**Support for the
In Situ Vitrification
Treatability Study at the
Idaho National Engineering
Laboratory: FY 1988 Summary**

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February 1989

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SUMMARY

The objective of this project is to determine if in situ vitrification (ISV) is a viable, long-term confinement technology for previously buried solid transuranic and mixed waste at the Radioactive Waste Management Complex (RWMC). The RWMC is located at the Idaho National Engineering Laboratory (INEL). In situ vitrification is a thermal treatment process that converts contaminated soils and wastes into a durable glass and crystalline form. During processing, heavy metals or other inorganic constituents are retained and immobilized in the glass structure, and organic constituents are typically destroyed or removed for capture by an off-gas treatment system.

The primary FY 1988 activities included engineering-scale feasibility tests on INEL soils containing a high metals loading. Results of engineering-scale testing indicate that wastes with a high metals content can be successfully processed by ISV. The process successfully vitrified soils containing localized metal concentrations as high as 42 wt% without requiring special methods to prevent electrical shorting within the melt zone. Vitrification of this localized concentration resulted in a 15.9 wt% metals content in the entire ISV test block. This ISV metals limit is related to the quantity of metal that accumulates at the bottom of the molten glass zone. Intermediate pilot-scale testing is recommended to determine metals content scale-up parameters in order to project metals content limits for large-scale ISV operation at INEL.

Particulate and gaseous releases during processing were characterized. The releases can be retained in an off-gas treatment system that is appropriately designed to meet necessary criteria established for these releases. The small quantity of particulate can be effectively retained by an appropriately designed conventional wet scrubbing and filtering system.

Metals become incorporated into the vitrified product primarily as pools near the bottom of the block. Metals are also incorporated as partially melted pieces, small dispersed spheres, and solubilized oxides within the glass structure. Materials Characterization Center MCC-1 28-day leach tests showed the dissolution rates of the vitrified INEL soil to be $0.036 \text{ g/m}^2 \cdot \text{d}$

(based on Si). This is approximately a factor of four less than the dissolution rate of vitrified Hanford soil, and is about an order of magnitude less than that of nuclear waste glass.

Based on the encouraging results of the FY 1988 activities, adequate technical bases exist to support further development and scale-up of the ISV process for improved confinement of buried wastes and soils at the RWMC.

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INTRODUCTION

As management of hazardous radioactive and chemical materials gains increased attention in the United States and throughout the world, new, more effective technologies are being sought to immobilize and/or destroy the materials either in situ for previously disposed wastes, or at the waste generation site. Pacific Northwest Laboratory^(a) (PNL) is developing a remedial action process for contaminated soils that is significant in its application to this need. The process, called in situ vitrification (ISV), was initially developed for the U.S. Department of Energy (DOE) by PNL to demonstrate a potential technology for the immobilization of soil contaminated with transuranic (TRU) radionuclides. Subsequent tests have shown that many hazardous chemicals are also destroyed or immobilized as a result of the treatment.

In situ vitrification is a thermal treatment process that converts contaminated soils into a chemically inert and stable glass and crystalline product. The ISV process has potential for application to many contaminated soil sites at the Idaho National Engineering Laboratory (INEL). The high temperature of the process would destroy or remove organic contaminants, and the glass and crystalline product would encapsulate any inorganic contaminants, such as plutonium and americium. All these functions would be performed in place and in one processing step.

The application of ISV at TRU and mixed waste burial grounds within INEL requires that the existing ISV data base be expanded to soils containing a high metals content. This report presents the results of FY 1988 work conducted in support of INEL, which included engineering-scale ISV testing. The primary objective was to determine the applicability of the ISV process to soils with a high metallic content. Test results determined the extent of

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free and soluble metals in the vitrified product. Particulate releases to the off-gas treatment system and leaching characteristics of the vitrified soil were determined.

The ISV process offers a technical alternative for managing the buried TRU and mixed hazardous wastes at INEL. By understanding the process design and its limitations, the applicability of ISV to site-specific disposal problems can be judged.

