Subsurface Planar Vitrification Treatment of Problematic TRU Wastes: Status of a Technology Demonstration Program

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ABSTRACT

This paper provides a status of the In Situ Transuranic Waste Delineation and Removal Project in which the GeoMelt® Subsurface Planar Vitrification™ (SPV™) process is being evaluated for the in situ treatment of burial sites containing remote handled mixed transuranic (TRU) waste. The GeoMelt® SPV™ process was invented and patented by Geosafe Corporation. AMEC holds the exclusive worldwide license to use this technology. The current project is part of a three-phase demonstration program to evaluate the effectiveness of the GeoMelt® SPV™ process to treat waste contained in vertical pipe units (VPUs) and caissons that were used for the disposal of remote handled transuranic wastes located at Hanford's 618-10 and 618-11 burial grounds. This project is being performed for the US Department of Energy (DOE) for use at the Hanford site and other DOE installations.

The Phase I evaluation determined that removal and treatment of the 618-10/11 VPUs are beyond what can be safely accomplished using conventional excavation methods. Accordingly, a careful stepwise non-intrusive delineation approach and treatment using the GeoMelt® SPV™ technology, followed by removal, characterization, and disposal of the resulting inert vitrified mass was identified as the preferred alternative. Phase II of the project, which started in July 2004, included a full-scale non-radioactive demonstration of AMEC's GeoMelt® SPV™ process on a mock VPU configured to match the actual VPUs. The non-radioactive demonstration (completed in May 2005) was performed to confirm the approach and design before proceeding to a radioactive ("hot") demonstration on an actual VPU. This demonstration took approximately 130 hours, processed the entire mock VPU, and resulted in a vitrified monolith weighing an estimated 90 tonnes.[1] Plans for a radioactive demonstration on an actual VPU are being developed for CY 2006.

In addition to demonstrating GeoMelt® SPV™, delineation techniques are being evaluated as part of the project to confirm the locations of the actual VPUs and to progressively determine their physical and chemical contents. The initial calibration and testing activities were completed in December 2005. The techniques included non-intrusive geophysical measurements from adjacent boreholes (ground penetrating radar, neutron-gamma radiography, etc.). Other methods available for use, on an as needed basis, include gas headspace sampling and boroscope examinations inside the VPUs/caissons.

INTRODUCTION

The GeoMelt® vitrification process is an electric melting process used to treat hazardous and radioactive wastes through Joule heating. The treatment results in the destruction, removal or permanent immobilization of contaminants. This process has been used successfully to treat a wide range of organic, heavy metal and radioactive contaminants. The GeoMelt® vitrification technologies can be configured in various ways to meet a wide range of site remediation and waste treatment requirements. The

configurations range from large in situ melts to above-ground melts in refractory-lined containers. The melt temperature typically ranges from 1300-2000ºC depending on the materials being treated and the configuration being used.

A broad range of organic contaminants has been treated successfully in commercial operations. Concentrated organic wastes up to 33-wt% in soil have been successfully treated with the process. Organic contaminants such as dioxins, pesticides and polychlorinated biphenyls (PCBs) are destroyed via pyrolysis and de-chlorination reactions at elevated temperatures and in reducing conditions around the melt. No organic contaminants remain in the melt due to the inability of organics to exist in the melt at such high temperatures. The destruction and removal efficiency (DRE) for organic contaminants achieved during commercial operations is greater than 99.9999%. This DRE includes the percentage destroyed by the melt (typically 90 to 99.9%) and the percentage destroyed and/or removed from the off-gas stream by the off-gas treatment equipment.[2]

The process can accommodate relatively high concentrations of heavy metals and radionuclides resulting in permanent immobilization in the vitrified product. Most metals and radionuclides are retained in the melt, with typical retention efficiencies (REs) of 99.99% or better for the non-volatile species. The degree of retention in the melt of semi-volatile heavy metals such as lead, cadmium and arsenic is quite high and generally around 80 to 90%. Cesium has been processed with resulting in REs of 99 to 99.99%.[3] Volatile metals such as mercury evolve from the melt and are captured by the off-gas treatment system.

