A VITRIFICATION PROCESS
FOR THE VOLUME REDUCTION
AND STABILIZATION OF ORGANIC RESINS

J. L. Buelt

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Pacific Northwest Laboratory
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ABSTRACT

Pacific Northwest Laboratory has completed a series of experimental tests sponsored by the U.S. Department of Energy (DOE) to determine the feasibility of incinerating and vitrifying organic ion-exchange resins in a single-step process. The resins used in this study were identical to those used for decontaminating auxiliary building water at the Three Mile Island (TMI) Unit 2 reactor. The primarily organic resins were loaded with nonradioactive isotopes of cesium and strontium for processing in a pilot-scale, joule-heated glass melter modified to support resin combustion.

The glass melter contains molten glass which is electrically heated. The resins are fed to the melter via a novel technique which forces contact of the combustion gases and molten glass. The glass, which absorbs cesium and strontium from the combustion gases, is poured and cooled in a disposal container. Thus the cesium and strontium from the loaded resin are incorporated in a chemically stable glass product.

The feasibility of the vitrification process for organic resins is determined by various parameters. These include:

- process rates
- volume reduction
- radionuclide retention in the glass
- material conformance to operating conditions
- process stability.

The feasibility tests demonstrated an average process rate of 3.0 kg/h. Based on this rate, if 50 organic resin liners were vitrified in a six-month campaign, a melter 2.5 times the size of the pilot scale unit would be adequate. A maximum achievable volume reduction of 91% was demonstrated in these tests. This means that the glass produced from the melter was 9% of the original volume of the resins. The unique feeding technique was successful in absorbing most of the radionuclides into the glass. The average cesium retention in the glass throughout the feasibility tests was 84%; strontium retention was held at 95%. The portion of radionuclides entrained with the gaseous effluents may
be trapped with a wet scrubber or filter system and recycled back to the melter. Construction materials exposed to molten glass at 1000 to 1200°C demonstrated insignificant corrosion. Overall, the process operated very smoothly with few difficulties.

In summary, test results show favorable performance of all the test parameters. Where less than optimum performance was achieved (i.e., process rates and radionuclide retention), alternative approaches to improve those characteristics are described in this report. Once it is demonstrated that the wet scrubbing or filter system decontaminates the effluent stream sufficiently, preparations could be made for a pilot-scale radioactive demonstration or a larger-scale nonradioactive demonstration.
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INTRODUCTION

Cleanup of the auxiliary and fuel handling buildings' water at Three Mile Island (TMI) generates certain radioactive waste byproducts. These byproducts include 50 liners of EPICOR II® ion-exchange resins. The resins were used to decontaminate 1.9 million liters of water from the auxiliary and fuel handling buildings. If necessary, more EPICOR II resins will be used as a polishing system for the 2.6 million liters of containment building and reactor coolant system water (UPDATE 1981). The U.S. Department of Energy (DOE) is conducting feasibility studies for the final conditioning and safe disposal of these waste byproducts.

One promising method for the conditioning of EPICOR II resins, which are primarily organic, is a single-step process that incinerates the resins and vitrifies the ash, radionuclides, and inorganic components. The resultant glass solidifies as a stable, nonleachable product that incorporates the Cs and Sr radionuclides loaded on the EPICOR II resins. By converting the EPICOR II resins into glass, their volume is significantly reduced for cheaper and safer disposal.

Under DOE sponsorship, the Pacific Northwest Laboratory (PNL) has completed a series of tests of the vitrification process using EPICOR II resins loaded with nonradioactive isotopes of cesium and strontium. For these tests, a pilot-scale version of a joule-heated glass melter was modified to support combustion. The modifications included installing a feed system unique to glass melters, so that the resins and simulated radionuclides would come in close contact with molten glass. The tests were conducted subsequent to screening tests performed for the same-type process in an in-can glass melter (Buelt 1981). The joule-heated melter tests were conducted to determine volume reduction, process rates, and retention of Cs and Sr in the glass. The results of the tests were used to produce a conceptual process flow diagram representative of full-scale operation.

*EPICOR II is a registered trademark of EPICOR Inc., Linden, New Jersey.

