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IN SITU VITRIFICATION: TECHNOLOGY STATUS AND A SURVEY OF NEW APPLICATIONS

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IN SITU VITRIFICATION: TECHNOLOGY STATUS AND A SURVEY OF NEW APPLICATIONS

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ABSTRACT

Pacific Northwest Laboratory (PNL) is developing a thermal treatment process called in situ vitrification (ISV) for remediating contaminated soils, underground structures such as tanks, and buried wastes. ISV was initially developed for contaminated soil applications in 1980 and has since become a mature technology for these applications. Relatively new applications of ISV to underground structures and buried wastes are currently in the development stages.

This paper will outline the development progress of the ISV technology, including the results of demonstrations and other field-scale testing performed to date, and examine the key remaining issues associated with new ISV applications. Progress on issues attendant to waste form performance and economics will be addressed.

INTRODUCTION

In situ vitrification (ISV) is a waste remediation technology currently under development by the U.S. Department of Energy (DOE) for application to radioactive and mixed waste contaminated soils. The process was developed by researchers at DOE's Pacific Northwest Laboratory (PNL).[a] Invented in 1980, the process has been developed primarily at PNL.[1] However, collaborative development efforts have been undertaken in recent years between PNL, Oak Ridge National Laboratory (ORNL) and the Idaho National Engineering Laboratory (INEL). Geosafe Corporation, the only commercial supplier for ISV remedial services, has typically been involved in the DOE development program.

Within the context of DOE, the primary development focus on ISV is to resolve the few remaining technical issues for application to contaminated soils. Secondary development efforts involve advanced applications to buried waste and underground storage tanks. However, the development for buried waste and tanks is being deemphasized until the remaining issues associated with contaminated soils applications are resolved. Once the technology is judged ready for substantive deployment on mixed and radioactive waste sites within the DOE complex, the development of advanced applications is expected to be pursued.

ISV Process Description

In situ vitrification has been developed as a remedial action process for soils contaminated with hazardous chemical wastes and/or radionuclides. Figure 1 illustrates the operation of the ISV process. An array of graphite electrodes is inserted a few inches into the ground. Because soil is not electrically conductive when its moisture has been driven off, a conductive mixture of flaked graphite and glass frit is placed between each electrode to serve as a starter path. An electrical potential is applied to the electrodes to establish an electrical current in the starter path. The flow of current heats the starter path and surrounding soil to well above the initial soil-melting temperatures of 1100° to 1400°C. The graphite starter path is eventually consumed by oxidation, and the current is transferred to the molten soil, which is processed at temperatures between 1450° and 2000°C. As the melt grows in size, nonvolatile radionuclides and inorganics are incorporated into the molten soil. The high temperature of the process destroys organic components by pyrolysis. The pyrolyzed byproducts migrate to the surface of the vitrified zone, where they combust in the presence of air. A hood placed over the area being vitrified directs the gaseous effluents to an off-gas treatment system. Upon cooling, the solidified glass and crystalline monolith is estimated to be

