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Synroc Processing Options

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Lawrence
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Synroc Processing Options

ABSTRACT

Synroc is a titanate-based ceramic material currently being developed for immobilizing high-level nuclear reactor wastes in solid form. Synroc D is a unique variation of Synroc. It can contain the high-level defense wastes, particularly those in storage at the Savannah River Plant.

In this report, we review the early development of the initial Synroc process, discuss modification and other options that simplify it overall, and recommend the future direction of research and development in the processing area.

A reference Synroc process is described briefly and contrasted with the Savannah River Laboratory glass-based reference case. Preliminary engineering layouts show Synroc to be a more complex processing operation and thus more expensive than the glass-based process. However, we believe that simplifications, which will significantly reduce the cost difference, are possible. Further research and development will continue in the areas of slurry processing, fluidized bed calcination, and mineralization. This last will use sintering, hot uniaxial pressing, or hot isostatic pressing.

SUMMARY

In the last two years of the LLNL Synroc program, the technical emphasis was on product synthesis, characterization, and leaching performance. There was a parallel effort to ensure that the synthesis techniques developed or used to fabricate the final waste form in the laboratory are feasible on a much larger scale in a remote canyon.

A Synroc D reference process was developed. In this process, we employ a ball mill to produce a fine particle slurry, a spray calciner to form an homogenous fine particle powder, and hot isostatic pressing (HIP) of large monoliths using in situ methods of redox-control with metal powders. A preliminary engineering layout and cost analysis made by the Savannah River Laboratory (SRL) showed the base process cell of the Synroc system to cost approximately twice that of the glass reference-process cell and, thus, the overall Synroc processing plant to be about 50% more than the total cost of the glass plant.

There are possible simplifications to the Synroc reference process that would significantly reduce the cost difference. These include reducing or eliminating the ball mill operation and replacing the spray calciner and metal redox powder mixing step by a slurry fed fluidized bed redox calciner. Additionally, sintering and hot uniaxial pressing (HUP) may offer some advantages over HIP, either in volume requirements or operational complexity.

Further research and development is required to determine whether the simplifications would be feasible and to what extent they improve the process. In the continuing Synroc program, high priority will be given to these areas. Equally important are the areas of washing and of liquid and solid separation at the head-end to remove sodium and silicon from the sludge. The presence of sodium and silicon produces a less resistant-to-leaching cesium host in the Synroc synthesis, and some research and development in reducing their presence is warranted.

INTRODUCTION

Synroc, a titanate-based ceramic material, is presently being developed for immobilizing high-level nuclear reactor wastes in solid form.¹⁻³ Synroc D, a unique variation of this waste form, will be able to contain high-level defense waste, particularly those in storage at the Savannah River Plant (Aiken, SC).⁴⁻⁷ Currently, there are three types of Synroc, i.e., Synroc B, the basic three-phase formulation containing no radioactive waste; Synroc C, containing simulated high-level commercial waste; and Synroc D, containing simulated high-level defense waste.

As part of a national DOE program on long-term high-level waste technology,⁸⁻⁹ the Synroc project was started at Lawrence Livermore National Laboratory (LLNL) in July 1979. During FY80 and FY81, the emphasis of the technical work was on

product synthesis and performance.¹⁰ In our program, small samples (<10 to 500 g) were synthesized, characterized, and leached. The major goal was to maximize the loading of the radioactive waste in a form having optimum nonleaching characteristics.⁵ A parallel effort was directed at ensuring that the synthesis techniques developed or used to fabricate the final waste form both were technically feasible on a larger plant size scale and could economically be operated in the environment of a remote canyon.

The purpose of this report is to describe the various processing options developed during the initial stages of the project, to discuss briefly some of the advantages and disadvantages of each option, and finally, to outline the future direction of research and development in the Synroc processing area.

