

IN SITU VITRIFICATION DEMONSTRATION FOR THE STABILIZATION OF BURIED WASTES AT THE OAK RIDGE NATIONAL LABORATORY

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ABSTRACT

A demonstration of In Situ Vitrification (ISV) technology for the stabilization of radioactively contaminated soil sites at the Oak Ridge National Laboratory (ORNL) was successfully completed during July 1987. This demonstration is the first application of the ISV process not performed at the Hanford Site, where the technology was developed and patented by Pacific Northwest Laboratory (PNL). The joint ORNL-PNL pilot-scale demonstration was performed on a 3/8-scale trench (2 m deep x 1 m wide x 10 m long) that was constructed to simulate a typical seepage trench used for liquid low-level radioactive waste disposal at ORNL from 1951 to 1966. In the ISV process, electrodes are inserted around a volume of contaminated soil, power is applied to the electrodes, and the entire mass is melted from the surface of the soil down through the contaminated zone, thus making a glassy-to-microcrystalline waste form that incorporates the contaminants. Gases produced during the melting are collected, treated, monitored, and released through an off-gas process trailer. In the ORNL demonstration, a 25-t mass of melted rock approximately 1.2 m thick x 2.1 m wide x 4.9 m long was formed during 110 h of operation that consumed approximately 29 MWh of power. Data obtained on the operational performance of the test and waste-form durability will be used to assess the feasibility of applying the ISV technology to an actual waste trench.

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- 1 Operated by Martin Marietta Energy Systems, Inc., under contract DE-AC05-84OR21400 with the U. S. Department of Energy.
 - 2 Operated by Battelle for the U. S. Department of Energy under contract DE-AC06-76RL01830.

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INTRODUCTION

A series of seven seepage pits and trenches (see Fig. 1) were used between 1951 and 1966 for the disposal of approximately 2.5×10^7 gal of liquid radioactive wastes at the Oak Ridge National Laboratory (ORNL). Approximately 200,000 Ci of ^{90}Sr and 600,000 Ci of ^{137}Cs , along with smaller quantities of other fission products, uranium, and transuranium elements, were disposed of in this series of pits and trenches. To facilitate the seepage of liquids, the trenches were constructed on the tops of ridges, were oriented perpendicular to the strike of the bedding of the formation, and were filled with crushed limestone or dolomite (see Fig. 2). As the liquids seeped out, the Cs and Sr remained within, or in close proximity, to the trenches. Cesium is generally irreversibly sorbed by the illite-rich soils at ORNL. Strontium, on the other hand, is poorly sorbed. To reduce the mobility of Sr, the pits and trenches were treated with a highly alkaline solution (NaOH) at the time of disposal. All of the pits and trenches are now covered with asphalt caps to reduce the direct flow of precipitation through the waste. Currently, the pits and trenches do not significantly contribute to surface-water contamination, and most of the Cs and Sr remains in close proximity to the bottom of the pits and trenches. However, the large inventory of ^{137}Cs and ^{90}Sr , the close proximity of the wastes to the surface, and the potential for significant releases in the future necessitates either a long-term site maintenance and monitoring program or some form of remedial action to allow the sites to be permanently decommissioned.

In Situ Vitrification (ISV) is one possible technology that could be applied to the pits and trenches (others include grouting and ground densification). ISV, developed and patented by Pacific Northwest Laboratory (PNL), involves placing four electrodes in a square array around the contaminated volume of soil, applying power to the electrodes, and melting the entire mass of contaminated soil into a homogenous and durable glassy-to-microcrystalline waste form. The melting begins at the surface of the soil and progresses downward through the contaminated zone. Gases produced during the high-temperature (1600 to 2000°C) operation are collected through an off-gas hood and scrubbed of possible contaminants in a process trailer. The ISV technology has been extensively tested by PNL at electrode spacings from approximately 0.3 to 6 m. The pits and trenches at ORNL are candidates for vitrification because of their small areal extent and shallow depth (<6 m). The potential for personnel exposure from the high concentrations of ^{90}Sr and ^{137}Cs in the pits and trenches makes an *in situ* technology highly desirable compared with one that would require excavation of the contaminated zone.