CONCLUSIONS

Feasibility testing of the ISV process on INEL soil with a high metals content was successfully performed during two engineering-scale ISV tests. Analyses of the data from these FY 1988 activities provide the following conclusions:

- Processing parameters from two engineering-scale ISV tests demonstrated the feasibility of using ISV to process INEL soils into a more compact and environmentally stable (immobilized) form.
- The ISV process successfully vitrified INEL soils containing localized metal concentrations as high as 42 wt% without incorporating special techniques to prevent shorting. The metals content in the vitrified soils was 12.5 wt% for Test 1 and 15.9 wt% for Test 2. The total percent metals achieved during engineering-scale ISV testing is difficult to relate directly to large-scale operations because of scale-up uncertainties. Intermediate pilot-scale testing is a recommended low cost and low risk method to evaluate metals content scale-up parameters.
- The small particulate and gaseous releases can be effectively retained by a conventional ISV off-gas treatment system consisting of a wet scrubber and filtering media.
- The dissolution rate of the vitrified INEL soil is $0.036 \text{ g/m}^2 \cdot \text{d}$ (based on Si). This is about 4 times less than the dissolution rate for vitrified Hanford soil, and about an order of magnitude less than that for a nuclear high-level waste glass.
- Metals are incorporated into the vitrified product primarily as a molten pool near the bottom of the block. Metals were also present as partially melted pieces, small spheres dispersed in the glass, and solubilized Fe_2O_3 , Cr_2O_3 , and NiO within the glass structure.
- The metal pool at the bottom of the molten glass appears to promote downward growth by skewing the power to the lower portion of the melt.

Initial testing indicates the potential for ISV treatment of high metal content waste presently held in burial trenches at INEL. Based on the encouraging results of the feasibility tests, adequate technical bases are available to support further feasibility testing and scale-up of the ISV process for immobilization of buried INEL wastes and soils.

PROCESS DESCRIPTION AND STATUS OF DEVELOPMENT

In situ vitrification has been developed as a remedial action process for soils contaminated with radionuclides and/or hazardous chemical wastes. Figure 1 illustrates the operation of the ISV process. A square array of four molybdenum (Mo)/graphite electrodes is inserted into the ground to the desired treatment depth. Because soil is not electrically conductive when its moisture has been driven off, a conductive mixture of flaked graphite and glass frit is placed among the electrodes to serve as a starter path. An electrical potential is applied to the electrodes to establish an electrical current in the starter path. The flow of current heats the starter path and surrounding soil to well above the initial soil-melting temperatures of 1100°C to 1400°C. The graphite starter path is eventually consumed by oxidation and the current is transferred to the molten soil, which is processed at temperatures between 1450°C and 2000°C. As the molten zone grows, it incorporates or encapsulates radionuclides and nonvolatile hazardous elements, such as heavy metals, into the glass structure. The high temperature of the process destroys organic components by pyrolysis. The pyrolyzed byproducts

FIGURE 1. Process of In Situ Vitrification

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migrate to the surface of the vitrified zone, where they combust in the presence of air. A hood placed over the area being vitrified directs the gaseous effluents to an off-gas treatment system.

The process can accommodate a staging operation alternative. For this processing alternative, retrieved wastes are placed on the surface of an area to be vitrified to increase the effective vitrification depth. Since ISV is a batch operation, this alternative will increase the amount of material vitrified in each batch setting, thereby improving the efficiency of the operation.

PNL began developing ISV technology in 1980. Since that time, numerous experimental tests under a variety of conditions and with a variety of waste types have been conducted (Buel et al. 1987, Timmerman and Oma 1984, Timmerman et al. 1983, Buel and Carter 1986). Table 1 describes the different scales of testing units that PNL used in developing and adapting ISV technology. The successful results of 59 bench-, engineering-, pilot-, and large-scale tests have proven the general feasibility and widespread applications of the process. Also, economic studies have indicated that tremendous economies-of-scale are attainable with the ISV process (Oma et al. 1983).

The extensive technical data establish the performance of the system and waste form, economics and safety, and waste site applications for radioactive and hazardous chemical wastes. Aspects of the existing ISV data base as applied to INEL wastes are summarized as follows:

TABLE 1. Testing Units for Developing In Situ Vitrification Technology

<u>Equipment Size</u>	<u>Electrode Separation, m</u>	<u>Vitrified Mass per Setting</u>	<u>Tests Completed As of 9/30/88</u>
Bench scale	0.11	1 to 10 kg	12
Engineering scale	0.23 to 0.36	0.05 to 1.0 t	26
Pilot scale	0.9 to 1.5	10 to 50 t	16
Large scale	3.5 to 5.5	400 to 800 t	<u>5</u>
			59

