CERAMIC PROCESS AND PLANT DESIGN FOR HIGH-LEVEL NUCLEAR WASTE IMMOBILIZATION*

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ABSTRACT

In the last 3 years, significant advances in ceramic technology for high-level nuclear waste solidification have been made. Product quality in terms of leach-resistance, compositional uniformity, structural integrity, and thermal stability promises to be superior to borosilicate glass. This paper addresses the process effectiveness and preliminary designs for glass and ceramic immobilization plants. The reference two-step ceramic process utilizes fluid-bed calcination (FBC) and hot isostatic press (HIP) consolidation. Full-scale demonstration of these well-developed processing steps has been established at DOE and/or commercial facilities for processing radioactive materials. Based on Savannah River-type waste, our model predicts that the capital and operating cost for the solidification of high-level nuclear waste is about the same for the ceramic and glass options. However, when repository costs are included, the ceramic option potentially offers significantly better economics due to its high waste loading and volume reduction. Volume reduction impacts several figures of merit in addition to cost such as system logistics, storage, transportation, and risk. The study concludes that the ceramic product/process has many potential advantages, and rapid deployment of the technology could be realized due to full-scale demonstrations of FBC and HIP technology in radioactive environments. Based on our finding and those of others, the ceramic innovation not only offers a viable backup to the glass reference process but promises to be a viable future option for new high-level nuclear waste management opportunities.

INTRODUCTION

In previous work,(1-3) it has been established that polyphase ceramic waste forms, including Synroc(3) (synthetic rock) and natural analogous minerals,(4) exhibit superior leach resistance when compared to the borosilicate glass waste form under the same leach conditions. Ceramic waste forms have been synthesized from several chemical compositions typical of the Savannah River, Hanford, NFS and Barnwell reprocessing plants.(5-7)

DEMONSTRATED PROCESS TECHNOLOGY

In the last year, the ceramic process for high-level liquid waste immobilization has been reduced in process complexity to two major processing steps: fluidized-bed calcination (FBC) and hot isostatic press (HIP) consolidation. Both of these steps are well-developed and well-understood commercial processing methods(8-15) and, in addition, have been demonstrated in DOE facilities for government-generated fuel and waste management needs.

Fig. 1. Typical Fluid-Bed Calcine Prepared from Simulated High-Level Waste Liquids and Slurries.

Fluidized-bed calcine (Fig. 1) has been produced from high-level liquid waste for 18 years at the Idaho National Engineering Laboratory (INEL).(11) Although the Waste Calcine Facility (WCF) was designed as a pilot plant, it has been used as a production plant since 1964. Over 1700 m^3 (60,000 ft^3) of high-level waste calcine has been produced. High-level waste calcination began in the $105 million New Waste Calcine Facility (NWCF) at INEL in the fall of 1982.(12) The NWCF is remotely operated and maintained in accordance with "as low as reasonably achievable" (ALARA) principles to minimize personnel radiation exposures, whereas the WCF was manually maintained.(15) Therefore, the remotization equipment and procedures required for operating and maintaining high-level liquid waste FBC systems have been developed. The WCF, which is 1.2 m (4 ft) in diameter, is smaller, and the 1.5-m (5 ft) diameter NWCF is about the same size as a calciner required to process HLLW from a 3000-tonne/year reprocessing plant or for a base design which has characteristics assumed to represent the Savannah River requirements for defense waste consolidation. Thus, the FBC technology for the first step of the ceramic process is well established and could be readily adapted with little development for processing high-level liquid waste in a full-sized ceramic waste form production facility.

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Hot isostatic pressing (HIPping) technology has been used in the ceramic, metallurgical, and cermet industries for over 30 years.(13,14) Radioactive materials and spent fuel rods are being consolidated in DOE and commercial HIP units much larger than that required for ceramic immobilization of high-level liquid wastes.(13,14) Most HIP units are or could be remotely operated; many units are automated so that the entire HIPping operation from loading to unloading is automatic and controlled from a remote console. Thus, the HIP technology for the second step of the ceramic process utilizes technology developed over many years with full-size systems operated in radioactive environments. Although nonradioactive formulations similar in composition to high-level waste calcine have been HIPped, actual high-level waste calcine has not been HIPped. Optimization of HIP processing parameters on large-scale waste forms and HIP can development is required to adapt this technology to the ceramic process.