(a) The Pacific Northwest Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

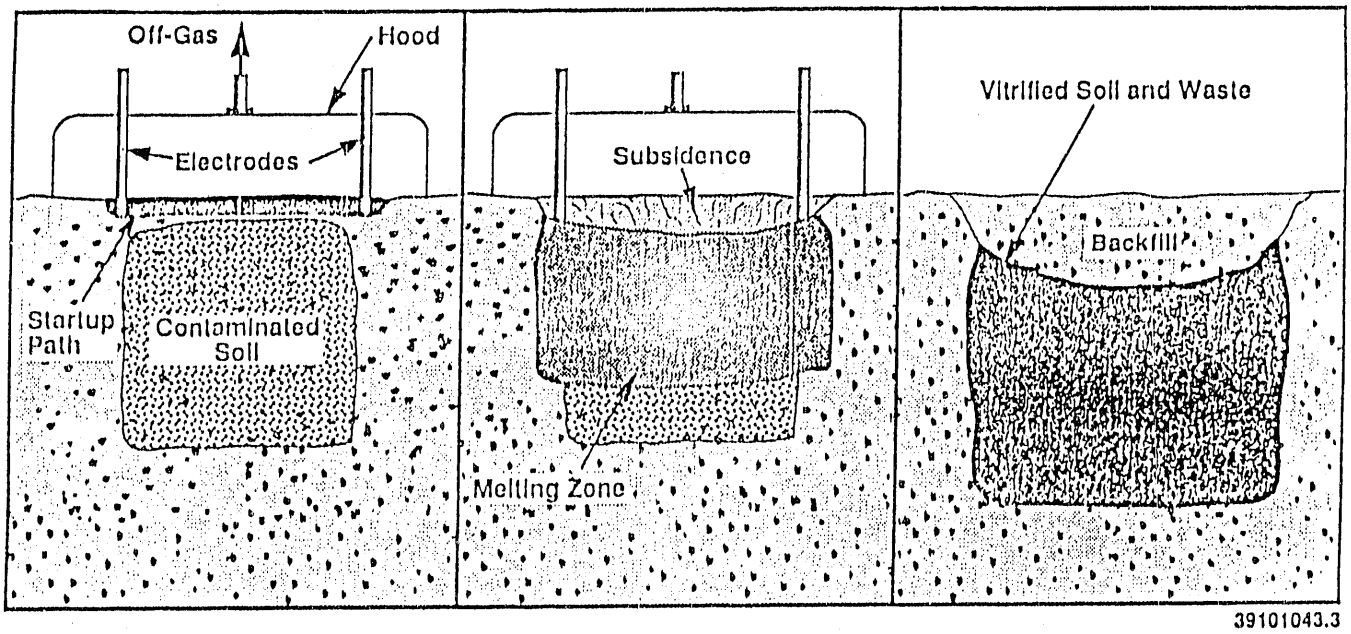


Figure 1. ISV process sequence.

stable for geologic periods and is highly resistant to leaching. For expansive contaminated areas, adjacent settings of the process result in the formation of a single contiguous monolith.

An electrode feed system allows the vertical position of the electrodes to be controlled as the melt progresses. Although the electrodes generally rest on the bottom of the melt, the electrodes can be retracted if desired. For example, retraction of electrodes a few centimeters from the bottom of the melt is often necessary during operations involving buried metal since molten metal pools at the base of the melt can short-circuit the power delivery system.

ISV Equipment Description

Three scales of processing equipment have been developed to date. Engineering-scale testing conducted in the laboratory typically operates at power levels between 15 to 20 kW and produces ISV blocks as large as 1000 kg. The equipment is designed with significant flexibility to allow testing of alternative electrical and process equipment configurations as well as to develop new operating techniques. Essentially all initial development testing is conducted in the laboratory at engineering-scale.

The DOE's pilot-scale equipment, operated by PNL, is depicted in Figure 2 and consists of a complete processing system in a single semi-trailer including a 500 kW transformer, off-gas treatment system, and process controls. A separate off-gas confinement hood is placed over the area being treated. The pilot-scale system can produce blocks as large as 50 tons, achieving melt depths of 2 to 3 m. At approximately 3/8 the size of a full-scale system, the pilot-scale system is ideally suited for acquiring near full-scale performance field data.

The large-scale system, considered full scale, uses a 3.75 MW power conditioning system to produce a vitrified soil monolith of 700 to 900 tons per setting. The equipment is mounted on 3 semi-trailers, one each for power conditioning and process cooling, off-gas treatment, and process control and data acquisition. The large-scale system includes many redundant and back-up safety systems for radioactive applications, such as a double containment glovebox around the off-gas treatment system. Geosafe Corporation owns and operates a similar system designed for soils contaminated with only hazardous wastes.

SURVEY OF APPLICATIONS

Development efforts during the 1990s have included radioactive and mixed waste contaminated soils, buried wastes, and underground storage tanks for DOE; but the primary development focus at this time is contaminated soils. The results summarized in this paper represent the work of primarily PNL, ORNL, and INEL. Although PNL has been involved in the development of all applications, the Laboratory has collaborated with INEL since 1988 to adapt the ISV technology to radioactively contaminated buried