- Although a 5-m depth has been demonstrated with the large-scale ISV system, the estimated vitrification depth limit is projected to exceed 10 m. This greatly exceeds the estimated required ISV depth of 6.1 m for the buried INEL wastes.
- Radionuclides and heavy metals are sufficiently retained within the melt, and nitrates and organics are destroyed during processing.
- An off-gas treatment system is included in the process to remove contaminants such as volatilized heavy metals and acid gases from the gaseous effluents. The off-gas system meets the Washington State Administrative Code and federal requirements for stack emissions.
- A depth transmitter system has been developed for reliable monitoring of the vitrification zone depth during processing.
- A reference electrode design has been established and successfully tested for future ISV applications. This design employs an Mo/graphite combination, which promotes subsidence of the surface of the melt below the original soil grade. Oxidation of Mo during processing is prevented by the graphite collar, plus a fused coating of MoSi_2 on the Mo electrode core.
- Metal inclusions do not significantly affect the ISV process unless a full electrical short circuit is approached. The power system is adequately protected against voltage and current surges.
- Cement inclusions are completely dissolved within the glass waste form.
- In situ vitrification is capable of processing various soil types throughout the United States. Soils from nine U.S. locations were found to be similar in the properties that could affect the ISV process. Similar ISV processing capability is anticipated for the INEL site or other soil types.
- The ISV waste form has been shown to surpass established Environmental Protection Agency (EPA) toxicity tests.

TEST DESCRIPTION

The primary objective of the engineering-scale ISV test was to determine the maximum metal content in INEL soils that can be processed without short-circuiting the ISV power supply. In addition, supplemental data on product durability and off-gas characteristics were obtained. In preparation for testing, the INEL soils were characterized, the engineering-scale ISV test unit was prepared, and an off-gas sampling train was set up.

SOIL CHARACTERIZATION

Samples of the uncontaminated soil from the INEL burial ground site have been analyzed to determine inorganic composition. The cation oxide data as analyzed by inductively coupled plasma (ICP) are presented in Table 2. The ICP analyses confirm that an acceptable ratio of glass-forming constituents exists to make an acceptable product. The high SiO_2 and Al_2O_3 concentrations contribute to product durability, while the K_2O and Na_2O concentrations maintain a melting point that is low enough to permit effective processing by ISV.

Table 3 presents soil property data for the INEL soil. The as-received soil sintered (fused into a solid mass without melting) at 1200°C . The sample started to melt at approximately 1300°C , and foaming was observed in the melt between 1300°C and 1450°C . At 1450°C , the foaming subsided into a thick melt (viscosity >5000 poise). The temperature was increased to 1600°C , where the melt viscosity was estimated at 4000-5000 poise and the melt would not pour from the crucible.

Comparison of the oxide composition of the INEL soil with other soil compositions indicated that adding approximately 5 wt% Na_2O would reduce the melt temperature and viscosity of the composition and improve the processability. A total of 5 wt% Na_2O as Na_2CO_3 was added to the INEL soil (see Table 3). This addition lowered the reaction temperature approximately 100°C and substantially decreased the viscosity.

TABLE 2. INEL Soil Sample Analyses

Oxide	Composition, (a) wt%
SiO ₂	62.60
Al ₂ O ₃	11.85
Fe ₂ O ₃	4.25
CaO	3.68
K ₂ O	2.99
MgO	1.72
Na ₂ O	1.37
TiO ₂	0.68
MnO ₂	0.10
BaO	0.09
ZrO ₂	0.05
B ₂ O ₃	0.05
NiO	0.04
SrO	0.02
Cr ₂ O ₃	0.02
Total Oxide	89.5%
Moisture	7.5%

(a) Average of two analyses of soil samples.

The melted glass sample (soil + 5 wt% Na₂O) was allowed to cool and was again heated to 1600°C. The foaming observed during the first melt was again observed, indicating that redox reactions within the glass liberated oxygen, which caused the foaming. Electrodes with graphite collars are used in the ISV process, which results in very reduced melts. To simulate these conditions, approximately 10 wt% graphite was added to the soil with 5 wt% Na₂O. The sample sintered and melted at the same temperature as the soil + 5 wt% Na₂O sample, but only a very small amount of foaming was observed at 1400°C. At 1300°C, the sample was completely molten and the viscosity was estimated to be >5000 poise. At 1600°C, the melt was very smooth with an estimated viscosity of 500 poise.

TABLE 3. INEL Soil Properties

<u>Properties</u>	<u>INEL Soil</u>	<u>INEL Soil with 5% Na₂O^(a)</u>
Sintering temperature	-1200°C	-1100°C
Melt temperature	-1450°C	-1300°C
Estimated viscosity at 1600°C	~4000-5000 poise	~500 poise
Soil tap density ^(b)	1.27 g/cm ³	--
Soil theoretical density	2.07 g/cm ³	--
Vitrified soil density	2.38 g/cm ³	--

(a) Na₂O added as Na₂CO₃.