Thus, scale-up of this technology has been demonstrated. Full-scale facilities, utilizing this technology to process radioactive materials, have been built and operated in the United States. Integration of the two processing steps into a single remotely operated and maintained processing line is required to adapt this technology to immobilization of high-level liquid waste.

Ceramic Advantages

Several processing advantages result from the utilization of this technology to manage high-level liquid waste. The process is completely flexible in that it can make glass, glass ceramics, or ceramic waste forms. All waste compositions evaluated to date can be used to make high-quality waste forms. The process eliminates the problem of corrosion in the glass melter and utilizes major components that have an expected lifetime far greater than the lifetime of the facility. This eliminates major component replacement and increases the on-line time of the waste facility. Better retention of the volatile radionuclides is predicted for the ceramic process since the process operates at lower temperatures and, in the higher temperature portions of the process, the material is canned so that no volatility occurs at all. For the same sized glass and ceramic facility, the throughput of the ceramic facility is about three times the throughput of the glass facility because of the higher waste loading and density of the ceramic waste form.

The ceramic volume reduction over glass is about a factor of three due to the higher waste loading and higher density of the ceramic waste form. Whereas, glass is limited to about 30-35 wt. % waste loading to avoid separation of soluble phases from the glass, the ceramic waste loading can be increased to 60-90%, depending upon the composition of the waste. The density of the ceramic waste form is 4-6 g/cm^3, while that of glass is limited to about 3.0 g/cm^3. This greater volume reduction of the high-level liquid waste reduces the required storage capacity, transportation, repository implacement, and repository sites by about a factor of three; this signficantly reduces the costs of waste management.

The waste form quality of ceramics prepared by this process are considerably better than that of glass prepared in a melter. Ceramic waste forms are composed of a number of natural minerals which have existed in the earth's crust for millions of years.(4) Thus, by placing the radionuclides in analogous mineral forms, there is every reason to believe that these minerals will contain these radionuclides indefinitely, even under geologic conditions of temperature, pressure, and hydrology. Ceramic waste forms are crystalline; therefore, the exact location of all radionuclides can be determined.(5-7) The leaching of these materials can be demonstrated, leach-resistant waste minerals can be designed, and long-term leach rates can be modeled.(7) This accounts for the much better leach-resistance of ceramics compared to glass. In fact, the actinides, which produce the long-term hazards to man if this material is leached to the biosphere, have a leach-resistance up to three orders of magnitude greater than that of glass.(3,7) Although the multiengineered barrier system is designed to prevent the entry of radionuclides to the biosphere under normal conditions, under upset conditions, the major barrier against entry of these materials to the biosphere is the waste form. In addition, many other properties of the ceramic, such as fracture resistance, high-temperature stability, and thermal conductivity, are also superior to glass.(3,7) Fracture resistance reduces the surface area exposed to the leaching media significantly. The high thermal stability and thermal conductivity permits greater waste loading of shorter cooled waste and longer handling times without cooling with no damage to the waste form.

Ceramic Process Description

The ceramic process flow diagram for SRP defense waste is shown in Fig. 2. The high-level liquid waste is blended with the tailoring additives (calcia, silica, titania, and zirconia) to assure that waste components are immobilized in the desired phases in the blending-feed tank. In the case of defense waste containing mercury, the high-level waste would be refluxed with formic acid to reduce the mercury to the elemental state prior to blending. The mercury would be separated form the wastes, purified, and recovered. Excess additives are generally added to assure ample tailoring materials during feed compositional variations; the excess tailoring agents form highly stable phases such as zirconolite with excess zirconia. The additives are generally premixed and added as slurry for convenience and reduction of dusting, although the individual powders could be used if desired. For a large inventory of fairly uniform waste, the additives would probably be procured as premixed powders. The blending-feed tank would be of sufficient capacity to supply the fluid-bed calciner requirements for several days. Two blender-feed tanks would be used so that the next feed batch could be prepared in the second tank while the calciner was being fed from the first tank.

