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VITRIFICATION PROCESS

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ABSTRACT

Results obtained from application of two novel approaches for extending the ultimate treatment depth attainable with in-situ vitrification (ISV) are presented. In the first, the electrode design is modified to concentrate the Joule heat energy delivered to the soil/waste matrix in the lower region of the target melt zone. This electrode design has been dubbed the "hot-tip" electrode. Results obtained from both computational and experimental investigations of this design concept indicate that some benefit toward ISV depth enhancement was realized with these hot-tip electrodes.

A second, alternative approach to extending process depth with ISV involves initiating the melt at depth and propagating it in either vertical direction (e.g., downward, upward, or both) to treat the target waste zone. A series of engineering-scale experiments have been conducted to assess the benefits of this approach. The results from these tests indicate that ISV may be effectively initiated and sustained using this subsurface start-up technique. A survey of these experiments and the associated results are presented herein, together with brief discussion of some considerations regarding setup and implementation of this subsurface start-up technique.

INTRODUCTION

In-situ vitrification (ISV) is a process by which electrical energy is supplied to a soil/waste matrix. The resulting Joule heat raises the temperature of the soil/waste matrix, producing a pool of molten soil. The ISV process was invented in the early 1980s by researchers at Pacific Northwest Laboratory (PNL) for the U.S. Department of Energy (DOE) and was patented in 1983 (Brouns et al. 1983). It has been licensed by DOE to Battelle Memorial Institute which has granted exclusive sublicense to the Geosafe Corporation for commercial applications.

Since its inception, there have been many successful applications of the technology to both staged and actual waste sites. However, there has been some difficulty in extending the attainable treatment melt depth to levels greater than 5 m. This difficulty may be the result of characteristics unique to the sites processed or it may be an inherent limitation in the design and application of the currently used ISV systems.

Numerous system modifications have been postulated to facilitate reaching the target melt depth at any given site. In this presentation, two different approaches to address the depth limitation issue are presented: 1) a modification to the standard electrode design and 2) a modification to the standard ISV operating sequence. The design concept for the modified standard electrode holds great promise for achieving target depths while simultaneously requiring minimal alteration to the ISV system and its operational procedures. In this concept, only the lower portion of the electrode supplies current to the melt pool. For the modification to the standard ISV operating sequence, initiation of the ISV process in the subsurface (rather than at the surface) is being evaluated. This concept holds great promise for achieving target depths by initiating the ISV process at depth and allowing the molten pool to grow downward, outward, upward, or a combination of all three directions, depending on the necessary treatment volume. PNL's recent experimental and

modeling experience using these two concepts will be presented.

ENGINEERING-SCALE HOT-TIP ELECTRODE RESULTS

Numerical simulations of the ISV process have been performed using the TEMPEST code (Trent and Eyler, 1993.). In these simulations, we compute the thermo-fluid response of the ISV system by solving a discretized form of coupled partial differential equations that describe the conservation of mass, momentum, energy, and electric charge. Temperature-dependent thermal, electrical, and transport properties included in the model were obtained from Buel et al. (1987). Multi-phase, multi-electrode systems are also explicitly modeled in these simulations.

A typical two-phase, four-electrode engineering-scale ISV system was modeled in these simulations. In this configuration, a Scott-Tee power supply was used to provide two-phase power (with 90° phase separation) to the melt through four 5-cm-dia graphite electrodes. These electrodes were placed in the soil at the vertices of a square pattern. Each diagonally opposed pair of electrodes receives power from one phase of the Scott-Tee system. Center-to-center spacing for these electrode pairs was 0.43 m. A nominal total power level of 25 kW (split equally between phases) was modeled. For the base-case electrode configuration, the entire length of the electrode supplied current to the melt. In the hot-tip electrode configuration, however, the upper portion of the electrode was electrically insulated from the melt. Current supply to the melt was therefore restricted to the lower portion of the electrode.