(b) ASTM Method C520, volume 4.06.

These data indicate that Na₂CO₃ (and possibly other additives) could reduce viscosity and improve the melting properties of INEL soils, if necessary. However, additives were not used in the tests described here. The ultimate need for additives for large-scale ISV of INEL wastes can be determined once the ISV scale-up effects on INEL soils are known. The scale-up effects can be determined by intermediate pilot-scale testing.

TEST CONFIGURATION

The engineering-scale ISV system consisted of a test container that measured 1.83 m (6 ft.) in diameter by 2.44 m (8 ft.) high. Figure 2 shows the test configuration. Four graphite electrodes having a 38.1-mm (1.5-in.)-diameter and a 0.305-m (12-in.) separation were used. Power was supplied to the electrodes using a 30 kVA Scott-tee-connected transformer, which converts a three-phase primary alternating current (AC) to a two-phase secondary AC. The transformer was equipped with 12 voltage taps that permitted near full power operation between 400 V and 59 V electrode potential. The power level was controlled by saturable reactors located on the secondary side of the transformer.

The test objective was accomplished by progressively increasing the metallic content of the soil as melt depth increased. A computer model was

FIGURE 2. Configuration of the Engineering-Scale ISV Test

used to predict the test parameters, including melt depth, shape, mass vitrified, etc. The metals were centrally placed in the regions indicated in Figure 2, based on the model projections for melt shape. The metals loading was slowly increased, starting with 5.8 kg near the surface and increasing to 210.4 kg in the bottom region. Total metals contents of 5, 10, 15, 20, and 25 wt% per foot of the depth (based on the total vitrified mass) were selected in the experimental design of the ISV test. The localized concentrations of the metals added to obtain the desired melt concentrations were higher, ranging from 11 to 57 wt% metals. Table 4 lists the quantity of metal placed at the specified depths prior to vitrification. The metals consisted of approximately 70 wt% stainless steels and 30 wt% carbon steels. Figure 3 shows the metals and their typical placement within the soil. In addition to metals, combustible scrap materials (wood and paper) were added to the soil to simulate the INEL burial ground composition of 0.5 wt% combustibles.

To monitor the progress of the melt, type K thermocouples were installed starting 0.15 m (6 in.) below the soil surface and at designated intervals to the bottom of the container. A high-temperature type C thermocouple located at the 0.76-m (30-in.) depth was used to monitor actual melt temperatures during the test.

TABLE 4. Metals Added with Soil Depth

Soil Depth, m (ft)	Metals in Total Vitrified Mass, wt%	Localized wt% Metals in Soil	Amount of Metals, kg	
			Added	Cumulative
0 - 0.30 (0 - 1)	5	11	5.8	5.8
0.30 - 0.61 (1 - 2)	10	14	24.7	30.5
0.61 - 0.91 (2 - 3)	15	26	65.3	95.8
0.91 - 1.22 (3 - 4)	20	42	125.3	221.1
1.22 - 1.52 (4 - 5)	25	57	210.4	431.5

FIGURE 3. Typical Placement of Metals for Engineering-Scale ISV Testing (88083146-99CN)

OFF-GAS SAMPLING

The engineering-scale ISV system was enclosed to permit collection and characterization of the off gases and particulates that emanated during melting operations. The test container was maintained at a slight negative pressure, and the lid was equipped with a back pressure HEPA-filtered relief to prevent particulate releases during the test. Off gases were routed from the test container plenum area to the building ventilation and filtering system. During testing at INEL, an off-gas flow of about 850 lpm (30 cfm) was maintained.

A sampling train was used to obtain particulate loading data and information on selective elemental volatilization during the ISV of the INEL soil. The sampling train generally consisted of an EPA Reference Method 5 sampling scheme (40 CFR 60, Appendix A). Figure 4 illustrates the sample train setup. Particulates were collected by drawing a representative sample from the 5-cm (2-in.) off-gas process line through a heated probe assembly leading into a glass fiber filter. The filter was housed in an oven compartment maintained at approximately 260°F. After exiting the filter/oven assembly, the gases were drawn through a series of three Greenburg-Smith impingers containing deionized water for condensate collection. A fourth empty impinger was placed in line to collect carry-over liquid/condensate, and a final impinger containing 300 g of 6-16 mesh silica gel was used to trap water vapor. A control unit was used to monitor and control sampling rates, record sample volume, and track off-gas sampling temperatures during the vitrification process.

