal shock to the alumina container or to the sintered billet. Approximately 77 L of beads, having a 75% free-packing factor, are required to completely submerge the sintered billet with fused glass during the subsequent hot-isostatic pressing cycle. Although this charge is only about 65% of the container void available after sintering, a certain amount of bead spillage is inevitable. Spillage is collected and recycled by a pump (vacuum type) and hopper system.

Equipment design specifications for the glass-bead charging station are summarized in Table G-11 (see Section G.9) and the station is shown in Figure G-8 (Section G.9). A special design is required to provide the remote features and reliability of operation in a radioactive environment. A spillage recycling system also needs to be developed and demonstrated. Finally, a reliable method of preventing the discharge of contamination when removing the alumina container cover must be developed.

G.6.10 CONTAINER LIFTING DEVICE WITH FILTER

A lifting device similar to that already described (Section G.6.7) is engaged with the alumina container when bead charging is complete and the articulated spout is removed. The lifting device facilitates transport of the container by overhead crane to the HIP and from the HIP to the overpack staging station. This lifting device also deposits a filter element in the container opening in the process of automatic engagement. The filter remains with the container during hot-isostatic pressing and traps residual, radioactive particulates during the de-airing and thermal cycles.

Lifting device design specifications are summarized in Table G-12 in Section G.9. Identification and experimental evaluation of an effective filter for containing radioactive species is an important addition to the developmental efforts described for the similar device in Section G.6.7.

G.6.11 HOT-ISOSTATIC PRESS

The principal functions of the HIP are to densify supercalcine to essentially 100% of theoretical, to obtain the desired supercalcine phase characteristics and to obtain a high degree of uniformity of these characteristics in a relatively large billet. These functions are expected to be accomplished by the isotropic application of high pressure to the sintered billet while at an elevated temperature to induce consolidation and molecular bonding.

In the envisioned process, pressure is applied by fused glass to the relatively porous billet. The glass composition is selected to provide viscosity and other characteristics at hot-isostatic pressing temperatures and pressure, such that the glass will not appreciably permeate the pores of the billet and will at the same time be essentially impermeable to the argon pressurization medium. As such, the fused glass transfers the isotropic gas pressure to the sintered billet (acting as a hydrostatic fluid), while maintaining hermetic encapsulation of the billet during its densification shrinkage. Thus, the initial steps in the hot-isostatic pressing process are to degas the sintered billet using vacuum and then to melt the glass beads under a partial vacuum to submerge and seal the billet in a relatively dense, fused-glass medium. The alumina container holds the fused glass and submerged billet.

When the glass is completely fused and the billet is thermally stabilized at the desired temperature, argon gas is pumped into the hot-isostatic pressing vessel and the vessel is pressurized. During a holding period at operating pressure and temperature, the sintered supercalcine billet consolidates to a density of about 3.9 kg/L (estimated theoretical) and undergoes molecular bonding. The sequence of pressure and temperature applications and the number and length of holding periods may be varied to obtain the desired crystalline phase characteristics and uniformity. As the submerged billet shrinks, the glass level drops in the alumina container to maintain pressure equilibrium on the billet. During hot-isostatic pressing, the alumina container does not undergo appreciable dimensional change because pressure on the container is equalized.

The HIP is then depressurized and cooled in a manner to preserve the integrity of the supercalcine billet and alumina container. When the HIP pressure is equalized to cell pressure, the vessel is opened to remove the alumina container. The container might need to be removed while the glass temperature is still near its softening point to avoid possible rupture of the container due to a thermal mismatch between the glass and container. (It may not be possible to develop a glass that meets this criterion as well as the more important hot-isostatic pressing criteria.)

Figure G-4 in Section G.9 shows an estimated pressure-temperature schedule for the selected design parameters. For supercalcine, a hot-isostatic pressing temperature requirement of about 1150°C is expected based upon limited supercalcine sintering experiments at PNL, whereas a pressure requirement of approximately 100 to 140 MPa (15,000 to 20,000 psi) is expected based upon industrial experience with hot-isostatically pressed ceramic powders.

The number of HIPs required is determined by dividing the hot-isostatic pressing cycle time by the time required to fill a sintering container at the design calcination rate. Thus,

for an estimated 10.6-h, hot-isostatic pressing cycle and a design fill time of 3.7 h, three HIPs are required. However, at the nominal filling rate of 5.6 h, two HIPs would suffice.

The hot-isostatic pressing equipment specifications for the HIP are summarized in Table G-13 (see Section G.9) and the equipment (including the HIP) is depicted in Figure G-9 (Section G.9). The equipment required for the SCHIP process consists of a pressure vessel, a vessel entry and closure system, an internal furnace, a vacuum system, a gas handling and pressurization system, a cooling system and a control system with instruments. These are covered in more detail below.

G.6.11.1 Pressure Vessel Entry and Closure System

A top-loading, yoke-type closure system with automatic cover handling was selected for use with an overhead crane for simplicity and reliability of container handling. (The 10.3:1 length-to-diameter ratio of the selected supercalcine container does not lend itself to bottom handling stability.) The yoke-type closure system should be easily automated and purportedly eliminates a stress discontinuity in the vessel wall typical of threaded closures. With this design, the potential for catastrophic vessel failure should be effectively minimized, if not eliminated. Seal failure is not considered to be catastrophic since the stored energy release is considerably throttled in the cover/vessel interfaces to prevent the formation of a shock-wave. The yoke-type design also lends itself to stainless steel cladding or enclosure for durability in the typical cell environment.

For the anticipated process requirement, a pressure vessel having a maximum working pressure of 170 MPa at 230°C (25,000 psi at 450°F) should be adequate. Vessels are typically designed to a 25% higher test pressure (213 MPa at 230°C) and are hydrostatically tested at an equivalent room-temperature pressure (247 MPa at 21°C). Rupture discs are normally provided to protect the integrity of the pressure vessel. For this design, a rupture setting of 196 MPa $\pm$ 5% would provide a 10% margin over maximum working pressure against premature rupture without exceeding actual test pressure.

G.6.11.2 Furnace

The furnace is located inside the pressure vessel and is sufficiently insulated to maintain a cold wall for mechanical strength considerations. The exterior of the vessel is jacketed and cooled with water for this purpose. Radiant heating elements are required to melt the glass in a partial vacuum. Molybdenum elements may be a viable choice since they are rated to 1700°C in a vacuum or an inert atmosphere. However, they require a special support design, limited heating and cooling rates and careful control of the element's environment since gas contamination (with O₂, N₂, H₂O, CO, etc.) embrittles molybdenum and causes thermal-cycle recrystallization above 1300°C. Primary furnace design considerations include:

- thermal durability of the heating element and support structure
- mechanical durability of the heating module during container exchange with an overhead crane
- remote replacement of the heating module and associated temperature sensors

G.6.11.3 Vacuum System

A vacuum system is required to degas the sintered billet and glass pack and to maintain a practical vacuum of about 13 Pa on the sintered billet and glass pack during glass fusion. An appropriately sized roughing pump is expected to develop adequate vacuum for this purpose. The exhaust gas from the pump is vented through the off-gas system to trap and recycle any residual, radioactive volatiles or airborne particulates.

G.6.11.4 Gas Pressurization System

An argon gas pressurization system with a 170-MPa (25,000-psi) capability is required to supply the maximum expected pressure for the selected supercalcine design parameters. A typical system having this capability consists of the following components:

- compressor package
- high-pressure, strain gauge transducers and signal conditions
- fail-safe, air-operated valving
- high-pressure gas recovery, compressor, and intercooler heat exchangers located in an isolated primary loop, and a secondary loop, heat-exchanger system
- flowmeter valving
- gas storage reservoir
- redundant pressure controllers
- cascaded and redundant, intermediate-pressure relief valves and high-pressure, rupture disc assemblies vented into the cell
- high-, intermediate- and low-pressure piping
- recovery system.

The compressor package consisting of four, single-stage compressors is arranged in two, two-stage sets. The triple diaphragm design of these compressors assures noncontaminating gas compression for furnace heating element longevity and allows automatic shutdown in the event of diaphragm seal failure.

The water-cooled, high-pressure, gas heat exchangers are included to protect the compressors while operating in series and to prevent the gas discharge components from overheating. All valving is electrically piloted and air-operated to enable remote operation with interlocked, sequential fail-safe controls. Cascading and redundant, pressure relief valves and rupture discs are located on each isolatable section of the gas pressurization systems. An argon gas storage reservoir having a capacity of 1500 m³ at 18 MPa and 21°C should be adequate, although reservoir capacity and pressure typically have a significant effect on performance of the pressurization system.

The three hot-isostatic pressing vessels required to meet a design throughput of 6.4 canisters per day might be arranged to utilize a single pressurization system. A single system

could incorporate back-up compressor capacity to enable routine maintenance or replacement concurrent with operation. Fail-safe, interlocking provisions would be included in the three-vessel configuration to prevent more than one vessel from being at pressure at a given time. This provision is a part of the overall safety hierarchy.

Gas recovery systems typically reclaim up to 90% of the argon required for a hot-isostatic pressing cycle. During depressurization, gas is cooled and automatically analyzed for contaminants (e.g., radioactive particulates, O_2 , CO , CO_2 , water vapor and hydrocarbons) using an in-line gas chromatograph to determine if the gas has to be exhausted to the off-gas system or purified by filtering and/or dialysis. Nonradioactive contaminant levels in the range of 1 to 50 ppm should permit furnace heating-element longevity. Recovery of gas below a vessel pressure of about 3-1/2 MPa (500 psig) is generally considered impractical. The balance of gas could be vented to the off-gas system.

G.6.11.5 Cooling System

The pressure vessel cylinder is jacketed and water-cooled to retain structural integrity during high-temperature pressurization and to reduce the hot-isostatic pressing cycle time. Integral cooling channels in the vessel closures provide similar functions plus maintain the high-pressure seals, furnace connections, thermocouple connections and reliable operating temperatures.

Cooling water is supplied from an isolated, closed-loop system consisting of circulating pumps, heat exchangers, an expansion tank and necessary valves, piping and control elements. Flow through the closed loop is monitored to ensure maintenance of minimum flow and to activate corrective procedures and emergency cooling, if required. Water flow in the open loop is controlled by an automatic thermostatic valve, and it is adequate to provide for permissible discharge temperatures.

G.6.11.6 Control and Instrument System

Pressure-temperature parameters in a hot-isostatic pressing system must be controlled accurately and reliably to safely obtain the desired results. Temperature control needs considerable attention to obtain the desired accuracy of heating rates, holding temperature uniformity and cooling rates.

An automated, totally integrated, computerized control system with instrument system redundancy may be required to obtain the desired level of operational safety due to the presence of radioactive material in a high-energy-level HIP. All routine operational functions including sequencing, timing, temperature-pressure control, interlocking, quality assurance record keeping, deviation control, system checks and others would be programmed and performed automatically. In addition to routine control, a basic function of the computer would be to monitor the hot-isostatic pressing system for abnormalities of operation, response, material stress levels, component temperatures, etc. and to provide diagnostic and trouble-shooting guidance to the operator to assure the proper maintenance and safe operating condition of the system. The computer would also monitor all significant parameters that affect the inherent

safety of the system, and would respond to situations requiring remedial action in a predetermined manner based upon a careful and thorough fault-free analysis of potential failure scenarios.

A panel with a symbolic system diagram, an instrument system readout of key operating parameters and light indicators (of the operating phase, normal operation, deviations outside set-point tolerances and fault alarms) would be provided for operator surveillance and verification of control response. Manual operation of the hot-isostatic pressing system for maintenance, repair, verification, etc. would be in a semi-automatic mode and would be limited by computer override to maintain safe operating conditions based on maximum limits or combinations thereof.

G.6.11.7 System Safety

Five levels of safety are designed into the envisioned hot-isostatic pressing system. The levels are prioritized in both the importance of a failure and the ability to correct or override that failure so that the process is affected only to the extent to maintain system integrity or to prevent potentially catastrophic conditions. The levels are as follows:

- Control Level. The control level is the normal operating mode for controlling sequence, time, temperature, vacuum and pressure in the system. At this level the HIP is operated within predetermined set-point tolerances. Pressure transducers, thermocouples, etc. provide feedback to control each support system; controller program and interlocks maintain the appropriate sequence. The basic quality of the total system design, appropriate maintenance and scheduled replacement of components is expected to minimize deviations from this level of control.
- Warning Level. An excursion outside set-point tolerances or other situations requiring remedial action would temporarily halt the processing cycle pending automatic diagnosis and correction or compensation for the fault. At this level, there is time for remedial action without adverse effect on the process or other more serious consequences. Visual and audible alarms would summon the operator for diagnosis verification. A prioritized list of potential faults might be generated by the computer depending on the complexity of the event. The sequence of remedial actions would be based on probability of occurrence and the effects monitored, diagnosed and recorded. Once the appropriate response was obtained, the process cycle would continue at the point of interruption. A maintenance and inspection check-list would be generated to prevent recurrence.
- Hazard Level. At the hazard level, a fault would result in the automatic and systematic abortion of a process run in such a manner as to maintain the integrity of the hot-isostatic pressing system and to preserve the alumina container if possible. The high-pressure valving, the cooling system reservoir and other critical subsystems would be designed and interlocked to revert to a fail-safe abortion mode in the event of loss of electrical power, compressed air, instruments or other support services. Control lights and a siren would be activated to warn the operator. In the event that the fault was

remedied during abortion, the sequence could be manually interrupted and the process run placed on hold for analysis of equipment and alumina container status and activation of a salvage process if possible.

- Danger Level. If through three levels of priority action a fault has not been corrected resulting in excessive pressure, then pressure-activated relief valving would bleed gas into the cells as required to protect the pressurization system and vessel. These valves would be backed up by a redundancy of rupture discs (previously described) designed to burst at a pressure below the actual vessel test pressure. Hot gas from the rupture disc would be expanded through a nozzle (to prevent formation of shock waves) and flow through the cell filtration system.

In a 2550-m³ hot-isostatic pressing cell, pressure would increase by about 0.11 kg/cm² (1.6 psi). The cell walls, ceiling and service feed-through ports would be designed to safely contain this pressure increase. An air circulation system, designed to change cell air 5 times/h, would require about 1 to 2 min to dissipate the resulting volume surge.

At the danger level, actions are not controllable, nor can they be overridden. Rapid depressurization could severely damage the vessel contents, but the primary intention is to ensure safety.

- Critical Level. The critical level represents the fifth and final level of the hot-isostatic pressing safety system. Based upon manufacturers' experience, with a safety-oriented hot-isostatic pressing system, the vessel is not expected to catastrophically rupture. Nevertheless, to protect operating personnel and to avoid the possible release of radioactive material to the atmosphere there must be a complete and thoroughly dependable answer to the question of what would happen should the pressure vessel rupture.

In consideration of this question, two scenarios are described to identify a possible range of energy levels involved in such an event. Should a crack occur (and escape detection) causing the vessel to suddenly rupture at maximum operating pressure (170 MPa), the energy release might be similar to an explosion of about 26 kg (57 lbs) of TNT (as typically equated by HIP manufacturers). Or, should all safety systems (previously described) fail to operate and a sound vessel be inadvertently pressurized beyond the capability of the pressurization system (by uncontrolled thermal expansion of the argon) to 2-1/2 times actual test pressure (530 MPa or 77,000 psi) before suddenly rupturing, the explosive results might approach that of about 80 kg (176 lb) of TNT.

The key to dealing with these energy levels in an enclosed cell may lie in the "suddenness" of pressure release. Sudden release of pressure, in itself, is not explosive, but more nearly an adiabatic expansion of gas from the volume of the vessel to that of the containment cell. At least one HIP manufacturer recognizes this in their vessel design and safety claims. All parts that withstand pressure are wound with a prestressed wire mantle. In vessel failure, the mantle purportedly contains fragments and sufficiently retards gas escape to prevent shock-wave formation. This approach should receive careful

consideration because aside from the explosion-like effects of a shock wave and fragment projectiles, the resulting increase in pressure in a 2550 m³ cell may reach 0.17 kg/cm² (2.5 psi). A cell design to safely contain this pressure increase should be reasonably possible considering the bulk of materials normally required for radiation shielding.

This approach might be further enhanced by locating the HIP below ground level and sufficiently oversizing the upper portion of the yoke to contain fragment projectiles. The doors to the adjacent isolated cell areas (used for HIP maintenance and repair, etc.) might also be constructed to blow out in the event that pressure in the primary containment cell exceeded design level. This would serve as a shock absorber and would further reduce the resulting pressure through expansion to a much larger volume.

G.6.11.8 Research and Development Required for Hot-Isostatic Pressing

Research and development of a process and equipment to hot-isostatically press supercalcine is expected to require considerable effort based upon the number and complexity of unknowns. Preliminary considerations include:

- An experimental hot-isostatic pressing process for sintered supercalcine needs to be developed. Samples need to be prepared to determine density, crystalline phase characteristics and other properties to evaluate the effectiveness of hot-isostatic pressed supercalcine in immobilizing high-level waste and to characterize process requirements. Simulated supercalcine materials should be evaluated in preliminary experimentation to utilize existing hot-isostatic pressing facilities. This work could be carried out using conventional "canning" techniques to encapsulate the billet.
- Based upon the above findings, a glass composition would need to be developed for the hermetic encapsulation and pressurization of the sintered supercalcine at the required process temperature and pressure. Simulated supercalcine materials should also be used in this phase to utilize existing hot-isostatic pressing facilities. This work would also further characterize the process requirements and would provide additional samples for evaluating high-level waste immobility and for characterizing the glass-supercalcine interface.

The findings from these preliminary experiments would form the basis for expanding and directing the scope of research and development as follows:

- evaluation of hot-isostatic pressing equipment to determine means for remote operation, maintenance, inspection, removal and installation
- analysis of safety concerns relative to the design and operation of a hot-isostatic pressing system and the containment cell to assess the probability of inadvertent loss of radioactive material to the atmosphere
- thermodynamic modeling and analysis of the hot-isostatic pressing process based on preliminary findings of material characterizations to identify a scaled-up process and sizing equipment. The number of HIPs required could be accurately estimated based upon this work. Decisions relative to equipment sizing for process research and development using

radioactive supercalcine could be analyzed for technical and economic feasibility. Due to the expected cost of full-scale, hot-isostatic pressing equipment and adequate cell containment, an intermediate-sized, experimental HIP coupled with carefully controlled scaling factors might be most cost effective.

- thorough and complete materials characterization within the range of expected process temperature
- design of the HIP container (alumina) including material selection and evaluation of its compatibility with processing conditions and materials. This work would encompass sintering requirements, handling considerations and storage concerns.
- design of an experimental facility and a remotely operable system for hot-isostatic pressing radioactive supercalcine, and the construction, procurement, fabrication, installation, and other functions in preparation for its operation
- experimental operation of the system to 1) develop a hot-isostatic pressing process for radioactive supercalcine utilizing a glass pressure/encapsulant medium in an open container, 2) prepare samples for evaluation and characterization, 3) demonstrate the remote aspects of hot-isostatic pressing, and 4) verify the thermodynamic model and scaling factors. Initially the system would be debugged and operated using simulated supercalcine to permit direct inspection and product evaluation for system and process verification.

The findings of this work would form the basis for identifying detailed process and equipment design criteria for a full-scale, remotely operable hot-isostatic pressing system and facility.

G.5.12 CONTAINER OVERPACK CANISTER

The function of the overpack canister is to provide radioactively clean containment to facilitate safe handling, intermediate storage and transport of the supercalcine billet to the final repository. The alumina container is placed in the overpack canister immediately after removal from the HIP. If this transfer is made while the container is relatively hot (to prevent thermal cracking), the lifting device is preheated to avoid excessive thermal stresses in the neck of the container. Once in the overpack canister, the integrity of the alumina container is no longer of importance since it is not a functional part of the immobilization of radioactive waste in hot-isostatically pressed supercalcine. Research and development considerations for this overpack canister are similar to those noted in the reference process and also include:

- canister design to accept the alumina container, facilitate closure welding and provide for subsequent handling
- thermal stress analysis of the alumina container during transport to the canister.

G.6.13 CANISTER PROCESSING

As covered in detail in the reference process, a helium source is inserted after the alumina container is placed in the canister (and crushed hulls added as an option). Then the closure lid is installed and seal-welded. The weld is then helium leak-checked to ensure its integrity. Canisters that fail to pass the leak test are rewelded and rechecked; the canisters may also be visually inspected at this time. Sealed canisters are then moved to a decontamination chamber where any loose contamination is removed. From here the canisters are placed in the in-cell storage racks or are transferred out of cell to an interim waste storage area as noted in the reference process.

The research and test efforts identified in the reference process for canister handling, closure, seal welding, inspection and decontamination are also required for canister processing in the SCHIP process. In addition, selected components and aspects of the SCHIP overpack canister processing system should be fabricated and tested to verify their capabilities and demonstrate their remote aspects.

G.7 RESEARCH AND DEVELOPMENT REQUIRED

Specific research and development requirements were identified by unit operation in Section G.6. Based upon the extent of work identified and the current state of product and process development, considerable effort will be required to research and develop the SCHIP process to the point where basic design criteria for a full-scale, remotely operable system can be identified. An encompassing program that integrates the sequence of product, process, and equipment research and development required is beyond the scope of this report, and perhaps academic at this point because of the number of fundamental unknowns. Nevertheless, a logical, prioritized research and development approach needs to be identified for the assessment of the potential technical and economic feasibility of the SCHIP process as described. Such an approach might embody the following primary objectives and priorities:

- develop a hot-isostatic pressing process for sintered, simulated supercalcine using existing hot-isostatic pressing facilities and conventional "canning" techniques
- using this process, prepare hot-isostatically pressed samples of simulated supercalcine and evaluate their potential for radioactive waste immobilization
- if promising, identify (or develop) a glass composition to hermetically encapsulate and pressurize the sintered supercalcine at the identified hot-isostatic pressing pressure and temperature and form into package
- identify a container material that 1) is thermally compatible with the identified glass composition and hot-isostatic pressing temperature, 2) facilitates melting the glass in a partial vacuum, and 3) shows promises of meeting projected in-process handling criteria; prepare experimental sample containers
- using existing hot-isostatic pressing facilities, if possible, develop a process for melting the glass-bead pack in a partial vacuum and for "open-can" hot-isostatic pressing of sintered, simulated supercalcine while submerged in fused glass
- concurrently, develop a process for sintering supercalcine using the experimental containers and prepare samples
- using all of the above processes, prepare hot-isostatically pressed samples of sintered, simulated supercalcine and evaluate 1) their potential for radioactive waste immobilization and 2) the glass-supercalcine interface for adverse effects
- based on the foregoing findings, re-evaluate the overall SCHIP product, process and equipment using 1) actual materials characterization data, 2) thermodynamic modeling of the sintering and hot-isostatic pressing process increments and of the final product in interim and repository storage, 3) computerized stress analysis of the container, and 4) the active involvement of hot-isostatic pressing equipment manufacturers. Expected results of this analysis would include relatively accurate estimates of the full-scale process, corresponding equipment requirements and criteria, final product characterization (including storage temperature profiles), required research and development, as well as an updated and greatly expanded version of this report.

- based on the above re-evaluation, assess the technical and economic feasibility of the SCHIP process
- if promising, research and develop equipment and process capabilities, demonstrating all remote aspects of the SCHIP process. An intermediate-scale version of the process would probably be developed because of the cost of hot-isostatic pressing equipment. Simulated supercalcine would be used to facilitate close monitoring, inspection and evaluation.
- concurrently, develop experimental capabilities to sinter and hot-isostatically press radioactive supercalcine to verify the simulated supercalcine process and prepare samples to evaluate radioactive waste immobilization characteristics of hot-isostatically pressed supercalcine
- based on these findings and demonstrations, identify the conceptual SCHIP product process and equipment including basic design criteria for a full-scale, remotely operable system.

G.8 ALTERNATIVE PROCESSES AND EQUIPMENT

A very limited time has been available to develop the preconceptual design bases for the SCHIP process. No doubt there are viable alternatives that have not been considered. Limited experimental knowledge of the process and the supercalcine material characterization in the intermediate process steps are handicaps. Some of the alternatives considered are noted in this section.

G.8.1 TRANSMITTAL OF HOT-ISOSTATIC PRESSING PRESSURE

Typically, powders to be consolidated are precompacted, degassed and sealed in a ductile container. During hot-isostatic pressing, the sealed container collapses or is otherwise deformed on the contained powder to transmit pressure to the powder without allowing the gaseous pressure medium to permeate the relatively porous material and thereby negate pressing. Several methods have been developed involving sheet-metal cans, vitreous containers, elastomeric bags, etc. as the sealing interface. Of the methods considered potentially feasible in a radioactive environment, all involve the task of completely filling the container and predensifying its contents. Accomplishing this task remotely while controlling excess powder and dust and using radioactive-resistant equipment is considered impractical. To avoid can failure from excessive deformation during hot-isostatic pressing, the contained material must be uniformly predensified to about 60% to 70% of theoretical density. Not only does this requirement complicate the filling task, it does not preclude occasional can failure. Under best conditions, can failure is expected to occur about 1% of the time based on typical industrial experience. This rate is considered intolerable for the SCHIP process, because can failure would probably grossly contaminate the inside of the HIP and the pressurization system, possibly damaging the furnace or causing the can to become wedged in the vessel (in the event of the rapid depressurization of a slow leaking container); this would require complicated cleanup, recovery and reprocessing procedures and further treatment of the breached can.

If technically feasible, the envisioned SCHIP process would eliminate many of the problems associated with conventional canning. The molten glass encapsulant tenaciously conforms to any contour of a sintered billet and accommodates nonuniformity of density within the billet. This process also facilitates in-process inspection of the size of the sintered billet and permits an excess glass charge to be made to assure complete submersion of the billet in the fused-glass sealing medium. Over- or undercharges of the sintering furnace should be easily accommodated. Furthermore, the open alumina container is not dimensionally affected during hot-isostatic pressing due to pressure equalization and is subjected to compressive pressures of one to two order of magnitudes less than typical alumina compressive strengths. However, should the container break during cooling, the tenacity of the glass adhering to the supercalcine billet would minimize contamination of the vessel interior and pressurization system. The recovery procedures may be complicated unless the HIP incorporates a removable shell to contain the molten glass. This possibility has not been fully explored.

G.8.2 PRE-HOT-ISOSTATIC PRESSING DENSIFICATION

A number of techniques have been developed and commercially applied to densify or consolidate powders, including vibration, tamping, cold-die pressing, cold-isostatic pressing, sintering, etc. The selection of a technique is generally based upon the powder's packing characteristics, which are a function of the particle shape and size distribution. Although the packing characteristics of supercalcine have not been fully explored, limited experiments indicate that the "as-calcined" material responds to sintering. This approach was selected because it lends itself to the "open-can" HIP utilizing glass as the sealing and pressure-transmitting medium. Sintering also facilitates relatively good control of powder and dust when compared to the other preconsolidation techniques. The mechanical complexity of sintering is expected to be comparatively simple, although this area has not been fully explored. Still another consideration in the selection of sintering is the expected variability, based on operating experience at PNL, of calcined material characteristics. Sintering precompaction should accommodate these variables, but it needs to be experimentally evaluated.

G.8.3 CONTAINER HANDLING

Sintering furnaces and hot-isostatic pressing equipment may be top- or bottom-loaded. The advantages of each approach need to be more fully explored before finalizing the container handling technique for the SCHIP process. Container handling with an overhead crane and top-loading was selected based on the relatively unstable container length-to-diameter ratio identified for the preconceptual product design. However, other considerations such as heat loss during hot transfers, potential damage to the container or furnace elements, cell layout, etc. may indicate the preferability of bottom loading.

G.8.4 HOT-TRANSFER TO HIP

An alternative to the described hot-isostatic pressing cycle is to vacuum-degas and melt the glass-bead pack to seal the sintered supercalcine billet prior to hot-transfer of the alumina container and contents to the HIP. This alternative might reduce the hot-isostatic pressing cycle by four hours (to 6.6 h) and reduce the number of HIPs required by one. Two or three vacuum furnaces and one or two additional transfers would be required for this alternative; however, their cost may be less than replacing a HIP. The scope of this report excludes cost analysis of this detail. An important advantage of this alternative is that it facilitates in-process inspection and measurement of the fused glass level to assure complete submersion of the sintered billet and provides a means of routine determination of compaction.

It might be possible to integrate the sintering, vacuum-degassing, glass-charging and billet-sealing steps to eliminate the cool and reheat thermal cycles incorporated to prevent volatilization of radioactive wastes during transfer and glass-bead charging. This alternative has not been fully explored due to time constraints. However, it would result in only one hot-transfer of the alumina container, thus conserving energy requirements to reduce overall process time by about six hours. It would also trim the equipment requirements to about four sintering/vacuum furnaces and two HIPs and would provide a sealed supercalcine billet during transfer, thereby improving the control of possible cell contamination.

G.9 DRAWINGS AND DATA SHEETS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

TABLE G-4. Equipment Data Sheet for the Supercalcine HLLW Feed System

Function

Prepare supercalcine solution and deliver to spray calciner.

Process Interfaces

HLLW supply; spray calciner.

Design Parameters

Same as reference process except has a 25% greater volume; 30 to 305 L/h feed rate; capable of handling soda-rich solutions.

Operating Conditions

Same as reference process except has a 25% greater volume; 203 L/h feed rate.

Features

Same as reference process except accurate HLLW sampling and analytical system, supercalcine chemical addition system and thorough mixing system.

Services or Auxiliaries

Same as reference process.

Space Requirements

Maintenance Methods

Remote

Remarks

TABLE G-5. Equipment Data Sheet for the Supercalcine Spray Calciner

Function

Calcine supercalcine solution into dry powder and an off gas.

Process Interfaces

Calciner feed system; 4-way diverter to sintering furnaces; off-gas system.

Design Parameters

Same as reference process except 28% greater capacity required (305 L/h supercalcine solution input, 31.3 kg/h dry particle output).

Operating Conditions

Same as reference process except 203 L/h supercalcine solution input and 20.9 kg/h dry particle output.

Features

Same as reference process except frit-feeding system not required.

Services or Auxiliaries

Same as reference process.

Space Requirements

Slightly larger than reference process.

Maintenance Methods

Same as reference process.

Remarks

Similar to reference design.

FIGURE G-3. Supercalcine Delivery System

TABLE G-6. Equipment Data Sheet for the Supercalcine Delivery System

Function

Directs continuous supercalcine output from spray calciner to one of four batch sintering furnaces and concurrently allows off gas to flow from the other three furnaces.

