IMPLEMENTATION OF IN SITU VITRIFICATION TECHNOLOGY FOR REMEDIATION OF OAK RIDGE CONTAMINATED SOIL SITES: PAST RESULTS AND FUTURE PLANS

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ABSTRACT

In situ vitrification is a thermal treatment technology being developed for remediation of contaminated soils. The process transforms easily leached, contaminated soils into a durable, leach-resistant, vitreous and crystalline monolith. This paper presents the results of the recent highly successful ISV demonstration conducted jointly by PNL and ORNL on a tracer-level quantity of radioactive sludge in a model trench at ORNL. A retention of ^{90}Sr in the vitreous and crystalline product of greater than 99.9999% was measured with a reduction in potential environmental mobility of more than two orders of magnitude. The paper also presents the current plans for continued collaboration on a two-setting treatability test on one portion of an old seepage pit at ORNL.

INTRODUCTION

In situ vitrification (ISV) is a thermal treatment technology developed by Pacific Northwest Laboratory (PNL) for the remediation of contaminated soils. The process is based on the joule-heating principle of glass melter technology, developed at PNL for immobilizing high-level nuclear waste. Since its inception in 1980, over 150 tests and demonstrations have been conducted, ranging from bench- and engineering-scale tests in the laboratory to pilot- and full-scale tests and demonstrations in the field. The full-scale field tests have produced vitrified soil monoliths as large as 800,000 kg and 12 m in diameter, and up to 5 m in depth.

In situ vitrification can provide a single comprehensive treatment for soils contaminated with inorganic and organic hazardous materials and radionuclides. The process can accommodate a wide variety of soil types and site conditions, including sand, silt, clay, sludge, rocks, debris, combustibles, and buried scrap metal. The technology has been transferred to the Geosafe Corporation for commercial remediation of soils contaminated with hazardous wastes.

Past waste disposal operations at Oak Ridge National Laboratory (ORNL) have left several contaminated seepage sites that were used to dispose of over one million curies of radioactive liquid waste (mostly ^{137}Cs and ^{90}Sr) between 1951 and 1966 (Spalding 1987). Testing of ISV is being performed as part of an ongoing remedial technology evaluation for the ORNL pits and trenches. These ORNL sites are good candidates for treatment by ISV because of their relatively small areal extent and shallow depth. An in situ treatment technology is preferred over conventional "retrieve and treat" operations because it will greatly reduce or eliminate the potential for personnel exposure to the high radioactive dose rates at the sites.

The primary driver for remediation of the ORNL sites is the large inventory of ^{90}Sr, which is highly mobile in the subsurface environment and presents the potential for significant release in the future. It should also be noted that the ^{137}Cs is irreversibly sorbed to the natural illitic clays and is highly immobile.

PROCESS DESCRIPTION

The soil melting process employed by ISV allows for single-step remediation of multi-component waste sites. Figure 1 depicts the ISV processing sequence as applied to contaminated soils. Melting is induced by applying electrical power to the soil through an array of graphite electrodes inserted vertically into the surface of the soil over the site to be melted. Power is controlled to the melt at a constant level and the molten soil mass grows downward and outward. Temperatures between 1400°C and 2000°C are typically maintained in the molten soil pool. Power to the melt is maintained until the desired treatment depth is obtained and the soil and its contents are melted. Off-gases generated by the melt during processing are confined in a steel hood placed over the site being melted and directed to an off-gas treatment system, where they are scrubbed and filtered to remove particulate, acid gases, and other vapors before being released to the atmosphere. Upon cooling, the molten soil solidifies to form a durable vitreous and crystalline mass. For application to expansive waste sites, the process can be applied in sequential and adjacent batch settings to create one contiguous monolith.

During processing, the electrode feed system (EFS) is used to control the vertical position of the energized electrodes in the melt. The electrodes typically are allowed to feed downward by gravity, constantly resting on the bottom of the melt. However, within the melt, they can be held, retracted, or advanced as necessary. The EFS allows for a simple setup process since it eliminates the need to intrude into the waste site for electrode placement.

