HLW COMPLIANCE PLAN FROM A WASTE PRODUCER'S PERSPECTIVE

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ABSTRACT

In 1980 the U.S. Congress passed the West Valley Demonstration Project Act which directs the U.S. Department of Energy (DOE) to solidify the High-Level Waste (HLW) stored at West Valley, New York into a form suitable for disposal. The immobilization medium selected is borosilicate glass, and the vitrification process is via the joule-heated, slurry-fed ceramic melter. To ensure that the HLW form generated by the West Valley Demonstration Project (WVDP) is suitable for disposal, the DOE implemented the Waste Acceptance Process (WAP), which is developing the Waste Acceptance Preliminary Specifications (WAPS).

As part of the WAP, the WVDP and other waste producers will have to develop several plans and reports of data on its waste form and process. This includes the WVDP Waste Form Description which was issued in July 1986. The Waste Compliance Plan (WCP) describes the methods and tests that will be used to demonstrate that the WAPS are met. The WVDP WCP will include discussions on the methodology for determining glass properties, techniques for insuring production of acceptable glass, and performance requirements for canisters.

The current WVDP schedule calls for initial production of radioactive waste glass in the Spring of 1989. To meet the requirements of the WAP, the project's WCP must be issued, reviewed, and approved well in advance of this date to allow sufficient time for preparation of all qualification data on waste form characterization.

INTRODUCTION

In 1980 the U.S. Congress passed the West Valley Demonstration Project Act (PL96-368) which directs the Department of Energy (DOE) to solidify the liquid high-level waste (HLW) remaining at the former commercial nuclear fuel reprocessing plant at West Valley, New York. To ensure that the Nuclear Regulatory Commission (NRC) is properly consulted during WVDP activities, the DOE and NRC agreed to a memorandum of understanding.

Borosilicate glass is the waste form selected by the DOE. The wastes that will be immobilized in the glass are PUREX solids, THOREX waste, and zeolite IE-96. The PUREX solids are predominately hydroxide precipitates from the neutralization of a nitric acid solution. The THOREX waste is predominately nitrates in a nitric acid solution. The cesium in the PUREX supernatant will be removed by the zeolite ion exchange material. The decontaminated supernatant will be solidified in the Cement Solidification System (CSS) and treated as low-level waste. The cesium loaded zeolite will be mixed with the PUREX solids, THOREX waste, and glass formers and melted in the West Valley melter. The nominal waste loading of the glass will be about 33 weight percent waste oxide: 23 percent from the PUREX solids and THOREX waste, and 10 percent from the zeolite.

A block diagram of the reference WVDP HLW vitrification process flow sheet is shown in Fig. 1. The zeolite and THOREX waste will be transferred to and mixed with the PUREX solids in the HLW Storage Tank 8D-2 where the PUREX waste is currently stored. The slurried waste will be transferred to the Concentrator Feed Makeup Tank (CFMUT), sampled, and concentrated. Alternately the waste may be transferred independently from the three waste tanks to the CFMUT and then mixed. After mixing, sampling, and analysis, an appropriate quantity of glass formers will be added to the CFMUT to make the appropriate melter feed. After concentration and final mixing, the waste will be transferred to the Melter Feed Tank and then metered to the Slurry Fed Ceramic Melter via an air displacement pump. After melting, molten glass will overflow into a stainless steel canister to become the solidified product. All of the major components shown in Fig. 1, except for the CFMUT, have been installed and are undergoing functional and checkout testing. The full scale equipment will be used for waste certification testing.

The DOE is developing WAPS for the WVDP HLW Form (1) that will establish minimum requirements that WVDP HLW must meet to be compatible for disposal in a repository. By showing compliance with these specifications, the WVDP will be assured that its canistered waste form will interface properly with the repository. To assist this process, the WVDP prepared "Description of the West Valley Demonstration Project Reference High Level Waste Form and Canister" (2). The techniques that are being developed to show that the specifications will be met will be provided in a WCP. Some of the techniques being considered for the WVDP WCP were reported previously (3).

