

**APPLICATION OF A NEW IN SITU VITRIFICATION TECHNOLOGY
FOR THE
REMEDICATION OF UNDERGROUND STORAGE TANKS^a**

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ABSTRACT

A new form of in situ vitrification (ISV) has been developed for remediating both underground storage tanks (USTs), and waste sites containing large quantities of vaporizable material coupled with relatively impermeable barriers (such as basalt or clay). The new process, called planar-ISV, was developed by Geosafe. Planar-ISV uses two vertical planes of starter material to initiate ISV processing. The molten planes then process the tank or site in a “downward and inward” fashion. During planar-ISV processing, the volatile and semivolatile contaminants in the waste are destroyed or processed by the planar-ISV melts that surround the site. Once the gas-generating materials have been processed and the region between the melt planes has dried out, the melt planes converge, to rapidly process the waste site area and its associated nonvolatile materials. The new process eliminates concerns over melt expulsion events during ISV processing.

Planar-ISV was recently evaluated in a treatability study for the Idaho National Engineering and Environmental Laboratory (INEEL). The treatability study evaluated the application of planar-ISV to large USTs containing various mixed, hazardous and radioactive wastes, including polychlorinated biphenyls (PCBs), which limit the potential technology options available for remediation. The treatability study included:

- A single 6.7-m (22-ft) long planar melt (needed to verify it could remediate 6.1-m long USTs)
- A dual-planar melt of a heavily instrumented, 2.4-m (8-ft) diameter x 3.7-m (12-ft) long UST, containing fill soil, 5,640 L (1,490 gal) of water, 1,800 g (4 lb) of hydrated cesium hydroxide ($\text{CsOH} \cdot \text{H}_2\text{O}$), 45.4 kg (100 lb) of hydrated lithium hydroxide ($\text{LiOH} \cdot \text{H}_2\text{O}$), and 11.3 kg (25 lb) of molybdenum oxide (MoO_3).

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The treatability study showed that planar-ISV effectively remediates USTs up to 7.3 m (24 ft) long and containing over 33 vol% aqueous liquids. In particular, the heavily instrumented tank was effectively processed without any significant pressure buildup within the tank (a prerequisite for melt expulsion events). In addition, the test showed up to 99.996% cesium retention in the ISV waste form, similar to that of conventional-ISV melts. Additional benefits associated with planar-ISV include a 50% reduction in energy consumption, vs. conventional-ISV, and the potential segregation of radioactive contaminants from hazardous contaminants in the ISV off-gas system (due to different condensation locations).

Based on the results of the treatability study, planar-ISV has been selected as the preferred technology for remediating the TAN site V-Tanks at the INEEL. A final decision on planar-ISV's use will be made as part of the Waste Area Group (WAG) 1 Record of Decision, currently scheduled for the end of FY-1999. The results of this test may also be potentially applicable to other, even larger USTs within the DOE complex.

INTRODUCTION

Four USTs await remediation at the INEEL Test Area North (TAN) Site. These tanks contain a mixture of radioactive and chemical contaminants, including PCBs. Three of the tanks (tanks V-1, V-2, and V-3) are approximately 3.0 m (10 ft) in diameter, with a length of 5.9 m (19-ft, 4-in). They were placed 6.1 m (20 ft) below grade surface (bgs), with their longitudinal axis parallel to the horizontal, and a separation between tanks of approximately 91 cm (3 ft). The tanks are buried approximately 2.4 m (8 ft) away from the nearest building (still in use). The fourth tank (tank V-9) is considerably smaller than the other three tanks, and is buried away from tanks V-1, V-2 and V-3. The presence of PCBs in these tanks, coupled with alpha contamination, has significantly complicated and restricted the choice of acceptable remediation techniques.

A review of potential remediation technologies found that ISV is the only currently available technology that can meet all potential Applicable Regulations and Requirements (ARARs) with regards to radioactive, hazardous, and PCB-contaminated wastes. The ISV process was patented by Battelle Pacific Northwest National Laboratory and DOE in 1983 (1), with commercial application exclusively licensed to Geosafe Corp. The efficacy of using ISV to treat radioactive and organic wastes has been well established by both Battelle Pacific Northwest National Laboratory and Geosafe. In addition, Geosafe has obtained a National Toxic Substances Control Act (TSCA) permit to use ISV to remediate PCB-contaminated wastes in a contaminated soil/waste matrix, at concentrations up to 17,680 ppm.

Conventional-ISV technology processes the contaminated soil/waste matrix in a top-down fashion. To initiate conventional-ISV, a series of graphite electrodes are partially inserted into the soil directly above the site to be processed. These graphite electrodes are connected to electrical power, and a conductive path of graphite and frit is placed between each graphite electrode, to complete the electrical circuit. The electrical circuit is then engaged. The electrical circuit causes Joule heating, which is then used to heat and melt the soil/waste matrix surrounding the graphite starter path. Once molten, the soil itself can support the flow of electrical current,

thereby dissipating enough Joule heat to propagate the melting process. Consequently, the melt develops in size to vitrify the region from near the top surface down through the column of soil and waste materials to the desired depth. In processing a tank from the top down, however, there is the potential to trap volatile materials between the bottom of the advancing melt and the yet-to-be processed tank walls. As the volatile materials in the tank heat up, pressures can build in this region until the vapors formed are forced to pass through the molten mass. This can lead to displacement of molten material from the melt pool and a consequent overheating of the off-gas treatment hood and appurtenances.

Recently, however, Geosafe has developed an enhancement to conventional-ISV that allows for safe, effective processing of USTs. This enhancement, called planar-ISV, appears to address concerns over melt expulsion by minimizing the potential pressure buildup within buried containers holding liquid wastes. With planar-ISV, the horizontal array of starter tubes used in conventional-ISV is replaced with two vertical planes of starter material. These starter planes form two independent melts during the initial stages of the process. This controls the initial melt process so that it can be focused for optimal treatment of the waste zone. Moreover, because the melts are separated laterally during their initial stages, the UST or buried container can be processed in a predominantly sideways-in fashion. This maintains a permeable zone of soil above the center of the tank, which allows for the controlled transport of gases generated during processing to the off-gas treatment system. Any gases generated from hazardous materials during processing are either pyrolyzed during melting or are removed to the off-gas treatment system. By the time the two melts have merged, the process will have effectively and safely removed all volatile materials (mainly water) in the treatment zone. Figure 1 illustrates the planar-ISV melting technique applied to an underground tank.