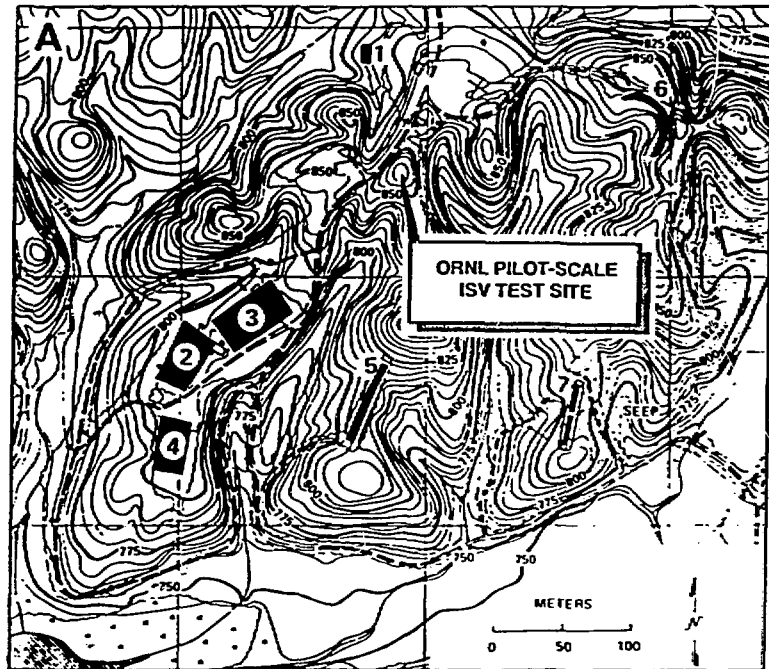
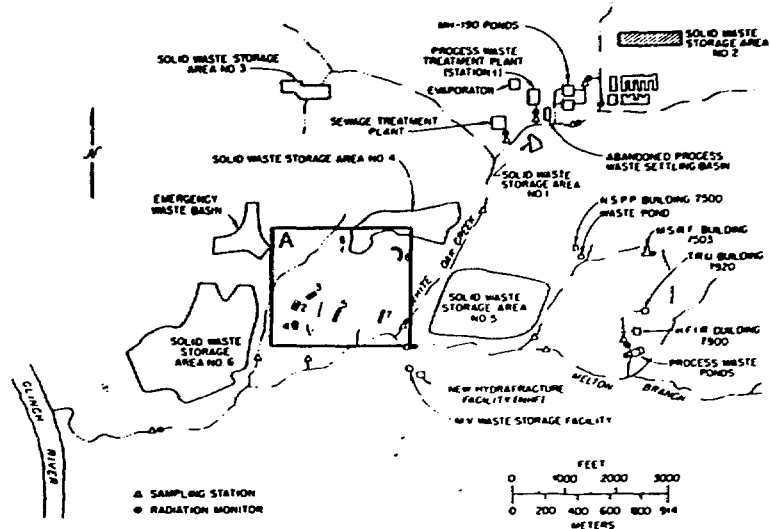


Fig. 1. Location of the radioactive liquid-waste disposal pits and trenches at ORNL and the pilot-scale ISV demonstration.

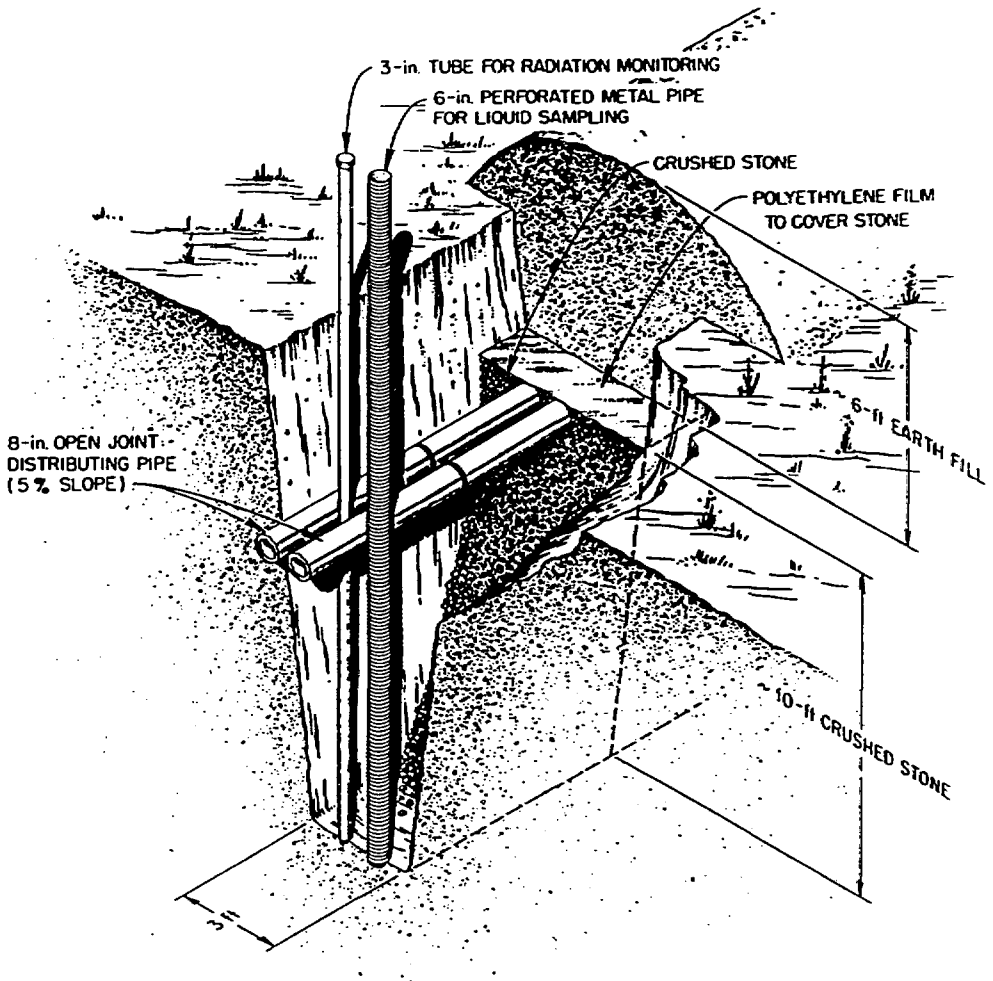


Fig. 2. Construction details of ORNL liquid-waste disposal Trench 7.

The pilot-scale demonstration of ISV technology was performed jointly by ORNL and PNL to achieve four main objectives: (1) complete an application of ISV technology away from the Hanford Site to evaluate the feasibility of technology transfer to ORNL, (2) assess the operational performance of ISV for applications in heterogenous, high-carbonate soils and rocks, (3) determine the retention factors (mass in melted soil divided by mass in off-gas) for Cs and Sr under field conditions, and (4) evaluate the durability of the waste form produced in the ORNL soil system. The results from this pilot-scale test will be used to determine the feasibility of applying the ISV technology to an actual waste trench at ORNL.