Process Interfaces

Spray calciner; 4 sintering furnaces.

Design Parameters

Capacity = 32 kg/h; automatic and remote operation; enclosed and ventilated operation; remote replacement and removal; stainless steel construction; temperature = 300° to 1250°C.

Operating Conditions

Temperature = 300° to 1150°C; capacity = 21 kg/h.

Supercalcine properties: shape--agglomerated, semi-spherical particles; particle density = 2.8 to 3.8 kg/L; bulk density = 0.8 kg/L; particle hardness = 5 to 7 mole scale; moisture/volatiles content = 10 wt%; particle size--90% are 1 to 10 μ .

Features

Independent weighted seals to furnace and to alumina container; closure furnace cover with an insulated plug during sintering; cascading uncoupling to facilitate furnace translation; sealed enclosure; 4-way indexing rotary diverter; off-gas vent to calciner; spray-flush cleaning system.

Services or Auxiliaries

Compressed air (pneumatic actuators); electrical (temperature sensors); nitric acid (cleaning); water (cooling); drain to sintering furnace enclosure.

Space Requirements

3.0 m high by 3.0 m in dia

Maintenance Methods

Remote; periodic spray-flush cleaning cycle.

Remarks

FIGURE G-4. Sintering Thermal Cycle

TABLE G-7. Equipment Data Sheet for the Sintering Furnace

Function

Densify supercalcine into a cylindrical billet having a volume approximately 19% of bulk supercalcine powder.

Process Interfaces

Supercalcine delivery system; overhead crane transport to decontamination station.

Design Parameters

Automatic and remote operation; remote replacement and installation; 32 kg/h design fill rate; 154 L design capacity; inside dimensions are 0.32 m in dia by 3 m in height; radiation and nitric acid resistant; temperature range is to 1250°C; cooling capability; enclosure with off-gas ventilation; cleanout capability.

Operating Conditions

21 kg/h fill rate; 146 L (117 kg) bulk supercalcine accumulation (properties same as specified for the supercalcine delivery system in Table G-6; thermal cycle (see Figure G-4) is normally 1150°C for 16.6 h; including 5.6 h fill time; heating and cooling rates are 300°C/h.

Sintered billet properties: shape is right circular cylinder; density = 3.8 kg/L; weight = 105 kg; volume = 28 L.

Features

Closed-loop cooling; closure of inlet with insulated plug during thermal cycle; radial translation of furnace to remove alumina container; off gas through delivery system; means of sealing furnace and container to delivery system; enclosed system during supercalcine accumulation and thermal cycle; spray-flush cleaning system.

Services or Auxiliaries

Electrical (furnace, temperature sensors); compressed air (pneumatic actuators); process water (cooling); drain to furnace enclosure; instrument air (pressure sensor); nitric acid (cleaning).

Space Requirements

4.6 m in dia by 3.4 m high for cluster of 4 furnaces and translation mechanism (integral with furnace enclosure).

Maintenance Methods

Remote

Remarks

FIGURE G-5. Sintering Furnace

TABLE G-8. Equipment Data Sheet for the Furnace Enclosure

Function

Limit spread of process dust; provide means to periodically clean delivery system and furnaces.

Process Interfaces

Encloses cluster of 4 sintering furnaces; interfaces supercalcine delivery system and overhead transfer.

Design Parameters

Remote operation, installation and removal; remote access to enclosed furnaces and mechanism for maintenance, installation and removal; dust tight; stainless steel construction.

Operating Conditions

Ambient temperatures = 25^o to 350^oC; air pressure is negative with respect to cell and positive with respect to furnace when connected to calciner.

Features

Dust tight access hatch; air curtain during installation and removal of sintering containers; spray-flush cleaning system; remote service connections to furnaces and enclosed mechanisms.

Services or Auxiliaries

Compressed air (port actuators); instrument air (pressure sensors); nitric acid (cleaning); pumping system to dispose of spent cleaning solution.

Space Requirements

5 m in dia by 4.5 m high

Maintenance Methods

Remote

Remarks

FIGURE G-6. Furnace Enclosure

TABLE G-9. Equipment Data Sheet for the Container Lifting Device with Cover

Function

Apply temporary cover to alumina container and automatically engage (disengage) container for transport by means of overhead crane.

Process Interfaces

Overhead crane; alumina container.

Design Parameters

Lifting capacity = 2 MT; stainless steel construction; remote operation; radiation resistant; temperature = 25^o to 45^oC; maximum operating rate = 6.5 cycles/d.

Operating Conditions

Temperature = 25^o to 450^oC; nominal operating rate = 4.3 cycles/d; superheated steam is 100^o to 350^oC.

Features

Automatic engagement and disengagement with alumina container; closes and seals opening of alumina container when engaged; remote coupling to crane.

Services or Auxiliaries

Compressed air (pneumatic actuators); overhead crane (lifting).

Space Requirements

30 cm in dia by 1.0 m in height

Maintenance Methods

Remote

Remarks

TABLE G-10. Equipment Data Sheet for the Alumina Container Decontamination System

Function

Limit spread of process dust by cleaning alumina vessel with steam spray.

Process Interfaces

Sintering furnace.

Design Parameters

Capacity = 6.5 containers/d; remote operation, installation and removal; stainless steel construction; temperature = 100° to 45°C; pressure is negative with respect to cell.

Operating Conditions

Nominal steam temperature = 350°C; capacity = 4.3 containers/d; off-gas condensation and collection; condensate recycled to feed make-up tank.

Features

Temperature and pressure sensors; steam condensation/recycling system; electrical (temperature sensors); instrument air (pressure sensors).

Services or Auxiliaries

Superheated steam (cleaning); lifting device with closure.

Space Requirements

Decontamination vessel: 1.0 m in dia by 4.0 m in height.

Maintenance Methods

Remote

Remarks

FIGURE G-7. Load Removal and Decontamination System

TABLE G-11. Equipment Data Sheet for the Glass-Bead Charging Station

Function

Charge alumina container with glass beads.

Process Interfaces

Alumina container.

Design Parameters

Design capacity = 6.5 charges/d at 77 L/charge; remote operation, installation and removal of all components; radiation resistant; bead heating capability; alumina container heating capability.

Operating Conditions

Nominal capacity = 4.3 charges/d at 77 L/charge; container holding furnace temperature = 350°C; glass bead preheat temperature = 350°C; charge rate = 15 L/min;
Glass beads: dense-packing particle size distribution; free flow bulk density = 1.7 kg/L; bead density = 2.3 kg/L.

Features

Glass bead preheat hopper; rotary air-lock metering valve; holding furnace for alumina container; overflow recycling system; articulated delivery tube; temperature and pressure sensors; glass bead fill level sensor.

Services or Auxiliaries

Electrical (furnace, temperature sensors, fill level sensor); compressed air (pneumatic valve actuators); instrument air (pressure sensor).

Space Requirements

2 m in dia by 6 m in height

Maintenance Methods

Remote; contact on bead preheat hopper.

Remarks

Special design required.

FIGURE G-8. Glass-Bead Charging Station

TABLE G-12. Equipment Data Sheet for the Container Lifting Device with Filter

Function

Apply filter to alumina container and automatically engage (and disengage) container for transport by means of an overhead crane.

Process Interfaces

Overhead crane to alumina container for transport from glass-bead charging to HIP to over-pack staging.

Design Parameters

Design rate = 6.5 cycles/d; lifting capacity = 2 MT; stainless steel construction; remote operation; radiation resistant; filter temperature = 25^o to 1250°C; lifting device temperature = 25^o to 700°C; filter pressure = 13 Pa to 120 MPa.

Operating Conditions

Nominal rate = 4.3 cycles/d; lifting device temperature = 25^o to 650°C ambient; filter temperature = 25^o to 1150°C; filter pressure = 13 Pa to 103 MPa.

Features

Automatic engagement and disengagement with alumina container; deposits filter in container opening; remote coupling to crane.

Services or Auxiliaries

Compressed air (pneumatic actuators); overhead crane.

Space Requirements

30 m in dia by 1.0 m in height

Maintenance Methods

Remote

Remarks

TABLE G-13. Equipment Data Sheet for the HIP

Function

Vacuum-degas sintered supercalcine billet and glass bead pack; fuse glass to hermetically encapsulate billet; densify supercalcine to near 100% of theoretical.

Process Interfaces

Glass-bead charging station; overpack station.

Design Parameters

Automatic operation; remote loading and unloading; remote installation and removal; stainless steel construction on exposed surfaces; minimum fatigue life = 26,000 cycles (650 cycles/yr, 40-yr life); maximum load = 0.7 MT; electrical resistance furnace capability = 230°C; maximum working pressure = 170 MPa; degassing vacuum = 10 Pa; work zone dimensions are 30 cm in dia by 3.0 m in height; pressurizing gas is argon; maximum heating and cooling rates = 500°C/h; ASME Boiler and Pressure Vessel Code Section VIII, Division 2 applies; argon gas and cooling water isolation to cell.

Operating Conditions

Thermal pressure cycle (see Figure G-4): degassing vacuum = 13 Pa; maximum temperature = 1150°C; maximum pressure = 103 MPa; turnaround cycle = 11.0 h.

Features

Top loading HIP consists of the following major subsystem features: stainless steel clad pressure vessel; rugged, easily repaired furnace and heat shield module; argon pressurization system, including a reliable, efficient, contamination-free, and isolated gas recovery and storage system; automatic, programmed fail-safe control system including an instrument system (pressure, temperature, leak detection, gas analysis etc.); yoke-type closure system including automatic cover handling; closed-loop, recirculating cooling system; vacuum system including exhaust gas isolation.

Services or Auxiliaries

Argon gas (pressurizing); electrical (furnace, pumps, temperature, strain gages, pressure sensors); water (cooling); compressed air (pneumatic actuators).

Space Requirements

Maintenance Methods

Furnace module, pressure vessel and closure system are remote; cooling system, vacuum system and pressurization system are semiremote.

Remarks

Hot-isostatic pressing systems of this size and configuration available from Autoclave Engineers, Inc. and ASEA, Inc; development effort required to obtain remote operation, maintenance, installation and removal, and radiation resistant and isolated pressurization vacuum and cooling systems.

FIGURE G-9. Hot-Isostatic Pressing Vessel

G.9 REFERENCES

McCarthy, G. D. 1977. "High-Level Waste Ceramics: Materials Consideration, Process Simulation and Product Characteristics." Nuclear Technology 32:92-105.

APPENDIX H

SYNROC HOT-ISOSTATIC PRESSING PROCESS

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APPENDIX H

SYNROC HOT-ISOSTATIC PRESSING PROCESS

H.1 INTRODUCTION

SYNROC hot-isostatic pressing is an alternative high-level radioactive waste immobilization process utilizing SYNROC (Ringwood 1978) as the host medium. SYNROC (synthetic rock) is an acronym for a series of synthetic, igneous rock systems consisting of a combination of thermodynamically compatible minerals. The selected minerals are purportedly known to have the capacity to accept and retain radioactive waste elements in their crystal lattices and to be stable in a wide range of geological and geochemical environments for periods ranging from 20 to 2000 million yr. In addition, the fundamental reasons underlying this capacity to immobilize radioactive elements are supposedly well understood in terms of the basic principles of crystal chemistry, mineralogy and geochemistry.

The basic philosophy of the SYNROC approach to radioactive waste immobilization is to retain the advantages of the supercalcine concept (Appendix G) and to minimize its disadvantages. Ringwood (Ringwood, Kessen, Ware, Hibberson and Major 1979) identifies the following disadvantages of supercalcine:

- The crystalline phase assemblage of supercalcine, having a 78 wt% oxide waste-loading, is dictated to a large degree by the composition of the waste; therefore, it is not necessarily ideal for long-term predictability of containment of radioactive waste components.
- Some radioactive species (for example ^{137}Cs , which ultimately transmutes to barium and ^{90}Sr , which in turn ultimately transmutes to zirconium) would not be stable at high concentrations in the supercalcine crystalline host phases due to the density changes that accompany radioactive transmutation. Thus, after a substantial portion of the species had decayed, the crystal lattice would probably be severely damaged, permitting ready leaching of the remaining species.
- Because of high loadings of radioactive waste in supercalcine, excessive radiation damage to some crystalline phases will occur. This may enhance solubility in some phases and cause pronounced volume expansion leading to cracking and increased permeability to leaching solutions.

Radioactive waste concentrations in the SYNROC systems do not exceed 10 wt%. One would naturally expect this to increase the relative cost of processing, transporting and disposal compared to the reference process and other alternatives having higher radioactive waste concentrations. However, major technical advantages are claimed by Ringwood and his colleagues including:

- At lower concentrations, the radioactive species are simply substituted in the crystal lattices determined by the SYNROC components. This provides unique flexibility in optimizing a crystalline phase assemblage with the most desirable and predictable immobilization characteristics.
- Because the radioactive waste atoms are not major components, the problems associated with transmutation and radiation damage are greatly reduced. Thus, mineral phases can be produced that are more likely to possess the ability to retain products of transmutation and radioactive species.
- Compositional flexibility makes it possible to utilize SYNROC systems consisting of host crystals of types that are known to be extremely resistant to leaching in appropriate geological environments.

Several different SYNROC compositions have been studied by Ringwood and his colleagues. One composition (SYNROC B) that has been studied in detail is composed of the minerals perovskite (CaTiO_3), hollandite ($\text{BaAl}_2\text{Ti}_6\text{O}_{16}$) and zirconolite ($\text{CaZrTi}_2\text{O}_7$). These minerals are intimately mixed with radioactive waste (10 wt%) and are synthesized by crystallization in the solid state at elevated temperatures and pressures. This synthesis is analogous to the formation of most natural metamorphic rocks found in the earth's crust.

In the envisioned SYNROC HIP process, the proper proportion of calcined HLLW is measured and intimately blended with SYNROC minerals. The minerals have a closely controlled particle size distribution to obtain the desired blend and dense-packing characteristics. The blended calcine and SYNROC minerals are heat-treated at about 1325°C to form large, sintered billets having substantial strength but appreciable porosity (about 23%). The billets are then vacuum-degassed, sealed, and isostatically pressed at about 1325°C and 100 MPa (15,000 psi) to complete recrystallization and to form the desired grain size, uniformity and dense phase assemblage approaching 100% of that theoretically possible. During solid-state recrystallization, the intimately dispersed, radioactive waste elements are accepted into the crystal lattices of the SYNROC minerals. The resultant synthetic, metamorphic rock assemblage should also have high strength and leach resistance based on the relative enhancement of these characteristics in other hot-isostatically pressed materials, including ceramics.

The SYNROC HIP process is expected to have a significantly lower risk of product abnormalities than either the reference or the SCHIP processes because of the lower concentration of waste. More safeguards and more equipment and cell design considerations will be required than in the reference process, and these considerations will no doubt lead to a more expensive process.

The SYNROC HIP process facility will have the capability to 1) receive and process HLLW, 2) calcine HLLW, 3) measure and thoroughly blend a controlled particle size distribution of SYNROC formers with the HLLW calcine, 4) collect and heat-treat the SYNROC blend into sintered billets, 5) vacuum-degas, seal and hot-isostatically press the sintered billet to a desired density, crystalline assemblage, and uniformity, 6) load the pressed billet into a canister, 7) close and seal-weld the canister, 8) check the canister seal, and 9) decontaminate the canister and transport to an interim storage area prior to final storage.

H.2 PROCESS DESCRIPTION

The SYNROC HIP process is depicted in the flow diagram shown in Figure H-1. As covered in the reference process (Appendix A), HLLW (the basic composition is given in the Overall Process Bases Section) is received in the waste facility and stored in the HLLW feed system. The HLLW is continuously fed through a pneumatic atomizing nozzle into a spray calciner at a nominal operating rate of 158 L/h. Calcine and an off gas are produced in the heated calciner chamber. The calcine falls into a continuous zig-zag (twin-shell type) blender at a nominal operating rate of 17.2 kg/h.

SYNROC-former minerals that have been manufactured to the desired composition and particle size distribution are preheated and continuously metered into the blender at the rate of 155 kg/h to form a 9:1 ratio with the waste calcine (10% oxide waste-loading). The solids are uniformly and intimately blended and gravity-fed through a delivery system into a sintering furnace at a nominal rate of 172 kg/h.

In the sintering furnace, the SYNROC blend is accumulated in an alumina container that is sealed to the delivery system. When the container is filled with the desired quantity, the semicontinuous flow from the blender is diverted to a second, adjacent sintering furnace. The first furnace is then closed off from the delivery system with an insulation plug in the cover, and the SYNROC oxides are heated to their sintering temperature.

During sintering the SYNROC oxides begin to recrystallize and uniformly shrink to approximately 15% of their bulk volume. Volatilization accounts for about 15 wt% loss. The resulting sintered billet is cylindrical and roughly proportional to the container dimensions. A sacrificial coating on the inside of the container is burned off to expel any radioactive waste adhering to the surface and to act as a parting agent.

The delivery system is then decoupled, and the furnace, container and hot, sintered billet are translated to a vacuum-degassing and glass-charging system. The outer shell of the furnace engages and seals to this system to form an air-tight vessel. The vessel is evacuated to about 13 Pa, which causes the molten glass level to rise about 4.5 m in a siphoning tube that connects the vessel with a glass remelt tank. The tube is preheated to accommodate free glass flow. When the sintered billet is completely degassed, pressure in the glass tank is increased slightly to force the fused glass through the siphon into the alumina container. The molten glass charge is controlled by the original level of glass in the remelt tank (relative to the siphon tube inlet) and is sufficient to completely submerge the billet. The pressure in the vessel remains at about 13 Pa during glass charging.

After glass charging, the vessel is pressurized to atmospheric pressure and is decoupled. The furnace, container and submerged sintered billet are then translated to another position. A special lifting device, appropriately preheated, engages the alumina container through a decontamination chamber. As the container is lifted, superheated steam is used to vigorously scrub the exterior of the container to remove any radioactive contamination. The spent steam is exhausted through a scrubber and filter system. Condensed steam is recycled to the spray calciner.

FIGURE H-1. SYNROC HIP Flow Diagram

The decontaminated container then passes through a cell lock to the hot-isostatic pressing cell and is placed on the bottom cover of the hot-isostatic pressing vessel. The preheated bottom cover is then transferred into position and raised to bottom-load the container into the hot-isostatic pressing vessel. The temperature in the vessel is maintained at an intermediate level during loading and unloading by natural convection. An argon purge is also maintained in the vessel by virtue of the lighter density of argon at these temperatures relative to the cell atmosphere. A yoke is then positioned to seal the vessel.

The pressure in the hot-isostatic pressing vessel is initially reduced to remove residual air and contaminated argon. Concurrently, the temperature of the contents are increased and stabilized at 1325°C. Argon gas is then pumped into the vessel and the vessel is pressurized to about 100 MPa (15,000 psi). Under high pressure and temperature, the fused glass acts as an impermeable, hydrostatic fluid to densify and recrystallize the submerged SYNROC billet.

The HIP is then depressurized and cooled somewhat for unloading the container and its pressed billet. The relatively hot container is transferred to an overpack canister with an overhead crane. At this time crushed hulls may be added to the void in the alumina container above the glass level.

A helium source is then added to the canister and its closure installed and seal-welded. After the weld is leak-tested, the canister is inspected, decontaminated and transported to temporary storage to cool.

H.3 BASES AND ASSUMPTIONS

Preconceptually designing a production process for SYNROC HIP requires a number of assumptions in terms of the process bases, canister, effluents, waste, scrap, utilities and materials. These are generally identified in the Overall Process Bases Section. Assumptions specific to the SYNROC HIP process are identified in the following subsections.

H.3.1 PROCESS BASES

As indicated previously, the process flow diagram for the SYNROC HIP process is shown in Figure H-1. The diagram is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. Design capacities are 1.5 times the nominal operating values. Other pertinent data on the HLLW is presented in the Overall Process Bases Section and in Appendix A. Additional processing data is listed below:

- HLLW solution feed to spray calciner
 - design rate = 237 L/h
 - operating rate = 158 L/h
 - heat loading = 3.46 W/L
 - oxides content = 92.6 g/L
- Calciner input to blender
 - volatile residue = 15 wt%
 - operating rate = 17.2 kg/h
 - design rate = 25.8 kg/h
 - bulk density = 0.6 kg/L
 - heat loading = 31.8 W/kg
- SYNROC input to blender
 - volatile residue = 15 wt%
 - design rate = 232 kg/h
 - operating rate = 155 kg/h
 - bulk density = 1.9 kg/L
 - ratio to HLLW calcine = 9:1 (by weight)
- SYNROC/calcine blend
 - volatile residue = 15 wt%
 - design rate = 258 kg/h
 - operating rate = 172 kg/h
 - bulk density = 1.8 kg/L
 - heat loading = 3.2 W/kg
- Sintered billet
 - bulk charge (with volatiles) = 962 kg (534 L)
 - heat loading = 3060 W

charge time--
 operating = 5.6 h
 design = 3.7 h
sintered density = 3.4 kg/L
sintered weight and volume (without volatiles) = 818 kg (241 L)
glass-sealing medium--
 density = 2.3 kg/L
 fused charge = 453 kg (197 L)
cycle time (accumulate, sinter, degas, seal, and decontaminate) = 14.8 h

- Hot-isostatic pressing
 cycle time (hot transfer in, warm transfer out) = 6.7 h
 hot-isostatically pressed SYNROC--
 density = 4.4 kg/L
 billet weight and volume = 818 kg and 186 L
- Overpack canister
 operating rate = 1288/yr (4.3/d)
 design rate = 1930/yr (6.4/d)
 heat loading = 3060 W

H.3.2 CANISTER

General requirements for waste canisters are given in the Overall Process Bases Section and are discussed in more detail in Appendix A. Specifics for the SYNROC HIP process canisters are noted here. The canister configuration shown in Figure H-2 represents the final product of the SYNROC HIP process. The canister consists of a sealed, 304L stainless steel outer shell with appropriate lifting means and an open alumina container that holds the SYNROC HIP billet completely submerged in glass. As with the SCHIP process, the alumina container and glass are for processing purposes only and are not considered part of the waste immobilization system because of their potential for thermal cracking during storage.

For SYNROC having a 10 wt% waste loading, the size of the canister is limited by the specified 60-cm-dia and 3-m-height maximums (Overall Process Bases Section). Within these limits an ID of 51.2 cm was selected for the alumina container, with a charge volume of 583 L. However, the 3060 W/canister heat load limitation requires the charge to be about 534 L. Thus, there is a 9% fill allowance to accommodate variations in accumulated bulk density, among other process variables. Based on these selections and on the previously identified process bases, the resulting SYNROC HIP billet dimensions are about 36 cm in dia by 1.8 m in height. Approximately 197 L of fused glass are required to submerge the billet during hot-isostatic pressing.

If desired, the void in the alumina container above the glass level may be filled with approximately 415 kg of crushed fuel rod hulls prior to closing the canister. This represents 100% of the hulls from which the contained radioactive waste was extracted.

WEIGHT	SYNROC	818 KG
	HULLS	415 KG
	GLASS	453 KG
	ALUMINA CONTAINER	432 KG
	304L CANISTER	613 KG
		<hr/> 2731 KG

FIGURE H-2. Canister

The total weight of the canister, including optional hulls, is estimated at 2731 kg. The contained SYNROC represents about 30% of this weight. An approximate breakdown of component weight is given in Figure H-2. About 1.8 MT of total canister weight will be required per MTHM processed.

Based upon estimated thermodynamic properties of the SYNROC billet and preliminary calculations, the steady-state centerline temperature of the SYNROC billet will be roughly 570°C if the canister is exposed to 50°C air storage. The corresponding maximum glass temperature is estimated at 430°C, which is well below its anticipated annealing point. The possible thermal expansion mismatch between the billet, glass and alumina container might crack the alumina container and glass. When the canister wall is maintained at 100°C, the billet centerline

temperature and glass temperature are estimated at 500°C and 350°C, respectively. Glass and alumina cracking is more likely at these temperatures unless a thermal expansion match can be practically attained.

H.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

For this study it is assumed that the effluents from the SYNROC HIP process are similar in content and order of magnitude in volume to those from the reference process. Except for the additional equipment for this expanded and more complex process that eventually will have to be discarded, it is assumed that the wastes from this process will be of the same order of magnitude in volume and contents as the reference process. Systems and provisions are assumed to be included in the fuel reprocessing plant to treat, reduce in volume and package both liquid and solid wastes. All solidified process scrap that may contain radioactive waste will be recycled and incorporated into the SYNROC HIP process.

H.3.4 UTILITIES AND MATERIALS

Estimates of the utility services and essential materials required for the SYNROC HIP process are given in Tables H-1 and H-2, respectively. These order-of-magnitude estimates are based on the requirements for the reference process with appropriate adjustments for differing process steps and anticipated space and volume changes.

As with the reference process, only the utilities used in the SYNROC HIP process and provided from the fuel reprocessing plant are shown; any emergency utilities required, such as electricity and cooling water, are supplied from the fuel-reprocessing-plant emergency services. Maintenance and replacement materials are also not shown. Only chemicals used for periodic facility decontamination and cleanup are included.

TABLE H-1. Estimate of Utilities Required for SYNROC HIP Process

<u>Utility</u>	<u>Required Capacity</u>	<u>Average Use Rate</u>	<u>Disposition</u>
Electricity	4000 kW	2600 kW	--
Raw water	2500 L/min	1100 L/min	Recycle or environment
Process water	800 L/min	60 L/min	HLLW
Sanitary water	40 L/min	20 L/min	Sanitary or chemical sewer-tile field
Air	2000 scmm	1000 scmm	As appropriate
Steam	32 kg/min	7 kg/min	As appropriate

TABLE H-2. Estimate of Materials Needed for SYNROC HIP Process^(a)

<u>Material</u>	<u>Amount Needed Per Unit Time</u>
Alumina containers	4.3/d or 1,288/yr
Canisters	4.3/d or 1,288/yr
Glass frit	1950 kg/d or 585 MT/yr
SYNROC formers	3,720 kg/d or 1,116 MT/yr
Caustic (19M)	8,000 L/yr
Nitric acid (12.2M)	12,000 L/yr
Helium sources	4.3/d
Argon	64 scm/d or 19,200 scm/yr
Detergent	1000 kg/yr
Ammonia	120,000 kg/yr

(a) Operating rate; excludes maintenance and apparel.

H.4 FACILITY AND EQUIPMENT REQUIREMENTS

The SYNROC HIP process is contained in a reinforced concrete structure similar to that shown in Figure D-3 in Appendix D. However, the number of cells, their size and general arrangement will differ from that shown in Figure D-3 and will be determined after more data is established on the process and equipment requirements.

The overall requirements for the waste facility and its support groups and services are covered in the Overall Process Bases Section. The requirements for remote operation and remote maintenance of all in-cell equipment are the major concern when establishing the bases for a process installation. The major pieces or equipment systems and their approximate space requirements are given in Table H-3.

TABLE H-3. Process Cell Equipment Requirements and Dimensions

<u>Equipment</u>	<u>Number Required</u>	<u>Dimensions (Width x Length x Height), m</u>
HLLW batch tank	2	2.5 x 2.5 x 4
HLLW feed tank	1	1.5 x 1.5 x 3
Spray calciner	1	3.5 x 4 x 6
Blender	1	3 x 5 x 3
Delivery system	1	4.5 x 4.5 x 5
Sintering furnace	4	See furnace enclosure
Glass-charging station	4	See furnace enclosure
Decontamination station	2	2 x 2 x 2
Furnace enclosure	2	5 x 5 x 4.5
Hot-isostatic press	2	5 x 10 x 10
Canister loading station	1	3 x 3 x 6
Lid-closure station	1	2 x 3 x 6
Leak-detection station	1	2 x 3 x 6
Decontamination station	1	3 x 4 x 7
Canister rack	1	3 x 3 x 6
Storage transfer	1	2 x 3 x 5

H.5 STATE OF PROCESS DEVELOPMENT

Although a moderate level of research has been conducted on simulated SYNROC by Ringwood (1978) and colleagues and at PNL, to date only small quantities of this material have been hot-isostatically pressed (and not in a glass pressure medium). Major problems have been precompaction of "as-calcined" SYNROC and metal can failure during hot-isostatic pressing. However, the fact that existing hot-isostatic pressing technology produces dense ceramic materials of increased strength and of desirable crystalline grain size and uniformity indicates the possible advantages of applying this process to the production of a SYNROC waste form.

Very small samples (5 cm^3) of simulated SYNROC B (calcined) have been successfully sintered in an alumina receptacle at PNL. However, material characterizations of sintered SYNROC have not been completed. The state of development of the hot-isostatic pressing process and equipment and of spray calcination and accessory systems in relation to the SYNROC HIP process is the same as that identified for the SCHIP process (Appendix G).

H.6 UNIT OPERATIONS

The overall SYNROC HIP process consists of a number of unit operations. The function and operation of each, as they differ from the reference process are described in greater detail in this section. The schematic drawings and equipment data sheets summarizing the design requirements can be found at the back of this appendix in Section H.9. Required research and development work specific to the SYNROC process is also identified.

H.6.1 HLLW FEED SYSTEM

The HLLW or calciner feed system for the SYNROC HIP process is basically the same as for the reference process. The function and requirements for the basic HLLW feed system are described in detail in Appendix A. This system has undergone considerable development and evaluation, and its operating parameters and design requirements have been well established. Additional research and development requirements are also identified in Appendix A.

H.6.2 SPRAY CALCINER

The function, design and operation of the spray calciner for the SYNROC HIP process is basically the same as for the reference process except that a system is required for feeding SYNROC formers along with the calcine output into the blender described in Section H.6.5. The SYNROC-formers feed system replaces the glass frit feed system used in the reference process. The research and development efforts required for the spray calciner include those noted for the spray calciner of the reference process.

H.6.3 OFF-GAS SYSTEM

The function and operation of, and the research and development requirements of the SYNROC HIP process off-gas system are the same as those specified for the reference process. However, the required capacity is about 10 times greater than that of the reference process. For operating efficiency, two parallel systems may also be required.

H.6.4 SYNROC-FORMERS FEED SYSTEM

SYNROC formers are manufactured to a desired particle size distribution and density. The prepared formers are preheated to about 700°C in a continuous rotary kiln before being fed into the cell for blending with the calcined waste. Because these operations are performed outside the cell, common manufacturing and processing techniques may be used. A variety of possibilities exist. Process and equipment research and development are required to apply existing technology to meet the materials criteria for the formers used in the SYNROC HIP process.