Testing has shown the ISV product to be highly durable and leach resistant. Extensive leach testing and comparison with natural analogs indicates that the ISV product should retain its integrity for millennia (Buelt et al. 1987; Callow et al. 1991). More recently, it was confirmed that the ISV product shows no significant changes during wet/dry and freeze/thaw cycles and that it passes EPA biotoxicity testing (Geosafe 1992). In general, the ISV waste form offers a tremendous--often orders of magnitude--improvement over

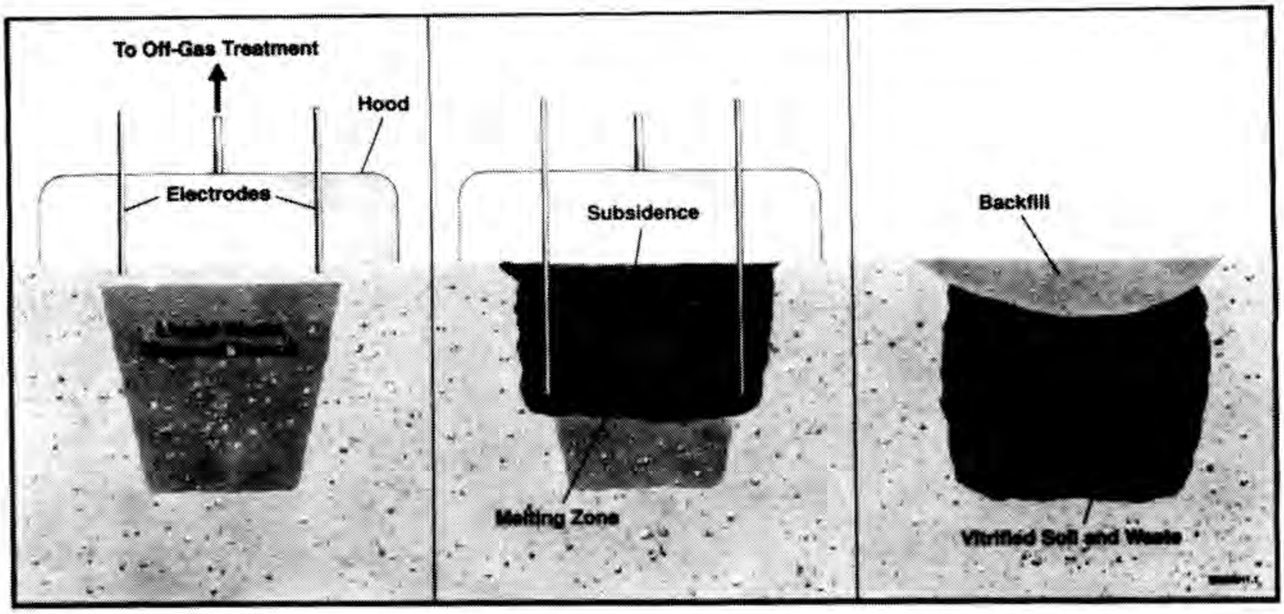

Fig. 1. The ISV processing sequence as applied to contaminated soils.

the original forms of the contaminants and greatly reduces potential impacts to human health and the environment.

PILOT-SCALE TEST RESULTS

In May 1991, a pilot-scale test was conducted on a scale-model of a radioactive liquid waste disposal trench at ORNL. The test site was staged in native soils and limestone gravel was used to simulate the trench. A tracer sample of radioactively contaminated sludge (containing 10.1 mCi of ^{137}Cs and 12.7 mCi of total activity) from an inactive seepage pit was placed into the trench for treatment. The test site was extensively instrumented during staging to collect thermal and pressure data during processing that would confirm the results of the test. Figure 2 shows a cutaway drawing of the pilot-scale ISV system used for the test.