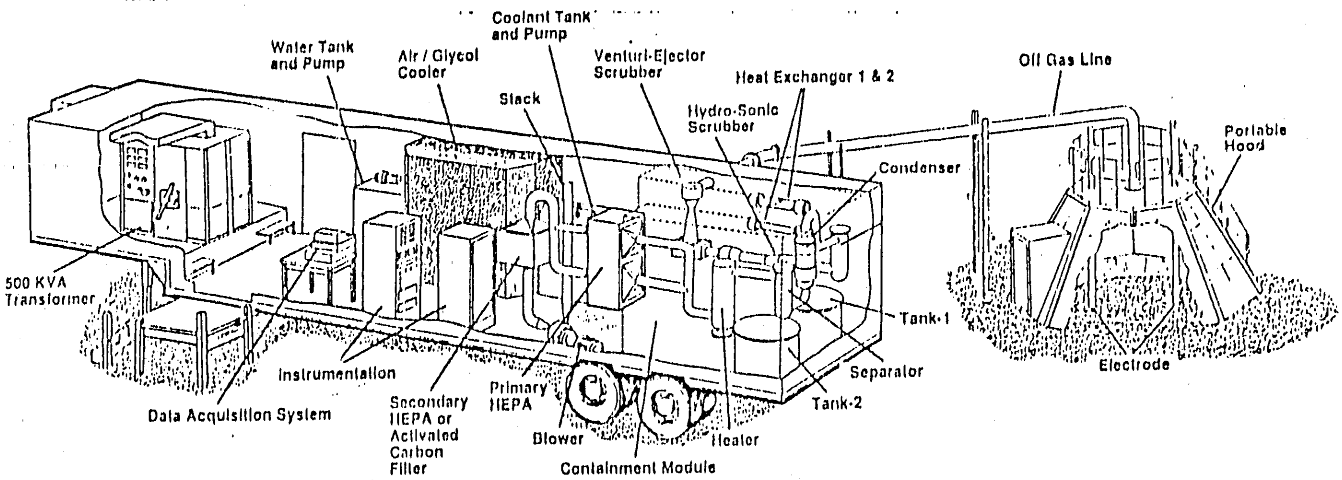


Figure 2. Pilot-scale ISV system.

wastes. Since 1986, PNL and ORNL have been jointly developing the ISV technology for applications to cesium contaminated seepage trenches at ORNL. Development for underground storage tank applications has been exclusively conducted by PNL since 1989.

In addition to the three applications described above, PNL has been investigating the feasibility and merit of using ISV to generate vitrified underground barriers. Research to date has been limited to engineering-scale testing and computer modelling to evaluate the potential for mechanical stress cracking in the barrier upon cooling. Under private contract with a research agency representing foreign corporations, Battelle has begun evaluating ISV for both vitrified barrier and civil engineering construction applications.

Contaminated Soils

Field testing during 1990 and 1991 included a large-scale test of a mixed waste disposal crib at the Hanford Site in Washington State and a radioactive pilot-scale test of a simulated liquid waste disposal trench at ORNL in Tennessee.

Mixed Waste Disposal Crib. The 116-B-6A crib site was vitrified in April 1990. The waste site is located in the 100-BC area on the Hanford Site near the Columbia River. Between 1951 and 1968, the crib received about 5000 L of decontamination wastes from the 111-Building. The site was retired in 1968. The estimated inventory of radioactive material included 900 mCi of ^{90}Sr and 150 mCi of ^{137}Cs. Small amounts of ^{60}Co and ^{239}Pu were also present. The crib also contained chrome, lead, and other hazardous constituents. The 3.6-m-square, 2.4-m-high crib was constructed of wooden timbers. The crib was then backfilled with rocks and covered by 1.8 m of fill dirt.[3]

The ISV demonstration at the crib site was significant because it was the first large-scale test of a mixed waste site. In addition, the site included a significant amount of combustibles that was considered a potential challenge to the off-gas treatment system relative to off-gas generation rate and heat loading. The equipment configuration included stationary electrodes installed to the target melt depth since electrode feeding had not been developed for full-scale applications at that time.

The demonstration occurred over a 12 day period in April 1990, achieving an 87% on-line operating efficiency and produced a block of approximately 800 t. A melt depth of 4.25 m was achieved, which was equivalent to the bottom of the crib; however, the target melt depth had been 6 to 7 m. No significant processing problems were encountered although three electrodes failed due to air oxidation and had to be replaced during the test. Post-test sampling and analyses indicate that the product is comparable to previous ISV products relative to leach resistance. Product samples taken from cores in several locations revealed a uniform composition.