H.6.5 BLENDER

Calcine discharged from the calciner at about 450°C and SYNROC formers preheated to about 700°C are continuously and intimately blended in a zig-zag blender. The continuous zig-zag

design is an outgrowth of the twin-shell (batch) design, known in the industry for its blending efficiency. With each half turn of the blender, part of the material moves forward and part of it moves backward within the legs of the blender. Random splitting, merging, tumbling and rolling bring particles into contact with each other. Particles move by gravity, and the slope and rotational speed of the blender determines the materials' residence time. In addition, comparatively short-term variations in the calcine production rate (i.e. through intermittent calciner filter blowback and vibrator activation) are leveled off by the time the blended material is ready to be discharged. Therefore, a relatively accurate and uniform quantity of material is discharged with each revolution.

Continuous blenders are commercially available with capacities of 10 to 10,000 ft³/h (283 to 283,170 L/h). Pilot-plant, production-sized equipment is also available to experimentally determine required residence time and other parameters of operation. Developmental design efforts are required to:

- adapt the design to remote operation in a radioactive environment, including construction materials, rotary seals, idler wheels, and drive components
- provide shell insulation and possibly a means of heating the shell to maintain material temperature during blending
- adapt the design for remote maintenance, installation and removal.

Table H-4 in Section H.9 gives the blender design requirements, and Figure H-3 (also in Section H.9) shows the configuration of the blender.

H.6.6 SYNROC/CALCINE BLEND DELIVERY SYSTEM

The semicontinuous output of the blender is directed to one of four, batch sintering furnaces by the SYNROC delivery system, which is similar to the SCHIP delivery system described in Appendix G. At the same time process off gas from the other three furnaces flows upward through the delivery system and blender to the spray calciner off-gas system.

Table H-5 (Section H.9) gives the design requirements and Figure H-4 depicts the configuration of the delivery system. The system consists of a four-way diverter and four identical delivery tubes. Each delivery tube has a mechanism for remote coupling to an alumina container positioned in the sintering furnace. A dust-tight coupling is accomplished by a weighted spout that compresses a seal retained in the container lip. A disc-bellow interfaces with the delivery tube and the spout to seal the system, while accommodating thermal expansion and spout decoupling for container exchange. An insulated plug is used to close the spout opening during the sintering cycle and container exchange. The plug allows off gas to escape, but prevents appreciable loss of residual SYNROC to the enclosure when closed. The spout nests into the furnace cover and is lifted with the cover to permit translation of the furnace for vacuum degassing and glass charging.

An offset funnel inside the diverter directs the flow of SYNROC to one of the delivery tubes, which is attached and sealed to the alumina container. A rotary actuator automatically

signals the funnel when the desired quantity of SYNROC has accumulated in the alumina container. The funnel signal may be synchronized with the intermittent flow of material from the blender to provide a "clean" switch. The quantity of SYNROC in the alumina container may be detected by a variety of methods (e.g., radiation-activated level sensor, fill time, accumulated weight, etc.) depending on the consistency of blender output and detailed equipment design considerations.

A two-position diverter has been developed and thoroughly evaluated at PNL as a part of ongoing process demonstrations. Nevertheless, a four-way diverter and SYNROC delivery system must be designed and demonstrated. Developmental considerations are:

- positive control of the SYNROC flow and dusting
- the seal between the spout and the alumina container
- identification of reliable, radiation-resistant actuators and bearings
- prevention of excessive SYNROC accumulations (caused by sintering, condensation of off gases, etc.)
- controlling the spread of residual SYNROC during container exchange.

H.6.7 SINTERING FURNACE

In the sintering furnace, calcined waste and SYNROC formers are fully dried and are densified into a rigid, relatively strong cylindrical billet. The density of the accumulated material is increased from about 1.8 kg/L to about 3.4 kg/L, or about 77% of theoretical.

Four identical sintering furnaces are arranged to mate-up with the furnace covers described as a part of the SYNROC delivery system. Each furnace is supported on a curved track and mechanized to translate out from under its cover to adjacent stations.

At the start of a cycle, an alumina container is transported and lowered into an empty sintering furnace. The neck of the container is shaped to facilitate remote coupling and decoupling with a lifting device, as well as to facilitate sealing to the delivery system. Guides in the furnace center the container. The furnace is remotely translated under its cover; the cover and delivery tube spout are then lowered to engage the furnace and to couple to the alumina container, respectively. The container is then preheated to about 650°C and the insulated plug is retracted in preparation for SYNROC delivery.

As previously described (Section H.2.2), the alumina container is sized to accumulate 534 L of SYNROC/calcline blend. At a blender, design output rate of 258 kg/h and a bulk density of 18 kg/L, 3.7 h are required to accumulate this quantity. During this time, the first SYNROC in the furnace self-heats to approximately 750°C at the center of the container. Preheat temperatures are selected to drive off residual moisture and nitrates without excessive volatilization of cesium and other radionuclides.

When the container is filled, the SYNROC flow is diverted to a container located in another furnace, and the insulated plug is inserted to thermally close off the delivery system.

The SYNROC/calcline charge is then heated to approximately 1325°C in a sintering thermal-cycle similar to that shown in Figure H-5 (see Section H.9). It is assumed here that cesium, ruthenium and other volatile species will be condensed and retained within the calciner/sintering furnace. Heating rates are moderated to prevent thermal gradients in the alumina container that might cause it to fracture. When sintering is complete, the delivery system is decoupled and the furnace is translated to the next station. In Section H.9, data on the sintering furnaces is given in Table H-6 and a furnace is depicted in Figure H-6.

A very small sample (0.01 L) of simulated SYNROC B powder (as-calcined) has been uniformly densified during sintering in an alumina container. The material did not adhere to the container walls. Scale-up experimentation is required to determine the applicability of this approach at volumes 50,000 times that demonstrated. A major factor is the time required to heat the bulk SYNROC material, due to the inherently low thermal conductivity of granular materials. This factor will largely determine the number of furnaces required. Another consideration is the stability of the sintered billet within the alumina container during subsequent transport and charging with molten glass.

Other research and development considerations include:

- characterization of the bulk SYNROC material (thermal conductivity versus temperature, density versus temperature-time, specific heat, etc.)
- effects of internal heat generation and radiation on the sintering shrinkage and the heating rates
- uniformity of densification within a large, sintered billet
- cost effective design of an alumina container for thermal and mechanical durability
- design of a durable furnace and remotely replaceable heating element to withstand the cleaning and thermal cycles
- development of a reliable, remotely operable furnace translator to withstand radiation and cleaning solutions
- determination of the method, frequency and amount of cleaning required
- development of a reliable, dust-tight seal between the alumina container and the delivery system
- positioning of the alumina container in the sintering furnace
- determination of the volatilization characteristics during sintering and their effects on the quality of the sintered billet.

H.6.8 VACUUM-DEGASSING AND GLASS-CHARGING SYSTEM

After sintering, the billet is vacuum-degassed and hermetically sealed with molten glass. This takes place within an enclosure shared with another furnace at a station adjacent to the delivery system. Each sintering furnace is located in the lower portion of a vacuum vessel,

which rides on a curved track and is translated by a pivot arm as shown in Figure H-7 in Section H.9. The upper portion of the vessel contains a glass delivery spout and vacuum-drawing parts. The spout is heated to maintain the delivery temperature of the glass. Data on the vacuum-degassing and glass-charging system is given in Table H-7 and it is depicted in Figure H-7 (see Section H.9).

A glass siphon is sealed to the vacuum vessel and extends into a glass remelt tank, which may be located outside the cell or in a special, radioactively controlled area. The remelt tank is air tight at relatively low pressures, i.e., about 35 kPa (5 psi). The tank has a capacity of about 1 m³, of which 197 L or about 20% of the tank's capacity is required to submerge the sintered billet. Glass cullet is fed into the remelt tank in the form of spheres to facilitate metering, flow and degassing. The spheres may be preheated with process heat to minimize remelt time. The siphon tube extends below the surface of the molten glass by the amount of glass to be siphoned to a container.

At the start of a cycle, the furnace/vacuum vessel is translated into position under the top portion of the vacuum vessel and the vessel halves are pneumatically connected. The temperature is stabilized and the pressure is decreased to about 13 Pa to vacuum-degas the billet. The glass level in the siphon tube raises about 4.4 m above the glass level in the tank to balance the vacuum vessel and remelt tank pressures, which are maintained near 1 atm. The height of the siphon tube is such that the capacity of the vacuum system is inadequate to prematurely suck glass into the vacuum vessel. When degassing is completed, the pressure in the remelt tank is increased slightly to force siphoning of glass into the alumina container while maintaining a vacuum on the billet. When the siphon clears, as indicated by a decrease in pressure differential, pressure to the remelt tank is shut off and the alumina container is degassed through the siphon. Glass cullet is then again fed into the remelt tank through an air lock to recharge the melter. When the tank is charged, the system is pressurized and the vacuum vessel is decoupled. The sintering furnace (and lower vessel portion) is then pivoted to a common unloading station as described in the next sections.

The glass remelt tank design and the vacuum-activated spout systems for intermittent delivery of glass (gobbing) are well known in the industry and are highly developed. While relatively long siphon-runs may not exist, there is no apparent technical reason precluding such a design. An important feature envisioned is the balance of pressure on the siphon tube in contact with the glass to avoid its collapse. In addition, considerable developmental effort is required to demonstrate the principles described above using the glass composition required to meet hot-isostatic pressing criteria. Other research and development considerations include:

- time and vacuum required to "adequately" degas the sintered billet for the designed results from hot-isostatic pressing
- design of remotely replaceable siphon and system
- determination of the volatile characteristics of sintered SYNROC at the required vacuum and temperatures and of the filtering and recycling requirements.

H.6.9 LOAD, REMOVAL AND DECONTAMINATION SYSTEM

The sealed billet is translated in the enclosure to a position for removal. A decontamination system is integrated into this position to minimize the spread of process dust in the enclosure. The decontamination system consists of a high-pressure, superheated steam spray and shrouding to collect the spent steam into an exhaust system that has appropriate scrubbers and filters. The exhaust and steam systems are balanced to prevent steam from escaping into the enclosure or through the enclosure opening. The steam is superheated to the temperature of the alumina container to prevent thermal shock to the container.

Design specifications for the decontamination station are summarized in Table H-8 (see Section H.9) and Figure H-8 (see Section H.9) shows the arrangement of the system. Experimentation is required to evaluate the effectiveness of continuous decontamination and to demonstrate the feasibility of maintaining an effective balance between the exhaust and steam flows.

The length of the sintering, vacuum-degassing and glass-charging cycles plus transfer and decontamination times determines the number of sintering furnaces required. If the turnaround cycle time is 14.8 h (including a 3.7 h fill time at the design rate), four sintering furnaces will be required. Since the fill and sintering times are probably much greater than the degassing and glass-charging times, two degassing and glass-charging stations would be adequate.

H.6.10 FURNACE ENCLOSURE

A furnace enclosure is required to limit the spread of SYNROC process dust during accumulation in the alumina container, sintering, vacuum degassing, glass charging and container exchange. Provisions for recycling the dust are also included. Although a fairly reliable, dust-tight seal is envisioned between the delivery system and the alumina container during filling, experience in delivering the reference process calcine underscores the difficulty of absolute dust control. The sintering of SYNROC before breaking this seal should appreciably decrease airborne dust, as does glass fusion in the reference process. Nevertheless, SYNROC dust contamination of the furnace and alumina container is expected.

Each enclosure isolates two furnaces, a vacuum-degassing and glass-charging station and a removal and decontamination position. The pivot arms and translation tracks are also enclosed; the actuating mechanism is outside the enclosure and may be located outside the cell. The enclosure is cleaned at appropriate intervals using a spray-flush system. The contaminated cleaning solution is blended with HLLW and recycled through the spray calciner.

The enclosure provides flexible service connections to the sintering furnace and furnace translator. It also includes provisions for the remote operation, monitoring, maintenance, removal and installation of the enclosed components. Table H-9 (see Section H.9) summarizes the specifications for the enclosure and its arrangement is shown in Figure H-9 (see Section H.9). A considerable developmental design effort is required to demonstrate an enclosure that is compatible with the internal equipment and all of the remote aspects.

H.6.11 CONTAINER LIFTING DEVICE WITH COVER

A lifting device (carried by an overhead crane) automatically engages the alumina container in the sintering furnace and lifts it through the decontamination station and enclosure hatch. The neck of the container is designed for this purpose. The lifting device has a filtered cover that seals the container opening. This cover prevents the entry of steam during container decontamination. After decontamination, the alumina container is transported to the HIP loading station where the filtered cover is left on to help minimize the spread of contamination in the HIP.

A developmental design effort is required to satisfy the lifting device specifications summarized in Table H-10 in Section H.9. Of particular concern is the integration of the design with the container neck design to provide positive, automatic engagement and disengagement and to prevent excessive thermal shock or mechanical stress to the alumina container. The prospect of handling broken containers must be considered in designing lifting devices and the equipment in which the containers are processed.

H.6.12 HOT-ISOSTATIC PRESS

The principal function of the HIP is to densify SYNROC to essentially 100% of theoretical, to obtain the desired SYNROC crystalline phase characteristics, and to obtain a high degree of uniformity of these characteristics in a large billet. These functions are expected to be accomplished by the isotropic application of high pressure to the sintered billet while at an elevated temperature, which induces consolidation and molecular bonding.

In the envisioned process, pressure is applied to the relatively porous billet by fused glass. The glass composition is selected to provide viscosity and other characteristics at hot-isostatic pressing temperatures and pressures, such that the glass will not appreciably permeate the pores of the billet and will at the same time be essentially impermeable to the argon pressurization medium. As such, the fused glass transfers the isotropic gas pressure to the sintered billet (acting as a hydrostatic fluid), while maintaining hermetic encapsulation of the billet during its densification shrinkage.

After a brief degassing cycle and when the glass and billet are thermally stabilized at the desired temperature, argon gas is pumped into the hot-isostatic pressing vessel and the vessel is pressurized. During a holding period at operating pressure and temperature, the sintered SYNROC billet consolidates to a density of about 4.4 kg/L (estimated theoretical) and undergoes molecular bonding. The sequence of pressure and temperature applications and the number and length of holding periods may be varied to obtain the desired crystalline phase characteristics and uniformity. As the submerged billet shrinks, the glass level drops in the alumina container to maintain pressure equilibrium on the billet. During hot-isostatic pressing the alumina container does not undergo appreciable dimensional change because pressure on the container is nearly equalized inside and outside.

The HIP is then depressurized and cooled somewhat in a manner to preserve the integrity of the SYNROC billet and alumina container. When the HIP pressure is equalized to cell

pressure, the vessel is opened to remove the alumina container. The container might need to be removed while the glass temperature is still near its softening point to avoid possible rupture of the container due to a thermal mismatch between the glass and container. (It may not be possible to develop a glass that meets this criterion as well as the more important hot-isostatic pressing criteria.)

Figure H-5 in Section H.9 shows an estimated pressure-temperature schedule for the selected design parameters. For SYNROC, a hot-isostatic pressing temperature requirement of about 1325°C is expected based upon limited SYNROC B sintering experiments at PNL, whereas a pressure requirement of approximately 100 to 140 MPa (15,000 to 20,000 psi) is expected based upon industrial experience with hot-isostatically pressed ceramic powders. Hot-isostatic pressing equipment specifications are given in Table H-11 (see Section H.9), and the equipment is depicted in Figure H-10 (see Section H.9).

H.6.12.1 Pressure Vessel Entry and Closure System

A bottom-loading, yoke-type closure system was selected for use in the pressure vessel. The yoke-type closure system should be easily automated and purportedly eliminates a stress discontinuity in the vessel wall typical of threaded closures. With this design, the potential for catastrophic vessel failure should be inherently reduced and effectively minimized (if not eliminated). Failure of a seal is not considered to be catastrophic since the stored energy release is considerably throttled in the cover/vessel interfaces to prevent the formation of a shock wave. The yoke-type design also lends itself to stainless steel cladding or enclosure for durability in the typical cell environment.

For the anticipated process requirement, a pressure vessel having a maximum working pressure of 170 MPa at 230°C (25,000 psi at 450°F) should be adequate. Vessels are typically designed to a 25% higher test pressure (213 MPa at 230°C) and hydrostatically tested at an equivalent room-temperature pressure (247 MPa at 21°C). Rupture discs are normally provided to protect the integrity of the pressure vessel. For this design, a rupture setting of 196 MPa $\pm$ 5% would provide a 10% margin (over maximum working pressure) against premature rupture, without exceeding actual test pressure.

H.6.12.2 Furnace

The hot-isostatic pressing furnace is located inside the pressure vessel and is sufficiently insulated to maintain a cold wall for mechanical strength considerations. The exterior of the vessel is jacketed and cooled with water for this purpose. Unlike the top-loaded SCHIP furnace, the SYNROC HIP furnace may be a forced-convection design. In this design, the element material can be either graphite, molybdenum or tungsten. Graphite is a probable choice since it is nearly twice as strong at 2500°C as at room temperature and has excellent thermal shock resistance. The forced-convection furnace design decreases localized temperature gradients in the alumina container because there is no direct radiation. The element structure is simple and lends itself to remote exchange. A smaller vessel diameter can also be used,

significantly reducing cost. Since graphite is less susceptible to high-temperature degradation than molybdenum, the purity of the argon pressurization medium is less critical, simplifying that system.

H.6.12.3 Vacuum System

A vacuum system may be required to degas the hot-isostatic pressing vessel before pressurization, depending on the level of argon purity required for system longevity and the effectiveness of the bottom-load purging system. An appropriately sized roughing pump is expected to develop adequate vacuum for this purpose. The exhaust gas from the pump is vented through the calciner off-gas system to trap and recycle any residual radioactive volatiles or airborne particulates.

H.6.12.4 Gas Pressurization System

An argon gas pressurization system having a capacity of 170 MPa (25,000 psi) is required to supply the maximum expected hot-isostatic pressing pressure for the selected SYNROC design parameters. A typical system having this capability consists of the following components:

- compressor package
- high-pressure, strain-gage transducers and signal conditioners
- fail-safe, air-operated valving
- high-pressure gas recovery, compressor, and intercooler heat exchangers located in an isolated primary loop and a secondary loop, heat-exchanger system
- flowmeter valving
- gas storage reservoir
- redundant pressure controllers
- cascaded and redundant, intermediate-pressure relief valves and high-pressure, rupture disc assemblies vented into the cell
- high-, intermediate- and low-pressure piping
- recovery system.

The compressor package consisting of four single-stage diaphragm compressors is arranged in two, two-stage sets. The triple diaphragm design of these compressors assures noncontaminating gas compression for furnace heating-element longevity and allows for automatic shutdown in the event of diaphragm seal failure.

Water-cooled, high-pressure, gas heat exchangers are included to protect the compressors while operating in series and to prevent the gas discharge components from overheating. All valving is electrically piloted and air-operated to enable remote operation with interlocked, sequential, fail-safe controls. Cascading and redundant, pressure relief valves and rupture discs are located on each isolatable section of the gas pressurization system. An argon gas

storage reservoir with a capacity of 2500 m³ at 18 MPa and 21°C should be adequate, although reservoir capacity and pressure typically have a significant effect on performance of the pressurization system.

The two hot-isostatic pressing vessels required to meet a design throughput of 6.4 canisters/d might be arranged to utilize a single pressurization system. A single system could incorporate back-up compressor capacity to enable maintenance of replacement concurrent with operation. Fail-safe, interlocking provisions would be included in the two-vessel configuration to prevent more than one vessel from being at pressure at a given time. This provision is a part of the overall safety hierarchy.

Gas recovery systems typically reclaim up to 90% of the argon required for a hot-isostatic pressing cycle. During depressurization, gas is cooled and automatically analyzed for contaminants (including radioactivity) using an in-line gas chromatograph. This is necessary to determine if the gas has to be exhausted to the off-gas system or purified by filtering and/or dialysis. Nonradioactive contaminant levels in the range of 1 to 50 ppm are necessary for furnace heating-element longevity. Recovery of gas below a vessel pressure of about 3-1/2 MPa (500 psi) is generally considered impractical; the balance of gas charge would be vented to the off-gas filter system. Radioactive testing of this system may reveal that the argon is so contaminated that an argon recovery system should not be included in the final equipment design.

H.6.12.5 Cooling System

The pressure vessel cylinder is jacketed and water-cooled to retain structural integrity during high-temperature pressurization and to reduce the hot-isostatic pressing cycle time. Integral cooling channels in the vessel closures provide similar functions and maintain the high-pressure seals, furnace connections, thermocouple connections, etc., at safe and reliable operating temperatures.

Cooling water is supplied from an isolated, closed-loop system consisting of circulating pumps, heat exchangers, an expansion tank and necessary valves, piping and control elements. Flow through the closed loop is monitored to ensure maintenance of minimum flow and to activate corrective procedures and emergency cooling, if required. Water flow in the open loop is controlled by an automatic thermostatic valve and is adequate to provide for permissible discharge temperatures.

H.6.12.6 Control and Instrument System

Pressure-temperature parameters in a hot-isostatic pressing system have to be controlled accurately and reliably to safely obtain the desired results. Pressurization and let-down rates usually do not require any control; therefore, they are quite simple in design. Temperature control in the SYNROC HIP process should be less complicated than for the SCHIP process because the SYNROC billet is transferred at hot-isostatic pressing temperature. However, a reliable system is needed for maintaining holding temperature uniformity.

An automated, totally integrated computerized system with instrument system redundancy may be required to obtain an acceptable level of operational safety due to the presence of

radioactive material in the high-energy-level HIP. All routine operations including sequencing, timing, temperature-pressure control, interlocking, quality assurance record keeping, deviation control, system checking, and others would be programmed and automatically performed. In addition to routine control, a basic function of the computer would be to monitor the hot-isostatic pressing system for abnormalities of operation, material stress levels, component temperatures, etc., and to provide diagnostic and trouble shooting guidance to the operator to assure proper maintenance and safe operation of the system. The computer would also monitor all significant parameters that affect the inherent safety of the system and would respond to all situations requiring remedial action in a predetermined manner based upon a careful and thorough fault-free analysis of potential failure scenarios.

A panel with a symbolic system diagram, an instrument system readout of key operating parameters and light indicators (of the operation phase, normal operation, deviations outside set-point tolerances and fault alarms) would be provided for operator surveillance and verification of control response. Manual operation of the hot-isostatic pressing system for maintenance, repair, verification, etc., would be in a semi-automatic mode and would be limited by computer override to maintain overall safe operating conditions based on maximum limits or combinations thereof.

H.6.12.7 System Safety

Five levels of safety are designed into the envisioned system for hot-isostatic pressing of radioactive materials. The levels are prioritized in both the importance of a failure and the ability to correct or override that failure so that the process is affected only to the extent to maintain system integrity or to prevent potentially catastrophic conditions. The levels are as follows:

- Control Level. The control level is the normal operating mode for controlling sequence, time, temperature, vacuum and pressure in the system. At this level the HIP is operated within predetermined set-point tolerances. Pressure transducers, thermocouples, etc., provide feedback to control each support system; controller program and interlocks maintain the proper sequence. The basic quality of the total system design, appropriate maintenance and scheduled replacement of components are expected to minimize deviations from this level of control.
- Warning Level. An excursion outside set-point tolerances or other situations requiring remedial action would temporarily halt the processing cycle pending automatic diagnosis and correction or compensation for the fault. At this level there is time for remedial action without adverse effect on the process or more serious consequences. Visual and audible alarms would summon the operator for verification of the diagnosis. A prioritized list of potential faults might be generated by the computer depending on the complexity of the event. The sequence of remedial actions would be based on probability of occurrence and on the effects monitored, diagnosed and recorded. Once the appropriate response was obtained, the process cycle would continue at the point of interruption. A maintenance and inspection check-list would be generated to prevent recurrence.

- Hazard Level. At the hazard level, a fault would result in the automatic and systematic abortion of a process run in such a way as to maintain the integrity of the system and to preserve the alumina container, if possible. The high-pressure valving, cooling system reservoir and other critical subsystems would be designed and interlocked to revert to a fail-safe abortion mode in the event of loss of electrical power, compressed air, instruments or other support services. Control lights and a siren would be activated to warn the operator. In the event that the fault was remedied during abortion, the sequence could be manually interrupted and the process run placed on hold for analysis of alumina container status and activation of a salvage process, if possible.
- Danger Level. If through three levels of priority action a fault has not been corrected resulting in excessive pressure, then redundant, pressure-activated relief valving would bleed gas into the cells as required to protect the pressurization system and vessel. These valves would be backed up by redundant rupture discs designed to burst at a pressure below the actual vessel test pressure. Hot gas from the rupture disc would be expanded through a nozzle to prevent formation of a shock wave and would flow through the cell filtration system. In a 1700-m³ hot-isostatic pressing cell, pressure would increase by about 14 kPa (2 psi). The cell walls, ceiling and service feed-through ports would be designed to safely contain this pressure increase. An air circulation system, designed to change cell air five times per hour, would require about two minutes to dissipate the resulting volume surge.

At the danger level, actions are not controllable, nor can they be overridden. Rapid depressurization could severely damage the vessel contents, but the primary intention is to ensure safety.

- Critical Level. The critical level represents the fifth and final level of the safety system. Based upon manufacturers' experience with a safety-oriented hot-isostatic pressing system, the vessel is not expected to catastrophically rupture. Nevertheless, to protect operating personnel and to avoid the possible release of radioactive material to the atmosphere, there must be a complete and thoroughly dependable answer to the question of what would happen should the pressure vessel rupture.

In consideration of this question, two scenarios are described to identify a possible range of energy levels involved in such an event. Should a crack occur (and escape detection) causing the vessel to suddenly rupture at maximum operating pressure (170 MPa), the energy release might be similar to an explosion of about 24 kg (53 lbs) of TNT (as typically equated by HIP manufacturers). Or, should all safety systems (previously described) fail to operate and a sound vessel be inadvertently pressurized beyond the capability of the pressurization system (by uncontrolled thermal expansion of the argon) to 2-1/2 times actual test pressure (530 MPa or 77,000 psi) before suddenly rupturing, the explosive results might approach that of about 76 kg (167 lbs) of TNT.

The key to dealing with these energy levels may lie in the "suddenness" of release. Sudden release of pressure in itself is not explosive, but more nearly an adiabatic

expansion of gas from the volume of the vessel to that of the contaminant cell. At least one HIP manufacturer recognizes this in their vessel design and safety claims. All of their parts that withstand pressure are wound with a prestressed wire mantle. In the event of vessel failure, the mantle purportedly contains fragments and sufficiently retards gas escape to prevent shockwave formation. This approach should receive careful consideration because aside from the explosive-like effects of a shock wave and fragment projectiles, the resulting increase in pressure in a 1700-m³ cell may reach 24 kPa (3.6 psi). Safety might be further enhanced by locating the HIP below ground level and sufficiently oversizing the upper portion of the yoke to contain fragment projectiles. The doors to the adjacent isolated cell areas (used for HIP maintenance and repair) might also be constructed to blow out in the event that pressure in the primary containment cell exceeded design level. This would serve as a shock absorber and would further reduce the resulting pressure through expansion to a much larger volume.

A cell design to safely contain a pressure increase of 24 kPa (3.6 psi) should be reasonably possible considering the bulk of materials normally required for radiation shielding. Although containment may be possible, damage may be severe to cell components. The severity of damage and the difficulty of recovering from the event should be assessed.

H.6.12.8 Research and Development Required

Research and development of equipment for the SYNROC HIP process is expected to require considerable effort based upon the number and complexity of unknowns. Preliminary considerations include:

- An experimental hot-isostatic pressing process needs to be developed for sintered SYNROC. Samples need to be prepared to determine density, crystalline phase characteristics and other properties to evaluate the effectiveness of hot-isostatically pressed SYNROC in immobilizing high-level waste and to characterize process requirements. Simulated SYNROC materials should be used in these preliminary experiments to utilize existing hot-isostatic pressing facilities. This work could be carried out using conventional "canning" techniques to encapsulate the billet.
- Based upon the above findings, a glass composition would need to be developed for the hermetic encapsulation and pressurization of the sintered SYNROC at the required process temperature and pressure. Simulated SYNROC materials should also be used in this phase to utilize existing hot-isostatic pressing facilities. This work would also further characterize the process requirements and would provide additional samples for evaluating radioactive waste isolation and for characterizing the glass-SYNROC interface.

The findings from these preliminary experiments would form the basis for expanding and directing the scope of research and development considerations as follows:

- Hot-isostatic pressing equipment would be evaluated to determine means of remote operation, maintenance, inspection, removal and installation.

- Safety concerns relative to the design and operation of a hot-isostatic pressing system and containment cell would be analyzed to assess the probability of inadvertent loss of radioactive material to the atmosphere.
- Thermodynamic modeling and analysis of the hot-isostatic pressing process based on preliminary findings of materials characterizations would be initiated to identify a scaled-up process and to size equipment. The number of HIPs required could be accurately estimated based upon this work. Decisions relative to equipment sizing for process research and development using radioactive SYNROC could be analyzed for technical and economic feasibility. Due to the expected cost of full-scale, hot-isostatic pressing equipment and adequate cell containment, an intermediate-sized, experimental HIP coupled with carefully controlled scaling factors might be most cost effective.
- Thorough and complete materials characterization within the range of expected process temperatures would be performed.
- The hot-isostatic pressing container (alumina or other materials) would be designed and its compatibility with processing conditions and materials would be evaluated. This work would encompass sintering requirements and handling and storage concerns.
- An experimental facility and a remotely operable system for hot-isostatic pressing radioactive SYNROC would be designed and built.
- The system would be operated to 1) develop a hot-isostatic pressing process for radioactive SYNROC utilizing a glass pressure/encapsulant medium in an open container, 2) prepare samples for evaluation and characterization, 3) demonstrate the remote aspects of hot-isostatic pressing, and 4) verify the thermodynamic model and scaling factors. Initially the system would be debugged and operated using simulated SYNROC to permit direct surveillance and product evaluation for system and process verification.

The findings of this work would form the basis for identifying detailed process and equipment design criteria for a full-scale, remotely operable hot-isostatic pressing system and facility.