Of specific concern in preparation for the test was the volatilization potential of the ^{137}Cs. A non-radioactive pilot-scale ISV test conducted at ORNL in 1987, using chemical simulants, had resulted in a volatilization of Cs measured at 0.12% (Spalding and Jacobs 1989). Even this relatively low amount could present a risk to personnel during actual remedial operations considering the extremely high levels of radioactivity in the pits and trenches. Therefore, a major objective of the 1991 test was to reduce the accumulation of ^{137}Cs in the off-gas treatment system. Therefore, a prefilter was specially designed to capture volatilized ^{137}Cs from the off-gas stream. Figure 3 shows the prefilter assembly placed between the off-gas hood and the off-gas treatment system.

Powered operations of the test were conducted over a 128-hour period with a total of 29 MWh of energy delivered to the melt. Approximately 12,000 kg of soil were melted and

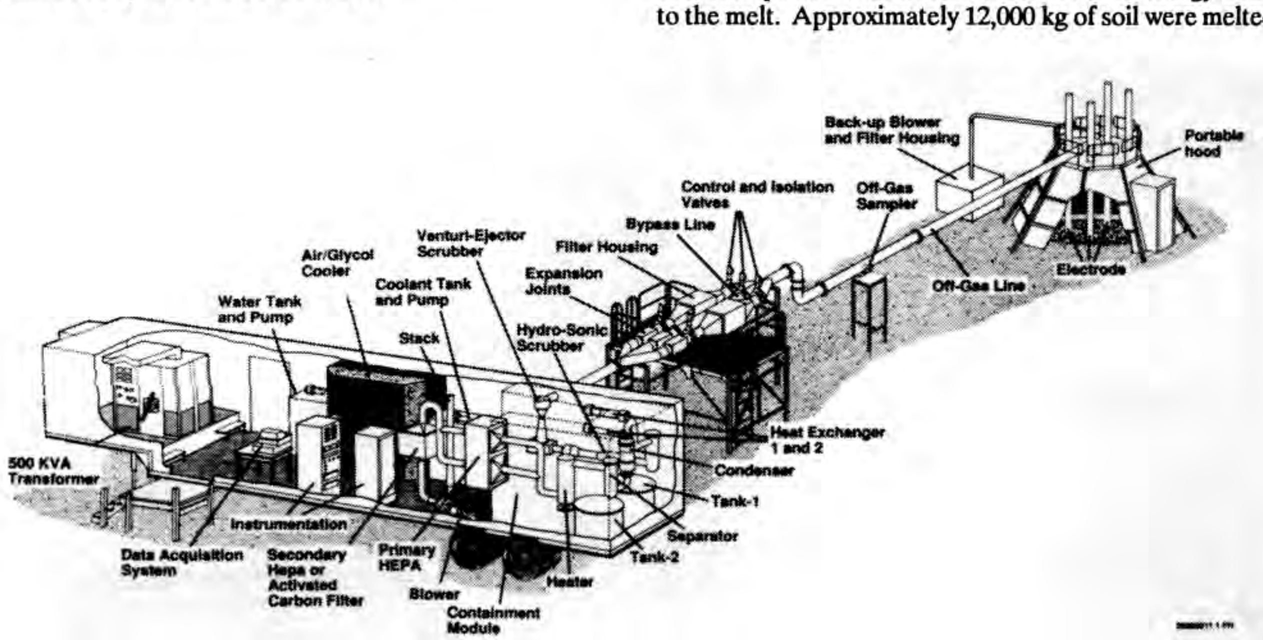

Fig. 2. Cutaway drawing of the pilot-scale ISV system used for the May 1991 ISV test at ORNL.