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TEST EQUIPMENT

The joule-heated glass melter chosen for this study is a key component of the reference process for high-level defense waste vitrification (Savannah River Operations Office 1981). Involvement with the development of this equipment has given PNL a great deal of experience in this area, which simplified the conversion of the joule-heated melter to an incineration/vitrification unit.

In the joule-heated melter, molten glass is contained inside a melting cavity lined with dense refractory and insulating ceramics. The glass is kept molten by passing an alternating electrical current between submerged electrodes at opposite ends of the melting cavity. Brouns et al. (1980) describe the development and operation of the melter in greater detail.

The joule-heated melter was chosen to vitrify the resin as opposed to another glass-melting process known as the in-can melter. The in-can melter produces glass directly inside a disposal canister which is inserted into an electrically-heated furnace (Blair 1979). Although EPICOR II vitrification screening tests employing the in-can melter showed that it operated well (Buelt 1981), the joule-heated melter was anticipated to achieve higher processing rates. Also, as resins are vitrified and more glass is formed, the glass level rises in the in-can melter, which can cause operational complexities in a remotely-operated cell or canyon. The joule-heated melter, on the other hand, maintains a near-constant level throughout the vitrification process.

The incineration/vitrification method differs significantly from presently proposed radwaste incineration/vitrification systems. For example, though the PNL joule-heated melter and the Penberthy Pyro-Converter (Oyen and Tucker 1960) both utilize a glass furnace, PNL's joule-heated melter incorporates a unique feeding system, which forces more contact of the glass and combustible resins. As shown in the process schematic (Figure 1), the PNL process adapts a vertical drop tube, the end of which is submerged 5 to 8 cm below the surface of the glass. The drop tube concept is similar to that used by a molten salt incinerator (Gay et al. 1981). Resins and oxygen are fed though the drop tube
into the melter shown in Figure 2, allowing the molten glass to scrub the radionuclides and residual particles from the off gas.

The resins are fed by a volumetric feeder shown in Figure 3. The drop tube and feeder move vertically with a scissors jack which could easily adjust the degree of submergence of the drop tube in a cell. The jack also withdraws the drop tube from the melt while the operation is idle.
The feed rate of resins into the melter is monitored by the feed level in the feeder's hopper. The walls of the hopper are steep to avoid bridging of the most resins, which tend to agglomerate. The feeder is equipped with an intromitter at its base, which also helps to keep the resins free-flowing at this constriction.

During fabrication the drop tube diameter was arbitrarily limited to 3.8 cm (1-1/2 in.), since it had to slip inside a small-diameter bellows which isolates the melter from the outside atmosphere (see Figure 1). A larger-diameter drop tube would have reduced the chances of plugging and decreased the pressure drop from combustion gases in the drop tube.
Process off gas is metered through a vacuum control valve into a high-efficiency particulate air (HEPA) filter. The gas is then scrubbed before being exhausted to the building stack. Before treatment, the off gas is sampled at two locations to determine the simulated radionuclide concentrations. The gas is first sampled within centimeters of the exit of the melter; the second sample is drawn after the off gas passes through the HEPA filter. Each sampling system consists of two wet scrubbers, a vacuum plump, and a wet-test meter to totalize the sample volume. The first system also contains a sample bomb for periodic mass spectrometry analysis of gaseous constituents. The sample systems depicted in Figure 4, provide data to determine the quantity of strontium and cesium evolved from the process and the HEPA-filter efficiency at removing the simulated radionuclides.
FIGURE 4. Off-Gas Treatment and Sampling System
SCREENING TESTS

Two screening tests were performed with the joule-heated glass melter prior to installing the feed system described in the preceding section. The objective of the screening tests was to verify that the joule-heated glass melter is the appropriate instrument for longer-term (12-h) tests. The tests were performed with minimal equipment modifications. The criteria for performance evaluation were retention of Cs and Sr in the glass, combustion behavior, and general processing observations.