H.6.13 CONTAINER OVERPACK CANISTER

The function of the overpack canister is to provide a radioactively clean, sealed shell to facilitate safe handling, intermediate storage and transport of the SYNROC billet to its final repository. The alumina container is placed in the overpack canister immediately after removal from the HIP. If this transfer is made while the container is relatively hot (to prevent thermal cracking), the lifting device is preheated to avoid excessive thermal stresses in the neck of the container. Once in the overpack canister, the integrity of the alumina container is no longer important since it is not a functional part of the immobilization of radioactive waste in hot-isostatically pressed SYNROC. Research and development considerations for the overpack canister are similar to those noted in the reference process and also include:

- canister design to accept the alumina container, facilitate closure welding and provide for subsequent handling
- thermal stress analysis of the alumina container during transport to the canister.

H.6.14 CANISTER PROCESSING

After the alumina container is placed in the canister, a helium source is inserted and the closure is installed and seal-welded, as covered in detail in the reference process. The weld is then helium leak-checked to ensure its integrity. Canisters that fail to pass the leak test are rewelded and rechecked; the canisters may also be visually inspected at this time. Sealed canisters are then moved to a decontamination chamber where any loose contamination is removed. From here the canisters are placed in the in-cell storage racks or transferred out of cell to an interim waste storage area as noted in the reference process.

The research and test efforts identified in the reference process for canister handling, closure, seal welding, inspection and decontamination are also be required for the canister processing in the SYNROC HIP process. In addition, selected components and aspects of the SYNROC HIP overpack canister processing system should be fabricated and tested to verify their capabilities and demonstrate their remote aspects.

H.7 RESEARCH AND DEVELOPMENT REQUIRED

Specific research and development requirements were identified by unit operation in Section H.6. Based upon the extent of the work identified and the current state of product and process development, considerable effort will be required to research and develop the SYNROC HIP process to the point where basic design criteria for a full-scale, remotely operable system can be identified. An encompassing program that integrates the sequence of product, process and equipment research and development required is beyond the scope of this report and is perhaps academic at this point because of the high number of fundamental unknowns. Nevertheless, a logical, prioritized research and development approach needs to be identified for the assessment of the potential technical and economic feasibility of the SYNROC HIP process as described. Such an approach might embody the following primary objectives and priorities:

- develop a hot-isostatic pressing process for sintered, simulated SYNROC using existing hot-isostatic pressing facilities and conventional "canning" techniques
- using this process, prepare hot-isostatically pressed samples of simulated SYNROC and evaluate their potential for HLLW immobilization
- if promising, identify (or develop) a glass composition to hermetically encapsulate and pressurize the sintered SYNROC at the identified hot-isostatic pressing pressure and temperature
- concurrently, identify a container material that is thermally compatible with the identified glass composition at hot-isostatic pressing temperature and that shows promise of meeting projected in-process handling criteria; prepare experimental sample containers
- also concurrently, develop a blending process and corresponding SYNROC-former particle size characteristics to provide the desired bulk density and intimate blending with simulated, calcined HLLW; existing batch blending equipment may be used for this work
- develop a laboratory process for sintering blended SYNROC using the identified experimental containers, for vacuum-degassing the sintered billet and for submerging the billet in molten glass while maintaining a vacuum
- using existing facilities, if possible, develop a process for "open-can" hot-isostatically pressing sintered, simulated SYNROC submerged in fused glass
- using all of the above processes, prepare hot-isostatically pressed samples of sintered, simulated SYNROC and evaluate their potential for HLLW immobilization and the glass-SYNROC interface for adverse effects
- based on the foregoing findings, re-evaluate the overall SYNROC HIP product, process and equipment using 1) actual materials characterization data, 2) thermodynamic modeling of the sintering and hot-isostatic pressing increments and of the final product in interim and repository storage, 3) computerized stress analyses of the container, and 4) the active involvement of the manufacturers of hot-isostatic pressing equipment, container blenders and glass delivery systems. Expected results of this analysis would include

relatively accurate estimates of the full-scale process, corresponding equipment requirements and criteria, final product characterization (including storage temperature profiles), required research and development as well as an updated and greatly expanded version of this report.

- based upon the above re-evaluation, assess the technical and economic feasibility of the SYNROC HIP process
- if promising, research and develop equipment and process capabilities to demonstrate all remote aspects of the SYNROC HIP process. An intermediate-scale version of the process would probably be developed because of the cost of hot-isostatic pressing equipment. Simulated SYNROC would be used to facilitate close monitoring, inspection and evaluation.
- concurrently, develop experimental capabilities to sinter, seal and hot-isostatically press radioactive SYNROC to verify the simulated SYNROC process and prepare samples to evaluate HLLW immobilization characteristics of hot-isostatically pressed SYNROC
- based on these findings and demonstrations, identify the conceptual SYNROC HIP product, process and equipment including basic design criteria for a full-scale, remotely operable system.

Initial hot-isostatic pressing experiments should include evaluation of a sealed vitreous container as an alternative to "open can" hot-isostatic pressing using molten glass as a sealing and pressurization medium. Section H.8.6 partially describes this potentially promising alternative.

Only a limited time has been available to develop the SYNROC HIP process preconceptual design. Limited experimental knowledge of the process and the radioactive SYNROC material characterizations are handicaps. Some of the alternatives considered are noted in this section.

H.8.1 TRANSMITTAL OF HOT-ISOSTATIC PRESSING PRESSURE

Typically, powders to be consolidated are precompacted, degassed and sealed in containers. During hot-isostatic pressing, the sealed container collapses or is otherwise deformed on the contained powder to transmit pressure to the powder without allowing the gaseous pressure medium to permeate the relatively porous material and thereby negate pressing. Several methods have been developed involving sheet-metal cans, vitreous containers, elastomeric bags, etc., as the sealing interface. Of the methods considered potentially feasible in a radioactive environment, all involve the task of completely filling the container. Although in itself this task is relatively simple, accomplishing it remotely while controlling excess powder and dust and using radioactive-resistant equipment is considered impractical from a reliability standpoint. In addition, to avoid can failure (from excessive deformation during hot-isostatic pressing), the contained material must be uniformly predensified to about 60% to 70% of the theoretical density. Not only does this requirement further complicate the filling task, it does not preclude occasional can failure. Under best conditions, can failure is expected to occur about 1% of the time based on typical industrial experience. This rate is considered intolerable for the SYNROC HIP process because can failure would probably contaminate the inside of the HIP and the pressurization system and would possibly damage the furnace or cause the can to become wedged in the vessel (in the event of the rapid depressurization of a slow leaking container); this would require complicated cleanup, recovery and reprocessing procedures and further treatment of the breached can.

If technically feasible, the envisioned SYNROC HIP process would eliminate many of the problems associated with conventional canning. The molten glass encapsulant tenaciously conforms to the sintered billet over a wide range of predensification and accommodates nonuniformity of density within a billet. This process also facilitates in-process inspection of sintered billet size and an excess glass charge can be made to assure complete submersion of the billet in a sealing medium. Thus, over- or undercharges of the sintering furnace should be easily accommodated. Furthermore, the open alumina container is not dimensionally affected during hot-isostatic pressing due to pressure equilization and is subjected to compressive pressures one to two orders of magnitude less than typical alumina compressive strengths. However, should the container break during hot-isostatic pressing, the tenacity of the glass would prevent exposure of the sintered billet and contamination of the vessel interior and pressurization system. Recovery procedures may be complicated, although it should be possible in the SYNROC HIP process to provide a back-up shell exterior to the sidewall and bottom insulation to contain any molten glass leakage. With the furnace located at the upper end, damage to

the interior of the HIP would be minimal. This approach would also simplify container alignment in the HIP, since the shell would be removed with the bottom plug for container exchange and could be mechanically referenced on the hoist-trolley with respect to the HIP vessel.

H.8.2 PRE-HOT-ISOSTATIC PRESSING DENSIFICATION

A number of techniques have been developed and commercially applied to densify or consolidate powders, including vibration, tamping, cold-die pressing, cold-isostatic pressing, sintering, etc. The selection of a technique is generally based upon the powder's packing characteristics, which are a function of the particle shape and size distribution. Although the packing characteristics of SYNROC have not been fully explored, limited experiments indicate that the "as-calcined" material responds to sintering. The described SYNROC HIP process approach was selected because it eliminates the volume of material to be calcined, and it assures a minimum as-blended packaging density based upon actual testing of 90% of the material of the blend.

Process complexity of sintering is expected to be comparatively simple, although this area has not been fully explored. Still another consideration in the selection of sintering is the expected variability, based on operating experience at PNL, of calcined material characteristics and variability of blender feed control. Sintering precompaction should easily accommodate these variables, but needs to be experimentally evaluated.

H.8.3 CONTAINER HANDLING

Sintering furnaces and hot-isostatic pressing equipment may be top- or bottom-loaded. The advantages of each approach need to be more fully explored before finalizing the container handling technique for the SYNROC HIP process. Container handling with an overhead crane between the sintering/sealing operations and hot-isostatic pressing (hot transfer) was selected because of the relatively unstable container length-to-diameter ratio (5.5:1). Bottom loading into the HIP was selected to prevent excessive heat loss from the HIP during hot transfer.

H.8.4 VOLATILIZATION OF RADIOACTIVE SPECIES

An underlying concern is the possible high percentage of volatilization of radioactive species during the lengthy SYNROC sintering process. This concern, although not experimentally substantiated, suggests the need for an alternative approach to the described process. A number of approaches might be appropriate, including the following:

- A crown of SYNROC formers only (without blended HLLW calcine) could be added to the accumulated material. The composition, packing characteristics and depth of this crown could be tailored to attract and filter the volatiles during sintering recrystallization. This approach could easily be incorporated into the described delivery system and would have the peripheral benefit of entraining residual radioactive waste calcine in the delivery system and controlling enclosure contamination during transfer of the sintering furnace. The quantity of crown material could be metered and preheated outside the cell and fed through an air lock.

- As a further enhancement of the selective crown, an additional crown of glass beads could be incorporated. By selecting a relatively low-fusion-temperature glass, the accumulated SYNROC could be sealed to the alumina container to further minimize the escape of volatiles. This approach would require vacuum-degassing the material during delivery or before sintering, because the glass seal would attenuate with the accumulation during sintering shrinkage and would prevent subsequent degassification. Continuous electrodynamic-degassing might be used; in this case the output of the blender falls through evacuated glass chambers containing electromagnetic and electrostatic fields and is totally degassed in seconds. The subcrown of nonradioactive SYNROC formers would prevent the potentially undesirable interface of radioactive waste calcine and glass (before radioactive waste recrystallization with SYNROC). Potential contamination of the enclosure would be decreased.
- Another alternative would be to integrate the above sintering station with the molten-glass charging station and to inject a charge of molten glass over the accumulated degassed material. This approach would require a shorter cycle time than required for melting a glass-bead pack and could be carried out at a lower accumulated material temperature to further reduce the likelihood of volatilization. Conversely, this approach would somewhat complicate the delivery system and would probably require two additional siphons and glass remelt tanks (compared to the described process).

H.8.5 PROCESS CONTAINER

A potential hazard of the "alumina" container described is mechanical breakage during transfer to and from the HIP or thermal breakage caused by process abnormalities. The scope of this study does not include the selection of an optimum process container material. Nevertheless, this is an important consideration to the process and comparative safety analyses. For instance, Ringwood (Ringwood et al. 1979) describes several alternative SYNROC compositions having melting points ranging from 1245⁰ to 1340⁰C. These would have correspondingly lower sintering and hot-isostatic pressing temperature requirements, i.e., 1000⁰ to 1200⁰C. The process container requirements are therefore an unknown at this stage of consideration; the number of possible candidate materials are temperature sensitive. Another consideration is the potential feasibility of maintaining an argon atmosphere in the SYNROC HIP process cells utilizing the unrecoverable argon from the hot-isostatic pressing cycle. It may well be feasible to utilize a metallic process container to avoid brittle fracture and eliminate the potential breakage hazard.

In an argon atmosphere, molybdenum, tantalum and columbium alloys would be candidates for temperatures above 1200⁰C. Below 1200⁰C in an oxygen atmosphere, appropriate designs might utilize Inconel[®] 601, Inconel 617, Incalloy 802, Haynes[®] 188 and Hastelloy[®] X. These "super alloys" are marginal at this temperature; however, for a one-time, short-duration service they

[®] Tradename of International Nickel Co.

[®] Tradename of Cabor Corporation (Haynes 188 and Hastelloy X).

may suffice. An overpack canister may not be required; however, the primary considerations are the characteristics of container material often being exposed to the process temperatures and the weldability of this material for final closure. Material characteristics could well be enhanced by the hot-isostatic pressing operation, although the pressure requirements for hot-isostatically pressed metals are generally higher than for ceramics.

H.8.6 VITREOUS PROCESS CONTAINER

A vitreous process container candidate requires separate mention since it significantly alters the overall SYNROC HIP process and may be a preferable approach. A vitreous material is selected whose softening point roughly corresponds to the required hot-isostatic pressing temperature. A glass composition high in silica should provide this basic characteristic based on the 1500°C softening point of 96%-silica glass. The envisioned container would be cylindrically shaped and would have a neck to seal to the delivery system and to facilitate subsequent fusion sealing. A high-temperature metallic shell would encase the vitreous container supported by a 2- to 3-cm interface of alumina beads on the sides and bottom to absorb handling impacts and to provide a path for pressure equalization on the vitreous container. The outer shell would provide a means of handling the vitreous container in the cell and may be suitable for final containment as described in H.8.5. The vitreous container would be coated internally with a layer of SYNROC formers (nonradioactive) using a slurry technique. The described composite container system would be inspected, introduced into the cell, sealed to the delivery system and evacuated to receive the blended SYNROC charge. The charge would be continuously electrodynamically degassed as described previously (Section H.8.4). When the desired charge was accumulated, a crown charge of nonradioactive SYNROC formers would be added. The container would then be fusion-sealed by impinging two gas flames, carrying opposite electrical charge, on the neck below its attachment to the delivery system. As the glass temperature increases, its electrical resistance decreases; this permits the flow of electrical current with the flames acting as electrodes. Glass temperature is rapidly and uniformly increased to the fusion point. Concurrently, the glass viscosity decreases and atmospheric pressure "collapses" the neck upon itself forming a positive fusion.

The delivery system is then decoupled, and the filled, sealed container is transported to a preheat furnace and then hot-transferred to the HIP. During hot-isostatic pressing, the vitreous container collapses on the SYNROC charge and tenaciously conforms to maintain pressure equilibrium on the SYNROC. The alumina interface or magnesia beads would tend to fall around the vitreous container during hot-isostatic pressing shrinkage and maintain it in an upright position. After removal from the HIP, this charge could be topped off to provide the desired heat transfer interface during storage. The interface material would also continue to protect the hot-isostatically pressed billet from mechanical shock and a thermal expansion mismatch with the metallic container; the vitreous container is expected to break away from the hot-isostatically pressed SYNROC during cooling for this reason.

This alternative eliminates the molten-glass charging system and thereby greatly simplifies the process, while providing encapsulant seal reliability during hot-isostatic pressing.

Conventional preheat furnaces could be used; approximately two to three furnaces would be required at the design rate. Only a two-position delivery system would be required based on an estimated fusion seal and transfer time of about 30 to 45 min. There may also be product advantages in a noninterrupted recrystallization cycle, and post-delivery volatilization will be contained. Finally, two HIPs are expected to have the required capacity for this alternative based on previous estimates and on using less expensive preheat furnaces.

Conversely, the delivery system will be somewhat more complex in that an air lock will be required in each leg to maintain a vacuum during delivery; a fusion system will also be required. Resistance fusion is widely used in the glass industry.

The principal new area of research and development required for this alternative involves the selection or development of the vitreous container and corresponding sealing techniques. Continuous electrodynamic degassing would also need to be experimentally evaluated to test its effectiveness on calcined, radioactive waste. In terms of equipment requirements, cell size, utilities and essential material requirements and process reliability assurance, this alternative may be preferable to the SYNROC HIP process and other SYNROC process alternatives identified.

H.8.7 CALCINED SYNROC HIP PROCESS ALTERNATIVE

Another alternative involves the mixing of HLLW and solutions of selected SYNROC minerals plus a colloidal suspension of nickel particles and then calcining the solution for subsequent processing in a manner described for the SCHIP process. This approach would require more calciners (3) and more canisters (X3) due to the low bulk density of SYNROC calcine in the maximum sized containers. It would also require more HIPs (12) to accommodate the greater number of containers.

H.9 DRAWINGS AND DATA SHEETS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

TABLE H-4. Equipment Data Sheet for Zig-Zag Blender

Function

Blends HLW calcine solids and SYNROC-former solids.

Process Interfaces

HLLW calciner; SYNROC delivery system.

Design Parameters

Calcine input = 25.8 kg/h; SYNROC-formers input = 232 kg/h; blend output = 258 kg/h or 143 L/h; stainless steel shell; variable speed controlled drive; enclosed operation; radiation resistant; remote replacement and removal.

Operating Conditions

Temperature = 300^o to 750^oC; throughput = 172 kg/h or 96 L/h.

HLW calcine properties: shape--agglomerated, semispherical particles; particle density = 1.2 kg/L; particle hardness = 5.7 mohs; particle size--90% are 1 to 10 ; bulk density = 0.6 kg/L; moisture volatiles content = 15 wt%.

SYNROC-formers properties: manufactured to particle size, shape and distribution to provide relatively dense packing characteristics when mixed with 10% calcine; particle density = 3.4 to 4.0 kg/L; bulk density = 1.9 kg/L; moisture/volatiles content = 15 wt%

Blend SYNROC output: bulk density = 1.8 kg/L; moisture/volatiles content = 15 wt%; ratio of high-level waste to SYNROC formers = 1.9 by weight.

Features

Solid-solid blending; sealed shell; no moving parts inside shell; variable speed; off-gas vent to calciner; spray-flush cleaning system.

Services or Auxiliaries

Electric (motor and temperature sensor); nitric acid and compressed air (cleaning); instrument air (pressure sensor).

Space Requirements

2 m in dia by 4 1/2 m long.

Maintenance Methods

Remote--periodic spray-flush cleaning cycle in conjunction with spray calciner and delivery system.

Remarks

Similar to Patterson-Kelly Co. continuous zig-zag blender; design modification required for remote drive, radiation resistance, operation at elevated temperatures, remote cleaning cycle and smaller capacity.

FIGURE H-3. Zig-Zag Blender

TABLE H-5. Equipment Data Sheet for SYNROC Delivery System

Function

Directs continuous SYNROC output of blender to one of four batch sintering furnaces; concurrently provides for off-gas flow from the other three furnaces.

Process Interfaces

Blender; four sintering furnaces

Design Parameters

Capacity = 258 kg/h; automatic and remote operation; enclosed and ventilated operation; remote replacement and removal; stainless steel construction; temperature = 300° to 1350°C.

Operating Conditions

Temperature = 750° to 1325°C; capacity = 172 kg/h.

SYNROC-calcine blend properties: bulk density = 1.8 kg/L; moisture/volatiles content = 15 wt%; calcine content = 10 wt%.

Features

Independent weighted seals to furnace and to alumina container; closure furnace cover with an insulated plug during sintering; cascading uncoupling to facilitate furnace translation; sealed enclosure; four-way indexing rotary diverter; off-gas vent to blender; spray-flush cleaning system.

Services or Auxiliaries

Compressed air (pneumatic actuators); electrical (temperature sensors); nitric acid; water (cooling, drain to sintering furnace enclosure).

Space Requirements

5.0 m high by 45 m in dia

Maintenance Methods

Remote, periodic spray-flush cleaning cycle

Remarks

FIGURE H-4. Delivery System

FIGURE H-5. Thermal Cycle

TABLE H-6. Equipment Data Sheet for Sintering Furnace

Function

Densify SYNROC into a cylindrical billet with a density of about 3.4 kg/L.

Process Interfaces

SYNROC delivery system; vacuum-degassing/glass-charging system (located inside sintering furnace enclosure); canister transport system.

Design Parameters

Automatic and remote operation; remote replacement and installation; design capacity = 583 L; inside dimensions are 0.60 m in dia by 3 m high; radiation and nitric acid resistant; temperature range = 25° to 1350°C; cooling capability; enclosure with off-gas ventilation; cleanout capability.

Operating Conditions

Fill rate = 172 kg/h; capacity = 534 L (962 kg) bulk SYNROC (SYNROC properties same as specified in Table H-5).

Thermal cycle: (see Figure H-5) normally 1325°C; 14.2 h (including 5.6 fill time); heating rate = 300°C/h.

Sintered billet properties: cooling rate = 40°C/min; shape--right circular cylinder; density = 3.5 kg/L; weight = 818 kg; volume = 241 L.

Features

Closed-loop cooling; closure of inlet with insulated plug during thermal cycle; radial translation of furnace to adjacent stations; off gas through SYNROC delivery system; means of sealing furnace and container to delivery system; enclosed system during SYNROC accumulation and thermal cycle; spray-flush cleaning system.

Services or Auxiliaries

Electrical (furnace, temperature sensors); compressed air (pneumatic actuators); process water (cooling); instrument air (pressure sensor); nitric acid (cleaning).

Space Requirements

See Table H-9.

Maintenance Methods

Remote

Remarks

FIGURE H-6. Sintering Furnace

TABLE H-7. Equipment Data Sheet for Vacuum-Degassing and Glass-Charging System

Function

Process Interfaces

Sintering furnace; load, removal and decontamination station (located inside sintering furnace enclosure, see Figures H-8 and H-9).

Design Parameters

Vacuum = 13 Pa; glass remelting tank capacity = 1 m³; air tight 13 Pa to 35 kPa; automatic and remote operation; remote installation and replacement; glass-melting rate = 75 kg/h.

Operating Conditions

Glass temperature = 1250°C; vacuum = 13 Pa; molten glass charge = 197 L; charge time = 0.9 h.

Features

Molten glass siphon; glass cullet supply hopper; vacuum and pressure control systems.

Services or Auxiliaries

Electrical (vacuum pump, glass-melting furnace, siphon heaters, temperature sensors); compressed air (glass transfer and pneumatic rotary and linear actuators).

Space Requirements

Remelt tank and cullet hopper located outside cell; siphon and vacuum vessel included in enclosure (see Figure H-8).

Maintenance Methods

Remelt tank is contact; other equipment is remote.

Remarks

FIGURE H-7. Vacuum-Degassing and Glass-Charging System

TABLE H-8. Equipment Data Sheet for Load, Removal and Decontamination System²

Function

Provides access to the sintering furnace enclosure and decontaminates alumina container upon its removal.

Process Interfaces

Sintering furnace enclosure; alumina container lifting device.

Design Parameters

Stainless steel construction; remote operation, installation and removal; guides for engaging alumina container with lifting device; superheated steam cleaning to avoid thermal shock of container; steam collection system built into sintering furnace enclosure.

Operating Conditions

Steam superheated to 1200°C; operation only during removal of container.

Features

Balanced steam flow and exhaust to develop a "steam curtain" flowing into the enclosure; ventilation system.

Services or Auxiliaries

Electrical (temperature sensors); steam (cleaning); instrument air (pressure sensors).

Space Requirements

2 m by 2 m by 2 m

Maintenance Methods

Remote

Remarks

FIGURE H-8. Load Removal and Decontamination System

TABLE H-9. Equipment Data Sheet for Furnace Enclosure

Function

Contains and limits spread of process dust; provides means to periodically clean delivery system and furnaces.

Process Interfaces

Sintering furnaces; SYNROC delivery stations; vacuum-degassing and glass-charging stations; load removal and decontamination station.

Design Parameters

Remote operation, installation and removal; remote access to enclosed furnaces and mechanism for maintenance, installation and removal; stainless steel construction; built-in cleanout capability.

Operating Conditions

Temperature = 250 to 350°C; pressure is negative with respect to cell and positive with respect to furnace when connected to calciner.

Features

Access hatch; air curtain during installation and removal of alumina containers; spray-flush cleaning system; remote service connections to furnaces and enclosed mechanisms; drain to HLLW recycle system.

Services or Auxiliaries

Compressed air (port activators); nitric acid (cleaning); instrument air (pressure sensors); ventilation (uses steam decontamination ventilation system).

Space Requirements

5 m in dia by 4.5 m high

Maintenance Methods

Remote

Remarks

FIGURE H-9. Furnace Enclosure

TABLE H-10. Equipment Data Sheet for Container Lifting Device with Cover

Function

Applies permanent cover to alumina container and automatically engages (and disengages) container for transport by means of an overhead crane.

Process Interfaces

Overhead crane; alumina container.

Design Parameters

Lifting capacity = 5 MT; stainless steel and ceramic construction; remote operation; means to preheat jaws to 1200°C.

Operating Conditions

Container contact temperature = 1300°C; superheated steam temperature = 1200°C.

Features

Automatic engagement and disengagement with alumina container; remote coupling to crane.

Services or Auxiliaries

Compressed air (pneumatic actuators).

Space Requirements

0.6 m in dia by 1 m high

Maintenance Methods

Remote

Remarks

TABLE H-11. Equipment Data Sheet for HIP

Function

Simultaneously apply pressure and temperature to densify SYNROC to nearly 100% of theoretical density.

Process Interfaces

Hot transfer container device.

Design Parameters

Automatic operation; remote loading and unloading through vessel bottom; remote installation and removal; stainless steel construction; minimum fatigue life = 40,000 cycles (965 cycles/yr, 40-yr life); load = 2.5 MT; forced-convection furnace, gas; temperature = 1350°C maximum; maximum working pressure = 170 MPa; workzone dimensions are 0.65 m in dia x 3.0 m high; working fluid is argon gas; argon purge during hot transfer; 500°C/h heating and cooling rates; ASME boiler pressure vessel code; Section VIII, Division 2, applies; argon gas and cooling water isolation to cell; wire around pressure vessel and yoke.

Operating Conditions

Thermal-pressure cycle (see Figure H-5): pressure = 103 MPa; temperature = 1325°C; turnaround cycle = 6.7 h.

Features

Bottom-loading HIP consists of the following major subsystem features: stainless steel clad pressure vessel; rugged, easily replaceable furnace and heat shield module; argon pressurization system, including a reliable, efficient, contamination-free, isolated gas recovery and storage system; automatic, programmed, fail-safe system; yoke-type closure system; closed-loop cooling system; bottom-load hot transfer mechanism.

Services or Auxiliaries

Argon (pressurizing gas, purge gas); water (cooling); electrical (furnace, forced-convection motor, strain gages, temperature sensors, pressure sensors); compressed air (pneumatic actuators).

Space Requirements

5 m by 10 m by 10 m high

Maintenance Methods

Furnace mobile--remote; pressure vessel--remote; closure system--remote; cooling system--semiremote; pressurization system--semiremote.

Remarks

HIP systems of this size and configuration available from Autoclave Engineers, Inc. and ASEA Inc.; considerable development efforts required to adapt to remote and radioactive process.

FIGURE H-10. Hot-Isostatic Pressing Vessel

H.10 REFERENCES

- Ringwood, A. E. 1978. Safe Disposal High-Level Nuclear Wastes--A New Strategy. Australian National University Press, Books Australia, Norwalk, Connecticut.
- Ringwood, A. E., S. E. Kesson, N. G. Ware, W. O. Hibberson and A. Major. 1979. The SYNROC Process: A Geochemical Approach to Nuclear Waste Immobilization. Research School of Earth Sciences, Australian National University, Canberra, A.C.T. 2600, Australia.

APPENDIX 1

TITANATE PROCESS

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APPENDIX I TITANATE PROCESS

I.1 INTRODUCTION

The titanate waste form, which is produced from HLLW, calcium hydroxide and granular titanate additives, may be a viable alternative to the glass monolith produced by the reference process (Appendix A). The production of a titanate waste form is amenable to quality assurance. In addition, some of the product's attributes, such as its thermal stability, leach resistance and resistance to damage on impact, appear to be better than those of the glass monoliths.

The titanate process facility will have the capability to mix, heat and densify materials, press the materials into a billet, enclose the billet in a canister, and process and handle the canisters. The development of a titanate waste form is in a very conceptual stage. Many aspects of the titanate waste form and the process to produce it need to be identified and/or resolved.

I.2 PROCESS DESCRIPTION

The titanate process is based on studies performed at Sandia Laboratories. The process is illustrated in Figure I-1. As discussed in Appendix A, the HLLW is received in the waste facility in storage tanks. Here, sampling is conducted to establish the thermal, chemical and acidic properties of the waste. When these properties have been determined, the HLLW is transferred to an ion-exchange/calcination vessel. Calcium hydroxide is added to lower the acidity of the HLLW and thereby improve ion-exchange efficiency. Calcium titanate is then added as the ion-exchange medium. The calcium titanate and calcium hydroxide are added in the required proportions to adjust the pH and to yield about a 25 wt% oxide waste-loading in the final waste form.

During the period of mixing using air spargers, the contents in ion-exchange/calcination vessels are heated to evaporate moisture from the titanate and HLLW mixture. When the material is dry, it is further heated to drive off the nitrates, increase the density and thereby "calcine" the batch.

Each batch of calcined titanate and waste is pneumatically transferred to a calcine surge tank for storage. As needed, the calcined titanate and waste mixture is discharged from the calcine surge tank to volumetric batchers, which allow a specific volume of the titanate waste to be fed into the hot presses. They can also serve as a monitoring point to obtain the heat loading of the specific volume that will be hot-pressed into its final form.

The hot presses yield a right cylindrical billet formed at 1100°C and 6.9 MPa (1000 psi). A 227-MT (250 ton) mechanical hot press is used to make the 59-cm- (23.2-in.-) dia by 30-cm- (11.8-in.-) high billets in an inductively heated graphite die. Each graphite die and billet is removed from the hot press after pressing for cooling and extrusion of the billet. After extrusion from its die, each billet is loaded into a stainless steel canister. The canister is seal-welded and then helium leak-checked. The canister is decontaminated, inspected and placed in storage to await shipment to a final disposal repository.

FIGURE I-1. Titanate Process Flow Diagram

I.3 BASES AND ASSUMPTIONS

Many of the criteria for the titanate process are given in the Overall Process Bases Section. Additional assumptions applicable to this process are included in this section.

I.3.1 PROCESS BASES

A major assumption for the titanate process is that the properties and quantities of titanate waste obtained by the ion exchange with the calcium titanate are the same or very similar to those obtained from ion exchange with sodium titanate described in a report by Lynch (Lynch et al. 1976). Sandia Laboratories recently demonstrated the calcium titanate process on a laboratory scale as an improvement over the sodium titanate process, but at present has issued no publications on the subject. Based on the encouraging results of this demonstration, Sandia Laboratories has redirected its efforts to the calcium titanate waste form and process. As a consequence, this study is based on the improved calcium titanate process.

