CONF-950216--35

WSRC-MS-95-0056

Development of Plasma Vitrification Technology for Contaminated Soil at the Savannah River Site

by

A. L. Kielpinski
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

J. C. Marra
WSRC
SC USA

J. Etheridge
Diagnostic Instrumentation and Analysis Laboratory
MS USA

R. Kirkland
Diagnostic Instrumentation and Analysis Laboratory
MS USA

R. F. Schumacher
WSRC
SC USA

V. Rogers
WSRC
SC USA

MASTER

A document prepared for WASTE MANAGEMENT '95 at Tucson from 02/26/95 - 03/02/95.

DOE Contract No. DE-AC09-89SR18035

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831; prices available from (615) 576-8401.

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161.

DEVELOPMENT OF PLASMA VITRIFICATION TECHNOLOGY FOR CONTAMINATED SOIL AT THE SAVANNAH RIVER SITE

Andrea L. Kielpinski[1], J. C. Marra[1], J. Etheridge[2], R. Kirkland[2], V. Rogers[1], and R. F. Schumacher[1]

[1] Westinghouse Savannah River Company
[2] Diagnostic Instrumentation and Analysis Laboratory, Mississippi State University

ABSTRACT

The Mixed Waste Integrated Program (MWIP) of the United States Department of Energy's Office of Technology Development is developing treatment technologies for a wide variety of materials containing mixed low-level waste, i.e., having low levels of radioactivity along with hazardous constituents. Vitrification is a promising treatment technology for many of these wastes, including contaminated soil such as that found at the Savannah River Site.

Proof-of-principle tests were performed to demonstrate the feasibility of both ex-situ and in-situ vitrification of contaminated soil by means of a plasma torch. A mixture of 89% as-excavated Savannah River Site sandy clay loam with 11% lime addition was tested. Vitrification of a mixture of this feed, in a 10" diameter crucible with a non-transferred arc plasma torch at a nominal 160 kW, was successful. The process produced homogeneous glass (albeit with local compositional variations), surrounded by a skull of incompletely reacted feed. Characterization of the resultant product durability using the Product Consistency Test [1] showed elemental leaching well below the Environmental Assessment glass (which is often used as a minimum standard of glass acceptability in high-level waste glass assessment) for both the glass and the skull regions. Future tests should include doping the soil with hazardous constituents to enable further verification of the wasteform integrity via the Toxic Characteristic Leaching Procedure [2].

In-situ operation was mimicked in the test crucible by segregating the lime additive from the soil within the crucible. Making full use of the available torch maneuvering capabilities (which would likely exceed those of a torch used in-situ) failed to produce a homogeneous melt. Therefore, intimate mechanical mixture of the additive with the soil appears crucial to the success of SRS soil vitrification, and must be included in design considerations for in-situ operation.

INTRODUCTION

The Mixed Waste Integrated Program (MWIP) of the United States Department of Energy's Office of Technology Development is funding efforts to develop treatment technologies for a wide variety of materials containing mixed low-level waste (MLLW), i.e., having low levels of radioactivity along with hazardous constituents. Vitrification is a promising treatment technology for many of these wastes, including contaminated soil. Soils with MLLW constituents are present at many sites across the Department of Energy (DOE) complex, as well as in industrial settings such as the naturally occurring radioactive material (NORM) associated with drilling operations in the oil and gas industries. High temperatures are required for soil vitrification, making plasma technology attractive for this application.

The processing requirements of one soil type may not be directly transferrable to another, due to differing morphology and chemical composition. Rather than testing a "generic" soil which might ultimately prove to have limited applicability, a representative contaminated soil material was desired for proof-of-principle testing of the plasma vitrification method. Savannah River Site (SRS) soil was used for these tests because SRS is a large ($\approx$300 square miles) DOE site where nuclear materials production and processing operations have been carried out over several decades, resulting in significant quantities of soil requiring remediation.

This paper describes the plasma vitrification tests of SRS soil which were performed at the Diagnostic Instrumentation and Analysis Laboratory (DIAL) of Mississippi State University. Both ex-situ and in-situ treatment methods are of interest; therefore, the tests were designed to demonstrate features of each.