The vitrified product normally consists of a mixture of glass and crystalline materials and often has an appearance similar to volcanic obsidian. The product is typically ten times stronger than concrete and is extremely leach resistant. The vitrified product readily satisfies the requirements of the US Environmental Protection Agency's (EPA) Toxicity Characteristic Leaching Procedure (TCLP).[4] The vitrified product is normally 10 to 100 times more durable and leach resistant than typical borosilicate glasses used to immobilize high-level nuclear waste. The durability and leach resistance of the glass is due to a high concentration (60 to 90 wt%) of glass formers (SiO_2 and Al_2O_3) and the ability to treat most soil and wastes without temperature-lowering additives such as sodium.

The vitrification process can accommodate complex mixtures of contaminant types including organic and inorganic contaminants. Large amounts of debris can be accommodated in individual melts. Types of debris treated in prior commercial operations include concrete, bitumen, bricks, steel, wood, plastic and automobile tires.

Off-gases that evolve from the melt are collected in a steel containment hood and directed to an off-gas treatment system. The off-gas treatment steps can vary depending on project requirements but generally consist of particulate filtration, quenching, wet scrubbing, a second stage of particulate filtration, and carbon adsorption and/or thermal oxidation.

SUBSURFACE PLANAR VITRIFICATION™ (SPV™)

The GeoMelt® Subsurface Planar Vitrification™ (SPV™) configuration is an application of the GeoMelt® technology that is used to treat subsurface contamination including buried waste or structures such as underground tanks, trenches and cribs. The melting process is initiated in the subsurface between pairs of electrodes (Figure 1). The planar melts can be initiated at the desired depth and separation within the subsurface, allowing for optimal treatment of the contaminated zone. If more than one pair of electrodes is used, they can be positioned so that individual planar melts coalesce as melting progresses.

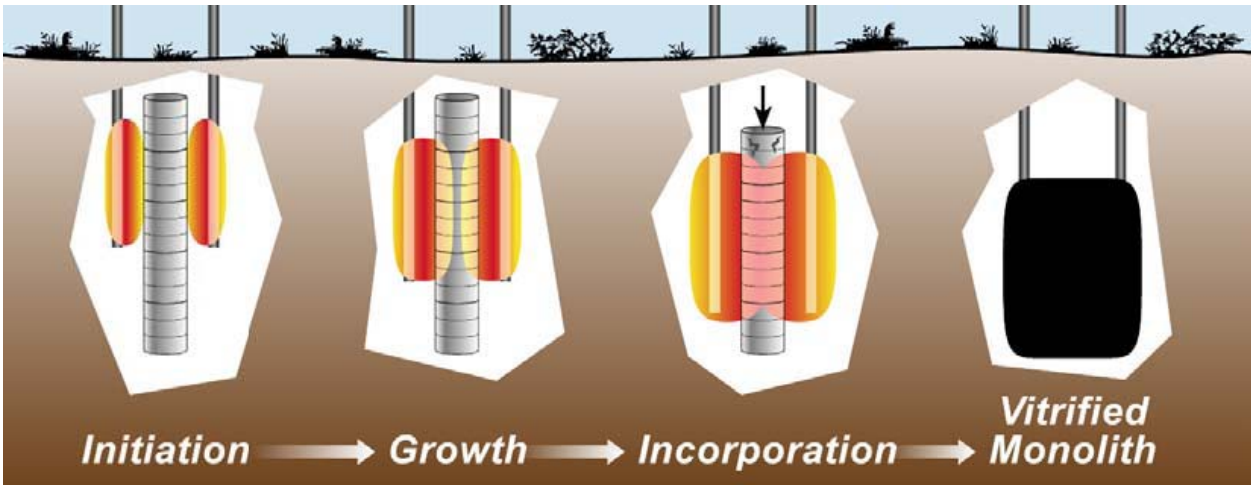

Fig. 1. GeoMelt® SPV™ Process.

The GeoMelt® SPV™ process was used to treat a portion of an inactive absorption bed at the Los Alamos National Laboratory site (in 2000) [5] and was previously demonstrated for the Idaho National Environmental Engineering Laboratory site for the treatment of buried tanks.[6] The current project is to evaluate the effectiveness of the GeoMelt® SPV™ process to treat waste contained in VPUs and caissons that were used for the disposal of remote handled mixed transuranic waste.

This technology was recommended because it results in the lowest worker, public, environmental, and programmatic risk. A key advantage of GeoMelt® SPV™ treatment is that by destroying the dangerous materials several feet below the ground, a safe distance always exists between workers and wastes until each VPU and its wastes have been converted into an inert glass form. Dangerous materials are treated in place, destroying organics, pyrophoric metals, and potential high energy wastes. The self-shielding properties of glass reduce the radiological dose rate at the glass surface and the resulting glass monolith is a suitable form for on-site disposal, if that option is selected.