This large-scale test brought attention to the problem of obtaining target melt depth. As a results, studies were initiated to identify the factors controlling melt growth, and engineering solutions are currently being evaluated both numerically and in engineering-scale tests to significantly enhance melt depths.

Simulated Liquid Waste Disposal Trench. In May 1991, a pilot-scale demonstration was conducted at ORNL on a simulated disposal trench. The 1/4 scale trench contained 10 mCi of ^{137}Cs and was designed to simulate the trenches at ORNL, many of which contain thousands of curries of ^{137}Cs and ^{90}Sr. The test was conducted over a 127 h period and achieved a melt depth of approximately 2.6 m satisfying expectations for the pilot-scale test. Key objectives and results included the following:

- test a particulate prefilter system installed on the off-gas line to determine its effectiveness in preventing cesium from entering the off-gas treatment system

  Between 97 wt% and 99 wt% of cesium was retained in the melt. However, the 1 wt% to 3 wt% that escaped could cause high radiation dosages to workers and create highly radioactive secondary liquid waste if the cesium were allowed to collect in the off-gas treatment system. During the test, approximately 97.3% of the cesium was retained in the melt, however, the off-gas line prefilter was effective since no measurable amounts of cesium were detected downstream of the prefilter.

- determine the fate of cesium relative to potential contamination of surrounding soil

  Posttest sampling and analyses determined that no surrounding soil became contaminated during processing. This finding is consistent with previous engineering- and pilot-scale tests.

- evaluate seismic imaging as a nondestructive geophysical monitoring method to monitor the shape progression of the melt

  The imaging method involves firing a seisgun into the ground at prescribed locations and then detecting the resulting compression waves in an array of passive geophones and hydrophones. Collected data are currently being evaluated. Future developments may lead to a seismic imaging system that would provide operators an on-line measurement capability to help ensure an acceptable melt depth and shape is achieved.

Additional posttest analyses of the results of this pilot-scale demonstration are underway. Comprehensive results will be reported in late 1992.

Buried Wastes

PNL and INEL have been jointly developing the ISV technology for application to buried waste since 1988. The joint effort culminated in two pilot-scale tests at INEL in June and July 1990.[4] The tests were designed to simulate typical radioactive and mixed buried wastes. Since testing was at the pilot scale, the waste packages were scaled accordingly. The 55-gal drums and 1.2-m x 1.2-m x 2.4-m burial boxes were represented by 9.5-L cans and 2-ft x 2-ft x 3-ft cardboard boxes, respectively. Test 1 involved a random dump arrangement of cans and boxes, while test 2 represented a stacked arrangement of cans and boxes. The stacked arrangement represented a more significant challenge for the ISV process since the waste constituted approximately 18 wt% of the total test pit. In both test 1 and 2, the contents of the cans and boxes were typical of common buried wastes and included paper, cloth rags, metal, glass, and sludge. Hazardous or radioactive materials were not used in the test; however, rare earth tracers were used as a surrogate for plutonium.

Conclusions from the work include the following:

- Based on MCC-1 leach testing, the durability of the ISV product was comparable to obsidian and granite and 4 to 10 times more durable than a typical high-level nuclear waste glass.

- Posttest observations revealed there was little potential for creating an underground fire when processing these types and configurations of waste materials. For example, unburned paper was found within a few centimeters of the melt edge. Note, however, that the presence of oxiders in buried waste could support subsurface combustion; and this aspect will be investigated further in subsequent tests.

- When processing through sealed containers, particularly in test 1, sudden gas releases from the cans caused the hood to momentarily pressurize.

  Normally the containment hood is operated at a slight vacuum to prevent contaminant leakage from the hood to the environs. In actual applications on radioactively contaminated buried wastes, the transient gas surges to the hood could result in momentary releases of contaminants. As a result of these tests, PNL has been developing engineered systems to accommodate these surges. It is expected that most containers in landfills are of low integrity due to damage sustained during burial or from corrosion once buried.[5] Low integrity containers would not be expected to retain appreciable quantities of gas at pressure. However, the possibility of transient gas releases is a key issue requiring further study.

Underground Storage Tanks

From 1989 to 1991, PNL's ISV development efforts have included underground storage tank applications. These development efforts are largely represented by two field tests: a pilot-scale test of a 720 L steel and concrete tank in September 1990, and a large-scale test of a 22,700 L tank in July 1991.

