IN SITU VITRIFICATION OF BURIED WASTE*

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ABSTRACT

This report describes the application of the in situ vitrification (ISV) process to buried mixed and transuranic wastes. The results of product evaluation activities from two ISV intermediate-scale field tests conducted on simulated buried waste pits are reported. The physical properties and durability of the resulting waste form is discussed. The resulting waste form was found to be more leach resistant than high-level nuclear waste glasses and to be very comparable to stable natural materials, such as obsidian.

INTRODUCTION

In situ vitrification (ISV) is being evaluated as a remedial treatment technology for buried mixed and transuranic (TRU) wastes at the Subsurface Disposal Area (SDA) at the Idaho National Engineering Laboratory (INEL) and can be related to buried wastes at other Department of Energy (DOE) sites. There are numerous locations around the DOE Complex where wastes were buried in the ground or stored for future burial. Waste volumes were reported in 1988 as follows (1):

Low-level wastes	2,472,000 m^3
Transuranic (contact)	58,196 m^3
Transuranic (remote)	1,485 m^3
Mixedlow-level	78,664 m^3
Total	2,600,000 m^3 (92 million ft^3)

The Buried Waste Program (BWP) is conducting a Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) remedial investigation/feasibility study (RI/FS) for the Department of Energy Idaho Field Office (DOE-ID). As part of the RI/FS, an ISV scoping study on the treatability of the SDA mixed low-level and mixed TRU waste is being performed for applicability to remediation of the waste at the Radioactive Waste Management Complex (RWMC). The ISV project being conducted at the INEL by EG&G Idaho, Inc., consists of a treatability investigation to collect data to satisfy nine CERCLA criteria with regards to the SDA. This treatability investigation involves a series of experiments and related efforts to study the feasibility of ISV for remediation of mixed and TRU waste disposed of at the SDA. TRU waste typically includes cloth, paper, plastic, metals, rubber, sludge, and concrete contaminated with TRU radionuclides. TRU waste is contaminated with primarily alpha-emitting radionuclides with an atomic number greater than 92, a half-life longer than 20 years, and a concentration greater than 100 nCi/g. Wastes were deposited into the burial pits and trenches in containers. Approximately 60% of the containers were steel drums (30 to 55 gal), 5% were plywood boxes, and 30% were cardboard and fiberboard containers. Hazardous materials include organics, inorganic, and metals associated with the TRU wastes.

PROCESS DESCRIPTION

ISV is an emerging, in-ground thermal treatment technology that fixes hazardous and radioactive contamination in the soil by converting it to a glass and crystalline waste form (2). The process involves the insertion of electrodes into the soil, typically in a square array (Fig. 1). A starter path for electrical current is established using a small amount of a graphite and glass frit mixture placed between the electrodes on the soil surface. Dissipation of power through the starter material creates temperatures high enough to melt a layer of soil, thus establishing a molten, conductive path. Current passing through this conductive path maintains the melt temperature by joule-heating. The joule-heated principle operates by internal resistance heating of the conductive material as an electrical current passes through the molten media. The molten zone continues to grow downward and encompasses the contaminated soil. The process continues until heat losses from the melt approach the energy delivered to the molten soil via the electrodes or until power is discontinued to the melt.

At the high temperatures created (approximately 1300 to 1800°C), organic materials pyrolyze, diffuse to the surface, and combust. Off-gases from the process are collected, treated, and monitored to ensure an environmentally safe release. Remaining ash and other noncombustible materials dissolve or become encapsulated in the molten soil. Natural convective currents within the molten soil help to uniformly distribute the stabilized materials. Molten soil cools to a durable glass and crystalline waste form, physically resembling natural obsidian or volcanic rocks.

Because the primary purpose of the ISV process is to stabilize and immobilize nuclear and toxic waste components, the chemical morphology and release characteristics of ISV waste forms must be known to provide for an accurate performance assessment. Data generated from durability testing will also be required as input to heath-based risk assessment models as part of the technical and CERCLA evaluation of the ISV process and waste form.

