Safety Assessment of the Liquid-Fed Ceramic Melter Process

**J. L. Buelt
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August 1980

**Prepared for the U.S. Department of Energy
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SUMMARY AND CONCLUSIONS

As part of its development program for the solidification of high-level nuclear waste, Pacific Northwest Laboratory assessed the safety issues for a complete liquid-fed ceramic melter (LFCM) process. The LFCM process, an adaptation of commercial glass-making technology, is being developed to convert high-level liquid waste from the nuclear fuel cycle into glass. This safety assessment uncovered no unresolved or significant safety problems with the LFCM process. Although in this assessment the LFCM process was not directly compared with other solidification processes, the safety hazards of the LFCM process are comparable to those of other processes. The high processing temperatures of the glass in the LFCM pose no additional significant safety concerns, and the dispersible inventory of dried waste (calcine) is small.

High-level liquid waste could come from a government defense plant or from a spent-fuel reprocessing facility for a nuclear power generator. This safety assessment was based on the nuclear power waste flowsheet, since power waste is more radioactive than defense waste at the time of solidification, and all accident conditions for the power waste would have greater radiological consequences than those for defense waste. The power waste flowsheet on which this assessment is based is designed to solidify waste generated from a 5-MTU/d reprocessing facility. The high-level waste has a maximum concentration of 7M, and the combined processing rate for high- and intermediate-level liquid waste is 138 L/h (assuming 662 L/MTU).

An exhaustive list of possible off-standard conditions and equipment failures was compiled. These accidents were then classified according to severity of consequence and type of accident. The severity of an accident (minor, moderate or severe) was determined by the potential for radioactivity release and the extent of equipment damage. The types of accidents were those characterized by liquid waste spills, molten glass releases, process pressurizations, failure of an off-gas system component, and process cell damage.

Radionuclide releases to the stack were calculated for each group of accidents using conservative assumptions regarding the retention and decontamination features of the process and facility. The most severe accident was found to be a design-basis tornado, which is assumed to cause all high-efficiency particulate air filters in the facility to rupture. The depressurization caused by a tornado would cause approximately 44 g of particulate (105 Ci) to be released from the plant stack. Using a simplified inhalation dose calculation for the assumed site parameters, the design-basis tornado results in a 50-yr whole-body dose of 0.58 rem due to acute exposure of an individual at the site boundary. While there are no firm guidelines by which to judge the severity of this site-boundary dose, it can be compared to reactor site regulations (Nuclear Regulatory Commission Reactor Site Criteria 1979), which state that reactors must be located such that a design-basis accident would not result in whole-body doses greater than 25 rem.

Two recommendations that should be considered by process designers are given in the safety assessment. First, the assessment points out the importance of cell high-efficiency particulate air filters as a barrier to radionuclide releases. Thus, process cell designers should consider filter protection, as well as ease of maintenance, when designing a solidification plant. Secondly, the assessment assumed no credit for decontamination of ruthenium once it volatilizes to the cell atmosphere, since the behavior of volatilized ruthenium through filters and ductwork is uncertain. If in the future this assumption is shown to be correct, ruthenium trapping at the stack should be considered.

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INTRODUCTION

The Pacific Northwest Laboratory (PNL) is developing the liquid-fed ceramic melter (LFCM) process for the U.S. Department of Energy (DOE) for use in a waste-solidification plant. The LFCM immobilization process is designed to convert high-level liquid waste (HLLW) into glass in one continuous step. The final product of the LFCM process is a chemically and radiologically stable glass (Ross et al. 1979). Work was begun on the joule-heated ceramic melter at PNL in 1973, and in 1976 the process of directly feeding liquid waste to a joule-heated melter was begun on a pilot-scale (Buelte and Chapman 1979). Since that time both pilot-scale and full-scale experiments with the melter have been conducted using simulations of both defense wastes from a government defense facility and power wastes from a nuclear power generator.

As an instrumental part of PNL's development of the LFCM process, PNL conducted this safety assessment. Off-standard conditions and equipment failures were postulated for a generic LFCM process, and the resulting equipment damage and releases of radioactivity were estimated. In addition, to help interpret the results a simplified inhalation exposure model was used to calculate a site-boundary dose for each accident group. Designers of an actual waste-solidification facility can use this assessment to help establish reliability standards for equipment, services and radionuclide barriers, and to reduce the expected occurrence of off-standard conditions to an acceptable, cost-effective minimum.

This safety assessment reflects the current knowledge and experience of the research personnel who have developed the LFCM process. It does not compare the LFCM process with other solidification processes, but concentrates on the unique features of the LFCM. The associated systems, such as the feed system, off-gas system, and process cell, are included because of their close interrelationship with the melter. Finally, since defense waste is more dilute than power waste and its activity has decayed during its longer storage period, the radiological consequences of accidents involving power wastes are more severe than they are for accidents involving defense wastes. Therefore, to evaluate worst-case accidents the radiological calculations in this safety assessment are based on the more radioactive commercial power waste.

PROCESS AND FACILITY DESCRIPTION

Since no LFCM vitrification facility yet exists for either defense or power waste, it is necessary to describe a generic LFCM process and facility. Defense and power waste flowsheets have similar process and equipment requirements. The flowsheets differ primarily in the composition, dilution, activity and acidity of the feed, the composition of the glass, the characteristics of the off gas, and the required processing rate. Since these differences do not appreciably affect process design, only one basic process and one facility are described for both flowsheets. An actual solidification plant may differ from this generic process and facility to accommodate the specific requirements of that plant.

The generic LFCM process shown in Figure 1 is based on the philosophy of zero liquid discharge. The process consists of the following:

- a feed system, through which the high-level waste is received, conditioned and metered to the vitrification equipment;
- the vitrification equipment, including the ceramic melter, the receiving canister, and the canister change-out and handling mechanisms;
- the off-gas system, which treats any radionuclide or chemically toxic effluents to meet regulations governing their release.

The LFCM process description that follows is based on many man-years of experience at PNL, including the solidification of over 14,000 L of nonradioactive, simulated HLLW. The developmental work is continuing and design improvements are being made in areas such as process rate boosting, melter capacity scale-up, aerosol entrainment control, ruthenium volatility control, and safety and operational design. However, this safety assessment does not take credit for design features that have not been experimentally proven.

FIGURE 1. Liquid-Fed Ceramic Melter Solidification Process

THE FEED SYSTEM

High-level and intermediate-level liquid wastes are introduced to the solidification plant through the feed system (see Figure 1). The first step in the feed system is the feed-preparation tank, which is equipped with instruments to remotely monitor and characterize the waste. It also has equipment to agitate the waste and water-cooling and steam-heating coils for controlling the temperature of the waste. The proper amount of glass formers is added to the liquid feed while it is in the feed-preparation tank. The steam coils are used to preheat the feed to 90°C. This is a precautionary step to completely eliminate any possibility of a steam explosion in the melter. (Steam explosions are discussed in more detail in Appendix A.) As a quality-assurance and processing-control step, the feed from this preparation tank is also sampled before it is transferred to a feed tank.

The feed from the feed-preparation tank is typically conditioned and transferred to the feed tank once per shift. This feed tank is equipped with water-cooling and steam-heating coils for temperature control. It is also agitated to keep solids in suspension and has instruments that monitor the tank level and solution temperature. From the feed tank, the HLLW is metered to the LFCM by an air lift. The air lift operates by injecting air into the longer side of a U-tube, creating a density difference between the two sides of the U-tube. This causes a controllable amount of liquid to flow into the head pot. Here, the air and liquid waste separate, the liquid waste flowing to the melter through a water-cooled nozzle and the air venting to the off-gas system.

LIQUID-FED CERAMIC MELTER

The LFCM described in this safety assessment is similar to the one that has been under development with nonradioactive, simulated waste at PNL for the past three years (Buelt and Chapman 1978). This melter operates on the same basic principle as any commercial electric melter in the glass industry. Molten glass is contained within a cavity that is bordered by glass-contact, backup, and insulating refractories. The glass is joule-heated by passing an alternating current between electrodes at opposite ends of the cavity. As the

liquid waste, which is premixed with glass formers, is fed to the melter, heat is transferred from the molten glass through a floating crust to the aqueous pool, as shown in Figure 2. Since the floating crust is physically stable, the LFCM has an added safety feature in that the amount of dispersible calcine is small.

As the operation continues decomposition gases and water of hydration, which are produced by the calcination of the floating crust, percolate up through the crust and aqueous pool. Sometimes when these gases break through the crust, molten glass comes into contact with the aqueous pool. The resulting increase in heat transfer creates surges in the off-gas flow of up to three

FIGURE 2. Liquid-Fed Melter Operation

times the normal rate for a few seconds; however, the off-gas system has been sized to accommodate such surges. As the dried crust is heated, it begins to flux with the glass formers and goes into solution with the glass. From that point on the glass flows through an overflow drain system and into the receiving canister. When the melter is in operation, the glass in the melter is typically 1200°C (1050°C at the electrodes). The safety concern with a high-temperature operation such as this is the release of volatile radionuclides during a glass spill; however, later sections of the report show that this is not the major factor contributing to radionuclide releases. Steady-state temperatures above the aqueous pool depend upon the degree of flooding on the molten glass surface and the amount of auxiliary power input supplied by the lid cavity heaters; temperatures may range from 200°C to 400°C.

Figure 3 shows the basic parts that make up the LFCM. They are:

- the refractory, which contains the molten glass by primary and secondary refractory layers and offers thermal and electrical insulation between the glass and the surroundings;
- the electrodes and power system, which control and provide the necessary power to sustain melting;
- the containment box, which supports the refractory and electrode materials, contains the effluents, and serves as a backup to the refractory for glass containment.
- the primary drain system, commonly known as the overflow, which drains glass from the bottom of the melter into an overflow trough for glass-pouring operations;
- the bottom drain, which provides for complete draining of the LFCM for scheduled and emergency shutdowns;
- the instrument system, which monitors and controls temperatures, electrical properties, cooling flow, and signals to the engineering safeguard systems.

FIGURE 3. Liquid-Fed Ceramic Melter

Each of the components listed above have been refined during the developmental period to increase reliability and safety of operation. Each is discussed in more detail below.

Refractory

The refractory is composed of three layers of composite materials. The first is the glass-contact refractory, which is a very dense, fused-cast material. Directly behind the glass-contact refractory is a castable, high-alumina refractory that is used for added thermal insulation and secondary glass containment. Behind this layer is a very porous, highly thermally insulative refractory. The melter cavity outlined by the glass-contact refractory measures 86.4 cm (34 in.) wide by 122.0 cm (48 in.) long by 66.0 cm (26 in.) high; 48.3 cm (19 in.) is the normal glass depth.

Electrodes and Power System

The electrodes are composed of a nickel and chromium alloy. Two independently controlled sets of electrodes are imbedded directly into opposite walls of the melting cavity. The two-zone arrangement is designed to control glass temperatures vertically in the melting cavity. The electrodes are connected to the power control system by an isolation transformer so that the electrical circuit on the melter is isolated from the ground. The power control system operates in a constant current mode so that if the glass overheats no additional power would be introduced, as would be the case with constant voltage control. Except for insulated electrical cables and electrode connectors, the power control system is located outside the process cell. Accidents from maintenance of the power control system outside of the cell are not addressed in this safety assessment.

Containment Box

The stainless steel containment box holds the refractory and electrodes in place and contains the effluents at a slightly negative gauge pressure (-2.5 kPa; -10 in. water). The box is divided into two sections, the main body and the lid. The main body, which encloses the refractory below the glass level, has water and air-cooling coils to control the temperature of the electrodes and specific sections of the overflow drain. These coils are designed so that failure will not allow the coolant to contact molten glass. The lid encompasses the plenum area above the glass level, which provides head space for aerosol separation. The lid also encloses electrical-resistant heaters imbedded in the insulation. These heaters provide extra heat to the glass surface for increasing the capacity of the LFCM. The containment box is 1.9 m long by 1.6 m wide by 1.5 m high. In the case of process upsets that may produce steam surges in excess of the off-gas system capacity, the integrity of the containment box is protected by a pressure-release vent to the process cell.

Drain Control

The primary drain control system draws glass from the bottom of the melting cavity, up through a tunnel in the refractory known as the riser block,

down a refractory trough, and into the receiving canister. Glass flow is controlled by the air-lifting principle; a platinum tube in the riser block bleeds in air, which causes glass to rise into the trough. The overflow section is adequately insulated and is heated by electrical heaters mounted behind a protective barrier.

The bottom drain, which uses a freeze valve, is a safety feature for draining the melter in a scheduled or emergency shutdown. By reducing the air flow to the freeze valve and applying heat with electrical-resistance heaters, the frozen glass plug softens and allows the glass in the melter to drain controllably into the bottom-drain canister.

Instrument System

The melter is fully instrumented with thermocouples, power system monitors, gas and liquid flow monitors, and a pneumatic transducer to monitor the vacuum in the melter plenum. The instrument system is tied into safeguard alarms and automatic shutdown devices. Engineered safeguards include:

- an automatic feed shutdown in the event of a melter pressurization or power failure;
- an automatic power-shutdown switch and alarm in the event electrical resistance between the electrodes suddenly increases beyond a specified range, e.g., during extensive foaming or a reboil (Pye 1979);
- temperature alarms for the glass, electrodes, containment box and overflow system;
- receiving canister high-fill alarms as indicated by load cells and canister neck temperatures.

THE OFF-GAS SYSTEM

The off gases from the vitrification process are condensed and decontaminated in the off-gas system (Hanson and Kaser 1976). The volume and decontamination capabilities of the system exceed the regulatory limits for normal operation and most off-standard conditions. The off gas is composed primarily of steam, air, decomposition gases, aerosols and volatile radionuclides.

Though the volume of the constituents depends on the process flowsheet, air flow due to inleakage is typically $0.57 \text{ m}^3/\text{min}$ (20 scfm), and aerosol entrainment is typically 0.5 wt% of the waste (Buelte and Chapman, 1979).

Melter off gases travel first to a venturi ejector. The venturi, located close to the melter, removes most of the particulates and condenses nearly all of the steam. After the gases are separated from the condensate, a tube-and-shell condenser provides a high decontamination factor (DF) for small particles as it removes the last of the condensables. It also offers backup capacity in the event of venturi failure or any other off-standard condition.

The scrubbed and condensed liquids from the venturi ejector and tube-and-shell condenser are decontaminated in a series of evaporators and condensers that serve as a condensate-cleanup loop. The evaporator bottoms from this loop are recycled to the HLLW feed system, while decontaminated condensate is evaporated through the plant stack.

The noncondensable gases from the tube-and-shell condenser are drawn through a mist eliminator to a series of high-efficiency particulate air (HEPA) filters, humidity control heaters, and ruthenium and iodine adsorbers. From there a blower moves the decontaminated noncondensables to the plant ventilation system, where they are mixed with the process cell ventilation air and are released to the atmosphere through the plant stack.

PROCESS CELL

The cells for the LFCM process serve as the Class I safety boundary as defined by Nuclear Regulatory Commission (NRC) guidelines and provide the primary shielding for personnel. No specific cell design is recommended in this report, but a brief description of the cells with a ventilation flow schematic is necessary for this safety assessment. This safety assessment is based on using two primary cells (the feed cell and the solidification cell), plus associated piping and service galleries. The entire cell area is within an operating gallery, which has a controlled ventilation system. The major

equipment in each primary cell and the approximate cell dimensions are given in Table 1. For this safety assessment it is not necessary to determine actual equipment layout in the cells.

The cell safety features assumed for this accident analysis are the following. A sloped, stainless steel-lined floor in the cell contains liquid and glass spills. Even though only a few combustible items are allowed in the cell, primarily electrical cable insulation, a fire-suppression system is included. The water, air and steam services contain isolation heat exchangers or otherwise provide containment of any contamination that may enter service lines during an accident. The cell also has lead-glass viewing windows, overhead cranes and equipment-access hatches.

TABLE 1. Conceptual Process Cell Specifications

<u>Cell Designation</u>	<u>Approximate Size</u>	<u>Major Equipment</u>
Feed cell	4.9 m x 7.3 m (floor) and 6.1 m in height (floor to ceiling)	2 feed tanks (~1200 L) 2 transfer pumps 1 overhead crane
Solidification process cell	4.9 m x 7.3 m (floor) and 15.7 m in height	1 feed air lift (~150 L/h) 1 joule-heated glass melter (~500 L) 1 canister being filled (~100 L) 5 canisters being processed 1 venturi-ejector tank (~3000 L) 1 condenser 1 condensate-cleanup system (evaporators and condensers) 1 ruthenium sorber 1 iodine sorber 1 overhead crane

Because of the massive cell structure and the zero-liquid-discharge philosophy, the ventilation system, which combines cell ventilation and process off-gas, is the principal path for radionuclide release to the environment. The prefilters and HEPA filters are the primary safeguard against radionuclide release through this path, though no credit is taken for decontamination by the prefilters in this assessment. The assumed layout of the ventilation system is shown in Figure 4. The decontamination factors (DF) for the HEPA filters are also indicated in Figure 4. Other potential engineered safeguards are not shown, such as fast-acting ventilation dampers and shock-wave arresters, which along with the prefilters provide extra protection for the HEPA filters.