Pilot-Scale Tank Test. The pilot-scale test involved a steel and concrete tank containing a simulated mixed waste sludge.[6] The recipe for the sludge was developed by evaluating the tank waste contents of 33 inactive tanks at ORNL. To represent the worst-case sludge, the quantities of chemicals composing the sludge equaled or exceeded concentrations of all key species in the 33 inactive tanks. The composition of the sludge is shown in Table I.

The pilot-scale test was initiated in early September 1990. However, after 19 h of powered operations, the test was halted at a melt depth of approximately 1 m due to a sudden release of gas from the

Table I. Chemicals used for simulated sludge for pilot-scale tank test.

Species	Total Quantity Used (g)	Wt% in Sludge
Chromium III Oxide (Cr_2O_3)	2,315	0.58
Cobalt Oxide (CoO)	420	0.10
Tributyl Phosphate (TBP)	990	0.25
Mercury II Oxide (HgO)	270	0.067
Cesium Nitrate ($CsNO_3$)	270	0.067
Strontium Nitrate [$Sr(No_3)_2$]	675	0.17
Lead (Pb)	4,620	1.16
Nickel (Ni)	1,000	0.25
Barium Oxide (BaO)	3,012	0.76
Cadmium Oxide (CdO)	62	0.016
Hydraulic Oil (Source of TOC)	9,900	2.48
Total	23,534	~5.9
Balance ORNL Soil	375 kg	

waste tank, which pressurized the off-gas containment hood. Up until that time, the tank had been venting gas and vapor through the melt at a regular, controlled rate. The resulting data were evaluated, and the mechanism responsible for the pressurization was identified. To solve the problem, a graphite vent pipe was installed through the vitrified soil to the bottom of the tank, and a radiant heat shield was placed in the hood over the vitrification zone after the melt had solidified and cooled. The test was restarted from grade and completed without incident, and the target melt depth of 2.4 m was achieved.

Toxicity Characteristics Leach Procedure (TCLP) results revealed that both the vitrified soil product and the metal ingot (from melted steel tank and reduction of some oxide species in soil and sludge) passed the leach test criteria as shown in Table II. While the TCLP results were not unexpected for the vitrified soil phase, that the metal phase passed TCLP criteria was largely due to the redox state of the melt. Species such as chromium, barium, and lead from either the sludge or stainless steel of the tank were predominantly oxidized and dissolved in the glass, rather than reduced as a metal. Other more volatile species, including all of the mercury and most of the cadmium, were volatilized from the metal phase and

Table II. TCLP results of glass and metal phases for pilot-scale tank vitrification test.

Species	Glass (ppm)	Metal (ppm)	Allowable Limit (ppm)
Arsenic	0.012	0.01	5
Barium	<0.05	0.08	100
Cadmium	0.27	0.72	1
Chromium	0.27	0.72	1
Lead	0.07	0.14	5
Mercury	<0.0004	0.0004	0.2
Selenium	<0.01	0.01	1
Silver	<0.05	0.05	5

captured in the off-gas treatment system. Relative to surrounding soils, low ppm traces of cadmium were found in one sample in the partial melt zone at the glass-soil interface, however, no other contamination of the surrounding soil was observed.

Large-Scale Tank Test. The large-scale test was initiated in July 1991 and involved a 22,700 L steel and concrete tank backfilled with pumice.[7] The composition of pumice is similar to that of soil. Pumice was used as a melt rate enhancement technique since a smaller amount of energy would be required to melt the smaller and drier mass of pumice relative to soil. The density of pumice (0.65 g/cc) was significantly less than soil (1.45 g/cc) even though the composition is very similar. No hazardous or radioactive materials were used in the test. The bottom 10% of the tank was filled with water saturated soil to represent a wet sludge and enable study of how gas and vapor release from the tank. Also, this test provided opportunity for the first large-scale use of electrode feeding.

The test operated for six days during which very rapid melt rates of up to 15 cm/h were attained. Three electrodes broke during the test. However, once a feeder was realigned and operators gained hands-on operating experience, the electrode feed system worked well for the balance of the test.