The fluid-bed calciner would be similar to the NWCF at INEL;(12) the same operating and maintenance procedures would be used.(12,15) The off-gas would pass through dual venturi scrubbers before passing through the ruthenium and iodine traps; after HEPA filtration, the gas would pass through a catalyst bed to destroy the NO$_x$ with ammonia (if necessary as with an acid fed calciner) before release to the stack.

The calcine can be withdrawn continuously or intermittently from the fluid bed by gravity and/or pneumatically. The calcine particulates (Fig. 1) are vibration packed in the HIP can to 50-60% theoretical density. The HIP can is then evacuated and sealed by crimp-welding the fill and evacuation tubes. In many cases, evacuation at 600°C is sufficient to assure correct redox conditions during HIPping. However, if

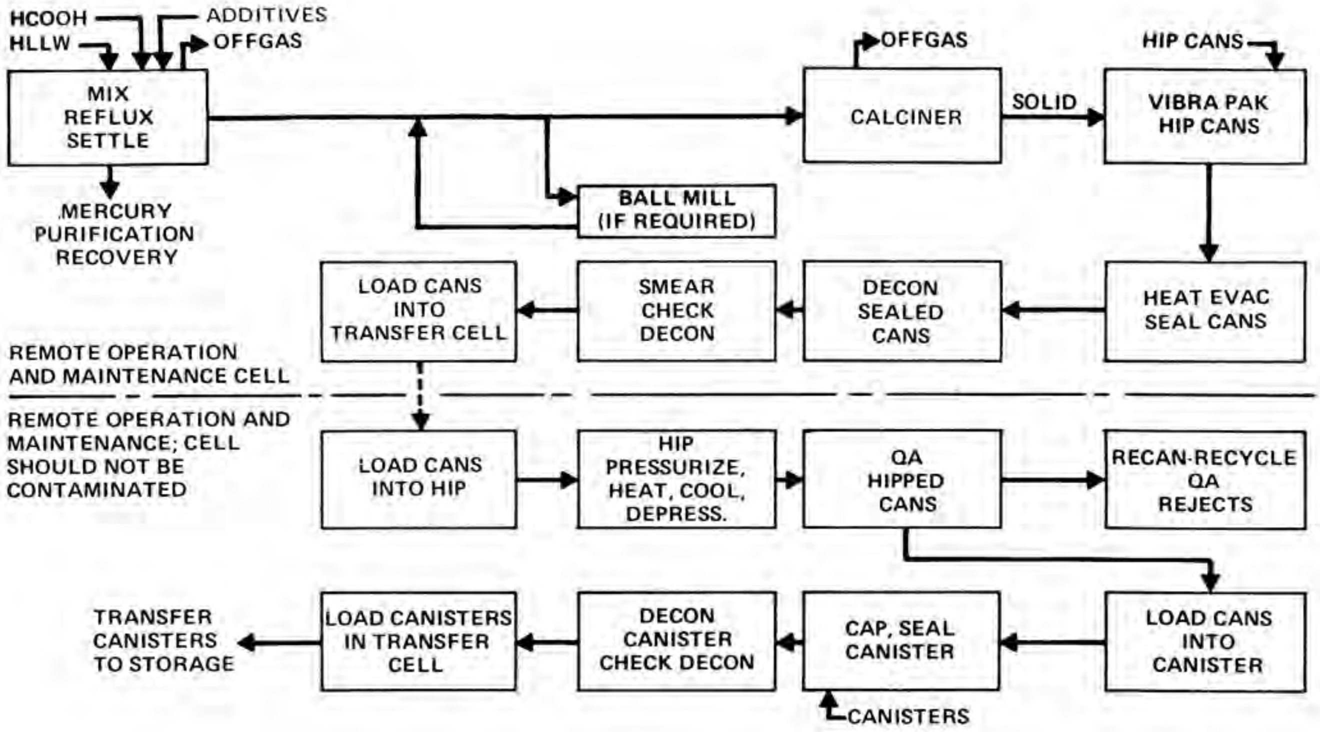

Fig. 2. Ceramic Preparation Flow Diagram for Neutralized Defense Waste.

necessary, the calcine particles can be reduced by short contact with a nonexplosive dilute mixture of hydrogen in nitrogen before sealing.(7) If desired, a small amount of metallic getter can be included in each HIP can to control gaseous NO_x, CO, CO_2, or H_2O that might form from incomplete denitration, dehydration, or removal of carbonaceous substances.(16)

The HIP cans are decontaminated (high-pressure water) and transferred to the HIP cell where they are HIPped and loaded into conventional defense or commercial waste canisters. These canisters are then sealed, decontaminated, and sent to storage. The rest of the waste management process is the same as glass except during storage, transportation, and handling consideration must be given to the higher radioactivity and decay heat contained in each package.