The properties of ISV waste forms are directly related to the composition of the waste, composition of surrounding soil, and the thermal history of materials reacted during vitrification and cooling. The application of the ISV process to buried

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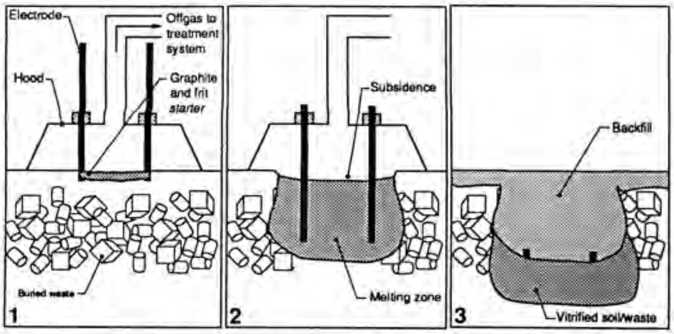

Fig. 1. The in situ vitrification process.

waste and soil presents unique conditions compared to the homogeneous soil/waste conditions previously tested.

PHYSICAL PROPERTIES

Samples of waste forms from laboratory and field tests (3,4) on buried waste have been evaluated. Laboratory tests have been conducted vitrifying soils containing a mixture of buried hazardous heavy metals (Ag, As, Ba, Cd, Cr, Hg, Pb, and Se) with stainless and carbon steels, combustibles, and organics. Actual site soil from the INEL was used and a basalt block was placed at the bottom of the soil to simulate the basalt layers below the SDA. The Intermediate Field Tests (IFT) 1 and 2 were conducted using simulated wastes with no hazardous or radioactive materials. The IFT 1 and 2 tests were designed to simulate a waste region containing randomly and stacked disposed drums and boxes intermixed with fill dirt with high metal contents.

In general, the product from these tests consisted of a black (with green tints), glassy material containing variable amounts of bubbles and crystalline material. Glass was the principal phase found within the monoliths. The outermost portion of the monolith, the most quickly cooled portion, was glassy with little devitrification (crystals). The IFT test monoliths consisted of an outermost zone of black glass about 5.1 cm (2 in.) thick followed by a white-to-beige-to-lavender zone with a very fine crystal structure (aphanitic) region 5.1 to 10.2 cm (2 to 4 in.) thick that graded into a coarser crystal structure (phaneritic) material. During cooling, devitrification occurred within the glass monolith producing a feather-like crystalline phase called augite (Fig 2). The mineral augite, a variety of clinopyroxene, is a calcium-magnesium-iron rich silicate. Augite is a common, naturally occurring pyroxene found in volcanic rocks, such as the basaltic rocks found at the INEL. Solids characterization of the ISV melt products showed that the ISV melts are reducing, resulting in melt $Fe^{+2}/\Sigma Fe$ ratios of $>90\%$.

Metallic materials were also found in both pits and were observed in three forms. One form was unmelted metal consisting of massive pieces of scrap metal concentrated into the lower one-quarter of the monoliths. Some spherical particles or nodular lumps of metal were observed. This morphology indicates that these materials have undergone some transformation, most likely melting, during the ISV processing. The third class of metallic materials included the microscopic ($<10\mu m$) spheres of opaque material, mostly metals, found in all samples that were examined in detail. Usually the amount of opaque material was one volume percent or less.

Tracers that were added to the tests to similar TRU contaminants indicate that the bulk elemental composition of ISV melts is relatively homogenous. These tracers were added to the field tests in single canisters rather than small amounts in each canister. This indicates that the ISV process will produce a relative homogeneous waste form.

Fig. 2. IFT 2 texture of vitrified monolith material.

DURABILITY

A series of tests were performed to determine the dissolution behavior of samples produced from the ISV processing of typical soils from the INEL Subsurface Disposal Area. Durability testing is divided into three types of leaching tests. The first, the Environment Protection Agency's toxicity characterization leaching procedure (TCLP), is the minimum regulatory testing requirements established for landfill disposal. However, the TCLP does not address radioactive waste components, provide a technical basis for assessing long-term durability, provide a basis for a comparison to highly durable waste forms or natural analogs, or provide an assessment of the source term release of the waste component for risk assessment models. To provide this information, additional durability tests must be conducted. These additional tests fall into two categories: comparative testing (comparing ISV waste forms to similar waste forms and natural analogs) and testing to determine the intrinsic rate (fastest) of waste form dissolution $(k+)$.

The ISV waste forms do not exhibit hazardous characteristics of TCLP toxicity. However, it is possible that molten metal at the bottom of the monolith may be TCLP toxic if excessive lead is present in the waste.