FIGURE 4. Sample Train Configuration

TEST RESULTS AND PERFORMANCE ANALYSIS

Initial plans called for one engineering-scale ISV test; however, because of events during the test, a second test was performed. Test 1 achieved a melt depth of 0.69 m (27 in.) before it was discontinued because of lost electrical conduction. The process effectively vitrified through areas with localized metal contents of 11 wt% and 14 wt% (see Table 4). The vitrified block was removed and a second test was continued into the region of the soil with a higher metals content. Before starting Test 2, soil and metals were removed to the 0.61-m (24-in.) depth (see Figure 2), new electrodes were installed, and 0.30 m (6 in.) of fresh INEL soil were placed above the metals-bearing soil. The second test effectively vitrified through an area with a localized concentration of 26 wt% metals and 0.30 m (6 in.) into an area with 42 wt% metals, as presented in Table 4. Performance of the ISV process during these two tests is discussed in the following sections.

POWER SYSTEM PERFORMANCE

The power system performed well during both tests and the saturable reactor controllers operated smoothly, despite the high metals content encountered. The tests were configured so there was not a direct metals short between electrodes, which would prevent normal ISV startup. Slightly oversizing the electrode placement holes would ensure against initial direct shorting during actual large-scale ISV operations at a TRU waste trench or pit [i.e., 0.46-m (18-in.) or larger casings instead of 0.36-m (14-in.) casings for the electrodes].

Performance data that show the differences between Tests 1 and 2 are presented in Table 5. The vitrified masses were very comparable, with the block produced during Test 1 only slightly larger than that produced during Test 2. Because higher concentrations of metals were encountered during Test 2, more pooling of molten metal between the electrical phases occurred, causing partial shorting and decreasing the maximum power input possible. As a result, Test 1 averaged 12.5 kW power input while Test 2 averaged 8.9 kW.

TABLE 5. Power System Performance During Engineering-Scale ISV

<u>Parameter</u>	<u>Test 1</u>	<u>Test 2</u>
Power time	16.2 hr	19.2 hr
Total processing time	18.9 hr	19.9 hr
Mass vitrified	155 kg (341 lb)	142 kg (313 lb)
Melt width	0.51 m (20 in.)	0.46 m (18 in.)
Melt depth	0.69 m (27 in.)	0.66 m (26 in.)
Average power	12.5 kW	8.9 kW
Total energy	202 kWh	171 kWh
Energy-to-mass ratio	1.3 kWh/kg	1.2 kWh/kg

The energy-to-mass ratios for Tests 1 and 2 were 1.3 and 1.2 kWh/kg, respectively. Energy-to-mass ratios typically are in the range of 0.8 to 1.0 kWh/kg; however, these ratios were higher because of the lower average power input during the tests. As the average power input decreases, the fraction of that power associated with heat loss increases, resulting in a higher energy-to-mass ratio.

The two vitrified blocks that were produced were narrower than the ISV computer model had projected. It appears that the presence of a molten pool of metal at the bottom of the melt enhanced downward melting. The electrical current was focused at the point of least resistance, creating a higher power density in the glass at the bottom of the melt and promoting downward growth.

Test 1: Electrical Behavior

The most significant observation made during Test 1 was that metals can cause loss of conduction between phases as opposed to a dead short, which was previously thought to be the primary failure mechanism. Electrical data for Test 1 are illustrated in Figure 5. Between 10 and 13 hours run time, the A-phase amperage began a steady decline and the resistance between A-phase electrodes began to increase. This transition occurred between a melt depth of about 0.36 to 0.51 m (14 to 20 in.), which was well into the second layer of metal-laden soil (refer to Figure 2). As a result of the declining amperage, the A-phase power also declined during this period to the point that essentially no power was dissipated between A-phase electrodes. Later in the

FIGURE 5. Test 1 Electrical Data

test, the resistance on B-phase also increased, thus reducing power input. At this point, Test 1 was discontinued. After examining the block, it was observed that large voids had formed in the upper 0.30-m (1-ft)-deep region between the electrodes. The INEL melt, which was more viscous than Hanford melts, trapped the gases that were generated. The soil densification (shrinkage) upon melting, combined with the retention of vitrified soil in the viscous upper region, may have supplemented the void formation by reducing the conductive path between electrodes. This may have attributed to the gradual loss of conductance within the melt. The high melting temperature (1450°C) and high viscosity properties of the native INEL soil were presented in Table 3.