ISV PROCESS DESCRIPTION

The ISV process as applied to the stabilization of contaminated soils at ORNL involves placing four molybdenum electrodes into a square array of augered holes that bound the contaminated zone. Figure 3 illustrates the sequence of the process. A starter path for electrical current is established by placing a small amount of a mixture of graphite and glass frit between the electrodes on the surface of the soil. Dissipation of power through the starter material creates temperatures high enough to melt a layer of soil, thus establishing a molten, conductive path. This molten zone continues to grow downward and encompass the soil. Less-dense material sometimes creates a layer of rock that floats near the surface of the melt until it is eventually incorporated into the molten mass.

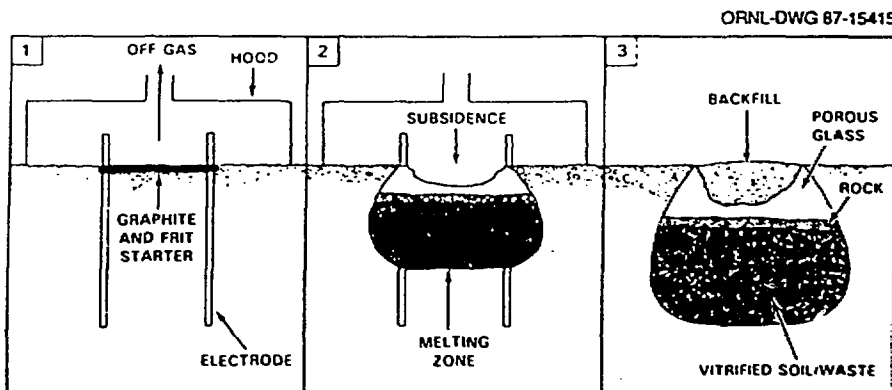


Fig. 3. Schematic illustration of the ISV operating sequence.

At the high temperatures (1600 to 2000°C) created, organic materials pyrolyze, and the pyrolysis products diffuse to the surface and combust. In addition, water vapor and carbon dioxide are released from the soil. The movement of the resultant gases through the melt can produce some porosity in the final product near the surface of the melt. A hood over the vitrification zone is maintained under a slightly negative pressure to collect off-gases and the small percentage (<0.01 wt%) of particulates that are released with the off-gas from the molten mass. The hood also provides support for the electrodes. The off-gases from the process are collected, treated, and monitored to ensure that only environmentally safe levels of potential contaminants are released. The remaining noncombustible materials dissolve or become encapsulated into the molten soil. Thermally induced convective currents within the molten soil help to homogenize the final waste form, which physically resembles natural obsidian or basalt glass.

The principles of ISV are based on developments from work performed at PNL on joule-heated melters for various nuclear waste immobilization projects (Buel et al. 1979). The joule-heating principle involves internal resistance heating as electrical current passes through the molten media. In ISV, the resistance decreases as the molten mass increases in size. To maintain a power level high enough to continue melting additional soil, the current must be increased. To accomplish the variable current during ISV processing, a power transformer with multiple voltage taps is used. At start-up, the ISV process requires high voltage and low amperage. As the melt progresses and resistance decreases, the lower voltage taps on the power transformer allow increased amperage to be applied to the melt, maintaining a high power level. The process continues until heat losses from the melt approach the energy delivered to the soil via the electrodes or until power is discontinued to the electrodes.

PILOT-SCALE TEST SYSTEM

The pilot-scale test system at ORNL used four electrodes having a 1.2-m separation and consists of a power-control unit and off-gas containment hood over the test site. A cutaway view of the support trailer and off-gas hood are illustrated in Fig. 4. Prior to the ORNL test, this same system had been used on 11 pilot-scale tests at the Hanford site.

Power-Delivery System

The pilot-scale power system uses a Scott-Tee connection to transform a three-phase input to a two-phase secondary load using diagonally opposed electrodes in a square pattern. The 500-kW power supply may be either voltage or current regulated. The alternating

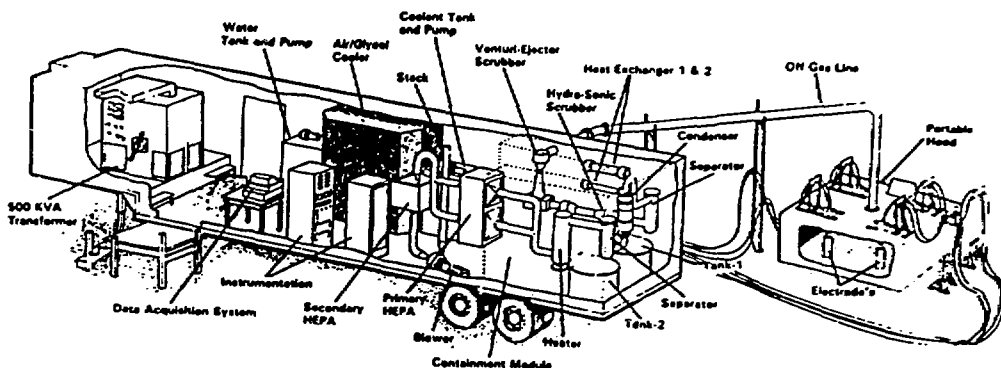


Fig. 4. Cutaway view of the pilot-scale process trailer and off-gas hood.

primary current is rated at 480 V, 600 A, 3 phase, and 60 Hz. The three-phase input feeds a Scott-Tee connected transformer (Fig. 5), which provides a two-phase secondary load. The transformer has four separate voltage tap settings of 1000, 650, 430, and 250 V. Each voltage tap has a corresponding amperage rating of 250, 385, 580, and 1000 A per phase, respectively. The amount of three-phase input power delivered to the transformer is controlled by adjusting the conduction angle of the thyristor switches located in each of the three input lines. These switches, in conjunction with selectable taps on the transformer secondary, regulate the amount of power deliverable to both secondary phases. The Scott-Tee connection provides an even power distribution among the three primary phases when the molten zone approaches a uniform resistance load. During the test at ORNL, this power system proved effective in maintaining a balanced load to the electrodes.