Before the treatability study, Geosafe had demonstrated this application in numerous tests, including engineering-scale tests on buried canisters filled with water-saturated soil, and pilot-scale testing on 420-L (110-gal) salvage drums containing fully saturated soils. Testing was then increased in scale to a 7,600-L (2,000-gal) tank with similarly successful results. Planar-ISV melts up to 4.6 m (15 ft) long had also been successfully

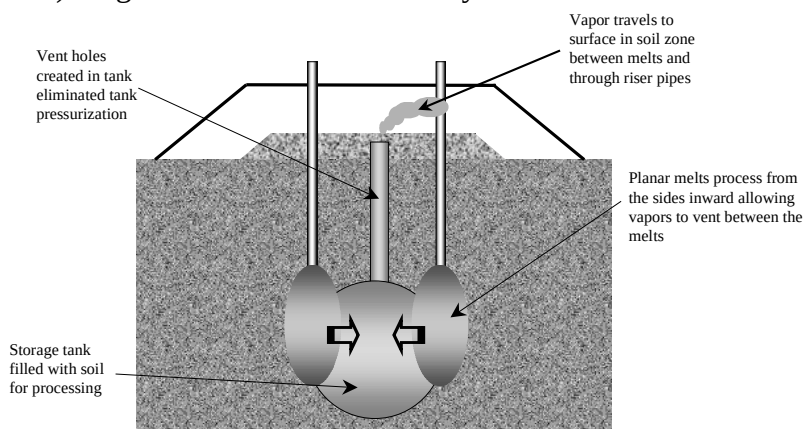


Figure 1. Illustration of Planar Melting Applied to an Underground Tank.

demonstrated. However, concerns still remained on whether the existing equipment was of sufficient size to effectively vitrify a 6.1-m (20-ft) long tank, containing over 30 vol% water. It is for these reasons that a planar-ISV treatability study was commissioned.

TEST OBJECTIVES AND SETUP

The purpose of the planar-ISV treatability study was to extend the scale of application and demonstrate the efficacy of the planar-ISV process for remediating the TAN-Site V-Tanks. This was needed to evaluate whether planar-ISV can be safely applied to the V-Tanks in a manner that meets the regulatory and safety requirements associated with their contents and the INEEL site. To meet this purpose, the treatability study was broken down into a series of two field-scale planar-ISV tests, which are described below.

Long Starter Plane Test

The first planar-ISV test was used to evaluate whether planar-ISV could meet the dimensional requirements for all TAN V-tanks under consideration. This involved staging a single planar-ISV melt at a depth, length, and height representative of that required for the largest of the TAN V-tanks. The largest (and deepest buried) of the V-tanks under consideration are tanks V-1, V-2 and V-3, which are 3.0 m (10 ft) in diameter, approximately 5.9 m (19.3 ft) long, and buried horizontally, so that the top surface is approximately 3.0 m (10 ft) below grade. Based on planar-ISV design requirements, this required a planar-ISV melt with an electrode spacing of 6.7 m (22 ft) to ensure that the ISV melt would surround the entire length of the tank during processing. In addition, the required height for the planar-ISV starter plane was 1.5 m (5 ft), one-half the total diameter (or height) of the actual tank. Per design requirements, the planar-ISV starter plane was buried at a depth 1.5–3.0 m (5–10 ft) below grade (top to bottom), and had a nominal thickness of 1.9 cm (0.75 in.). To attempt to make the test representative of INEEL site conditions, the melt was performed in non-contaminated soil obtained from the INEEL TAN Site. The singular planar-ISV melt was to be initiated in situ, then propagated until the planar-ISV melt had grown to a thickness greater than 0.9–1.2 m (3–4 ft). Type K thermocouples were placed throughout the INEEL soil to monitor planar melt growth during the test.

Simulated V-Tank Test

The purpose of the second field-scale ISV test was to verify that planar-ISV could be used to effectively process a large subsurface tank, containing large amounts of aqueous or combustible liquids, without process upset. Of primary concern was the potential of a melt expulsion, which would be expected if the planar-ISV process did not allow a sufficient pathway for transporting vaporizable material from the tank (other than through the ISV melt). The second test involved planar-ISV processing of a cylindrical, horizontally buried carbon-steel tank, slightly smaller in diameter than the larger TAN V-tanks. The tank used in this test was 2.4 m (8 ft) in diameter and 3.7 m (12 ft) long, and was buried at a depth of 3.0–5.5 m (10–18 ft) below grade (top to bottom). Total volume for the test tank is 17,000 L (4,500 gal). This represents a scale-down of approximately 20%, based on tank diameter, and 38%, based on tank

length. Reducing the diameter of the test tank allowed for a field-scale evaluation of planar-ISV implementability on large subsurface tanks, at less than 50% of the cost associated with remediating a full-size prototype of tanks V-1, V-2, and V-3.

To simulate the contents of the V-tanks, the test tank was filled with soil and at least 5640 l (1490 gal) of water. The total volume of water in the tank is equivalent to approximately 33% of the total volume of the tank (discounting the initial moisture content of the added soil). This level is similar to the average volume of aqueous waste and sludge expected to be present in each of the large V-tanks, after effluent transfer from tank V-3. The inclusion of soil in the tank is in accordance with the preconditioning requirement of filling the residual void volume of the tank with soil, prior to initiating planar-ISV.

Included in the mixture of soil and water was 1,800 g (4.0 lb) of hydrated cesium hydroxide ($\text{CsOH}\cdot\text{H}_2\text{O}$), 45.4 kg (100 lb) of hydrated lithium hydroxide ($\text{LiOH}\cdot\text{H}_2\text{O}$), and 11.3 kg (25 lb) of molybdenum oxide (MoO_3). These chemicals were added to the bottom of the tank, to simulate the contamination in the sludge layer present in the actual V-tanks. The nonvolatile lithium hydroxide and molybdenum oxides were added to the test to assist in evaluating waste form homogeneity following planar-ISV processing. The nonradioactive cesium hydroxide was added to the soil and water mixture to evaluate the potential volatility of radioactive cesium during planar-ISV processing. Based on compositional data, it appears that Cs^{137} is one of the more prevalent radionuclides in the V-tanks. It is also one of the more potentially volatile, and there were concerns that the use of planar-ISV may result in a higher degree of volatility than has been observed in past "conventional" ISV melts. The increased cesium volatility, if real, could cause increased exposure concerns than originally anticipated. As a result, it became a major data gap that needed to be explored in the simulated tank test. The results of the cesium volatility evaluation are discussed in a separate section of the report.

After filling the tank with soil, water and the simulated contaminants, five large vents, 35.5 cm (14 in.) in diameter and 3.8 m (12.5 ft) long, were placed on top of the test tank. The vents extended approximately 61 cm (2 ft) into the subsurface tank. The vent pipes provided a low resistance pathway for transporting volatile vapors from the tank during planar-ISV melting. The inclusion of the vent pipes is consistent with plans for the TAN V-tanks. However, the existing V-tanks have flanges on top, which may be used as vents, therefore requiring less additional vent pipe placement.