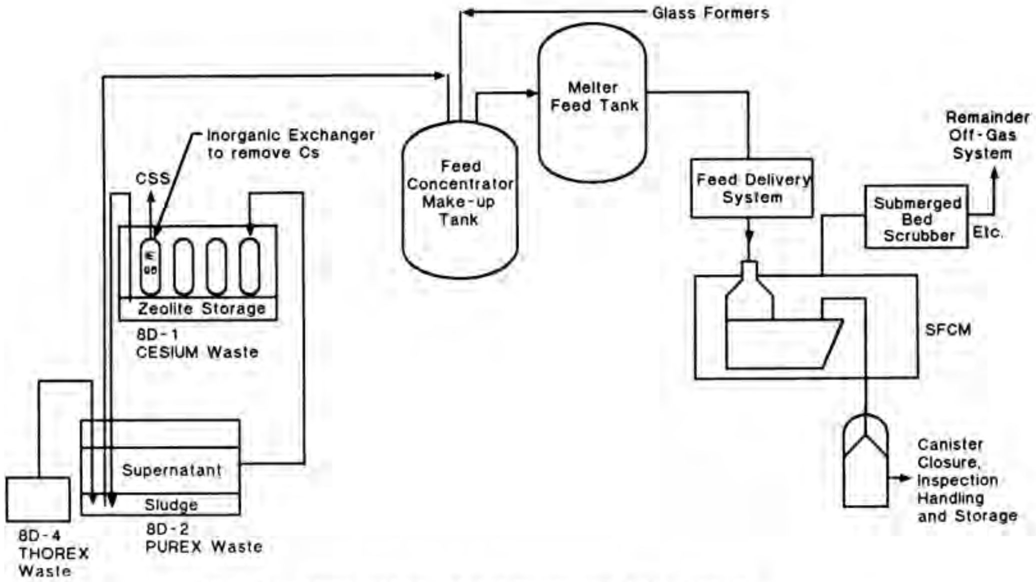

Fig. 1. West Valley HLW Processing Flow Sheet.

This paper updates those techniques. From the perspective of the WVDP, it is imperative that the specifications and data necessary to show compliance is available and approved to support the start of waste vitrification during the spring of 1989. A summary logic diagram of these activities is shown in Fig. 2. For waste acceptance, process qualification testing will verify that the techniques used to provide the glass composition are adequate. West Valley glass testing has begun as has preliminary process testing. The reference West Valley canister has been designed and is being tested to assess its integrity. Administrative controls are being developed for those specifications that require inspections, measurements, etc., to show compliance. The reporting of the test date will be through the Waste Qualification Report (WQR). To ensure that sufficient time is available for review of the data, the WVDP plans to submit its WQR in sections. As data is generated, it will be made available to the interested organizations.

TECHNIQUES FOR PROVIDING GLASS COMPOSITION

Two methods are being considered for providing the chemical composition and radionuclide inventory of the waste glass: sampling the feed and relating its composition to that of the glass by way of a process mass balance model, or sampling the radioactive waste glass. These are discussed below. The decision on which method to use will be based upon the overall achievable precision and accuracy, capability to develop and implement the method by the beginning of waste vitrification, ease of use during system operation, and overall cost.

Compliance Method No. 1

The waste form chemical composition would be based upon a simplified process mass balance model that would predict the composition of the glass in the canister. The WVDP would report the waste glass composition by sampling and analysis of the waste slurry, and by adding known amounts of nonradioactive chemicals to the waste. The mass balance would assume that either the constituents go into the canister or to the process off-gas.

During full-scale nonradioactive testing in the actual waste vitrification equipment, frequent samples of the slurry feed, draining glass, and canistered glass would be taken and analyzed. This would form the data base that assures that the waste glass is as predicted by the model. An example of a process model for waste vitrification and how it can be used is discussed in Ref. 4.

During production, the following would be inputs to the model:

- Chemical analysis of the waste based on the sample from the CFMUT that corresponds to the glass in the particular canister.
- Concentrator tank level at the time of sampling.
- Chemicals added to the CFMUT to make the slurry melter feed.
- Melter plenum temperature and average slurry feed rate.

The advantage of this method is that equipment to remove and handle radioactive glass samples from the canister or glass pour stream does not have to be designed, fabricated, and tested.