A total of 5.73 kg of EPICOR II resins loaded with nonradioactive Cs and Sr were divided evenly for the two screening tests. The loadings were based on the analytical detection limits of Cs and Sr in the glass (0.02 wt%). Since the mass of glass in the melter (21 kg) is determined by the volume of the melting cavity, at least 4.2 g each of Cs and Sr had to be added to be detectible. The resins were thus loaded to 2.0% Cs and Sr each on a dry-resin basis so that 1 kg of dry resins would approach five times the detection limits of Cs and Sr in the glass. These resin loadings are many orders of magnitude higher than the loadings expected from TMI.

Each run was carried out at an average processing rate of 2.3 kg/h, which is considerably higher than the 0.6 kg/h demonstrated with the in-can melter screening tests (Buelt 1981). Mass spectrometry of five off-gas samples showed no detectable carbon monoxide. Only 0.24% and 0.31% of the mass of resins fed collected on the HEPA filter during each run, indicating most resins were destroyed by combustion with very little particulate entrainment.

Because of the higher velocities and turbulence due to combustion at the higher processing rates, the retention of Cs and Sr in the glass product was not expected to be as high as with the in-can melter screening tests. Still the retention of Sr in the glass was 99.5% [decontamination factor (DF) = 200] in both screening tests, and 98.9% and 98.0% (DF = 91 and 50, respectively) of the Cs was retained in the glass during the two runs. In comparison, Buelt (1981) reported retention figures of 99.3% and 99.8% for Cs and 99.8% and 99.9% for Sr at the lower processing rate of 0.6 kg/h in the in-can melter. The retention values for the joule-heated melter show that all but a small portion
of the radionuclides can be contained in the glass without the benefit of effluent scrubbing equipment. The tests also demonstrate that higher processing rates can be achieved with the joule-heated glass melter without seriously increasing the radionuclide or particulate losses.
TWELVE-HOUR TESTS

Based on the successful screening tests with the joule-heated glass melter, two 12-h tests were then conducted. The EPICOR II resin loadings for the longer-term tests were more closely representative of the anticipated true loadings of resins at TMI than those for the screening tests. Because more resins were vitrified during the longer-term tests, lower Cs and Sr loadings were required to allow detection in the glass; Cs was loaded to 0.070 wt% and Sr to 0.11 wt% on a dry resin basis. To prepare the resin, cesium and strontium nitrates were added with distilled water to a 140-liter batch of resins and mixed thoroughly. The supernate was filtered from the resins, which were allowed to drain overnight. The residual moisture content of the resins fed to the melter was 40%. The resins were not dried to better simulate the resins in the TMI liners.

The 12-h tests were conducted to:

- determine the maximum processing rates with the pilot-scale equipment
- demonstrate the attainable reduction in volume from converting resins to glass
- verify postulated Cs, Sr and other effluent characteristics
- demonstrate a method of recycling Cs, Sr and particulates into the vitrification process
- determine drop tube material conformance to processing conditions
- develop a conceptual process flow diagram based on test results.

PROCESSING RATES

One of the important objectives of these tests was to determine the processing rates achievable in the joule-heated melter. Two tests were carried out over 10.2- and 12.0-h time spans, with average feed rates of 2.9 and 3.1 kg/h, respectively. If the time allotted for momentary process shutdown (oxygen bottle changeout, HEPA filter replacement, and off-gas line and glass overflow inspection) is not included, the average feed rate for both
runs was 3.7 kg/h. Refinement of operating techniques could achieve the 3.7 kg/h feed rate on a total elapsed time basis. The limiting factor for process rates was surprisingly not the melter size nor the vacuum on the system, but a resin-flow restriction in the drop tube whose diameter was limited by the flexible bellows. Once the 3.7-kg/h feed rate was exceeded, the agglomerated resins bridged inside and plugged the 3.8-cm (1-1/2-in.) diameter drop tube. Although the demonstrated feed rates are respectable for the pilot scale equipment used in this study, even higher rates could have been achieved had a larger-diameter drop tube been used.