Three factors are often mentioned as causing problems in the fluid-bed calcination: nozzle plugging, high-sodium feed, and volatile nuclide evaluation. All of these factors can be handled by proper design and operation.(11,12,15) The nozzles at the NWCF are designed so they can be changed while calcination continues. Even though self-cleaning nozzles have been developed for commercial applications, nozzle plugging is so infrequent the changeout method of operation is preferred. High-sodium calcines tend to agglomerate in storage; since calcine is not stored in the ceramic process, this is not a problem. Proper control of feed composition will permit fluid-bed calcination of much higher sodium content wastes than currently processed at INEL. Even though nuclide volatilization is low during calcination operation, improved retention methods would be desirable during fluid-bed calcination as in all solidification methods. Off-gas scrubbers are designed to remove volatile nuclides and recycle them to the calciner; volatile retention in the calciner appears to improve during the second pass through the system.

ECONOMICS

The laboratory process knowledge, in terms of process flexibility plus the knowledge of key process steps in scale-up operations, established the practicality of ceramic technology for high-level nuclear waste solidification. On the basis of the above premise, preliminary generic designs of ceramic and glass nuclear immobilization facilities, which did not take into account possible site-specific considerations, were used to develop process layouts for both technologies (Fig. 3 and 4). Since a better data base exists for Savannah River waste management, this data base was used in this analysis. The overall comparative costs of managing high-level nuclear waste by the glass and ceramic options from immobilization through repository emplacement were developed using a methodology similar to that used by McDonell(17) at SRL and Rozsa(18) at LLNL. The plant layouts given in Fig. 3 and 4 are intended to be generic and not site-specific; therefore, site-specific considerations may alter these layouts somewhat.

The capital cost (Table I) of the ceramic and glass plants were derived from the immobilization plant capital costs used by McDonell by using the 0.6 power factor to adjust the size. McDonell's 1093-m^2 (11,764 ft^2) facility was estimated at $455 million; therefore, our 875 m^2 (9,424 ft^2) glass and 879 m^2 (9,462 ft^2) ceramic plants were estimated to cost $398 million and $399 million, respectively.

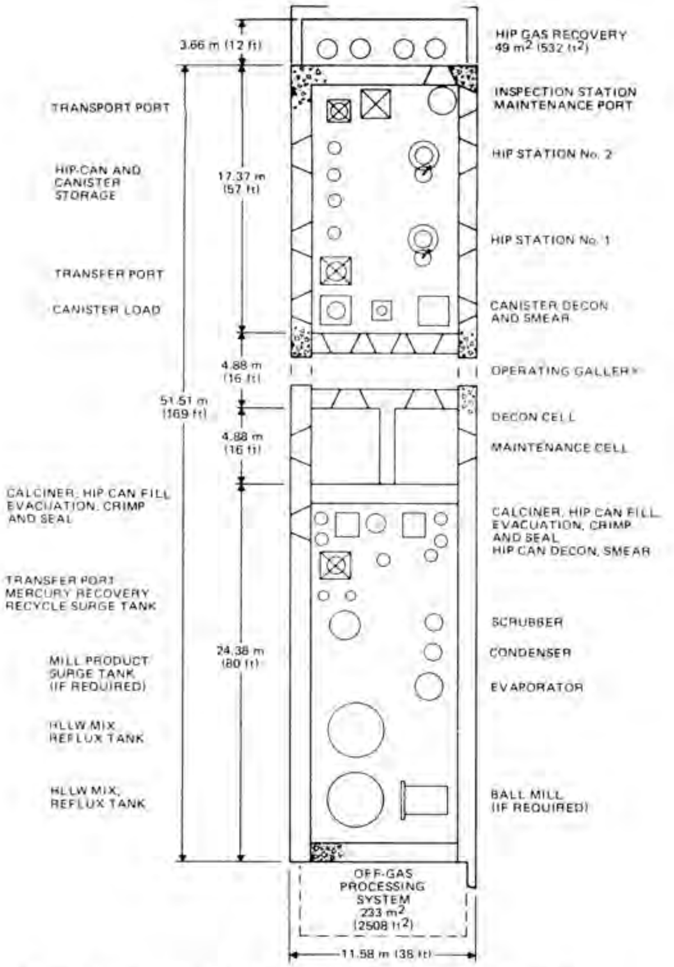

Fig. 3. Ceramic Process Layout for Neutralized Defense Waste.