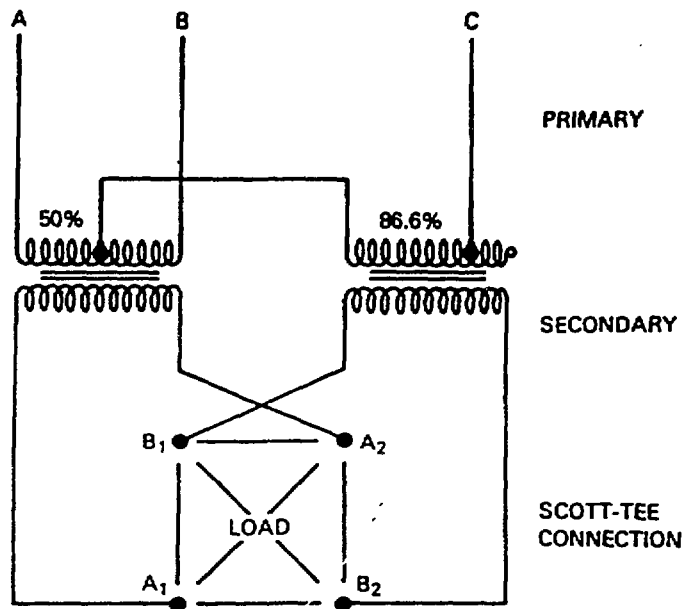


Fig. 5. Scott-Tee electrical connection for the pilot-scale ISV system.

Off-Gas Containment and Electrode-Support Hood

The off-gas containment and electrode-support hood, constructed from seven panels of 20-gauge stainless steel bolted together, is 3.1 m wide, 5.5 m long, and 0.9 m high. Four leveling supports are attached to the corners of the hood, which also has a port for viewing (and access to) the surface of the melt. A central off-gas port allows direct coupling of the hood to the processing trailer and off-gas treatment system. The hood is equipped with heat fins installed on the surfaces of panels to help cool the hood to which radiant heat is transferred from the partially molten surface during processing. The hood, designed to withstand a water vacuum of 18 cm, was sealed to the surface of the soil surrounding the molten zone by means of a flexible skirt of tightly-woven, high-temperature-resistant fiber attached to the bottom of all side panels. The skirt extended approximately 0.6 m away from the hood to allow for a hood-to-ground seal when covered with a layer a soil. Electrical bus bars connected to the molybdenum electrodes protrude through the hood; these were surrounded by electrically insulated sleeves that allowed adjustment of the electrode positions. The electrodes and bus bars were supported by insulators above the sleeves. The insulators were designed to withstand movement of the molten mass against the electrodes from convective currents and the gravitational and buoyant forces exerted on the electrodes.

Off-Gas Treatment System

The off-gas treatment system is shown schematically in Fig. 6. The off-gas passes through a venturi-ejector scrubber and separator, a Hydro-Sonic^{*} scrubber, a separator, a condenser, another separator, a heater, two stages of HEPA filtration, and a blower. Liquid to the two wet scrubbers is supplied by two independent scrub recirculation tanks, each equipped with a pump and heat exchanger. The entire off-gas system has been installed in a 13.7-m-long semitrailer to facilitate transportation (see Fig. 4). All of the off-gas components except the second-stage HEPA filter and blower are housed within a removable containment module that has gloved access and is maintained under a slightly negative pressure to protect workers from potential contamination. Heat is removed from the off-gas by a closed-loop cooling system consisting of an air/liquid heat exchanger, a coolant storage tank, and a pump. A 1:1 mixture of water and ethylene glycol is pumped first from the storage tank through the shell side of the condenser and the two scrub-solution heat exchangers, and then through the air/liquid exchanger, where heat is removed from the coolant.

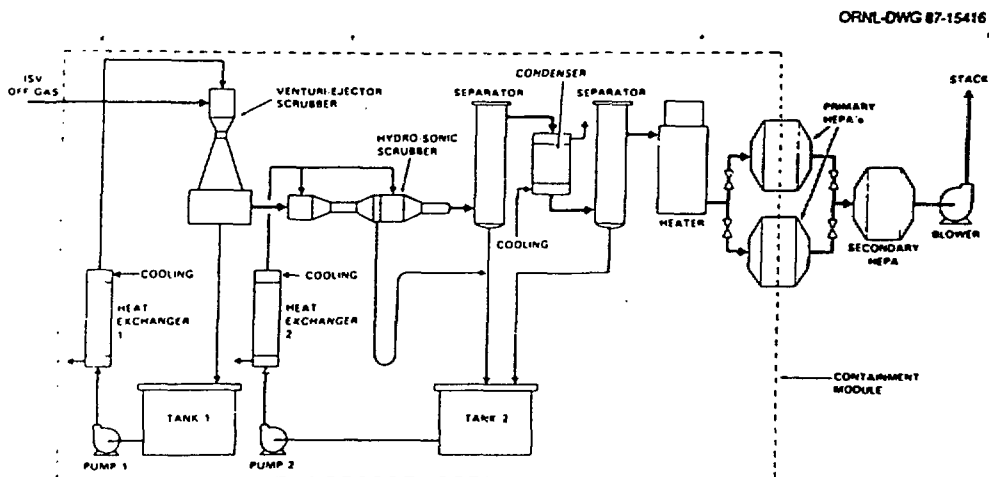


Fig. 6. Schematic drawing of the pilot-scale ISV off-gas treatment system.