OPERATION

Other than occasional plugging of the drop tube when the maximum feed rate was exceeded, the process operated very smoothly. Although most of the combustion took place below the surface of the glass, occasionally a bright flash of flame could be seen through a view port at the glass surface. Fluctuations in the melter vacuum, which was generally operated at 25 cm H₂O, were minimal. The oxygen concentration in the off-gas stream was controlled arbitrarily between 15% and 25% which kept particulate entrainment low. No carbon monoxide was detected in any of the eight off-gas samples analyzed by gas chromatography or mass spectrometry. However, if the oxygen supply was inadequate (near 0% in the off gas), carbonaceous, sooty particulates immediately formed and were entrained in the off gas. Particulate losses, based on the wet resin feed, were 0.45% for the two experiments. The low particulate losses kept the 2.5-cm (1-in.) tubing off-gas line between the melter and particulate filter generally free of obstructions. Only on two occasions was an off-gas restriction problem detected. These incidents both occurred at the vacuum control valve between the melter and filter. The ball valve had interfered with the fully developed, laminar flow (Re₀ ≈ 570 for 2.5-cm tubing) at that point, creating stagnant areas for particulates to collect. This problem could be avoided in the future by placing the vacuum control valve downstream of the filter.
VOLUME REDUCTION

Wet EPICOR II resins from the long-term tests contained a fraction of zeolite resins, which restricted the volume reduction that might have been obtained from purely organic resins. The theoretical volume reduction limit, restricted by the quantity of zeolite, is 91%, meaning the volume of the glass product is 9% of the original volume of the resins. This theoretical limit was demonstrated during the two 12-h tests. The final glass product consisted primarily of vitrified zeolite, glass formers and absorbed radionuclides. Glass formers have already been developed for LINDE IONSIV IE-96® zeolite resins in another program conducted at PNL. These glass formers, whose composition is given in Table 1, were added to the zeolite resins in the second of the two tests, yielding a glass former:zeolite ratio of 3:7.

CESIUM AND STRONTIUM RETENTION

In order to totalize the Cs and Sr losses in the effluent stream, mass balances are necessary. The off-gas sampling system determines the concentration of radionuclides in the off gas. The mass balances determine total off-gas flow at various time periods throughout the run. Together they totalize radionuclide losses and thus the radionuclide retention in the glass can be calculated. The oxygen input rate was continually metered with a rotometer. The volumetric feed rate of the wet resins, whose compacted density was 0.78 g cm\(^3\), was monitored by recording the level in the feeder hopper.

<table>
<thead>
<tr>
<th>Component</th>
<th>Weight %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Borax (Na(_2)B(_4)O(_7)-10H(_2)O)</td>
<td>32</td>
</tr>
<tr>
<td>Soda ash (Na(_2)CO(_3))</td>
<td>23</td>
</tr>
<tr>
<td>Lithium carbonate (Li(_2)CO(_3))</td>
<td>29</td>
</tr>
<tr>
<td>Titania (TiO(_3))</td>
<td>16</td>
</tr>
</tbody>
</table>

*Linde Ionsiv IE-96 is a registered trademark of Union Carbide Corp., New York, New York.*

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composition of the main components of the off gas was determined by gas chroma-
matography and mass spectrometry of gaseous samples. From this data, a mass
balance was performed to determine the rate of air in-leakage and total off-gas
flow. The mass balance was necessary because condensate interfered with flow
measurements by a pitot tube installed in the off-gas line. The results of
these conditions are plotted in Figure 5 for the second test.

Retention of Cs and Sr in the glass was determined by analysis of three
sampling techniques:
- off-gas sample
- HEPA filter
- glass product.
The off-gas sampling system and mass balances provide the most accurate mea-
surement of radionuclide retention. The analyses of the glass product and
filter particulates serve as a check for the retention values measured by the
off-gas sampling system. The retention values measured by the various tech-
niques are tabulated in Table 2 for different periods of each run. The overall
retention for Cs was 84% while that of Sr was 95%. (DF = 6.3 and DF = 20,
respectively.) These values are somewhat less than observed during the
screening tests for two reasons:
1. Higher processing rates were achieved during the longer-term tests.
2. Resin loadings of Cs and Sr were reduced for the longer-term tests.

Processing Rate Effect
For the same drop-tube diameter, higher processing rates result in reduced
contact time between the combustion gas and molten glass phases. Therefore the
higher processing rates could account for some decrease in retention. However,
the average rate during the long-term tests was only 30% higher than the rates
achieved during the screening tests. This rate increase could not be the total
cause of the reductions of Cs from 50 DF to 6.3 DF. Even so, retention values
might be increased with a larger drop-tube diameter thus increasing the contact
time.