Compliance Method No. 2

During full-scale nonradioactive testing in the actual waste vitrification equipment, frequent samples of the draining glass and canistered glass would be taken, analyzed, and reported. Samples of the canistered glass would be removed from the top of the glass and compared with the results from the bulk of the casting and with the drain glass samples. It would be demonstrated that the shards and glass "hair" that can be removed from the canister opening would be representative of the bulk glass composition. It would further be determined how frequently the canisters must be sampled to demonstrate that the composition of the waste form falls within the acceptance region when process variations and upsets are considered.

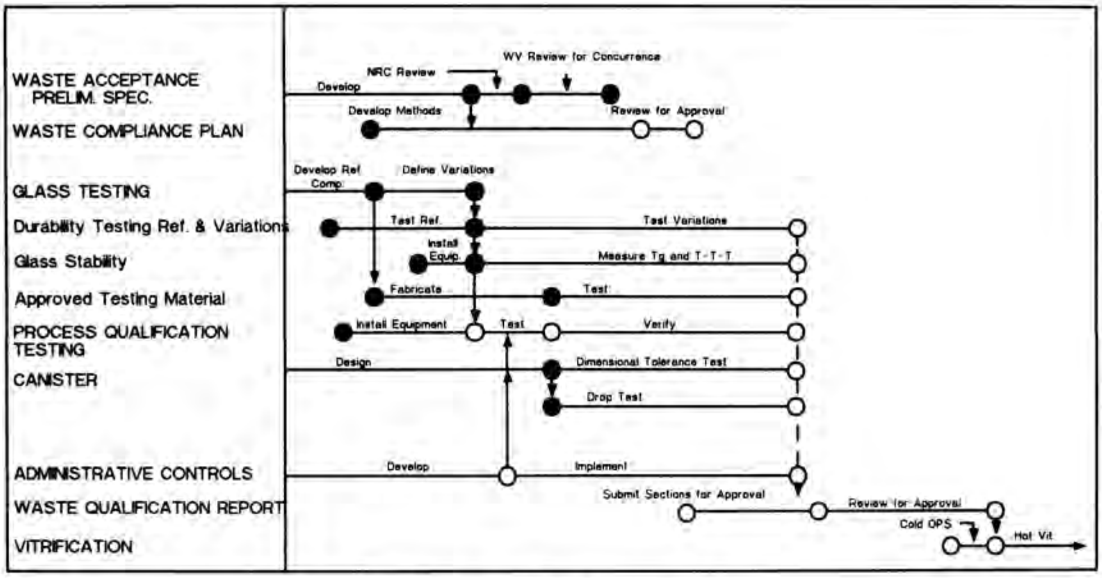

Fig. 2. Waste Compliance Logic.

During waste vitrification, the glass would be sampled after the canistered waste form is removed from the turntable. The frequency that samples would be removed will be determined during cold testing; it would be based on the sampling variability observed. A glass shard or glass "hair" would be removed from the top of the canistered glass, transferred to the analytical laboratory, and chemically analyzed for composition. The production records would include the chemical analysis results of the glass shard or "hair" that was analyzed.

The advantage of this method is that a fully developed statistical process control program is not needed. Such a process control program requires that a large amount of data be collected and analyzed during cold test melter runs. A process control program with technical specifications will be needed to ensure that a processible melter feed is fed to the melter, but a much smaller data base will be required. Limiting the amount of data collection and analysis is a significant cost and schedule savings.

Radionuclide Inventory

The same approach used to provide chemical composition during waste vitrification will be used to provide radionuclide inventory in a canistered waste form, i.e., one of two methods will be used. Either a sample of the waste feed will be analyzed for the radionuclide inventory and related to the inventory in the glass by a process model, or a glass shard or "hair" will be removed from the top of the canistered glass and analyzed for the radionuclide inventory.