REFERENCE GLASS PROCESS

The high level wastes at the Savannah River Plant are mixtures of acid wastes, which were neutralized with excess NaOH, and alkaline wastes. Most of the radionuclides are contained in a precipitate as an insoluble sludge. The exception is cesium, which is soluble and in the supernatant

The radionuclide concentration in the sludge is relatively low (<3%). The stored wastes are very heterogeneous, since there are a variety of reactor and separation processes that generated the wastes. In Table I are summaries of the average (composite) and two extreme (high iron and high

TABLE I. Summary of average (composite) and extreme compositions (high Fe and high Al) of Savannah River Plant waste calcines.⁷ Included are only major components.

	Without Al removal, wt%			With Al removal, wt%		
Fe ₂ O ₃	53.17	36.13	5.32	55.90	47.16	14.80
Al ₂ O ₃	4.78	28.26	76.05	1.29	9.24	50.77
MnO ₂	3.56	9.94	4.37	3.74	12.98	12.16
U ₃ O ₈	12.34	3.26	1.28	12.98	4.25	3.57
CaO	3.62	2.69	0.35	3.80	3.51	0.97
NiO	9.08	4.47	0.78	9.54	5.84	2.17
SiO ₂	0.40	0.85	0.56	0.40	1.12	1.57
Na ₂ O	4.52	5.98	1.96	4.75	6.63	5.41
Na ₂ SO ₄	0.50	0.93	0.50	0.50	1.21	0.50
Zeolite ^a	8.82	8.93	9.33	8.00	8.05	8.58

^aCesium returns to the main sludge stream fixed on a zeolite having a composition approximately Na₂O·Al₂O₃·55SiO₂·H₂O.

aluminum) calcined wastes or equivalent fully oxidized compounds. Because of this heterogeneity, extensive head-end processing is planned, it is largely independent of the final immobilization process and includes blending, mixing, screening, washing, separating liquids and solids, controlling pH and redox reactions, evaporating fluids, and perhaps reducing particle size.

The early glass process - referred to as the reference or base-case process - is schematically shown in Fig 1. The supernatant containing almost all the cesium and sodium and some strontium is separated from the sludge. The cesium and strontium in the supernatant is then removed using cation exchange columns. In this initial version of the base-case flowsheet, the cesium returns to the sludge processing stream fixed on an aluminosilicate zeolite. The strontium returns in a Na EDTA solution stream. An alkaline washing step removes approximately 75% of the aluminum and is included because a high aluminum concentration is deleterious to the glass process. The sludge plus the cesium and strontium streams are combined and then spray calcined to produce a

finely divided homogenous powder. Then, the powder is mixed with glass-forming components and placed in a glass melter.

In FY81, some significant modifications were made by Savannah River Laboratory (SRL) to the reference process (see Fig 2). To save on initial capital expenditures, the sludge and supernatant will be processed in two separate stages; the sludge will be the first. In processing sludge, the spray calciner is replaced by a liquid-fed continuous melter. This reduces the required process cell height and combines two separate steps, thus simplifying the system. In addition, the base-case cesium-stream treatment was changed. Cation exchange is still used to separate the cesium from the liquid stream, but now it is followed by eluting with formic acid. The eluate is then fed directly back to the sludge process. The composition of the resultant sludge and recycle streams is also changed somewhat from the first reference process. This is due largely to the elimination of the zeolite, the increased sodium from the cesium eluate, and the aluminum washing step, which was found to be less effective than originally believed.

FIG. 1. Reference process (FY80) for immobilizing Savannah River Plant radioactive waste (modified from Smith et al.7).

FIG. 2. Flow diagram for the first stage of the glass reference process of the defense waste processing facility.¹⁰ Sludge will be processed first, before the supernatant, to keep the initial capital expenditures down.

REFERENCE SYNROC D PROCESS

Basically, Synroc immobilizes the radionuclides as a dilute solid solution in the titanate minerals zirconolite ($\text{Ca} \cdot \text{ZrO}_2 \cdot 2\text{TiO}_2$), perovskite ($\text{CaO} \cdot \text{TiO}_2$), and a hollandite analogue ($\text{BaO} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{TiO}_2$). The large fraction of relatively inert components in the defense waste reacts to form solid solutions of spinel [$(\text{Fe}, \text{Mn}, \text{Ni}) (\text{Al}, \text{Fe})_2\text{O}_4 - (\text{Fe}, \text{Mn}, \text{Ni})_2 \text{TiO}_4$] and nepheline ($\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) phases.