Resins Loading Effect
The simulated radionuclides were absorbed into the glass product during
these tests simply by mass transfer of Sr and Cs chemical compounds from the
FIGURE 5. Off-Gas Component Balance at Various Times for Second 12-h Test
TABLE 2. Cesium and Strontium Retention as Measured by Three Independent Detection Mechanisms

<table>
<thead>
<tr>
<th>Time Period</th>
<th>Off-Gas Sample Analysis, %</th>
<th>Filter Particulate Analysis, %</th>
<th>Glass Product Analysis, %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cs</td>
<td>Sr</td>
<td>Cs</td>
</tr>
<tr>
<td>Run 1: 0.0 to 4.6 h</td>
<td>70</td>
<td>86</td>
<td></td>
</tr>
<tr>
<td>Run 1: 5.0 to 10.2 h</td>
<td>82</td>
<td>95</td>
<td></td>
</tr>
<tr>
<td>Run 1: Overall</td>
<td>78</td>
<td>92</td>
<td></td>
</tr>
<tr>
<td>Run 2: 0.0 to 4.5 h</td>
<td>95</td>
<td>97.8</td>
<td></td>
</tr>
<tr>
<td>Run 2: 4.9 to 8.7 h</td>
<td>95</td>
<td>98.7</td>
<td></td>
</tr>
<tr>
<td>Run 2: 8.9 to 12.0 h</td>
<td>77</td>
<td>95.7</td>
<td></td>
</tr>
<tr>
<td>Run 2: Overall</td>
<td>89</td>
<td>97.8</td>
<td></td>
</tr>
<tr>
<td>Runs 1 and 2 Combined</td>
<td>84</td>
<td>95</td>
<td>94</td>
</tr>
</tbody>
</table>

Combustion gases into the molten glass phase. Assuming that the Cs and Sr are present in the vapor phase, the mass transfer rate is governed by the equation

\[ N_{A2} = K_G (p_{AG} - p_{A*}) \]  

(Welty, Wicks and Wilson 1969)

where

- \( N_{A2} \) = mass transfer flux
- \( K_G \) = overall mass transfer coefficient
- \( p_{AG} \) = bulk composition of the radionuclide in the gas phase
- \( p_{A*} \) = partial pressure of the radionuclide in equilibrium with the bulk composition in the liquid phase.

Reducing the resin loading, as done in the 12-h tests, reduces the radionuclide's partial pressure gradient (the driving force for mass transfer). This explains, at least in part, the reduction of Cs and Sr retention in the glass.

The resin loading used in the longer-term tests, although more representative, may still be higher than the true Cs and Sr loadings. To find out if even lower loadings might affect radionuclide retention further, \( p_{A*} \), the equilibrium partial pressure, must be considered along with the reduction of resin loading (\( p_{AG} \)).
The expected Cs concentration in the glass from vitrifying EPICOR-II resins from TMI can be estimated. If the mass fraction of Cs isotopes in the resins and the volume reduction (demonstrated at 91%) are known, the maximum Cs concentration in the glass can be calculated. Radiation surveys of one liner from TMI indicate the mass fraction of all Cs isotopes is $3.2 \times 10^{-5}$ based on a wet resin content of 680 kg per liner. (The ratio of Cs-137 detected by the radiation survey to the combined Cs isotopes was established from existing ORIGEN runs. Although an ORIGEN run was not available for the conditions at TMI-2, a range of established conditions from 2% to 3.3% enrichment and from 33,000 MWD/t to 10,000 MWD/t showed that variance in the isotope ratio was less than 1%.)

The mass-fraction Cs loading used in these tests was $4.2 \times 10^{-4}$ (wet resin basis). Assuming 100% retention, the final glass product concentration with the 91% volume reduction would be 0.012% for the actual TMI resins, somewhat less than the 0.071% achieved during the two tests conducted in this study. Even though the resin loading in this study was higher than that expected for actual resins, the radionuclide concentration in the glass was correspondingly higher. Assuming $p_{AG}$ and $p_{A*}$ are proportional to resin loading and radionuclide glass concentration, the partial pressure driving force for mass transfer very nearly approximates the anticipated partial pressure gradient for the TMI resins. Thus, the value for cesium retention in the glass demonstrated in this study is expected to be representative of the cesium retention for actual TMI resins. The driving force may actually be increased, thus obtaining a higher retention value at the expense of adding more glass formers and reducing the radionuclide concentration in the glass.

