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Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management

Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
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ABSTRACT

The U.S. Department of Energy's (DOE) Hanford Site has an inventory of 217,000 m^3 of nuclear waste stored in 177 underground tanks. The DOE, the U.S. Environmental Protection Agency, and the Washington State Department of Ecology have agreed that most of the Hanford Site tank waste will be immobilized by vitrification before final disposal. This will be accomplished by separating the tank waste into high- and low-level fractions. Capabilities for high-capacity vitrification are being assessed and developed for each waste fraction. This paper provides an overview of the program for selecting preferred high-level waste melter and feed processing technologies for use in Hanford Site tank waste processing.

1.0 INTRODUCTION

The U.S. Department of Energy's (DOE) Hanford Site, located in southeastern Washington State, has the largest and most diverse amount of highly radioactive waste in the United States. This high-level radioactive waste (HLW), consisting of approximately 217,000 m^3 (57 Mgal) of alkaline liquids, slurries, salt cakes, and sludges, has been stored in 177 large, underground tanks since 1944. In addition, significant amounts of ^{90}Sr and ^{137}Cs were removed from the tank waste, converted to salts, doubly encapsulated in metal containers, and stored in water basins.

A Tank Waste Remediation System (TWRS) Program has been established by the DOE to safely manage and process Hanford Site tank waste in preparation for permanent disposal. The mission of the TWRS Program is to store, treat, and immobilize the Hanford Site tank waste and capsules in an environmentally sound, safe, and cost-effective manner. The goal of the TWRS Program is to pretreat tank waste by fractionating it into low-level waste (LLW) and HLW streams followed by immobilization into a vitrified product. The LLW fraction will undergo near-surface disposal onsite and the HLW fraction will go to the national geologic repository.

The current program framework for achieving this goal was established in 1993 under the *Hanford Federal Facility Agreement and Consent Order*.[1] This agreement, signed by the Washington State Department of Ecology, the U.S. Environmental Protection Agency, and the DOE, is commonly referred to as the Tri-Party Agreement. A revised plan adopted at that time sets time lines for establishing technical capabilities, processing plant operations, and completion of processing for all tank wastes. This agreement establishes specific milestones for selection and demonstration of vitrification technologies before detailed design of the LLW and HLW vitrification plants. The pertinent major milestones for each waste fraction are as follows.

<u>LLW</u>

- Begin LLW melter testing with simulants - September 1994
- Select reference melter and glass formulation - June 1996
- Begin detailed plant design - November 1996
- Initiate hot operations - June 2005

<u>HLW</u>

- Complete melter tests and select reference melter - September 1998
- Initiate definitive design - December 1998
- Complete construction - December 2007
- Initiate hot operations - December 2009.

The following sections provide: information on the general characteristics of LLW and HLW to be processed; the TWRS Program approach to vitrification process technology selection and evaluation with emphasis on Hanford Site HLW processing needs; and a review of the criteria and selection process used in HLW melter technology evaluation; HLW melter technology candidates and development considerations; key conclusions of the evaluation; and program structure for development of selected HLW melter technologies.

2.0 GENERAL CHARACTERISTICS OF TANK WASTES AND VITRIFICATION FEEDS TO BE PROCESSED

Hanford Site radioactive tank wastes were produced primarily from reprocessing of irradiated fuel from plutonium production reactors using the bismuth phosphate process (1944 to 1956), reduction oxidation process (1952 to 1966), and plutonium-uranium extraction

(PUREX) solvent extraction process (1956 to 1972, 1983 to 1988). Over a 12-year period, high-heat wastes were reworked in the Hanford Site's B Plant to recover ^{137}Cs and ^{90}Sr by ion exchange and solvent extraction followed by encapsulation.[2]

The wastes converted into mixed sludges and salt cake were initially routed to single-shell tanks (SST). More recent wastes from the PUREX process and supernates removed from the SSTs have been consolidated in double-shell tanks (DST). The Hanford Site wastes are stored in 149 SSTs containing approximately 136,800 m^3 (36 Mgal) of salt cake, sludge, and residual liquid with 460 x 10^{16} Bq (125 MCi) and 28 DSTs containing 80,000 m^3 (21 Mgal) of liquids, salts, and sludges (the majority of tank volume is liquid) with 310 x 10^{16} Bq (85 MCi). In addition to the wastes stored in the tanks, there are approximately 1,900 6.7-cm-diameter by 52-cm-long Cs/Sr capsules containing approximately 600 x 10^{16} (160 MCi) total. The predominant chemical inventory in the SSTs and DSTs is shown in Table I.[3]