FIGURE 4. Location of HEPA Filters in The Ventilation System

FLWSHEETS

To conduct this safety assessment, the process flowsheet assumptions had to be delineated. A comparison of defense and power waste flowsheets indicated that defense wastes are less hazardous than power wastes. Defense wastes are seven times less active than power wastes, and their ruthenium concentrations (which account for the major release in off-standard conditions) are considerably lower. In addition, the flow rates of aqueous and off-gas streams and most equipment capacities are lower for defense wastes. Because of the less hazardous nature of defense wastes, no accidents could be identified involving defense wastes that would have more severe consequences than accidents involving power wastes. Thus, for simplicity this safety assessment was conducted only for the power waste flowsheet.

This approach is also justifiable because the defense waste and power waste flowsheets are extremely similar. The only accident identified that may be unique to the defense waste flowsheet is a release of a concentration of mercuric iodide which contains the long-lived I-129. However, the radiological consequences of such a release would be small compared to other releases calculated in this safety assessment due to the fact I-129 has a low specific activity. The hazards of mercury vapor release were also considered in the preliminary analysis to determine if the chemical toxicity would approach the radiological hazards of other accidents. The Environmental Protection Agency's (EPA) atmospheric emission standards (National Emission Standards for Mercury 1979) for some industries permit up to 2.3 kg of mercury to be released over a 24-h period for normal operation. Therefore, for a mercury release to be significant during an accident it would have to be orders of magnitude greater than the EPA standard.

In this section, both the power waste and defense waste flowsheets are briefly summarized. The defense waste flowsheet is included primarily for comparison purposes. For more detailed information on the particular power waste flowsheet on which this safety assessment is based, refer to Appendix B.

POWER WASTE FLOWSHEET

The reference power waste flowsheet is based on the solidification of 3-yr-old HLLW. The predicted isotopic concentration of the HLLW is based on an ORIGEN (Bell 1973) code for fuel having the following power and exposure history:

Reactor type	pressurized water reactor
Burnup	29,000 MWd/MTU
Specific power	32 MW/MTU
Initial enrichment	2.8% ^{235}U
Reactor operating factor	80%
Age at reprocessing	3 yr

These fuel characteristics were selected by Hall (1977) in his description of a potential vitrification facility for Allied Gulf Nuclear Services. Hall's assumption of 3-yr-old fuel was warranted by: 1) a sufficient supply of old fuel through the foreseeable future, 2) economic advantages of processing aged fuel, and 3) a reduction in population dose commitments as a result of this age restriction.

Power waste is produced from reprocessing spent fuel. During reprocessing, 99.5% of the uranium, plutonium and iodine and all of the krypton and tritium are assumed to be extracted. This, with chemical additions, results in the HLLW composition given in Table 2. The maximum nitric acid concentration of 7M reflects high estimates for process chemical additions and off-gas capacity.

The capacity of the vitrification facility is assumed to be identical to the reprocessing facility, which operates 300 d/yr at 5 MTUWE^(a)/d. Therefore, for a HLLW concentration of 378 L/MTUWE, the vitrification facility must vitrify 78.8 L of HLLW/h. In addition, intermediate-level liquid waste (ILLW), another reprocessing side stream with a 284-L/MTUWE concentration, must be vitrified at 59.2 L/h. Finally, part of the condensate from the condensate-cleanup loop is recycled to the feed tank at 14 L/h. Thus, the combined HLLW,

(a) MTUWE--metric ton of uranium waste equivalent, which is the equivalent proportion of waste generated from 1 MTU of fuel.

TABLE 2. Characteristics of Power Waste Feed Solution

<u>Feed Characteristics</u>	<u>HLLW</u>	<u>ILLW</u>	<u>Recycled Condensate</u>	<u>Total</u>
Constituents, g/L				
Inerts				
H ⁺	4-7	2.5	4.0	4.97
Na	---	17.64	---	6.87
K	---	0.36	---	0.14
Cr	0.123	0.0323	---	0.076
Ni	0.065	0.017	0.07	0.047
Fe	0.458	0.121	---	0.285
Gd	64.15	---	---	33.24
Mn	---	0.5	---	0.189
NO ₃ Max	644.0	217.0	248.0	441.0
PO ₄	0.045	23.0	---	8.98
SO ₄	---	0.11	---	0.043
Cl	0.053	0.32	---	0.164
Shear fines	0.66	---	---	0.34
H ₂ O min	<u>424.0</u>	<u>844.0</u>	<u>832.0</u>	<u>625.0</u>
TOTAL	1141.0	1105.0	1084.0	1121.0
Fission Products				
Ba	3.5	Negligible	Negligible	1.81
Ce	5.78	Negligible	Negligible	3.0
Cs	6.09	Negligible	Negligible	3.16
La	2.96	Negligible	Negligible	1.53
Nd	9.57	Negligible	Negligible	4.96
Pd	3.2	Negligible	Negligible	1.66
Pr	2.79	Negligible	Negligible	1.45
Rb	0.78	Negligible	Negligible	0.408
Rh	0.90	Negligible	Negligible	0.466
Ru	5.1	Negligible	Negligible	2.64

TABLE 2. (contd)

<u>Feed Characteristics</u>	<u>HLLW</u>	<u>ILLW</u>	<u>Recycled Condensate</u>	<u>Total</u>
Fission Products (contd)				
Sm	2.18	Negligible	Negligible	1.13
Sr	2.04	Negligible	Negligible	1.05
Tc	1.95	Negligible	Negligible	1.02
Te	1.32	Negligible	Negligible	0.684
Y	1.09	Negligible	Negligible	0.564
Zr	8.58	Negligible	Negligible	4.45
Other	1.35	Negligible	Negligible	0.63
I (0.5%)	<u>0.003</u>	<u>0.0043</u>	<u>0.0773</u>	<u>0.011</u>
TOTAL	59.2	---	---	30.62
Actinides				
U (0.5%)	12.68	16.91	Negligible	13.22
Np	1.06	---	Negligible	0.556
Pu (0.5%)	0.119	0.152	Negligible	0.119
Am	0.645	---	Negligible	0.338
Cm	<u>0.053</u>	<u>---</u>	<u>Negligible</u>	<u>0.028</u>
TOTAL	14.55	17.07	Negligible	14.2
Insoluble Solids (constituents included above)	14.55	2.0	---	8.32
Physical Properties				
Maximum specific gravity	1.122	1.084	1.166	
Watts/L	7.02	0.012	Negligible	3.64
L/MTHM	378.0	284.0	67.0	729.0
Age at solid-yr	3.0	3.0	---	3.0

ILLW and recycled condensate must be processed at a rate of 152 L/h. Table 2 lists the constituents and properties of the HLLW, the ILLW and the recycled condensate. See Tables B.1 and B.2 in Appendix B for the mass and activity balances for the process lines and the equipment, respectively. Table B.3 lists the major pieces of equipment and includes a brief description of the purpose, size and operating characteristics of each.

The power waste solidification flowsheet does require some special design considerations that are not shown on the generic process flowsheet in Figure 1. These are the feed denitration step in the feed system and the NO_x destruction step in the off-gas system. Vitrification experiments with simulated high-level wastes have shown that ruthenium volatility is high for nitric acid concentrations above 2M (Christian and Pence 1977; Buelte and Chapman 1979). Buelte and Chapman showed that ruthenium volatility can be reduced to 5% by denitrating the feed in the preparation tank with stoichiometric proportions of formic acid. This feed-preparation step is carried out in a batch process and involves analyzing the waste and adding formic acid and glass formers. Each batch is equivalent to the amount of feed required for one 8-h operating shift (1200 L). The analysis determines the specific gravity and acid molarity of the combined HLLW, ILLW and recycled condensate. Formic acid is then added to denitrate the nitric acid to an equilibrium concentration of 0.5M, releasing CO_2 , NO_x and steam to the effluent system. Denitration takes place at 95°C to control the reaction rate (Bradley and Goodlet 1972). After the reaction is complete, the glass formers are added to the agitated feed tank to produce a glass-former-to-waste-oxide ratio of 2:1.

In the power waste flowsheet, there is also a high concentration of NO_x in the off gas from the decomposition of nitric acid and nitrate salts. Because of the chemical toxicity of NO_x , the quantity must be reduced to within legal limits in a NO_x destructor where ammonia reacts to form nitrogen and water. Ammonia requirements are based on the assumption that all NO_x is in the form of NO_2 .

Off-gas decontamination in the power waste flowsheet is based on operational limits defined by the NRC (Nuclear Regulatory Commission Standards for Protection Against Radiation 1979) and by EPA regulations (Environmental

Radiation Protection Standards for Nuclear Power Operations 1979), which are discussed in more detail in Appendix C.

The constituents of a typical waste glass from this power waste flowsheet are given in Table 3. Note that the melter produces 254 kg of glass/MTUWE or 84.7 L of glass/MTUWE. The waste glass has a specific gravity of 3.0 and a heat generation density of 31.3 W/L.

TABLE 3. Power Waste Glass Constituents

<u>Constituents</u>	<u>g/L of Glass</u>	<u>kg/MTHM</u>
Waste		
Inerts	480	40.6
Fission products	377	32.0
Actinides	143	12.1
Subtotal	1000	84.7
Glass formers		
Na ₂ O	240	20.3
K ₂ O	60	5.1
SiO ₂	1060	89.8
B ₂ O ₃	280	23.7
TiO ₂	180	15.2
CaO	60	5.1
CuO	90	7.6
Al ₂ O ₃	30	2.5
Subtotal	2000	169.3
Total	3000	254.0

DEFENSE WASTE FLOWSHEET

The generic defense waste flowsheet for this safety assessment is typical for 5-yr-old wastes generated from the United States' defense program. The feed compositions, activity levels and processing rates were taken from a data summary for a defense waste processing facility at Savannah River Laboratory (Landon 1980). The design processing rate is 116 kg of glass per hour, which

is 33% more than the required average annual processing rate. For a 17.8-wt% concentration of solids (excluding glass formers), the high-level waste must be processed at 225 L/h. Although achieving this rate for a liquid-fed melter of the size described in this report is conceivable with adequate boosting techniques (e.g., lid heaters), it has not been demonstrated. Therefore, for this safety assessment, it is assumed that two melters would be required to meet the daily rate commitment.

Since two melters would be required to meet the daily rate commitment, only one-half of the design processing rate is considered in this safety assessment; thus, HLLW must be processed at a rate of 112 L/h. Assuming the same recycled-condensate-to-feed ratio as in the power waste flowsheet, the recycled condensate flow rate must be 10 L/h. Finally, assuming 720 kg of frit is added for every 1000 kg of glass produced and assuming that the theoretical specific gravity of the glass formers is 2.5, the volume increase associated with the glass formers is 16.2 L/h. Therefore, the overall processing rate is 138 L/h, which is less than that assumed for the power waste flowsheet. Table 4 lists the constituents and feed rates of the defense waste feed as it enters the solidification facility.

In addition to the conservative assumption that two melters would be used, this safety assessment assumes that the defense high-level waste is only aged for five years, which is considerably shorter than the actual average age of defense wastes stored in this country. The radionuclide activity for 5-yr-old waste is 113 Ci/L, which is considerably lower than the 780-Ci/L level for power waste. Appendix B contains a more detailed defense waste flowsheet for the LFCM process.

TABLE 4. High-Level Defense Waste Feed Constituents and Feed Rates

Constituents	HLLW Feed Rate, kg/h(a)
H ₂ O	104
NaNO ₃	0.0918
NaNO ₂	0.0345
NaAlO ₂	0.0368
NaOH	0.208
Na ₂ CO ₃	0.0150
Na ₂ SO ₄	0.0195
Other salts	9.59 x 10 ⁻⁴
Na-EDTA	1.11
FeOH ₃	9.18
Al(OH) ₃	2.12
MnO ₂	1.97
UO ₂ (OH) ₂	0.686
Ni(OH) ₂	1.04
CaCO ₃	0.900
Hg(OH) ₂	0.325
HgI ₂	0.914
Other solids	1.75
Zeolite	1.51
SiO ₂	0.455
Total salts and solids	22.4

(a) Average flow.

THE SAFETY ASSESSMENT METHODOLOGY

The methodology used in this safety assessment was to define all off-standard and accident conditions unique to the generic LFCM vitrification process and to determine the consequences of those accidents. The consequences are described qualitatively for all accidents, but only representative accidents were quantitatively analyzed in terms of radioactivity releases.

Some assumptions were made to simplify the scope of this assessment. First, this safety assessment is an evaluation of the HLLW solidification process and process cells independent of other associated processes and facilities. However, in the case of defense wastes, or in the event commercial reprocessing is resumed, the solidification process will be part of a larger operation consisting of spent-fuel storage, spent-fuel reprocessing, HLLW storage, HLLW solidification and waste canister storage. Since many of the other processes have larger inventories of waste, a safety analysis of the entire facility may indicate that the most severe design-basis accident(s) will result from processes other than the solidification process.

Another simplifying assumption is the disregarding of the defense waste flowsheet in hypothesizing and analyzing accidents. As discussed in the previous section, because of the relatively low activity of defense wastes, in all cases the consequences of off-standard conditions for defense wastes were judged to be less severe than those for power wastes. In addition, no dissimilar off-standard conditions could be identified for defense wastes, except mercury releases, and short-term releases of the amount of mercury processed during defense waste solidification would have insignificant consequences.

The accidents for the power waste flowsheet were categorized according to the severity of their consequences (minor, moderate or severe) in terms of radionuclide release and extent of equipment damage. No attempt was made to assign probabilities to any of the accidents other than to indicate whether they were relatively frequent, infrequent or highly unlikely events.

The accidents were grouped further by type of accident. Then an estimate of the release term for a representative accident in each subgroup was calculated. The radionuclide source term for the accident resulting in the largest radionuclide release in each group is known as the bounding source term and is conservatively assumed to reflect every accident in that group. In keeping with the philosophy of nuclear reactor safety analyses, one engineered safeguard was assumed to be unavailable for severe accidents. However, all combinations of one unavailable engineered safeguard were not considered. The bounding source term was used to analyze the consequences of radionuclide release from the stack in terms of a site-boundary dose. Because this method is very conservative, the consequences and radionuclide releases for most accidents are less than those selected for analysis.

ACCIDENT DEFINITIONS AND CLASSIFICATIONS

The accidents were grouped according to the following three categories:

1. Minor accidents or off-standard conditions--relatively frequent, rapidly recoverable process interruptions with potential for minor radionuclide release;
2. Moderate accidents--infrequent events that could potentially result in some radiation release outside the facility or equipment damage that would require significant recovery time;
3. Severe or design-basis accidents--highly unlikely events with the potential for significant radiation release or major equipment damage; these accidents may approach the limits of the engineered safety features that serve to contain radioactive releases. The resulting releases may approach the guideline exposure limits in the Nuclear Regulatory Commission Reactor Site Criteria (1979).

As mentioned earlier, the minor, moderate and severe categories of accidents were further subdivided by type of accident so that the worst-case accident in each subgroup could be analyzed for radionuclide release. These subgroups are:

- the accidents characterized by a release or spillage of liquid waste from the process equipment into the cell,
- those accidents characterized by the release of molten glass to the cell,
- those accidents characterized by process pressurizations and release of process gases into the cell,
- those accidents characterized by a failure of an off-gas system component resulting in higher than normal release to the stack,
- those accidents characterized by process cell damage, such as ventilation HEPA filter failure.

The tables in this section contain the power waste flowsheet accidents, their causes and consequences, and the safeguards that help mitigate the

consequences. In presenting the information in these tables several general guidelines were followed. First, common mode failures were generally not addressed since they depend heavily on the final design for the cell services, such as electrical, compressed air, cooling water, and steam. (A common mode failure analysis for combined failure of primary and backup systems will certainly be warranted for a specific facility.) Secondly, the tables do not emphasize the safety features that reduce the probability of an accident occurring, only the safeguards that help to reduce the effects. Examples of safety features that are not discussed include double process containment, electrical-mechanical interlocks, limit switches and administrative controls such as quality assurance, operator training and detailed operating procedures. These safety features will be applied in the final design in a cost-effective manner to reduce the probability of those accidents with the more severe consequences. Finally, in describing the consequences an attempt was made to estimate the potential radionuclide release at worst-case while at the same time reflecting the realistic consequences of the accident.

While the consequences of minor and moderate accidents will be important in the plant design and operation, severe accidents will receive significant attention in a review of plant safety. The severe accidents represent the design-basis accidents for the solidification plant. During the history of nuclear reactor licensing, a standard set of safety challenges that are accepted as design-basis accidents has evolved. Examples of these are double-ended pipe breaks in the main coolant lines and extremely powerful tornados and earthquakes. While licensing of fuel reprocessing and waste solidification plants does not have the extensive precedents of nuclear reactors, some accidents have become standard because of their frequent inclusion in similar safety studies. For example, the design-basis tornado and the design-basis earthquake (also referred to as the safe-shutdown earthquake) are commonly analyzed.

Design-basis accidents associated with reprocessing plants include red-oil (nitrated forms of degraded solvents) explosions in tanks and nuclear criticalities in tanks and ion-exchange columns. The red-oil explosion was included in this analysis. A criticality in a HLW tank was not considered since the

majority of plutonium is stripped from the HLLW and neutron poisons are added. In addition, the potential of a criticality is higher in a reprocessing plant and therefore is not unique to the LFCM process. (The possibility of a criticality in the glass tank of the melter was considered in Appendix D despite very strong arguments that the prerequisites for this event could not occur.) An ammonium nitrate explosion was not considered in this assessment because of the inconceivability of forming large quantities of crystalline ammonium nitrate downstream of the NO_x destructor and of initiating the reaction by a shockwave.