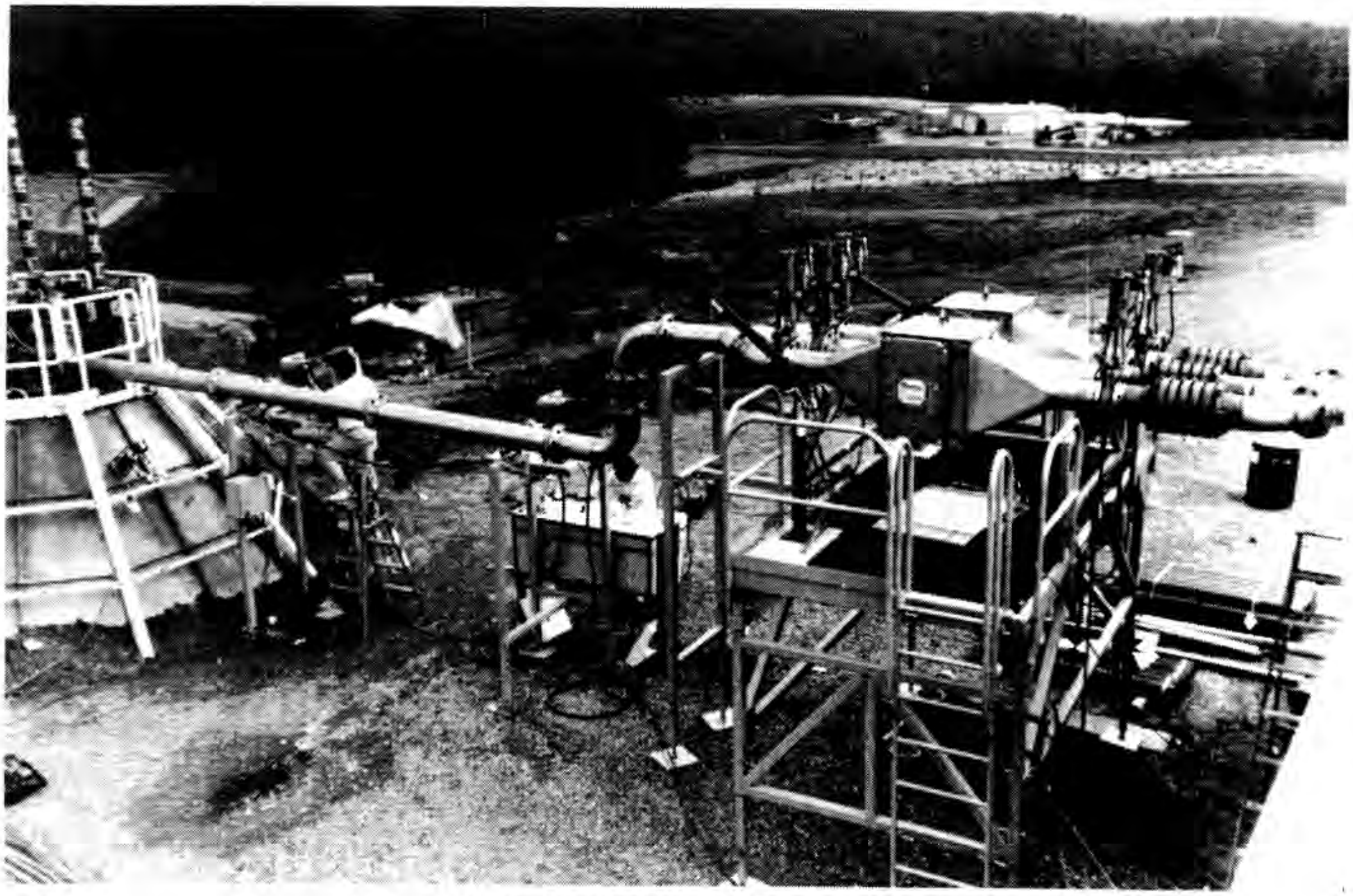

Fig. 3. The ISV off-gas prefilter assembly.

the process reached a treatment depth of 2.7 m, fully processing the trench and exceeding the target treatment depth. Extensive analysis and evaluation of test data showed that retention in the vitreous and crystalline product was >99.9999% for ^{90}Sr and 97.7% for ^{137}Cs. Furthermore, the prefilter, which used a high-temperature high-efficiency particulate air (HEPA) filter, was 100% effective in capturing volatilized ^{137}Cs since no increase in radioactivity was detected in the off-gas treatment system downstream of the prefilter.