The overall treatment method for retrieval, pretreatment, vitrification, and storage of Hanford Site tank waste and Cs/Sr capsules is shown in Figure I.[4] Retrieval and pretreatment will prepare waste for vitrification. Most tanks will be retrieved in a manner to obtain separation of soluble and insoluble material. Soluble salts and supernate solutions will be staged for pretreatment as LLW vitrification feed. Sludge retrieved from SSTs will be consolidated in DSTs for in-tank pretreatment and staged to HLW vitrification.

Pretreatment of the Hanford Site tank wastes is intended to minimize the volume of HLW chemicals to be vitrified and to separate radionuclides to achieve acceptable regulatory criteria imposed on the LLW vitrified glass product. Cesium and possibly other radionuclides will be removed from the LLW stream by ion-exchange processes and combined with the HLW tank fraction (washed solids resulting from HLW pretreatment process).

Encapsulated Cs and Sr waste may be blended into the HLW feed stream or packaged for disposal in a repository.

The LLW vitrification feed stream is significantly larger than the HLW stream (1,070,000 MT of LLW versus approximately 154,000 MT of HLW).[5] An example of an all-tank blended LLW feed stream, Cs ion-exchange stream, and HLW sludge chemical inventory is shown in Table II.

The HLW feed compositions could vary significantly from the all-tank blend composition (Table II) because of tank retrieval

Table I. Estimated Chemical Inventory of Hanford Site Single- and Double-Shell Tank Waste

Major components	SST (in MT) (% in SSTs)	DST (in MT) (% in DSTs)
Na^+	56,000 (23%)	10,000 (11%)
NO_3^-	99,000 (42%)	7,000 (7%)
H_2O	52,000 (22%)	62,000 (67%)
Others	30,000 (13%)	14,000 (15%)
Phosphate	4,700 (2%)	--
Hydroxide	5,800 (2%)	1,800 (2%)
Nitrite	6,500 (3%)	2,900 (3%)
Cancrinite	3,000 (1%)	3,800 (4%)
Aluminum	2,000 (1%)	--
Carbonate	1,700 (<1%)	1,800 (2%)
Sulfate	1,600 (<1%)	--
Potassium	--	500 (<1%)
Organic carbon	--	900 (1%)
Zirconia	--	300 (<1%)
Balance	4,700	2,000
Total	237,000 MT	93,000 MT

sequencing and blending constraints. Without blending, many of the individual tanks contain components that are expected to limit the amount of waste that can be incorporated in the glass (waste loading).[2]

3.0 PROGRAM APPROACH TO VITRIFICATION PROCESS TECHNOLOGY SELECTION AND EVALUATION

As determined by the Tri-Party Agreement milestone structure, timing of the technology selection for HLW and LLW is different in that the LLW melter selection is required to support facility design 3 years ahead of HLW. The TWRS Program approach to selection of the LLW vitrification melter system is to target adaptation of large-scale commercial or mixed/hazardous waste vitrification process technology through a competitive procurement demonstration process. Details and status of low-level tank waste vitrification technology assessment are provided by Wilson et al. at this symposium.[6]

The approach for HLW seeks to build on the existing technology base for fully remote, high-radioactivity level processing systems. An evaluation and selection process established by

Figure I. Treatment Methods for Hanford Site Tank Waste and Capsules.