Another energetic accident, one that is unique to the solidification process, was also considered. It is the steam explosion, which is caused by a thermal reaction between molten glass and water. In a steam explosion the energy yield could be very large, but again there are substantial theoretical arguments and experiences that indicate the prerequisites for such an interaction could not occur (Postma, Chapman and Buelte 1980; see Appendix A).

The principal reason for including severe accidents despite substantial evidence that they would not occur is to serve as design-basis accidents against which the process facility could be tested. Follow-on safety analyses for specific facilities will probably include specific shock wave and pressure time histories and structural responses to these accident assumptions.

The minor accidents are described in Table 5, the moderate in Table 6, and the severe in Table 7.

TABLE 5. Minor Accidents and Off-Standard Conditions

Accident Description	Events or Sequence of Events Leading to Accident	Engineered Safeguards	Consequences
<u>LIQUID</u>			
Leakage in a feed tank transfer line.	Pipe corrosion; Weld failure.	Cell sump activity detector; Cell drainage and alarm system; Cell HEPA filters.	Estimated that maximum of 200 L leaks before corrected by operator; Assume it all evaporates.
Evaporator overflows.	Evaporator heater failure; Failure of tank level indicator and alarm;	Cell sump activity detector; Cell drainage system; Cell HEPA filters.	Estimated that maximum of 20 L of evaporator bottoms spilled before corrected; however, evaporator overflow would almost universally flow into second evaporator or condensate tanks where radiation detectors are present.
Feed tank or feed-preparation tank overflows.	Failure of tank level indicator and alarm; Unintentional water addition to feed tank; Cooling coil leaks into HLLW; Steam coil rupture.	Cell sump activity detector reduces response time; Cell drainage system; Cell HEPA filters.	Estimated that maximum of 200 L of HLLW spilled on feed cell floor before corrected; Assume it all evaporates; Detailed design may call for containment of these types of spills so that no consequence occurs.
<u>GLASS</u>			
Canister dropped during transfer.	Failure of canister lifting device or overhead crane.	Cell HEPA filters.	Estimated spill of 50 L (~1/2 canister) of 1000°C glass onto cell floor.
Loss of control on glass air lift.	Operator inattention and failure of canister overflow alarms; Control valve failure.	Canister seal to overflow; Glass rate reduces as melt level is lowered.	Estimated spill of 50 L (~1/10 melter) of 1000°C glass onto cell floor.

TABLE 5. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>PROCESS PRESSURIZATION</u>			
HCOOH added at excessive rate.	Operator fails to comply with procedures; Control valve failure.	Off-gas system is oversized to accommodate steam surges from the melter and larger-than-normal air leaks; Cell drainage system; Cell sump activity detector; Cell HEPA filters.	Too rapid HCOOH addition leads to an uncontrolled exothermic reaction that causes excessive gas evolution and may pressurize a feed tank and cause HLLW spillage; Assume 20-L spillage.
Excessive HCOOH added to feed tank.	Operator error and tank sample analysis in error; HCOOH flow control in error.	Off-gas system has reserve DF capacity; Off-gas system has reserve vacuum capacity.	May react rapidly in melter or evaporator with nitrates; Assume 200-L spill from evaporator and 10-min vapor production from melter (25 kg).
Feed tank steam-heating coil leaks.	Coil ruptured by corrosion; Coil ruptured by erosion; Coil ruptured by mechanical failure of pump or agitator.	Off-gas system instruments detect steam surge; Off-gas system can accommodate steam surges; Cell drainage system; Cell sump activity detector; Cell HEPA filters.	Pressurization of tank may entrain HLLW through tank vent or failed gasket; Assume spill of 200 L.
Ruthenium buildup in off-gas system lines.	RuO ₂ plateout in off-gas system lines.	Detected by off-gas system instruments since it occurs gradually for rapid compensation; Automatic feed shutdown; Cell HEPA filters.	Assume 10 min of melter vapor production is vented to cell (25 kg).

TABLE 5. (contd)

Accident Description	Events or Sequence of Events Leading to Accident	Engineered Safeguards	Consequences
PROCESS PRESSURIZATION (contd)			
Melter pressurization due to loss of steam condensation units.	Loss of cooling to venturi-ejector tank; Loss of venturi-ejector recirculation pump; Loss of cooling to condenser.	Gradual occurrence and detected by off-gas system instruments; Extra flow capacities in venturi and condenser; Automatic feed shutoff; Melter pressure relief.	Assume pressurization results in 10 min of melter vapor release to cell atmosphere (25 kg).
Loss of off-gas system blowers.	Mechanical failure; Failure of check valves in parallel blower; Mechanical damage by one blower to the adjacent blower; Loss of all electrical power, including emergency power.	Cell HEPA filters; Venturi-ejector creates vacuum; Automatic feed shutdown.	Assume 10 min of melter vapor production is vented to cell (25 kg).
Small water-glass steam explosion.	HLLW feed preheating fails so that feed temperature is significantly lowered below 90°C while satisfying all other steam explosion criteria. See Postma, Chapman and Buelt (1980) for a more detailed discussion.	Oversized venturi-ejector and condenser; Cell HEPA filters; Melter has pressure relief to cell; Automatic power shutdown.	Assume 1 min of melter vapor production is vented to cell (2.5 kg).
Order-of-magnitude increase in cold cap vaporization.	Melter accumulates excessive cold cap from excessive over-feeding and, coincidentally, breaks up to produce a steam surge.	Oversized venturi-ejector and condenser; Cell HEPA filters; Melter has pressure relief to cell; Automatic feed shutdown.	Assume 10 min of melter vapor production is vented to cell (25 kg).
Melter pressurizes due to large pressure drop in off-gas system.	Flooded condenser; Wet HEPA filters; Line pluggages.	Melter has pressure relief to cell; Cell HEPA filters; Automatic feed shutdown.	Assume 10 min of melter vapor production is vented to cell (25 kg).

TABLE 5. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>PROCESS PRESSURIZATION</u> (contd)			
Breach of containment box above glass level.	Weld failure due to thermal stresses or corrosion; Gasket failure from vapor attack or excessive off-gas system vacuum; Damage from overhead crane operation.	Excess off-gas system blower capacity; Cell HEPA filters; Automatic feed shutoff on loss of vacuum; Limited liquid holdup in melter (<20 L).	Assume 10 min of melter vapor production is vented to cell (25 kg).
Power failure.	Power supply interruption.	Automatic feed shutoff; Backup power supply to off-gas equipment.	Assume 1 min of melter vapor production is vented to cell until backup power supply re-energizes blower and scrubber; Glass in melter freezes in 12 h but no safety implications.
<u>OFF-GAS SYSTEM</u>			
No HCOOH added to feed tank.	Operator error and tank sample analysis in error.	Off-gas system has reserve DF capacity.	Ruthenium DF in the melter drops from 20 to 5.
Excess air inleakage.	Failed gasket; Crack in process system piping or equipment.	Off-gas system instruments; Oversized off-gas system and blowers; Cell HEPA filters.	Small drop in adsorber bed DF; System more susceptible to pressurization from melter steam surge.
Feed tank boiling.	Boiling point lowered due to failure of tank vent orifice causing excessive vacuum on feed tank; Loss of cooling; Loss of temperature control; Operator error.	Feed tanks are vented to the off-gas system; Off-gas system is oversized.	No immediate consequences.

TABLE 5. (contd)

Accident Description	Events or Sequence of Events Leading to Accident	Engineered Safeguards	Consequences
<u>OFF-GAS SYSTEM</u> (contd)			
Evaporator boiled dry.	Tank level malfunction; Operator error; Excessive purge.	Multiple evaporative stages to clean up liquid effluent; Opportunity to recycle this effluent back to feed tanks.	Entrainment of 10^{-4} particulates and 10^{-2} ruthenium in volatile form adds negligible activity to off-gas system flow.
Loss of cooling water to condenser.	Plugged line; Line rupture; Radionuclides detected in cooling water requires immediate cooling water shutdown.	Venturi-ejector provides principal condensation; Off-gas system has a DF margin.	Loss of DF provided to condenser; System may pressurize if large steam surges occur in the melter.
Loss of off-gas system heater control.	Malfunction in temperature control system. This must take place at low processing rates since power limits at design rates would prevent overheat.	Off-gas system instruments will alert operators; Off-gas system has a DF safety margin.	Loss of DF in temperature-sensitive adsorber beds; Ruptured HEPA filter due to high gas temperature.
HEPA filter rupture.	Heater over temperature; Water carryover past condenser; Off-gas system blower overspeed; Off-gas system pressure pulse.	HEPA filters have backup; Off-gas system has a DF margin.	Above normal release of activity until corrected.
Ruthenium adsorber bed overheats.	Loss of temperature control on off-gas heaters. This must occur at low processing rates since power limits would prevent overheat at design capacity.	NO_x destructor provides additional ruthenium DF.	DF on silica gel bed decreases with increasing temperature.
Moisture in ruthenium adsorber bed.	Loss of temperature control on off-gas system heater; Water carryover past condenser.	NO_x destructor provides additional ruthenium DF.	Water may elute the ruthenium from the silica gel bed; The ruthenium that escapes the NO_x destructor may be released to the stack.

TABLE 5. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>OFF-GAS SYSTEM</u> (contd)			
Chemical contaminants degrade ruthenium adsorber bed.	The surface chemistry of ruthenium adsorption has not been completely described; Degraded performance has been observed with silica gel.	NO _x destructor provides additional ruthenium DF.	Above normal release of ruthenium occurs until stack monitoring detects the off-standard condition.
Iodine silver zeolite bed overheats.	Loss of off-gas system heater temperature control at low processing rates.	None.	Possible reduction in DF.
Moisture in iodine silver zeolite adsorber bed.	Water carryover from condenser; Off-gas system heater malfunction.	None.	Increases DF.
Contaminants in iodine silver zeolite adsorber bed.	Contamination has been observed in the laboratory.	None.	Possible reduction in DF.
NO _x destructor bed too hot.	Heater control malfunctions.	Cell HEPA filters.	Ammonium nitrate crystals may be formed in bed; Inconceivable chance of explosion however.
NH ₃ flow out of tolerance.	Malfunction in control system; Operator error.	Off-gas system has ruthenium DF margin.	Ruthenium DF may be reduced; Additional NO _x released to stack.
Ruthenium adsorber bed dropped during change-out.	Operator error.	Cell HEPA filters.	Silica gel spills onto cell floor; No desorption occurs.
Iodine adsorber bed dropped during change-out.	Operator error.	Cell HEPA filters.	Silver zeolite spills onto cell floor; No desorption occurs.

TABLE 5. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>OFF-GAS SYSTEM</u> (contd)			
Melter feed line plugged.	Intermittent feeding leads to sludge buildup and dryout in line; Failure of water-cooled tip during startup.	Process indications during gradual flow reduction would detect pluggage and minimize consequences-- these are a melter lid temperature increase, off-gas flow and pressure drop changes, and glass flow reduced or stopped; HLLW would flow to evaporator and undergo multiple cleanup cycles; Activity monitors in evaporators will help detect occurrence.	HLLW would be pulled through air vent into off-gas system condenser; Assumed to occur for 10 min before detection.
<u>PROCESS CELL</u>			
Contamination enters cooling water.	Break in cooling coils in the feed tanks, condenser or evaporator system.	Activity detector in cooling system; Anticipated in design so that any contamination is isolated or contained.	Repair required.
Contamination enters service steamline.	Break in steam coils in the feed tanks or evaporator.	Activity detector in steam system; Anticipated in design.	Repair required.
Contamination enters air supply system.	System design not complete enough to specify, though unlikely because of pressurized air supply.	Activity detectors in piping and equipment areas.	Primarily occupational exposure to operational personnel; Minor release to biosphere limited because of cleanup.
Major equipment replacement.	Decontamination, encapsulation and removal of contaminated equipment such as a melter may cause small contamination release.	Hot maintenance cell and hot storage area are negative pressure and ventilation is discharged through HEPA to stack.	Occupational exposure to workers during contamination cleanup; About 4 kg of powdered glass released to cell atmosphere.

TABLE 5. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>PROCESS CELL</u> (contd)			
Canister failure during storage.	A weld failure on a canister stored in a pool.	Pool water cleanup system; Pool building ventilation goes to stack.	Occupational exposure during contamination control.
Electrical shock from contact with manipulator.	Operator protrudes electrical shield; Major equipment damage from severe accident causes failure of electrical shields.	Contact with one electrode has no shock hazard. Must contact at two points at different potentials (i.e. opposite electrodes, electrode and glass thermowell, or ground and electrode if containment box has existing electrical short to another electrical potential).	No effect if all conditions are not satisfied; No effect on radionuclide release.

TABLE 6. Moderate Accidents

Accident Description	Events or Sequence of Events Leading to Accident	Engineered Safeguards	Consequences
<u>LIQUID</u>			
Feed tank rupture.	Tank weld fails; Pump impeller or agitator impeller fails and ruptures tank; Corrosion; Erosion.	Cell sump activity detector; Cell drainage system; Cell HEPA filters.	Assumed that complete tank (1200 L) drains to cell sump.
Break or major leak in in venturi recirculation line.	Weld failure; Damaged by overhead crane operation.	Cell sump detector; Cell drainage system; Cell HEPA filters; Off-gas system instruments; Visual observation.	Assume 10 min of leakage before correction (1140 L) causes maximum humidity and aerosol in vitrification cell.
Excessive, undetected overfeeding for prolonged period.	Operator error; Loss of feed air-lift control and operator ignores warning signs.	Drastic change in electrical resistance between electrodes; Plenum temperatures decrease; Excessive rate in level reduction in feed tank; Excessive glass production rate; Sealed box construction helps prevent liquid spills.	Small liquid spill (10 L) through seals in melter; May cause process pressurization; May require melter restart.
Prolonged blockage of glass flow through overflow.	Spinels or other crystalline forms collect on floor and plug riser; Inadequate overflow heat supply causes collection of frozen glass at overflow pour tip.	Change in electrical resistance between the two electrodes and no glass production helps detect situation; Sealed box construction helps prevent spills.	Glass production flow ceases and melt level rises; Assume small liquid spill (10 L) through seals in melter; Requires shutdown and high overflow and melt temperatures to correct problem.

TABLE 6. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>GLASS</u>			
Canister overfilled.	Operator error; Failure of canister weight and temperature monitor system; Melter pressurization and pluggage of equilization line; Unplugging of temporarily plugged riser or overflow.	Cell floor lined with stainless steel; Cell HEPA filters.	Estimated spill of 10 L of 1100°C glass when canister is disconnected.
Canister ruptured during filling operation.	Undetected, improper canister seam weld.	Cell floor lined with stainless steel; Cell HEPA filters.	Estimated 50 L (~1/2 canister) of 1100°C glass is spilled.
<u>PROCESS PRESSURIZATION</u>			
Moderate water-glass steam explosion.	HLLW preheating fails while satisfying all other criteria. See Postma, Chapman and Buelt (1980) for a more detailed discussion.	Oversized venturi-ejector and condenser; Cell HEPA filters; Automatic feed shutdown.	Assume 40 L of 1100°C glass is spilled and 25 L of HLLW vaporized to the cell; Gasket failure to melter or other minor damage.
Extensive glass foaming fills melter head cavity and escapes to cell through pressure relief.	Abnormal glass melting behavior not detected by laboratory screening and analysis.	Cell HEPA filters; Automatic feed shutoff upon loss of vacuum; Stainless steel-lined cell.	Assume 25 L of HLLW in cold cap is vaporized to the cell and 36 kg glass foam is released to cell through pressure relief vent, breach in canister connection, and gaskets in melter lid.
<u>OFF-GAS SYSTEM</u>			
Breach in off-gas system line	Equipment drop or weld failure leads to complete off-gas system breach and line displacement.	Off-gas system instruments; Cell HEPA filters.	Assume 10 min of melter vapor production (25 kg) is vented to cell.

TABLE 6. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>OFF-GAS SYSTEM (contd)</u>			
All cooling to off-gas system components lost.	Plugged line; Line rupture; Radionuclides detected in cooling water; Operator error; Steam from melter is not condensed and goes through off-gas system.	Off-gas system instruments; Melter has pressure relief to the cell.	Increased activity in the off-gas system plus venting of melter vapors to cell; Assumed to occur for 10 min before corrected.
HEPA filter dropped during change-out.	Operator error and protective bag fails when dropped.	Cell HEPA filters.	Assume 1/2 solids loading on HEPA filters is dispersed to cell.
<u>PROCESS CELL</u>			
Equipment or canister transfer breaks HEPA filter.	Overhead crane hook or cable breaks; Crane controls fail in unsafe mode.	Detection of failure by visual observation, high activity level alarm in ventilation duct, and differential pressure meters; Backup filters.	Some additional occupational exposure due to cleanup; Some cell contamination (assume 1 kg calcine) may be swept into ventilation duct.
Fire in the cell.	Electrical cable insulation or decontamination supplies burn and breach cell HEPA filters.	Sprinkler system in cell; Ventilation passes through building HEPA enroute to stack.	Some additional occupational exposure during cleanup; Cell contamination may be swept past ruptured HEPA filter (assume 1 kg calcine).