In Situ Transuranic Waste Delineation and Waste Removal Project

The DOE is conducting this demonstration project to provide technical data to support an accelerated remediation approach in support of a 2018 regulatory milestone. Hanford's 618-10 and 618-11 Waste Burial Grounds contain approximately 10,300 cubic meters of TRU waste and are considered high-risk sites because of their radioactive inventory (Table I) and their potential to continue to contaminate ground water. The two sites include VPUs and caissons used in the 1960s for the disposal of remote handled mixed TRU waste. The high risk associated with these sites is due to three factors. First, the 300 Area laboratories handled highly radioactive and highly energetic materials and that the 618-10/11 burial grounds were used from 1954 to 1967 to dispose of the most dangerous wastes from site laboratories and fuel development facilities. Second, wastes that were too radioactive and/or otherwise dangerous for trench disposal were placed in the VPUs and caissons. Third, insufficient historical recordkeeping and lost records make every activity associated with those sites highly uncertain.

Table I. 618-10 and 618-11 VPU and Caisson Inventory of TRU and Other Dangerous Wastes.[7,8]

Volume of remote-handled TRU (cubic meters)	Pu-239 (Curies)	Sr-90 (Curies)	Cs-137 (Curies)
102 [a]	630	2000	2000

[a] Radiological and chemical hazards include cesium, strontium, thorium, americium, uranium, plutonium, curium, neptunium, zirconium, beryllium, pyrophoric sodium-potassium metals, oils, and solvents.

The VPUs were constructed by welding five bottomless 205 L (55-gal) drums end-to-end to result in 4.6-m (15-ft) long shafts. The caissons were constructed of 2.4-m (8-ft) diameter corrugated metal pipe, 3.05-m (10-ft) long, with the top of the caisson being 4.6-m (15-ft) below grade, and connected to the surface by an offset 0.91-m (36-in) diameter pipe with a dome cap lid (Figure 2).