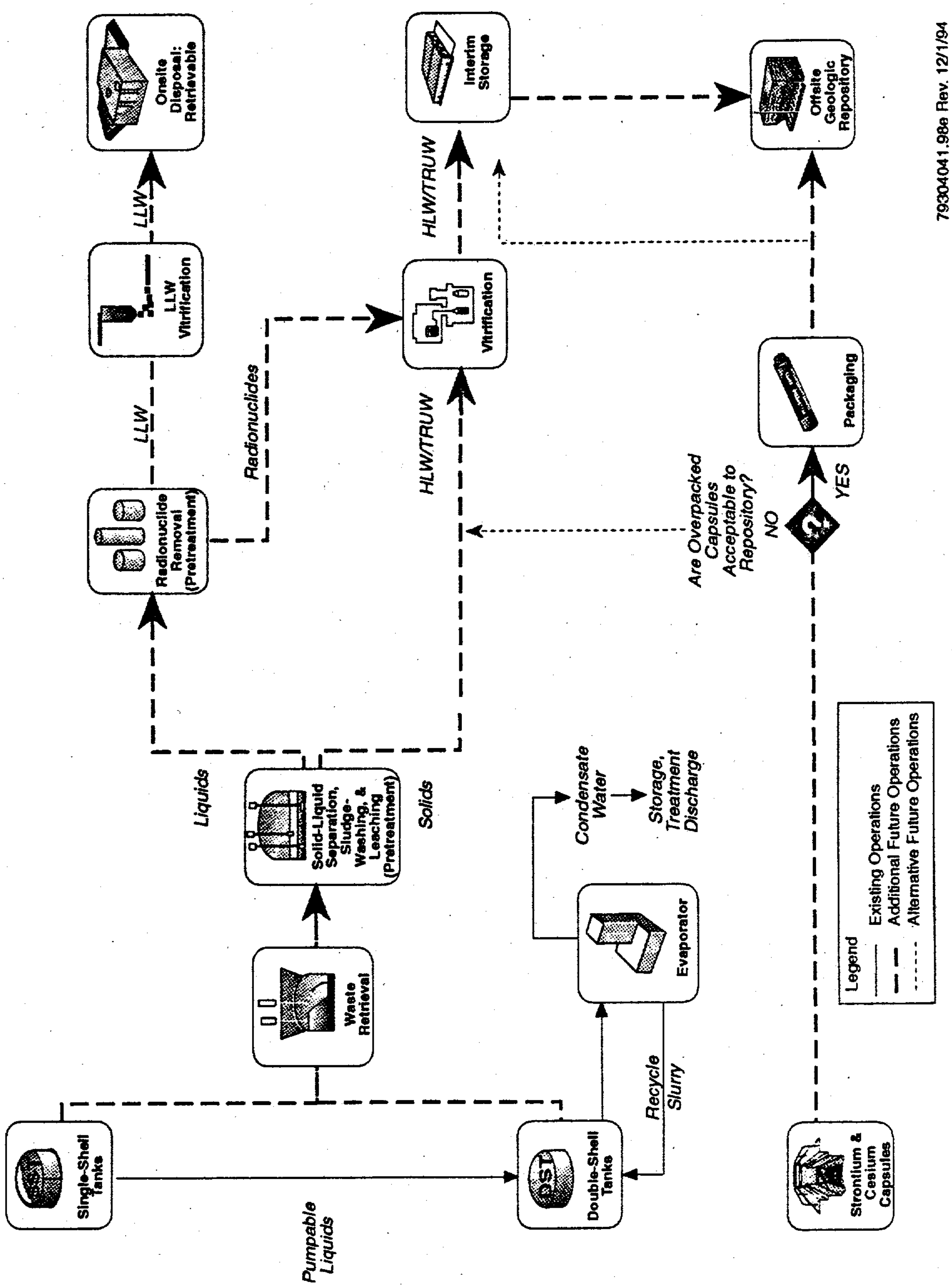


Table II. Waste Separations - Streams to Vitrification

	LLW feed stream	Cs ion-exchange stream	HLW sludge
Total mass flow (MT)	1.07 E+6	4.88 E+3	1.49 E+5
Component flow (MT)			
Al	4.11 E+3	7.95 E+0	7.16 E+2
Fe	9.92 E+0	6.77 E+0	8.04 E+2
Cr	1.32 E+2	3.30 E-1	3.23 E+1
Na	7.84 E+4	2.58 E+2	1.72 E+3
Si	1.35 E+1	3.26 E+1	4.89 E+2
P	1.67 E+3	2.59 E+0	2.14 E+2
NO_2^- and NO_3^-	1.21 E+5	1.60 E+3	1.30 E+3
H_2O	8.23 E+5	2.79 E+3	1.36 E+5
Radionuclides			
Cs and Ba (MCi)	7.69 E-1	9.49 E+1	1.11 E+1
Sr and Y (MCi)	5.05 E+0	1.30 E+0	1.34 E+2
Tc (kCi)	3.50 E+1	4.07 E-1	9.15 E+0
TRU (kCi)	7.97 E+0	1.48 E+0	1.76 E+2
Total (MCi)	5.86 E+0	9.62 E+1	1.46 E+2

Westinghouse Hanford Company (WHC) has identified technologies consistent with TWRS Program needs and defined the development program.