Test 2: Electrical Behavior

Electrical data from Test 2 are presented in Figure 6. This test is characterized by more oscillations in the current and resistance due to shifting of the metal pool that formed in the bottom of the melt zone. Approximately 8 hours into Test 2, when the melt was about 0.30-m (12-in.) deep (0.15 m into the metals zone), A-phase resistance decreased and the power input to A-phase dropped to near zero. This occurred again at 11 hours and 15 hours run time. The B-phase resistance also dramatically decreased at 15 hours, resulting in a near zero power input. These fluctuations in resistance could be caused by the metal pool contacting an electrode and then pulling away as the metal settled to a lower point in the melt zone. When metal contacts an electrode, joule heating of the soil near that electrode ceases and localized cooling begins. When the metal then moves away, the resistance between the affected electrode pair increases and power drops. Depending on the extent of the cooling, power may or may not be restored between the electrode pair. At 18 hours run time, B-phase resistance dropped to less than 0.01 Ω , indicating a near dead short between B-phase electrodes. Power was discontinued a short time later.

Test 2 illustrated an operating experience that is unique to the ISV process. This test went through a number of resistance oscillations that were caused by glass melting and metallic shorting periods. A typical ISV operation follows a declining melt resistance profile as melt depth

FIGURE 6. Test 2 Electrical Data

increases. The presence of metals in the melt adds a variation to this resistance curve. Resistance proceeds normally until a shorting condition exists on one or both of the ISV electrical phases. During a partially shorting condition, as seen many times during this test, the molten glass begins to cool and eventually the metals shift to free the shorted condition. At this point, the cooled glass is more resistive than it was previously and requires a higher potential (voltage) to supply power to the melt. As power input is increased, the molten soil and its high temperature begins melting more metals in the soil. Operations continue until another shorting condition is experienced. Test 2 had a series of these low resistance, metallic shorting, and glass-cooling cycles, followed by an increased resistance, glass reheating, and metallic melting operational cycle. This series of resistance and power oscillations are illustrated by the Test 2 electrical data in Figure 6. These resistance oscillations continued until A phase was shorted and B phase conduction was lost at 18 hours of run time.

PRODUCT CHARACTERIZATION

Post test analysis included visual examination of the two vitrified blocks, a determination of the free and soluble metals, and leach testing to determine product durability.

Visual Examination

Upon visual examination, both test blocks showed only slight subsidence compared with that projected by the ISV computer model. This can be explained by the high melt viscosity of INEL soil at the normal ISV operating temperature (refer to Table 3). The melt viscosity of Hanford soil is approximately 100 poise at 1600°C, while the viscosity for INEL soil was 4000 to 5000 poise. The high viscosity, in conjunction with normal gas generation from within the melt, attributed to large voids that formed in the upper portion of the two blocks, and thus minimized any melt subsidence. These voids and the highly viscous melt probably attributed to the loss of electrical conduction during Test 1.

Free and Soluble Metals

Table 6 presents a summary of the metals incorporated into the vitrified soils during the two engineering-scale ISV tests. Metals accounted for 12.5 wt% of the Test 1 block and 15.9 wt% of the Test 2 block. The bottom of each block was fractured to examine the amount and location of free metal in the melt. Metals were present mainly as pools at or near the bottom of the melt. The metal pool removed from the Test 1 block is shown in Figure 7. Figure 8 shows some of the partially melted or encapsulated metals protruding from the sides and bottom of the Test 2 block. Small spheres dispersed through the glass phase were observed in both Test 1 and Test 2. The spheres range in size from 1 μm to over 1 mm, and were determined by a scanning electron microscope (SEM) to be primarily iron. This is consistent with the reducing conditions in the molten soil. Figure 9 shows a micrograph of the dispersed spheres from Test 1. The spheres are calculated from the micrograph to comprise 0.1 vol% (0.3 wt%) of the glass phase.

Iron and stainless steel were also observed to have solubilized into the glass in the form of Fe_2O_3 , Cr_2O_3 , and NiO . Table 7 shows the ICP analysis of vitrified soil from different locations within each vitrified block. Concentrations of Fe_2O_3 , Cr_2O_3 , and NiO all increased over the original soil composition (see Table 2). Based on the mass of glass produced and the average concentration of these oxides within each block, calculations indicate

TABLE 6. Metals Content of Vitrified Soil

Metals Form	Metals Mass, kg	
	Test 1	Test 2
Main pool at bottom of melt	6.8	16.1
Smaller pool(s)	3.6	2.5
Partially melted or encapsulated	8.2	3.4
Small dispersed spheres	0.45	0.39
Solubilized metal	0.29	0.24
Total metals in melt	19.3 kg	22.6 kg
Percent metals in melt	12.5 wt%	15.9 wt%

FIGURE 7. Metal Pool from Bottom of Test 1 Block (88083116-43CN)

that 0.29 kg of the metals (1.4% of total metals) solubilized during Test 1, compared with 0.24 kg (1.0% of total metals) during Test 2. With larger scale ISV operations, the metals solubilization could increase since longer operational time is expected to favor the kinetics of solubilization.