TABLE 7. Severe Accidents

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>LIQUID</u>			
Feed tank explosion.	Detonation of red oil (would require coincidental inclusion of significant quantities of the degraded solvent plus loss of agitation plus boiling of feed tank for prolonged period); Foaming or reboil causes glass overheat and automatic power shutdown fails.	Feed tank ability to contain or dissipate detonation energy; Cell drainage system; Cell HEPA filters.	Event assumed to create maximum aerosol saturation in the feed cell (220 L = 31% MTUWE). Cell HEPA failure assumed as a single-failure criterion.
<u>GLASS</u>			
Breach of melter containment box below glass level.	Ruptured by overhead crane operation; Unusually corrosive glass that has gone undetected in feed and process analysis; for prolonged period. Operator inattention and failure of glass and/or electrode overtemperature alarms.	Glass must penetrate three layers of refractory in sufficient quantity to heat box <800°C for extended period of time; Melter temperature instruments; Operators can drain melter through bottom drain into glass sump tank; Stainless steel-lined cell; Cell HEPA filters.	All or part of melter glass (1370 kg = 500 L) spills to cell floor; Release to cell atmosphere due to some volatile radionuclides in glass assumed to be at 1150°C for 30 min; Assume cell HEPA ruptured as single-failure criterion.
Criticality in melter tank.	Preferential settling out of PuO ₂ in melter tank over prolonged operation (See Appendix D for analysis that indicates this accident is impossible).	Neutron detectors in cell; Cell HEPA filters.	Foam-up of glass in melter; This accident release not calculated since there is no evidence criticality could occur in the melter.

TABLE 7. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>PROCESS PRESSURIZATION</u>			
Severe water-glass steam explosion ruptures the melter.	HLLW feed preheating fails while fulfilling all other criteria. See Postma, Chapman and Buelt (1980) for a more detailed discussion.	Massive cell boundary; Stack HEPA filters.	Complete rupture of the melter containment box would release all glass (~500 L) plus disperse the calcine cold cap (~10 L) plus generate approximately 124 kg of small glass particulates due to glass-water reaction. The cell and cell windows are massive and are expected to survive any missiles. The cell HEPA filters may also be protected in the final design, but rupture is assumed to be consistent with the single-failure criterion.
<u>OFF-GAS SYSTEM</u>			
(No severe accidents have been identified.)			
<u>PROCESS CELL</u>			
Design-basis tornado.	A tornado having the characteristics defined in Atomic Energy Commission (1974) Regulatory Guide 1.76 is assumed to rupture one set of HEPA filters in the plant ventilation system.	Plant designed to withstand this event with only minor damage	Assume melter operation continues for 10 min after design-basis tornado with a release of the melter vapor to the cell atmosphere. The second set of ventilation HEPA filters is assumed to be ruptured to meet the single-failure criterion.

TABLE 7. (contd)

<u>Accident Description</u>	<u>Events or Sequence of Events Leading to Accident</u>	<u>Engineered Safeguards</u>	<u>Consequences</u>
<u>PROCESS CELL</u> (contd)			
Design-basis earthquake.	The plant will be designed to withstand an earthquake sized according to site history with only minor equipment damage and without breaching the cell boundary.	All ventilation inlet and outlet ducts have HEPA filters.	Assume 10 min of feed (25 L) is spilled to floor, 10 min of melter vapor pro- duction (25 kg) escapes to the cell, and filled canis- ter is over-turned spilling 50 L of glass. Assume the cell HEPA filter is breached to be consistent with single- failure criterion.

RADIONUCLIDE RELEASES FOR EACH ACCIDENT SUBGROUP

The classification of accidents according to severity and type was described in the previous section. In this section, the calculated release term for each subgroup of accidents is given. The release terms are based on the operating parameters of the process, but many assumptions were also required to calculate the release terms. The assumptions are intended to be conservative in that they maximize the accidents' consequences. However, again the intent was to describe the accidents realistically. A discussion of the major assumptions is presented first followed by a brief summary of the calculation for each accident subgroup.

GENERAL ASSUMPTIONS FOR ANALYSIS OF RADIONUCLIDE RELEASES

High-level liquid waste contains a large number of elements. Except for a few nuclides, it is assumed that the radionuclides maintain their relative proportion to each other whether in a liquid or solid state. Therefore, in this release term analysis most of the radionuclides are referred to collectively as particulate.

There are two exceptions to the above assumption. First, during molten glass spills some radionuclides will volatilize to the cell atmosphere. Second, ruthenium can form volatile gas species, especially when a waste solution with a high nitric acid concentration is heated. These volatile elements were taken into consideration when calculating the release terms for the accident subgroups.

Gray (1976) has measured the volatility of cesium, molybdenum, tellurium, rubidium and ruthenium from molten glass as a function of temperature. Gray found that the volatility of these elements increases exponentially with the temperature of the glass. However, with the possible exception of ruthenium, all of these volatile elements condense and form micron-sized particles that can be trapped by the HEPA filters. Preliminary calculations indicated that only cesium volatilizes in quantities large enough to warrant a separate release term. However, as a conservative measure this analysis assumes that ruthenium forms volatile gas species that do not condense in time to be

trapped by the HEPA filters. Therefore, for the molten glass spills a cesium release term and a ruthenium release term were calculated.

Ruthenium volatility must also be considered in all of the other accident calculations since the formation of volatile gas species (RuO_4 and a nitrosyl ruthenium complex; Christian 1976) can occur under some special conditions. During normal operation a ruthenium adsorber bed in the off-gas system removes any volatile ruthenium. However, under accident conditions the volatile gases may be released to the cell atmosphere where they are assumed to be entrained in the cell ventilation flow. This accident analysis assumes that this entrained volatile ruthenium passes untrapped through the ventilation system's HEPA filters. This may be a realistic assumption for the gaseous nitrosyl ruthenium complex which is formed under the high nitric acid concentration conditions, but it is probably overly conservative for RuO_4 which is often formed when a waste solution is undergoing drying. During experiments RuO_4 has been observed to plate out in metal process lines and in ventilation ducting. It also readily decomposes to its particulate form, RuO_4 , at room temperatures.

Another consideration when calculating the radionuclide release terms is determining how much of the volatile elements is released to the cell atmosphere. For the molten glass accidents the volatile release rates measured by Gray (1976) were used. It is necessary to integrate the release over time, temperature and surface area to accurately predict the total release. For these release term calculations a simple, but conservative model was applied. The spill was assumed to produce a thin glass pool that remains at the initial glass temperature for 30 min, at which time no additional volatile release occurs. (The volatility data was based on a slightly different glass composition than the one used as the basis of this study, but any variations are judged to be within the accuracy of the other assumptions required.) The volatility data for molten glass is given in terms of weight loss per unit area per unit time. To convert this into MTUWE of a particular isotope, it was necessary to find the ratio of the radioactive isotopes for each element. For cesium, the Cs-134 isotope is 2% and the Cs-137 isotope is 46%. For ruthenium, the Ru-106 isotope is 0.7%. The other isotopes of these elements are either stable or have negligible activity.

The calculations of release terms for liquid waste spills and melter off-gas releases to the cell atmosphere require assumptions regarding the entrainment of particulates (all radionuclides, including ruthenium) and the volatilization of ruthenium. Liquid waste releases would be either from the feed system, the venturi scrubber or the evaporator. The release can take the form of a spill onto the cell floor or a spray into the cell atmosphere. For spills the radionuclides become mobile through entrainment during evaporation. Entrainment rates for particulates are reported as less than 10^{-4} (Christian and Pence 1977). Ruthenium is generally entrained at the same rate as particulates for neutralized solutions of HLLW. However, volatilization has been observed to predominate at high nitric acid concentrations. Above $8M$ HNO_3 the volatilization rate of Ru-106 may approach 10^{-1} . In measurements on simulated waste compositions, ruthenium total loss factors of up to 1.5×10^{-3} were obtained with $6M$ - HNO_3 concentrations (Hanson 1980). Therefore, those liquid releases involving HLLW spills ($7M$ - HNO_3 maximum) assume the conservatively high release rate of 10^{-2} of a volatile form of ruthenium, most likely a nitrosyl ruthenium gas complex (Christian 1976). The exceptions to these assumptions are the entrainment values for the melter itself. Based on operating experience the entrainment of all radionuclides including cesium, but excluding ruthenium, into the melter off-gas will not exceed 5×10^{-3} for normal operating conditions. The entrainment of ruthenium as volatile species is conservatively assumed to be 5×10^{-2} .

There is one additional volatile element that was considered in the safety assessment. There is a significant amount of iodine in spent fuel and it is very volatile. However, by the time the spent fuel is reprocessed the majority of the iodine has decayed to stable isotopes. The remaining radionuclide is I-129. The long-term buildup of I-129 in the environment from an expanded nuclear fuel cycle may be a problem, and for that reason the EPA requires that most of the iodine be trapped and immobilized (Environmental Radiation Protection Standards for Nuclear Power Operation 1979). Most of this iodine control will take place in the reprocessing plant since flowsheets generally indicate 99.5% release during reprocessing. There are only 1.7×10^{-4} Ci of I-129 in one MTUWE of HLLW. Release of several MTUWE of iodine would not cause significant exposure at the site boundary; therefore, a separate release term for

iodine was not calculated. This accident analysis did address the concern over the possibility that the loaded iodine adsorber bed could be exposed to some type of accident conditions that would release many MTUWE of iodine. However, no viable accident condition appeared capable of releasing the chemically adsorbed iodine on a silver zeolite adsorber bed. Rejuvenation of this type of bed requires passing hydrogen at approximately 400°C to 500°C through the bed to strip off the iodine. However, if the bed is pushed beyond capacity, any physically adsorbed iodine can be relatively easily released by an increase in gas temperature or by bed washing. In future, more advanced safety analyses this potential accident should be examined more closely, especially if normal operation requires the rejuvenation of the iodine adsorber bed.

Another assumption required to calculate the release terms is the model for evaporation and entrainment of liquid releases. Pool releases were modeled by conservatively estimating how much liquid could remain on flat surfaces in the process cell and be completely evaporated. All the liquid that was evaporated was assumed to give off particulate and ruthenium source terms equivalent to the entrainment factors described earlier. For accidents that are characterized by a release of liquid under pressure, there is the potential that a fine aerosol will be formed. A typical line break is more likely to result in a jet or stream of liquid that merely forms a pool of liquid. On the otherhand, if it were possible to have a leak exhibiting a great deal of atomization, the evaporation of these fine droplets would be greatly increased and some entrainment of the dried aerosols and the mist into the ventilation flow may be expected. However, it is difficult to produce atomization of a significant amount of liquid, as has been shown in the spray calciner development program at PNL (Larson and Bonner 1976), and it would be unlikely that an accident condition could produce a significant amount of aerosol. The settling rates for liquid droplets range from 10 to 75 cm/s for 70- to 200- μ m-dia particles (Perry and Chilton 1973). The model used in this analysis assumes that while a spray of liquid continues it will evaporate at the maximum possible rate and there will be 100% entrainment of the evaporated liquid. The maximum evaporation assumes that the ventilation air enters the cell at 0% humidity and exits at 100% humidity at 50°C, producing a moisture content of

22 m³/L
131 g/m³ (Perry and Chilton 1973). The ventilation flow rate in the feed cell is assumed to be 1300 m³/h. Of the total ruthenium that is entrained into the ventilation air in this form, 99% is treated as a particulate and 1% is assumed to be volatile. The normal DF given earlier for HEPA filters are assumed to apply to the particulates arising from these liquid spray accidents.

For some accidents a source term of calcine (dried HLLW) was projected. The source of this calcine would be from either the cold cap in the melter or from evaporated HLLW contamination, presumably from a small leak over a period of time onto a hot surface.

Appendix C contains a listing of all the radionuclides and their activities per MTUWE. The release terms were calculated and expressed in terms of a fraction of a MTUWE. Although this is an unorthodox unit of activity, it is compatible with the flowsheet information and can easily be converted to total curies ($6 \times 10^{+5}$ Ci/MTUWE for the 3-yr-old power waste). The tabulation of release terms is presented in both MTUWE and curies.

RELEASE TERM ANALYSES

Summaries of the release term calculations are presented below. Only one accident from each subgroup (the bounding source term) is presented, but all accidents were reviewed to determine which accident in each subgroup would result in the largest release for that group. The releases are reported in MTUWE.

Minor Accidents Involving Liquids

Description. Leakage in a feed tank transfer line spills 200 L of HLLW onto the cell floor where it dries into a sludge.

Assumptions. 10^{-4} of the particulates are entrained in the vapor and 10^{-2} of the ruthenium is volatilized. This occurs over a 70-min period. Cell and stack HEPA filters provide a DF of 1 for ruthenium and a DF of 10^8 for particulates.

Discussion. A spill of 200 L would correspond to 10 min of tank-to-tank transfer or a spill of 0.56 cm in depth on the feed cell floor. Much of this is expected to drain into the sump, but credit is not taken for this possibility. Evaporation is assumed to occur as fast as the ventilation turnover permits. The high volatilization release fraction is used for ruthenium although tank-to-tank transfer takes place after denitration, after which time the ruthenium release would be due to entrainment only and would drop to approximately 10^{-4} .

Release to Stack. The ruthenium released to the stack would be equal to 2.8×10^{-3} MTUWE. The particulate released to the stack would be equal to 2.8×10^{-13} MTUWE.

Moderate Accidents Involving Liquids

Description. A major leak in the venturi recirculation line creates an aerosol that saturates the process cell for 10 min.

Assumptions. The activity of the recirculation fluid was calculated (Appendix B) as 3.2 Ci/L due to ruthenium and 3.8 Ci/L due to particulate matter. The accident conditions and the low nitric acid concentration do not lead to high ruthenium volatility. Therefore, the volatile ruthenium is assumed to be 10^{-3} of the total. Maximum saturation of the air (131 g/m^3 at 50°C) is assumed. The cell volume is 566 m^3 and this is turned over once during the 10-min accident. With these assumptions 150 L of recirculation liquid is evaporated creating an aerosol of particulates. The DF values for a cell atmosphere release are 1 for volatile ruthenium and 10^8 for particulates.

Discussion. The expected course of such an accident would involve a small leakage that does not form an aerosol and that would undergo little evaporation before being detected and stopped and the residual spillage collected in the cell sump. It is very difficult to generate a liquid aerosol that could evaporate and create the particulate aerosol that is assumed for this accident. The volatilization of ruthenium is also expected to be much less than the 10^{-3} assumed.

Release to Stack. The ruthenium released to the stack would be equal to 1.1×10^{-5} MTUWE. The particulate released to the stack would be equal to 1.0×10^{-11} MTUWE.

Severe Accidents Involving Liquids

Description. An unstable compound of nitrated, degraded solvents termed "red oil" is formed in the reprocessing plant and is carried over to the feed tank where it detonates.

Assumptions. The danger of a "red oil" explosion is greater in the reprocessing plant, and process design and operation will reduce this possibility to an acceptable risk there. Therefore, the primary assumption is that "red oil" could collect and detonate in the feed tank. The explosion is assumed to generate the maximum saturated air in the feed cell (218 m^3). With these assumptions 28.6 L (4×10^{-2} MTUWE) of HLLW is distributed in the cell atmosphere. In addition, 200 L are assumed to dry on the cell floor as was calculated in the feed tank rupture. The failure of the cell HEPA filter is assumed in keeping with the overall safety analysis philosophy of assuming a single failure in the engineered safeguards for the severe accidents. Ruthenium volatility is assumed to be 10^{-2} since the feed could be as high as 7M nitric acid. The DF values for the stack HEPA are assumed to be 1 for volatile ruthenium and 10^4 for particulates.

Discussion. The "red oil" explosion has been included in the safety analysis, but a more thorough analysis of an actual plant design may show that the probability of this accident is negligible. The assumption of maximum evaporation and associated particulate aerosol formation is conservative, but maximizes the source term for this accident.

Release to Stack. The ruthenium released to the stack would be equal to 2.8×10^{-3} MTUWE. The particulate released to the stack would be equal to 4.0×10^{-6} MTUWE.

Minor Accidents Involving Glass

Description. A newly filled canister is upset during transfer and a portion of the glass pours onto the cell floor.

Assumptions. The canister contains 100 L (300 kg) of glass. One-half of the total (50 L) is assumed to still be molten. The average temperature of the spilled glass is 1000°C. The glass spill is 1 cm thick with a surface area of 5 m². The principal elements volatilized are cesium and ruthenium. The volatilization is modeled by assuming the glass spill remains at 1000°C for one-half hour and then cools immediately. (The choice of major volatile elements and the time-at-temperature model are discussed at the beginning of this section.) The DF factors for radionuclides released to the cell atmosphere are 10⁸ for the cesium and 1 for the ruthenium. The volatilization rates at 1000°C are 0.55 mg/cm²-h for cesium and 0.01 mg/cm²-h for ruthenium. The radionuclides of each element that contribute the principal radioactivity are: Cs-134 is 2%, Cs-137 is 46%, and Ru-106 is 0.7%.

Discussion. The possibility of a canister spill is recognized, but canister transfer equipment and procedures will insure that this event has a low probability of occurrence.

Release to Stack. The ruthenium released to the stack would be equal to 1.3×10^{-4} MTUWE. The cesium released to the stack would be equal to 6.2×10^{-11} MTUWE.

Moderate Accidents Involving Glass

Description. A canister fails during the filling operation and the newly poured molten glass spills onto the cell floor.

Assumptions. One-half the canister volume (50 L) is spilled. The glass temperature is 1100°C. The glass spill is 1 cm thick with an area of 5 m². The principal elements volatilized are cesium and ruthenium. The volatilization is modeled by assuming the glass spill remains at 1100°C for one-half hour and then cools immediately.