Studies performed and decisions made in 1992 and 1993 resulted in the need for significant expansion of vitrification capacity for HLW. The DOE's decisions to retrieve all the DST and SST waste for processing and to use only in-tank processing for pretreatment drastically changed the volume of waste to be processed. The increased waste volume is estimated to require a glass production rate of 400 to 800 kg/h versus the previous Hanford Waste Vitrification Plant design capacity of 100 kg/h.[7] The need for increased capacity in a new plant design was the primary driver for evaluating melter systems with higher throughput capability and potential for higher waste loading.

4.0 ASSESSMENT OF VITRIFICATION TECHNOLOGIES FOR DEVELOPMENT TO MEET HANFORD SITE HLW PROCESSING NEEDS

The initial step in the HLW melter system assessment[8] was a survey of potentially viable HLW melter system technologies. Melter system technologies currently being used for vitrification of HLW, those previously considered for HLW processing, and other existing technologies with potential application were identified by the Pacific Northwest Laboratory (PNL) and approved by WHC for initial consideration by an assessment team. The assessment team established the final list of technologies. Pacific Northwest Laboratory also provided best available information relative to each technology considered in the assessment.

Participants in the assessment included a core team of WHC technical staff augmented by outside experts. These outside experts were responsible for developing and preparing the final summary recommendations. Other participating technical support personnel included PNL and Savannah River Site staff and representatives from several foreign HLW vitrification programs.

The assessment was performed in two phases, each culminating in a workshop to bring participants together to perform the review, ranking, selection, and recording of results.

The technologies were evaluated using a list of minimum requirements in Phase I with six of the most viable technologies selected for a more in-depth evaluation in Phase II. For those technologies considered in Phase II, data packages[9] were prepared by PNL and reviewed by peers for use in the Phase II workshop.

The top six technologies selected for Phase II evaluation were judged on probability of success against minimum screening requirements in Phase I. Phase I screening requirements were (1) capability to produce consistent, acceptable waste form; (2) basis to successfully develop required technology within TWRS Program constraints, including Tri-Party Agreement milestones; and (3) reasonable total program cost compared to other technologies.

All the initial melter system candidates are presented in the following list. The list is separated into those technologies considered in Phase II of the assessment and technologies dropped from consideration in Phase I. The two technologies dropped from consideration in the Phase II assessment are identified separately at the bottom of the list for Phase II technologies.

<u>Technologies considered in Phase II assessment</u>

- Low-temperature (≤1200 °C), ceramic-lined, joule-heated melters

- Low-temperature (1050 °C), metal-lined, stirred, joule-heated melters

- High-temperature (>1200 °C), ceramic-lined, joule-heated melters

- High-temperature, metal-lined, joule-heated melters (cold wall)

Technologies dropped during Phase II assessment

- Low-frequency, metal-can, induction-heated melters
- High-frequency, cold-wall, induction-heated melters.

Technologies considered in Phase I but not considered in Phase II assessment

- Plasma torches (entrained and indirect)
- Transferred-arc plasma melters
- Arc furnaces
- Conventional combustion melters
- Cyclone combustion melters
- Microwave melters
- Rotary kilns
- Hot isostatic presses
- In-can melters.

5.0 CRITERIA/ATTRIBUTES AND SELECTION PROCESS KEY FEATURES

For the six technologies selected for detailed assessment in Phase II, a significant information base was compiled by PNL staff for each candidate in preparation for the Phase II workshop.[9] This information and the expert judgement of the assessment team were used to assess each technology relative to evaluation criteria. The Phase II evaluation criteria included the ability to do the following.

- Process a range of compositions.

- Control product quality.

- Develop technology on schedule.

- Integrate with process and facility.

- Minimize total cost.

- Minimize safety and environmental risk.

- Minimize other risks and limitations (e.g., institutional barriers).

The technologies were compared with each Phase II evaluation criterion to establish a ranking. The criteria and specific attributes that were addressed are shown in Figure II.[8]

6.0 KEY CONCLUSIONS FROM THE ASSESSMENT

Two technologies [(1) the high-temperature, metal-lined, joule-heated melters (cold wall), and (2) the low-frequency, metal can, induction-heated melters] scored well below the other four Phase II technologies relative to the majority of criteria. The assessment team reached consensus to drop these technologies from consideration as potential HLW melter system technologies.