Questions must be resolved regarding the metals content that is achievable as the ISV process progresses from engineering scale to large scale. Scale-up could be proportional to the percent metals in the soil, the depth of the metal pool that forms at the melt bottom, or some other parameter such as total metal mass. The ratio of the electrode separation to the melt depth could also affect the metals content limit. Because of these factors, the total percent metals achieved during engineering-scale ISV testing is

FIGURE 8. Partially Melted or Encapsulated Metals Protruding from Test 2 Block (8809121-62CN)

difficult to relate directly to large-scale operations. Intermediate pilot-scale testing is a recommended low cost and low risk method for evaluating scale-up parameters.

Leach Testing

The dissolution properties (durability) of vitrified INEL soil samples have been evaluated using the Materials Characterization Center MCC-1 test procedure. The free metals encased in the vitrified soil were not characterized because their characteristics will be very dependent on the actual waste makeup. The MCC-1 static leach test measures the elemental mass loss of a monolith sample of glass as a function of time. For this test, a glass sample is suspended within a sealed Teflon container by a Teflon support

FIGURE 9. Dispersed Metal Spheres in Vitrified Glass from Test 1 (39737)

grid. The test results are based on leachate ICP analysis from which the total concentrations of materials removed from the sample are determined. For this evaluation, testing was conducted with a surface area/volume ratio (SA/V) of 10 m^{-1} in a leachate of deionized water (DIW) at 90°C for 28 days. To allow the direct comparison of data, the leachate, temperature, and SA/V used in testing the ISV waste form are the same as those parameters used to test high-level nuclear waste borosilicate glasses.

To allow the comparison of leach test results from waste forms with different compositions, the results are given in terms of normalized elemental mass release for the MCC-1 test. Normalized elemental mass releases were calculated with the following equation:

$$NR_i = m_i / (F_i \times SA)$$

TABLE 7. Vitriified Soil Sample Analyses

Oxide	Reference Soil Composition, wt%	Test 1 Compositions, wt%				Test 2 Compositions, wt%			
		Lower Center	Near Electrode	Cold Cap	Test 1 Average	Lower Center	Bottom Edge	Side	Test 2 Average
SiO ₂	62.60	67.60	65.00	65.40	66.00	67.30	67.00	66.60	66.97
Al ₂ O ₃	11.85	13.40	13.30	12.90	13.20	13.20	13.40	12.90	13.17
Fe ₂ O ₃	4.25	4.58	4.61	5.17	4.79	4.35	5.14	4.98	4.82
CaO	3.68	3.85	3.80	4.38	4.01	3.62	3.49	3.93	3.68
K ₂ O	2.99	2.30	2.90	2.60	2.60	3.10	2.80	2.90	2.93
MgO	1.72	1.92	2.01	2.00	1.98	1.90	1.92	2.02	1.95
Na ₂ O	1.37	1.50	1.36	1.75	1.54	1.42	1.54	1.49	1.48
TiO ₂	0.68	0.80	0.80	0.76	0.78	0.80	0.84	0.81	0.82
MnO ₂	0.10	0.28	0.26	0.10	0.21	0.33	0.11	0.12	0.19
BaO	0.09	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
ZrO ₂	0.05	0.09	0.08	0.28	0.15	0.12	0.08	0.07	0.09
B ₂ O ₃	0.05	0.10	0.06	0.33	0.16	0.08	0.06	0.06	0.06
NiO	0.04	0.06	0.04	0.05	0.05	0.06	0.05	0.04	0.05
SrO	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03
Cr ₂ O ₃	0.02	0.21	0.19	0.03	0.14	0.19	0.03	0.04	0.08
Total	89.5	96.81	94.54	95.86	95.74	96.59	96.58	96.09	96.42

where NR_i = normalized mass release of element i (g/m^2)
 m_i = mass of element i in leachate (g)
 F_i = fraction of element i in glass (dimensionless)
 SA = surface area of sample monolith (m^2)

The 28-day MCC-1 leach tests were conducted on both the Test 1 and Test 2 glasses. The results from the MCC-1 leaching tests are presented in Table 8, along with leaching data for vitrified Hanford soil and a high-level nuclear waste glass for comparison.