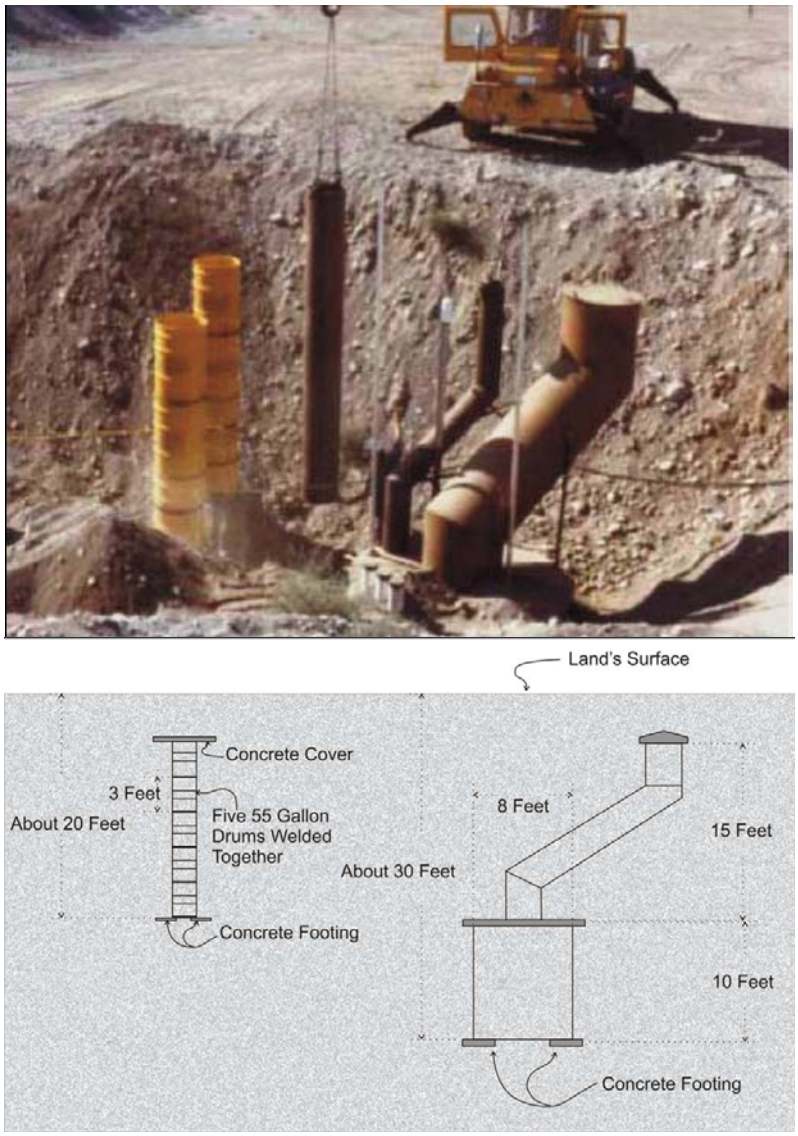

Fig. 2. Typical vertical pipe unit (left) and caisson (right).

Phase II of the In Situ TRU Waste Delineation and Removal Project included a non-radioactive demonstration of AMEC's GeoMelt® SPV™ process on a mock VPU configured to match the actual VPUs.[1] AMEC successfully completed the demonstration at its test site in Richland, Washington in May 2005. A mock VPU was constructed and loaded with a variety of non-hazardous materials (Table II). A non-radioactive cerium oxide tracer, to simulate plutonium and verify vitrified product homogeneity, was placed in the vertical center of the mock VPU, approximately 2.74-m (9-ft) below the ground surface.

Table II. Mock VPU Contents

Contents	**Description**	**Concentration**
Metallic Debris	364 kg steel fragments/canisters	20 wt%
Organic Debris	41 kg wood	5 wt%
Concrete	Concrete blocks	10 wt%
Soil	Sandy soil	65 wt%
Non-Radioactive Tracer	CeO_2	725 g

Electrodes were installed and starter paths were injected below grade. Two starter paths were installed in the subsurface between the two electrodes using a pressure injection technique to provide electrically conductive pathways to initiate the melting process. The starter paths formed two 0.91-m (3-ft) tall starter planes, surrounding the VPU, in the approximate shape of an ellipse. Thermocouples assemblies were positioned in vertical and horizontal arrays adjacent to the VPU to monitor the growth of the melt. A hood was installed over the melt area to capture off-gases for treatment and an overburden delivery system was deployed to compensate for subsidence by maintaining a cover of soil/gravel above the melt. An electrode feed system was employed to periodically insert electrodes as the depth of the melt increased (Figure 3).

Fig. 3. GeoMelt® SPV™ off-gas containment hood, electrode feed assembly and overburden delivery system.

The demonstration took approximately 130 hours and resulted in a vitrified monolith weighing an estimated 90 tonnes. All equipment worked as designed and no abnormal conditions were experienced. The total depth of the melt from grade was in excess of 6.1-m (20-ft) upon completion, indicating complete treatment of the mock VPU and its contents. The overburden delivery system successfully fed pea gravel to the melt over the entire 6-day melt period without incident.

Thermal modeling is used to predict moisture and heat movement in the subsurface beyond the extent of the melt. Data collected during this demonstration were used to calibrate a recently updated thermal model called TEMPEST [9], which was successfully used to correlate predicted and observed temperature profiles in a sufficiently accurate manner to design melts in similar geologic strata, such as found in the 618-10/11 burial grounds (Figure 4).