Each vent pipe was filled with a zeolite material that is effective in capturing any volatilized cesium released during planar-ISV processing. The presence of zeolite in these vent pipes limits cesium transport to the ISV off-gas system, where it could become a potential exposure concern if the rate of volatilization was high enough. The zeolite material may also effectively capture other volatile inorganic contaminants in the tank, such as mercury.

To better simulate tank conditions at the INEEL, the simulated tank was surrounded with INEEL soil. During back filling, graphite electrodes and planar-ISV starter planes were placed in their appropriate position. Two electrodes were placed on each side of the tank, 4.6 m (15 ft) away from each other. Each pair of electrodes was placed parallel to the side of the tank, and

located 30 cm (1 ft) away from the edge of the tank. This resulted in a rectangular electrode pattern, 3.0 m (10 ft) by 4.6 m (15 ft), that surrounded the simulated V-tank. The bottom of the graphite electrodes were buried at a depth of 3.7 m (12 ft). The starter planes of graphite and glass frit were placed between the electrodes on each side of the tank. The starter planes had dimensions of 4.3 m (14 ft) long by 1.2 m (4 ft) high by 1.3 cm (0.5 in.) thick, and were buried at a depth of 2.4–3.7 m (8–12 ft) below grade, top to bottom.

Type K thermocouples were placed at numerous locations within the tank, the vent pipes, and the surrounding soil, to provide a relatively complete profile of the melt progression during planar-ISV processing. Pressure transducers were also attached to the tank to measure the potential buildup in pressure that could occur within the tank during planar-ISV processing. In addition, an array of type-K thermocouples was positioned at a considerable distance from the tank to estimate both the maximum growth of the 100°C isotherm in the soil surrounding the ISV melt. This information is important, due to the presence of adjacent buildings within the vicinity of the subsurface V-tanks.

Prior to initiating planar-ISV processing, a 30-cm (1-ft) thick layer of gravel and an off-gas collection hood were placed over the site. The gravel was used to enhance subsidence control during ISV processing, while the off-gas hood collected and transported the off-gas to an off-gas treatment system. Because there were no hazardous contaminants in the test, there was no need to use wet scrubbing in the off-gas system. Nevertheless, the off-gas system was periodically monitored to determine the degree of cesium volatilization that resulted during off-gas sampling.

Planar-ISV processing continued until the planar melts had come together, and melted the entire tank volume, as well as the soil 30 cm (1 ft) below the tank bottom. Termination of planar-ISV melting was based on type K thermocouple data within the areas of concern. Following planar-ISV melting, the type K thermocouple data continued to be recorded for a time sufficient to define the estimated melt and isotherm growth that resulted from planar-ISV processing.

PRE-TEST INEEL SOIL EVALUATION

To simulate the expected conditions of the TAN V-tanks, both planar-ISV tests were to be performed using INEEL soil representative of that surrounding the TAN V-tanks. The INEEL soil that was sent to Geosafe came from a gravel pit north of TAN. This soil was expected to be representative of the soil surrounding the TAN V-tanks. However, it was found (via whole rock analysis) that the gravel pit soil had an extremely high concentration of lime, which lowered the expected soil melting temperature to 1,370– °C, slightly less than the melting temperature of the tank itself (1,515°C). These results were significantly different from what was expected, based on whole-rock analysis from a bench-scale sample of soil that was taken near the tanks. The low melting temperature of the soil was of concern, since it could result in melting the contents of the tank, as well as the surrounding soil, without melting the tank itself. Based on the differences in expected soil melt temperatures for both whole rock analysis samples, it was concluded that the high-lime soil that was shipped to Geosafe was probably not representative of the soil surrounding the TAN V-tanks. As a result, it was decided to add soil from Geosafe's test site to the high-lime soil that was shipped to Geosafe from the INEEL. The Geosafe test site soil

was higher in silica, and therefore raised the expected melt temperature of the surrounding soil to a temperature above that of the carbon steel tank.

Following the simulated V-tank test, coring and whole rock analysis was performed on samples of the soil surrounding the TAN V-tanks. This was necessary to determine whether the “high lime” soil was representative of the actual soils surrounding the TAN V-tanks. A comparison of the normalized oxide composition of the simulated test soil, with that of the TAN site soil, is shown in Table 1. The results found that there was much less calcium oxide (i.e., lime) in the TAN site soils than that in the simulated V-tank soils. The reduced lime in the TAN site soils will result in an expected melt temperature above 1,600°C, reducing the need to add refractory material.

LONG STARTER PLANE TEST RESULTS

The long starter plane test was initiated on March 11, 1998, and was completed approximately 29 hours later. The test included a linear ramp-up to 1,060 kW during the first 19.5 hours of operation, and maintenance of this power level through the remaining 9.5 hours of the test. A total of 21,300 kWh was delivered to the target treatment zone during this test.

After allowing sufficient time to cool, the vitrified block was excavated. The general shape of the monolith was fairly consistent over the length of the block. The length of the monolith was approximately 7.4 m (24.25 ft), while the width was approximately 2.3 m (7.5 ft). The top surface of the monolith was approximately 2.4 m (8 ft)

Table 1. Comparison of Simulated V-Tank Soil Composition to TAN Soil Composition.

Oxide	Simulated V-Tank Soil, wt%	TAN Soil, wt%
Al ₂ O ₃	5.4	11.9
CaO	29.2	13.4
Fe ₂ O ₃	2.0	4.3
K ₂ O	1.1	2.7
MgO	2.1	3.1
Na ₂ O	0.9	1.3
P ₂ O ₅	0.1	0.3
SiO ₂	57.6	62.4
TiO ₂	0.3	0.5

below grade surface. The monolith had a thickness of approximately 1.0 m (3.25 ft) in its center, which tapered off to 46 cm (1.5 ft) at the lateral extremities. Therefore, its maximum depth was approximately 3.5 m (11.5 ft) below the surface.

The density of the vitrified product was approximately 2.8 g/cm^3 . As a result, it is estimated that the vitrified monolith weighs 42,000 kg (92,600 lb). Since 21,300 kWh were used to generate this block, it is estimated that the specific energy consumption for this test was 510 kWh/tonne of glass. Assuming a total carbonate and moisture content in the INEEL soil of 25 wt% (based on whole rock analysis), this converts to a specific energy consumption of 380 kWh/tonne of soil, substantially less than the 800 kWh/tonne energy efficiency that has been reported for conventional top-down ISV processing.