The distribution of radionuclides among the Synroc phases has been investigated by microprobe analysis (see Table 2).⁹ Both perovskite and zirconolite can incorporate the fission products and actinide elements into their crystal lattices as dilute

solid solutions. (Radionuclides do not partition into the spinel phases.) Cesium is incorporated as a dilute solid solution (2 to 5 wt%) in barium hollandite.

The presence of more than a few weight-percent sodium in the sludge negates using hollandite as a cesium host. If cesium is to be processed with the sludge containing several weight-percent sodium and silicon, then nepheline or, possibly, pollucite ($\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) will act as the host. To use the more leach-resistant hollandite as a cesium host would require a separate processing stream, unless the sodium and silicon levels in the sludge could be reduced.

TABLE 2. Distribution of principal elements in high level wastes between coexisting phases of Synroc D, based on electron-probe microanalysis. Symbols in parentheses indicate that small amounts of these elements will also enter the designated phase. A possible minor metal phase incorporates Ni, S, Te, Bi, Pd, Rh, etc.

Zirconolite	Peroxykite	Spinel	Nepheline	Hollandite
U ⁴⁺	Na ⁺	Fe ²⁺	Na ⁺	Ce ⁴⁺
Th ⁴⁺	Sr ²⁺	Ni ²⁺	K ⁺	Rb ⁺
Pu ⁴⁺	Pu ³⁺	Mn ²⁺	Rb ⁺	K ⁺
U ⁶⁺	Am ³⁺	Fe ³⁺	Ba ²⁺	Ba ²⁺
Am ³⁺	U ⁶⁺	Mn ³⁺		
Np ⁴⁺	Np ³⁺	Al ³⁺		
(ACU ³⁺) ^a	(RRE ³⁺) ^b	Cr ³⁺		
(RRE ³⁺) ^b	(ACU ⁴⁺) ^c			
(Sr ²⁺)				

^aACU³⁺ are trivalent actinides

^bRRE³⁺ are rare earth elements

^cACU⁴⁺ are quadrivalent actinides

The processes needed to produce Synroc may be divided into three areas (see Fig. 3)

- Slurry processing e.g. blending, spray drying and evaporating

- Powder processing e.g. calcining and pelletizing

- Mineralizing and densifying e.g. sintering and hot pressing

Since it is better to carry out the mineralizing step under subsolidus conditions, the powders must be very homogeneous on a fine particle scale to help insure chemical equilibrium. A typical sequence of processing steps is shown in Fig. 3. The sludge and Synroc additives are mixed and spray dried or calcined to produce a free flowing dehydrated powder of micron sized particles (primarily nitrate and hydroxide compounds). The spray dried powders are calcined at higher temperatures (600 to 800 °C) to decompose them to oxides. The calcined powder is then redox calcined using CO and CO₂ mixtures to fix the valence state of several of the elements (primarily iron) before the mineralization step. This redox control aids in the final formation of spinel. Redox control may

FIG. 3. Basic Synroc processing options. The process may be divided into three areas, i.e., slurry processing, powder processing, and mineralizing (densifying).

also be done in situ using metal powders, i.e., nickel, iron, or titanium.

The mineralizing step may be carried out by hot uniaxial pressing (HUP) at temperatures of 1000 to 1200°C and pressures of 2 to 4 ksi. Sintering or hot isostatic pressing (HIP) are also possible methods for this step.