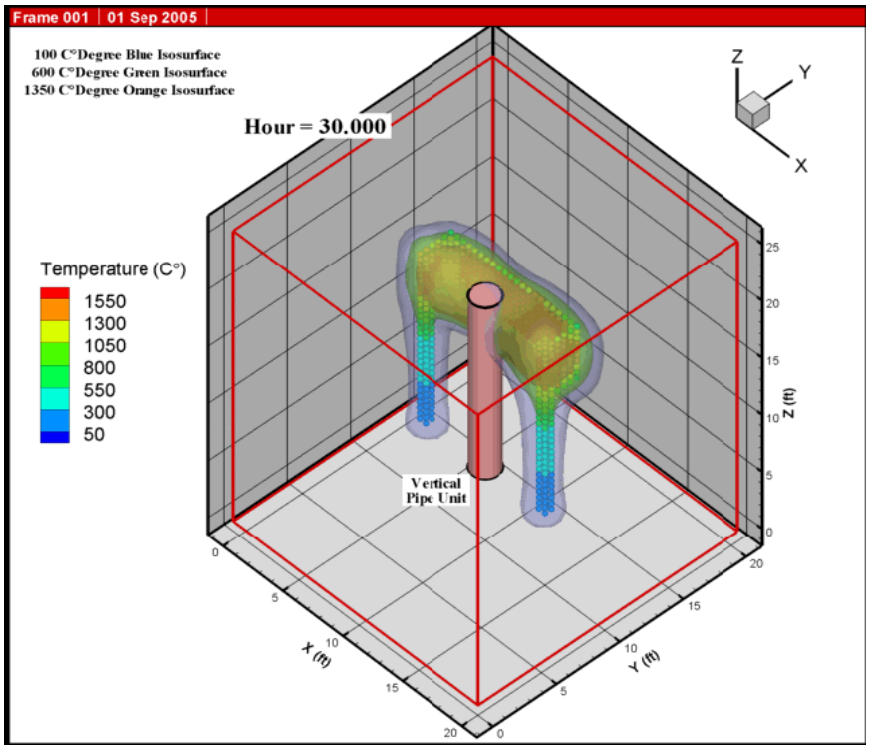

Fig. 4. Computer model showing progress of VPU vitrification at hour 30 of the 130 hour demonstration.

In June of 2005 the vitrified monolith was excavated to facilitate cooling in preparation for sampling and analysis. In July 2005 samples were collected and analyzed to determine product homogeneity using cerium oxide as a non-radioactive tracer to mimic the physicochemical properties of plutonium. The vitrified monolith is shown in Figure 5 after it was partially excavated.

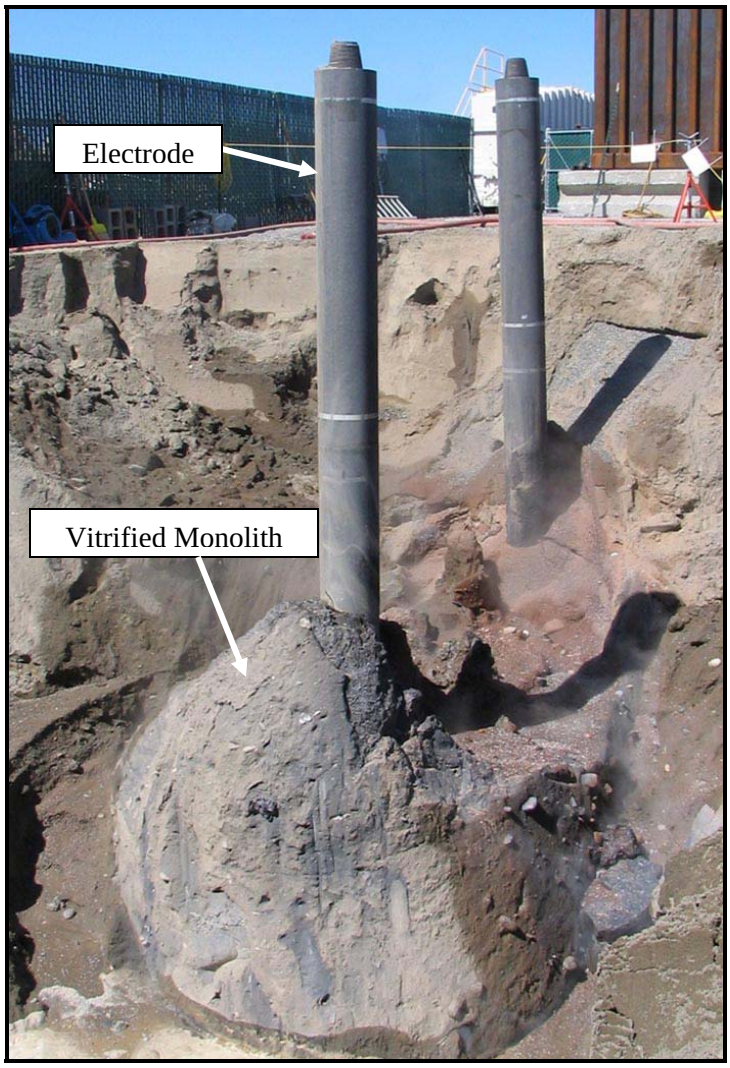

Fig. 5. Vitrified monolith after partial excavation to promote cooling and sampling. Two graphite electrodes are visible extending upward from the monolith.