* Hydro-Sonic scrubber is a product of Hydro Sonic Systems, Dallas, Texas.

The venturi-ejector scrubber serves both as a quencher and high-energy scrubber. The second scrubber is a two-stage Hydro-Sonic scrubber (tandem nozzle scrubber) as illustrated in Fig. 7. The first section condenses vapors, removes large particles, and initiates growth of the finer particles so that they are easily captured in the second stage. Particles are captured when the gas is mixed with fine water droplets produced by spraying water into the exhaust of the subsonic nozzle. Mixing and droplet growth continue down the length of the mixing tube. Large droplets containing the particles are then removed by a vane separator and drained back into the scrub tank. When operated at a differential pressure of 127 cm of water, the unit is designed to remove over 90% of all particles greater than $0.5 \mu\text{m}$ in diameter. Efficiency of removal increases with an increase in pressure differential.

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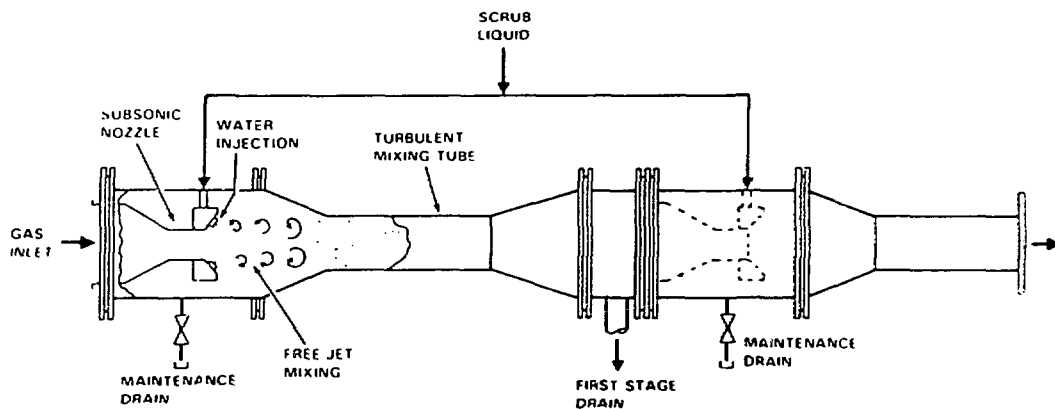


Fig. 7. Tandem Nozzle Hydro-Sonic Scrubber (Hydro Sonic Systems, Dallas, Texas).

Additional water is removed from the gas system by a condenser having a heat exchange area of 8.9 m^2 and a final separator. The gases are then reheated to approximately 25°C above the dew point in a 30-kW heater to prevent condensate carryover to the HEPA filters. The first stage of filtration consists of two $61 \times 61 \times 29 \text{ cm}$ HEPA filters in parallel. During operation, one filter is used and the other remains as a backup in case the operating filter becomes loaded. The primary filter can be replaced during operation. The second-stage HEPA filter acts as a backup in case a first-stage filter fails.

Test Description

The objectives of the pilot-scale demonstration of ISV technology at ORNL were developed to address key differences between the conditions during previous tests of the ISV technology and those anticipated for contaminated sites at ORNL. For example, the waste trenches at ORNL contain large quantities of ^{137}Cs that could volatilize at high temperatures and be carried into the process trailer with the off-gas, resulting in additional operator exposure and excess wastes. Therefore, it was necessary that the retention factor of $>10,000$ for Cs (Cs in melted soil divided by Cs in off-gas) obtained in engineering-scale tests (Carter et al. 1987) be confirmed at a larger scale under field conditions. Also, the soils at ORNL are more structurally and chemically heterogeneous than those used in previous tests at Hanford. The trench design, with a significant quantity of crushed carbonate rock present, results in a bulk composition of the ISV melt lower in silica and higher in calcium and magnesium than previously studied compositions (Oma et al. 1983). Therefore, the operational performance of the ISV technology as well as the long-term durability of the resulting waste form need to be addressed specifically for sites at ORNL. Some of these issues have been investigated during lab- and engineering-scale tests conducted at PNL during 1985-1986 (Carter et al. 1987). Evaluation of ISV technology on the pilot scale in actual soil at ORNL will provide for more confident scale-up predictions.

Trench 7 (see Figs. 1 and 2) was chosen as a model because of its size, inventory characteristics, and the fact that much characterization of the trench has already been accomplished (Olsen et al. 1983). To enable the pilot-scale ISV system to be used, a 3/8-scale model of Trench 7 was constructed in a pristine (i.e., uncontaminated) portion of ORNL (see Fig. 1). The site, located on top of a ridge in the Maryville Limestone (an interlayered limestone-shale), was chosen for the similarity of its physical and geological characteristics to the areas used for seepage disposal in the past at ORNL. After preparation of the site (clearing, leveling, electrical service, etc.), the trench was constructed perpendicular to the strike of the bedding. The 9.2-m-long trench was 1 m wide at the top and tapered to 0.4 m at the bottom. A schematic cross section at the midpoint of the trench is shown in Fig. 8 (see Fig. 10 also). The trench was constructed to a depth of 1.5 m, except for the central section, which was excavated to a depth of 2.5 m to allow for the placement of a vertical array of eight type-K thermocouples (one thermocouple at every 0.31 m depth) for monitoring the depth of the melt. To simulate the contaminated sludge that is present in Trench 7, 526 kg of a mixture of 18 wt% Cs- and 82 wt% Sr-carbonate was placed in the central portion of the trench. These quantities of Cs and Sr were selected to yield a waste form with sufficient concentrations of Cs and Sr that their leach characteristics could be