Additional assumptions include:

- The density of the hot-pressed titanate waste is 4 g/cm^3 .
- The density of the calcined titanate waste is 2 g/cm^3 .
- The waste-loading is 25 wt% based on 25% waste oxides and 75% TiO_2 and CaO .
- Ion exchange occurs rapidly.
- Decay heating does not exceed the sintering temperature of the calcined titanate waste.

I.3.2 CANISTER

The major limiting factor in the canister design is the allowable heat loading of 3.06 kW per canister. This loading is achieved with approximately 82 L of the titanate waste in a canister. Specifically, the canister has a 61 cm (2 ft) ID, is 30.5 cm (1 ft) high and has .64-cm- (.25-in.-) thick walls. Each canister accommodates a single, large billet. The canister design is shown in Figure I-2. The lid is designed to encompass any fluctuations in the height of the billet and still minimize the possible air gap between the top of the billet and the lid.

I.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

The effluents from the titanate process are similar in contents to those from the reference process (Appendix A). The volume of process off gas should be slightly greater, but not by a significant amount. This slight increase is due to 1) the air needed for the pneumatic transfer system and 2) an additional CO_2 load on the system if the spent graphite dies are burned in an oxidizing oven.

As with the effluents, the wastes from the titanate process are assumed to be of the order of magnitude in volume and contents as the reference process. It is also assumed that the fuel reprocessing plant has systems and provisions to treat, reduce the volume and package both liquid and solid wastes. Any and all solidified scrap from the titanate process that contains high-level waste is recycled back into the process.

FIGURE I-2. Canister

I.3.4 UTILITIES AND MATERIALS

Estimates of the utility services and essential materials required are shown in Tables I-1 and I-2, respectively. These estimates are based on the requirements for the reference process with adjustments for the additional process steps and anticipated space and volume changes.

TABLE I-1. Estimate of Utilities Needed for Titanate Process

Utility	Required Capacity	Average Use Rate	Disposition
Electricity	3100 kW	1837 kW	--
Raw water	1556 L/min	460 L/min	--
Process water	600 L/min	20 L/min	Recycled as HLLW or ILLW
Sanitary water	40 L/min	20 L/min	Sanitary or chemical sewer-tile field
Air	750 scmm	500 scmm	As appropriate
Steam	26 kg/min	6 kg/min	As appropriate

TABLE I-2. Estimate of Materials Needed for Titanate Process(a)

<u>Material</u>	<u>Amount Needed Per Unit Time</u>
Ca (Ti ₂ O ₅ H) ₂	884 kg/d
Ca (OH) ₂	282 kg/d
Canisters	4.3/d
Caustic (19M)	8,000 L/yr
Nitric acid (12.2M)	12,000 L/yr
Helium sources	5.0/d
Argon	4,250 to 5,660 L/d
Detergent (for example, Tide)	1,000 kg/yr
Ammonia	120,000 kg/yr

(a) Excludes maintenance materials.

I.4 FACILITY AND EQUIPMENT REQUIREMENTS

The general requirements for the equipment and process cell for a waste facility are covered in the Overall Process Bases Section. The major pieces of equipment are listed in Table I-3. Figure I-3 indicates the general arrangement of the titanate process equipment.

TABLE I-3. Major Processing Equipment

<u>Equipment</u>	<u>Number Required</u>
HLLW feed system	1
Ion-Exchange/calcination vessel	2
Off-Gas system	1
Pneumatic transfer system	1
Calcine surge tank	1
Volumetric batcher	2
Hot press	4
Cylinder extruder and canister loader	1

FIGURE I-3. Titanate Process Cell and Equipment Arrangement

I.5 STATE OF PROCESS DEVELOPMENT

The calcium titanate process is in the very preliminary stages of development. Encapsulation of simulated HLLW using calcium titanate was demonstrated on a laboratory scale by Sandia Laboratories and was shown to be quite promising. No documentation on the calcium titanate process is available, but much has been written on the sodium titanate process covering extensive research, cold and hot demonstrations and material characterization. However, because the use of calcium titanate has indicated better processing and material qualities, it was chosen as the ion-exchange media for this study of the titanate process.

Favorable characteristics of the titanate waste form are:

- better than or comparable to leach resistance of glass (Schwoebel and Johnstone 1976)
- higher density than glass (Schwoebel and Johnstone 1976)
- higher thermal conductivity than glass by a factor of 2.8 (Schwoebel and Johnstone 1976)
- 25 wt% tested and potentially up to 50 wt% waste-loading capacity (optimum waste loading hasn't been determined). This feature may be useful in accommodating the variable concentration ranges of HLLW.

The major drawback of the titanate process is the hot-pressing operation. Operational experience with a hot press in a containment cell is limited and hasn't been demonstrated on a continuous processing basis. The time needed for hot pressing and cooling may also be a constraint.

Most of the unit operations for the titanate process have been demonstrated in cold and hot operations. The ion-exchange mixing and calcination vessel is in essence a pot calciner; the ion-exchange/calcination vessel has been demonstrated by McElroy (McElroy et al. 1968) using actual radioactive wastes. The other major unit operation in the titanate process is the hot-pressing operation. Hot pressing has been used to produce 5.1-cm- (2-in.-) dia billets containing simulated wastes at Sandia Laboratories (Johnstone 1978) and 1.3-cm- (.5-in.-) dia billets containing radioactive wastes at ORNL (Johnstone 1978). Scaling up this process to produce 59-cm- (2-ft-) dia billets will no doubt present many engineering difficulties. Although nonradioactive experience does exist with this sized hot-pressing operation (Haskell Sheinberg, personal communication, Los Alamos Scientific Laboratory), actual hot-cell processing with a full-scale press of that size will require considerable developmental work.

The laboratory-scale demonstrations with the titanate solidification process have indicated it to be a potentially viable process. Further research and demonstration on a pilot-scale, along with characterization of the calcium titanate waste form, needs to be accomplished before this process can be considered applicable to hot-cell operations.

I.6 UNIT OPERATIONS

The titanate process is divided into 11 unit operations. In this section, each of these unit operations is discussed in detail, including the equipment, operations and areas needing further research and development. Drawings and data sheets on the equipment and operations discussed in this section can be found at the back of Appendix I in Section I.9.

I.6.1 HLLW FEED SYSTEM

The HLLW feed system for the titanate process is identical to the reference process HLLW feed system excluding the recirculating feed system to the spray calciner. The cooling, agitation, ventilation, transfer, start-up and shutdown operations are all similar to those covered in Appendix A for the reference process.

I.6.2 FEED AND ADJUSTMENT TANK

The function of the feed and adjustment tank (see Figure I-4 in Section I.9) is to receive a specific volume (950 L) of HLLW from the HLLW feed system. This tank provides for measuring by volume the amount of HLLW necessary for a single, batch ion-exchange and calcination operation. This tank also allows for holdup during which analytical measurements necessary to make adjustments to the HLLW feed are made. Specifically, the acid molarity, heat content and chemical content of the HLLW are determined. Calcium hydroxide is added to adjust the acid molarity. If the heat content or chemical content is too high, some of the HLLW can be removed from the tank for processing later. Table I-5 in Section I.9 gives the design and operational features for the feed and adjustment tank.

I.6.3 ION-EXCHANGE/CALCINATION VESSEL

The primary functions of this vessel are 1) acid neutralization, 2) ion exchange and 3) calcination. A 950-L batch of HLLW is added to the tank. Approximately 71 kg of calcium hydroxide $[\text{Ca}(\text{OH})_2]$ are added to the HLLW to neutralize the solution to a pH of about 1. After neutralization, ~221 kg of calcium titanate $[(\text{Ca}(\text{Ti}_2\text{O}_5\text{H})_2)]$ ion-exchange media are mixed with the neutralized HLLW. The calcium hydroxide and calcium titanate additions yield a final waste form having an oxide waste-loading of approximately 25 wt%. The general equation for the ion-exchange reaction (Lynch et al. 1976) is given below:

In this reaction a nonspecific cation in the aqueous waste (M^{+2}) exchanges with the calcium. The calcium titanate and waste complex is very likely a much more complex structure than indicated by the equation. Therefore, the above equation is only a generalization of the chemical reaction.

The residence time for each batch should be long enough to ensure fairly complete ion exchange since the ion-exchange reaction has been reported, by R.B. Dosch at Sandia

Laboratories, to occur instantaneously. The operating cycle for the ion-exchange/calcination vessel is broken down in Table I-4. The heating times in Table I-4 were calculated assuming a 50% power efficiency input requirement. As shown, the ion-exchange process has potentially up to 7 h of mixing and exchange time.

Once all the materials have been fed into the ion-exchange/calcination vessel, the batch is heated to 700°C and the calcination process begins. Three induction-heating zones divide this vessel. The lower 150-kW^(a) zone provides liquid heating, evaporative heating and calcination heating. The middle 150-kW^(a) zone and the upper 100-kW^(a) zone supply strictly liquid and evaporative heating functions. The ion-exchange/calcination vessel shown in Figure I-5 (see Section I.9) is designed so that after most of the water vapor and nitrates are driven off, the material to be calcined resides in the lower and smaller-diameter portion of the tank. In this way, as the volume in the tank is reduced, liquid-level indicators automatically turn off the various heating zones since they are no longer needed.

Air sparge rings in the vessel provide the agitation for mixing the liquid during the ion-exchange and evaporation steps. As the liquid level in the tank is reduced, the air spargers are shut down accordingly. Vibrators are also provided at two levels in the vessel to aid in the discharge of the calcined titanate and waste material.

To meet the design capacity flow rate of 5700 L HLLW/d, the titanate process requires two ion-exchange/calcination vessels. Other design and operating constraints are given in the ion-exchange/calcination vessel equipment data sheet (see Table I-6 in Section I.9).

Besides the investigation into whether or not the neutralization procedure to improve the ion-exchange process is necessary, other areas requiring research include methods of removing material from the vessel and minimizing the processing time. The pot-type calciner has been shown to be effective for calcination, but the removal of the calcined material has not been demonstrated with a pot calcination vessel. Methods to prevent caking or to dislodge and removed caked material must be developed for such problems. The cycle time for the actual

TABLE I-4. Operating Cycle Times for Ion-Exchange/Calcination Vessel

<u>Operation</u>	<u>Time, h</u>
Filling	0.6
Liquid heating	1.0
Evaporation	6.6
Calcination	1.0
Product removal	1.4
Time lags	<u>0.4</u>
TOTAL	11.0

(a) 150 kW and 100 kW are average values.

batch operation on a pilot-scale, and later on a full-scale, needs to be confirmed. Actual operating experience is necessary to optimize the ion-exchange, evaporation and calcination time.

I.6.4 OFF-GAS SYSTEM

The requirements for and the operation and function of the titanate process off-gas system are similar to those specified in Appendix A for the reference process off-gas system. Volatility during calcination and hot pressing has not been estimated. While atomizing air is not used as in the spray calciner, the use of air spargers, the pneumatic transfer system and increased air inleakage in the pressing equipment may result in similar off-gas processing rates. Operating efficiencies may necessitate parallel systems.

I.6.5 PNEUMATIC TRANSFER SYSTEM

The function of the pneumatic transfer system is to transport calcined titanate and waste from the ion-exchange/calcination vessel to the calcine surge tank. Because radioactive material is being transferred, a negative-pressure system is used. The driving force for the transfer system is a steam jet that creates a vacuum for transporting this material up to 9.1 m (30 ft) in height. The steam jet discharges to the off-gas system's condenser. A schematic of the off-gas system is shown in Figure I-6 in Section I.9.

The design transfer rate of 400 kg/h in 5.1-cm (2-in.) tubing provides a line velocity of approximately 18.3 m/s (60 ft/s) and a solids-to-gas mass ratio of approximately 3.0 (Skriba et al. 1977). At the design rate the total titanate waste transfer will require approximately one hour. Table I-7 (see Section I.9) provides additional information on the pneumatic transfer system.

The flow, measured by pressure-sensing devices located in the ion-exchange/calcination vessel, in the calcine surge tank and at the inlet and exit flanges of the transfer line is monitored and controlled from the control room. Transfer temperatures are determined by temperature indicators located in the sending and receiving vessels. Plugs in the line will be cleared with vibrators and a blowback system. This transfer line is also insulated to prevent condensation.

Research and development requirements for the pneumatic transfer system include development and testing of pneumatic transfer procedures and equipment for titanate waste materials. Optimization of the transfer system to obtain sufficient flow with a minimal vacuum supply is another developmental consideration.

I.6.6 CALCINE SURGE TANK

The calcine surge tank functions as a material holdup point prior to the hot-pressing operation. The design capacity of this tank is approximately 1200 L (42.4 ft³) or a full processing day's holdup.

The calcined titanate and waste is transferred to the calcine surge tank via the pneumatic transfer system. The material enters the upper section of the calcine surge tank as illustrated in Figures I-7 and I-8 (see Section I.9). This upper section is designed to operate as a cyclone separator, stripping the titanate waste from the pneumatic air transport. Vacuum is supplied by the pneumatic transfer system when material transfers are taking place and by the off-gas system when no material is being transported. The off gas discharged passes through a sintered-metal filter with blowbacks before entering the main off-gas system. Table I-8 in Section I.9 is the data sheet for the calcine surge tank.

From the calcine surge tank, the material is gravity-fed into one of two volumetric batchers, as needed. An air vibrator is attached to the calcine surge tank to facilitate the discharge of calcined titanate waste to the volumetric batchers.

Areas of future research and development for the calcine surge tank include:

- heat transfer determinations to see if the tank needs fins added to increase the cooling rate
- testing of the vibratory and gravity flow properties of the titanate and waste material to determine if it can be removed from this tank by a gravity-fed method.

I.6.7 VOLUMETRIC BATCHERS

The volumetric batcher measures a specified volume of calcined titanate waste in preparation for discharging and loading into a hot press. The volumetric batcher illustrated in Figure I-9 (see Section I.9) is designed to contain 164 L (5.79 ft³) of calcined titanate waste material with a density of 2.0 g/cm³. Gamma ray spectrometry is used to indicate the level and volume of calcine in the batcher. When the volumetric batcher is full and the hot press is ready, the batch is transferred to the hot press by gravity and vibration. Gamma ray spectrometry is used to monitor any buildup of material in the transfer lines. Other information on the volumetric batchers is given in Table I-9 in Section I.9.

It is assumed that a sufficient amount of the internal heat is dissipated without the use of fins to prevent the titanate and waste calcine from sintering and agglomerating in the batcher. Testing the calcium titanate waste complex's sintering point, and even its melting point, is an area for future research. Heat transfer calculations for this type of batching vessel configuration along with the ease of material transfer from these vessels must also be determined. Material transfer throughout the titanate process is a major area requiring research and development since no transfer experience with this type of material has been developed.

I.6.8 HOT PRESS

The purpose of the hot-pressing operation is to compact a batch of titanate-waste calcine into a dense, ceramic billet 59 cm (≈2 ft) in dia by 30 cm (≈1 ft) high. The maximum temperature required for this operation is 1100°C (Johnstone 1978) and the required pressure is

6.9 MPa (1000 psi). Heating is provided by induction-heating coils, and the pressure is delivered through a unidirectional mechanical press with a 227-MT (250-ton) pressing capacity.

Right cylindrical billets are hot-pressed in dies made from low-grade graphite. The die assembly is shown schematically in Figure I-10 (see Section I.9). A Grafoil[®] disk is placed between the calcine batch and the plunger to prevent a reaction between the waste and the plungers. The Grafoil disks are backed by graphite spacers of the same composition as the die body, followed by the rams. Two thermocouple wells lined with closed-end alumina sleeves are positioned diametrically opposite one another. One thermocouple is attached to a programmer that controls the heating rate, maximum temperature and residence time. The second thermocouple provides back-up temperature monitoring. The graphite die body also serves as a susceptor for the inductive-heating systems.

The illustration of the hot-pressing mechanism (Figure I-10 in Section I.9) shows that the calcine enters the hot press from the volumetric batcher. With the upper plunger raised above the feed inlet, the material is gravity and vibratorily fed into the pressing zone while the lower plunger is held in the stationary filling position. During and upon completion of the filling process, the hot press vibrates the calcine to level and partially compact it. When the batch of calcine is loaded, a pressure load of 6.9 MPa (1000 psi) is applied to the two rams. At the same time, the press is inductively heated under vacuum with the graphite die serving as a susceptor. The heating rate is controlled to 35°C/min (Johnstone 1978), but it is assumed that the hot press can be preheated to approximately 500°C since the titanate material has already been calcined to 700°C, which reduces the pressing time. When the maximum temperature of 1100°C is reached, the billet is maintained at this temperature for a residence time to assure a fairly uniform temperature distribution and densification throughout the billet. The desired density is 4.0 to 4.1 g/cm³, which is an increase by a factor of two over the density of the calcine. Additional hot-pressing information is supplied in Table I-10 in Section I.9.

Research and development necessary for hot pressing include:

- development of an automatic-loading hot press
- scaling the hot press up to the large sized billet being produced
- testing large-scale operations to check the cycle time estimate
- material characterization studies of the calcium titanate waste product
- determining the average number of hot pressings a die can withstand
- improving scaled-up demonstration under radioactive hot-cell conditions.

I.6.9 DIE AND BILLET COOLING AND TRANSFER

After the hot pressing and initial cooling to 550°C, the formed waste billet and die assembly are removed from the hot press, loaded on a die transfer cart and transferred to the

[®] Tradename of Amax Specialty Metals Corporation.

billet extrusion station. The die transfer cart is designed to hold the billet in the die to prevent the billet from prematurely falling out of the die. This transfer is performed partially by rolling the die transfer cart and partially by lifting the transfer cart with a crane up to the extrusion station (see Figure I-3). If the hot-pressing operation is staggered, the transfer system needs to be operated once every 3.5 h at design capacity and every 5.5 h under normal operating conditions.

Determining whether the cart and crane system will be effective in transferring the die and billet is an area for research and development. Another transfer method might be the use of a conveyor system, although its use would necessitate the need for a complex mechanical system that could potentially have a higher degree of maintenance associated with it.

I.6.10 BILLET EXTRUSION AND CANISTER LOADING STATION

The billet extrusion device is a mechanical ram similar to that used with the hot press. Figure I-11 (see Section I.9) illustrates the billet removal and canister loading procedure. The canister lift raises the canister up to the extrusion station for loading. When the canister is in position, the die transfer cart is positioned at the extrusion station. Then, in one continuous motion, the billet ram extrudes the billet from the die assembly and force-loads the billet into the canister. It is anticipated that minimal force will be needed on the billet ram due to the fact that the thermal expansion coefficient of the graphite die is expected to be lower than the billet. A vacuum cleaning system may be required in this operation to control the spread of radioactive dust.

After the billet has been loaded into the canister, the canister and its contents are transferred for lid emplacement, welding and further processing. The die is removed from the loading depression and is recycled back to the hot press for future pressings. The present estimate of die life is about 20 pressings (Lynch et al. 1976). The interaction between the graphite and waste form is the main factor affecting die life. When the graphite dies are spent, they could be placed in a low-temperature ($\sim 700^{\circ}\text{C}$), oxidizing oven and burned to form CO_2 . The majority of the waste contamination that remains behind in the form of ash could be recycled in the waste solidification process. Table I-11 in Section I.9 supplies additional processing information.

Required developmental work on the billet removal operation includes 1) testing the ease in which the billet is extracted from the die and loaded into the canister, 2) demonstrating the operation on a remote pilot or full scale, 3) doing material characterization studies to determine the thermal properties of the titanate waste form such as the thermal expansion coefficient, and 4) doing studies on limiting the interaction between the die and the solidified waste, thus increasing the die life.

I.6.11 CANISTER PROCESSING

The full canister is moved to the weld station, a helium source is inserted and a lid is placed on the canister and seal-welded. Next, the canister is helium leak-tested to ensure its

integrity and is visually inspected for any flaws. Canisters that fail to pass the leak test are rewelded, if possible. Sealed canisters are then moved to a decontamination chamber where loose contamination is removed. From here the canisters are placed in in-cell storage racks or are transferred outside the cell to an interim storage facility to await final disposal. Canister processing is covered in detail under the reference process (see Appendix A).

I.7 RESEARCH AND DEVELOPMENT REQUIREMENTS

In the titanate process, a need exists to test and develop each component of the system. This will involve operating individual equipment components on a small scale and the integrated system in the cold mode. In addition, it will require both cold and hot operation in a remote mode. The installation and test demonstration will also aid in verifying the cell equipment system and arrangement.

Characterization of the calcium titanate waste form is a major area for future research since only limited work has been performed on this to date. Specific thermal properties of interest are:

- thermal conductivity before and after calcination
- thermal conductivity of the pressed product
- specific heat of the calcined and pressed titanate
- thermal expansion coefficient of the hot-pressed material
- sintering temperature of the calcined titanate.

Other characterization studies needed for the calcium titanate product are leach testing and process volatilization testing.

The material transfer systems will need development since none have ever been demonstrated with the titanate material. The pneumatic transfer system may experience plugging, which must be considered in the design. It must be determined if the titanate material can be made to flow in the gravity-vibratory feeding system.

The lack of heat capacity and conductivity data prevented the calculation of the heat dissipation within the various process vessels. Once the heat transfer rates are determined, vessel designs can be updated or modified, if necessary, to maintain the material below the sintering temperatures or to allow it to flow and transfer easily.

I.8 ALTERNATIVE PROCESSES AND EQUIPMENT CONSIDERED

Only a very limited time has been available to develop the preconceptual design bases described in this document for the titanate process. Considering this, there are probably a number of viable alternatives that have not been considered or that should be explored further in preparing a finalized process design.

A processing alternative for this process is to replace the titanate ion-exchange material with a zirconate material. The zirconate process is the same as the titanate process with minor physical changes in the final zirconate waste form as compared to the titanate waste form. Levine (1976, 1978; Levine and Nowak 1977) has discussed the possibility of using zircalloy waste hulls as a feedstock to a chemical process for preparing the zirconate ion-exchange material. Enough zirconate could be prepared from the waste hulls to stabilize the HLLW generated, and in this way two waste streams are combined into one stable product for storage. Preliminary studies (Levine 1978) have indicated that the zirconate conversion from waste hulls and encapsulation with HLLW should be technically and economically feasible. With these encouraging results the zirconate alternative should be considered in the final decision of this process. The emphasis should be on whether the titanate or zirconate is the better waste form as far as leach resistance, thermal and physical properties and economics are concerned.

I.8.1 CELL ARRANGEMENT

The cell arrangement displayed in Figure I-3 is one possible arrangement of the remote processing equipment described for the titanate process. Alternatives to this arrangement should be investigated for possible improvements of the process material flow and the remote requirements for operation, maintenance and replacement of equipment.

I.8.2 ION-EXCHANGE/CALCINATION VESSEL

A batch or pot-type calcination vessel was selected for this process. From the information Sandia Laboratories has provided, pot calcination appeared to be a reliable method by which evaporation and calcination could be conducted. According to Sandia personnel, the calcined titanate and waste flowed easily out of a laboratory-scale calcination container; therefore, the material transfer properties of the calcined titanate and waste seemed to be quite good. If the material was not calcined in the same vessel as the ion-exchange mixing process, the resultant slurry could be pumped to and calcined in a spray calciner or in a fluidized-bed calciner. However, Sandia indicated that the mixture was too viscous to pump. If future testing shows the material can be pumped without diluting the mixture, then an ion-exchange mixing vessel coupled to a spray calciner could be a process improvement. However, decrepitation of the waste and titanate complex grains may occur during transfer, which could adversely affect powder compaction and pressing operations.

Because of the higher thermal conductivity of the calcined titanate waste as compared with that of directly calcined HLLW, a pot calciner may have a large enough volume and still

be able to transfer the internal decay heat generated. It should be noted that for the two calcination vessels in this report only the heating requirements for evaporation and calcination and the design flow capability were taken into consideration in developing the process flow requirements. If the decay heat generation proves to be too great for these vessels and the evaporative heat transfer coefficient proves to be too low and/or if the thermal conductivity of the titanate and waste calcine proves to be too low, more vessels with reduced volumes may be needed. The nuclear heat dissipation requirements are an area of future research and development for this process.

If processing constraints (including processing time, product quality, heat transfer problems, etc.) make it impossible to use the pot calcination vessel, the option of diluting the ion-exchange slurry for material transport convenience still exists. Once the titanate material is in a pumpable form, it can be fed into a spray calciner or a fluidized-bed calciner for calcination of the titanate and waste. However, since these calciners do not adequately denitrate the calcine, a second, higher-temperature calcination process must be used before hot pressing.

I.8.3 VOLUMETRIC BATCHERS

Instead of measuring the batches to be fed to the hot press on a volume basis, these batches could conceivably be measured on a weight basis using load cells. Material characterization studies supplying the average calcine density, and its range, may provide greater insight into which method is better for this particular use. The volume method was chosen to prevent the potential of overloading the press volume, which could occur if a lower-density calcine was measured on a weight basis.

I.8.4 HOT PRESS

The hot-isostatic press is an alternative to the uniaxial hot press for consolidating the titanate and waste into a ceramic form approaching its theoretical density of 4.1 g/cm^3 . However, additional processing steps would be required to further densify the titanate waste calcine before it could be hot-isostatically pressed. An alternative to this scheme would be the introduction of chemical fluxes, such as iron compounds, into the calcined waste to form a low-melting ceramic complex that could be melted via an ICM as in the reference process.

I.8.5 PNEUMATIC TRANSFER SYSTEM

Alternatives to the negative-pressure pneumatic systems described would include a positive-pressure flow system which is not recommended for handling radioactive nuclear material, or the conveyance of the material by gravity which would increase the cell height by a factor of approximately two.

I.8.6 DIE AND BILLET COOLING AND TRANSFER

After the hot pressing and initial cooling to 550°C , the formed waste billet and die assembly are removed from the hot press for further cooling and transfer to the billet extrusion

station. This transfer step can be performed either with a crane or a conveyor system on an average of every 3.5 h at design capacity and once every 5.5 h under nominal operating conditions. Research may be required to determine whether the transfer logistics and dust containment requirements favor crane or conveyor transfers.

1.9 EQUIPMENT DATA SHEETS AND DRAWINGS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

FIGURE I-4 Feed and Adjustment Tank

TABLE I-5. Equipment Data Sheet for Feed and Adjustment Tank

Function

Supply a specified volume of HLLW to the ion-exchange/calcination vessel and allow for any adjustments and measurements necessary to the HLLW feed.

Process Interfaces

HLLW feed system; ion-exchange/calcination vessel.

Design Parameters

1425 L/batch; 4 batches/d; remote operation, monitoring, maintenance and replacement; HLLW sampling capability; cooling capability; chemical addition capability; ventilation; transfer capability to HLLW storage tank and to ion-exchange vessel; drain.

Operating Conditions

Approximately 950 L/batch at 4 batches/d.

Features

Agitator; temperature sensors; sampler; atmospheric pressure sensors; level sensor; cooling glands.

Services or Auxiliaries

Chemical addition for adjustments and cleaning; vacuum (vessel ventilation); electrical (agitator and temperature sensors); instrument air (pressure and level sensors); water (cooling); compressed air (agitator alternate, air lift transfer system).

Space Requirements

1.22 m by 1.22 m by .99 m (4 ft by 4 ft by 3.25 ft)

Maintenance Methods

Remote

Remarks

FIGURE I-5. Ion-Exchange/Calcination Vessel

TABLE I-6. Equipment Data Sheet for Ion-Exchange/Calcination Vessel

Function

HLLW neutralization to pH of 1; ion exchange; calcination of titanate waste.

Process Interfaces

HLLW, $\text{Ca}(\text{Ti}_2\text{O}_2\text{H})_2$ and $\text{Ca}(\text{OH})_2$ feed systems; process off-gas system; pneumatic transfer system.

Design Parameters

2104 kg/batch at 2 batches/d, 11/h batch; heating capacity = 115 kW for upper zone, 170 kW for middle zone and 170 kW for lower zone; vibration capability; flushout capability; remote operations, monitoring, maintenance and replacement; chemical addition capability, including calcium hydroxide and calcium titanate; mixing capability.

Operating Conditions

1403 kg/batch at 2 batches/d, 7.5 h/batch; heating to 700°C.

Features

Induction-heating furnaces (3 zones); vibrators; air sparge rings; temperature sensors; pressure sensors; liquid level sensors; high temperature block valve; internal heat transfer plates.

Services or Auxiliaries

Chemical addition $\text{Ca}(\text{OH})_2$ and $\text{Ca}(\text{Ti}_2\text{O}_5\text{H})_2$; electrical (heating, temperature sensors); instrument air (pressure and liquid level sensors); compressed air (air spargers, vibrators); water (induction coil cooling).

Space Requirements

1.52 m in OD by 3.2 m high (5 ft in OD by 10.5 ft high)

Maintenance Methods

Remote

Remarks

FIGURE I-6. Pneumatic Transfer System

TABLE I-7. Equipment Data Sheet for Pneumatic Transfer System

Function

Transfer calcined titanate waste from ion-exchange/calcination vessel to calcine surge tank.

Process Interfaces

Ion-exchange/calcination vessel; calcine surge tank; process off-gas system.

Design Parameters

528 kg/batch; specific gravity = 2.0; pipe routing to minimize line length and turns; transfer rate = 400 kg/h; line velocity = 78.3 m/s (60 ft/s); line size = 5.1 cm (2 in.) in ID; solid-to-gas mass ratio = 3.0; remote operation, monitoring, maintenance and replacement.

Operating Conditions

352 kg/batch; 1 batch every 6 h; maximum inlet vacuum = 15 in. Hg; system pressure drop = 5 psi

Features

HEPA filter; vacuum jet; pressure sensor; blowback flush system and vibrators.

Pipe: stainless steel; 2 in. schedule 40; mill run 2B interior finish; 61-m section bend radius

Connectors: Cherry Burrell's I line or similar; remote flange joints.

Services or Auxiliaries

Dry compressed air (pneumatic transfer air, vibrators); instrument air (pressure sensors); steam (pneumatic transfer vacuum jet); chemical addition (blowback flush system).

Space Requirements

6 cm in OD by 9.14 m in length (2.4 in. in OD by 30 ft in length)

Maintenance Methods

Remote

Remarks

FIGURE I-7. Calcine Surge Tank

FIGURE I-8. Schematic of Calcine Surge Tank

TABLE I-8. Equipment Data Sheet for Calcine Surge Tank

Function

Holdup tank for the calcined titanate waste.