During FY80 a base-case flow sheet for the Synroc process was developed in cooperation with SRL personnel. The process is shown in Fig. 4. We used the initial reference glass-process as much as possible to take advantage of previous extensive engineering studies and experience. The aluminum-dissolution step was eliminated since aluminum is not deleterious to the Synroc process. However, the required head-end processing separating cesium and strontium and all associated off-gas and recycle systems are the same. The main differences are that a HIP operation is used instead of a glass melter and a ball milling step is added to prepare a fine particle ($<10\ \mu\text{m}$) feed for the spray drier. The HIP step to mineralize and densify was chosen for the reference process over sintering and HUP, because large monoliths could be produced in a closed system.

We visited several production facilities where large pieces of super alloys, used for jet engine parts, are made from powder. We evaluated their typical operating conditions and requirements relative to ours (see Appendix A). Working volumes of approximately $2 \times 8\ \text{ft}$, temperatures to 1500°C, and pressures to 15 ksi compare favorably with our reference case requirements of $2 \times 4\ \text{ft}$, 1100°C, and 15 ksi. If less than full density is acceptable, lower pressure will be required. To produce one ton per day requires cycle times of 18 h or less. The above production units have cycle times of approximately 8 h.

The composition of the working sludge is slightly different than the tank farm sludge shown in Table 1 due to the addition of some side streams (most notably Na₂O and SiO₂). The composition of the final product for the composite case is shown in Table 3, along with a breakdown of additives and major side streams. TiO₂ is first used as a grit blasting agent for final canister decontamination and then added to the sludge mixture. In the glass reference case, SiO₂ is the grit blasting agent.

With the above reference process as a guideline, a preliminary engineering layout was

(a)

FIG. 4. A base-case flow sheet for the Synroc process. This reference process was developed in cooperation with Savannah River Laboratory personnel.¹¹ (a) Feed and slurry preparation. (b) Calcination. (c) Ceramic formation and consolidation.

(b)

(c)

FIG. 4. (cont'd.)

TABLE 3. Flow rates and compositions of the major streams in the Synroc process as equivalent oxides. The tank farm sludge is only part of the material in the system. A major part of the sodium and silicon comes in from the recycle streams and from filter flushes. The final compositions resulting from the overall process are shown here.

	Total sludge from the tank farm		Recycle streams flow rate, lb/h				Flow rate of		Final product in canisters	
	Flow rate lb/h	wt %	Strontium ^a concentrate	Cesium ^b zeolite	Sand-filter flush	Canister decontamination	Total waste from sludge and recycle streams lb/h	Synroc additives flow rate lb/h	Final flow rate lb/h	wt %
Fe ₂ O ₃	20.46	40.1					20.46		20.46	24.3
Al ₂ O ₃	16.16	31.7		0.00			17.15		17.15	20.4
MnO ₂	5.44	10.7					5.44		5.44	6.5
Na ₂ O	2.70	5.3					2.70		2.70	3.2
Na ₂ SiO ₃	2.42	4.7	2.24	0.002	0.381		5.64		5.64	6.7
U ₃ O ₈	1.93	3.8					1.93		1.93	2.3
CaO	1.54	3.0					1.54	2.46	4.00	4.8
SiO ₂	0.40	0.8		2.91	1.57		4.88	5.93	10.81	12.8
ZrO ₂								4.10	4.10	4.9
TiO ₂						12.00	12.00		12.00	14.3
Total	51.0	100.1					65.67		84.2	100.0

^aThe major component is Na-EDTA. Assume that EDTA will be destroyed by calcination, except for the sodium content.

^bZeolite is (Na₂O, MgO, CaO, Al₂O₃, 4-6SiO₂, H₂O). Assume Na₂O, Al₂O₃, 5SiO₂, H₂O for material balance purposes.

FIG. 5. Schematic of the overall building floor plan for the most recent (FY81) glass reference process.¹¹

proposed and a canyon schematic detailed by E. I. DuPont engineers (Wilmington, Del.) under contract to SRI. The same engineering staff did a workshop for the glass reference process and similar ones for several of the alternative waste forms. The preliminary engineering studies allow us to compare the glass base-case process and several alternative waste-form cases and will help guide future research and development efforts in the processing area.