CENTRAL CROSS SECTION OF ISV DEMONSTRATION TRENCH

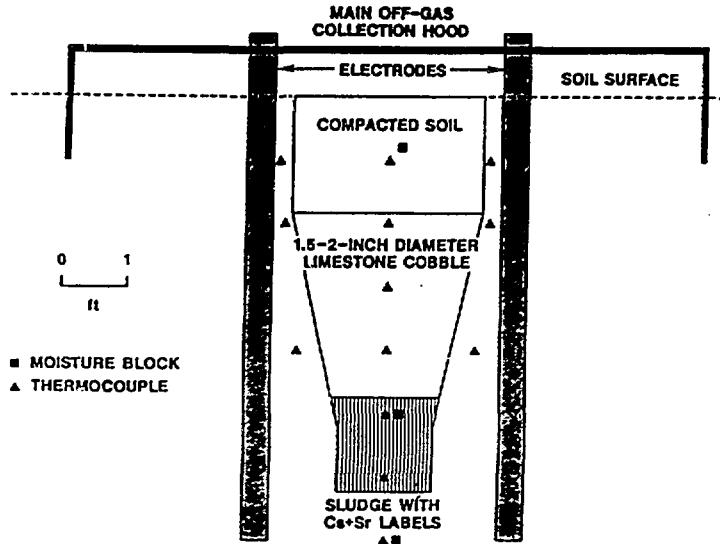


Fig. 8. Central cross section of the pilot-scale ISV demonstration trench (approximate scale).

determined. The entire trench was then filled from the 0.6- to 1.5-m level with crushed carbonate rock. The upper 0.6 m of the trench was backfilled with original soil. In addition to the array of thermocouples in the center of the trench, type-K thermocouples were placed at depths of 0.6, 1.2, and 1.5 m along both sides of the trench at distances of 2.1 and 3.1 m from the center of the trench. Three type-R thermocouples, which have a higher maximum operating temperature than type-K thermocouples, were placed in a vertical array at the center of the trench at depths of 0.9, 1.5, and 2 m in an unsuccessful attempt to monitor melt temperatures. Moisture detection cells were placed in the trench at several locations but did not function properly and will not be discussed further. The four molybdenum electrodes (5 cm diameter and 3.7 m long) were placed in graphite sleeves (15 cm O.D.) and placed 1.2 m apart in a square array of augered holes approximately 2.5 m deep. The off-gas hood was then placed over the trench and connected to the off-gas treatment system. The electrodes were connected to the power-delivery system. Run data (temperatures, off-gas flow rate and CO and CO₂ concentrations, electrical parameters, etc.) were recorded every 6 min and logged into a computerized data storage system. Off-gas samples were collected approximately every 2 h (duplicate samples were obtained for analysis at both PNL and ORNL).

RESULTS

After one false start on June 26, 1987 (a result of transformer circuitry malfunctions), the ISV test was started on July 15, 1987. The test ran continuously for 110 h until power was shut off on July 19. Soil temperatures and manual probing of the melt with a steel rod confirmed that the desired depth (approximately 2.1 m) of melting had been reached. Final analysis of the test is still in progress - preliminary results and interpretations are presented here.

Figure 9 illustrates key operational parameters as a function of run time. As discussed earlier, the trends of decreasing voltage and increasing amperage during operation are necessary to maintain melting. Tap changes, indicated by the sharp increases in the total power, were made at run times of approximately 10, 20, and 30 h. The total power consumed during the test was approximately 29 MWh. The depths of melting were determined by observing the maximum temperature reached at each thermocouple in the vertical array located at the center of the trench. Upon contact with the melt, type-K thermocouples generally burn out, giving a sharp response on the data-logging system and making depth determinations relatively easy. Based on visual observations of the surface of the soil and temperatures recorded by the thermocouples throughout the trench, the final dimensions of the melted mass are approximately 4.9 m long x 2.1 m wide x 1.2 m thick (see Fig. 10). Assuming that approximately 1.2 kWh are required to melt 1 kg of soil at the test site (based on water and CO₂ content), the estimated mass of soil that was vitrified is 25 t. This power-to-melt conversion factor is an estimate and will be refined through further analysis of the run data and melt characteristics. As of August 28, 1987, the edges of the mass had cooled to temperatures <70°C.

The flow rate of the off-gas at the stack and the concentration of CO₂ in the off-gas are illustrated in Fig. 11. It is interesting to note that the time (approximately 20 h) of the increase in CO₂ concentration corresponds to the time that the depth of melting reached 0.6 m, where the melt should have first encountered the crushed carbonate rock. Samples of the off-gas scrub solutions are now being analyzed so that the Cs and Sr retention factors can be determined. Samples of the crushed carbonate rock and soil used in the trench, wall-smears from the off-gas conduits, and HEPA filters are also being analyzed for Cs and Sr so that a mass balance for the system can be calculated. Based on results from an engineering-scale (1/12-scale of Trench 7) test (Carter et al. 1987), the retention factors are anticipated to be >10,000 for both Cs and Sr.