A primary goal of the demonstration project was to validate that the melt generates a glass monolith in which hazardous materials not destroyed by the melt (e.g., toxic metals) are homogeneously distributed throughout the melt. The homogeneity is attributable to convection currents within the molten glass (which has a viscosity similar to honey) that circulate molten materials (Figure 6). The resultant convective current effectively distributes the contents of the material within the VPU and the soil surrounding the VPU into a homogenous mass.

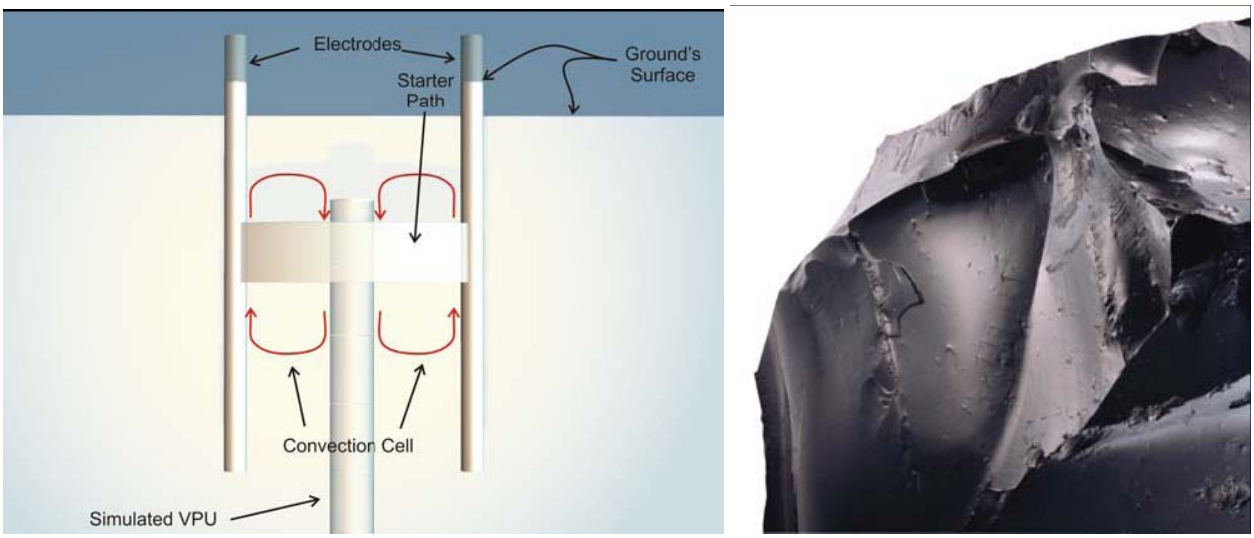

Fig. 6. Convective currents in the melt (left) produce a homogeneous glass product (right).

Cerium oxide was used to assess the homogeneity of the resulting monolith. A total of 725 grams of cerium oxide was placed in a single location within the VPU. From these initial quantities and the target mass of 100 metric tons for the monolith, the concentration of cerium above the background is calculated to be 5 mg/Kg of glass. The sampling plan identified the acceptance criteria for homogeneity - cerium concentration in the melt are within ±15% of the measured mean of the sample population.

A total of 60 representative glass samples were collected. Fifteen samples were collected from the surrounding soil. The mean concentration values of cerium in the soil (background), the glass monolith and the difference in the means were determined. These values are presented in Table III.

Table III. Cerium Concentrations

Background Mean (mg/Kg Ce)	Glass Monolith Mean (mg/Kg Ce)	Delta (mg/Kg Ce)
48.09	53.37	5.28

From Table II, the difference is 5.28 mg/Kg, which correlates well with the estimated increase of 5 mg/Kg, due to the 725 grams of cerium oxide added to the VPU. While the background concentration of cerium in the soil was higher than anticipated, at the 95% confidence level, the glass sample data (minimum and maximum values for the mean) is above the range of the background data. This demonstrates that the measured increase is due to the cerium oxide material added to the VPU. This fact, combined with the high correlation between the anticipated concentration increase and the mean values, indicates that the cerium was well mixed within the monolith, resulting in 29 of the 30 data points being within the ±15% of the mean target for homogeneity.

Downhole Delineation

In addition to demonstrating GeoMelt® SPV™, delineation techniques have been evaluated as part of the project to confirm the locations of the actual VPUs and to progressively determine their physical and chemical contents. The delineation of the VPUs must provide sufficient information to design the melt, ensure minimal hazard to personnel, public and the environment, and only minimally intrude upon the buried waste.