During vitrification, the sample will be analyzed for Sr-90, U-235, Pu-238, Pu-239/240, and gamma emitting isotopes. Values for the remaining radionuclides will be obtained by using scaling factors developed during the West Valley HLW Characterization Program (5-7). Scaling factors for those radionuclides that will not be directly measured but have half-lives greater than ten years and are expected to be in concentrations greater than 0.05 percent of the radioactivity inventory will be developed from a waste sample removed after sludge homogenization. This will ensure that a representative sample is used to develop the scaling factors. The weight of glass in each canister will be measured and will be included in the production records for each canister. From this, and the specific activity in the sample, the amount of each radionuclide present in each canister will be estimated and reported in production records. ORIGEN2 calculations will be used to calculate the radionuclide content of each canister at time of shipment and to ensure that all radionuclides with half lives greater than ten years in concentrations greater than 0.05 percent of the total radioactive inventory at any time up to 1,100 years after production will be reported.

GLASS TESTING

Durability Testing

To meet the intent of the Specification for Radionuclide Release Properties, the WVDP is performing tests to characterize its glass for radionuclide release properties. This is the most important property of the waste form, because it describes how the glass behaves after disposal in the repository.

The WVDP is performing Interactive Powder Leach Tests (MCC-3 type), and Partial Exchange Interactive Flow Tests (8-9) on the sets of glasses discussed below. The flow test is a repository relevant test where leachate is removed and replaced at intervals to simulate flow through a repository. The powder test provides a quick response, but does not provide as much information as does the flow test. For example, flow test data can be analyzed to assess the leaching mechanism. However, the powder test results can be related to the flow test. The test temperature is 90^{0}C. Generally, the test durations are seven days for the Interactive Powder Leach Test, and until equilibrium is established for the flow test, generally 6 to 12 months. The flow test can be allowed to continue beyond this time to obtain longer term results. The replacement interval for the Flow Test is being selected to simulate the flow expected in a repository. This selection will be based on reviewing the repository projects technical literature and on discussions with the repository projects. Each test performed on an acceptable glass will be in triplicate.

The sources of the test glasses are:

- ATM-10, an Approved Test Material fabricated by the Materials Characterization Center (MCC) incorporating elements in WVDP HLW; it is doped with Am, Np, Pu, Tc, Th, and U. See Table I for the target composition.
- A small scale melter; the glass is doped with full levels of Th and U. Various compositions around the nominal composition will be melted.
- The WVDP Component Test Stand (CTS) melter; appropriate elements will be used as substitutes for the radioactive elements. Various compositions will be melted.
- Lab scale crucibles; some glass will be doped with Th and U and some will have appropriate elements substituting for the actinides. Various compositions will be melted.

TABLE I

TARGET COMPOSITION FOR ATM-10 GLASS

Oxide	Target Wt. %
Al_2O_3	6.50
AmO_2	0.0068
B_2O_3	9.26
BaO	0.05
CaO	0.56
CeO_2	0.07
Cr_2O_3	0.29
Cs_2O	0.07
Fe_2O_3	11.31
K_2O	3.33
La_2O_3	0.03
Li_2O	2.82
MgO	1.21
MnO_2	1.22
Na_2O	10.17
Nd_2O_3	0.17
NiO	0.32
NpO_2	0.0208
P_2O_5	2.33
PuO_2	0.0085
RhO_2	0.01
RuO_2	0.07
SO_3	0.27
SiO_2	44.90
SrO	0.03
Tc_2O_7	0.0030
ThO_2	3.34
TiO_2	0.91
UO_2	0.52
Y_2O_3	0.02
ZrO_2	0.27
TOTAL	100.09

ATM-10 is a homogenous glass and will be used to obtain durability information especially on those radioactive elements that are used as dopants. The MCC will also make ATM-10 available to the repository projects for their testing programs.

Glasses doped with Th and U of varying compositions will be melted and tested to provide a phase field in which the glass will be characterized. The compositional boundary for the elements listed in Table II will be investigated singly, i.e., one element will be raised or lowered at a time. Periodically, the results will be reviewed statistically to ensure that the results are meaningful and that the next set of components to be studied are the proper ones. This will provide an envelope within which the glass is acceptably durable. Processibility properties, e.g., viscosity, will also be measured. There will be an intersection of two composition sets, i.e., durable compositions and processible compositions, within which the WVDP will generate glass (See Fig. 3).