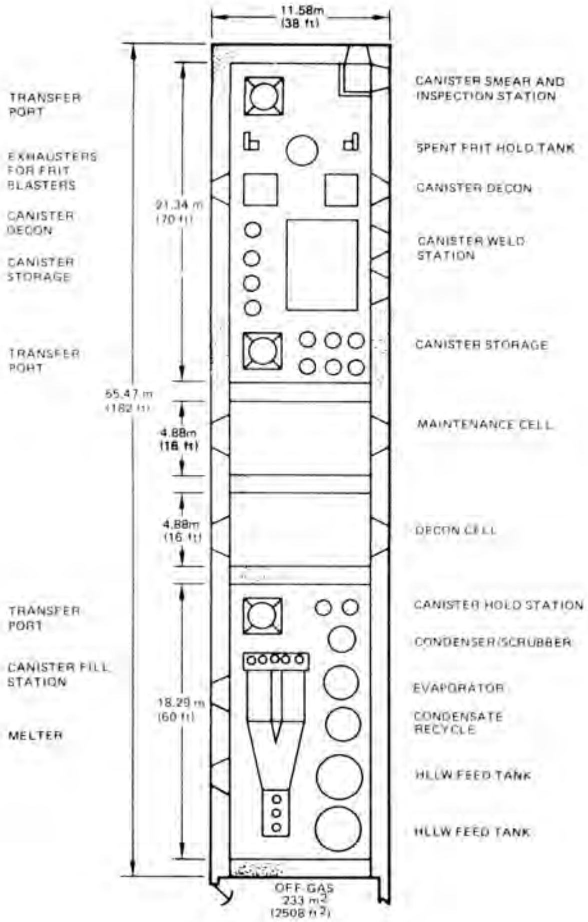

Fig. 4. Layout of Glass Production Canyon for Neutralized Defense Waste.

In addition, lump-sum allocations of $50 million for equipment, $26 million for storage, and $25 million for shipping were added for a total capital cost of $499 million and $500 million, respectively (see Table I). These same lump-sum allocations were used by McDonell and Rozsa.

SRP has a large inventory of waste and continues to produce a small amount of waste each year. For this study, it was assumed that waste would be processed through the Year 2013 but that the current waste inventory would be completely processed by 2009 if the glass plant starts up in 1989 as planned. Thereafter, the glass plant would process the waste as produced by the reprocessing plant.

The ceramic plant shown in Fig. 3 can process the current inventory as well as the current waste produced at the SRP at three times the rate of the glass plant shown in Fig. 4; thus, both plants have the same waste form package production rate, but the ceramic plant requires three times the waste feed rate as the glass plant. The higher waste loading and density of the ceramic waste packages account for this difference. Thus, at three times the waste processing rate, the ceramic plant could immobilize the current SRP inventory in ~7 years, whereas the glass plant would require ~20 years; thereafter (15 years), the ceramic plant would process the waste as it was produced by the reprocessing plant. For this analysis, it was assumed that the ceramic plant would start up in 1992. Costs associated with processing the current SRP waste inventory in the ceramics plant over a 7-year period were calculated; however, operating costs associated with processing the current waste inventory over the 22-year period were essentially the same. Thus, with the ceramic option, the current waste inventory could be immobilized before it could be immobilized in glass, with little or no cost penalty.

It was assumed that $80,000 per man-year for plant operators, first-line supervision, and first-line engineering would cover all labor and labor-associated overhead costs such as management, payroll, purchasing, general maintenance, etc. Sixty-seven percent of labor costs were assumed to cover materials and supplies. Annual equipment replacement costs were assumed at 15% of glass capital equipment costs and at 10% of the ceramic capital equipment costs; different percentages were used since the major item in the glass plant, the melter, must be rebuilt every 2 to 3 years, whereas the major items in the ceramic plant, calciner and HIP units, should last the lifetime of the plant without replacement. Operating costs are summarized in Table I.