PILOT-SCALE ISV OPERATIONAL PARAMETERS

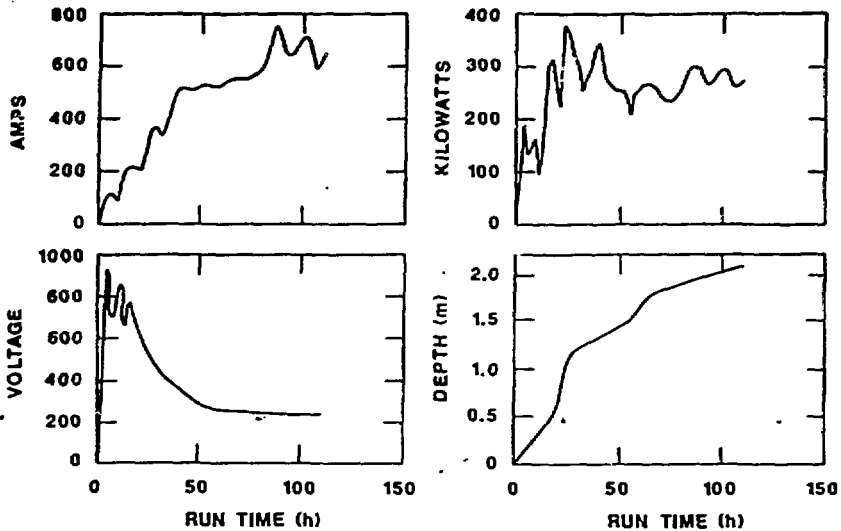
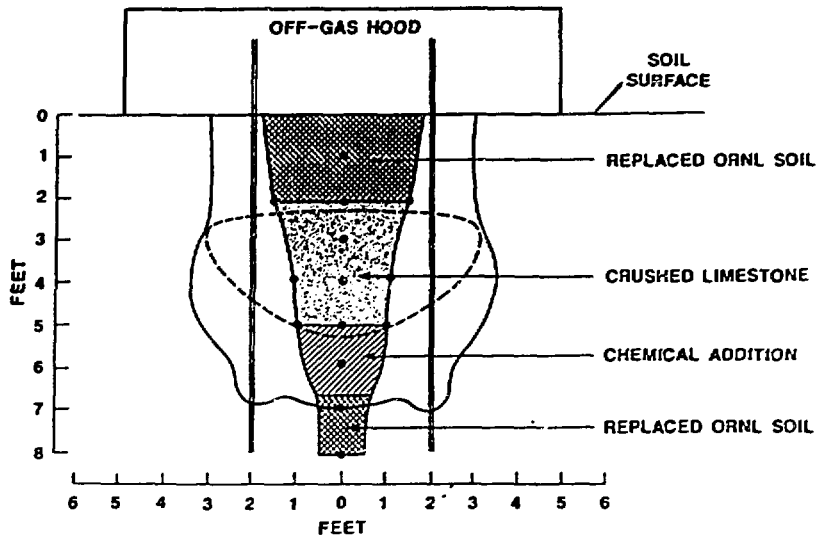
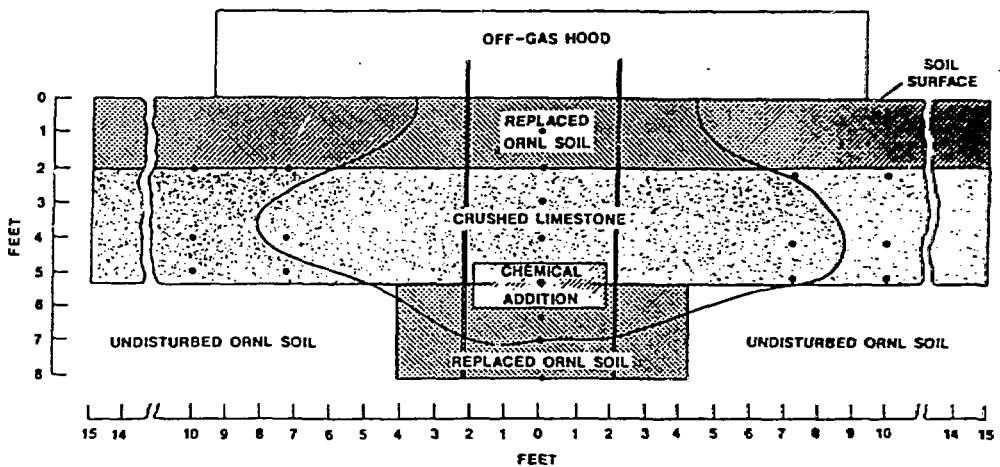


Fig. 9. Voltage, amperage, power, and melt depth as a function of run time. Voltage and amperage are averages of the A and B phases.

The vitrified mass will be cored so that samples of the waste form can be characterized and tested for its durability. Results from the engineering-scale test suggest that the waste form will have a bulk composition (weight percent) approximated by 48% SiO_2 , 16% Al_2O_3 , 16% CaO , 12% MgO , and 5% FeO . Depending on the cooling rate achieved in the pilot-scale mass, the final product may be either a glassy, obsidian-like solid or a cryptocrystalline mixture of mineral phases. Carter et al. (1987) performed heat treatments designed to simulate the slow cooling of a bulk composition similar to that expected in a full-scale test. The resulting product was a microcrystalline mixture of diopside, enstatite, and Al-silicates (likely hosts for Cs and Sr). MCC-1 28-d leach tests on this material resulted in normalized elemental releases of approximately 20 and 4 g/m^2 for Cs and Sr, respectively. These values are equal to or lower than values reported for PNL-76-68 glass (a waste form considered for high-level nuclear waste). Values for Si, Ca, and Al are also equal to or lower than those for PNL-76-68 glass. Samples of the pilot-scale waste form will be subjected to a variety of durability and simulated-weathering tests so that the long-term performance of the waste form can be quantified. Results from the off-gas analyses, characterization of the waste form, and durability tests will be used to assess the feasibility of a full-scale application of ISV to an actual waste trench at ORNL.