A nominal glass composition with Th and U will be remelted at various redox states and characterized by the above tests to assess the effect of glass redox state on durability. The value of the redox state of the glass is a result of processing conditions. The redox state that processes optimally will be identified. Glasses of that redox state and of redox states on either side will be tested as described above in DIW. This will provide the effect of changes in redox state that could be generated during waste vitrification.

During cold testing the cooling rate of the glass in the canister will be monitored such that the test specimens fabricated in the laboratory can be heat treated to simulate the canistered glass. After monitoring the glass temperatures in a series

TABLE II

ELEMENTS THAT WILL BE VARIED TO DEFINE THE DURABILITY BOUNDARY

Element	High	Low
Si	X	X
Th	X	X
Fe	X	
P	X	X
B	X	
Na	X	
Cr		X
Al	X	X
U	X	X

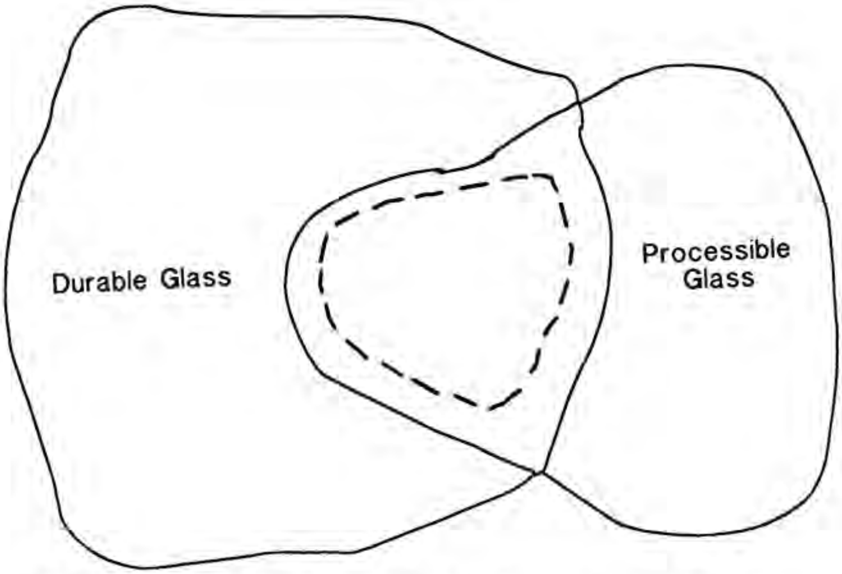

Fig. 3. Strategy for Achieving a Durable and Processible Glass.

of canisters, a reference cooling heat treatment will be developed, it will simulate the heat treatment of the glass at about 4 cm from the surface. Specimens of a nominal glass composition containing Th and U will then be subjected to this heat treatment and characterized by the above durability tests. This will provide an assessment of the effect of the cooling rate on durability.

In addition to deionized water, ground waters expected to be found in repositories will be used in the above tests. The tests will be performed using a glass of nominal composition. This testing will provide a correlation between different leachates.

To monitor the potential bias of the results within an individual laboratory, several facilities will be used to perform these tests. The Vitreous State Laboratory (VSL) of the Catholic University of America is testing the glasses of various chemical compositions and redox states, the heat treated glasses, and glass in repository ground waters. Pacific Northwest Laboratory (PNL) will overlap the testing of selected chemical compositions with the Interactive Powder Leach Test. The MCC will test ATM-10 and a glass of similar composition doped with Th and U. As a courtesy, the Savannah River Laboratory (SRL) will be performing static tests on WVDP glasses to compliment their testing program. The WVDP will continue discussions with the repository projects about the use of WVDP glasses in their testing programs.

To assist in correlating the WVDP data with other glass data, the results of this testing will be compared to models on glass durability. This will include models developed at VSL, (9-10), SRL (11), and the Hahn Meitner Institute (HMI) (12). Generally these models explain the long term behavior of the glass by using thermodynamic and kinetic approaches. The VSL model relates the test results to behavior in the repository by comparing the experimental and repository leachate contact time and the experimental and repository glass surface area to leachate volume ratio. The SRL model correlates the free energy of hydration of the glass (which is related to its composition) to MCC-1 leach rates of Si. HMI uses the PHREEQE computer code, a geochemical modeling program, which follows a reaction path for the dissolution reaction; it considers hydrolysis and complexing of ions in solution and precipitation of secondary minerals.