(The choice of major volatile elements and the time-at-temperature model are discussed at the beginning of this section.) The DF factors for radionuclides released to the cell atmosphere are 10^8 for the cesium and 1 for the ruthenium. The volatilization rates at 1100°C are $2.5 \text{ mg/cm}^2\text{-h}$ for cesium and $0.018 \text{ mg/cm}^2\text{-h}$ for ruthenium. The radionuclides of each element that contribute the principal radioactivity are: Cs-134 is 2%, Cs-137 is 46%, and Ru-106 is 0.7%.

Discussion. The canisters are thoroughly inspected before use and the duty imposed by filling with glass from a joule-heated melter is not severe. Therefore, large seam splits are an unlikely event. In addition, the glass spill and volatilization models are conservative.

Release to Stack. The ruthenium released to the stack would be equal to 2.2×10^{-4} MTUWE. The cesium released to the stack would be equal to 2.8×10^{-10} MTUWE.

Severe Accidents Involving Glass

Description. A breach of the melter containment box below the glass level results in a glass spill.

Assumptions. Entire melter content of 500 L spills onto the process floor. The glass temperature is 1150°C . The glass spill covers the 36-m^2 cell floor to a depth of 1.4 cm. The volatilization is modeled by assuming the glass spill remains at 1150°C for one-half hour and then cools immediately. (The choice of major volatile elements and the time-at-temperature model are discussed at the beginning of this section.) The principal elements volatilized are cesium and ruthenium. The rate of volatilization at 1150°C is $5.5 \text{ mg/cm}^2\text{-h}$ for cesium and $0.05 \text{ mg/cm}^2\text{-h}$ for ruthenium. The cell HEPA filter is assumed to be ruptured in keeping with the single-failure criterion for severe accidents. The stack HEPA filter provides a DF of 10^4 for cesium and 1 for ruthenium.

Discussion. The failure of the cell HEPA filter is presumed to occur from fire or heat damage and is in keeping with the assumption of a single-failure criterion.

Release to Stack. The ruthenium released to the stack would be equal to 4.6×10^{-3} MTUWE. The cesium released to stack would be equal to 4.4×10^{-5} MTUWE.

Minor Accidents Resulting in Process Pressurization

Description. Loss of cooling to the venturi-ejector and the main condenser causes pressurization of the melter.

Assumptions. The pressurization is assumed to occur for 10 min before the aqueous pool in the melter evaporates to the cell. The normal melter vapor production is assumed to be released to the cell atmosphere for 10 min and is assumed to be equivalent to the maximum estimate of the volume of the aqueous pool (25 L). Ten minutes of operation are equivalent to 3.5×10^{-2} MTUWE. The vapor entrainment factor in the melter is 5×10^{-3} for particulates and 5×10^{-2} for ruthenium. All of the ruthenium is assumed to be of the volatile form. The DF values provided by the cell and stack HEPA filters are 1 for ruthenium and 10^8 for particulates.

Discussion. There are 11 accidents assigned to this subgroup. Several of these are postulated to cause a pressurization of the melter cavity, which would cause a release of the melter vapor to the cell atmosphere. The release terms from these accidents are greater than from the other process pressurization events. Since the liquid-fed melter may experience steam surges, both the venturi-ejector and the condenser are designed for over capacity. Loss of cooling to the venturi-ejector would not instantaneously result in a pressurization. Current design philosophy calls for an automatic interlock to stop feed input upon a signal of melter pressurization.

Release to Stack. The ruthenium released to the stack would be equal to 1.75×10^{-3} MTUWE. The particulate released to the stack would be equal to 1.75×10^{-12} MTUWE.

Moderate Accidents Resulting in Process Pressurization

Description. Extensive foaming takes place in the melter and foam escapes to the cell through the pressure-relief outlet.

Assumptions. The glass temperature is assumed to be 1150°C . In lieu of a phenomenological model for volatile release from glass foaming, the assumption is made that one-fifth (36 kg) of the melter inventory is released as foam and the volatile release of cesium and ruthenium from this high-surface-area foam is 20% of the total contained in the foam. In addition, the melter vapor release of 25 L of HLLW is added to the source term. The DF values are 1 for ruthenium and 10^{-8} for particulates and cesium.

Discussion. While glass reboil and foaming can occur in the waste vitrification process, the assumptions made here represent an exceptionally severe occurrence. Foaming can normally be controlled by the automatic shutdown of feed and a reduction of power to the melter. The assumptions regarding the amount of glass foam escaping the melter and the subsequent release of volatile cesium and ruthenium are judgmental in lieu of experimental data. The assumption that there would be 25 L of HLLW in the melter represents the maximum flooding that has been observed in the developmental melters.

Release to Stack. The ruthenium released to the stack would be equal to 1.26×10^{-2} MTUWE, the cesium would be equal to 0.216×10^{-8} MTUWE, and the particulates would be equal to 1.75×10^{-12} MTUWE.

Severe Accidents Resulting in Process Pressurization

Description. A severe water-glass thermal reaction causes a steam explosion that ruptures the melter.

Assumptions. For this safety assessment it is assumed that this thermal reaction takes place despite all evidence that it could never occur. The release term consists of fragmented glass, cold-cap calcine, volatile cesium and ruthenium from molten glass, and vaporized HLLW. The quantity of fragmented glass (124 kg) was determined from the thermal balance of the heat required to vaporize the 25 L of HLLW and raise it to a temperature of 800°C and a pressure of approximately 22,060 kPa (3200 psia). The specific heat of the glass was assumed to be $0.25 \text{ cal/g-}^{\circ}\text{C}$. The 10 L of cold-cap calcine are equivalent to a

1-cm-thick cold cap over the entire molten glass surface area. The glass volatile release is based on the same assumptions used for the glass release severe accident. The accident is assumed to rupture the cell HEPA filter. The DF factors for the stack HEPA filter are 1 for ruthenium and 10^4 for cesium and particulates.

Discussion. The phenomenon of energetic thermal reactions as related to molten glass-water systems is discussed by Postman, Chapman and Buelt (1980). This accident is postulated as a design-basis accident here, but future analyses and experiments are expected to prove that it can not occur in this process. The large source term of particulates is a result of the assumption that all of the glass involved in the thermal interaction is dispersed into the cell atmosphere and is entrained into the ventilation airflow; this is a very conservative assumption. The ruthenium releases are treated as the volatile form and no credit for decontamination factors is taken in the analysis. The single failure of an engineered safeguard, in this case the cell HEPA filter, is assumed.

Release to Stack. The ruthenium released to the stack would be equal to 6.4×10^{-3} MTUWE, the cesium would be equal to 4.4×10^{-5} MTUWE, and the particulate would be equal to 5.6×10^{-5} MTUWE.

Minor Accidents in the Off-Gas System

Description. Water or extensive condensation enters the ruthenium adsorber bed and elutriates a portion of the ruthenium from the silica gel.

Assumptions. The adsorber bed has a volume of approximately 30 L. After one year's operating time it will hold approximately 73 g of ruthenium, which is equivalent to 3.75×10^{-2} MTUWE. All of this ruthenium is assumed to be elutriated and revolatilized downstream. The NO_x destructor is assumed to provide a DF of 300.

Discussion. There are 18 minor accidents and off-standard conditions associated with the off-gas system. Many of them do not result in releases exceeding the normal operating release level. In the past

the greatest concern has been with the release of the radionuclides accumulated on an adsorber bed. The elutriation of ruthenium from a silica gel adsorber bed is considered very unlikely, but is analyzed to indicate the maximum source term from this event.

Release to Stack. The ruthenium released to the stack would be equal to 1.25×10^{-4} MTUWE.

Moderate Accidents in the Off-Gas System

Description. The double HEPA filter from the off-gas system is dropped in the process cell during replacement.

Assumptions. The HEPA filter is the first double filter downstream from the melter. It contains one operating year's accumulation of particulates. Dropping the filter would release one-half of the accumulation to the cell atmosphere. The combined DF provided by the cell and stack HEPA filter banks is 10^8 .

Discussion. The bounding accident chosen for this accident subgroup again addresses the possibility of the release of an accumulation of activity in the off-gas system. The operating life is assumed to be one year at which time the filters would contain 1.9 kg of particulates, which is equal to 7.5×10^{-3} MTUWE.

Release to Stack. The particulate released to the stack would be equal to 3.75×10^{-11} MTUWE.

Severe Accidents in the Off-Gas System

Description. None identified.

Assumptions. Not applicable.

Discussion. The classification of an accident as severe required either extensive equipment damage or a large release of radionuclides to the environment. No accidents involving the off-gas system that meet these criteria have been identified.

Release to Stack. None.

Minor Accidents That Affect the Process Cell

Description. Dismemberment of the melter during replacement releases a quantity of calcine-like glass dust to the cell atmosphere.

Assumptions. The melter may be dismantled in the process cell, and approximately 4 kg (~2 L) of powdered glass will be released to the cell atmosphere during this operation. No auxiliary decontamination system for dust is used. There are 254 kg of glass per MTUWE. The cell and stack HEPA filters provide a combined DF of 10^8 for particulates.

Discussion. The joule-heated melter is not expected to last the life of the waste solidification facility. Replacement may require dismantling the melter in the process cell. Since this is a controlled process, it will be possible to control any radionuclide releases to a safe operating level. The above assumptions were made to quantify the maximum source that might result from such an operation.

Release to Stack. The particulate released to the stack would be equal to 1.6×10^{-10} MTUWE.

Moderate Accidents That Affect the Process Cell

Description. A fire occurs in the cell and results in failure of the cell HEPA filter.

Assumptions. The fire ruptures the double HEPA filter on the cell ventilation outlet. Approximately 1 kg of radioactive dust, which is assumed to be powdered glass, is set free by the fire. There are 254 kg of glass in a MTUWE. The DF provided by the stack HEPA filters is 10^4 .

Discussion. The amount of combustibles in the cell will be limited. The principal combustible will be the power cabling insulation. There will be a fire-detection system and at least one fire-suppression system in the cell. The danger to the HEPA filter from heat, fire and smoke will be considered in the design. The exact source of the 1 kg

of particulates is not defined; it is assumed to represent cell contamination. However, this amount of unrecovered contamination is very unlikely.

Release to Stack. The particulate released to the stack would be equal to 3.9×10^{-7} MTUWE.

Severe Accidents That Affect the Process Cell

Description. A design-basis tornado as defined by the NRC (Nuclear Regulatory Commission 1974) passes over the plant.

Assumptions. The worst conditions specified by the NRC are assumed. A 21-kPa pressure drop at the rate of 14 kPa/s occurs. The depressurization causes one set of the HEPA filters to rupture in the cell and building ventilation system. The melter vapor production is assumed to be vented to the cell atmosphere for 10 min. In keeping with the criterion that assumes the failure of a single engineered safeguard during design-basis accidents, the remaining HEPA filter is assumed to be ruptured. The DF for particulates and ruthenium is 1.

Discussion. The process equipment and vital services must be protected from the wind-generated forces and associated missiles by the building structure in order to meet the requirements of the regulatory guide. In addition, two cases of depressurization must be analyzed. Based on experimental data for HEPA filters exposed to over pressures and shock waves (Anderson and Anderson 1966), the depressurization represents the approximate rupture point of HEPA filters. Whether further analysis would prove they could withstand the assumed depressurization would depend on details of the ventilation system and the size and type of structure employed in the HEPA filter itself. The source term reflects interruption of the process and a boil-off of the maximum liquid hold-up in the melter to the cell atmosphere as opposed to the normal path in the off-gas system.

Release to Stack. The ruthenium released to the stack would be equal to 1.75×10^{-3} MTUWE. The particulate released to the stack would be equal to 1.75×10^{-4} MTUWE.

TABULATION OF RELEASE TERMS FOR ALL ACCIDENT SUBGROUPS

The release terms for volatile ruthenium, volatilized cesium (condensed to particulate form), and particulates are presented in Tables 8, 9 and 10 for minor, moderate and severe accidents, respectively. The release terms are given both in terms of MTUWE and curies. Ruthenium and cesium are listed separately because they can form volatile compounds that are released preferentially compared to the other radionuclides in the waste.

The ruthenium releases are large for the minor and moderate accidents relative to the release of all other radionuclides for these classes of accidents. This is a result of the conservative assumptions that ruthenium will have a high conversion factor to a volatile form and that no effective decontamination of the volatile ruthenium forms will take place in the cell and plant ventilation systems.

The severe accident releases are largest as a result of both the greater process upset and the assumption that the cell HEPA filter is unavailable as an engineered safeguard. This assumption is in keeping with design-basis accident analyses for nuclear reactors where the assumption that an engineered safeguard is unavailable is part of the licensing philosophy.

The largest releases for all classes of severity result from process pressurizations and damage to the cell boundaries. These results support the intuitive judgment that the design of a solidification facility must eliminate any possibility of an explosion in the process and must protect the process enclosure from damage due to any accident so that the radionuclides are contained.

The particular accident resulting in the largest particulate release is a design-basis tornado. It was assumed that the depressurization caused by a tornado ruptures the HEPA filters in the plant ventilation system, and that the cell HEPA filters are unavailable. While a tornado causes very little process upset, with no decontamination in the ventilation system all of the radionuclides escaping from the melter off-gas are released to the stack. Similar or larger releases would also be expected if the same accident assumptions were applied to solidification processes other than the LFCM process. The second

largest particulate release term results from a steam explosion in the melter, which is caused by a thermal reaction between the molten glass and the aqueous feed. Although there is considerable dispersion of the waste material in the process cell, the plant ventilation system limits the release from this accident.

Presenting the release terms only in units of activity (curies) obscures the fact that some radionuclides are biologically more of a hazard than others. Therefore, a simplified inhalation dose calculation was performed and is included in Tables 8, 9 and 10. The dose calculation predicts the whole-body dose that a person would accumulate over 50 yr as a result of standing at the site boundary at the worst location while the release cloud passes. The dose factors used in the calculation were taken from the DACRIN code (Houston, Streng and Watson 1976), but the dose calculation was carried out by hand. The dose is due only to inhalation during the release cloud passage and does not take into consideration long-term exposure to contaminated land, water or crops. A preliminary calculation of the dose due to cloud shine during the exposure period showed it to be about one-tenth of the inhalation exposure; thus, cloud shine was not included.

1X/10
The site and meteorological assumptions for the dose calculation are a 50-m stack, a plant boundary at a 2-km radius, and the 0- to 8-h atmosphere diffusion factors given in the Regulatory Guide 1.3 (Atomic Energy Commission 1974). The X/Q ratio for these assumptions is $3.5 \times 10^{-5} \text{ s/m}^3$.

The largest whole-body dose, 0.57 rem, is due to the particulate release during the design-basis tornado. There are no firm guidelines for judging the acceptability of public exposure as calculated for the very low-probability design-basis accidents at a solidification plant. However, some insight may be gained by comparing the design-basis accident doses with those given in the NRC criteria for picking a reactor plant site (Nuclear Regulatory Commission Reactor Site Criteria 1979). The regulation states that a design-basis accident should not lead to a 25-rem dose to the whole body nor a 300-rem iodine dose to the thyroid for an individual exposed for two hours in the exclusion area, nor should an individual in the surrounding low-population zone (beyond the site boundary) receive this dose if exposed to the entire cloud release.