TEST MATERIAL

At SRS, the soil most likely to be contaminated with low levels of radioactivity is that found near processing facilities. For building stability, these facilities were generally sited on a particular group of soil series. Soil excavation for construction of facilities and burial of waste resulted in mixing of diagnostic horizons to the extent that individual soil series cannot be identified. The resulting mix of material is classified, using United States Department of Agriculture (USDA) soil taxonomy, in the Great Group Udorthents. The particle size distribution for this group typically is sandy clay loam (USDA nomenclature), with an estimated breakdown of 55% quartz sand, 30% kaolinite clay particles, and 15% silt. Thus, despite the inevitable local variability in soil composition throughout the site, there is a high probability that most of the contaminated soil at SRS is quite similar in particle size distribution and mineralogy. Although a sandier soil is generally found as topsoil throughout the site, this sandy top layer would have been removed during facility construction, exposing the sandy clay loam. For the tests described herein, clean (nonradioactive) sandy clay loam was excavated from the soil pile associated with the construction of a non-radioactive site facility.

Proof-of-principle tests of sandy clay soil vitrification have been performed previously [3]. From those tests, it had been determined that use of a fluxing additive was required for homogeneous vitrification, and that commercial, agricultural limestone was suitable for this purpose. Two tests had been run: one with no additive and one with 30% by volume of lime additive. Although lack of a flux additive precluded complete vitrification of the soil, too much flux was used in the second test, which resulted in precipitation of multiple crystalline phases upon cooling, again impairing product homogeneity.

Based on these findings, the feed composition for the present tests was approximately 11 weight percent agricultural dolomitic lime, (a mixture of $CaCO_3$ and $MgCO_3$, with 20% elemental Ca and 10% elemental Mg), and 89% as-received soil (i.e., with a moisture content of approximately 15% and containing small amounts of plant material, gravel, etc.).

TEST DESCRIPTION

The objective of the tests was to examine the potential for both ex-situ and in-situ treatment of contaminated soil. Processing issues for ex-situ vitrification, in which feed material is introduced into a processing vessel, include feed composition (ratio of soil to additive), feed material handling, and off-gas emissions. Ex-situ vitrification allows the use of either the transferred-arc or nontransferred-arc mode of torch operation. Transferred-arc operation may be more efficient; since heat is conducted through the melt directly, it is more likely to promote axial mixing and avoid temperature stratification.

In-situ treatment involves inserting the plasma torch into a series of boreholes at the contaminated site, withdrawing the torch as the surrounding material at a given axial location is vitrified. Any required additives would be introduced down the borehole. In-situ treatment is attractive, in that handling and transportation of contaminated material is minimized; off-gas treatment requirements may also be reduced if the soil acts as a percolation filter for off-gas. However, the feasibility of in-situ vitrification depends on the ability to achieve product quality under in-situ processing conditions. Since a fluxing agent appears to be necessary for this type of soil to vitrify, adequate mixing of the flux with the soil must be ensured. In-situ vitrification precludes the use of transferred-arc operation, since there would be no bottom attachment point for the arc when moving the melt upwards in the borehole. The increased gas flow through the torch in non-transferred arc mode might, however, increase mixing through forced convection by the gas impinging on the melt.

With these considerations in mind, two different types of tests were performed on the SRS soil:

1. The soil and lime were intimately mixed in the crucible. The torch was fixed both radially and azimuthally.

2. The soil and lime were segregated in the crucible. The torch was manually manipulated laterally as well as axially.

The torch was run in non-transferred arc mode for all tests to permit a direct comparison of the two test types. The first test type, essentially an ex-situ demonstration, will be rerun in transferred arc mode at a later date. The second type of test was meant to mimic the in-situ processing conditions, where the flux would be introduced into the soil without intimate mixing. The second test type was repeated twice, once with only limited lateral movement, and a second time with lateral movement specifically aimed at promoting mixing between the flux and the soil.

In all tests, the torch was first run several inches above the melt for 10-15 minutes to allow the test material to soften, thus reducing blow-off, and then lowered to a distance of approximately two inches above the surface of the test material. The duration of each test was 45-60 minutes of torch operation.