The improvement is attributed to the enhanced thermal efficiency that results from performing the process below the soil surface. In a conventional top-down ISV process, a "cold cap" forms at the top of the melt. This cold cap is about 30 cm (1 ft) thick. Typically, a very large temperature gradient exists across this 30-cm (1-ft) thickness, causing a considerable amount of energy loss from the melt. By contrast, the thickness of dry soil covering the planar-ISV test was 1.5–1.8 m (5–6 ft) thick. Since dry soil is a very effective thermal insulator, the applied energy to the melt was redistributed, with a considerably greater fraction going to process the soil. The results indicate that additional soil overburden is effective in limiting heat losses during ISV processing.

A geochemical evaluation of the monolith found that the composition of the melt was similar to what was expected, based on whole rock analysis of the soil. As a result, it can be safely assumed that the entire portion of the soil (including the limestone-based rock) was effectively dissolved in the ISV melt. Elemental X-ray dispersive spectroscopy, wavelength dispersive spectroscopy, and X-ray fluorescence were used to evaluate the structure of the vitrified product. The results suggest that the resultant ISV monolith is highly crystalline, with substantial quantities of wollastonite and diopside. Based on a previous investigation (2), the ability to retain contaminants within these crystalline materials is as good, if not better, than if the product was completely vitreous.

Based on the results of the long starter plane test, it appears that planar-ISV will meet the dimensional requirements for processing the subsurface V-tanks at TAN.

SIMULATED V-TANK TEST RESULTS

The simulated V-tank test was initiated on April 7, 1998, and was completed on April 13, 1998, a total operation time of 146.75 hours. The test was initiated by power ramp-ups to 525 kW in the first 6 hours and 1,300 kW in the next 25 hours, followed by a decrease in power level to 1,100 kW over the next 71 hours of the test, due to amperage limitations. The ISV system was then powered down for approximately 2 hours, so that additional equipment cables could be added to the system, increasing the available amperage. After adding the cables, the

system was powered up to 1,500 kW and 1,750 kW over the next 20 hours and 14 hours, respectively, followed by idling the melt at 450 kW over the final 8 hours of the test. Based on the electrical data, a total of 157,800 kWh were consumed in processing this tank.

The amperage limitations during the first part of the test were due to the planar-ISV melts melting into the large carbon-steel tank. This resulted in a significant drop in resistance, due to the electrically conductive nature of the metal tank. Increasing the system's amperage normally offsets such a drop in electrical resistance. Unfortunately, the number of cables that were initially used on this test limited the maximum amperage to 2,200 amps/phase. This amperage limitation was less than what was required to maintain a desired power level of 1,500 kW during ISV processing. The amperage limitation was overcome by adding a third electrical cable to the system, after 102 hours of operation. The increased power level was still significantly under the limitations of the present Geosafe system, which can accommodate four cables per electrode, with a corresponding limit of 4,000 amps/phase.

The graphite electrodes used to power each ISV melt began feeding into the melt after 24 hours. Electrode feeding then continued throughout the test, accompanied by a number of time periods in which one or more electrodes were gripped or raised to arrest their downward progression. This was done to maintain fairly uniform electrode depths for all four electrodes, during processing, and whenever it was necessary to maintain an adequate separation between the bottom tip of the electrodes and the molten metal layer at the bottom of the ISV melt. As a result, the electrodes tended to feed into the melt in discrete motions, rather than at a smooth, continuous rate. The average electrode insertion rate was approximately 3.8 cm/hr (1.5 in/hr) during the first 24 hours (after initiating electrode feeding) and 1.3 cm/hr (0.5 in./hr) over the next 48 hours. The electrodes were gripped at an average depth of 5.2 m (17 ft) over the final 50 hours of the test, to concentrate power to the bottom tank contents (and keep the electrode tips out of the conductive metal layer at the bottom of the ISV melt).

During the simulated V-tank test, the pressure transducers that were placed in the tank (at the tank bottom, the tank side, and the tank end) never rose by more than 2800 N/m² (0.4 psig). The slight build-up in pressure was followed by a gradual, steady decline. This was due to the substantial volume of water that was present in the tank. In essence, the pressure transducer at the bottom of the tank simply served as a real-time indication of the head capacity or total volume of water left in the tank during planar-ISV processing. This indicates that the combination of the net effective flow area and path resistance provided by the vent pipes, together with the controlled melt processing rate, were sufficient to allow the steam generated within the tank to pass to the off-gas plenum without consequence during planar-ISV processing. The pressure transducers continued to operate until the advancing planar-melts had melted through their electrical connections.

Due to the extensive placement of thermocouples in the simulated V-tank, vent pipes and surrounding soil, a relatively good estimate can be made of the shape of the developing melt planes around the tank, at the plane crossing through the center of the simulated V-tank. This was achieved by defining a function that represents the time at which a particular thermocouple burned out, based on the thermocouple coordinates on this plane. Using this data, contour maps

could be generated for each melt plane at various times during planar-ISV processing. Figures 2, 3, and 4 show the estimated shapes of the melt planes at 48 hours, 96 hours, and 106 hours after initiating planar-ISV processing.

The contour diagrams show that the planar melts stay apart from each other, until the entire volume of water in the tank has been evaporated and vented to the ISV off-gas system. The presence of the tank limits joule heating of the tank contents (until the tank is melted) while the wet contents of the tank limit the ability of the melt planes to grow into the tank. As long as water remains in the tank, it is expected that the vaporization pathway to the ISV off-gas system will remain open. This is the primary reason that Geosafe developed planar-ISV.

The melt continues to grow laterally outward of the electrode envelope while the tank is off gassing. Once the water in the tank has been volatilized, the tank and its contents were rapidly melted to complete the remediation process. The rapid melt growth is illustrated in Figures 3 and 4 by the large increase in melt shape that occurs between the 96-hr and 106-hr time frame. During the remainder of the test, the upper shell of the tank and the 30-cm (1-ft) thick zone of soil beneath the tank were processed.