To allow for direct comparison of waste forms, the test method, leachate, temperature, and surface area/volume (S/V) used in testing the ISV waste form should be the same as those used in testing glass waste forms chosen for high-level nuclear waste. The differences in application (repository conditions such as groundwater saturation and temperature) and the nature of the waste form (multiphase ceramic-glass versus single-phase glass) make such comparisons difficult. Most high-level nuclear waste glasses have been tested at 90°C. The Materials Characterization Center (MCC) MCC-1 and MCC-3 test methods were originally developed, and the current procedures written, for application to high-level waste repositories in deep geological environments. The primary applications of the MCC tests will be to compare ISV waste forms and will have limited utility for mass transport analysis. By conducting the MCC-1 and MCC-3 leach testing at 90°C, the results can be compared to the existing large data base of leaching data on the high-level glasses.

A summary of MCC-1 leach testing results is presented in Fig. 3. Based on MCC-1 leach testing data, the durability of the IFT waste form is comparable to obsidian and granite, and 4 to 10 times more durable (based on MCC-1 testing) than typical high-level nuclear waste glasses.

The fastest rate at which a glass/ceramic will dissolve is the intrinsic rate of dissolution (k+). This glass parameter has the most technical relevance when evaluating and predicting the dissolution (durability) behavior of the glass. The intrinsic rate will be used as the source term in health-based risk assessment models to determine the risk associated with leaving the waste form in place. This will be a conservative value for the source term as saturated or partially saturated groundwater will slow the dissolution of the waste form significantly.

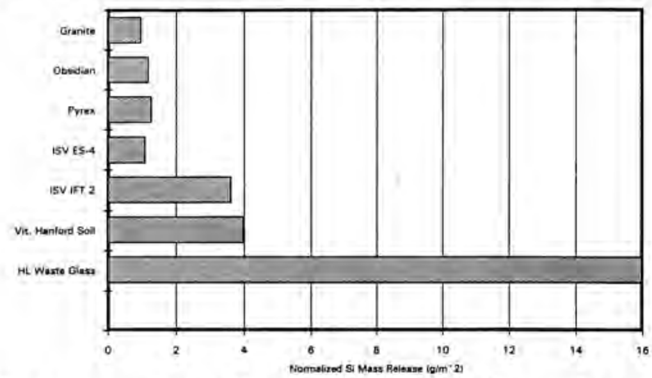

Fig. 3. Comparison of leach resistance of ISV waste forms to selected materials (MCC-1, 28 day, 90°C.

To understand the intrinsic rate, it is beneficial to discuss the three different regimes typically observed in glass dissolution: (a) the period in which the glass first contacts a leachate and the glass dissolution rate is uninhibited by any solubility effects, (b) a transient regime where the increasing concentrations of dissolved glass components in the leachate slows the dissolution rate through solubility effects, and (c) a steady-state regime in which the dissolution rate is constant because alteration processes (saturation) have reached a steady-state. At early times, the glass matrix dissolves with the intrinsic dissolution rate (k+) because there is nothing in solution or on the surface of the glass to impede the dissolution process.

Preliminary results from intrinsic rate constant measurements showed that the dissolution rates of the ISV samples range from 0.01 to 0.06 g/(m^2 d) at 90°C and pH 7. These values are 10 to 100 times smaller than measured for a typical borosilicate nuclear waste glass as shown in Fig. 4.

Additional work still needs to be conducted on technical issues associated with ISV processing on buried waste. These include pressurization of the hood due to sealed containers being processed, rapid vapor release from within or under the melt, waste composition variations, waste component transport due to thermal gradients, and depth limitations, for example. These technical issues are being address at the INEL and coordinated with other laboratories conducting ISV testing (5).

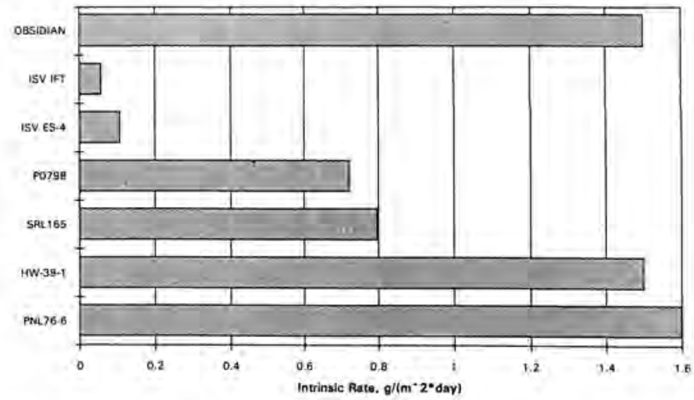

Fig. 4. Comparison of intrinsic dissolution rates of ISV waste forms to selected mateials (90°C).

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