Some of the results of this testing are reported elsewhere (13-16). They indicate that WVDP glass behaves comparably to glass expected from the Defense Waste Processing Facility and will be acceptable for disposal.

Thermal Stability

To ensure that the temperature effects during storage and repository disposal are known and that the glass is phase stable, the thermal characteristics of the glass are being analyzed. This includes measuring the glass transition temperature and developing time-termperature-transformation (TTT) data.

The glass transition temperature is a commonly used property. The glass transition temperature is that temperature reached on cooling when a super cooled liquid becomes a glass. It is a marked change in atomic mobility. Typically, the glass transition occurs over a short temperature range of about 5°C, below which atomic mobility is too slow to allow secondary phase formation, and above which the glass becomes more like a liquid (atomic mobility is accelerated). Nucleation and growth of second phases are possible up to the liquidus temperature (the temperature at which the material essentially finishes melting on heating) of the system, beyond which these phases redissolve.

The glass transition temperature, for WVDP test glass that includes uranium and thorium will be determined using differential scanning calorimetry and dilatometry. In detection by thermal expansion, as a glass sample is heated, a sharp increase in thermal expansion is noted at the glass transition temperature range. The glass transition temperature is determined by the intersection of extrapolations of the lower expansion response and the higher expansion response. For differential scanning calorimetry, an endothermic peak is detected at the glass transition temperature range, and the glass transition temperature is defined as the onset of this endothermic response.

As discussed above for the glass transition temperature, atomic mobility can permit nucleation and growth of secondary phases. Characterizing this behavior is facilitated through the use of a TTT diagram. The TTT diagram is a graphical representation of isothermal heat treatments of glass samples for specific lengths of time.

The WVDP will produce a TTT diagram for a nominal glass composition that contains Th and U. Phases resulting from isothermal heat treatments will be identified for type and volume percent abundance by standard analytical techniques (e.g., optical microscopy image analyses, x-ray diffraction, and scanning transmission electron microscopy). Selected specimens will undergo durability testing in deionized water according to the Interactive Powder Leach Test and Pulsed Flow Test methods and assess the influence of heat treatment on durability.

The temperatures used to develop the TTT curve will be between the glass transition temperature and the liquidus temperature. The time length of heat treatments will be between 0.5 hours and as long as canister cooling data dictates for reaching the glass transition temperature.

Following initial cool down of the canister, the canister storage temperature will not exceed the glass transition temperature. Therefore, the TTT diagram will be relevant only during waste form production, and will forecast the type and extent of secondary phases which may exist in the waste form.

CANISTER TESTING

The reference WVDP canister is shown in Fig. 4. It has a 42 cm diameter opening and a minimum wall thickness of 0.34 cm. It is fabricated from Type 304 stainless steel. The waste form canister plays no part in restricting radionuclide release from the repository waste package. The canister only serves as a receptacle for the vitrified waste. However, the canister must be shown not to have a negative impact on the other components of the repository system.

The change in canister dimensions caused by filling will be monitored during cold testing to ensure that a filled canister fits inside the repository container. Preliminary testing at West Valley shows that when the canisters are fabricated according to nominal values given in the specification, the canisters are well within the specification tolerances. This provides a potential opportunity to save cost by increasing the canister volume i.e., canister outer dimensions, until the canister outer dimensions approach the maximum tolerance. Fewer canistered waste forms will be generated for disposal, and therefore, the overall cost will decrease.

To assess the integrity of the canister, a glass filled canister filled during nonradioactive testing will be dropped on an essentially unyielding surface with the canister center of gravity over its bottom center. This is the most likely case for a drop of significant height. This is the case where the canister falls through a shield floor and back into a transportation cask from which it was being unloaded. After the drop, the canister will be leak tested and the strain in the area of the impact will be assessed.