The tests were performed using a nominal 250 kW plasma torch manufactured by Plasma Energy Corporation. The torch was run in non-transferred mode, using air as the working gas. In this mode, the power output fluctuated between about 130-160 kW. The airflow through the torch varied from 12-20 scfm for these tests. The torch is installed with both axial and lateral movement capabilities. A viewport equipped with video camera enabled visual observation during the test. As described below, this capability was used in one of the tests to move the torch in order to best achieve a mixed melt.

For each test, a total of 13.44 lb of feed was used (12 lb soil and 1.44 lb lime). The test material was processed in batch mode in a 10" diameter crucible which was placed inside a larger test vessel lined with refractory brick. The test vessel was sealed and under vacuum for the tests. Off-gas was monitored by a variety of techniques, including infrared, FTIR, two-color imaging, and spectroscopic pyrometry. These measurements will be reported separately.

RESULTS

Homogeneous Feed Test

The first test, with soil and lime homogeneously mixed in the crucible, produced a homogeneous, glassy material in the center of the crucible, with a 1-2 inch "skull" of incompletely reacted feed adjacent to the crucible walls and bottom. Presumably, a more powerful plasma torch, or use of the torch's lateral maneuvering capabilities, would have completely vitrified the material. To assess product quality as a function of radial distance from the torch, samples of the product were taken at three locations: (1) from the center of the melt; (2) from the transition region between glass and skull; and (3) from the skull region itself. These were examined by X-ray diffraction (XRD) and scanning electron microscopy coupled with energy dispersive X-ray spectroscopy (SEM-EDS) (Figure 1).

PLACE FIGURE 1 HERE

XRD of samples 1 and 2 (from center and transition regions, respectively) confirmed that homogenous glass was produced, with no crystalline phases observed. XRD of sample 3 (skull region) showed the presence of both quartz (the sand in the original soil) and cristobalite, with quartz predominating. Note that the transition temperature between these two phases is 1470°C, which implies that temperatures in this range were experienced even in the incompletely reacted region.

SEM results of the three samples are shown in Figure 1. All figures are at the same scale, as shown in the figures. SEM-EDS examination of sample 1 showed the presence of a number of species: Si, Fe, Ni, Cu, and Ca. The same species were identified in sample 3, with slightly higher peaks for Ca, Fe, and Ni showing some compositional variation between the glassy center and the incompletely reacted soil/lime mix in the skull region. However, SEM-EDS examination of an area in sample 2 showed the presence only of Si--none of the other species were identified except for a very minor peak for Cu. This SEM-EDS result was obtained at two locations within sample 2 which were about 1 mm apart. This implies that this area of the transition region between glass and unreacted batch is essentially pure silica glass.

The Product Consistency Test (PCT) [1], now ASTM C-1285 procedure for determining waste glass durability, was performed for the three samples, yielding leachate concentrations as shown in Table 1. Note that non-zero concentrations are measured for various cations in sample 2. This implies that constituents other than silica were indeed present in this sample, contrary to the SEM-EDS indications. Therefore, we assume that sample 2's average chemical composition is similar to that of the other samples; the SEM-EDS result can be attributed to the small scale of the SEM-EDS sample. This shows, however, that although a uniform glass product was formed in the transition region, local (microscale) variations in chemical composition resulted.

TABLE 1. PCT Results for Homogeneous Feed Test Product Samples

Elemental Release, ppm	Sample 1 (Center)	Sample 2 (Transition)	Sample 3 (Skull)	EA Glass [3]
Si	20.28	20.56	20.67	893
Na	0.08	0.10	0.15	1662
Li	0.02	0.05	0.02	190
B	0.10	0.07	0.06	587
Al	3.29	3.24	2.64	
Mg	1.46	1.46	ND	
Ca	2.26	2.29	14.73	

Table 1 shows that all three areas had releases of Si, Na, Li, and B lower than the Environmental Assessment (EA) glass which has been used as a minimum standard for durability for high level waste glass. The skull region had a significantly higher level of Ca release, however, indicating insufficient reaction between the soil and lime. The skull region also showed a slightly lower Al release than the glassy regions.