The normalized releases of Si, Al, and Na from the vitrified INEL soils averaged at least 4 times less than releases for the vitrified Hanford soil, and averaged at least an order of magnitude lower than the releases from the high-level nuclear waste glass. The leach rate for Fe in the vitrified INEL soils was higher for the INEL sample than for the Hanford and SRL165 waste glass. This anomaly is probably due to the higher free iron content of the vitrified INEL soils. Both Cr and Ni concentrations (present in the stainless steels added to the test) were below detection limits in the INEL leachates. Based on the most prominent element, Si, the dissolution rate of the vitrified soil is very low at $0.036 \text{ g}/\text{m}^2 \cdot \text{d}$. Because the ambient storage temperature of the ISV waste form will be nearer 20°C than 90°C , the actual

TABLE 8. Normalized Elemental Releases of Vitrified Materials(a)

Element	Normalized Releases, g/m^3			
	Vitrified INEL Soil Test 1	Vitrified INEL Soil Test 2	Vitrified Hanford Soil(b)	SRL165 Nuclear Waste Glass(c)
Si	1.0	0.65	4	17
Al	0.67	0.46	4	12
Fe	0.31	2.6	0.1	0.18
Ca	0.83	0.51	4	0.87
Na	1.1	0.81	5	21

(a) 28-day MCC-1, 90°C , DIW.

(b) Buel et al. 1987.

(c) Chandler et al. 1984.

release rate for the vitrified INEL waste form should be one to two orders of magnitude lower than shown due to the temperature reduction (Grambow 1984).

OFF-GAS CHARACTERIZATION

Off-gas particulate data were obtained via an EPA modified method 5 sample train (see Figure 4). Table 9 lists the data obtained from the off-gas particulate sampling train. The concentration of particulate in the off-gas is reported as grams/dscf, grains/dscf, and mg/m^3 . The solids content of the untreated off-gas was quite low, averaging 23.3 mg/m^3 during Test 1 and 56.5 mg/m^3 for Test 2. This is nearly an order of magnitude below the Hanford standards for nonradioactive stack emissions of 230 mg/m^3 for particulates.

Solids were scraped from the glass filter and analyzed for cation composition by ICP. Composition of the filter particulate from the tests is compared to the original soil and filter blank compositions in Table 10. The absence of SrO in the Test 1 and 2 particulate samples indicates that very little filter material was in the samples that were analyzed. The more volatile components within the soil, such as sodium, boron, lithium, phosphorus, lead, and zinc, are enriched in the off-gas particulate. Elements predominant in steel and stainless steel appeared in lower concentrations in the

TABLE 9. Off-Gas Particulate Sampling Data

<u>Parameter</u>	<u>Test 1</u>	<u>Test 2</u>
Average stack temperature, °C	66	62
Moisture in stack, %	4.0	<1.0
Volume off-gas sampled, dscf	928.1	638.0
Weight particulate collected, g (blank corrected)	0.61	1.0
Concentration of particulate in off-gas		
g/dscf	6.6×10^{-4}	1.6×10^{-3}
grains/dscf	1.0×10^{-2}	2.4×10^{-2}
mg/dry std m^3	23.3	56.5

TABLE 10. Off-Gas Particulate Composition

<u>Oxide</u>	<u>Composition, wt%</u>			
	<u>Reference INEL Soil</u>	<u>Filter Blank</u>	<u>Test 1 Particulate</u>	<u>Test 2 Particulate</u>
SiO ₂	62.60	50.5	64.0	24.1
Al ₂ O ₃	11.85	13.4	0.89	0.48
Fe ₂ O ₃	4.25	0.32	0.72	0.71
CaO	3.68	20.2	0.30	0.28
K ₂ O	2.99	--	1.72	4.07
MgO	1.72	0.56	0.46	0.34
Na ₂ O	1.37	0.78	3.79	7.37
TiO ₂	0.68	0.60	0.05	0.02
MnO ₂	0.10	0.10	0.14	0.12
BaO	0.09	0.006	0.014	0.003
ZrO ₂	0.05	--	0.05	0.04
B ₂ O ₃	0.05	6.90	8.48	6.90
NiO	0.04	--	0.02	--
SrO	0.02	0.12	0.01	--
Cr ₂ O ₃	0.02	0.02	0.01	0.13
Li ₂ O	0.012	0.011	0.75	0.88
P ₂ O ₅	--	--	1.19	3.18
PbO	--	--	0.55	0.92
ZnO	--	--	1.57	3.60
Total	89.5%	93.5	84.7	53.1

off-gas particulate than in the original soil. These particulate releases can be effectively retained by a conventional off-gas treatment system consisting of a wet scrubber and filtering media.

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