The relative improvement in the quality of the waste form produced by the ISV treatment in terms of the ^{90}Sr and ^{137}Cs in the soil was measured using a leach test regime on several pretest and posttest samples. The leaching procedure has been fully described by Spalding and Jacobs (1989) and is designed to measure the effects of dilute salt and acid solutions on the waste. The test consists of a series of five sequential washings of each sample with a 0.1 N $CaCl_2$ solution followed by five more sequential washings with a 0.1 N HCl solution. The samples subjected to the leach test included the pretest radioactive sludge, posttest ISV product fragments (retained on 10 mesh screen), and posttest pulverized ISV product powder (passing 100 mesh screen). Each sequential extract was assayed for ^{90}Sr and ^{137}Cs (Spalding et al. 1992).

Figure 4 shows the results of the cumulative leach extraction for ^{90}Sr. The results show that about 10% of the ^{90}Sr in the waste sludge is leached by exchangeable cations in the dilute salt solution. When followed by exposure to the dilute acid, almost quantitative dissolution of the ^{90}Sr resulted. These results clearly demonstrate the poor quality of the ambient waste form in the ORNL sites. However, considerable improvement was observed in the performance of the ISV samples. Notably, less than 1% of the ^{90}Sr was ultimately leached from the fragmented samples, a decrease of two orders of magnitude. Considering the difference in surface area between the product fragments and the actual field-size ISV product, a further reduction in leachability of at least two orders of magnitude would be expected.

Figure 5 shows the results of the cumulative leach extraction for ^{137}Cs. The results confirm the existing immobility of the Cs in the native illitic soils since the sludge exhibited only about 1% total leachability. The leachability of ^{137}Cs in the ISV product was decreased to well below 0.1%. Again, considering the difference in surface area between the product fragments and the actual field-size ISV product, a further reduction in leachability of at least two orders of magnitude would be expected in the field.

PLANS FOR TREATABILITY TESTING AT PIT 1

Following the successful test on a simulated ORNL waste site as described above, plans are being developed for a 1995 ISV test on one of the ORNL seepage pits. This treatability study will support an Interim Record of Decision (IROD) for closure of one or more of the seven seepage pits and trenches in ORNL Waste Area Grouping 7 (WAG 7) in 1996. The test will be conducted on a portion of Pit 1, which was constructed in 1951 as a 6-m-wide by 30-m-long trench with a nominal depth of 4.5 m. The pit was intended to be a

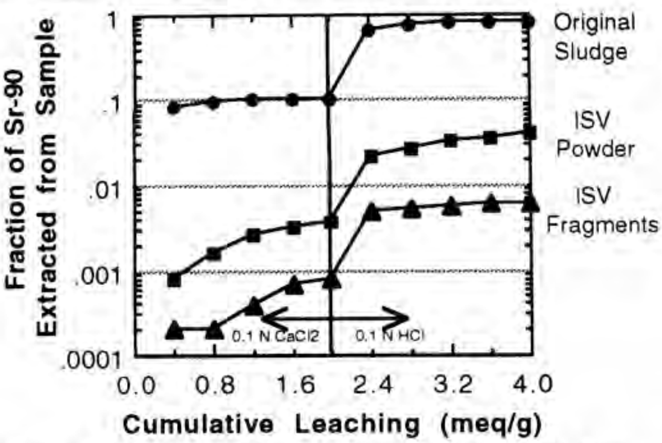

Fig. 4. Results of sequential leach extraction for ^{90}Sr from ORNL sludge and the ISV product.