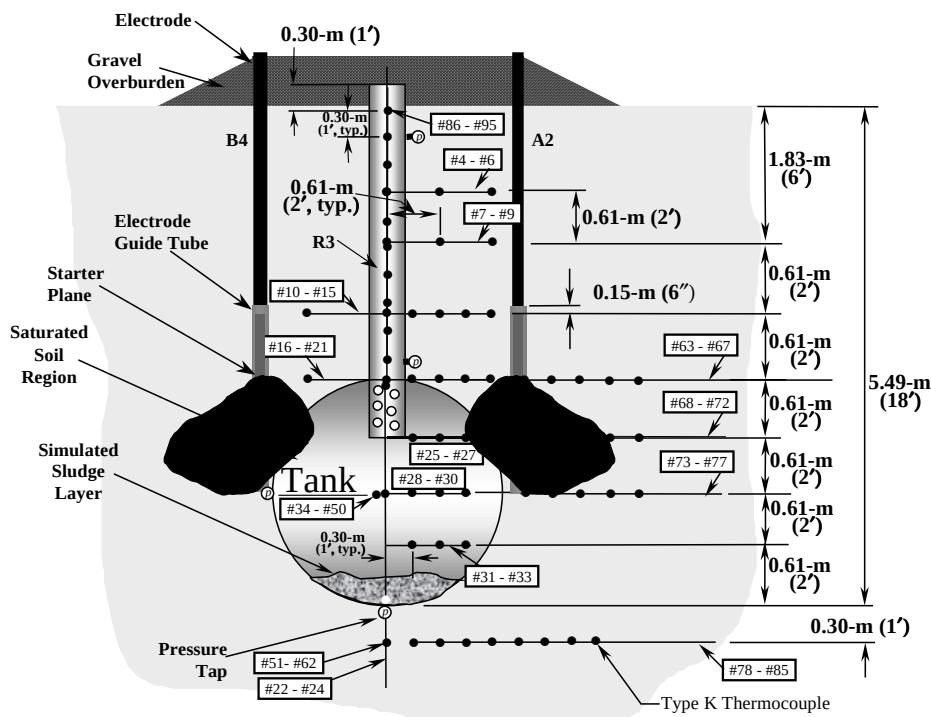


Figure 2. Simulated V-Tank Treatability Test – Estimated Melt Shape @ 48 hours

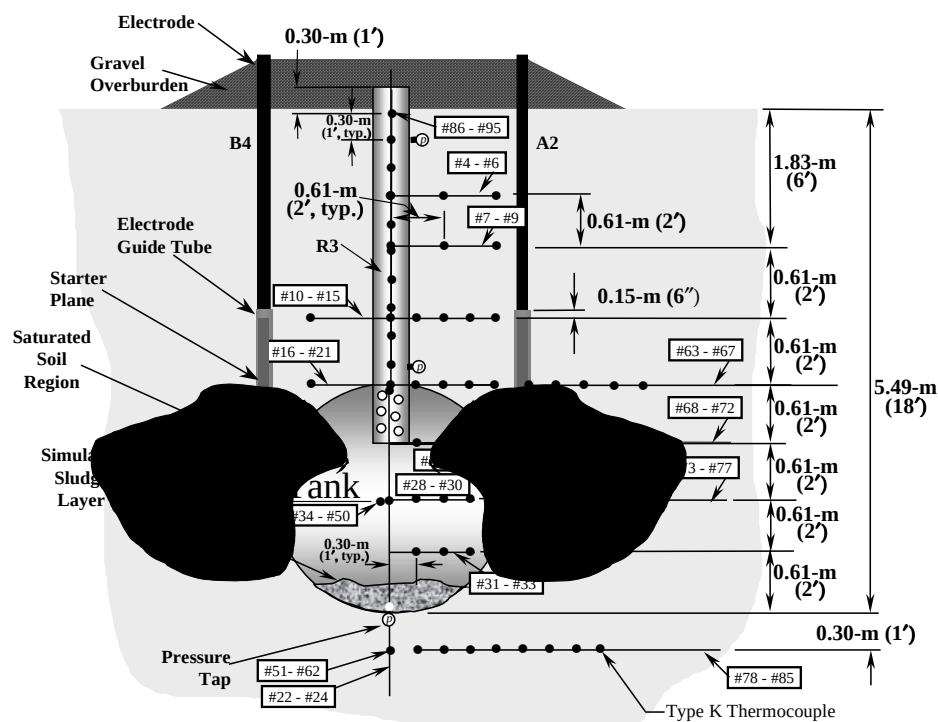


Figure 3. Simulated V-Tank Treatability Test – Estimated Melt Shape @ 96 hours

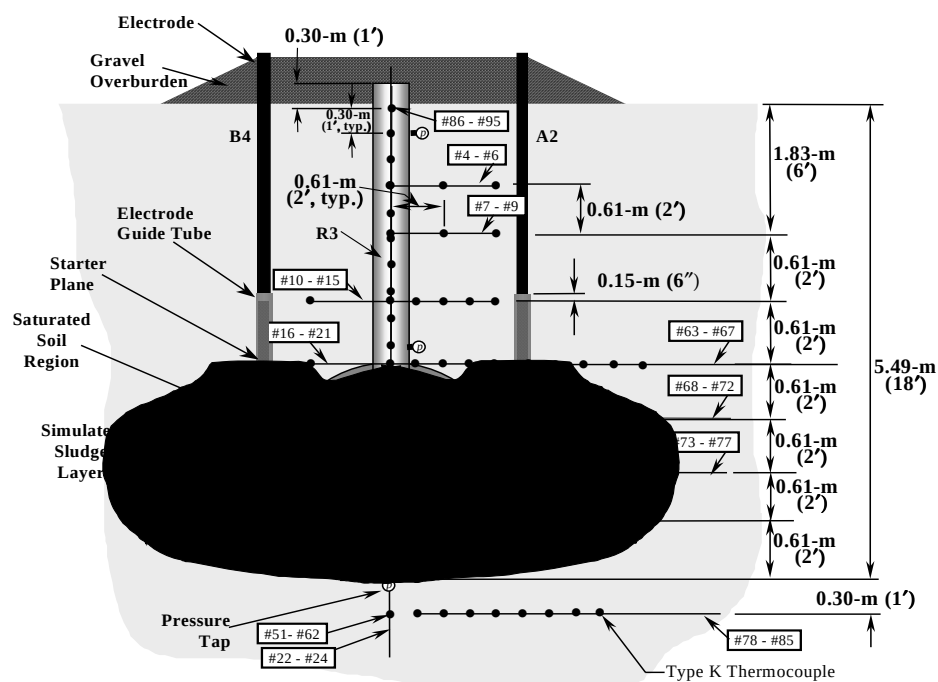


Figure 4. Simulated V-Tank Treatability Test – Estimated Melt Shape @ 106 hours

For situations where there are neighboring tanks on either side of the target tank (as with the TAN V-tanks), it is likely that the accentuated outward lateral melt growth would be inhibited, while the contents of the neighboring tanks are volatilized (along with the target tank contents). This would result in a narrower melt cross-section, and a more efficient melting process. Both types of situations could be effectively processed, however.