A delineation/calibration facility was designed and built at the AMEC test site in Richland, Washington to demonstrate the proposed technologies for delineation of the contents and orientation of the undisturbed VPU. The facility consists of two demonstration areas:

- A site for calibration of a neutron-gamma tool to delineate VPU contents (Figure 7) – This technique requires a scintillation detector and a small neutron source and can be used to identify of materials types, voids and layers (e.g., cement, sand).
- A site to calibrate and validate the use of down-hole ground penetrating radar – This tool is used to estimate the precise location and orientation of a buried VPU.

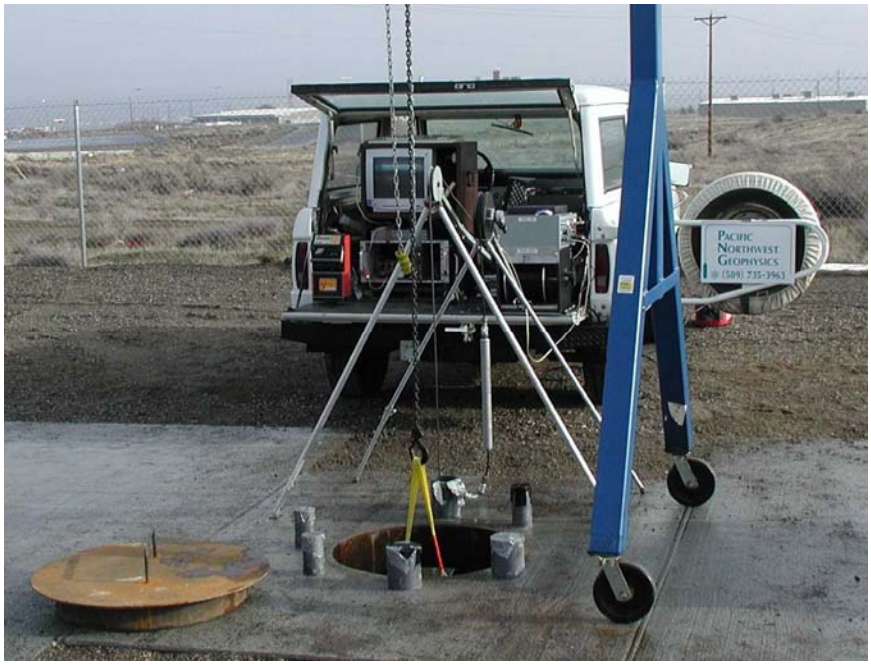

Fig. 7. Neutron-Gamma calibration facility.

The initial calibration and testing activities were completed in December 2005. At the time of this writing, the data were being analyzed.

Life Cycle Analysis

A life cycle analysis [1] was conducted to further examine the applicability of vitrification versus excavation. As summarized in Figure 8, the GeoMelt® SPV™ approach offers substantial advantages in terms of reduced worker risk, public risk, environmental risk, and programmatic risk relative to other approaches that bring the waste to the surface without first putting it into a safe, inert form. The life cycle cost is also minimized by using the GeoMelt® SPV™ approach.