Process Interfaces

Pneumatic transfer system; volumetric batchers.

Design Parameters

Designed to hold one day's processing volume or ~1200 L (2400 kg); maintains temperature below sintering point; stainless steel construction; vibration to aid in draining; level sensing capability; pneumatic transfer dust separation capability; remote operation, monitoring, maintenance and replacement.

Operating Conditions

0 to 2400 kg titanate waste calcine content; 500°C maximum temperature.

Features

Vibrators; temperature sensors; pressure sensors; cyclone separator; sintered-metal filters with blowback; level sensors (gamma ray spectrometry).

Services or Auxiliaries

Compressed air (vibrators, filter blowback); electrical (temperature sensors); water (cooling as required); instrument air (pressure sensors).

Space Requirements

1.22 m by .91 m by 1.22 m high (4 ft by 3 ft by 4 ft high)

Maintenance Methods

Remote

Remarks

FIGURE I-9. Volumetric Batcher

TABLE I-9. Equipment Data Sheet for Volumetric Batchers

Function

Measure a specified volume (164 L) of titanate waste calcine in preparation for hot pressing.

Process Interfaces

Calcine surge tank; hot press.

Design Parameters

Designed to measure material (specific gravity = 2.0) for one hot-pressing operation; maintains temperature below sintering point; stainless steel construction; vibration to aid in de-airing and draining; full and empty sensing capability; ventilation capability; remote operation, monitoring, maintenance and replacement.

Operating Conditions

Temperature is less than 500°C.

Features

Vibrators; temperature sensors; level sensors (gamma ray spectrometry); block valves.

Services or Auxiliaries

Vacuum (vessel ventilation); compressed air (vibrators); electrical (temperature sensors)

Space Requirements

.61 m by .61 m by .91 m high (2 ft by 2 ft by 3 ft high)

Maintenance Methods

Remote

Remarks

FIGURE I-10. Hot Press

TABLE I-10. Equipment Data Sheet for Hot Press

Function

Form a ceramic titanate waste billet at high temperature and pressure.

Process Interfaces

Volumetric batchers; billet and die cooling and transfer area.

Design Parameters

Heating to 1200°C (induction); graphite die/susceptor; graphite spacers and graphoil disks between plungers and pressing material; 7.6 MPa (1100 psi) or a 250-ton pressing capacity for mechanical hot press required; length-to-diameter ratio = 1; beginning specific gravity = 2.0 and final specific gravity = 4.0; remote operation, monitoring, maintenance and replacement; inert gas purge; water cooling capability.

Operating Conditions

Temperature = 1100°C; pressure = 6.9 MPa (1000 psi); filling and hot pressing-time = 6 h; cooling and billet and die removal time = 6 h; 2 hot-pressing operations/d/press.

Features

Uniaxial mechanical hot press with 60-cm-(2-ft-) dia rams capable of 7.6 MPa (1100 psi) and 1200°C; vibrators; temperature sensors; movement indicators.

Services or Auxiliaries

Hydraulic fluid (pressing); electrical (heating, temperature sensors, movement indicators); compressed air (vibrators); vacuum (vessel ventilation); water (die cooling).

Space Requirements

1.83 m by 1.83 m by 3.05 m high (6 ft by 6 ft by 10 ft high)

Maintenance Methods

Remote

Remarks

FIGURE I-11. Billet Extrusion and Canister Loading Station

TABLE I-11. Equipment Data Sheet for Billet Extrusion and Canister Loading Station

Function

Removes the billet from the die assembly and in the same downward ram motion loads the billet into the canister.

Process Interfaces

Billet and die cooling and transfer; canister processing.

Design Parameters

One billet is received every 3.5 h; remote operation, monitoring, maintenance and replacement; dust vacuuming and containment capability.

Operating Conditions

Billet removal time = 1 h; one billet is received every 5.5 h.

Features

Mechanical ram for extruding the billet from the die; mechanical lift to position the canister in place for loading; a preformed loading depression with hole for the billet to pass through and position for the die to rest in; position indicators.

Services or Auxiliaries

Vacuum (dust collection, ventilation); electrical (ram, position indicators); steam; water.

Space Requirements

.91 m by .91 m by 3.05 m high (3 ft by 3 ft by 10 ft high)

Maintenance Methods

Remote

Remarks

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APPENDIX J

CONCRETE PROCESS

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APPENDIX J

CONCRETE PROCESS

J.1 INTRODUCTION

The concrete process facility will have the capability to 1) calcine the HLLW, 2) mix the calcine with concrete-forming materials, 3) load these into a canister, 4) heat and pressurize the canister and its contents and accelerate the hardening of the concrete, 5) decontaminate the canister, 6) store the filled and vented canister to allow free water to escape, and 7) seal the canister before moving it to an interim storage facility or a final disposal repository.

The concrete process is a fairly simple process, although it appears somewhat more complex than the ICGM process. Its primary disadvantage may be the relatively low quality of the concrete waste form. Concrete has been found to exhibit relatively poor leaching characteristics and contains a high percentage of water, which may be released through radiolysis or through heat in an accident involving fire.

J.2 PROCESS DESCRIPTION

A flowsheet depicting the concrete process is shown in Figure J-1. As in the ICGM process described in Appendix A, HLLW is pumped from the HLLW feed system to a spray calciner for drying. The resulting dry powder, or calcine, is allowed to fall into a vibrated calcine receiver located below the calciner. When the level of calcine in the receiver reaches a certain height, as determined by gamma emission measurement (Nielson and Brodzinski 1978), the calcine flow to the calcine receiver is valved off and the batch of calcine in the receiver is pneumatically transferred to the volumetric batcher. The volume of the waste transferred to the batcher is determined and compared with the volume measured in the receiver. Next, the calcine is discharged to a receiving hopper where appropriate amounts of cement, fly ash and illitic clay are added through a weight batcher. Storage silos and the additive weight batchers are located in a nonradioactive area, and the material is introduced into the cell through air locks or back-flow preventors.

When waste and additives in the proper proportions have accumulated in the receiving hopper, the material is discharged to the concrete mixer which already contains an appropriate amount of water. Due to heat generation from radioactive decay and chemical reactions, the concrete mixer must be cooled to maintain a temperature of less than 50°C. This is necessary to prevent early setting of the concrete and will be accomplished with a water-cooled jacket around the mixer.

After the waste and additives have been mixed the desired length of time, they are discharged to a canister located beneath the mixer. While filling, the canister is vibrated to release trapped air and to pack the concrete material in the canister.

The filled canister is transferred to an autoclave where it is pressurized to 600 psi and then heated to 110°C at the walls of the canister over a period of 1.5 to 7 h. The high pressure prevents the concrete in the center of the canister from boiling. The canister is maintained at the elevated pressure and temperature for 24 h to permit the concrete to set, and then it is cooled for another 24 h while still in the autoclave (Moore, Newman and Rogers 1979). When the canister has cooled to ambient temperature, it is decontaminated and placed in preliminary storage for an unspecified number of years. During preliminary storage, free water in the concrete waste form slowly diffuses through the concrete and escapes through a filter in the canister lid. After the preliminary storage, the canisters are sealed and transported to either an interim or final storage facility.

Liquid condensed from the spray calciner off gases is routed to a waste concentrator for recycling, while the liquid from the autoclaves is drained to a holding tank for recycling to the concrete mixer.

FIGURE J-1. Preconceptual Concrete Flow Diagram

J.3 BASES AND ASSUMPTIONS

The concrete process will be designed and operated according to the general criteria discussed in Section 3, Overall Process Bases. In this section, process bases and assumptions specific to the concrete process are presented.

J.3.1 PROCESS BASES

The characteristics of the process materials and the concrete waste form are given in Table J-1.

During the setting process, the centerline temperature of the canister in the autoclave is maintained below the boiling point of water (250°C at 600 psi). The equilibrium centerline temperature (T_1), which is a function of configuration, decay heat and heat of dehydration, can be determined from the following equation:

$$T_1 = T_2 + \frac{AR^2}{AK} = 181^\circ\text{C} ,$$

where

T_2 = wall temperature = 100°C

A = setting heat = (4.0 W/L + 10.6 W/L) × (1000 L/m³) = 14,600 W/m³

R = radius of waste = 0.1496 m

K = thermal conductivity of wet concrete = 1.15 W/m-°C^(a).

TABLE J-1. Characteristics of Process Materials and Concrete Waste Product

Characteristics	Process Materials		Concrete Waste Product
	Calcine	Wet Concrete	
Thermal conductivity, W/m-°C	0.25	1.15	0.6
Specific gravity, kg/L	1.2	2.0	1.6
Decay heat, W/L	42.6 ^(a)	10.6	10.6
Temperature, °C	350	--	--
Volatile content, wt%	5	--	--
Heat of hydration over 24 h, W/L	--	4.0	--
Total heat of hydration, W-h/L	--	10.6	--
Water losses, %	--	--	70
Volume/canister, L	--	--	171
Total canister heat loading, kW	--	--	1.82

(a) 44.8 W/L for oxides.

(a) 1.92 times the conductivity of dry concrete.

After hydration, the concrete is maintained at a temperature high enough to promote the diffusion and escape of water during air storage, but not so high that water of hydration is released. The equilibrium centerline (T_1) and wall (T_2) temperatures are determined using the following two equations:

$$T_2 = \frac{Q}{2h\pi RL} + T_{\text{air}} = 120^{\circ}\text{C} ,$$

where

$$T_{\text{air}} = \text{air temperature} = 50^{\circ}\text{C}$$

$$Q = \text{canister heat loading} = 1820 \text{ W}$$

$$h = \text{heat transfer coefficient} = 11.35 \text{ W/m}^2\text{-}^{\circ}\text{C}$$

$$R = \text{radius of waste} = 0.1496 \text{ m}$$

$$L = \text{length of waste} = 2.44 \text{ m} ,$$

and

$$T_1 = T_2 + \frac{AR^2}{4K} = 219^{\circ}\text{C} ,$$

where

$$A = \text{decay heat} = 10,600 \text{ W/m}^3$$

$$R = \text{radius of waste} = 0.1496 \text{ m}$$

$$K = \text{thermal conductivity of dry concrete} = 0.6 \text{ W/m-}^{\circ}\text{C}.$$

J.3.2 CANISTER

Some general requirements for waste canisters are given in the Overall Process Bases Section and are expanded on in Appendix A. Specific details on the concrete process canister are noted here.

The canister used in the concrete process is depicted in Figure J-3 (see Section J.8) and is described in Table J-5 (Section J.8). Approximately 7 canisters are produced daily, each of which contains about 51 kg of high-level waste oxides and about 1.82 kW of heat. Since gases are generated inside the canister after sealing, the canister must be treated as a pressure vessel.

J.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTE

It is assumed that the concrete waste form is flexible enough to accommodate most high- and intermediate-level waste streams generated while processing HLLW. These waste streams include decontamination solutions,^(a) scrubber solutions, incinerator ash, and others that are considered too high in activity to be economically processed in the cementation facility; the cementation facility is considered to be a part of the fuel reprocessing plant. The liquid

(a) Decontamination solutions are steam, H_2O or dilute HNO_3 .

wastes that are destined for recycling are concentrated and added to the HLLW storage tanks. Condensates from the autoclave are collected in a storage tank and are recycled as water additions to the mixers or to the autoclaves. Due to the fact that the concrete process is more expanded and complex than the reference process, there may be more worn equipment that eventually will have to be discarded. With the exception of additional off-gas processing capacity to accommodate the air from the pneumatic transfer system, the effluents from the concrete process are similar in content and volume to the effluents from the reference process.

For the concrete process, it is assumed that all solidified process scrap that may contain high-level wastes is recycled and incorporated into the process stream. As with the other processes in this study, the extra processing requirements for recycling effluents, wastes and scrap have not been fully identified. Although recycling requirements for the concrete process are likely to be less than for more complex processes, they add to the process requirements and complexity when this process is compared to the reference process.

J.3.4 UTILITIES AND MATERIALS

The estimated utilities and essential materials required for the concrete process are listed in Tables J-2 and J-3, respectively.

TABLE J-2. Estimate of Utilities Needed for the Concrete Process

<u>Utility</u>	<u>Required Capacity</u>	<u>Average Use Rate</u>
Electrical, kW	800	500
Cooling water, L/min	1500	700
Process water, L/min	400	10
Sanitary water, L/min	20	12
Instrument air, scmm	1	1
Ventilation air, scmm	750	500
Steam, kg/min	50	10

TABLE J-3. Estimate of Materials Needed for the Concrete Process

<u>Material</u>	<u>Amount Needed per year^(a)</u>	<u>Amount Needed per Canister^(b)</u>
Canisters, lids and helium sources	2100 each	1.0
Nitric acid, L	1200	--
Cement, kg	2.6×10^5	128.3
Fly ash, kg	6.3×10^4	30.4
Illitic clay, kg	6.3×10^4	30.4
Suspending agent, kg ^(c)	2.8×10^3	1.4

(a) Multiply by 1.5 for design capacity.

(b) Requirement based on current studies.

(c) May be omitted.

J.3.5 INTERIM STORAGE

The concrete waste form is stored in air in open canisters for several years to permit the diffusion and escape of water vapor. If the concrete waste form is to be eventually disposed of in a salt repository and its wall temperature is to be limited to 100°C or less, as described in Section 3, Overall Process Bases, an interim storage period of 27 yr is required to reduce the decay heat rate to allowable levels.

It is assumed that the fuel reprocessing plant and the concrete waste processing facility are located at the site of the repository. This is because the concrete waste form may not be thermally stable when insulated inside transporting vehicles and exposed to the proposed fire scenarios. Locating the waste facility next to the repository could reduce overall shipping and handling costs.

J.4 FACILITY AND EQUIPMENT REQUIREMENTS

The general process cell and equipment requirements for a waste facility are covered in the Overall Process Bases Section. The major pieces of equipment required are listed in Table J-4. Figure J-2 is a diagram of the process cell and the arrangement of the equipment for the concrete process. The dimensions of the cell are shown in the figure.

TABLE J-4. Major Processing Equipment Required

<u>Equipment</u>	<u>Number Required</u>
Calcine feed system	1
Calciner	1
Condenser	2
Calcine receiver	1
Pneumatic transfer system	1
Filter assembly	4
Volumetric batcher	2
Receiving hopper	2
Weigh batcher	1
Concrete mixer	2
Sampling system	2
Canister holder	2
Autoclave	32
Blowdown tank	4

FIGURE J-2. Preconceptual Concrete Process Cell and Equipment Arrangement

J.5 STATE OF PROCESS DEVELOPMENT

All of the equipment in the concrete process has been widely used in industrial applications. Most of the equipment has been demonstrated in nuclear applications as well. Although concrete has been successfully used in the United States and in Europe (Lokken 1978) to immobilize low- and intermediate-level wastes, the production of large, concrete monoliths incorporating high-level wastes has not been demonstrated to the best of our knowledge. Thus, a number of the steps for this process will require demonstrations to verify they can be operated in a fully remote, production-scale process.

Plant-scale HLLW storage and feed tanks have been in use since the mid 1900's in the Waste Calcination Facility at Idaho National Engineering Laboratory. This and the research and development efforts conducted at PNL and other laboratories have established a high level of knowledge on this system and its appurtenants that can be applied to the concrete process.

An engineering-scale spray calciner has been demonstrated with HLLW in tests at PNL. These tests occurred during the WSEP operations and during the NWVP. A plant-scale spray calciner has also been demonstrated at PNL using simulated high-level wastes. These efforts have established a good and firm basis for this major piece of equipment that can be applied to the concrete process.

The development and use of pneumatic conveying of material is not a new application, except in the field of handling powder or fines of nuclear material. Information contained in documents authored by Skriba, Wu and Yang (1977) and Lyon and Laroueve (1979) and from the Technology for Commercial Radioactive Waste Management (U.S. Department of Energy 1979) shows that nuclear powders can be transported via negative-pressure systems. However, considerable research and development efforts will be required to establish the design of each individual system.

Concrete mixing and casting of low- and intermediate-level nuclear wastes is a well developed technology. Large, concrete castings using low-level waste have been produced in the United States, as well as in Europe. Intermediate-level wastes have been mixed with a concrete grout and injected into bedrock at ORNL. Extensive lab-scale testing of high-level, defense waste concrete matrices was performed at SRL. The prospect of incorporating high-level wastes in concretes has been advanced by J. G. Moore (Moore, Newman and Rogers 1979) and others at ORNL as the "FUETAP" (formed under elevated temperature and pressure) process. The higher temperatures and pressures used in this process may enable large monoliths of concrete bearing high-level wastes to be cast without boiling. Boiling would likely compromise the quality of the waste form and complicate equipment decontamination requirements.

Plant-sized autoclaves are routinely used in the rubber and plastics industry. Smaller autoclaves have been used in a variety of nuclear applications. Although a plant-sized, remotely operable autoclave has not yet been used with high-level wastes, the development of such an autoclave appears to be feasible.

The curing and dewatering of concrete in applications such as foundations and dams is well developed and documented. Relatively little information exists on the curing and dewatering of concrete containing high-level radioactive wastes (Weeren and Perona 1979; McCormack, Postma and Shur 1979). Adequate dewatering may be important in assuring that sealed canisters of concrete do not overpressurize.

J.6 UNIT OPERATIONS

The concrete process consists of 11 unit operations. The following subsections describe and analyze each of the unit operations. Drawings and data sheets on the equipment and systems discussed in this section can be found at the back of this appendix in Section J.8.

J.6.1 HLLW FEED SYSTEM AND SPRAY CALCINER SYSTEM

The function, the requirements and the operation of these systems are basically the same as those identified under the reference process (Appendix A). Only one spray calciner is required for the concrete process. The research and development efforts identified in the reference process are also applicable to the concrete process.

High-level liquid waste is jetted from the fuel reprocessing plant to a storage tank. When full, the waste is analyzed for chemical, curie, heat and solids content. Following analyses, the waste is air-lifted to a feed tank from which it is pumped to a spray calciner at a nominal rate of 158 L/h. In the spray calciner the liquid waste is converted to a dry powder, or calcine. As calcine is produced in the spray calciner, it falls to the base of the calciner and through a remotely made connection into the calcine receiver. Actual processing rates may be slightly higher if decontamination solutions, dissolved scrap and intermediate-level wastes are included.

For drawings and other details on the HLLW storage tanks and spray calciner unit operations, see Appendix A, Sections A.6.1 and A.6.2.

J.6.2 CALCINE RECEIVER

The calcine receiver is a batch collection hopper for calcine produced in the spray calciner. A batch of calcine is the amount of high-level waste that is ultimately contained in a single waste canister. The calciner receiver is depicted in Figure J-4 (see Section J.8) and described in Table J-6 (Section J.8).

Calcine falls to the bottom of the calcine receiver where it is prevented from draining out by a closed, remotely actuated valve. A vibrator mounted on the side of the calcine receiver is periodically operated to compact and level the calcine to a fairly repeatable density. The level of calcine and, to some extent, its heat generation rate may be monitored by strategically placed temperature sensors in the calcine receiver. When the level of calcine reaches that required to constitute a batch, as precisely indicated by gamma ray emission, a remotely actuated valve located between the spray calciner and the calcine receiver is closed. The valve at the base of the calcine receiver is simultaneously opened, which allows the charge to drain into the pneumatic transfer system.

Vacuum to the calcine receiver is constantly supplied via the calciner or the pneumatic transfer system. Calcine in the calcine receiver is kept from overheating due to its own decay heat through the use of internal cooling fins welded to the walls of the calcine receiver. The

use of fins allows the calcine temperature in the calcine receiver to be maintained at or below 550°C. A pressure sensor constantly monitors the pressure differential between the calcine receiver and the calciner, pneumatic transfer system and cell. A chemical addition capability enables liquid flushing of the calcine receiver.

Further research and development may be required to:

- determine the range in calcine bulk densities to establish the feasibility of basing batch size on volume
- determine the range in calcine thermal conductivities to ensure that the calcine in the calcine receiver will not overheat and self-agglomerate or cause damage to the calcine receiver
- prove that gamma ray spectrometry is a reliable method of determining calcine levels
- select or design valves.

J.6.3 PNEUMATIC TRANSFER SYSTEM

The pneumatic transfer system is used to convey batches of calcine from the calcine receiver to a volumetric batcher. The system is shown in Figure J-5 (see Section J.8) and described in Table J-7 (Section J.8).

When a calcine batch is discharged from the calcine receiver, an interlocked steam jet is simultaneously activated, which provides the vacuum and air movement necessary to transfer the calcine batch to one of two volumetric batchers. The routing to the designated volumetric batcher is previously established. The transfer system uses a solids-to-air ratio of approximately two to transfer batches of calcine weighing as much as 68 kg. Calcine is transported at approximately 9 m/s through a pipe having an ID of approximately 2.5 cm. The time required to complete a batch transfer is approximately 15 min. The progress of the transfer is followed with pressure sensors located in the calcine receiver, volumetric batchers and at the inlet and exit flanges of the connection line. The calcine-laden air stream enters the volumetric batcher where a cyclone separator and a filter separates the air and calcine. The steam-air mixture is treated in the same process off-gas system used to treat gases from the spray calciner.

Calcine is very hygroscopic and is likely to form plugs if moisture is present. To minimize the possibility of plugging, the transfer line is insulated to prevent condensation. In addition, dry air is added at the upstream end of the transfer system to avoid drawing moist air into the system. If plugs do form, they will be cleared with air or liquid blowback systems. The use of such a blowback system increases the risk of radioactive contamination of the pressure sensing and dry air systems, as well as the cell itself. Special backflow preventers and administrative procedures will be required to prevent this.

Further research and development may be required to:

- assess the potential for plugging of the transfer system with calcine produced in the spray calciner

- establish methods of unplugging transfer lines
- obtain a reliable valving system capable of diverting calcine-air streams to selected sources
- determine if calcine could flow by gravity to the mixing system if equipment were stacked; however, this would result in very high cell requirements.

J.6.4 VOLUMETRIC BATCHER

The volumetric batcher receives the calcine from the calcine receiver via the pneumatic transfer system. Here it is confirmed that the volume of the batch is the same as that measured in the calcine receiver. The heat content of the batch is also monitored. The volumetric batcher is shown in Figure J-6 (see Section J.8) and described in Table J-8 (Section J.8).

As the calcine is pneumatically transferred to one of two volumetric batchers, the majority of the calcine is separated from the transfer air by a cyclone separator. The calcine remaining in the air stream is separated out in a filter chamber located above the volumetric batcher. As calcine accumulates in the volumetric batcher, a vibrator mounted on the side of the batcher is activated to compact the calcine to approximately 1.2 g/cm^2 . The vibrator is also used to help empty the volumetric batcher. Internal cooling fins or pins are used to maintain the temperature of the calcine below 500°C . When an entire batch has been transferred to the volumetric batcher, as inferred by pressure differences between the calcine receiver, filter chamber and volumetric batcher, vibration is continued for several minutes to ensure adequate calcine compaction. The volume of the batch is then determined by gamma ray spectrometry. If the volume content is satisfactory, the batch is prepared for mixing with concrete-forming additives.

Further research and development may be required to:

- determine optimum placement of temperature sensors
- correlate temperature data with heat content
- design cyclone separator for good efficiency
- optimize placement and design of vibrator.

J.6.5 FILTER ASSEMBLIES

A filter assembly located directly above each volumetric batcher is used to separate the remaining calcine from the air stream used to transfer calcine to the volumetric batcher. Identical filter assemblies are also located above each receiving hopper in which calcine and concrete-forming additives are combined before mixing. The filter assembly is shown in Figure J-7 (see Section J.8) and described in Table J-9 (Section J.8).

Four filter assemblies are used. Dust entrained in the processes is collected on the filters. The steam jet used in the pneumatic transfer system provides the vacuum and the flow of air that carries dust to the filters. The filters may be comprised of banks of sintered-metal

candles. Each filter assembly is equipped with an air pulse blowback system, which removes collected dust from the filters and allows it to fall into the vessel below. Each filter assembly is also equipped with pressure sensors on each side of the filters and a chemical addition line for flushing filters. A temperature sensor is required to ensure that the filter assembly is maintained above 100°C, the condensation point of water.

Further research and development may be required to:

- optimize air pulse pressures and cycles
- select a filter medium
- assess filter pluggage potential and life.

J.6.6 RECEIVING HOPPER

The receiving hopper is used to collect and blend concrete-forming materials, i.e., calcine, cement, clay, fly ash and suspending agents, before discharging them to the mixer. The receiving hopper is shown in Figure J-8 (see Section J.8) and is described in Table J-10 (Section J.8).

A batch of calcine is emptied from a volumetric batcher into one of two receiving hoppers. A batch of inert, concrete-forming additives is then added to the receiving hopper. The inert, concrete-forming additives are measured out in a nonradioactive area of the waste facility where the batch weights are checked twice before the batch is introduced through air locks to the receiving hopper. Vibrators are used to ensure that additives are completely transferred to the receiving hopper. When all materials have been added, an air sparging system is used to blend and cool the materials. As previously mentioned, a filter assembly is located above each receiving hopper to collect dusts that are entrained by the sparging air.

A valve is located between the receiving hopper and the mixer. When opened the batch of material in the receiving hopper is discharged to the mixer. The mixer is maintained at a slightly higher vacuum than the receiving hopper during the discharging operation to prevent moisture from being drawn into the hopper.

Further research and development may be required to:

- select valves and air locks
- optimize design of sparging system
- determine the need for methods of assuring complete drainage of the receiving hopper.

J.6.7 CONCRETE MIXER

The concrete mixer is used to mix water with the mixture of calcine and inert, concrete-forming materials discharged from the receiving hopper. The wet concrete mix is discharged into a waste canister. The mixer is shown in Figure J-9 (see Section J.8) and is described in Table J-11 (Section J.8).

Before calcine and inert, concrete-forming chemicals are added to the mixer, most of the concrete water requirements for the batch are added to the mixer to prevent concrete residues

from hardening in the mixer. The mixer may be operated continuously to ensure that all surfaces are kept wet. After all materials are added to the mixer, the mixer motor amperage is monitored continuously as an indication of the torque required to mix the concrete. Extra water may be added as a function of torque and total mixing time.

The mixer is the shape of a horizontal drum. Mixer blades radiate from a concentric drive shaft, which has an operating range of 0 to 300 rpm. The mixer contains a water-cooled jacket to help maintain the concrete temperature below 50°C if found to be necessary. The mixer has access doors to aid in cleaning and inspection. As with other process vessels, the mixer is maintained at a slight vacuum relative to the cell environment. The mixer off-gas system and discharge valves are specially designed to minimize and/or clean concrete buildups. A concrete grout sampler may be located below the discharge valve, if required.

Further research and development may be required to:

- ensure long-lived shaft and access door seals
- provide methods for cleaning the mixer, discharge valve and off-gas line
- ensure adequate water in concrete mix; water may evaporate or leak from the mixer between mixer batches
- determine the value and need for concrete grout sampling
- determine optimum mixing speed
- finalize and test the design of a remotely maintainable unit.

J.6.8 CANISTER HOLDER

The canister holder contains and vibrates the canister while it is being filled with concrete and provides a sump to collect material that drips from the concrete mixer. The canister holder is shown in Figure J-10 (see Section J.8) and is described in Table J-12 (Section J.8).

An empty canister is placed in one of two holding receptacles. It is then moved via a trolley into vertical alignment with the discharge flange of the concrete mixer and coupled with that flange. After the contents of the concrete mixer are emptied into the canister, the discharge valve is closed and is flushed with a small amount of water. The flush water falls onto the surface of the newly cast concrete. At this time, vibrator(s) that are built into the frame of the canister holder are activated to facilitate the rising of air bubbles that may be present in the concrete. When flushing and vibrating are completed, the canister is disconnected from the discharge flange and the canister holder is retracted. This action positions the canister holder sump under the discharge flange to catch material that may fall from the area of the flange. The sump is equipped with a level detector, a hot nitric acid addition line and jet out capability to one of the HLLW tanks. The nitric acid is used to dissolve and/or slurry the spilled materials.

Additional research and development may be required to:

- optimize the method of connecting the canister and mixer
- determine vibration requirements
- design discharge valve flush system.

J.6.9 AUTOCCLAVE AND ASSOCIATED EQUIPMENT

The autoclave is used to accelerate the setting and curing of the newly cast concrete waste form while preventing it from boiling. The autoclave is the key to the FUETAP (Moore, Newman and Rogers 1979) concrete process on which this process design is based. The autoclave and associated equipment are shown in Figure J-11 (see Section J.8) and described in Table J-13 (Section J.8).

As soon as practical, a loose-fitting lid is placed on the concrete canister; then the canister is placed in one of 32 autoclaves. When the autoclave is sealed, warm water is added to the annulus between the autoclave and the canister to enhance heat transfer between the two items. Pressure in the autoclave is then increased to and maintained at approximately 600 psi through the introduction of compressed air. Heat is then applied to the autoclave via electrical resistance heaters located outside the pressure vessel. When a temperature of 110°C in the water between the autoclave and canister is reached, heating is stopped. Since considerable heat is released from the canister due to radioactive decay as well as by hydration of the concrete, cooling of the water may be necessary to maintain the 110°C temperature. This may be accomplished through the use of water cooling coils welded to the surface of the pressure vessel. The canister is contained in the 110°C water bath for 24 h to allow the concrete to harden. During this time, the centerline temperature reaches a maximum temperature of 181°C. (see Section J.3). Boiling of the canister contents will occur only if the temperature inside the canister reaches 250°C, which is the equilibrium steam temperature at 600 psi. The 69°C difference between the calculated centerline temperature and the equilibrium steam temperature provides a margin for error in determining the heat content of a calcine batch and for variability in the conductivity of the concrete form. After the concrete has completely hardened, the pressure in the autoclave is slowly vented to a blowdown tank while the water between the canister and the autoclave is cooled to 50°C. The pressure reduction/cooling process may require 24 h to avoid the generation of excessive internal pressures in the concrete as free water in the concrete escapes. When the autoclave and cell pressures are equalized, the canister surface is decontaminated with a hot water spray. Water is removed from the autoclave by jetting it to a holding tank or HLLW concentrator. The decontaminated canister is transferred to an air storage facility, which is described in the following subsection. The entire autoclave cycle, from canister loading to canister unloading, requires 64 h.