The high-temperature, metal-lined (cold wall), joule-heated melter system scored low primarily because of fundamental technical concerns about control of bulk melter temperatures, electrode performance, product quality, and the relative immaturity of the technology for HLW vitrification.

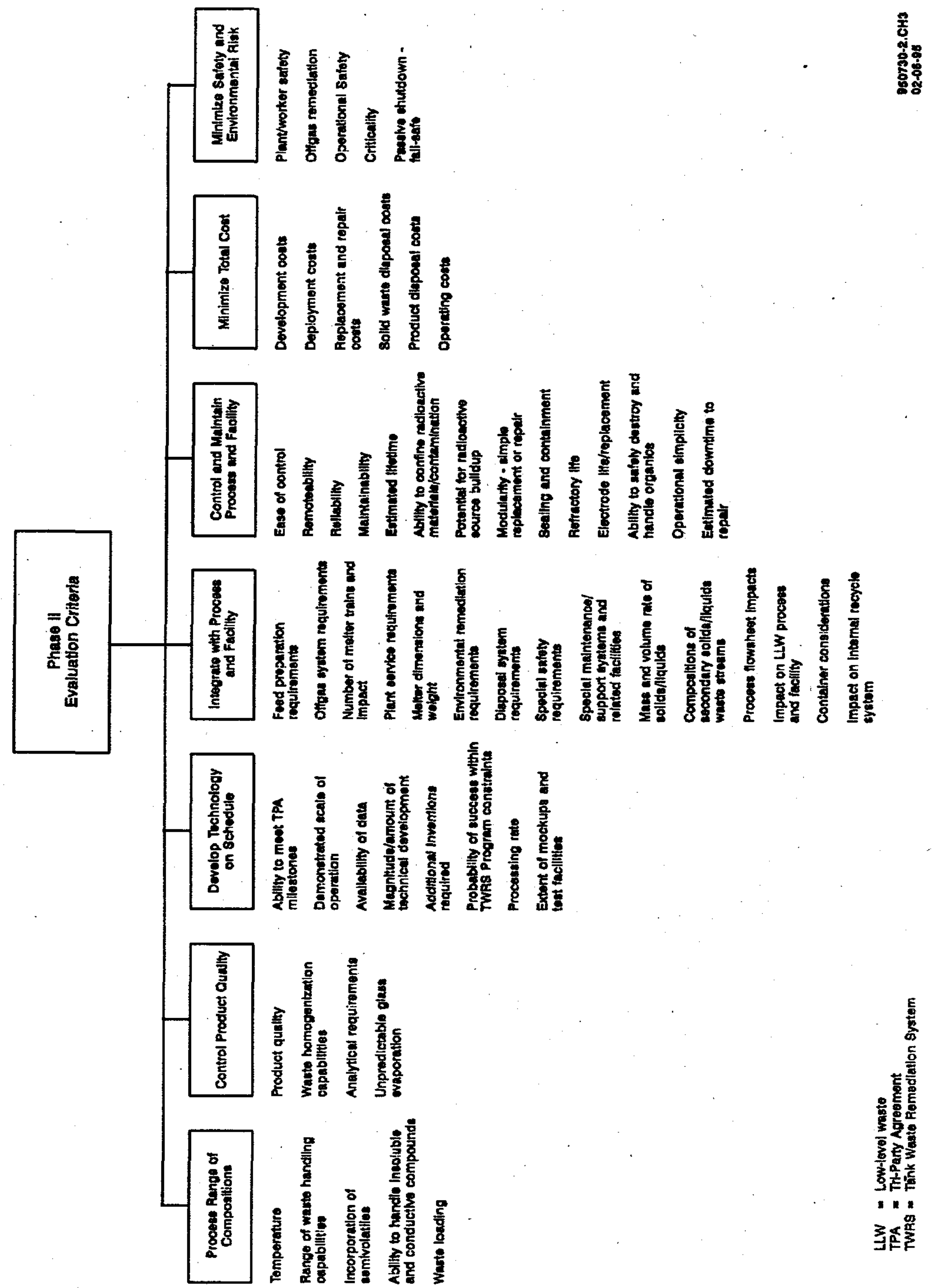
The low-frequency induction technology was dropped based primarily on cumulative concerns regarding required feed drying of Hanford Site alkaline feeds, limited operating temperature range, and limited unit capacity. The large number of melter units and supporting equipment required to process Hanford Site wastes in these melters would pose significant challenges in designing, operating, and maintaining such a plant.