Activities that can be performed at a later date before shipout to the repository will be delayed. These include final canister closure and canister decontamination. These activities need to be performed to reduce the spread of contamination at the repository handling facility. However, at West Valley the canistered waste forms will be stored in decontaminated cells that will have residual contamination. Also, failed process equipment that may spread contamination will be stored next to the canistered waste forms. The canisters may have to be decontaminated just before shipout. By delaying methods development, the WVDP will be able to assess improvements to techniques available at that time.

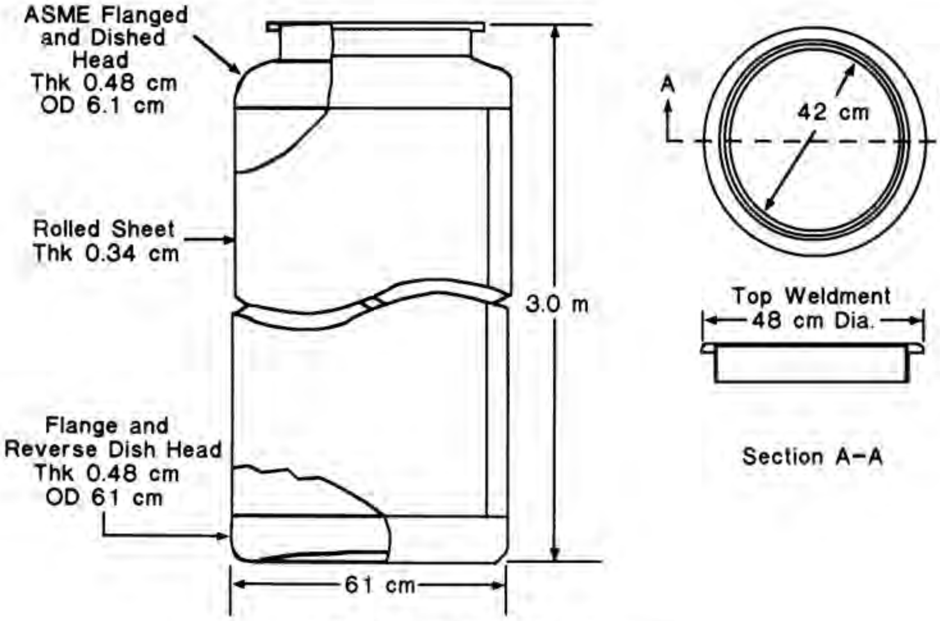

Fig. 4. West Valley Canister.

USE OF EXISTING DATA

Existing data will be used to show compliance with the specifications when appropriate. For example, the known effects of a component in a glass influenced the selection of the elements to be varied in defining the durability envelope. Previous studies are being reviewed to show that the glass has radiation stability, i.e., the durability results will be applicable to glass that has been exposed to a radiation environment. Also, previous studies have been reviewed to show that the glass and canister are chemically compatible (17-19). The conclusion of this review is that waste glass does not cause the canister to corrode from the inside during the repository operation period. Gas generation from the waste form which could cause pressurization inside the canister is another example where existing data has been used to show the need for no additional work (20-21).

ADMINISTRATIVE CONTROLS

Administrative controls will play a major part in showing compliance with the WAPS. Inspections of the empty canisters will be performed to ensure that no restricted materials (free liquids, explosives, pyrophorics, combustibles, and organics) are not in, nor on, the canisters. Administrative controls will also be used to show that the canisters are fabricated as specified.

SUMMARY AND CONCLUSIONS

WVDP waste compliance activities have begun and are proceeding to support the start of hot vitrification. The references give an indication of the amount of effort that has already been expended to support HLW vitrification. The WAP has enabled the WAPS to be sufficiently developed for the WVDP to start a testing program.

Waste compliance activities at the WVDP include developing techniques for providing the glass composition with the full scale process equipment, performing durability tests on glass specimens with Th and other radioactive components, phase stability studies, and full scale canister testing. A broad based testing program has begun.

The testing will be performed on nonproduction canister and glass; the properties of the radioactive canistered waste forms will be inferred from these tests. Compliance on canister closure and decontamination will be demonstrated at a later date, because the time they are performed does not influence the final product. When the technical literature indicates that a property is documented, the test will not be repeated. This testing will show that full-scale West Valley process equipment can generate acceptable glass.

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