Each autoclave is protected from overpressurization by rupture discs. The blowdown tanks are designed to accept an uncontrolled blowdown from several autoclaves simultaneously. Check valves protect other autoclaves from uncontrolled pressure surges. Gases from the blowdown tanks are vented to the process off-gas system.

Future research and development may be required to:

- produce a remote, mechanical autoclave lid system
- determine the thermal conductivity of concrete in full-scale castings and increase thermal conductivity, if warranted
- assess the variability in heat contents and rate of change of individual concrete batches
- assess the effectiveness of decontaminating canister surfaces in autoclaves
- determine an optimum pressure reduction rate in the autoclave
- optimize safety considerations in dealing with a pressurized system.

J.6.10 AIR STORAGE FACILITY

The air storage facility is used to enable free water to diffuse through and escape from the concrete and to permit the decay heat to decrease to levels that permit final disposal in a federal repository.

After the concrete-filled canister is removed from the autoclave, a filtered lid is placed on the canister and the canister is further decontaminated. The canister is then placed in a storage rack in the air storage facility for interim storage before being placed in a federal repository. Years of storage may be required to sufficiently dewater the concrete before a final, leak-tight lid is welded onto the canister. The decay heat will aid in the dewatering process. A total of 27 yr of storage may be required for canisters to cool from a heat content of 1.8 kW to 0.5 kW, the maximum heat content assumed in Appendix A for repository storage.

Further research and development may be required to:

- determine if filtered canisters can be safely and economically stored in an air storage facility
- determine time requirements for dewatering concrete.

J.6.11 OFF-GAS SYSTEM

The process off-gas system removes radioactive particulates and gases and noxious, nonradioactive gases from contributing unit operations. It discharges the cleaned streams to the fuel-reprocessing-plant air protection system. The equipment requirements are very similar to those described in Appendix A for the ICGM process. Equipment capacities and sizes may be increased somewhat due to the expected additional load caused by the pneumatic transfer system, the autoclave blowdown system and air inleakage through additional connections between the greater number of unit operations. Further research and development may be required to more precisely determine the off-gas system arrangement, volumes and rates and then size the equipment accordingly. The research and development efforts specified for the reference off-gas system described in Appendix A are also applicable to this system.

J.7 ALTERNATIVE PROCESSES AND EQUIPMENT CONSIDERED

Other means exist for producing calcine from HLLW. These include fluidized-bed calcination and rotary-kiln calcination. These processes are discussed in Appendix A, Section A.7.

Pneumatic transferring of calcine is an element of the concrete process. Pneumatic transferring could be eliminated altogether if the unit operations were located in an essentially vertical alignment to take advantage of gravity flows. However, stacking equipment in this manner may require a very tall and costly operating cell.

Volumetric batching of calcine is also a key element of this concrete process design. Variations in the density of the calcine will result in variations in batch weight and heat content, which may limit the amount of calcine permitted in a volume-based batch. This could lead to an increase in the number of concrete packages produced. An alternative to volumetric batching is gravimetric batching based on load cell weight measurements. However, since the batching equipment must be physically connected to other equipment to contain dust, the batching equipment may be constrained by the connections between equipment which could result in faulty weight measurements.

The autoclave step could be eliminated if the concrete could be kept from boiling during the setting process. If canisters were made of a small-diameter pipe, for example 0.1 m in ID, the heat transfer distance would be significantly decreased and boiling might not occur. Further gains might be made if the newly cast concrete canister were placed in a cool water bath. The smaller canisters, having a smaller radius for diffusion of free water and a lower heat content, might also require only a short period of preliminary storage before interim storage or final disposal. However, smaller-diameter canisters will increase the number of canisters and canister handling operations required. A nine-fold increase would be required for canisters with a 0.1 m ID; the number of calciners, batchers and mixers might also have to be increased to keep up with the higher batch demand. The elimination of the autoclave step might also have a negative effect on the quality of the concrete waste form.

Storage in air was selected for the temporary storage of newly produced concrete canisters. Storage in water could also be used, but the higher waste form temperature achieved in air storage may accelerate the rate at which free water is removed from the canister. This would shorten the time required between concrete casting and canister sealing. This time could be further shortened for small-diameter canisters if the canisters were placed in a furnace heated to approximately 100°C. Larger canisters of the size used in this study may not be amenable to fast removal of free water since concrete centerline temperatures may exceed 200°C for storage in air at 50°C. Exposure to too high a temperature will result in driving off some water of hydration, which is responsible for many of the important properties of the concrete waste form.

A major process alternative is the elimination of the spray calciner and the direct mixing of HLLW with concrete-forming additives. To retain the same water-to-waste oxide-to-additives ratio and the same canister size as used in this design study, the number of canister

requirements is increased by about five fold. As previously discussed, this would decrease preliminary storage requirements but would increase canister handling requirements. A modification of this approach is to mix the concrete in a canister that contains a built-in mixing apparatus. This approach eliminates the mixer but increases the cost per canister. The quality of mixing may be reduced and may be difficult to assess remotely. The direct use of HLLW in concrete may also have a deleterious effect on concrete quality since concrete is basically incompatible with acid; although much of the two-molar acid provided by the HLLW may be destroyed by radiolysis. This may result in increasing the porosity and solubility of the concrete.

J.8 EQUIPMENT DATA SHEETS AND DRAWINGS

The reader should refer to Section A.8 in Appendix A for a glossary of the symbols and abbreviations used in the figures in this section.

FIGURE J-3. Canister

TABLE J-5. Equipment Data Sheet for Canister

Function

To contain the concrete waste form during casting and to act as a barrier during disposal.

Process Interfaces

Concrete mixer discharge flange.

Design Parameters

Volume = 212 L; dimensions = 0.30 m in OD by 3 m in length; material--304-L stainless steel schedule-40 pipe; wall thickness = 0.01 m; meets the intent of the ASME boiler and Pressure Vessel Code Section III, Division I; must withstand 600 psi after sealing; remote operations, monitoring, maintenance, removal and replacement.

Operating Conditions

Nominal capacity = 171 L; maximum centerline temperature = 250°C; maximum surface pressure = 600 psi.

Features

- Porous stainless steel liner and filter cover facilitate dewatering during storage.
- The filter cover allows gases to escape, but helps retain particulate matter in the canister.

Services or Auxiliaries

None

Space Requirements

0.30 m OD by 3 m in length

Maintenance Methods

Remote

Remarks

FIGURE J-4. Calcine Receiver

TABLE J-6. Equipment Data Sheet for Calcine Receiver

Function

Collection of calcine.

Process Interfaces

Spray calciner; pneumatic transfer system.

Design Parameters

Maximum batch weight = 66.2 kg; vibrated calcine density = 1.2 g/cm³; calcine heat density = 44.8 W/L; ball valves at entrance and exit for batch containment and isolation; remote operation, monitoring, maintenance, removal and replacement.

Operating Conditions

Nominal batch weight = 51 kg; nominal batch accumulation rate = 3 h; nominal calcine entrance temperature = ~350°C; maximum calcine receiver centerline temperature = 550°C.

Features

Heat transfer fins; temperature sensors; pressure sensors; electric-activated ball valves; vibrator; chemical addition ring; gamma-emission level detection device (external); vibrator (external).

Services or Auxiliaries

Instrument air (pressure sensor); compressed air (vibrator); electrical (temperature and gamma sensors); chemical addition.

Space Requirements

0.32 m (12.75 in.) OD by 0.91 m (36 in.) high

Maintenance Methods

Remote

Remarks

FIGURE J-5. Pneumatic Transfer System

TABLE J-7. Equipment Data Sheet for Pneumatic Transfer System

Function

Transfer calcine from calcine receiver to the volumetric batcher.

Process Interfaces

Calciner receiver; volumetric batchers.

Design Parameters

66.2 kg/batch calcine; some particles up to 1 cm in dia; low spots in transfer lines to be avoided to minimize plugging; line size = 0.05 m in ID; line velocity = 9 m/s; solids-to-gas ratio ~2; remote operation, monitoring, maintenance, removal and replacement.

Operating Conditions

Nominal batch weight = 51 kg; nominal batch transfer cycle = 3 h; maximum inlet vacuum = 0.38 m Hg; system pressure drop = 2.27 kg/m.

Features

HEPA filter; vacuum jet (dry air-inlet); pressure sensors; chemical addition flush system.
Pipe: stainless steel; 0.035 m in ID; mill run 2B interior finish; 0.61-m sections bend radius.

Connectors: Cherry Burrell's I line or similar type; remote flange joints if required for rigidity; 3-way valve system (to two volumetric batchers); backflow preventers.

Services or Auxiliaries

Compressed air (plug blowback); steam (jet vacuum); instrument air (pressure sensors); chemical addition (line cleanout).

Space Requirements

0.03 m OD by 30 ft in length (minimum)

Maintenance Methods

Remote

Remarks

FIGURE J-6. Volumetric Batcher

TABLE J-8. Equipment Data Sheet for the Volumetric Batcher

Function

To collect calcine from pneumatic transfer system, verify volume and heat content of calcine; discharge calcine to receiving hopper.

Process Interfaces

Pneumatic transfer system; filter assembly and receiving hopper.

Design Parameters

Maximum batch weight = 66.2 kg; calcine density = 1.2 g/cm³; internal fins or pins for cooling; cyclone separation (upper section); remote operation, monitoring, maintenance, removal and replacement.

Operating Conditions

Nominal batch weight = 51 kg; calcine density ~1.2 g/cm³; centerline temperature ≤500°C; batch/3 h.

Features

Temperature sensors; pressure sensor; chemical addition capability; vibrator calcine level detection capability.

Services or Auxiliaries

Instrument air (pressure sensor); compressed air (vibrator); chemical addition (clean out); electrical (level detector, temperature sensors).

Space Requirements

0.03 m OD by 1.32 m high

Maintenance Methods

Remote

Remarks

FIGURE J-7. Filter

TABLE J-9. Equipment Data Sheet for the Filter Assembly

Function

To collect particulate entrained in air over the volumetric batcher and the receiving hopper.

Process Interfaces

Volumetric batchers; receiving hoppers.

Design Parameters

Maximum vacuum = 0.5 m Hg; operates at 0.26 m air/min; filter surface area $\sim 1 \text{ m}^2$; 13 to 14 filter candles 0.5 m in length with 0.075 m^2 of surface area per filter assembly.

Operating Conditions

Filter blowback is 0.25 s in duration at 60 psig; normal operating vacuum = 2 to 5 cm H₂O; calcine transfer vacuum = 0.38 m Hg.

Features

Blowback system; pressure sensor; temperature sensor; chemical addition capability; flow and vacuum control capability.

Services or Auxiliaries

Compressed air (blowback); instrument air (pressure sensors); chemical addition (clean out); electrical (temperature sensors).

Space Requirements

0.36 m in OD by 1.22 m high

Maintenance Methods

Remote

Remarks

TABLE J-10. Equipment Data Sheet for Receiving Hopper

Function

Collect all concrete-forming materials and air-blends prior to discharging to mixer.

Process Interfaces

Volumetric batcher; weight batcher; mixer.

Design Parameters

Holding capacity = 0.6 m³; contains 363 kg of materials; air blender; remote operations, monitoring, maintenance, removal and replacement.

Operating Conditions

Operating capacity ~0.2 m³; blender operation ~1 min.

Features

Air blender; temperature sensor; pressure sensor; air locks; discharge valve; vibrator.

Services or Auxiliaries

Compressed air (vibrator); instrument air (pressure sensor, blender); electric (valves, air locks and temperature sensors).

Space Requirements

0.76 m in OD by 1.22 m high

Maintenance Methods

Remote

Remarks

FIGURE J-8. Receiving Hopper

FIGURE J-9. Concrete Mixer

TABLE J-11. Equipment Data Sheet for the Concrete Mixer

Function

Mixes the concrete-forming materials prior to discharge into the waste canister.

Process Interfaces

Receiving hopper; canister.

Design Parameters

Capacity 0.3 m²; maximum mixing speed = 300 rpm; maximum concrete temperature = 50°C; torque indication of mixing motor; access doors for inspection and cleaning; remote operations, monitoring, maintenance, removal and replacement.

Operating Conditions

Batch volume ~0.2 m²; mixing speed is variable; concrete temperature <50°C; mixing time ~5 min.

Features

Mixing motor torque indication (amperage); temperature sensors; pressure sensor; cooling jacket; remote-controlled discharge valve; water and chemical addition; variable mixing speed.

Services or Auxiliaries

Cooling water; instrument air (pressure sensor); electrical (motor, temperature sensors, valve); chemical addition (concrete water and cleaning).

Space Requirements

0.91 m in dia by 2.1 m long by 1.52 m high

Maintenance Methods

Remote

Remarks

FIGURE J-10. Canister Holder

TABLE J-12. Equipment Data Sheet for Canister Holder

Function

Contains the canister for filling, vibrates canister and provides a sump for collection of material inadvertently spilled from the mixer.

Process Interfaces

Canister; concrete mixer.

Design Parameters

Contain canister 0.3 m in dia by 3.0 m high; canister connection capability; sump capacity = 0.2 m^3 ; sump flush capability; vibrators for bubble removal; remote operations, monitoring, maintenance, removal and replacement.

Features

Vibrator(s); sump; chemical addition; flush to sump; transfer line; and liquid level sensors.

Services or Auxiliaries

Compressed air (vibrators); chemical addition (cleaning); steam (jet); instrument air (liquid level sensors).

Space Requirements

0.91 m by 2.13 m by 2.59 m high

Maintenance Methods

Remote

Remarks

FIGURE J-11. Autoclave

TABLE J-13. Equipment Data Sheet for Autoclave

Function

Accelerates the setting and curing of concrete through elevated temperatures and pressures.

Process Interfaces

Canister; off-gas system.

Design Parameters

Contain canister 0.3 m in OD by 3.0 m high; pressure = 15 to 1000 psi; refreshed or static pressure modes; vessel temperature = 50° to 375°C; heating and cooling capability = 50° to 110°C; canister decontamination capability; pressure safety system; remote operation, monitoring, maintenance, removal and replacement.

Operating Conditions

Pressure = 15 to 600 psi; atmospheric temperature = 50° to 250°C; water temperature = 50° to 110°C.

Features

Cooling coils; pressure sensors; temperature sensors; transfer line pressure delivery system; off-gas system; pressure relief disc and metering valves; heating system; remote closure of autoclave.

Services or Auxiliaries

Water (cooling, heat transfer and decontamination); compressed air (autoclave pressure); steam (decontamination); electrical (heating, temperature sensors).

Space Requirements

0.7 m in dia by 3.1 m high

Maintenance Methods

Remote

Remarks

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APPENDIX K

CERMET PROCESS

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APPENDIX K CERMET PROCESS

K.1 INTRODUCTION

A cermet waste form, which is a composite material that has ceramic particles dispersed in a leach-resistant metallic phase, may be a viable alternative to the glass monoliths produced by the reference process (Appendix A). The production of cermet is amenable to quality assurance, and some of the attributes of the cermet waste form, such as thermal conductivity and stability, appear to be better than those of monolithic glass.

The cermet process facility will be capable of calcining HLLW, forming extrusions of the calcine, sintering and reducing these to a metallic state, enclosing the cermet monolith in a canister and processing and handling the canisters. The development of cermet forms containing high-level nuclear waste has progressed to the level of producing cermets containing waste from Nuclear Fuel Services and SRL on a laboratory scale in hot cells. However, full-scale processing of cermet waste forms is at a very conceptual stage. Many aspects of the waste form and the process need to be identified.

K.2 PROCESS DESCRIPTION

The preconceptual process flow diagram for the production of a cermet waste form is depicted in Figure K-1. High-level liquid waste is received at the waste facility from the fuel reprocessing plant and is directed to one of three dissolver tanks. When a tank is filled to a specified level, the contents are agitated to assure homogeneity and then are sampled for characterization of the HLLW. The HLLW in the tank is not processed further until the sample analyses are completed.

Following characterization of the HLLW, urea and metal- and cermet-forming additives, in the form of solids or aqueous solutions, are measured by weight outside the processing cell and are added to the dissolver tank. The tank is then heated to raise the temperature of the contents to 60°C to complete the solution of HLLW elements and additives.

While the HLLW in one tank is being prepared, as described above, HLLW from the fuel reprocessing plant is delivered to another tank and an already-prepared solution is withdrawn from the third tank to feed a heated-wall spray calciner. Each dissolver tank has the capacity to process enough solution to feed the spray calciner for 24 h.

The HLLW and additives are pumped from the dissolver tank to the spray calciner. The solution enters the calciner through a pneumatic atomizing nozzle at a nominal rate of 406 L/h. The elevated temperature in the calciner chamber causes the urea to react with the nitrates, which produces a calcine composed of waste elements and additives in the form of oxides and other ceramic compounds.

The gaseous effluents are exhausted from the calciner and treated as described under the reference off-gas system (Appendix A, Section A.6.5). The calcine flows from the calciner through a diverter into one of two batch mixers, where it is cooled, mixed with a liquid binder that is sprayed into the mixer and consolidated. Again, while the contents of one mixer are being processed, the already-processed contents of the second mixer are being discharged into a piston extrusion press.

The press extrudes the calcine and liquid binder mixture through a die to form a right circular cylinder approximately 0.27 m in dia. The extruded cylinder is then cut into 1-m-long sections. The extrusion press has a vacuum de-airing system that exhausts back into the off-gas system. The cylinder sections are inspected and the rejects are put through a crusher. Scrap from the extrusion and crushing operations is recycled to the batch mixer by a mechanical conveyor.

The extruded cylinder sections are passed through an air lock into the retort of a continuous sintering furnace. As the cylinders pass through the furnace 1) the liquid binder is driven off, 2) an Ar + 4% H₂ atmosphere reduces the appropriate oxides to a metallic state, and 3) the metals alloy and sinter to form a metal matrix that encapsulates ceramic crystals of waste. The cermet cylinders are partially cooled in the furnace and are then removed through another air lock. After a cermet cylinder has completely cooled, it is inspected and may be sampled. Then it is loaded into a canister which is sealed and processed as described in the reference process.

FIGURE K-1. Cermet Process Flow Diagram

K.3 BASES AND ASSUMPTIONS

Many of the bases and assumptions for the cermet process are given in the Overall Process Bases Section. Bases and assumptions specific to the cermet process are noted in this section.

K.3.1 PROCESS BASES

Data on the HLLW and its waste forms are presented in the Overall Process Bases Section and in Appendix A. The cermet process is based on a nominal operating capacity of 2000-MTHM waste equivalents per 300-d yr. Design capacities used are 1.5 times the nominal operating values. Additional data on the cermet process are listed below:

- Cermet waste form

cermet composition = 70 wt% alloy and 30 wt% ceramics

alloy composition = 70 wt% Fe, 20 wt% Ni, 5 wt% Cu, and 5 wt% other metals

ceramics composition = pollucite ($2\text{Cs}_2\text{O} \cdot 2\text{Al}_2\text{O}_3 \cdot 9\text{SiO}_2 \cdot \text{H}_2\text{O}$)

sodium aluminium orthosilicate ($\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$);

all other elements present primarily as oxides

yield = 244 g cermet/L HLLW

mass equivalent to 3.06 kW/can = 216.2 kg/can

monolith volume = 32.1 L/can

monolith dimensions = 0.2 m in dia by 1 m long

production rate = 6.45 cans/d design; 4.28 cans/d operating

specific heat = 0.109 BTU/lb - $^{\circ}\text{F}$

- Calcine (assume only oxides and no metals present)

Oxidation states of alloy formers are Fe_2O_3 , NiO , MoO_3 , RuO_2 , PdO , TeO_2 and CuO .

- Liquid feed solution (assumes volumes of HLLW, aqueous solutions of additives and molten urea are additive)

specific gravity of HLLW in urea = 1.01 kg/L

heat density of HLLW in urea solution = 1.51 W/L (assumes there is no reaction or gas released when urea is added to HLLW at 60°C)

- Additives

bulk density of urea pellets = 0.70 g/cm³

bulk density of H_2SiO_5 = 0.9 g/cm³

solubility of ferric nitrate in water = 1.59 g/ml

assumed 150% additions of Al and Si sources

- Sintering

furnace efficiency = 50%

reducing gas composition = 4% H_2 and 96% Ar

If all H_2 reacts to form H_2O , 12.3 g H/L HLLW are required to reduce oxides to desired metals.

assumed 50% efficiency for all water cooling applications.

Additional data on the cermet process materials and waste form are given in Tables K-1 and K-2.

K.3.2 CANISTER

The specified canister heat loading of 3.06 kW is the limiting factor in determining the size of the cermet waste form and canister. Given the specified heat loading, approximately 216 kg of the cermet waste can be placed in a canister. Oak Ridge National Laboratory personnel have reported cermet densities ranging between 6.2 g/cm^3 and 8.2 g/cm^3 . A density of 6.5 g/cm^3 was assumed for this study; thus, a canister volume of 33.3 L is required. A cermet cylinder, or monolith, that is 0.2 m (~ 8 in.) in dia by 1 m (~ 39 in.) long was selected. This size and shape is also easily handled.

It is assumed that the canister will fit closely around the cermet column. It is also assumed that the canister need not be heavy-walled construction because the cermet waste form has high mechanical strength.

K.3.3 SECONDARY RADIOACTIVE AND NONRADIOACTIVE WASTES

It is assumed that the effluents from the cermet process are greater and differ from those of the reference process only in that ammonia and CO_2 are present. Treatment of the ammonia in the off-gas system should require no new technology. Studies at ORNL have shown that ammonium nitrate is not formed in the effluents from the cermet process.

Because of the reducing environment in the calciner (produced by the urea) and in the sintering furnace, it is believed that there will actually be less volatile radioactive materials, such as ruthenium oxide, in the cermet process effluents than in those from the reference process (Aaron et al. 1979b).

Except for additional worn equipment (due to the fact that the cermet process is more complex than the reference process) that will eventually have to be discarded, it is assumed that the waste from the cermet process is of the same magnitude and composition as the waste from the reference process. It is also assumed that there are facilities in the fuel reprocessing plant to treat, consolidate and package both solid and liquid wastes. Finally, the assumption is made that all process scrap that may contain high-level waste is recycled back in the cermet process.

TABLE K-1. Cermet Process Constituents

Major Constituents	Solutions		Major Constituents	Gases					Major Constituents	Solids		
	HLLW, g/L	Molten Urea, g/L		Dissolver, Relative	Atomizing, scfm	Calciner, g-mole/d	Forming, kg/h	Firing, g-mole/d		Additives, g/L HLLW	Calcine, g/L HLLW	Cermet, g/L HLLW
H+	2.0	0.87	AR		95			42,210	Urea	192.8	0	0
Na	0.15	0.06	H ₂ O	Trace			1.8	30,900	Fe	113.7	115.7	115.7
Fe	2.0	50.52	HNO ₃	Trace		None			Ni	32.9	33.0	33.0
Ni	0.13	14.41	H ₂			Trace		11,640	Cu	8.3	8.3	8.3
Cr	0.33	0.14	NH ₃	Trace		122,755			Al	0.4	0.4	0.4
Gd	15.90	6.94	N ₂			Major			Si	0.6	0.6	0.6
P	0.87	0.38	CO									
NO ₃	199.0	290.83	CO ₂	Trace		61,377						
U	8.43	3.68	O ₂									
Pu	0.11	0.05	N ₂ O									
Other AM ^(a)	1.96	0.86	NO ₂									
AP ^(b)	0.5	0.22	NO									
FPM ^(c)	44.7	19.52	Total	Insignificant	287.8 kg/L	478.6 kg/h	1.8 kg/h	94.4 kg/L				
Urea	-	645.2										
Cu	-	3.62										
Al	-	0.17										
Si	-	0.26										

(a) Actinide metals.

(b) Activation products.

(c) Fission product metals.

TABLE K-2. Properties of Process Materials and Cermet Waste Form

Properties	Process Materials			Cermet Waste Form
	HLLW	Molten Urea	Calcine	
Specific gravity, kg/L	1.17	1.01	0.8	6.5
Temperature, °C	40	60	60 to 300 ^(a)	1050 to 1060
Heat loading, W/L	3.46	1.51	8.9	95.3
L/MTHM	567	1298.4	219.3	20.6
Nominal operating rate, L/h	158	362.9	61.1 (48.9 kg/h)	5.7 (38.6 kg/h)
Design rate, L/h	238	546.7	92.0 (73.6 kg/h)	8.6 (58.2 kg/h)
g oxide/L	92.6	136.1	309.4	--
Density, g/cm ²	--	--	0.8	6.5

(a) Temperature out of calciner = 300°C; temperature in mixer = 60°C.

K.3.4 UTILITIES AND MATERIALS

The estimated utility services and essential materials to sustain the cermet process at the design operating rate are presented in Tables K-3 and K-4, respectively.

TABLE K-3. Estimate of Utilities Needed for Cermet Process

Utility	Required Capacity	Average Use Rate	Disposition
Electricity	2000 kW	1040 kW	
Raw water	3000 L/min	1350 L/min	Recycle or discharge
Process water	400 L/min	10 L/min	Recycle as HLLW or ILLW
Sanitary water	40 L/min	20 L/min	Sanitary or chemical sewer
Air	700 scfm	500 scfm	As appropriate
Steam	26 kg/min	6 kg/min	As appropriate

TABLE K-4. Estimate of Materials Needed for Cermet Process

Material	Amount Needed Per Year, kg	
	Design	Operating
$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	9,400	6,300
$\text{Cu}(\text{NO}_3)_2 \cdot 2\text{H}_2\text{O}$	53,800	35,700
$\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	1,409,500	935,700
$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	279,300	185,400
H_2SiO_5	5,100	3,400
Urea ($\text{H}_2\text{NCO NH}_2$)	1,105,600	734,000
Argon	3.8×10^6	2.5×10^6
Hydrogen	21,100	14,000
Detergent	1000	1000
Canisters, each	1,920	1,290
Helium sources, each	1,920	1,290

K.4 FACILITY AND EQUIPMENT REQUIREMENTS

The cermet process is contained in a reinforced concrete structure similar to that shown in Figure D-3 of the MIL process (Appendix D). The overall requirements for the waste facility and its support groups and services are covered in the Overall Design Bases Section. The capability for remote operations and maintenance of all in-cell equipment is the major factor in subsequent process evaluations and research.

The major equipment and systems for the cermet process and the space requirements of each are given in Table K-5.

TABLE K-5. Process Cell Volume Requirements for Major Processing Equipment

<u>Equipment</u>	<u>Number Required</u>	<u>Dimensions (Length x Width x Height), m</u>
Additives' storage and batching system	1	5 x 4 x 7
Dissolver and feed system	3	2.5 x 2.5 x 6
Spray calciner	1	3.5 x 3.5 x 8
Calcine agglomerator	2	4.5 x 0.5 x 0.5
Extruder	1	3 x 0.5 x 0.5
Reject crusher and return elevator	1	1 x 1 x 10
Sintering system	1	8 x 0.5 x 1
Canning system	1	6 x 4 x 3
Canister handling system	1	(same as reference process)
Off-Gas system	1	(twice that of reference process)

K.5 STATE OF PROCESS DEVELOPMENT

Work in developing and evaluating cermet as an alternative high-level waste form was initiated in mid 1977 at ORNL. The cermet process is based on technology developed at ORNL for the preparation of neutron dosimeters and various other special isotopic materials. High-level liquid waste from both SRL and Nuclear Fuel Services is being used in the ORNL development program (Aaron et al. 1979a). The work is performed on a laboratory bench scale using small batches of both simulated waste and actual waste samples.

Significant improvements have been made in simplifying the cermet process. One recent accomplishment was the successful demonstration of a heated-wall spray calciner for continuously decomposing the urea and for calcining the precipitate. To date, samples of aluminum-urea solutions and simulated Thorex acid waste-urea solutions have been calcined in a 10-cm-dia calciner. The off gas from this demonstration has been analyzed, but the processing characteristics of the calcine have not been determined. Use of the calciner resulted not only in continuous processing, but in a major reduction in the quantity of urea required for the process. As little as a 1:1 urea-to-nitrate ion ratio was achieved using the calciner (Aaron et al. 1979b).

A variety of techniques for densifying the calcine are being investigated. Hot pressing, isostatic pressing and piston extrusion have been performed successfully. Of these methods extrusion, using water as a binder, followed by sintering in a reducing atmosphere is considered to be the most promising densification method. However, liquid-phase sintering is also being studied in an effort to reduce the temperature and sintering time necessary to achieve satisfactory densification. Typical sintering conditions are 30 min at 1050°C for 1/2-in.-dia samples.

A different approach to cermet densification has also been investigated, and the results from early tests are promising. It was found that cold-hydrostatic pressing of calcine that was produced in a reducing atmosphere resulted in very dense, strong monoliths. The ductility of the metals present in the reduced calcine permits cold welding of the particles at room temperature by isostatic pressing.

Personnel at ORNL are attempting to investigate all methods of densification. Those methods that appear most promising are being tested in the laboratory so that the quality of the cermets produced by each may be evaluated. In addition, the effluents from each method, the potential for engineering development and the reliability of each method will be evaluated.

To determine the volatility of Sr, Tc, Ru and Cs during cermet processing, laboratory-scale, batch experiments have been performed using radiotracers in simulated waste. Volatility loss measurements made during the processing of real HLLW support the results obtained with the radiotracer experiments. However, the losses due to the volatility of these elements and their oxides during processing in the spray calciner have not been determined.

In 1978, small cermet samples were produced in hot-cell facilities at ORNL using actual high-level waste from three different sources: 1) Nuclear Fuel Services' acid Thorex waste,

2) Savannah River Plant untreated acid waste, and 3) Savannah River Plant dried sludge (Aaron et al. 1979b). These samples are being evaluated using optical and scanning-electron-microscopy metallography, thermal conductivity, calorimetry and dimensional measurements. Limited leach testing of these samples is planned for the future.

Leaching tests have been performed on the cermetes that were prepared using simulated wastes (Aaron et al. 1979a). However, interpreting the results of the leach tests is complicated due to corrosion of the samples caused by galvanic coupling between the cermet samples and the metal testing equipment. Thus, to obtain quantitative leach rate data for cermetes leach-testing methods are being prepared that will not result in galvanic coupling. The fact that galvanic coupling occurred does indicate a characteristic of cermetes that must be considered when specifying the material for the cermet canister. The corrosion caused by the galvanic coupling is being studied with the aid of scanning electron microscopy.