Total waste management costs are given in Table II. A 17.7% contingency was added to the facility costs; this same contingency was used by McDonell. Since full-scale components of the major ceramic processing equipment has been constructed and

TABLE I

SRP NEUTRALIZED GOVERNMENT-GENERATED WASTE IMMOBILIZATION COSTS*

Item	Cost ($ millions) Glass	Cost ($ millions) Ceramic
Immobilization facility capital		
Facility	398	399
Equipment	50	50
Storage	26	26
Shipping	25	25
Total	499	500
Annual operating (inventory processing)		
Labor	11.6[a]	13.8[b]
Materials	7.8	9.2
Equipment repair	7.5	5.0
Total (annual)	26.9	28.0
Annual operating (post inventory processing)		
Labor	10.6[a]	10.0[b]
Materials	7.1	6.7
Equipment repair	7.5	5.0
Total (annual)	25.2	21.7

*60-cm (24 in.) diam. canisters used, resulting in 10,300 canisters of glass and 3,400 canisters of ceramic.

[a] Assumed current inventory processed in 20 years with 5 year operation thereafter.

[b] Assumed current inventory processed in 7 years with 15 years operation thereafter.

TABLE II

COMPARATIVE WASTE MANAGEMENT COSTS FOR SRP GOVERNMENT-GENERATED WASTE

Item	Costs ($ millions) Glass	Costs ($ millions) Ceramic
Immobilization plant capital		
Facility	398	399
Facility contingency (17.7%)	70	71
Auxillaries and equipment	101	101
Subtotal	569	571
Immobilization plant operating		
Production and inventory (20 yr glass, 7 yr ceramic)	538	196
Production (5 yr glass, 15 yr ceramic)	126	326
Subtotal	664	522*
Process development	65	80
Tank farm	9	0
Canisters ($5,500 ea.)	57	19
HIP cans ($3,000 ea.)	0	30
Subtotal	57	49
Storage	78	30
Transportation ($12,000 ea.)	124	41
Repository		
Capital ($31,250 ea.)	322	106
Operating ($60,500 ea.)	623	206
Subtotal	945	312
Total waste management costs	2,511	1,605

*Total operating costs were estimated at $\$515 \times 10^6$ if the ceramic plant was operated at a constant waste (current inventory plus new production) processing rate over the entire 22-year period.

operated, we have estimated development costs at $85 million, including designing, construction, and operation of an integrated pilot processing line. The tank farm and storage costs are the same as assigned by McDonell(17), who also assumed the ceramic process would start up in 1992. The canister, HIP can, transportation, and repository costs are shown in Table II. The transportation and repository costs are the same values as used by McDonell, but different repository capital allocations were used than those used by McDonell.

The data in Table II show that the immobilization plant capital and operating costs for the glass and ceramic options are essentially the same. The major portion in the $0.9 billion difference arises from different allocations in the repository. If the same method of allocating the repository costs were used, as used by McDonell, the difference would be $0.7 billion. Thus, when overall waste management costs are considered, the ceramic option has the potential of significant cost savings.

The Defense Waste Processing Facility (DWPF) initiative to manage SRP high-level wastes was launched in 1977 at which time glass was chosen as the reference waste form. Consequently, large investments have been made in glass development for this application and all preliminary design investments for the DWPF have been made, assuming glass would be the SRP waste form. The costs derived in this report have not considered factors associated with this development work and costs associated with modifying the environmental assessment and preliminary design to use ceramics instead of glass. Furthermore, repository cost methodology has not been finalized and therefore there can be large changes in repository cost. However, it should be pointed out that since only about one-third as many canisters of ceramic require repository emplacement as glass, the ceramic costs are much less sensitive to repository cost changes than glass costs.

The SRP waste management costs derived by McDonell(17) and Rosza(18) are given in Table III along with the data derived in this paper.