It is notable that all three samples had roughly similar release levels. That is, even the crystalline skull region material has a durability better than EA glass, as measured by the PCT. (Note that these PCT results are on a non-normalized basis, so this conclusion must be confirmed by chemically analyzing the samples and normalizing the PCT results.) The feed was not doped with any hazardous constituents, so its ability to retain such constituents could not be assessed by a direct measure such as the Toxic Characteristic Leaching Procedure [2]. This should be done to verify that both the glass and the incompletely reacted material represent a viable waste form.

Segregated Feed Tests

The main focus of these tests was to explore the ability to mix the lime and soil in-situ. From visual observation during the test and of the resultant product, there was clearly a lack of mixing between the flux and soil. In the first of these tests, with relatively little lateral torch movement, the lime clearly boiled, spattering the crucible sides and vessel viewports, while the soil remained stiff. Visual examination of the crucible contents showed extreme heterogeneity in the product, with "ledges" of unreacted soil lining the sides of that portion of the crucible which contained soil (i.e., about 270° of the circumference). Even in the center of the crucible, directly below the torch, the melt extended only a short depth below the surface, and at the sides of the crucible, the glassy portion had a thickness of only about 0.25 inches.

The test was then repeated, making full use of the visual capabilites and torch movement capabilities to try to drive the lime melt into the soil and soften the soil "ledges" in order to mix them into the melt. Although some mixing was achieved by this method, visual observation during the test still showed differences between the lime and soil regions of the crucible. Post-test examination of the material confirmed that soil "ledges" were still present around the sides of the crucible, although the product appeared more homogeneous than in the previous test.

Quantitative measurements such as those performed on the homogeneous feed will be performed on the products of these tests, but are not expected to change the following conclusions based on visual observation. The first of these two tests represents the simplest configuration for in-situ operation, that is, with additive introduced in a mass down the borehole. While some torch maneuverability may be available, it will likely be limited and the torch movement will not be "tailored" to optimize the mixing operation. The second test represents an "upper bound" to the optimization of torch operation to promote mixing. Even under these conditions, a homogeneous melt could not be produced. These tests indicate that in-situ vitrification of tyis soil type would require apparatus to mix in the flux prior to treatment with th plasma torch, which will require additional design effort and feasibility testing.

CONCLUSIONS

Proof-of-principle tests were performed to demonstrate the feasibility of both ex-situ and in-situ vitrification of contaminated soil by means of a plasma torch. To represent the SRS contaminated soils, a sandy clay loam was used; a mixture of 89% as-excavated soil with 11% lime addition was tested. Crucible vitrification of this mixture was successful, producing glass (albeit with local compositional variations) without crystallization.

In-situ operation was mimicked in the test crucible by segregating the lime additive from the soil within the crucible. Making full use of the available torch maneuvering capabilities (which would likely exceed those of a torch used in-situ) failed to produce a homogeneous melt. Therefore, intimate mechanical mixture of the additive with the soil appears crucial to the success of soil vitrification, and must be included in design considerations for in-situ operation.

ACKNOWLEDGEMENTS

John Andol and the operating crew of DIAL operated the equipment for these tests. C. A. Cicero and M. K. Andrews of WSRC performed the PCT on the samples. This work was funded by the Mixed Waste Integrated Program of the Office of Technology Development, U. S. Department of Energy.