The mass transfer equation among two dissimilar phases (combustion off gas and molten glass) may be studied further to arrive at some additional conclusions. Cesium oxide, as it exists in the glass, is a semivolatile component. When heated above 640°C, Cs$_2$O volatilizes from glass melts according to the equation:

$$\text{Cs}_2\text{O} \rightarrow \text{Cs}_\text{(g)} + \text{CsO}_\text{(g)}$$

(Albrethsen and Schwendiman 1967)
<table>
<thead>
<tr>
<th>Filter No. 1, Run 1: 0 to 5.3 h</th>
<th>Cs</th>
<th>Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filter No. 2, Run 1: 6.5 to 10 h</td>
<td>1.6</td>
<td>1.1</td>
</tr>
<tr>
<td>Filter No. 2, Run 2: 0 to 4.7 h</td>
<td>6.4</td>
<td>2.1</td>
</tr>
</tbody>
</table>

To obtain a higher DF, the off gases would probably have to be scrubbed in a wet scrubber. The off-gas sampling and scrub systems consistently removed all detectable Cs and Sr in the first scrubbing step. Nevertheless, the simulated radionuclides collected on the filter during the 12-h tests were recycled to the molten glass where all detectable radionuclides were incorporated into the glass. Thus, if a more efficient radionuclide filter can be designed, the filter and associated radionuclides could be recycled to the glass melter.

**MATERIAL CONFORMANCE**

The drop tube was constructed of a nickel/chromium alloy known as Inconel 601®. Following the feasibility tests the drop tube was withdrawn from the melt. The drop tube displayed no significant corrosion at or below the metal line (glass melt level), where oxidation might have been suspected. Thus the temperature and oxidizing conditions did not cause material conformance problems.

**CONCEPTUAL PROCESS FLOW DIAGRAM**

One of the objectives of the 12-h tests was to provide data for a conceptual process flow diagram. The conceptual process presented in this report is scaled up to vitrify the resins from all 50 liners in a six-month campaign. The mass balances and flow diagram are shown in Figure 6.

*Inconel 601 is a registered trademark of Huntington Alloys, Inc., Huntington, West Virginia.*
FIGURE 6. Conceptual Process Flow Diagram for Resin Vitrification
Assuming each liner contains 680 kg of wet resins, the total mass to be processed is 34,000 kg. The average required process rate is thus 8.3 kg/h, assuming 24-h operation and 5% down time. Glass formers premixed with the resins would comprise an additional 0.50 kg/h of the feed.

At this higher feed rate (8.3 kg/h) a larger-diameter drop tube is necessary to keep the agglomerated, wet resins from plugging up the tube and to keep the upward vertical velocities of the combustion gases in the glass at or below those experienced in the pilot-scale tests. Assuming that vertical velocity is a function of bubble size, the size of bubbles in the glass must be kept constant. For design purposes, the bubble size is assumed to be proportional to the circumference or diameter of the drop tube. Thus, the drop-tube diameter was scaled proportionally to the feed rate. At the 8.3 kg/h rate, a 15-cm-dia drop tube is required. The melting cavity should allow the same amount of clearance between the drop tube and melter cavity walls. Thus the melting cavity in the process flow diagram should be 30 cm x 46 cm x 20 cm deep.

The oxygen requirements, 260 std L/min, were calculated proportionally from the oxygen to average feed ratio (94 std L/min:3.0 kg/h) demonstrated during the tests. The oxygen combusts the resins in the drop tube yielding primarily CO₂ and H₂O. The air in-leakage, which comprises 61% (by volume) of the total off gas, is determined from acceptable design criteria of the joule-heated melter. The total off-gas rate is 940 std L/min. The off gases also entrain 0.037 kg/h of particulates based on the 0.45% losses determined in these tests.