For their economic evaluation, Du Pont Engineering and SRL(17) used a preliminary ceramics flowsheet that was developed by LLNL and Rockwell ESG early in the Alternative Waste Form Development Program sponsored by DOE through SRL. Since very little process development work had been completed when the preliminary flowsheet was developed, a conservative approach was used; the processing equipment was not optimized for the ceramic process but developed largely from a knowledge of early glass processes. A processing canyon of 3125 m^2 (22,984 ft^2) was required to contain the equipment used in the preliminary process flowsheet. Because of the early stage of development, SRL assessed a 54% contingency on the capital cost and a large contingency (~30-35%) on operating costs due to the complexity of the preliminary process.(17)

TABLE III

COMPARATIVE COST USING GLASS AND CERAMIC WASTE FORMS TO MANAGE SRP WASTE
(Millions of Dollars)

	SRL(57) McDonell (4-82)		LLNL Rosza (8-82)		Rockwell, ESG This Report (2-83)	
	Glass[a]	Ceramic[b]	Glass[a]	Ceramic[c]	Glass[a]	Ceramic[b,d]
Development	65	180	65	180	65	80
Tank farm	9	--	9	9	9	--
DWPF Stage 1 capital and support	650	1100	636	817	569	571
DWPF operating	535	870	535	680	664	522
Canister procurement	103	131	103	105	57	49
Interim storage	78	30	78	30	78	30
Offsite transport	124	41	124	33	124	41
Repository	945	528	826	261	945	312
Total	2509	2880	2376	2115	2511	1605

[a] 10,300 canisters of glassified waste, 60-cm-diameter canisters at 25-28% waste loading.
[b] 3,400 canisters of ceramic waste, 60-cm-diameter canisters at 52-55% waste loading.
[c] Using maximum alumina dissolution from waste to reduce number of waste packages to 2,729 at 65% waste loading.
[d] Using new flowsheet with well-developed technology.

LLNL(18) reduced the size of the processing canyon designed by Du Pont Engineering and SRL(17) using the preliminary ceramic flowsheet by eliminating the ball mills and one HIP unit and used a (15%) contingency for those parts of the processing canyons which were the same (i.e., the auxiliary off-gas processing, feed preparation, and maintenance areas) but used a 35% contingency on those parts of the processing areas that were different for ceramics. Accordingly, LLNL also reduced the ceramic operating costs developed by SRL by eliminating the operating costs associated with the parts of the canyon that were no longer required. In addition, LLNL used more recent tuff repository costs. With these reductions, the overall waste management costs for Savannah River waste, as derived by LLNL using the ceramic option, was $2.1 billion compared with the $2.9 billion SRL estimate.

The further cost reduction in this report to $1.6 billion resulted from a redesign of the ceramic plant and a reduction in development costs and contingencies because full-scale demonstration of the major process steps has been accomplished. In addition, different capital repository allocations were used than used by McDonell.

CONCLUSIONS

The results tabulated in Tables II and III show that the differences in estimates dealing with the ceramic option are primarily due to different base designs and contingencies used for the solidification plant. The difference in waste management costs between the glass and ceramic options deduced from our calculations can be attributed to the lower repository cost allocation since only about one-third as many waste packages needs to be isolated.

These studies suffer from the same issues characteristic of all cost estimates dealing with future activities; i.e., lack of a quantitative data base. The merit of these studies, therefore, is to point out the critical cost parameters early in the decision making process and to focus on developing more quantitative data to test key cost premises.

Models for overall waste management costs and their allocations are still at the very early stages of development; this study provides some insight on important operating parameters and conditions. Repository costs could decrease or increase, depending on many future factors and decisions in waste management. The ceramic process, however, because of its flexibility and volume reduction, offers an alternative less sensitive to these future events and decisions.

With further ceramic development aimed at process integration and parametric optimization, the operating knowledge of full-scale demonstration of the key ceramic process steps should be rapidly adaptable to scale-up of the ceramic process to full plant size. It is concluded from this and other studies that the ceramic form offers important advantages over glass in thermal stability, structural integrity, leach-resistance, waste loading, density, logistics, and process flexibility. In addition, status of technology evaluations and preliminary economic calculations indicate that ceramics must be considered a leading candidate for the form to immobilize high-level wastes. The ceramic option not only offers a viable backup to the reference glass system, but promises to be a competitive future option for new nuclear waste management opportunities.

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