Figure 1 illustrates the isotherms on a vertical plane cut through the melt when the system is at a melt depth of 1 m. This plane also passes through two adjoining electrodes of the engineering-scale setup (NOT the diagonally opposed electrode pair, however). These isotherms indicate the temperature distribution in the melt in this state. Both the base case (Figure 1A) and hot-tip (Figure

1B) results are presented in this figure. A hot-tip length of 27 cm was assumed in these "case B" results. Therefore, the hot-tip length represents approximately 27% of the melt depth below grade at this point in the process. As indicated in this figure, the hot-tip electrodes effectively lower the hot zone of the melt to the lower region of the melt's depth. This has the beneficial effect of increasing the heat flux out the bottom of the melt relative to the base-case configuration results. Since the melt growth rate is proportional to the local heat flux, this increase in heat flux out the bottom of the melt effectively increases the melt's downward growth rate. As a consequence, downward progression of the melt is enhanced by the presence of these hot-tip electrodes. The net effect of these hot-tip electrodes is to reduce the time required to achieve a specified melt depth. For example, in this engineering-scale configuration with a hot-tip length of 27 cm, the simulations indicate that a 1-m melt depth will be attained in 24 hr (or approximately 30% less time than with standard electrodes). The hot-tip electrodes should also serve to increase the ultimate depth attainable with ISV, since the energy is more efficiently distributed throughout the melt zone.

A series of two engineering-scale experiments were performed in collaboration with the Geosafe Corporation in November, 1993 to assess the efficacy of the hot-tip electrode concept. The first of these tests used the standard electrode and engineering-scale system described in the modeling discussion above. This test served as a baseline against which to compare the results obtained in follow-on hot-tip electrode tests. A steady-state power level of approximately 26 kW was maintained throughout much of this test. Each test was performed in sandy soil (similar to that found on the Hanford site in Washington state). Every effort was made to reproduce the conditions from one test to the other except for the electrode designs, which differed between tests. For the second test, a castable, refractory, insulative coating was applied to the electrode segments to produce a hot-tip electrode with a hot-tip length of 30 cm. For the base-case test, the electrodes were set in the soil to an initial depth of 7.5 cm. In the hot-tip test however, the electrodes were inserted to an initial depth of 18 cm. This was done to ensure that the top of the uppermost, coated

electrode segment was below the level of the top plate on the off-gas hood. The insulative coating was sufficiently thick so that it could not extend through the electrode seals of the existing off-gas hood. As will be seen in the electrode depth results, this difference had virtually no effect on the overall electrode depth penetration rate after the initial start-up transient. In both tests, power was ramped up to the 26 kW steady-state level in approximately 2.5 hr. Both tests were heavily instrumented with over 200 type K thermocouples to provide a detailed representation of the melt's growth. A plan view illustrating thermocouple placement employed in each test is presented in Figure 2.

Figure 3 illustrates the electrode depth transients realized in both the base case and hot-tip case tests. Note that the hot-tip electrode case reached the targeted 1-m melt depth in approximately 25 hr. The base-case test reached the targeted 1-m depth in 29 hr. Thus, this hot-tip configuration resulted in a 17% reduction in the time required to reach the target depth. Figure 4 illustrates the location and shape of the 500°C, 300°C, and 200°C isotherms in both the base-case and hot-tip case tests. These isotherms suggest the outline of the melt profile along the 0° axis (reference coordinate system presented in Figure 2) at 25 hr after the start of each test. As these contours indicate, the hot-tip electrodes provide a deeper, narrower melt shape than the base case. This shape is another manifestation of the concentration of Joule heat in the lower regions of the melt that is realized with the hot-tip electrodes.

The base-case test consumed 749.1 kWh to produce a 727-kg block. This corresponds to a specific energy consumption of 1.03 kWh/kg. The hot-tip test consumed 664.8 kWh and produced a block of 636 kg, corresponding to a specific energy consumption of 1.045 kWh/kg. These values are consistent with the nominal 1 kWh/kg specific energy consumption typical of engineering-scale melts in sandy soils like those found at the Hanford site in Washington state. The slightly higher specific energy consumption of the hot-tip case may be due to differences in

heat loss out the top surface of the melt.