Glass would be produced from this process at a rate of 2.1 kg/h or 0.85 L/h. If disposal criteria allows, the glass could be poured into a container that would fit in a 200-L (55-gal) burial drum for disposal. All 50 liners of resins could be disposed in 21 burial drums.
Summary and Conclusions

The nonradioactive feasibility tests for vitrifying EPICOR II resins have provided an important data base for development of this process. Process rates and their limiting factors have been determined for the pilot-scale equipment. A method for predicting attainable process rates on a larger scale has been proposed in the process flow sheet. The maximum theoretical volume reduction from converting the resins to glass is 91%. This has been demonstrated during the tests. The quantities and mechanisms of radionuclide losses as well as a method to recycle the radionuclides have also been identified.

The average process rate during the two 12-h tests was 3.0 kg/h (wet resin basis). The process rate was limited by an agglomeration of wet resins in the 3.8-cm (1.5-in.)-dia drop tube. Although demonstrated process rates were acceptable, higher rates would likely have been achieved if the resins were dried before feeding since dry resins become free-flowing granular particles. Wet resins were used in this study to more accurately simulate worst-case conditions in the TMI liners. Process rates would also have increased in the pilot-scale melter if a larger-diameter drop tube had been used.

Nevertheless, the process flow diagram for the larger-scale unit was based on the 3.0-kg/h demonstrated process rate. The flow diagram suggests that the drop-tube diameter be scaled proportionally with the feed rate to keep the upward combustion gas velocities in the glass constant. The melter design must accommodate the drop tube because the combustion gases must contact the glass freely without interference from the melter cavity walls. The electrodes must also be separated from the drop tube enough to minimize the electrical shorting effect created by the drop tube.

Although less Cs and Sr were retained in the longer-term tests than in the screening tests, it has been argued that these values are close to the minimum retention expected with EPICOR II resins. The test results indicate that as the ratio of resin loading to final radionuclide concentration in the glass decreases, less retention can be expected. Since the resins contain zeolite, which becomes part of the glass product, the radionuclide concentration in the glass is low. The ratio of Cs mass fraction loading in the resins to the Cs
mass fraction in the glass gives a good indication of relative mass-transfer driving forces. A high ratio indicates a high driving force. The ratio during the screening tests, which demonstrated 98% to 98.9% retention, was 4.7 while that during the 12-h tests, which averaged 84% retention, was 0.56. According to the process flow diagram, every unit weight of resins is converted to 0.25 units of glass; thus the expected Cs ratio for resins stored at TMI is 0.25. Obviously, the longer-term tests more closely approximated the mass transfer driving force expected for the true resins. Expected retention values for Cs would remain near 80% to 90%. These relationships may not explain Sr behavior, since SrO is a nonvolatile component of molten glass. Most Sr losses might be attributed to particulate entrainment.

The supply of pure oxygen rather than air into the drop tube had some merits. The data presented in this report suggests that Cs is absorbed into the glass as an oxide. Thus the use of pure oxygen for combustion may be an important factor in attaining the demonstrated Cs retention figures. The pure oxygen supply also helped maintain thorough combustion, keeping particulate entrainment low with negligible concentrations of CO and pyrolysis products.

A method for recycling volatilized or entrained radionuclides was demonstrated in these tests. Although the HEPA-filter decontamination factors were not as high as desired, the basic principle of recycling the filter material and absorbed radionuclides to the melter is feasible.

Overall, the process feasibility tests did not present any insurmountable technological difficulties. Vitrifying organic resins appears to be a technically feasible approach. The process is based upon equipment which has been under development for nuclear waste vitrification for many years and is now part of the reference process for high-level defense waste stabilization. The construction materials of the joule-heated melter conform well with resin vitrification-processing conditions. The tests performed in this study demonstrate the advantages of using the joule-heated melter for vitrification of radioactive wastes based on process rates, volume reduction, radionuclide retention, and material conformance. A successful radionuclide decontamination scrubber or filter needs to be demonstrated for the process. Once that is
completed, the resin vitrification process will be to the development stage so that preparation can then be made for a pilot-scale, radioactive demonstration or a larger scale, nonradioactive demonstration.
REFERENCES