Additional information on how the planar melts grow into each other was obtained from a preliminary planar-ISV tank test that Geosafe privately performed, just prior to the simulated V-tank test. The preliminary test involved planar-ISV processing of a slightly smaller subsurface tank, 1.8 m (6 ft) in diameter. The preliminary tank test was stopped before the tank was processed, however, in order to evaluate how the planar melts grow into the tank. Upon excavating the end of the partially processed tank, it was observed that both melt planes initially tend to grow around the tank, rather than directly into it. This phenomenon supports the theory that water in the tank limits horizontal melt growth into the tank. The absence of water outside of the tank allows for enhanced horizontal melt growth at the ends (relative to the sides of the tank). The “surrounding” nature of the planar-ISV melts also supports the rapid rate of tank melting that occurs, once all of the water in the tank has evaporated.

Thermocouple data from the test indicated that the soil in the region between the starter planes, 61 cm (2 ft) above the tank, was brought up to 100°C within 24 hours after test start-up—well before a sizable melt had been established. Soil 1.2 m (4 ft) above the top of the tank reached a temperature of 100°C, 36 hours after start-up, while soil 91 cm (6 ft) above the tank acquired a temperature of 100°C after 72 hours of planar-ISV processing. Therefore, any materials that may volatilize at or below this temperature (e.g., tritiated water, and most of the hazardous halogenated hydrocarbons in the tank) will have been exposed to temperatures at or above their boiling points for a considerable period of time. The results show that the planar-ISV approach can be used to pre-treat a soil column between the planes, removing residual volatile materials in the soil/waste matrix by thermal desorption.

During planar-ISV processing of the simulated V-tank, the vent pipes in the tank effectively transported the water vapor from the tank to the off-gas system in a relatively steady-state manner (without undesirable surges in off-gas). The presence of zeolite in the vent pipes, coupled with the insulative nature of the surrounding soil, caused the off-gas hood plenum to remain at temperatures under 65°C throughout the entire test. In addition, the hood vacuum was effectively maintained throughout the test. This resulted in the effective removal of all condensate and entrained off-gas particulate from the off-gas flow stream, with much of the entrained particulate being captured in the zeolite material in the vent pipes. The resulting low particulate levels in the off-gas system were a direct consequence of the subsurface operation of this melt. The reduced particulate levels also extended the useful life of the HEPA filters that were used in the off-gas treatment system. The extended HEPA filter life is expected to result in a smaller volume of secondary waste generated from used HEPA filters during planar-ISV processing. In addition, it is expected that the presence of zeolite in the vent pipes, coupled with its capture potential, may allow for certain condensed contaminants (such as mercury and

cesium) to be separated from each other in the secondary waste stream. This is due to differing condensation temperatures for each contaminant, and the linear temperature gradient that is observed in the zeolite-filled vent pipes.

An average off-gas flow rate of $57.8 \text{ m}^3/\text{min}$ ($2040 \text{ ft}^3/\text{min}$) was maintained during the test. In addition, the average moisture content in the gas stream was approximately 3 wt%, but appeared to be decreasing throughout the test. The steady decrease in off-gas moisture content was a result of the planar-ISV process driving off the moisture contained both within the tank and within the surrounding soil.

After the tank test ended, the resultant melt block was allowed to cool. During cooling, extensive thermocouple monitoring was performed in the soil surrounding the vitrified monolith. This was necessary to define the extent of post-test isotherm growth in the surrounding soil. The results found that the 100°C isotherm grew to a position 2.3 m (7.5 ft) away from the edge of the melt, after terminating power to the planar-ISV melt. The resulting data is not expected to be representative of actual isotherm growth during full-scale remediation of the TAN V-tanks, for the following reasons:

- The reduced-scale of the simulated V-tank (less than half the size of the large TAN V-tanks)
- The need to partially excavate portions of the glass monolith, prior to terminating far-field temperature collection
- The location of the far-field thermocouples (slightly above the resultant monolith)
- The contributing latent heat resulting from three adjacent, full-scale melts, during TAN V-tank remediation.

Nevertheless, the data will be useful in predicting (via computer modeling) the amount of 100°C isotherm growth expected in the field-scale, multi-melt remediation of the TAN V-tanks. The evaluation of 100°C isotherm growth is necessary to define the degree of engineered cooling needed to protect adjacent buildings and structures that surround the TAN V-tanks, since the closest adjacent building is about 2.4 m (8 ft) away from the end of tanks V-1, V-2 and V-3.^b

After allowing sufficient time to cool, the resultant ISV monolith was excavated. Upon excavating the block, it was observed that the tops of the vent pipes had undergone an average net subsidence of 1.3 m (4.2 ft) from their initial positions, while the overlaid gravel layer had subsided a total of 2.0 m (6.5 ft). During

^b The building in question (Building 616) is currently not in use, and may be dismantled before remediating the TAN V-tanks. If this is the case, it may not be necessary to supply engineered melt cooling during planar-ISV processing. An evaluation of surrounding structures still needs to be made, however.

excavation, it was observed that the vent pipes had been cast into the ISV monolith, and had not become separated from the melt. As a result, the vent pipes continued to channel vaporizable material from the tank to the off-gas system, until all of the vaporizable material had been removed.

Figure 5 provides a view of the excavated ISV monolith, after it had been completely excavated and fractured (to allow sampling of the vitrified product). The fracture plane shown in this figure corresponds to a cross sectional cut, similar to that shown in Figures 2, 3 and 4, but only 46 cm (1.5 ft) inside the East wall of where the tank was originally staged. The mounds on the top of the fractured block are locations for two of the four graphite electrodes.

The ISV block was approximately 6.1 m (20 ft) wide and 8.5 m (28 ft) long. The top surface of the block resided approximately 4.3 m (14 ft) below the surface, and was approximately 1.5 m (5 ft) high in its central region (corresponding to the original location of the tank). The height of the block then thinned-out to 1.2–1.4 m (4–4.5 ft) as you moved away from the central region. Based upon these dimensions, and a measured glass density of 2.65 g/cm^3 , the estimated mass of the resultant block is 195 tonnes.

Since 157,800 kWh were used to generate this block, it is estimated that the specific energy consumption for this test was 810 kWh/tonne of glass. With the 195 tonnes of glass being equivalent to 218 tonnes of soil and water (based on knowledge of the moisture content in both the glass and surrounding soil), the estimated energy consumption for this test was 720 kWh/tonne of soil. This value, while higher than the 380 kWh/tonne of soil realized in the long starter plane test, is still below the nominal value realized in conventional top-down ISV applications in unsaturated soils (800 kWh/tonne of soil). In addition, the energy consumption is substantially less than the standard energy ratio expected for conventional-ISV processing of subsurface tanks with high volumes of aqueous liquids. The reduced energy consumption is a direct result of the subsurface nature of this test.