Based on assessment scoring results, it was apparent that key discriminators among the remaining four technologies were development status of the technologies and operating temperature regime. The operating temperature influences waste processing flexibility and the total volume of glass product requiring disposal. These two major factors strongly influenced the regrouping of the final four technologies and the basis for selection of primary and backup technologies. As an example of the effect of melter operating temperature on the waste loading, an all-tank HLW vitrification feed is estimated to increase from 45 wt% at 1050 °C to 62 wt% at 1350 °C.[2]

The final four technologies were regrouped into one primary technology and a backup technology. The primary technology is joule heating with various electrodes (three variants). The backup technology is high-frequency induction heating.

The primary technology was judged to provide a solid technical base for HLW vitrification of Hanford Site wastes with reasonable plant life-cycle costs. This technology has nickel-based electrodes with an operating temperature limited to 1150 °C.

Figure II. Phase II Evaluation Criteria.



Alternate electrode materials capable of higher temperature operation (up to 1500 °C) could allow higher waste oxide loadings and process flexibility. Higher temperature electrode materials for this technology are being developed and tested at PNL.[10] In addition, the size and number of melters required to process Hanford Site feeds could be significantly reduced by feeding drier feed and by agitation of the melt pool thereby increasing glass production capacity. Melter designs that allow for sludge accumulation or periodic sludge removal may provide extended melter life.

High-frequency induction melting[11, 12] was judged to be a strong backup technology because of its development status for vitrification of HLW, the benefit of high-temperature operation, and potential for long melter life. This technology was recommended as backup because of a lack of test data with Hanford Site-type feeds, need to dry the melter feed to provide optimum capacity, and current lack of large-scale system and power supply demonstrations.

7.0 SUMMARY OF PROGRAM STRUCTURE FOR DEVELOPMENT OF HIGH-TEMPERATURE MELTER, COLD-CRUCIBLE MELTER, AND BACKUPS

The Hanford Site Program HLW melter development strategy is based on general testing activities used to develop melter systems at the Savannah River Site and Hanford Site, and international vitrification programs for radioactive waste immobilization. The approach is to test the selected melter technologies in a phased development sequence. The smallest, most cost-effective test systems that use nonradioactive simulants will be tested in the initial phases to resolve key technical issues. The major issues are associated with process chemistry, melter performance, feed processibility, and glass product quality. In the subsequent phase, large-scale, nonradioactive test systems will be used to confirm small-scale test results and demonstrate plant-scale performance and operability. Testing with radioactive waste feeds will be limited to small-scale systems. That work will develop the necessary correlation between nonradioactive simulants and actual radioactive waste feeds for equivalence in process chemistry, feed processibility, and glass product properties. All phases of testing are timed to provide the data to support key TWRS HLW Program decisions. Development and testing of the high-temperature systems (joule-heated, ceramic-lined, and high-frequency induction-heated melter systems) will be emphasized in view of significant potential cost savings (1 to 3 billion U.S. dollars) to be realized from reduced glass volume requiring expensive disposal.

8.0 CONCLUSIONS

Key conclusions from the HLW melter system assessment and program implementation plan are as follows.

- Diversity of waste source compositions on the Hanford Site represents incentives for blending wastes and for flexibility in glass melting process capability.
- All electric, cold-top melters with nickel-based electrodes, developed initially at the Hanford Site and further developed for worldwide use, provide a solid base technology for HLW vitrification of Hanford Site wastes.
- Increased melter temperatures offer the potential for significant reductions in total program costs. The first choice for increasing temperature is to develop alternative high-temperature electrode concepts. Other methods for improving melter performance, such as agitation, feed drying, and sludge mitigation geometries, will be investigated.
- High-frequency induction melting offers potential relief from problems of high-temperature electrode attack, but is considered a backup technology because of the relative immaturity of the melters and power supplies. Initial testing will focus on potential compatibility issues specific to the high-pH, high-sodium level of Hanford Site wastes.
- Development and testing will employ small- and large-scale test systems with the balance determined by cost/benefit considerations of the development program.

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