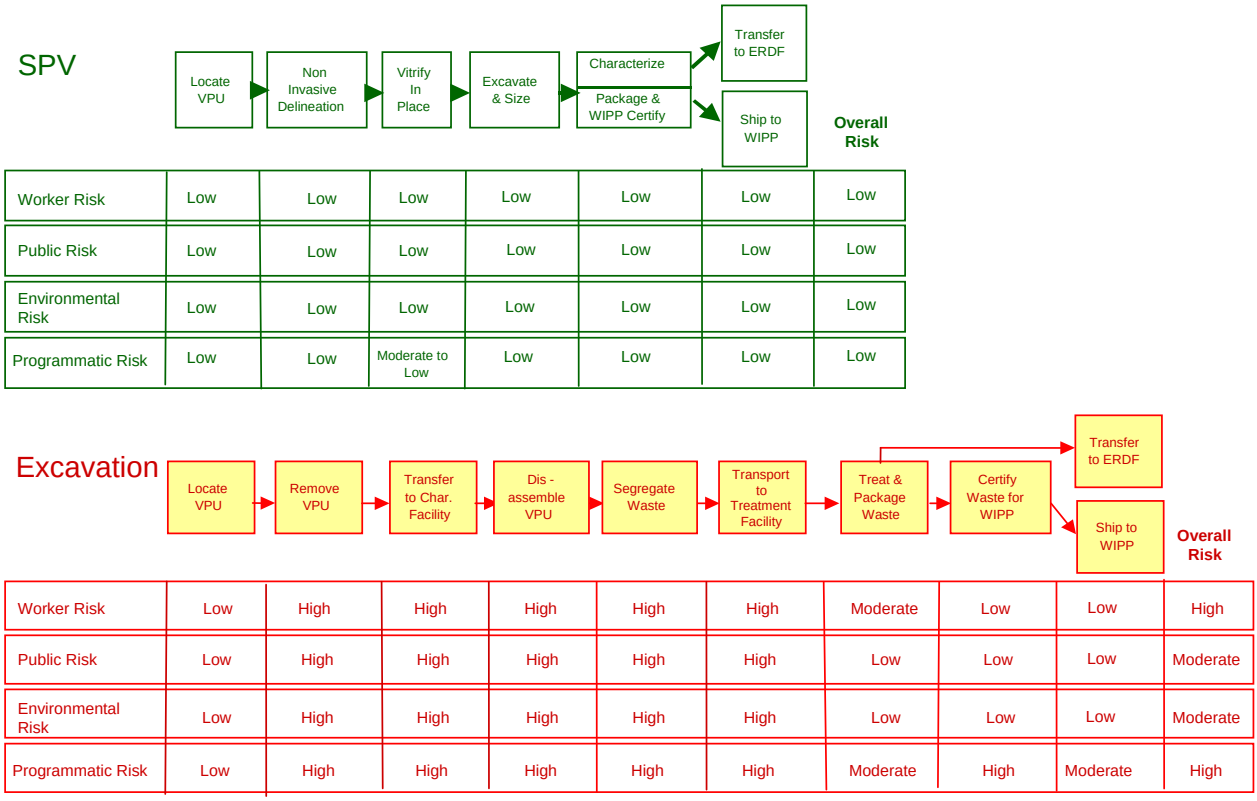

Fig. 8. Life cycle analysis showing benefits of GeoMelt® SPV™ treatment relative to baseline technologies.

Although excavation and removal appears to be simple and fast, the untreatable waste must be transported over the road to a facility where the contents of the VPU– which are still unknown with regard to chemical stability, energy potential, and flammability – must be glovebox sorted and characterized for appropriate treatment. Bulk stabilization of the waste (e.g. macroencapsulation for subsequent bulk disposal without characterization) would not address the waste disposal criteria for reactive materials, the potential for liquids, and radionuclide inventory limits. Substantial costs are associated with building the waste sorting and treatment capabilities required. Substantial worker and public risks are associated with every step of the baseline process.

With the GeoMelt® SPV™ technology, by contrast, nearly all of the vitrified waste is estimated to be suitable for bulk disposal in Environmental Restoration Disposal Facility at Hanford, Washington or at another appropriate disposal facility. Once homogenized in the glass, its transuranic (TRU) element concentrations will be below TRU levels and the vitrified waste could be disposed of in alternative facilities rather than the Waste Isolation Pilot Plant in New Mexico. Thus GeoMelt® SPV™ treatment avoids substantial TRU waste certification, shipment, and disposal costs.

CONCLUSIONS

The Phase II demonstration of AMEC's SPV™ process was a success both from a design and operational standpoint. The melt process treated the entire mock VPU and achieved a melt depth of over 6.1-m (20-ft). No operational difficulties were encountered. The resulting vitrified product satisfied the project

criteria for homogeneity with 29 of the 30 data points being within the ±15% of the mean target for homogeneity.

The life cycle analysis for the GeoMelt® SPV™ approach indicates it offers substantial advantages in terms of reduced worker risk, public risk, environmental risk, and programmatic risk relative to other approaches that bring the waste to the surface without first putting it into a safe, inert form. The life cycle cost is also minimized by using the GeoMelt® SPV™ approach.

The calibration of the neutron-gamma tool and demonstration of the down-hole ground penetrating radar provides the information needed to ensure that VPUs/caissions can be safely and effectively vitrified in place. Calibration work continues and preparations are underway to perform delineation activities on an actual subsurface structure on the Hanford site in 2006.

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