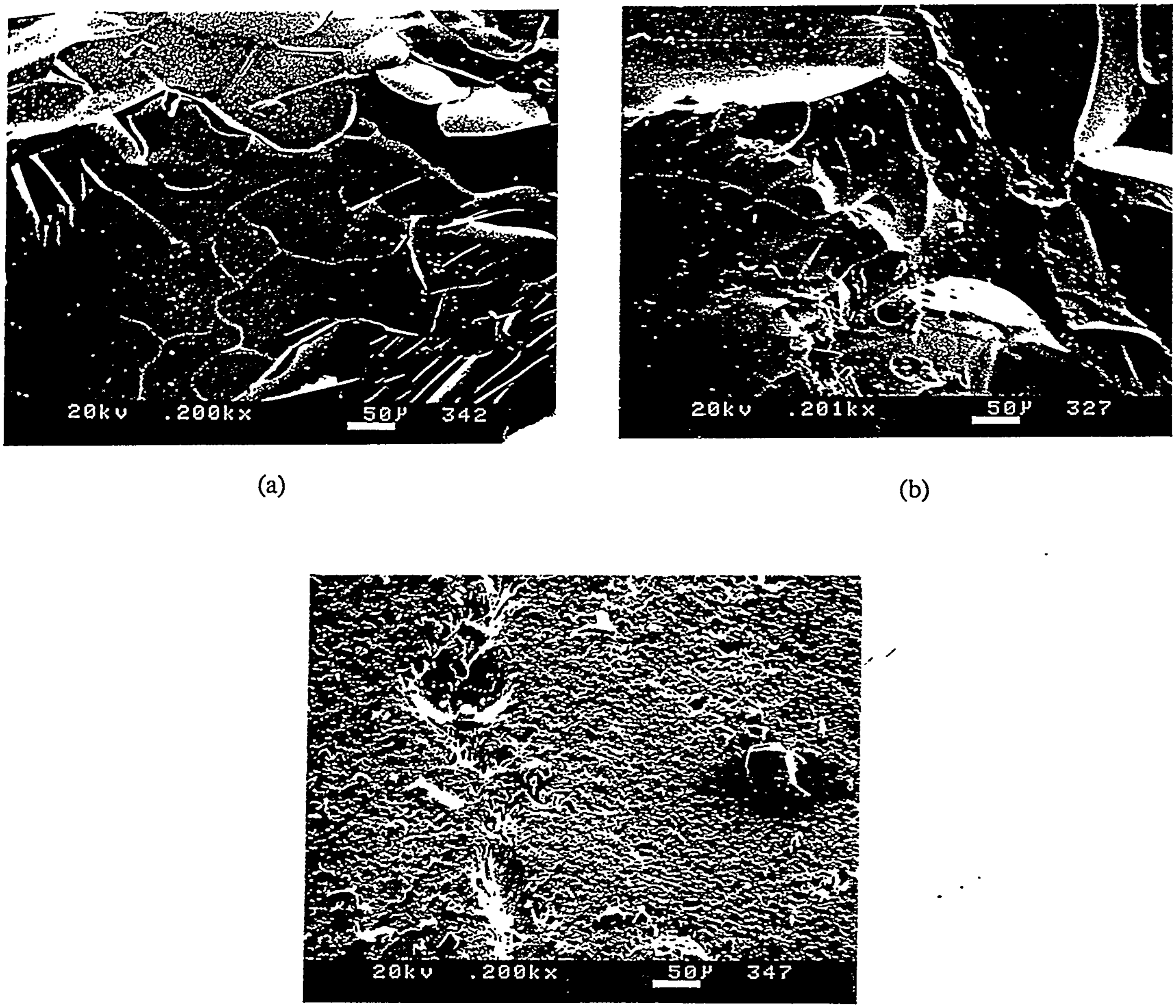
REFERENCES

[1] Jantzen, C. M., Bibler, N. E., Beam, D. C., and Ramsey, W. G., "Nuclear Waste Glass Product Consistency Test (PCT)--Version 7.0" (U), WSRC-TR-90-539, Revision 3, 1994.

[2] Federal Register Rules and Regulations, "Toxic Characteristic Leaching Procedure," Vol. 55, 11862, March, 1990.

[3] Schumacher, R. F., and Applewhite-Ramsey, A., to be published.

[4] Jantzen, C. M., Bibler, N. E., Beam, D. C., Crawford, C. L., and Pickett, M. A., "Characterization of the Defense Waste Processing Facility (DWPF) Environmental Assessment (EA) Glass Standard Reference Material" (U), WSRC-TR-92-346, Rev. 1, June, 1993.



(a)

(b)

(c)

Figure 1. SEM photographs of product from mixed-feed test: (a) center of melt, (b) transition region, (c) skull region.