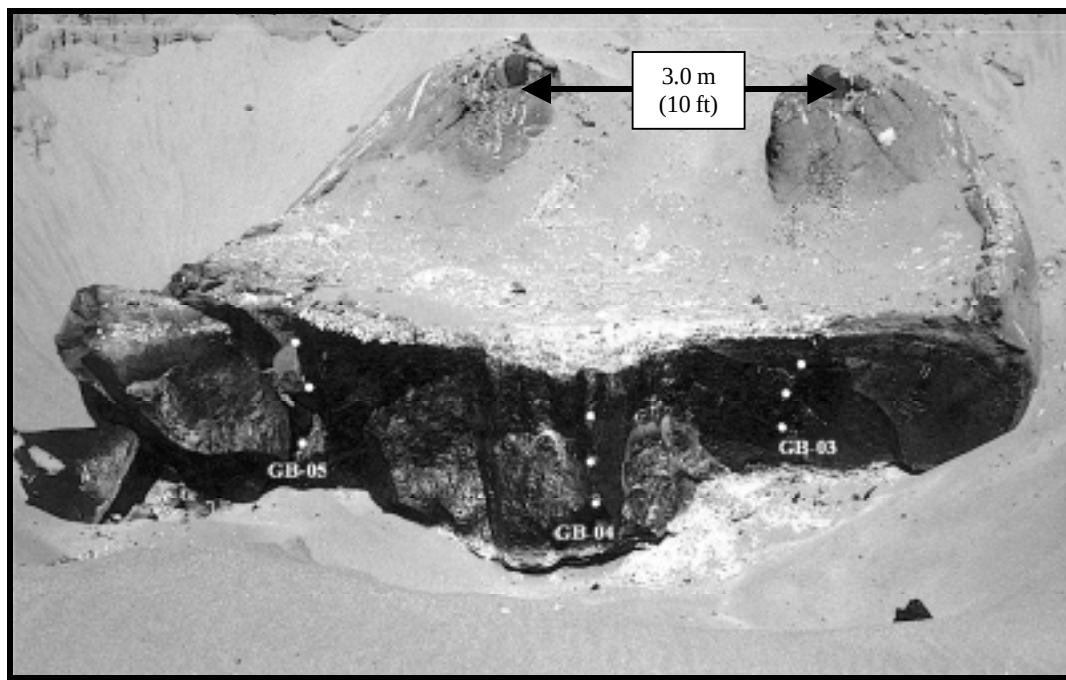


Figure 5. Cross-Section of the ISV Monolith from the Simulated V-Tank Treatability Test (GB-numbers and dots denote various glass sampling locations)

During the fracturing process, it was observed that not all of the simulated V-tank had melted and pooled at the bottom of the melt, during planar-ISV processing. Segments of the tank wall material present in the vitrified monolith are also shown in Figure 5, as light gray regions on either side of the GB-04 sample location. In particular, a crescent-shaped piece of metal can be observed at mid-height in the block, just left of the GB-04 sample location. The thickness of this metal layer was approximately 0.8 cm (5/16 in.) thick, indicating that it had been partially processed (the original thickness of the simulated V-tank was 0.95 cm, or 3/8 in.). The larger slab of metal to the right of the GB-04 sample location (block mid-height) was still 0.95 cm (3/8 in.) thick, however, indicating little reduction in thickness. Both of these metal segments are tightly embedded in the ISV monolith. A significant quantity of fully processed metal was also observed at the bottom of the ISV monolith.

The presence of a significant quantity of fully processed metal in the ISV melt bottom suggests that the metal segments were likely remnants of the final-processed sections of the simulated tank. The original location for these segments was most likely the top portion of the simulated tank, near the intersections of the tank wall and vent pipes. Estimates are that these tank segments would also be processed by the melt, if planar-ISV processing had continued. Furthermore, with the higher melting soils expected during actual remediation of the TAN V-tanks, it is expected that all of the tank metal will pool at the bottom of the ISV melt. Nevertheless, the presence of these incompletely processed metal remnants should not detract from the effectiveness of the planar-ISV remediation process. In particular, it is expected that any fixed or smearable actinide contamination on the interior of the tank would have been

effectively processed and incorporated in the surrounding ISV melt. If necessary, both the interior tank fill material and surrounding soils can also be fluxed with additives to raise the ISV melt temperature.

Samples of the vitrified waste form were then obtained and analyzed for lithium and molybdenum. The samples were taken at 15 locations within the ISV monolith, nine of which are shown in Figure 5. The results of these analyses found a significant uniformity in lithium and molybdenum concentration amongst all of the glass samples, despite lithium and molybdenum being originally placed in discrete locations at the bottom of the simulated V-tank. This demonstrates that the resultant ISV monolith is relatively homogeneous. The homogeneity is a consequence of convective currents in the melt, which tend to redistribute any inhomogeneities in the target waste.

Product consistency testing (PCT) was also performed on portions of five glass samples obtained from this block. The purpose of the PCT analysis was to confirm the high durability of the resultant vitrified waste form. The results of the PCT analysis confirmed that the excellent leach rate and durability properties associated with the ISV waste form are the same for planar-ISV processing as they are for conventional-ISV processing. Samples of the vitrified waste form were also structurally evaluated, in a manner similar to that of the long starter plane test. The results confirm the presence of diopside in the glass.

CESIUM VOLATILITY EVALUATION

As previously stated, the simulated V-tank test included 1,800 g (4 lb) of hydrated cesium hydroxide. The purpose of including cesium in this test was to evaluate its potential volatility during planar-ISV processing. Although previous conventional-ISV melts have shown significantly high retention of cesium in the glass, there were concerns that the different orientation of the planar-ISV process may result in increased cesium volatility. A high level of cesium volatility could become a potential exposure issue during planar-ISV processing of the TAN V-tanks, due to the relatively high quantities of Cs^{137} in the tanks.

To perform the cesium volatility evaluation, various off-gas system samples were analyzed. These included off-gas samples, off-gas condensate samples, off-gas hood and line smears, HEPA filter samples, and zeolite samples. The off-gas samples were taken during planar-ISV processing, while the smear, condensate, and HEPA filter samples were taken immediately after planar-ISV processing, and the zeolite samples were taken after allowing sufficient time for the ISV monolith and surroundings to cool.

The concentration of cesium in the off-gas condensate was 10.9 ppb. The average net concentration of cesium found in the hood smears was 8.4 ppb, while the average net concentration in the off-gas pipe smears was 9.7 ppb. The average concentration of cesium in the HEPA filter was found to be 2.2 ppb. Based on the results of these samples, it is estimated that approximately 154 mg of cesium collected on the off-gas hood, 98 mg of cesium collected on the off-gas line, 86 mg collected in the off-gas condensate, and 5.6 mg collected on the HEPA filter. Based on these results, it is safe to assume that the total amount of Cs^{137} expected to be

volatilized during planar-ISV processing of the TAN V-tanks will be insufficient to raise any exposure concerns, above grade.