TABLE 8. Radioactivity Releases for Minor Accidents

Source Term	Emitted from Stack		50-yr Whole-Body Dose Due to Acute Inhalation at Site Boundary, rem
	MTUWE	Ci	
Liquid			
Ruthenium	2.8E-03	1.3E+02	8.3E-03
Cesium	---	---	---
Particulates	2.8E-13	1.7E-07	9.1E-10
Total	---	1.3E+02	8.3E-03
Glass			
Ruthenium	1.3E-04	6.1E+00	3.7E-04
Cesium	6.2E-11	9.5E-06	4.7E-09
Particulates	---	---	---
Total	---	6.1E+00	3.7E-04
Pressurization			
Ruthenium	1.75E-03	8.2E+01	5.0E-03
Cesium	---	---	---
Particulates	1.75E-12	1.1E-06	5.8E-09
Total	---	8.2E+01	5.0E-03
Off-Gas System			
Ruthenium	1.25E-04	5.9E+00	3.7E-04
Cesium	---	---	---
Particulates	---	---	---
Total	---	5.9E+00	3.7E-04
Process Cell			
Ruthenium	---	---	---
Cesium	---	---	---
Particulates	9.6E-10	9.6E-05	5.2E-07
Total	---	9.6E-05	5.2E-07

TABLE 9. Radioactivity Releases for Moderate Accidents

Source Term	Emitted from Stack		50-yr Whole-Body Dose Due to Acute Inhalation at Site Boundary, rem
	MTUWE	Ci	
Liquid			
Ruthenium	1.1E-05	5.2E-01	3.1E-05
Cesium	---	---	---
Particulates	1.0E-11	6.0E-06	3.4E-08
Total	---	5.2E-01	3.1E-05
Glass			
Ruthenium	2.2E-04	1.0E+01	6.2E-04
Cesium	2.8E-10	4.3E-05	2.1E-08
Particulates	---	---	---
Total	---	1.0E+01	6.2E-04
Pressurization			
Ruthenium	1.3E-02	6.1E-02	3.6E-02
Cesium	2.2E-07	3.4E-02	1.7E-05
Particulates	1.8E-12	1.1E-06	5.9E-09
Total	---	6.1E+02	3.6E-02
Off-Gas System			
Ruthenium	---	---	---
Cesium	---	---	---
Particulates	3.8E-11	2.3E-05	1.2E-07
Total	---	2.3E-05	1.2E-07
Process Cell			
Ruthenium	---	---	---
Cesium	---	---	---
Particulates	3.9E-07	2.3E-01	1.2E-03
Total	---	2.3E-01	1.2E-03

TABLE 10. Radioactivity Releases for Severe Accidents

Source Term	Emitted from Stack		50-yr Whole-Body Dose Due to Acute Inhalation at Site Boundary, rem
	MTUWE	Ci	
Liquid			
Ruthenium	2.8E-03	1.3E-02	8.3E-03
Cesium	---	---	---
Particulates	4.0E-06	2.4E+00	1.2E-02 ✓
Total	---	2.4E+00	2.0E-02
Glass			
Ruthenium	4.6E-03	2.2E+02	1.3E-02
Cesium	4.4E-05	6.7E+00	3.3E-03
Particulates	---	---	---
Total	---	6.7E+00	1.6E-02
Pressurization			
Ruthenium	6.4E-03	3.0E+02	1.9E-02
Cesium	4.4E-05	6.7E+00	3.3E-03
Particulates	5.6E-05	3.4E+01	1.9E-01
Total	---	3.4E+02 340 Ci	2.1E-01
Off-Gas System			
Ruthenium	---	---	---
Cesium	None	---	---
Particulates	---	---	---
Total	---	---	---
Process Cell			
Ruthenium	1.75E-03	8.2E+01	5.0E-03
Cesium	---	---	---
Particulates	1.75E-04	1.1E+02	5.7E-01
Total	---	1.9E+02 190 Ci	<u>5.8E-01</u>

Total

CONCLUSIONS

The release term calculations showed that the largest releases will result from accidents associated with process pressurization and from those that occur at the same time a cell HEPA filter is damaged. These results support the intuitive judgment that the design of a solidification facility must eliminate any possibility of an explosion in the process and must protect the process enclosure from damage so that the radionuclides are contained. The cell HEPA filters serve as the major barrier to radionuclide releases and protection of these filters against pressure differentials, shock waves, excessive dust or smoke loading, wetting and fire must not be compromised by the desire for ease of maintenance. The designer could, if desired, also rely on redundant engineering safeguards, such as an auxiliary quench tank for the process pressurization vent, ruthenium adsorption at the stack, and double containment for HLLW process lines and tanks.

The calculated ruthenium releases for the minor and moderate accidents in terms of dose were found to be large relative to the releases of particulates. This is a result of the conservative assumptions that 1) ruthenium will have a high conversion factor to a volatile form and 2) no effective decontamination of volatile ruthenium will take place in the cell or plant ventilation systems. When a specific solidification plant design is analyzed, the ruthenium release estimates will undoubtedly be lower. However, if not it may be necessary to consider ruthenium adsorbers in the plant ventilation system to provide decontamination for any accidental release of ruthenium.

Since the process cell and the plant ventilation system effectively contain radionuclides, the LFCM process presents no safety hazards that are significantly greater than any other solidification process. In fact, the accident that resulted in the largest release term was a design-basis tornado and not an internal process disruption.

In summary, the release terms calculated for this safety assessment are useful for comparing the consequences of the different types of accidents. As solidification plant designs become more detailed, it will be possible to revise the release terms and reduce the conservatism used in the analytical

assumptions. An equally important contribution of this safety assessment is the thorough description of the accident scenarios. These accident descriptions reflect the knowledge gained through developing the LFCM process and will be used by the plant designers to increase the reliability and safety of the process and facility.

REFERENCES

- Anderson, W. L. and T. Anderson. 1966. "Effect of Shock Overpressure on High Efficiency Filter Units," In Proceedings of 9th AEC Air Cleaning Conference, Report CONF-660904, United States Atomic Energy Commission, Washington, DC.
- Atomic Energy Commission. 1974. Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Boiling Water Reactors, Regulatory Guide 1.3, Revision 2, Washington, DC.
- Bell, M. J. 1973. ORIGEN - The ORNL Isotope Generation and Depletion Code. ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Bradley, R. F. and C. B. Goodlet. 1972. Denitration of Nitric Acid Solutions by Formic Acid. DP-1299, Savannah River Laboratory, Aiken, South Carolina.
- Buelt, J. L. and C. C. Chapman. 1978. Liquid Fed Ceramic Melters: A General Description Report. PNL-2735, Pacific Northwest Laboratory, Richland, Washington.
- Buelt, J. L. and C. C. Chapman. 1979. Slurry Feeding of Nuclear Waste to an Electric Glass Melter. PNL-SA-7571. Presented at the ANS Meeting in Atlanta, Georgia.
- Christian, J. D. 1976. "Process Behavior and Control of Ruthenium and Cesium," Presented at the ANS-AICHE Meeting on Controlling Airborne Effluents from Fuel Cycle Plants, ANS-AICHE Meeting, August 5-6, 1976.
- Christian, J. D. and D. T. Pence. 1977. Critical Assessment of Methods for Treating Airborne Effluents from High-Level Waste Solidification Processes, PNL-2486, Pacific Northwest Laboratory, Richland, Washington.
- Environmental Radiation Protection Standards for Nuclear Power Operations, 40 C.F.R. Section 190 (1979).
- Gray, W. J. 1976. Volatility of a Zinc Borosilicate Glass Containing Simulated High-Level Radioactive Waste. BNWL-2111, Pacific Northwest Laboratory, Richland, Washington.
- Hall, J. C. 1977. Conceptual Liquid Waste Storage and Solidification Baseline Description. NMS-7, Allied-General Nuclear Services, Barnwell, South Carolina.
- Hanson, M. S. 1980. Spray Calcination/In-Can Melting: Effluent Characterization and Treatment. PNL-3109, Pacific Northwest Laboratory, Richland, Washington.
- Hanson, M. S. and J. O. Kaser. 1976. Design of Off-Gas Cleaning Systems for High-Level Waste Vitrification. BNWL-SA-5686, Paper presented at the 14th ERDA Air Cleaning Conference, Sun Valley, Idaho.

- Houston, J. R., D. L. Streng, D. C. Watson. 1976. DACRIN, A Computer Program for Calculating Organ Dose from Acute or Chronic Radionuclide Inhalation. BNWL-B-389, Pacific Northwest Laboratory, Richland, Washington.
- Larson, D. E. and W. F. Bonner (eds.). 1976. High-Level Waste Vitrification by Spray Calcination/In-Can Melting. BNWL-1092, Pacific Northwest Laboratory, Richland, Washington.
- Landon, L. F. 1980. Preliminary Technical Data Summary No. 3 for the Defense Waste Processing Facility. DPSTD-77-13-2, Savannah River Laboratory, Aiken, South Carolina.
- National Emission Standards for Mercury, 40 C.F.R., Section 61 (1979).
- Nuclear Regulatory Commission. 1974. Design-Basis Tornado, Regulatory Guide 1.76, Washington, DC.
- Nuclear Regulatory Commission Standards for Protection Against Radiation, 10 C.F.R., Section 20 (1979).
- Nuclear Regulatory Commission Reactor Site Criteria, 10 C.F.R., Section 100 (1979).
- Perry, R. H. and C. H. Chilton. 1973. Chemical Engineer's Handbook. Fifth Edition, McGraw Hill Book Co, New York, New York.
- Postma, A. K., C. C. Chapman and J. L. Buelt. 1980. As Assessment of Water/Glass Interactions in Waste Glass Melter Operation. PNL-2622, Pacific Northwest Laboratory, Richland, Washington.
- Pye, L. D. 1979. An Investigation of Controlling Mechanisms in Simulated Nuclear Waste Glass Melting and Gas Generation. New York State College of Ceramics, Alfred University, Alfred, New York.
- Ross, W. A., et al. 1979. "A Comparison of Glass and Crystalline Waste Materials," Ceramics in Nuclear Waste Management. Technical Information Center, U.S. Department of Energy, Springfield, Virginia.

APPENDIX A

STEAM EXPLOSIONS

APPENDIX A

STEAM EXPLOSIONS

Although violent water-glass interactions are never expected to occur, varying magnitudes of them were considered in this safety assessment. However, to emphasize the unlikelihood of their occurrence and methods undertaken to prevent them altogether, the Summary and Conclusions from a report investigating water-glass interactions is included here (Postma, Chapman and Buelt 1980, pp. 5 through 8).

A study of available information was made to evaluate the possibility of a vapor explosion in a LFCM. Vapor explosions (sometimes called violent glass-water interactions) need to be considered in the liquid-fed melter operation because experience in other industrial processes has shown that violent explosions can occur if a hot liquid is mixed with a cooler, vaporizable liquid. Many necessary conditions must be satisfied before a vapor explosion can occur, and the present study was aimed at determining whether all of the necessary conditions could be met in the liquid-fed melter operation.

Considerable experience has accrued world-wide in the operation of glass melters fed with aqueous solutions. No violent interactions have occurred so far. This experience and theoretical analyses indicate that destructive water-glass interactions are low-probability events, if they are possible at all. However, the information presently available does not permit one to totally rule out the possibility of a vapor explosion in a liquid-fed glass melter. Additional experimental studies in which molten glass and aqueous waste are purposely mixed are recommended to demonstrate that vapor explosions are not possible within the range of conditions that could be obtained in the melter.

Several precautions have been identified that will minimize the possibility of steam explosions during the time required to complete the follow-on experiments. The four precautionary actions listed below appear to be practical and will not adversely affect the operation of the existing melters.

1. The aqueous feed should be preheated prior to entry into the melter. Based on literature data that shows that vapor explosions do not occur if the cool liquid is close to its boiling point, we conservatively recommend that feed not be cooler than 10°C below its boiling point.

2. Cooling-water circuits used to cool the various parts of the melter should be examined to verify that a rupture in the cooling water conduits would not allow cool water to contact molten glass.
3. Feeding unheated process water to a hot melter should be avoided.
4. Glass product formulations should be designed to avoid the formation of glasses of unusually low viscosity. As shown later in this report, viscous dissipation inhibits vapor explosions. For typical waste glasses which have viscosities greater than 50 poise at 1050°C, the viscous retardation effect is expected to prevent violent interactions. However, if the glass viscosity was orders of magnitude lower than typical values, the inhibiting effect of viscosity might not alone be sufficient to prevent an interaction.

Other conclusions supported by the work completed are as follows:

- Vapor explosions occur when an appreciable volume of liquid becomes superheated. If the temperature exceeds the spontaneous nucleation temperature, then sudden flashing of liquid to vapor can generate shock waves and potentially destructive pressure pulses. We have concluded that such interactions are unlikely under standard operating conditions in a liquid-fed glass melter.
- In order to transfer enough heat to superheat a large volume of liquid, the hot liquid must be disintegrated and dispersed into an appreciable volume of the cold liquid. Experimental studies have shown that the collapse of a vapor film can initiate the breakup process. Several vapor collapse and growth cycles may occur in a feedback process leading to pressure pulses large enough to disperse the fluids. In the liquid-fed glass melter such disintegration is prevented by stable film boiling and by the high viscosity of molten glass.
- Vapor explosions never occur when the hot material is solid. This is because: 1) solids cannot be finely dispersed by collapsing vapor films and 2) solids act as nucleators, and it is not possible to appreciably superheat a liquid in the presence of vapor-forming nuclei. In the liquid-fed glass melter one predicts that a typical waste glass would be solidified before its temperature was low enough to allow a collapse of film boiling. This behavior alone would prevent vapor explosions under standard conditions.
- Small-scale tests with molten glass have shown that vapor explosions did not occur with glass under test conditions that produced explosions when the hot fluid was a molten metal. This supports the statement that the lower thermal diffusivity and higher viscosity of glass work against steam explosions.

- A very large effort is currently underway throughout the world to understand vapor explosions. However, to date, there is not an agreed-upon single model that describes all the sufficient conditions for a vapor explosion. Therefore, it is not possible to predict solely from theory whether a vapor explosion will occur for glass and water under all specified thermal conditions.
- Conditions in the waste glass melter that oppose vapor explosions include: 1) the feed contains suspended solids which would aid nucleation, thereby preventing the glass from becoming superheated; 2) the feed is preheated (experience has shown that steam explosions do not occur when water is hotter than 60°C); and 3) glass is highly viscous, thus preventing breakup in the short time required for a vapor explosion.
- Aqueous waste feeds containing powdered frit have a solid-particle concentration, which decreases the level of superheating that could occur. If these particles acted as nucleating sites, superheating-type vapor explosions would be impossible. However, not enough data are available to allow one to totally discount explosive interactions because of the suspended particles.
- Experience shows that dissolved gases, such as CO₂, can prevent violent interactions altogether. Hypothetically, the uncondensable gases provide a cushion between the two liquids thus disallowing vapor film collapse. This prevents fragmentation of hot liquid and thus no violent interactions occur.
- Experiments involving the intentional mixing of tens of kilograms of molten glass with waste solutions are needed to demonstrate that violent water/glass interactions are not credible under conditions which could be obtained in a liquid-fed melter operation.

Because of the findings of this report, research and development procedures at PNL have been updated to eliminate even the very slight possibility of steam explosions. Beforehand, a few liters of water with no suspended solids have been introduced into the melter before the slurry to cool the nozzle and prevent caking and plugging of the nozzle. Now, a water-cooled feed nozzle has been installed to eliminate the introduction of unheated process water. Also, procedures have been changed to preheat the feed to 90°C which will guarantee no explosions. In addition engineering calculations are being performed to determine whether enough CO₂ can be dissolved into the feed during the air-lifting step. This report shows that if the CO₂ concentrations in water up to 80 ml kg⁻¹ can be attained, which is 10% of saturated conditions, it would provide still another barrier to the possibility of a vapor explosion.

REFERENCES

Postma, A. K., C. C. Chapman, and J. L. Buel. 1980. An Assessment of Water/Glass Interactions in Waste Glass Melter Operation, PNL-2622, Pacific Northwest Laboratory, Richland, Washington.

APPENDIX B

LIQUID-FED CERAMIC MELTER PROCESS USED
IN THE SAFETY ASSESSMENT

APPENDIX B

LIQUID-FED CERAMIC MELTER PROCESS USED IN THE SAFETY ASSESSMENT

Because power wastes have higher activity levels than defense wastes, the off-standard conditions and consequences were postulated only for the power waste flowsheet. A more detailed description of the flowsheet used for this safety assessment is given in the figures and tables in this appendix. Figure B.1 shows the power waste solidification schematic, Tables B.1 and B.2 assign waste characteristics in the flow lines and equipment, and Table B.3 gives a detailed description with assigned decontamination factors for each piece of equipment. For comparison, a detailed process schematic is also given for defense waste in Figure B.2.

FIGURE B.1. Power Waste Solidification Flowsheet

TABLE B.1. Mass and Activity Balances for Power Waste Solidification Process Lines

Process Parameter	HLLW & ILLW 1 ^(a)	Recycled Condensate 2	Formic Acid 3	Denitration Off Gas 4	Glass Formers 5	Feed 6	Feed to Air Lift 7	Air 8	Air-Lifted Feed 9	Feed to Melter 10	Air 11	Glass Stream 12
LIQUIDS												
Flow rate, L/h	138	14	95			148	148		148	148		22
Acid molarity (g moles/L)	5	4	15			0.5						
Specific gravity	1.243	1.084	1.165									
GASEOUS												
Condensables, g-moles/h				1930								
H ₂ O												
HNO ₃												
Noncondensables, g-moles/h				2590				27	27		27	
N ₂												
O ₂												
CO ₂												
NO												
NO ₂												
NH ₃												
Temperature, °C							90		90	90		
Pressure, kPa											--	
SOLIDS												
Mass flow rate, kg/h					35							53
ACTIVITY												
Ru-106, Ci/min										160		
Ru-106, Ci/L										67		
Particulates, Ci/min										1.9+3		
Particulates, Ci/L										780		

(a) The process lines are labeled by number in Figure B.1.

(b) Negligible.

TABLE B.1. (contd)

Process Parameter	Off Gas 13 ^(a)	Condensate 14	Scrubbed Off Gas 15	Scrub Solution 16	Scrub Purge 17	Condensate 18	Noncondensables 19	Mist Condensate 20	Evaporator Off Gas 21	Condensate 22
LIQUIDS										
Flow rate, L/h		31920		31800	134.8	34.8		(b)	156	156
Acid molarity (g moles/L)										
Specific gravity										
GASEOUS										
Condensables, g-moles/h	7367		1930							
H ₂ O	7300		1930							
HNO ₃	67									
Noncondensables, g-moles/h	1799		4389				4389			
N ₂	1185		1324				1324			
O ₂	391		430				430			
CO ₂			1447				1447			
NO			965				965			
NO ₂	223		223				223			
H ₂										
NH ₃										
Temperature, °C	110		50	30						
Pressure, kPa				621						
SOLIDS										
Mass flow rate, kg/h										
ACTIVITY										
Ru-106, Ci/min	8	1.7E+3	0.8		7.2		4.0E-3			
Ru-106, Ci/L	2.3E-3	3.2	3.3E-4		3.2		2.4E-6			
Particulates, Ci/min	9.5	2.0E+3	0.95		8.6		9.5E-3			
Particulates, Ci/L	2.8E-3	3.8	4.0E-4		3.8		5.8E-6			

(a) The process lines are labeled by number in Figure B.1.

(b) Negligible.