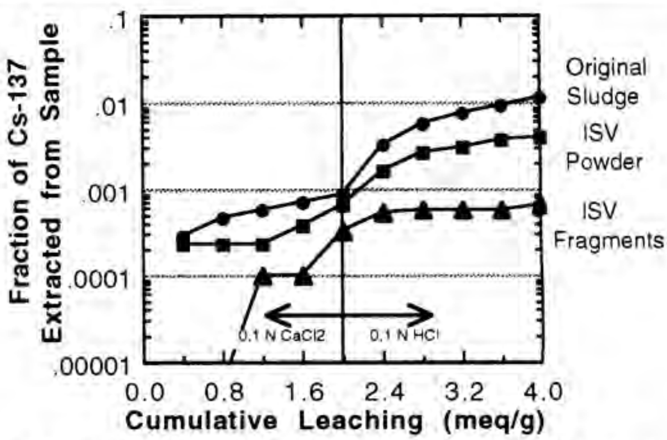

Fig. 5. Results of sequential leach extraction for ^{137}Cs from ORNL sludge and the ISV product.

proof-of-principle operation, but its use for disposal of liquid wastes continued long after the 3-month test period. It is estimated to have received less than 400 Ci of activity during its 30 years of operation (compared to the hundreds of thousands of curies deposited in each of the other six pits and trenches). While in operation, Pit 1 was spanned by a roof to prevent infiltration by precipitation. In 1981, the pit was filled with soil and capped with asphaltic concrete. Known conditions of Pit 1 and environs have been described by Carrier, Williams, and Roberts (1989).

Significant site characterization information will be collected before the implementation of ISV at Pit 1 or any of the other pits and trenches in WAG 7. The inventory and spacial distribution of radioactivity will need to be confirmed along with the actual depth and lateral extent of the pit. Likewise, depth to groundwater and/or perched water within and around the pit will need to be determined. These data can be collected via driven well points, hole logging, and penetration resistance probes to reduce radiation exposures to personnel. Once radiation levels have been determined, samples of backfill will be obtained. The samples will be used to measure permeability and melting characteristics of the soil. The site characterization will allow more confident assessments of the radiation hazards and ISV processing parameters.

The treatability study will address several key development needs of the ISV technology. First, there is the need to demonstrate that ISV can attain the required 4.5-m depth. Although greater depths have been demonstrated at other sites, the geologic, hydrologic, and geometric conditions of the ORNL seepage pits and trenches need to be confirmed.

Second, a technique for handling potentially as much as 5 Ci of ^{137}Cs that may volatilize during the ISV treatment needs to be demonstrated. The 1991 test showed that the volatilized ^{137}Cs could effectively be captured using a HEPA prefilter technique. An improved design for the prefilter will need to be established so that the loaded filters can be processed by ISV in subsequent melts, minimizing generation of secondary wastes and reducing exposure levels to personnel. Studies will continue to determine the controlling mechanism(s) of ^{137}Cs volatilization. Further understanding will contribute to the capability for reducing or controlling the amount of ^{137}Cs volatilization.

Third, the ability to move a contaminated hood and establish a new overlapping melt in a contaminated environment needs to be established. Due to their areal extent, all of the seepage pits and trenches will require multiple melt settings to complete vitrification of all source material. Multiple settings with a radioactively contaminated hood have not previously been demonstrated.

Like the previous field tests at ORNL, the Pit 1 treatability test will be a collaborative effort between PNL and ORNL. It is also anticipated that Geosafe Corporation will have a significant role in the operations of the test.

CONCLUSION

Results of recent ISV tests on actual wastes, specifically the 1991 tracer-level radioactive test at ORNL, have demonstrated the potential of ISV to provide a cheaper, faster, better, and safer remedial treatment option for contaminated soils compared to existing alternatives. It is expected that the development needs that must be addressed before applying ISV to the ORNL pits and trenches will be met through the activities associated with the treatability study on Pit 1. It is also expected that sufficient site characterization can be performed to accurately predict processing kinetics, processing temperatures, ^{137}Cs volatilization rates, and product quality. Finally, it is anticipated that by coordinating the treatability study within the regulatory closure process, and by demonstrating its safety, implementability, and product quality, regulatory and public acceptance of the use of ISV technology on the ORNL pits and trenches will be promoted.

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