Future tests will investigate the effect of hot-tip length and electrode design on the melt rate.

Pending the results of these tests, additional study is are planned to assess the impact of scale (viz., Engineering vs. Pilot & Large Scale systems) on the performance of these electrodes.

INITIATION OF THE ISV PROCESS AT DEPTH

The standard ISV process is initiated at the surface and propagated downward and outward until a target volume of a soil/waste matrix has been treated. To initiate the process, starter material (a mixture of graphite and glass frit) is placed in linear trenches between an array of four electrodes. This starter path connects diagonally opposed electrodes as well as adjacent electrodes. A starter path is necessary because soil is not sufficiently electrically conductive until it becomes molten. Once the soil/waste matrix becomes molten from the heated starter path, ions in the molten soil (such as sodium and potassium) become conductors for the supplied electrical power.

For the initiation of the ISV process at a target depth, the key step is the creation of a conductive region between the electrode array that is sufficient for starting the vitrification process. Multiple techniques evaluated included such technologies as directional drilling, direct injection, pneumatic fracturing, and hydraulic fracturing. Each of these techniques could be used to place starter material at a target depth. However, during evaluation of these different techniques, it became apparent that the two fundamental questions exist for the application of ISV in the subsurface are: 1) What starter path geometries are suitable for initiating the ISV process? and 2) Can the ISV process be initiated in the subsurface and propagated in a downward direction?

To answer the first question, a series of engineering-scale ISV tests were performed with different

starter-path geometries. Three-electrode and four-electrode configurations were investigated for both a planar and a tubular starter-path arrangement. Tests performed with a planar starter-path geometry utilized a 1-cm thick layer of ISV starter material that encompassed the entire electrode array. The starter-tube arrangement for the four-electrode test connected only diagonally-opposed electrode pairs. In the three-electrode test with tubular starter paths, however, all electrode pairs were connected with the starter tubes. The results of these tests indicate that all of the different starter-path geometries were suitable for initiating the ISV process. Based on the results of these engineering-scale tests, it was not possible to eliminate any potential methods for creating a conductive region between an electrode array based on the resulting geometry formed.

To answer the second question related to initiating the ISV process at a target depth, an engineering-scale test was performed in which the process was started at depth. This test was the second engineering-scale test performed in the soil subsurface (Tixier, Stottlemire, and Murphy 1991) and the first to explore the propagation of the ISV melt. Previous work was focused on the establishment of a vitrified barrier in the soil subsurface. Figures 5 and 6 illustrate the engineering-scale test layout plan and elevation views, respectively. (For a complete description of PNL's engineering-scale unit and the three-electrode power supply system, see Luey et al., 1992). A planar starter path 85 cm in diameter and approximately 1 cm thick was used to initiate the ISV process. The starter path was buried at a depth of 60 cm below grade. Stove pipes (15-cm-dia) were used to simulate well casings for the 5-cm-dia electrodes as well as a central vent pipe. A total of 19 type K thermocouples were used to monitor temperatures above, below, and to the side of the vitrification zone. The thermocouple array above the starter path was suspended from above the soil surface to ensure a constant position. The lateral thermocouple arrays and arrays located below the starter path were staged as the engineering-scale unit was backfilled with soil.

The engineering-scale test was conducted in the first half of 1994. Power was terminated after

22.8 hr of operation when the shutdown criterion was reached (i.e., the type K thermocouple at a depth of 120 cm reached a temperature of 1200°C). Figure 7 illustrates the power time-history for the engineering-scale test. During the start-up phase of the test, the power supplied to the soil was increased at a rate of 1 kW every 6 min until the target power level of 30 kW was achieved. Figure 7 also illustrates the total energy consumption for this test. Approximately 645 kWh were used in this test to produce a final block weighing 840 kg. This results in a specific energy consumption of 0.77 kWh/kg.