In this report, we will not detail the preliminary engineering layout, since that will be published by SRL. However, a brief summary is appropriate. The requirements of the processing cell dimensions for the latest glass and Synroc reference processes are shown in Figs. 5 and 6. The Synroc process requires a larger cell length (390 vs 192 ft), due to the increased area and volume needed by the ball mill and HIP units. This results in a significant difference in process cell costs, because the costs scale linearly with volume (length in our case, since width and height for the two reference cases are the same). The cost is approximately one million dollars per linear foot. Thus, the current reference-process cell for Synroc would

cost about 100% more than the current reference process cell for the glass process. The difference in total cost of the overall processing impacts much less, however, since the head-end process and peripheral and direct support operations and hardware (which are the same) constitute about half the total. Thus, the overall building and operational costs for the Synroc process are approximately 50% more than that of the glass reference process. We haven't examined the total costs of either process, including the costs of canisters, transportation, and ultimate repository storage. However, if they are significant, Synroc could be more economical, because it has nearly three times higher waste-loading capacity than glass and there would require fewer canisters and less repository storage space.

Two operations in the Synroc system contribute substantially to the increased process-cell length. The ball mill requires about 86 ft and HIP units about 175 ft. Both operations have two complete systems for redundancy. A large portion of the HIP requirement results from storing the working gas in the canyon and from the related compression operation. With more detailed analysis

and demonstration, the needed reduction in the ball mill and HIP operations might be relaxed for a potential reduction of 100 ft. Other simplifications also may be possible. These include eliminating the particle reduction operation and varying the calcination and redox control operations (see Mix and Mill Step, below).

MIX AND MILL STEP

Current synthesis is with micron sized particles. The existing sludge is composed mostly of particles $<20\text{ }\mu\text{m}$, although some agglomerations may be present. Larger particles, several hundred microns in size, are mainly sand and coal from filter flushes. These could be screened out and the remainder blended in a high shear mixer. If the pH were adjusted by adding acid, some components would dissolve and the particle-size distribution would be reduced. To test the effect of using large starting particles on the final product, some Syntrox synthesis should be done using large sized hydroxide particles, perhaps 20 and $50\text{ }\mu\text{m}$ diam. If these tests are successful, the milling requirement may be relaxed or even eliminated.

CONTROL STEP FOR DEHYDRATION, CALCINATION, AND REDOX REACTIONS

At this time, synthesis is carried out using one spray-drying step at 135°C to dehydrate the slurry and two separate calcination steps. The two calcination steps are at 600°C , to decompose to oxides, and at 800°C under O_2 to 1, CO_2 to CO gas atmospheres for valence control of iron, manganese, and uranium. An in situ redox control using metal additives (titanium, iron, or nickel) would eliminate one calcination step, but it requires a metal

blending step. Spray calcining combines dehydration and decomposition to oxides and has been extensively investigated and developed at Battelle Pacific Northwest Laboratory (Richland, Wash.) for the earlier glass reference process (i.e., spray calcination under a redox controlled atmosphere (CO and CO_2)) would eliminate several steps but requires a research and development effort to demonstrate feasibility. We can simplify this even further. The use of a fluidized bed for redox calcination that employs a slurry feed would be very space efficient compared to a spray dryer or calciner. In addition, it would combine all the above operational steps into one unit (dehydration, decomposition and redox control). Much experience has been gained at the Idaho Chemical Processing Plant (Idaho Falls, Idaho) with fluidized bed calciners and a wide variety of feeds,¹¹ but additional research and development is needed to demonstrate the feasibility of slurry feed and redox control for this specific Syntrox application.

MINERALIZATION INVOLVING HIP, SINTERING, AND HUP

For equipment and operational reliability, sintering is the most conventional and well understood of the three densifying operations. HUP and HIP under modest pressures each readily gives $>90\%$ densification at temperatures lower than required for sintering. The equipment and complexity of operation for high production rates using HIP or HUP however are significantly greater. For HUP there are concerns of die and punch lifetimes and operational reliability. For HIP there is the storage requirement of large working gas volumes as well as questions of equipment and operational complexity. Research and development of the process application of all these densification options will continue.