The test was halted after six days when a sudden gas release pressurized the containment hood and splattered molten soil on the stainless steel hood. Although sudden gas release events were anticipated, the magnitude of this event was unexpected and damage to the hood resulted in the test being terminated. As in the pilot-scale test, a graphite vent pipe had been installed, but the vent failed approximately 6 h before the event. Gas had been released through the melt at a regular rate even after the vent failed, however, some as yet unknown mechanism resulted in a dramatic increase in the gas release rate that resulted in molten soil being expelled from the melt. Data analyses, modelling, and excavation of the monolith are planned during 1992 to help provide an understanding of mechanisms controlling the event.

Vitrified Underground Barriers

PNL is evaluating the feasibility and merit of using ISV to generate vitrified underground barriers. The vitrified barrier approach may provide a technology option for DOE for both interim remedial actions and long-term applications where isolation is desired. In some instances, waste retrieval may not be economically feasible or technology for safe retrieval may not be available. Therefore, interim measures such as vitrified barriers may be desirable. In situ vitrified barriers could be formed without any significant soil movement and would be virtually impermeable to groundwater flow or waste migration for geologic periods as long as the barrier remained intact. In 1990, PNL initiated a modest development effort including engineering-scale testing and modelling work. In 1992, PNL is currently evaluating additional barrier configurations and processing enhancements via engineering-scale testing and computer modelling. This work is being conducted through a contract with a private research agency representing a consortium of foreign corporations.

Engineering-scale testing in 1990 involved the formation of planar, two-dimensional vertical walls and a subsurface horizontal floor.[8] Testing was conducted to evaluate various methods of controlling wall shape and enhancing the process efficiency. Additionally, a horizontal subsurface vitrified floor was established during engineering-scale testing that indicated the feasibility of such configurations in the field. Again, various methods were used to determine the best means to initiate a subsurface melt. For field application, it is envisioned that electrodes would be installed to depth and directional drilling techniques will be used to place a conductive starter path mixture between the electrodes at the desired depth below the waste.

Modelling work to date has involved a finite element analysis and a more complex creep model to predict the thermally induced mechanical stresses within the block as a result of the cooling process. Results indicate development of a structurally favorable residual stress pattern consisting of compressive stresses at the surface of the cooled block and tensile stresses in the center of the cooled block.[9] Additional work in this area is planned for 1992.

Cost Studies

For DOE applications, only two recent ISV cost studies have been produced. The two studies are part of larger systems studies, one for buried waste applications and the other for Hanford's single-shell tanks.