TABLE B.1. (contd)

Process Parameters	Noncondensables 23(a)	Noncondensables 24	Decontamination Off Gas Feed 25	Ammonia 26	NO _x Destructor Feed 27	Stack Off Gas 28	Evaporator Condensate to Stack 29	Purge 30	Purge 31
LIQUIDS									
Flow rate, L/h								(b)	14
Acid molarity (g moles/L)									4.8
Specific gravity									1.1
GASEOUS									
Condensables, g-moles/h						1781			
H ₂ O						1781			
HNO ₃									
Noncondensables, g-moles/h	(b)	4389	4389		5577	4204			
N ₂		1324	1324		1324	2512			
O ₂		430	430		430	245			
CO ₂		1447	1447		1447	1447			
NO		965	965		965				
NO ₂		223	223		223				
NH ₃				1188	1188				
Temperature, °C									
Pressure, kPa									
SOLIDS									
Mass flow rate, kg/h									
ACTIVITY									
Ru-106, Ci/min		4.0E-3	2.0-5			6.7E-8			
Ru-106, Ci/L		2.4E-6				3.0E-11			
Particulates, Ci/min		9.5E-3	9.5-11			9.5E-11			
Particulates, Ci/L		5.E8-6				4.3E-14			

(a) The process lines are labeled by number in Figure B.1.

(b) Negligible.

TABLE B.2. Mass and Activity Balance for Power Waste Solidification Process Equipment

Process Parameter	Feed Prep Tank A (a)	Feed Tank B	Vent Orifices C	Air-Lift D	Head Pot E	Melter F	Canister G	Venturi H	Condenser I	Scrub Tank J	Evaporator K	Condenser L	Mist Eliminator M
LIQUIDS													
Flow rate, L/h				148	148	148 (waste)		31800	35			156	
Specific gravity													
Capacity, L	2000	2100								8000	1500		
Normal inventory, L	1184	1184								5300	500		
GASEOUS													
Condensables, g-moles/h								7367					
H ₂ O								7300					
HNO ₃								67					
Noncondensables, g-mole/h				27 (air)				1799					4389
N ₂								1185					1324
O ₂								391					430
CO ₂													1447
NO													965
NO ₂								223					223
Temperature, °C	95	90				1200				30	100		
Pressure, in. H ₂ O						-10							
SOLIDS													
Mass flow rate, kg/h						53 (glass)	53						
Normal inventory, kg						1350 (glass)	100						
ACTIVITY													
Ru-106, Ci/min													
Ru-106, Ci/L	67	67				489	448			3.2			
Particulates, Ci/min													
Particulates, Ci/L	780	780				6.0E+3	5.5E+3			3.8			

(a) The pieces of equipment are labeled by letter in Figure B.1.

(b) Trapped.

(c) Ci/yr.

TABLE B.2. (contd)

Process Parameter	Heater N	Filter O	Ruthenium Bed P	Iodine Bed Q	Reheater R	Filter S	Heater T	Startup Heater U	NO _x Destructor V	Blower W	Stack X	Canister Y
LIQUIDS												
Flow rate, L/h												
Specific gravity												2.75
Capacity, L												712.0
Normal inventory, L												0
GASEOUS												
Condensables, g-moles/h									1781	1781		
H ₂ O									1781	1781		
HNO ₃												
Noncondensables, g-moles/h	4389	4389	4389	4389	4389	4389	4389	4389	4204	4204		
N ₂	1324	1324	1324	1324	1324	1324	1324	1324	2512	2512		
O ₂	430	430	430	430	430	430	430	430	245	245		
CO ₂	1447	1447	1447	1447	1447	1447	1447	1447	1447	1447		
NO	965	965	965	965	965	965	965	965				
NO ₂	223	223	223	223	223	223	223	223				
Temperature, °C												
Pressure, in. H ₂ O												
SOLIDS												
Mass flow rate, kg/h												
Normal inventory, kg												
ACTIVITY												
Ru-106, Ci/min			4.0E-3 ^(b)								2.0E-5 ^(b)	
Ru-106, Ci/L			1.7E+3 ^(c)								8.6 ^(c)	
Particulates, Ci/min		9.5E-3 ^(b)					9.5E-7 ^(b)					
Particulates, Ci/L		4.2E+3 ^(c)					0.42 ^(c)					

(a) The pieces of equipment are labeled by letter in Figure B.1.

(b) Trapped.

(c) Ci/yr.

TABLE B.3. Process Equipment Description

Equipment	Description
Feed-preparation tank (A)	2000-L volume. Normal inventory is 1200 L. Used for denitration and glass-former mixing. Equipped with water and steam coils for temperature control (90°C to 95°C). Tank is recharged each 8-h shift in batch operation. Tank design and agitation system prevent sludge accumulation. Equipped with dip tubes and maximum-level alarm.
Feed tank (B)	2100-L volume. Normal inventory is 1200 L. Cooling-water coils and steam-heating coils automatically control temperature of feed to 90°C. This tank feeds into the air-lift line leading to the melter. Tank design and agitation system prevent sludge accumulation. Equipped with dip tubes and maximum-level alarm.
Vent orifices (C)	Installed on vents to Tanks A, B and K. Orifice provides flow reduction to off-gas system so that tank vent flow is minimal.
Feed air lift (D)	Meters liquid waste from supply tank to melter. Length depends on tank and melter elevations. Air supply is higher than tank level and incorporates a check valve as backup protection. Line diameter is approximately one inch. Air supply is approximately 10 L/min.
Disengagement/head pot (E)	Located above melter elevations. Permits separation of air and liquid. Overflow pipe to protect against overflow.
Liquid-fed ceramic melter (F)	Particulate DF is 200 and ruthenium DF is 20 as determined by experiments with simulated waste (Buelte and Chapman 1979). (Described in report.)
Canister (G)	Repository heat-loading limits may limit canister inventory to approximately 100 L (300 kg). Therefore, the canister size is specified as 30.5 cm in dia by 180 cm in length. A prime candidate for canister material is 1/4-in.-thick stainless steel 304. Canister connection is equipped with a knife gate valve (pneumatic operator) for canister change-out. A flexible bellows permits alignment and accommodates thermal expansion. Heaters are installed to prevent or correct glass pluggage in this region.

TABLE B.3. (contd)

Equipment	Description
Venturi scrubber-ejector (H)	The primary purpose of this off-gas treatment stage is to condense the melter off gas while simultaneously scrubbing large amounts of particulates from the melter off gas. The condensate is cooled in a collection tank and recycled as the scrubbing solution. The normal melter off-gas stream contains 136 kg/h of condensables and 56 kg/h of noncondensables. However, the venturi was sized to accommodate steam surges of up to 408 kg/h. This requires scrubbing of 530 L/min through a 9-cm-dia nozzle at 620 kPa and 30°C. The DF provided by the venturi is at least 10 for particulates and 10 for the volatile ruthenium (Christian and Pence 1977).
Main condenser (I)	The main condenser accommodates any steam carry-over from the venturi, condenses the steam coming from the denitration process, and provides a DF of 10^2 for particulates and 2×10^2 for volatilized ruthenium (Christian and Pence 1977). The normal flow to the condenser is 35 kg/h steam and 9.8×10^4 std L/h of noncondensables. It is sized to accommodate additional steam surges of up to 400 kg/h.
Venturi scrub tank (J)	This 3000-L tank holds, agitates and cools the condensate from the melter. As condensate accumulates, it continuously overflows to the evaporator. A maximum-level detector is provided.
Evaporator (K)	This is the primary evaporator in the off-gas system. It receives the condensate from the venturi scrub tank (J), the condenser (I) and the mist eliminator (M). The evaporator bottoms are recycled to the feed tanks. The evaporator holds up to 1400 L of condensate (the equivalent of one shift).
Evaporator condenser (L)	Condenses approximately 155 kg/h steam. Condensate is sent to multiple evaporator/condenser stages known as the condensate-cleanup system. Decontaminated condensate is evaporated to the plant stack to comply with the zero-liquid-discharge philosophy.

TABLE B.3. (contd)

Equipment	Description
Mist eliminator (M)	No credit taken for DF. Uses baffles and wire mesh to eliminate any mists from the off-gas stream.
Heater (N)	Automatically controls the temperature of the off gas to prevent condensation in the HEPA filters and the adsorber beds. Gas flow is approximately 1650 std L/min. Overtemperature cut-outs are incorporated to prevent possibility of damage to downstream HEPA filters.
HEPA filters (O)	Two-stage HEPA filters provide a 10^4 DF for particulates. The normal gas flow is 1650 L/min (60 scfm). A 30.5-cm-square by 30-cm-deep HEPA filter will have a 0.25-kPa (1-in.-H ₂ O) pressure drop with a 7100-L/min (250 scfm) flow. Therefore, this size filter will permit up to a factor of ten flow increase before incurring a significant pressure drop (Long 1978). This filter can withstand shock pressures of up to approximately 21 kPa (3 psi) before rupturing (Anderson and Anderson 1966). The materials are heat resistant.
Ruthenium adsorber beds (P)	Silica gel is used as the medium for trapping ruthenium. The bed can be partially rejuvenated by washing it with water. The DF is greater at room temperature than it is at elevated temperatures. A DF of 2×10^2 is a reasonable working value; however, measured values have been both higher and lower (Newby and Barnes 1975). The bed velocity should be less than 12 cm/s. Therefore, the bed cross-sectional area should be 0.23 m ² or greater. To adsorb the 0.049 g Ru/MTUWE for one year of operation at a loading safety margin of two, a column 13 cm high with a pressure drop of 0.25 kPa (1 in. H ₂ O) is required.
Iodine adsorber beds (Q)	Silver-impregnated zeolite is an effective iodine adsorber with a DF of 10^3 (Burger and Burns 1979). Only 0.5% (1.2 g) of the 245 g in each MTUWE is assumed to be in the HLLW. Assuming a capacity of approximately 7 g iodine to 100 g sorbent, a bed of approximately 100 kg sorbent will accommodate a year's (1500 MTUWE)

TABLE B.3. (contd)

Equipment	Description
Iodine adsorber beds (Q) (contd)	operation with a loading safety margin of two. The beds should each be approximately 0.23 m ² in flow area and approximately 30 cm high. The pressure drop will be approximately 1 kPa (4 in. H ₂ O).
Heater (R)	Same as N.
HEPA filter (S)	Same as O.
Heater (T)	Same as N except that this heater also uses the heat obtained from the catalytic reduction of NO _x to preheat the incoming off gas to the ideal temperature for the reaction (~500°C).
Startup heater (U)	A heater sufficient to raise off-gas temperatures to the point that the NO _x destructor bed can function.
NO _x destructor bed (V)	A zeolite bed that serves at the catalytic matrix for the NH ₃ reduction of NO _x to N ₂ , O ₂ and H ₂ O. These reactions take place at 350°C to 500°C. Approximately 99% of the NO _x is removed. This bed also reduces the volatile RuO ₄ ; DF of 300 for Ru are realistic (Christian and Pence 1977).
Off-gas blowers (W)	Two parallel blowers (one spare), each rated at 125% of maximum expected off-gas flow, provide the pressure differential for the melter and off-gas system.
Plant stack (X)	Stack height and building air dilution will be site specific.

FIGURE B.2. Defense Waste Solidification Flowsheet

REFERENCES

- Anderson, W. L. and T. Anderson. 1966. "Effect of Shock Overpressure on High Efficiency Filter Units," In Proceedings of 9th AEC Air Cleaning Conference, Report CONF-660904, United States Atomic Energy Commission, Washington, DC.
- Buelt, J. L. and C. C. Chapman. 1979. Slurry Feeding of Nuclear Waste to an Electric Glass Melter. PNL-SA-7571. Presented at the ANS Meeting in Atlanta, Georgia.
- Burger, L. L. and R. E. Burns. 1979. Technical Requirements for the Control of ^{129}I in a Nuclear Fuels Reprocessing Plant. Pacific Northwest Laboratory, Richland, Washington.
- Christian, J. D. and D. T. Pence. 1977. Critical Assessment of Methods for Testing Airborne Effluents from High-Level Waste Solidification Processes, PNL-2486, Pacific Northwest Laboratory, Richland, Washington.
- Long, J. T. 1978. Engineering for Nuclear Fuel Reprocessing. American Nuclear Society, LaGrange Park, Illinois.
- Newby, B. J. and V. H. Barnes. 1975. Volatile Ruthenium Removal from Calcliner Off-Gas Using Solid Sorbents. Allied Chemical Corporation, ICP-1078, Idaho Falls, Idaho.

APPENDIX C

ACTIVITY OF THE RADIONUCLIDES IN THE HIGH-LEVEL LIQUID WASTE

APPENDIX C

ACTIVITY OF THE RADIONUCLIDES IN THE HIGH-LEVEL LIQUID WASTE

As part of the process flowsheet development, it was desirable to determine approximately what DF would be required during normal operation. There are two principal regulations to be met. One is the Nuclear Regulatory Commission Standards for Protection Against Radiation (1979, Section 20.106), which basically limits the permissible concentration of radionuclides in the atmosphere at the site boundary. The second regulation is the Environmental Radiation Protection Standards for Nuclear Power Operations (1979). The EPA regulation limits the total release of Kr-85, I-129 and alpha-emitting transuranics during the nuclear fuel cycle, and limits the annual dose-equivalent received by any member of the public as a result of planned discharges of radioactive materials in the fuel cycle. The EPA standards are much more restrictive than the NRC concentration limits, but the EPA standards have just become effective and calculational models for determining compliance have not been well established. In this study only the restrictions imposed by the EPA regulations relating to total release of Kr-85, I-129 and alpha-emitting transuranics were calculated.

To make the calculations for NRC's regulations it was necessary to know the average meteorological conditions and the site characteristics since the concentrations are measured at the site boundary. An atmospheric diffusion factor, X/Q , of 10^{-7} s/m³ was used. This value has been reported (Christian and Pence 1977) as an average measured value for the Allied General Nuclear Service reprocessing plant located at Barnwell, South Carolina.

Table C.1 is a listing of the radionuclide activities at three years after discharge from the reactor and after reprocessing as calculated by ORIGEN (Bell 1973). The maximum permissible concentrations (MPC) allowed for each radionuclide by NRC are given in column two. Since the importance of each nuclide is really reflected by the ratio of activity to MPC, this quantity is given in the third column.

TABLE C.1. Radionuclide Activities of Three-Year-Old Reprocessed Fuel
Versus NRC's Maximum Permissible Concentrations

<u>Isotope</u>	<u>Activity Ci/MTU</u>	<u>MPC, Ci/m³</u>	<u>Activity/MPC</u>
Light Elements			
H-3	5.28E-02	2E-07	2.6E+09
Se-79	3.5E-01	1E-10	3.5E+09
Sr-89	3.72E-02	3E-10	1.2E+08
Sr-90	6.14E-04	3E-11	2.0E+15
Y-90	6.15E-04	3E-09	2.1E+13
Y-91	3.36E-01	1E-10	3.4E+09
Zr-93	1.65E-00	4E-09	4.1E+08
Nb-93m	3.76E-01	4E-09	9.4E+07
Zr-95	2.02E+00	1E-09	2.0E+09
Nb-95m	4.28E-02	1E-10	4.3E+08
Nb-95	4.49E-00	3E-09	1.5E+09
Tc-99	1.28E+01	2E-09	6.4E+09
Ru-106	4.72E+04	2E-10	2.4E+14
Rh-106	4.72E+04	3E-06	1.6E+10
Pd-107	1.05E-01	1E-10	1.1E+09
Ag-110m	1.09E+02	3E-10	3.6E+11
Ag-110	1.42E+01	3E-06	4.7E+06
Cd-113m	8.04E+00	1E-10	8.0E+10
Sn-119m	4.60E-01	1E-10	4.6E+10
Sn-123	7.88E+00	1E-10	7.9E+10
Sb-125	3.38E+03	9E-10	3.8E+12
Te-125m	1.40E+03	4E-09	3.5E+11
Sn-126	5.08E-01	3E-06	1.7E+05
Sn-126m	5.08E-01	3E-06	1.7E+05
Sb-126	5.03E-01	1E-10	5.1E+09
Te-127m	5.15E+00	1E-09	5.2E+09
Te-127	5.09E+00	3E-09	1.7E+09
I-129	3.39E-02	2E-11	1.7E+09
Cs-134	6.52E+04	4E-10	1.6E+14
Cs-135	2.24E-01	3E-09	7.5E+07

TABLE C.1. (contd)

<u>Isotope</u>	<u>Activity Ci/MTU</u>	<u>MPC, Ci/m³</u>	<u>Activity/MPC</u>
Light Elements			
Cs-137	8.85E+04	5E-10	1.8E+14
Ba-137	8.28E+04	3E-06	2.8E+10
Ce-144	4.47E+04	2E-10	2.2E+14
Pr-144	4.47E+04	3E-06	1.5E+10
Pm-147	4.04E+04	2E-09	2.0E+13
Sm-151	1.13E+03	2E-09	5.7E+11
Eu-152	1.02E+01	4E-10	2.6E+10
Go-153	9.06E-01	3E-09	3.0E+08
Eu-154	5.31E+03	1E-10	5.3E+13
Eu-155	1.81E+03	3E-09	6.0E+11
Heavy Elements			
Tl-208	1.11E-04	4E-08	2.8E+03
Pb-212	3.07E-04	6E-10	5.1E+05
Bi-212	3.07E-04	3E-09	1.0E+05
Po-212	1.97E-04	2E-14	9.9E+09
Po-216	3.07E-04	2E-14	1.5E+10
Rn-226	3.07E-04	2E-12	1.5E+08
Ra-224	3.07E-04	2E-11	1.5E+07
Th-228	3.06E-04	2E-13	1.5E+09
Th-234	1.58E-03	1E-09	1.6E+06
Pa-233	2.84E-01	1E-10	2.8E+09
Pa-234m	1.58E-03	3E-06	5.3E+02
U-234	7.41E-04	4E-12	1.9E+08
U-236	1.21E-03	4E-12	3.0E+08
U-237	1.04E-02	1E-10	1.0E+08
U-238	1.58E-03	3E-12	5.3E+08
Np-237	2.84E-01	1E-13	2.8E+12
Np-239	1.66E+01	2E-08	8.3E+08
Pu-236	6.11E-04	2E-14	3.1E+10
Pu-238	9.77E+01	7E-14	1.4E+15