END VIEW LOOK SOUTH TO NORTH



SIDE VIEW LOOK WEST TO EAST

Fig. 10. Predicted shape of the pilot-scale ISV mass (overlaid on the original trench dimensions). Solid circles are locations of type-K thermocouples (see discussion on page 12).

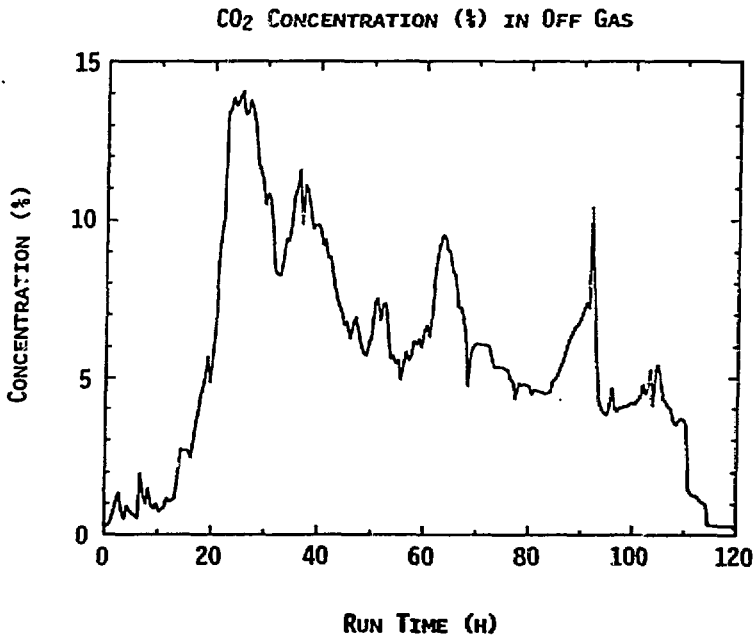
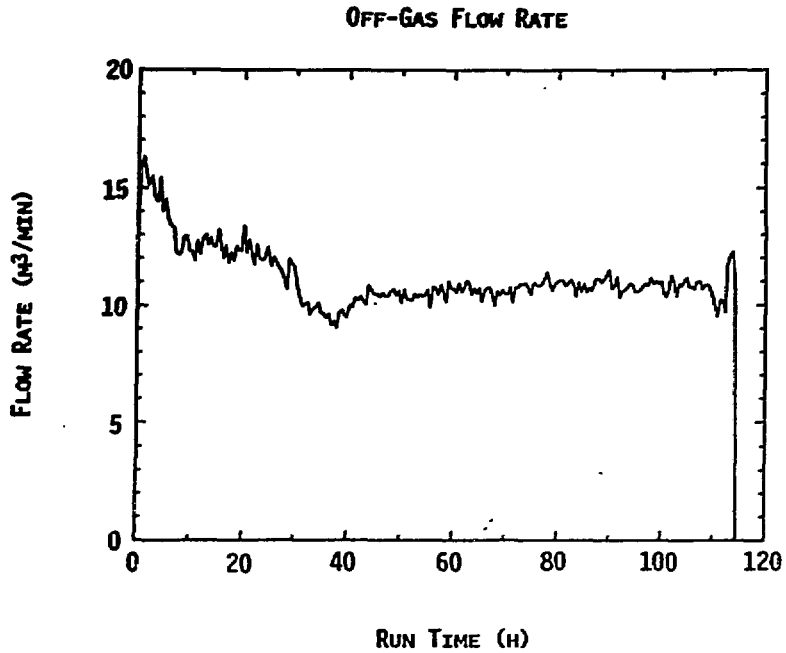


Fig. 11. Flow rate and CO₂ content of off-gas from the pilot-scale ISV demonstration.

SUMMARY

A demonstration of ISV technology for the stabilization of radioactively contaminated soil sites at ORNL was successfully completed during July 1987. This demonstration is the first application of the ISV process not performed at the Hanford Site, where the technology was developed and patented by PNL. The joint ORNL-PNL pilot-scale demonstration was performed on a 3/8-scale trench (2 m deep x 1 m wide x 10 m long) that was constructed to simulate a typical seepage trench previously used for liquid low-level radioactive waste disposal at ORNL from 1951 to 1966. A 25-t mass of melted rock approximately 1.5 m thick x 2.5 m wide x 5 m long was formed during 110 h of operation that consumed approximately 29 MWh of power. Data being obtained on the operational performance of the test and waste-form durability will be used to assess the feasibility of applying the ISV technology to an actual waste trench.

ACKNOWLEDGEMENTS

Staff at ORNL that contributed to the success of the demonstration included C. Abner, C. Brown, C. Bruce, D. Farmer, C. Thomas, R. Thomas, and R. Todd. Staff from PNL who helped to prepare the site and run the test included J. Buelte, K. Eliason, T. Hinkle, M. Longaker, K. Oma, T. Powell, and C. Timmerman. We gratefully acknowledge the help of numerous other persons at ORNL who helped in the preparation of the site. We thank K. L. Von Damm and A. D. Kelmers for their constructive comments that helped to improve this manuscript. Publication No. 3007, Environmental Sciences Division, ORNL.

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