As a crosscheck for these calculations, an estimate was made of the total volume of cesium that flowed into the HEPA filter. This estimate was based on the analysis of cesium concentrations in the off-gas flow directly in front of the HEPA filter, during ISV processing. The analysis found an average cesium concentration of 0.003 ppb in the off-gas flow. Using the sampled values, together with the measured off-gas flow rates, it was determined that approximately 10 mg of cesium was carried into the off-gas treatment system during the course of this test. Subtracting the 5.6 mg of cesium that was collected on the HEPA filter, it can be assumed that only 4.4 mg of cesium were not collected by the off-gas. This represents only 0.00025 wt% of the total cesium source term (1,788 g). This includes the cesium from the tank (1,470 g), the estimated amount of cesium processed from soils outside of the tank (313 g), and the total amount of cesium present in the zeolite (5 g).

Table 2 summarizes the results of the cesium condensation/collection inventory in the flow stream between the off-gas hood and exhaust. As indicated in this table, approximately 348 mg of cesium was transported from the soil or tank to these ex-situ locations. Based on the previously described cesium source term of 1,788 g, the ex situ inventory represents approximately 0.02 wt% of the source material. In other words, the planar-ISV process had an effective cesium retention efficiency of at least 99.98 wt% in this application. The amount retained by the zeolite material is included in this retention efficiency, since the cesium captured in the zeolite could be collected for subsequent treatment, or treated in place with a follow-on ISV process.

Analysis of the zeolite samples found that the zeolite material in the vent pipes collected a total of 25.6 mg of cesium. The primary source term for this cesium (1470 g) included the 1425 g of cesium that was staged in the tank (as $\text{CsOH} \cdot \text{H}_2\text{O}$) and the 45 g of cesium that was present in the tank fill and "heel sand" materials that were also in the tank. Based on this data, the cesium collected in the vent pipes represented approximately 0.0017 wt% of the total cesium source term. Furthermore, the only cesium that was found in the zeolite was restricted to the bottom 2-ft of the vent pipe columns. The excellent cesium capture exhibited by the zeolite suggests that much of the "ex situ" cesium came from the background cesium in the soils, rather than the tank itself. Assuming this is true, the cesium retention efficiency **for the tank contents** would be 99.996 wt%.

The cesium retention efficiency value of 99.98-99.996 wt% is consistent with values of 99.23-99.996 wt% obtained in previous tests involving conventional top-down ISV of treatment zones containing cesium (3,4,5,6,7,8). The only existing ISV data on cesium retention that is inconsistent with these results is a test that was performed for Oak Ridge National Laboratory (ORNL) in 1991. During this test, a cesium retention efficiency of only 97.6 wt% was obtained. Upon subsequent investigation, it was found that the low retention efficiency was due to cesium metal reacting with chlorides in the melt, which produced a more volatile cesium chloride compound than was

Table 2. Post-Test “Ex Situ” Cesium Inventory

Off-Gas Collection Site	Cesium Loading (mg)
Hood Surfaces	154
Off-Gas Pipe Surfaces	98
Off-Gas Condensate	86
HEPA Filter	5.6
Uncaptured Cesium	4.4
Total	348

originally planned (9). Note that the cesium hydroxide used in this test was in its hydroxide form, which is even more volatile than cesium chloride (10). Therefore, it can be safely assumed that cesium volatility will not be a concern during planar-ISV processing of the TAN V-tanks, no matter what form of cesium is present in the tank.

In addition, samples of both the vitrified waste form and the metal sublayer were analyzed for cesium. The results of these analyses confirm that cesium is present in the glass (at levels consistent with high cesium retention efficiency), and not in the metal pool at the bottom of the ISV monolith.

CONCLUSIONS AND APPLICATION

The results of the planar-ISV treatability study confirm the ability of planar-ISV to safely and effectively remediate the subsurface PCB-contaminated V-tanks at TAN. Planar-ISV is effective in eliminating concerns over melt expulsion events during ISV processing of subsurface tanks, by ensuring a continuous, relatively-permeable path for vapors to be transported to the ISV off-gas hood. The subsurface nature of the planar-ISV process also minimizes secondary waste volume, due to reduced particulate entrainment in the ISV off-gas system. Some of the secondary wastes generated during planar-ISV processing may be further segregated into radioactive and hazardous secondary wastes, due to the partitioning nature of the various volatile and semivolatile contaminants that are released during processing of the TAN V-tanks, reducing the total amount of secondary mixed-waste that is generated. In addition, use of planar-ISV appear to have similar cesium retention efficiencies and waste form product durability as that demonstrated with conventional-ISV processing, as well as potentially lower energy consumption requirements. Finally, the long starter plane test, in combination with the simulated V-tank test, demonstrated that planar-ISV can be effectively applied to subsurface tanks with dimensions and depths similar to that of the TAN V-tanks.

Based on the results of the planar-ISV treatability study, in combination with existing ISV data, planar-ISV has been selected as the preferred technology for remediating the PCB-contaminated tanks at TAN (11). The primary reason for this selection is that planar-ISV meets

all of the potential Applicable, Relevant, and Appropriate Requirements (ARARs) associated with cleanup of the PCB-contaminated tanks. Current efforts are now underway to reach a final Record of Decision (ROD) for WAG 1 cleanup by the end of FY-1999.

As part of Geosafe's final report on the planar-ISV treatability study, an applicability analysis section has been prepared, outlining a possible plan for remediating the TAN V-tanks (12). The proposed plan would be to set-up a series of four ISV starter planes around tanks V-1, V-2, and V-3. The starter planes would be put in place after trenching around and electrically isolating each tank from the surrounding tanks and buildings. The trenches would then be filled with soil (of desired composition), and vent pipes would be placed over the tanks. The liquid contents of the tanks would then be equilibrated, and the tanks would be filled with soil or other fill material of desired composition. A large ISV off-gas collection hood would then be placed over all three of the large V-tanks, and connected to the ISV offgas treatment system.

Planar-ISV would then be initiated by first processing the center V-tank (tank V-2) with the innermost ISV starter planes, then switching to the outermost starter planes, and processing tanks V-1, and V-3 concurrently. During planar-ISV processing, engineered methods would be used to prevent damage to adjacent buildings and structures, if necessary. Because of its significantly reduced size and slight separation from the other V-tanks, tank V-9 can be planar-ISV processed separately. The total estimated cost for ISV processing of the TAN V-tanks, per the proposed applicability analysis, is \$4.3 M. This does not include the costs associated with identification, management, and control of the ROD at the INEEL.

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