TABLE C.1. (contd)

<u>Isotope</u>	<u>Activity Ci/MTU</u>	<u>MPC, Ci/m³</u>	<u>Activity/MPC</u>
Heavy Elements (contd)			
Pu-239	1.62E+00	6E-14	2.7E+13
Pu-240	2.97E+00	6E-14	5.0E+13
Pu-241	4.35E+02	3E-12	1.5E+14
Pu-242	6.50E-03	6E-14	1.1E+11
Am-241	1.59E+02	2E-13	8.0E+14
Am-242m	9.45E+00	2E-13	4.7E+13
Am-242	9.45E+00	1E-09	9.5E+09
Am-243	1.66E+01	2E-13	8.3E+13
Cm-242	1.75E+02	4E-12	4.4E+13
Cm-243	3.38E+00	2E-13	1.7E+13
Cm-244	1.88E+03	3E-13	6.3E+15
Cm-245	2.94E-01	2E-13	1.5E+12
Cm-246	5.66E-02	2E-13	2.8E+11
Bk-249	1.71E-04	3E-11	5.7E+06

The NRC requirement for a mixture of volatile (a), potentially volatile (b) and solid (c) radionuclides is that the concentration not exceed unity as expressed by the following relationship: $a/\text{MPC}_a + b/\text{MPC}_b + c/\text{MPC}_c = 1$. Therefore, the principal radionuclides that belong to these three groups are given in Table C.2 as a more convenient list to work with. The DF that would be required to meet the MPC, if each radionuclide were released by itself, are listed in column three. These DF values are obtained by multiplying the ratio of activity to MPC by the product of the release rate, 5.79×10^{-5} MTUWE/s, by the atmospheric diffusion, 10^{-7} s/m³. The total shown for each group is essentially the number of times the concentration would exceed the permitted value if all radionuclides were released together, which of course they are. To combine the three groups, volatile, potentially volatile, and solids, it is conservative to say that the DF required for each group is a factor of 10 greater than the sum for each group.

TABLE C.2. Summary Table of Principal Radionuclides

Isotope	Activity/MPC	DF Required to Meet MPC for an Individual Radionuclide
Volatiles		
H-3	2.6E+09	1.5E-02
Kr-85	2.6E+10	1.5E-01
I-129	1.7E+09	9.8E-03
TOTAL		1.7E-01
Potentially Volatile		
Se-79	3.5E+09	2.0E-02
Tc-99	6.4E+09	3.7E-02
Ru-106	2.4E+14	1.4E+03
Rh-106	1.6E+10	9.3E-02
Te-125m	3.5E+11	2.0E+00
Te-127m	5.2E+09	3.0E-02
Te-127	1.7E+09	9.8E-03
Sb-125	3.8E+12	2.2E+01
Sb-126	5.1E+09	3.0E-02
Cs-134	1.6E+14	9.3E+02
Cs-135	7.5E+07	4.3E-04
Cs-137	1.8E+14	1.0E+03
TOTAL		3.4E+03
Major Solids		
Sr-90	2.0E+15	1.2E+04
Y-90	2.1E+13	1.2E+02
Ce-144	2.2E+14	1.3E+03
Pm-147	2.0E+13	1.2E+02
Eu-154	5.3E+13	3.1E+02
Pu-238	1.4E+15	8.1E+03
Pu-239	2.7E+13	1.6E+02
Pu-240	5.0E+13	2.9E+02
Pu-241	1.5E+14	8.7E+02
Am-241	8.0E+14	4.6E+03
Major Solids		
Am-242m	4.7E+13	2.7E+02
Am-243	8.3E+13	4.8E+02
Cm-242	4.4E+13	2.5E+02
Cm-243	1.7E+13	9.8E+01
Cm-244	6.3E+15	3.6E+04
TOTAL		6.5E+04

Therefore, a DF of 2 is required for the volatiles, a DF of 4×10^4 is required for the potentially volatile, and a DF of 7×10^5 is required for the solids.

Part of EPA's requirements (Environmental Radiation Standards for Nuclear Power Operations 1979) states that the release quantities for the entire fuel cycle supporting one gigawatt-year of electricity are limited to no more than 50,000 Ci of Kr-85, 5×10^{-3} Ci of I-129, and 0.5×10^{-3} Ci of alpha-emitting transuranics with half lives greater than one year. In comparison, it has been determined that approximately 40 MTUWE are equivalent to a gigawatt-year (Department of Energy 1979). The limitations on release imposed by EPA's annual equivalent dose are very site specific and require an elaborate calculational model. Therefore, the dose limitations were not computed for this study. The DF required by the first part of the EPA regulations are given in Table C.3.

As a result of EPA's requirements the DF required for both I-129 and the solids (particulates) are increased by 270 and 286, respectively, from NRC's requirements (remember the latter are also site-specific numbers).

Table C.4 summarizes the DF values assumed for the process equipment and the DF values required by NRC and EPA regulations.

TABLE C.3. Decontamination Factors Required by EPA

<u>Radionuclides</u>	<u>Decontamination Factor Required</u>
Kr-85	No Kr-85 in HLLW
I-129	270
Transuranics	2E+08

TABLE C.4. Process Decontamination Factors and Decontamination Factors Required by Regulations

<u>Process/Regulation</u>	<u>Decontamination Factor</u>			
	<u>Particulates</u>	<u>Volatilized Ruthenium</u>	<u>Iodine</u>	<u>NO_x</u>
Process				
Liquid-fed melter	2E+02	20	1	NA
Venturi ejector	10	10	1	1
Tube-and-shell condenser	10 ²	2E+02	1	1
Mist eliminator	1	1	1	1
Two-stage HEPA filter	10 ⁴	1	1	1
Ruthenium adsorber bed	1	2E+02	1	1
Iodine adsorber bed	1	1	10 ³	1
Two-stage HEPA filter	10 ⁴	1	1	1
NO ₂ destructor	10	3E+02	1	10 ²
Overall DF	2E+14	2.4E+09	10 ³	10 ²
Regulation				
10 C.F.R. 20 ^(a)	7E+05	4E+04	2	NA
40 C.F.R. 190 ^(b)	2E+08	NA	3E+02	NA
40 C.F.R. 50 ^(c)	NA	NA	NA	10 ²

(a) Nuclear Regulatory Commission Standards for Protection Against Radiation (1979).

(b) Environmental Radiation Protection Standard for Nuclear Power Operations (1979).

(c) National Primary and Secondary Ambient Air Quality Standards (1979).

REFERENCES

- Bell, M. J. 1973. ORIGEN - The ORNL Isotope Generation and Depletion Code.
ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Department of Energy. 1979. Draft Environmental Impact Statement: Management
of Commercially Generated Radioactive Waste, Volume II. DOE/EIS-0046, U.S.
Department of Energy, Washington, D.C.
- Environmental Radiation Protection Standards for Nuclear Power Operations,
40 C.F.R. Section 190 (1979).
- National Primary and Secondary Ambient Air Quality Standards. 40 C.F.R.
Section 50 (1979).
- Nuclear Regulatory Commission Standards for Protection Against Radiation,
10 C.F.R. Section 20 (1979)

APPENDIX D

CRITICALITY ANALYSIS OF THE JOULE-HEATED MELTER

APPENDIX D

CRITICALITY ANALYSIS OF THE JOULE-HEATED MELTER

Accumulations of fissile material, which may lead to a criticality, are always a concern in a fuel reprocessing or a HLLW solidification plant. A combination of administrative controls, process geometry, process sampling, and neutron poisons are used to prevent criticalities. The joule-heated ceramic melter operates with continuous throughput as opposed to batch operation. A safety question that must be answered is whether there is an accumulation of sedimentation in the melter that may form a critical mass.

Current evidence (Walker and Riege 1979) indicates that the actinides in the HLLW will not precipitate in the melter as a sediment. The solubility of the actinides (UO_2 , NpO_2 , PuO_2 , AmO_2 and CmO_2) in glass containing 20 wt% simulated HLLW and 10 wt% Gd_2O_3 is approximately a factor of five or greater than the nominal concentration of these actinides in the glass. Therefore, the initial conditions for a criticality accident in the melter are not possible.

Due to the large neutron absorption cross section that many of the actinides have, there was some doubt as to whether a critical mass could be reached even assuming such a sedimentation was formed. Therefore, exploratory calculations using very conservative (even to the point of physical impossibility) assumptions were carried out (Partain and Kirby 1978) to investigate the possibility of a criticality in the joule-heated ceramic melter.

MELTER

Independent criticality calculations were made using the NULIF (Wittekopf et al. 1976) and ANISN (Engle 1967) codes. With NULIF, infinite homogeneous mixtures of the melter contents were examined utilizing ENDF/B-IV cross sections. ANISN, which is a one-dimensional transport code, was used to investigate criticality for a simple, reflected slab model of the melter contents. Hansen-Roach cross sections were used for those calculations.

Neither the NULIF nor ANISN (Hansen-Roach) cross-section libraries fully support all the isotopes contained in the waste material. Most of the important fissile and fertile isotopes were supported, but in some cases substitute materials were used. For the NULIF calculations, the isotopes U-232, U-237, Pu-236, Pu-244, Am-242m, Am-242 and all curium isotopes were modeled as U-223. A simple equivalence of thermal fission cross section based on data from Mughabghab and Gorber (1973) was used to determine the U-233 concentration. Although crude, considering the number of isotopes to be represented and the capture-versus-fission characteristics of U-233, the substitution was felt to be conservative. For the ANISN calculations a similar technique was used to model U-232, U-237, Pu-244 and the isotopes of americium and curium. We obtained Np-237 cross sections from HEDL and adapted them to the Hansen-Roach library structure in an approximate fashion. The method was felt to be sufficiently accurate for the exploratory calculations, but since Np-237 is the most predominant actinide isotope in the waste following U-238, additional efforts are probably warranted for accurate studies. However, the NULIF library, using ENDF/B-IV cross sections, does include Np-237.

MODEL

Typical waste and glass compositions were used for reference. The waste is identified as PW-7 with the composition given by Bonner, Blair and Romero (1976). The high-level waste is considered to have originated from the reprocessing of light-water reactor fuel having a burnup of 33,000 MWd/t and a specific power of 30 MWt. Chemical separation removing 99% of the uranium and plutonium is considered as well as 100% removal of the hydrogen, helium, krypton and xenon. A significant conservatism in estimating melter criticality was introduced by omitting gadolinium and fission products from the model; nitric acid and phosphoric acid were also omitted. The relative concentrations of isotopes for each element present in the waste were based on ORIGEN (Bell 1973) data at 1.5 yr after separation that followed a 1100-d irradiation period and a 150-d cooling time. The waste-glass identified as 76-68 by Mendel et al. (1977) was used. The deletion of the fission products and gadolinium meant

that the total waste concentration was less than the nominal 33 wt% of the mix, but the actinide portion of the waste has the appropriate concentration.

RESULTS

A series of criticality calculations for infinite homogeneous mixtures of the waste were performed and the results are summarized in Table D.1. The first calculation used the nominal concentrations of the constituents that are shown in Table D.2. The nominal concentrations assume uniform mixing of the actinide waste oxides with the glass. As noted in Table D.1, the calculated multiplication factor for this composition was 0.168. The potential effect on criticality of waste concentration or solidification was explored by assuming that only the actinide oxides are concentrated as a whole to a density of 12 g/cm^3 . This appears to be a reasonable upper limit for concentration since it is essentially the theoretical density of PuO_2 . The relative composition of the constituent isotopes in the concentrated waste are the same as for the nominal concentrations, but no glass constituents are included. As shown in Table D.2, the concentrated waste is more dense than the nominal values by a factor of about 46. For a homogeneous mixture of the concentrated waste with omission of the glass constituents, the multiplication factor was calculated to be 0.524. An additional calculation that attempted to consider the influence of filling the melter with water and concentrated waste (12 g/cm^3) was also performed. A uniform slurry mixture was developed with the relative fractions based on assuming the melter to be filled with concentrated actinide waste to the design level (48 cm) with the remainder filled with water (79 cm). (NULIF models this as a homogenized slurry.) For the concentrated slurry, the multiplication factor for the homogeneous mixture was calculated to be 0.539. The dominant spectrum was hard.

As the values of the calculated multiplication factors show for the cases calculated, a significant degree of subcriticality exists. The concentrated cases are physically unrealistic, and yet they indicate a substantial margin to criticality even for this extreme possibility.

TABLE D.1. Summary of Liquid-Fed Melter Criticality Calculations

<u>Waste Melt Composition</u>	<u>Type of Calculation</u>	<u>Multiplication Factor</u>
Nominal	Infinite homogeneous	0.168
Concentrated	Infinite homogeneous	0.524
Concentrated waste and water slurry	Infinite homogeneous	0.539
Concentrated waste and water reflectors	One-dimensional slabs	0.576

TABLE D.2. Reference Homogenized-Waste Melt Compositions

<u>Constituent</u>	<u>Atoms/barn-cm</u>	
	<u>Nominal</u>	<u>Concentrated</u>
U-233	1.64E-06	7.46E-05 ^(a)
U-234	1.19E-07	5.50E-06
U-235	5.47E-06	2.53E-04
U-236	2.57E-06	1.19E-04
U-238	5.85E-04	2.70E-02
Np-237	4.59E-05	2.12E-03
Np-239	6.03E-12	2.78E-10
Pu-238	7.16E-07	3.13E-05
Pu-239	2.58E-06	1.19E-04
Pu-240	1.36E-06	6.28E-05
Pu-241	5.21E-07	2.41E-05
Pu-242	2.32E-07	1.07E-05
Am-241	3.91E-06	1.81E-04
Am-243	5.74E-06	2.65E-04
Na	2.93E-03	---
Fe	5.40E-04	---
Cr	6.48E-05	---
Ni	2.70E-05	---
Si	1.20E-02	---
O	4.89E-02	7.85E-02

(a) Includes amount to simulate other U, Pu, Am and Cm isotopes not specifically noted in list.

The NULIF calculation would be expected to be more reactive due to the omission of leakage effects in the model. Other differences obviously exist; the NULIF code was developed for analysis of light-water reactor lattices and could be inappropriate for cases in which the spectrum was grossly different from that for a light-water reactor. The conservatism of the approach and computational method appears to be verified, however. To allow additional independent assessment and to provide consideration of some geometric modeling and leakage effects, a one-dimensional model was analyzed with the ANISN code. This code is capable of accurately calculating a critical assembly with a fast spectrum. The concentrated waste as noted in Table D.2 was modeled as a 48-cm slab reflected on both sides by 79-cm slabs of water. Sixteen-group Hansen-Roach cross sections were used and the calculated multiplication factor was 0.576. This value tends to compare favorably with the NULIF value for the concentrated slurry, even considering the different codes and cross sections used, and further indicates a significant degree of subcriticality.

CONCLUSIONS

A sediment consisting of actinides will not occur in the melter. Even if such a sediment occurred, it would be substantially subcritical due to the predominating neutron absorption characteristics of the actinide mix present in the HLLW. Inclusion of the fission products and Gd_2O_3 , a neutron poison, in the criticality calculations would further reduce the multiplication factors.

REFERENCES

- Bell, M. J. 1973. ORIGEN - The ORNL Isotope Generation and Depletion Code. ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Bonner, W. F., H. T. Blair and L. S. Romero. 1976. Spray Solidification of Nuclear Waste. BNWL-2059, Pacific Northwest Laboratory, Richland, Washington.
- Engle, W. W., Jr. 1967. A User's Manual for ANISN, A One-Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering. K-1693, Union Carbide Corp., Nuclear Division, Oak Ridge, Tennessee.
- Mendel, J. A., et al. 1977. Annual Report on the Characteristics of High-Level Waste Glasses. BNWL-2252, Pacific Northwest Laboratory, Richland, Washington
- Mughabghab, S. S., and D. I. Gorber. 1973. Neutron Cross Sections. BNL-325, Brookhaven National Laboratory, Long Island, New York.
- Partain, W. L. and K. D. Kirby. 1978. Safety Analysis of High Level Waste Solidification Processes. SSA-112, Southern Science Applications, Inc., Dunedin, Florida.
- Walker, C. T. and U. Riege. 1979. "Compatibility of Actinides with HLW Borosilicate Glass: Solubility and Phase Formation," Ceramics in Nuclear Waste Management. Technical Information Center, U.S. Department of Energy, Springfield, Virginia.
- Wittkopf, W. A., et al. 1976. Fuel Utilization and Performance Analysis Code (FUPAC) System, NULIF--Neutron Spectrum Generator, Few-Group Constant Calculator, and Fuel Depletion Code. BAW-426, Power Generation Group, Babcock & Wilcox, Lynchburg, Virginia.

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