The cermet process is in its very preliminary stages of development, and much remains to be done in the development of the cermet waste form and on the process for producing it. Some areas where little is yet known include continuous processing, alloy composition optimization and calcine treatment. Making the transition from bench-scale batch operations to pilot-scale continuous processing with remote equipment is only now beginning to be explored on paper. No work has been done on recycling of scrap and rejected materials or canning the cermet pieces. The sintering cycle must also be studied to determine how to best form the desired oxides, aluminosilicates and titanates with the radioactive elements while simultaneously forming the metal matrix. However, the high thermal conductivity, good corrosion resistance and high mechanical strength of cermet waste forms justifies further development.

K.6 UNIT OPERATIONS

The cermet process can be divided into nine unit operations. These operations are discussed in detail in the following subsections. Drawings and data sheets for the equipment and systems discussed in this section can be found at the back of this appendix in Section K.8.

K.6.1 ADDITIVES' STORAGE AND BATCHING

The functions of the additives' storage and batching operations are to 1) store the additives in an accessible manner and 2) measure the precise amounts that must be added to the HLLW to produce the desired cermet composition. To assure that the proper amount of each additive is combined with a batch of HLLW, each batch of waste added to a dissolver and feed tank is characterized for quantity, heat content and composition. The appropriate quantities of each additive are then determined for that batch.

The iron, nickel and copper for alloy formation and aluminum for pollucite formation are added to the HLLW as water-soluble nitrates. Because the nitrates are deliquescent and do not flow freely, they are transferred in the form of aqueous solutions. Tanks having the volumes specified in Table K-6 (see Section K.8) will contain sufficient saturated solutions for eight days of operation at the design rate. These tanks are equipped with heating coils and agitators to help dissolve the nitrates in the water.

Very accurate, completely automated liquid batching systems are provided to accumulate the desired quantities of nitrate solutions in the batch tank. The batch tank has a capacity of 6500 L so that enough additives for a 24-h cycle may be batched for addition to a dissolver tank. When the desired quantities of the additives are present in the batch tank, the solution is pumped into the waste solidification cell or canyon through a one-way valve and is diverted to the appropriate dissolver tank. A redundant measurement of solution quantity may be required to guarantee proper proportions of cermet ingredients.

Urea is commonly supplied in pelletized form and has excellent flow properties so that it can be stored, transported, and measured out as a dry solid. A bin having a capacity of 1300 ft³ is sufficient to contain enough urea for one week of operation. A hopper train car has double this capacity; therefore, a storage bin sized to accommodate bulk urea shipments by rail will be used. Because the pelletized urea has excellent flow characteristics, gravity flow is used to transport it from the bin to the batch weight hopper. The valve controlling the flow of urea from the storage bin to the batch hopper is governed by feedback from the batch weighing system.

Oak Ridge National Laboratory personnel recommend the addition of silica as silicic acid for pollucite formation. Assuming the bulk density of dry silicic acid is 0.9 g/cm³, a bin of at least 6 ft³ capacity is required to contain enough material for one week of operation. Transfer and flow control of this material is the same as for the urea.

The solid additives are measured out by accumulating them in the hopper of a weighing system. The hopper must have a minimum capacity of 190 ft³ to accommodate enough solid

additives for a 24-h cycle in the dissolver tanks. A batch weighing system that has a precision of 0.5% and is programmable and completely automatic is available without additional development. A redundant weighing system may be required to guarantee that solids are added in their desired proportions.

When the desired amounts of solid additives are collected in the weight hopper, they are transferred by gravity flow into the processing cell or canyon through an air lock. A diverter then directs the additives to the appropriate dissolver tanks containing the HLLW. At PNL experience with GEMCO Dust-Tite spherical solids valves in air locks has been good, and they should work for a diverter as well.

All of the equipment, except the diverters, required for measuring out the additives are located in a nonradioactive area where they can be easily maintained. The solid and liquid batching systems, valves, air locks and pump are all standard items that should not require any additional development. However, should the stockpiles of contaminated metals held by DOE be used as the source of additives as personnel at ORNL have suggested (Aaron et al. 1979a), then the additives operation will be greatly complicated and process and remote equipment development will be required.

K.6.2 DISSOLVER AND HLLW FEED SYSTEM

The function of the dissolver and feed tank operation is to dissolve the waste elements and the cermet-forming additives in a hot urea nitric acid solution. This solution is then delivered under pressure to the calciner. The advantages of a urea solution over the acid solution are that a more complete and homogeneous blend of the waste materials and additives is possible. The urea is also a basic, reducing agent that destroys the acid and minimizes the formation of nitrates and volatile ruthenium oxides during the calcining process. The urea decomposition products produced during calcination are simple compounds containing no oxidants.

Three HLLW feed tanks are at the head end of the in-cell, HLLW processing equipment. Each tank has a volume of 18,000 L, which permits it to contain enough solution of HLLW, urea and other additives to feed the calciner at the design level for 24 h. The tank volume is also sufficient to accommodate the foaming that may occur if the solution is overheated. Figure K-2 shows the cycle the three tanks will follow. At any time during the processing of HLLW, one tank is receiving waste solution from the fuel reprocessing plant while the addition of urea and cermet-forming additives to the HLLW is taking place in another tank; at the same time, prepared solution is being withdrawn from the third tank to feed the spray calciner operation.

The feed tanks are made of 304L stainless steel. They must be heated to raise the temperature of the mixture of HLLW, urea and additives to 60°C and maintain this temperature so that the urea will go into solution. Cooling must also be available to assure that the contents of the tank never exceed 180°C to prevent the urea from reacting with the water. This heating and cooling is accomplished using stainless steel coils that are suspended from the tops of the tanks. Either superheated steam or cooling water is circulated through the coils. A propeller agitator is located in each tank to aid in heat transfer by convection, to

FIGURE K-2. Cycling of Dissolver and Feed Tanks

homogenize the solution of HLLW, urea and additives, and to suspend the insolubles, such as sand, zirconia and thoria. During the heating of the contents in the tank to 60°C, some water vapor and nitric acid are released. Therefore, the tanks are vented through a demister to the off-gas system.

When preparation of the molten urea solution is complete, it is transferred to the spray calciner by a feed recirculation system, as described under the reference process (Appendix A). A feed pressure of 25 psi is required at the nozzle of the calciner. The feed line from the tank to the nozzle and the return line are heated to 60°C to keep the urea in solution.

Further research is necessary to fully determine the kinetics of the urea and acidic HLLW reaction, so that scaling this operation up may be done more precisely. There is very little experience with pumping the solution of HLLW in hot urea; therefore, a pilot-scale demonstration of the operation is needed. Additional data on the dissolver and feed system components are given in Table K-7 (see Section K.8).

K.6.3 SPRAY CALCINER

The function, design and operation of the heated-wall spray calciner for the dissociation of the urea and the precipitation of the waste and additives are basically the same as for the reference process spray calciners. However, this process requires one large spray calciner, and argon or nitrogen rather than air is used as the atomizing fluid. The dissociation of the urea in the spray calciner is an exothermic reaction that produces some of the heat necessary to accomplish the calcination. In addition, no further solids additions are necessary during the calcination operation such as the frit in the reference process.

As shown in Figure K-3 in Section K.8, the solution of HLLW and additives in urea is injected into the heated chamber of the spray calciner through an atomizing spray nozzle. Argon is used as the atomizing gas. As the solution enters the heated chamber, it is heated to above 180°C and the urea decomposes to N_2 , NH_3 , H_2O and CO_2 . Traces of CO , NO_2 , N_2O and NO are also produced as the nitrates are converted to oxides. The waste and additives are precipitated as oxides, although it is possible that some of the heavy metal elements may be reduced to the metallic state. The gases and vapors are exhausted through sintered-metal filters to the off-gas system. The size of the filter system will be considerably greater than that of the reference process spray calciner because of the need to pass gases produced in the dissociation of urea. The solids are collected by the funnel at the bottom of the calciner and are diverted to one of two mixers.

Control of the calciner operation is accomplished by:

- proper ratioing of the solution and atomizing gas flow rates and pressures
- maintaining the desired temperature in the spray chamber
- maintaining the necessary vacuum in the chamber.

These operations are all accomplished in a manner similar to that of the reference process.

Spray calcination of the waste and molten urea solution has only recently been demonstrated on a laboratory scale at ORNL (Aaron et al. 1979b). An ultrasonic spray nozzle is being used in the demonstration unit. At PNL an internal-mix, atomizing spray nozzle has proven most satisfactory for calcining HLLW. Further research and development to identify the best nozzle for the cermet process will be required as the process is scaled up.

Another area requiring additional study is the effect that the exothermic reaction, which occurs as the solution is heated above 180°C, will have on the control of the process and on the size of the chamber. In addition, the appropriate atomizing gas and calcining temperature for obtaining the desired oxidation states of the solids and minimizing the volatilization of waste materials must be determined. Data on the spray calciner are given in Table K-8 in Section K.8.

K.6.4 CALCINE AGGLOMERATOR

The function of this unit operation is to mix a lubricant and binder (water) with the calcine and to form it into small pellets of the desired size, density and moisture content. However, before the binder (water) will adhere to the calcine, the calcine must be cooled enough so that the water will not vaporize. The calcine is agglomerated during this process to provide a free-flowing material suitable for charging into the extruder.

Personnel at ORNL have determined that a 10 ± 1 wt% addition of water to the calcine results in a material that can be extruded. Experience at PNL has shown that the temperature of the calcine exiting the calciner is a nominal 300°C. Therefore, it is cooled so that the water will not boil away when mixed with the calcine.

The operation is accomplished in two large batch mixers that are similar to a pugmill or pin mixer. The average design capacity is 74 kg/h, or 92 L/h, assuming a bulk density of

0.8 kg/L for the calcine. The batch size for this operation is 5% to 10% more than that required to produce one cermet log. One cermet log requires 270 kg of calcine and 5.5 h of calciner operation to produce enough calcine for a batch.

As calcine flows from the calciner, it is directed into one end of an agglomerator. Water is sprayed into the calcine to cool it and to act as a binder for the agglomeration that occurs in the agglomerator and for the extrusion operation that follows. Since vapors flow from the agglomerator to the calciner which is connected to the process off-gas system, water may cause caking of calcine in the connecting piece between the agglomerator and the calciner. This section is heated as necessary to prevent water condensation. To aid in cooling the calcine and in preventing the loss of too much water as steam, the agglomerator shell is enclosed in a water-cooled jacket. The agglomerator is also cooled to prevent decay heat from raising the temperature of a calcine batch during any loss of mixing. The agglomerator converts the calcine to spherical pellets that range in size from 8 mesh to 200 mesh and have a bulk density of 5 to 6 times that of the calcine. Mixing torque, or amperage, may indicate binder requirements and, hence, pellet quality. While the calcine is being charged into one mixer for cooling and agglomeration, the contents of the other mixer are discharged into the extruder.

The agglomerator is a high maintenance item because it has moving parts in contact with very fine abrasive material and because lubrication and rotating seals are required. Development and testing will be necessary to identify an agglomerator design and construction materials that will minimize maintenance requirements and maximize remote servicing efficiency. In addition, methods of monitoring the conditions in the mixer by remote means that will permit consistent production of agglomerated calcine of the desired consistency must be proven. The agglomerator is shown in Figure K-4 (Section K.8) and additional data are given in Table K-9 (Section K.8).

K.6.5 EXTRUSION

The function of the extrusion operation is to consolidate and form the waste into dense, circular cylinders. Monoliths that are 0.20 m in dia by 1.0 m long, when fired, will comply with the specified maximum of 3.06 kW/can. Because the cermet has excellent heat transfer properties, the maximum-sized monoliths permitted by the heat loading limit can be made so that the surface area of the waste form is minimized.

Calcine has been formed into shapes at ORNL on a laboratory bench scale by uniaxial pressing, isostatic pressing, hot pressing and piston extrusion. The latter is recommended for continuous production of large cermet forms. Piston extrusion should be less difficult and have a higher production rate per piece of equipment than either isostatic or hot pressing in a remote facility. It should produce a large body with a lower density gradient than uniaxial pressing. Hot-isostatic pressing has also been considered, but appears too complicated when compared to extrusion. Auger extrusion has not been demonstrated with calcined nuclear waste containing additives for cermet formation, but it may be an option for a continuous production process.

The agglomerated calcine with the right moisture content is discharged directly from the cooler/mixer into the extrusion press chamber. The chamber contains about 5% to 10% more material than is required to produce a cylinder of the specified size. The chamber is then sealed and evacuated to de-air the material. The vacuum pump exhaust is discharged in the process off-gas system. It contains air and some water vapors.

After the material in the chamber has been de-aired, the hydraulic ram is activated to compress the material and extrude it through the die. Personnel at ORNL report that 4500 psi are required to extrude calcined cermet material with their bench press, which has a 1/2-in.-dia disc and a 1-in.-dia piston. They also report that the dia shrinkage during firing is 25%. Therefore, the extruder die has an approximate dia of 27 cm.

When the cylinder is extruded to the desired length, which will be approximately 1.05 m because the linear fired shrinkage is 5%, the ram is stopped and the cylinder is cut off the extruded column and inspected. Fines from the extrusion and cutting operations and rejected cylinders are crushed and returned to the mixer via a bucket elevator and auger.

Inspection of the extruded cylinders includes dimensional measurements, density measurement and visual inspection for laminations. The tolerances on the green bodies will have to be determined as more is learned about the sintering behavior of large shapes made with the cermet composition, but they should not be too tight. Therefore, the gauging should be easily accomplished with remote go-no-go equipment. Rejected, extruded cylinders are crushed before they are returned to the mixer. A hammermill^(a) should be effective for this purpose.

Scaling up the extrusion process to production capacity and making it fully remote will require considerable development, design and test efforts even though large, piston extrusion presses are commercially available and are in routine use. Charging the agglomerated calcine from the cooler/mixer into the extrusion chamber may require some mechanical assistance, such as vibration or an auger. The extrusion pressure and rate, exact die size and shape, amount of de-airing, and the amount of moisture will have to be determined. The potential for spread of contamination in the process cell via the extrusion/recycling system may be high. Elaborate containment measures may be required to avoid excessive decontamination problems. The extruder is shown in Figure K-5 (Section K.8), and additional data on the extruder are given in Table K-10 (Section K.8).

K.6.6 REDUCTION AND SINTERING FURNACE

The functions of the reduction and sintering operation are:

- to drive off the residual water from the extruded cylinders by heating
- to form the desired ceramic compounds of the waste materials
- to reduce the appropriate metal oxides to the metallic state
- to form a metal alloy and sinter it into a matrix that encapsulates the ceramics.

(a) Koppers Co. Inc. one direction hammermill model No. 507 (100 to 150 hp).

The result of heating the cylinders in a reducing atmosphere is a dense, high-integrity waste form that has good thermal conductivity.

After a cylinder is extruded, inspected and accepted, it is moved through an air lock into the retort of a horizontal, continuous electric furnace. The retort is continuously purged with Ar + 4% H₂. The cylinders are moved through the furnace in a direction normal to their major axis. Because the design production rate of the cylinders is one every 3-3/4 h, their progress through the furnace is relatively slow. Thus, thorough drying and complete reduction is assured. The maximum temperature in the firing cycle is 1050°C. After the cylinders have passed through the maximum temperature zone of the furnace, they are cooled by a gas purge and then exit through another air lock.

Establishing the optimum firing cycle requires an understanding of the reaction kinetics involved in producing the ceramic compounds and alloy composition desired. The firing cycle not only affects what phases are present in the final product, but it determines the morphology of the finished product. The grain structure and size in turn determine the mechanical properties of the cermet. Much research and development remains to be performed in this area of the cermet waste form process. Nevertheless, the firing cycle shown in Figure K-6 in Section K.8 is proposed for the purpose of establishing a furnace size and a processing time table. Moving the cylinders through the furnace at a rate of 6 in./h will require a 12-ft-long retort with about a 4-ft heated section.

The exhaust from the retort is argon, water and traces of hydrogen. The formation of volatile waste species during the reduction and sintering operation is minimized by the reducing atmosphere.

The cermet cylinders exit the retort at approximately 400°C and enter a cooling chamber where the residual heat is removed by air circulation and water-cooled heat exchangers. Because the extruded cylinders have low green strength but weigh approximately 295 kg each, the design of the mechanisms for supporting and conveying them remotely through the air locks and retort will require major development work.

Control of the reduction and sintering operation is accomplished by:

- maintaining the proper temperature profile over the length of the retort
- moving the cylinders through the furnace at the correct rate
- maintaining the proper flow rate of reducing gas mixture through the retort.

An oxygen analyzer located in the exhaust line from the furnace's retort aids in detecting leaks in the air locks.

The air locks and cylinder-conveying mechanism are the equipment in this operation that require the most servicing. Because elastomers can not be used in a radiation environment, sealing the air-lock doors and conveyor drive shafts requires a special design. Moving parts in high-temperature environments have a high failure rate, so the conveyor must be designed for remote access and servicing. The handling of extruded cylinders, both before and after the reduction/sintering operation, is likely to result in the spread of contamination unless special containment provisions are made.

The size and complexity of this process step in a remote operations and maintenance mode will require considerable research and development to provide a viable system. The reduction and sintering furnace is shown in Figure K-7 (Section K.8), and other requirements for the furnace are given in Table K-11 (Section K.8).

K.6.7 CERMET INSPECTION

The function of this operation is to assure that each cermet cylinder produced meets the requirements established for solidified waste forms prepared for disposal. Because such requirements have not yet been established, it is difficult to specify the samples and tests that will be necessary. The cermet cylinders should be homogeneous because of the solution of all the constituents in urea to start with. Therefore, the number of samples required to be representative should be small. It is hoped that all inspections can be nondestructively performed, but if samples are required for analyses the cylinder may be drilled. Further development in this area will include demonstrating remote inspection methods to satisfy whatever requirements are established and to determine the required sampling levels.

K.6.8 CANNING OPERATION

The function of this operation is to encapsulate the cermet cylinder in a canister. The canister is similar to that described in the reference process except that the diameter of the opening at the top is the same as the inside diameter to accommodate the cermet cylinder. The clearance between the canister inside diameter and the cermet cylinder is small to promote heat transfer. Steel shot may be added if necessary to improve heat transfer. Closure, testing, inspection, and further processing of the sealed canister are very similar to those described under the reference process.

Two areas of the canning operation require special attention and further development. First, the dimensional control possible with the extruded and sintered cermet cylinder must be determined so that the canister diameter tolerances can be established. Second, during the performance of leach tests on samples of cermets prepared in a laboratory, galvanic corrosion of the alloy in contact with stainless steel was noted. Therefore, the canister alloy and the cermet alloy must be compatible to minimize corrosion of either.

K.6.9 OFF-GAS SYSTEM

The function and operation of and the research and development required for the cermet off-gas system are basically the same as specified for the ICGM process, except that the capacity of the cermet off-gas system is greater than the reference off-gas system. In addition, the cermet off-gas system must handle and process CO₂ and ammonia. Operating requirements and efficiencies will determine if dual systems should be used.

K.7 ALTERNATIVE PROCESSES AND EQUIPMENT CONSIDERED

An alternative to the use of metal nitrates as the source of the alloy formers has been suggested by personnel at ORNL (Aaron et al. 1979a). The DOE has stockpiles of contaminated metals that are unsuitable for commercial use. They are estimated to contain 30,000 tons of steel, 17,000 tons of nickel, and 1,700 tons of Cu; future inventory increases are estimated to range between 500 to 7,000 tons/yr. The waste facility could include processing equipment to convert these contaminated metals to a form suitable for adding to the HLLW for the formation of the metal matrix of the cermet. This equipment would most likely include cranes and manipulators for handling large items, cutting equipment and acid dissolvers. This alternative would definitely increase overall process complexity.

An alternative ceramic component in the cermet composition being considered is an alkaline earth titanate. Thus, the addition of a soluble titanium compound such as TiCl_3 would be required. The corrosive effects of chlorides must be considered in this approach.

An alternative to the silicic acid added to the HLLW to form aluminosilicates is colloidal SiO_2 . This has been successfully used as an additive to simulated HLLW feeds for the heated-wall spray calciners operated at PNL.

Continuous addition of the urea and the other additives to the HLLW stream as it flows through a system of weirs has been proposed as an alternative to the batch preparation of calciner feed described in this preconceptual design. Such an operation would result in a more continuous process, but would require close control of the HLLW flow rate and proportional addition of the additives. Another option is to increase the dissolver batch size to reduce the amount of analytical work and additives batching that is required.

It may be possible to produce the cermet waste form without the intermediate urea dissolution operation. If this is possible, the operation of the calciner would be the same as in the reference process. The volume of the off gases would also be greatly reduced. This approach could greatly simplify the overall process.

An internal-mix, pneumatic spray nozzle is recommended for use in this preconceptual design because it has been proven on large-scale spray calciners at PNL. An ultrasonic spray nozzle is being evaluated at ORNL for use in the calcining of the urea solution (Aaron et al. 1979a).

The iron-nickel-copper alloy used in this study is not considered a final, optimum composition. Alternative alloy compositions that are compatible with the canister alloy and the ceramic compounds desired will surely evolve as the process and product are developed. Maximum leach resistance and ease of sintering will undoubtedly be the dominating criteria in alloy development.

In Section K.5, State of Process Development, experiments with the reduction of the alloy-forming materials to the metallic state in the calciner followed by isostatic pressing of the resulting powder were described. This is a significant alternative to the preconceptual

process, because it would eliminate the sintering operation which is likely to be a maintenance problem and a source of volatile waste materials in the process effluents. However, such an operation may require the use of hydrogen in the atomizing gas. Also, operation of an iso-static press in a remote facility will require considerable development.

Auger extrusion of the calcine has been suggested as an alternative to extrusion using a press because it is a continuous process. This forming method has not been demonstrated with the calcined cermet composition. It may be difficult to obtain large, cylindrical shapes of sufficient density and homogeneity using an auger extruder.

Another cermet-forming alternative is to pelletize the calcine into small spheres that are then sintered and cast in a matrix such as lead. The cermet has such good thermal conductivity that such additional processing may not be justified. However, the elimination of the extrusion step and the simplification of the reduction/sintering step which may accompany this approach may warrant further investigation.

An alternative to the continuous sintering operation is to use periodic furnaces to fire the cermet cylinders. Such an operation in a remote facility may have several advantages over a continuous furnace. It would eliminate such high maintenance items as the air locks and conveyor. It would also eliminate the potential for a jam-up of cylinders in the furnace. However, it will require thermal cycling of the furnaces which shortens the life of the elements and other components.

As an alternative to using $\text{Ar} + 4\% \text{H}_2$ as the reducing gas in the sintering furnace retort, a mixture containing 12% CO as the reducing agent may be used.

As evaluation of the cermet waste form and the processes for its production continues at ORNL, SRL and PNL, additional alternatives may be identified and some of those suggested here may be discarded. These ongoing efforts are pertinent to the development of an optimum cermet waste form and a viable process to produce it.

K.8 EQUIPMENT DATA SHEETS AND DRAWINGS

The reader should refer to Appendix A, Section A.8 for a glossary of the symbols and abbreviations used in the figures in this section.

TABLE K-6. Equipment Data Sheet for Chemical Bins, Tanks and Batch Weigher

Function

Provide capability for chemical makeup, storage and batch weighing.

Process Interfaces

Dissolver; feed system.

Design Parameters

Atmospheric pressure for bins and -10 in. H₂O for tanks; temperature = 25° to 100°C; propeller agitator in nitrate solution and batch tank; material--304L stainless steel.
Bin volumes: urea--1300 ft³; silicic acid--6 ft³; batch hopper 190 ft³(a)
Tank volumes: Fe(NO₃)₃ · 9H₂O--46,200 L; Ni(NO₃)₂ · 6H₂O--3900 L; Cu(NO₃)₂ · 3H₂O--1280 L; Al(NO₃)₃ · 9H₂O--490 L; batch--6500 L.

Operating Conditions

All bins and tanks at atmospheric pressure (may want negative exhaust on tanks); bins at ambient temperature; tanks may be heated to 80°C to improve nitrate solubility.
Bin volumes: urea--860 ft³; silicic acid--5 L; batch hopper--125 L.
Tank volumes: Fe(NO₃)₃ · 9H₂O--38,000 L; Ni(NO₃)₂ · 6H₂O--3200 L; Cu(NO₃)₂ · 3H₂O--1050 L; Al(NO₃)₃ · 9H₂O--400 L; batch--5300.

Features

Temperature sensors; vessel ventilation; level sensors; agitators; steam heating capability.

Auxiliaries or Services

Steam (heating); electrical (agitators, temperature sensors, level sensors); vacuum (vessel ventilation); water and nitric acid (chemical makeup); instrument air (level sensors).

Maintenance Methods

Manual

Remarks

(a) General Resource Corp. hopper scale model No. FHS85-A.

TABLE K-7. Equipment Data Sheet for Dissolver and Feed Tanks

Function

Dissolve HLLW and cermet-forming additives in molten urea and serve as spray calciner feed tank.

Process Interfaces

Additives' storage and delivery system; fuel-reprocessing-plant HLLW delivery system; spray calciner process off-gas system.

Design Parameters

Capacity = 18,000 L (1 day's requirements); material--304L stainless steel; pressure = 25 cm H₂O; temperature = 40°C during filling, sampling and additions and 60°C during dissolving, melting and feeding; heating capability = 400 kW; cooling capability; agitator is vertically mounted, propeller-type; ventilation to process off-gas system.

Pumping system: cantilever slurry pump; 15 to 25 hp; 23,000 L/h @ 30,500 kg/m²; magnetic flowmeter; diaphragm flow control valve; recycle loop.

Operating Conditions

Nominal batch volume = 8750 L; cooling during filling, sampling and additions; heating during dissolving and melting; continuous pumping and agitation.

Features

Temperature, pressure and level sensors; cooling and heating coils and jackets.

Services or Auxiliaries

Electrical (pump, agitator, flowmeter, pressure air locks, temperature sensors, liquid sensor); water (cooling); steam (heating); instrument air (level sensor, atmospheric pressure sensor, diaphragm valve).

Space Requirements

2.3 m in dia by 6.0 m high (includes pump and agitator motors).

Maintenance Methods

Remote

Remarks

FIGURE K-3. Schematic Diagram of the Spray Calciner

TABLE K-8. Equipment Data Sheet for Spray Calciner

Function

Convert molten urea solution to metal oxide powder.

Process Interfaces

Dissolver and feed tanks; calcine agglomerator; process off-gas system.

Design Parameters

Capacity = 30 to 547 L/h; temperature = 180° to 800°C; pressure = 0.5 to 2.0 atm; material--310 stainless steel; vibration to remove wall accumulations; heated surface area = 12 m²; filter system with blowback capability; inert atomizing gas.

Operating Conditions

Capacity = 363 L/h; temperature = 300°C for filter chamber and 800°C for calcining chamber; pressure = 0.97 atm; periodic vibration.

Features

Furnace with 3 zones @ 200 kW/zone; vibrators; sintered-metal filters; temperature and pressure sensors; ceramic nozzle insert.

Auxiliaries or Services

Electrical (heating system, temperature sensors); compressed air (filter blowback, vibrators); instrument air (pressure sensor); argon (atomizing gas).

Space Requirements

3 m in dia by 8 m high

Maintenance Methods

Remote

Remarks

FIGURE K-4. Calcine Agglomerator

TABLE K-9. Equipment Data Sheet for Calcine Agglomerator

Function

Convert calcine to dense spheres suitable for extrusion.

Process Interfaces

Spray calciner; extruder.

Design Parameters

Capacity = 92 L/h calcine; binder water = 7.4 L/h; cooling water = 1000 L/h; materials-- 304 series stainless steel with carbide or nitride leading edges; temperature = 20° to 400°C.

Operating Conditions

Capacity = 61 L/h; binder water = 4.9 L/h; temperature = 50°C.

Features

Temperature and pressure sensors; torque indication; cooling water taps; binder water addition tap; heater on calcine addition pipe.

Auxiliaries or Services

Electrical (motor, heater, temperature sensors); water (binder, cooling; instrument air (pressure sensor)).

Space Requirements (per agglomerator)

0.5 m (wide) by 0.5 m (high) by 1.5 m (long)

Maintenance Method

Remote

Remarks

FIGURE K-5. Piston Extrusion Press

TABLE K-10. Equipment Data Sheet for Extruder

Function

Compacts calcine pellets into dense, green cylinders suitable for reduction/sintering.

Process Interfaces

Calcine agglomerator; green cylinder inspection station.

Design Parameters(a)

Tonnage = 100 MT; extrusion speed = 40 cm/min maximum; hydraulic pumping system is 20 hp; vacuum pump = 28 scfm, 20 hp; maximum cycle time = 3 h.

Operating Conditions

Pressure = 4500 psi; estimated cycle time = 20 min.

Features

Temperature and pressure sensors.

Auxiliaries or Services

Cooling water (hydraulic system); electrical (pumps, temperature and pressure sensors).

Space Requirements

3 m long by 0.5 m in dia

Maintenance Methods

Remote

Remarks

(a) Data based on Mohr Machinery Co., Inc. Model Number MEP - 23000 Piston Extrusion Press.

FIGURE K-6. Firing Cycle for Cermet Cylinders

FIGURE K-7. Reducing and Sintering Furnace

TABLE K-11. Equipment Data Sheet for Reduction and Sintering Furnace

Function

Reduce and sinter green extruded calcine cylinders into cermet waste forms.

Process Interfaces

Extruded cylinder inspection station; cermet inspection station.

Design Parameters

Operating rate = 6.4 cylinders/d (56.2 kg/h); conveying rate = .15 m/h; temperature capability = 1200°C; pressure = 0.5 to 2.0 atm; Ar • 4% H₂ flow rate = 73.2 kg/h; furnace has 2 zones, each 10 kW; retort--Inconel 600 or Hastalloy X; cooling capacity = 5.4 kW.

Operating Conditions

Operating rate = 4.3 cylinders/d (37.3 kg/h); sintering temperature = 1050°C; pressure = 1.03 atm; Ar • 4% H₂ flow rate = 50.5 kg/h; cermet temperature in exiting cooling chamber = 400°C.

Features

Loading and conveying mechanism; air locks; pressure and temperature sensors; oxygen analyzer.

Auxiliaries or Services

Electrical (furnace, temperature sensors); Argon • 4% H₂ (reducing gas); water (cooling); instrument air (pressure sensor).

Space Requirements

8 m (long) by 1 m (high) by 0.5 m (wide)

Maintenance Methods

Remote

Remarks

K.9 REFERENCES

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