Figure 8 illustrates the average melt rate downward, as determined from the insertion depths of the electrodes. Between hours 3 and 5, the melt rate was nearly 7.5 cm/hr, which is higher than the normal 2.5 cm/hr rate seen for the remainder of the engineering-scale test. From experience gained with the engineering-scale tests investigating starter-path geometries, this period of higher than normal melt rate can be attributed to the fact that the molten soil pool begins to form around the electrodes rather than uniformly throughout the starter-path region. This is further supported by the fact that the rate at which the temperature gradient propagated downward along the centerline thermocouple array lagged the insertion rate of the electrodes.

Figure 9 shows the centerline thermocouple temperatures below the starter path. In this figure, the thermocouple type and location are identified with a 12-character tag. The first three letters of this tag (e.g., TCK) indicate thermocouple type (in this case, type K). The following nine numbers indicate the thermocouple's depth, radial, and azimuthal location (i.e., zzzrrrθθθ in units of cm, cm, and degrees, respectively) relative to the soil surface ($z = 0$ cm) and horizontal coordinate system indicated in Figure 5. At 7 hr after startup, the thermocouple at a depth of 65 cm (TCK065000000) reached 1200°C, indicating that this region was beginning to fuse (or melt). At this time, the average electrode depth was 82.5 cm. From these data, it appears that the molten

pool became a continuous region between the electrode array at approximately run time hour 8. As the test progressed, the centerline temperatures began to catch up to the depths of the electrodes. At the time of shutdown, the centerline thermocouple at 120 cm was at 1200°C, with the average electrode depth at 120 cm.

Figure 10 shows the centerline thermocouple temperatures above the starter path (the thermocouple tags are as described in the preceding paragraph). This array was suspended from above grade within the engineering-scale unit to ensure a constant position. It was also placed slightly off center to avoid any influence from and interference with the central vent pipe. From the temperature profile, it appears that the thermocouple at 45 cm (TCK045012270) remained just above the molten soil pool throughout the test. The sudden downward spikes seen in Figure 10 can be attributed to subsidence events, in which soil overburden sloughed onto the surface of the molten pool. As this relatively cooler soil moved downward, it affected the temperature profile being recorded by the thermocouple. Subsidence during the engineering-scale test was very pronounced and needs to be addressed for field-testing of the subsurface ISV process concept. This will ensure that, in actual applications, the off-gas hood is sufficiently sized to envelop the top surface of the melt zone.

These results demonstrate that the ISV process can be initiated from a subsurface starter path. They also confirm the utility of using a planar starter path (as opposed to a standard starter path, which consists of discrete, linear starter tube paths between electrodes). In addition, test results show that the process can be propagated downward and has a very good specific energy consumption ratio (0.77 kWh/kg vs. approximately 1 kWh/kg as previously reported). Based on the results from the engineering-scale tests, further study is warranted for evaluating subsurface initiation of the ISV process. However, observations during the engineering-scale test identify soil subsidence as an issue that needs to be addressed in advance of field-testing of the subsurface-

started ISV process.

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(a)

(b)

Figure 1. In-situ vitrification melt isotherms from TEMPEST simulations of engineering-scale systems employing: (a) standard and (b) 27-cm hot-tip electrodes.

⊗ - Arrays of 5 type K thermocouples spaced 20 cm apart (80-cm array length)

X - Arrays of 10 type K thermocouples spaced 10 cm apart (90-cm array length)

Figure 2. PNL/Geosafe engineering-scale in-situ vitrification test thermocouple layout schematic – plan view.

Figure 3. PNL/Geosafe engineering-scale in-situ vitrification electrode depth transients – standard vs. 30-cm hot-tip electrode configuration results.

Figure 4. Base case vs. hot-tip isotherms along 0° axis at t = 25 hr.

Figure 5. Initiation at depth, engineering-scale test layout – plan view.

Figure 6. Initiation at depth, engineering-scale test layout – elevation view.

Figure 7. Power and energy consumption for initiation at depth, engineering-scale test.

Figure 8. Average melt rate for initiation at depth, engineering-scale test.

Figure 9. Temperature profile for centerline thermocouples below starter path.

Figure 10. Temperature profile for centerline thermocouples above starter path.

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