REFERENCES

1. A.I. Ringwood, S.F. Kesson, N.G. Ware, W.D. Hibberson, and A. Major, "The SYNROC Process: A Geochemical Approach to Nuclear Waste Immobilization," *Geochim. J.* **13**, 141 (1979).
2. A.I. Ringwood, S.F. Kesson, and N.G. Ware, "Immobilization of High Level Nuclear Reactor Wastes in SYNROC," *Nature* **278**, 219 (1979).
3. A.I. Ringwood, W. Sinclair, and G.M. McLaughlin, *Nuclear Waste Immobilization*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL 15147 (1979).
4. I.D. Tewhey, C.I. Hoeng, H.W. Newkirk, R.B. Rozsa, D.G. Coles, and F.I. Ryerson, *The Application of SYNROC to High Level Defense Wastes*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL 85032 (1980).
5. I. Tewhey, H. Newkirk, F. Ryerson, D. Coles, C. Hoeng, R. Rozsa, C. Rossington, and F. Bazan, *Phase Equilibria, Leaching Characteristics and Ceramic Processing of SYNROC D Formulations for U.S. Defense Wastes*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL 83658 (1981).
6. H.W. Newkirk, C.I. Hoeng, F.I. Ryerson, I.D. Tewhey, G.S. Smith, C.S. Rossington, A.I. Brackman, and A.I. Ringwood, *Status of SYNROC D Technology for Immobilizing U.S. Defense Wastes*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL 84395 (1980).
7. F.A. Stone, S.F. Gotroth, Jr., and P.K. Smith, *Preliminary Evaluation of Alternative Waste Forms for Immobilization of Savannah River Plant High Level Waste*, Savannah River Laboratory, Aiken, S.C., DP-1545 (1979).
8. F.A. Stone, S.F. Gotroth, Jr., and P.K. Smith, *Long Term High Level Waste Technology Program*, Department of Energy, Washington, D.C., DOE SRW-79-3 (1979).
9. I. Campbell, C. Hoeng, F. Ryerson, F. Bazan, T. Wolery, R. Van Konvenenburgh, M. Gauran, and R. Rozsa, *Immobilization of High Level Defense Wastes in SYNROC: An Appraisal of Product Performance*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL 85913 (1981).
10. I.H. Gould, Jr., Savannah River Laboratory, Aiken, S.C., private communication (1981).
11. S.F. Gotroth, Jr., Savannah River Laboratory, Aiken, S.C., private communication (1980).
12. F.A. Miller, *Design and Performance of a Full-Scale Spray Calciner for Nonradioactive High Level Waste Vitrification Studies*, Battelle Pacific Northwest Laboratory, Richland, Wash., PNL-3742 (1981).
13. R.E. Schindler, J.R. Berreth, G.C. Simpson, J.H. Valentine, and M.S. Walker, *Development of a Fluidized-Bed Calciner and Post-Treatment Processes for Solidification of Commercial Fuel-Reprocessing Liquid Wastes*, Idaho National Engineering Laboratory, Idaho Falls, Idaho, ICP 1136 (1977).

APPENDIX A. TRIP REPORT OF VISITS TO HIP FACILITIES

June 9, 1980

C&G 80-27

TO John Tewhey

FROM Clarence Hoernig R. Rozsa

SUBJECT Trip Report for Visiting Large Hot Isostatic Pressing Facilities Regarding MWED Program
Time Period: May 19-21, 1980

The Mineral Waste Form Development Program (MWED) is seriously considering the use of HIP (Hot Isostatic Pressing) as one method to process synthetic minerals as applied to U.S. Defense Waste Storage. In this regard large cylindrical shapes in the order of 2 ft dia. $\times$ 8 ft ht are under consideration for final storage in a repository and the HIP facilities that we visited were capable of handling canisters in this size range.

Two engineers from Savannah River Laboratory, Sam Gortoth and Marvin Jones, accompanied us on the trip. The main purposes of this trip were as follows:

- 1) To introduce the nature and scope of U.S. Defense Waste storage problem to engineers who have industrial production experience with HIP.
- 2) To jointly observe with SRI personnel the size and nature of the physical plant necessary for large scale HIP operations and to consider the feasibility of remote handling problems associated with the safe processing of radioactive materials.

- 3) To explore industrial canister design and failure experience in order to minimize or eliminate the possibility of contaminating a large HIP facility.

The companies contacted were:

- 1) Wyman Gordon Corp.
Worster Road
N. Grafton MA
- 2) Industrial Materials Technology, Inc.
155 River Street
Andover, MA 01810
- 3) United Technologies Research Center and Pratt-Whitney Corp.
Silver Lane
East Hartford, CO 06108

The HIP facilities visited were of two types. Both Wyman-Gordon and Pratt-Whitney have ASEA units which are vertical wire wound vessels with wire wound frame supported end closures. The IMT units are vertical forged vessels with threaded end closures manufactured by National Forge Corporation.

The ASEA units are the largest HIP facilities in existence. The furnaces enclosed in the pressure vessel have an available working space approximately 47 inches dia. $\times$ 60 inches height and are capable of achieving a temperature and pressure of 2300°F (1260°C), 20 KSI respectively. Temperature variations throughout the furnace cavity are held to less than $\pm 25^\circ\text{F}$ using a four zone furnace. Replacement costs for a new furnace are about \$300,000.

Because of size and long cooling times, the furnaces are never cooled below 1550°F (850°C) during normal operation. The work load is preheated in a separate furnace and then bottom loaded into the main HIP furnace. Finished work is unloaded at 850°C and slow cooled in a separate facility.

These ASEA units had been in operation about 2 years. Over that period of time ~ 140 heating cycles had been accomplished. During that time the furnace had to be repaired 3-4 times. Furnace failure was due to the unpredictable expansion after several cycles of an insulating mantel over the furnace. Design

changes are still in progress and these large HIP systems are still in the development stage. During normal operation 2 cycles per day can be accomplished without difficulty. A typical cycle is 2 hours from 850 °C to 1140 °C and one atmosphere argon to 25 KSI, a 3 hour soak at max temperature and pressure, and approximately 15 min to decompress to atmospheric pressure for unloading.

Both ASIA units are used almost exclusively to hot press super alloy metal powders to 100% theoretical density and near net shapes for high performance jet engine parts. Near net shapes are important to reduce machining costs as well as the loss of expensive alloy material.

Large parts up to 26 inch dia. x 20 inch ht. have been processed to full density with no variation in microstructure. To accomplish this HIP temperatures were pushed to 2050 °F or about 90% of the melting point.

Metal powders are 150 mesh (105 um) and with 50% below 325 mesh (44 um). Powders are transferred to evacuated HIP containers from special containers. Loading densities are in the range of 28-65% theoretical density. For our application it was felt that we could achieve good shape control if we could load our powder to a bulk density of at least 50-55% T.D.

At IMI the threaded HIP vessels are used primarily to improve the density of investment castings. There was a strong feeling that threaded enclosures were as safe as the ASIA design HIP facilities. The first HIP at IMI had a furnace with a working volume 38 inches dia. x 96 inches ht. and temperature/pressure capability of 2650 °F/15 KSI. Their furnaces were pulled for inspection after every 25 cycles.

The space requirement of the threaded vessels is much less than the ASIA unit. This made a tremendous impression on the SRI engineers because the overall size of the proposed remote processing facilities at Savannah River has the greatest impact on cost. The equipment represents only about 10% of the total cost.

Finally, in regard to container design, the current practice at IMI is the same as that used in industry. Bob Widmer at IMI felt that a secondary container could be designed to give assurances against the loss of radioactive debris in case leakage occurred.

There was general agreement by all SRI and IMI personnel and the representatives from industry that HIP processing of U.S. Defense Waste has considerable potential and HIP development should be actively pursued in our development program.