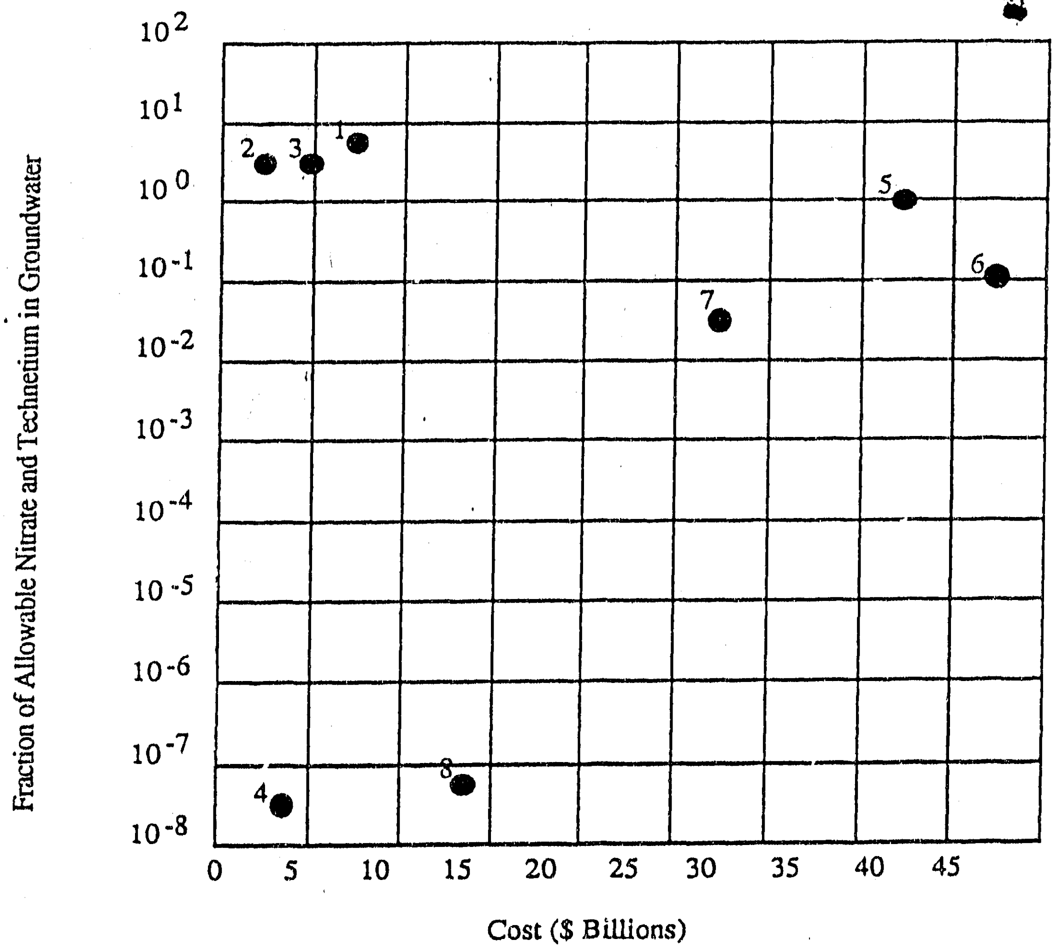
For radioactive and mixed waste contaminated buried waste at the INEL, rough order of magnitude cost estimates have been developed by INEL as part of a System Design Study that evaluated the life-cycle costs for both in situ and ex situ processing options (Mayberry, Quapp reference).[10] The cost estimates include the remaining research and development costs for evaluation and resolution of the remaining technical and engineering issues, design costs, construction and life-cycle operating costs. The lowest cost option involves using ISV as a final treatment, backfilling the site with soil (to fill the subsidence created by volume reduction of the wastes) and planting a vegetative cover. Based on the results of this study, the rough order of magnitude cost of ISV when used as a final treatment is about 16% of the cost for the least-costly ex situ system producing a vitreous waste form. Even if ISV is followed by retrieval of the ISV waste form, the cost is about 25% of the ex situ option. The study identifies an estimate life cycle operations cost for ISV of $648 per cubic yard for this complex application.

A systems engineering study for closure of Hanford's single shell tanks is being conducted by Westinghouse Hanford Company.[11] The study evaluates various remedial action alternatives for the tank wastes, the tank and associated structures, and outlying contaminated soils in those cases involving tanks that have leaked. Many of the options were estimated to cost between $25 and $50 billion. The least costly option (approximately $2 billion) required stabilizing the tanks by filling the dome and installing surface barriers. The solely ISV alternative was estimated to cost about $3 billion; it exceeded the environmental performance of the dome fill/barriers option by several orders of magnitude. Figure 3, adapted from Boomer et al., illustrates that this option and another involving ISV were judged significantly more effective in terms of reducing the potential for long-term environmental consequence by several orders of magnitude and were determined to be less costly by approximately an order of magnitude.

CONCLUSION

In situ vitrification is a significant, viable alternative for treatment of contaminated soils. The cost effectiveness of ISV technology, its applicability to a wide variety of waste types, and its outstanding waste form performance establish ISV as a premier treatment option for DOE. ISV appears ready for many near-term applications in spite of several technical issues yet to be resolved, perhaps most significant is the need to enhance melt depth. Methods to enhance melt depth are key to the success of ISV if the technology is to be broadly deployed within the DOE complex.

Further development will be required before ISV can be considered for applications such as buried wastes and underground storage tanks. However, ISV offers a significant potential treatment alternative from the current baseline remedial method shown in Figure 4. Limited, dangerous, and costly alternatives should ultimately drive further development and use of ISV for these advanced applications.



Legend of Closure Options

Deferred Action Option:
1. Continued storage

In Situ Alternatives:
2. Stabilize with dome fill and barriers
3. Immobilize with grout fill
4. In situ vitrification

Waste Retrieval Options:
5. HLW are vitrified, LLW are grouted, and tank filled with grout
6. HLW are vitrified, LLW are grouted, and tank shell removed
7. HLW conv. to ceramic logs, LLW calcined, dome filled and barriers established
8. Selective retrieval and use of several vitrification technologies including ISV

Figure 3. Costs and relative potential for groundwater contamination of selected closure options for Hanford's single-shell tanks (adapted from Boomer et al., 1991).

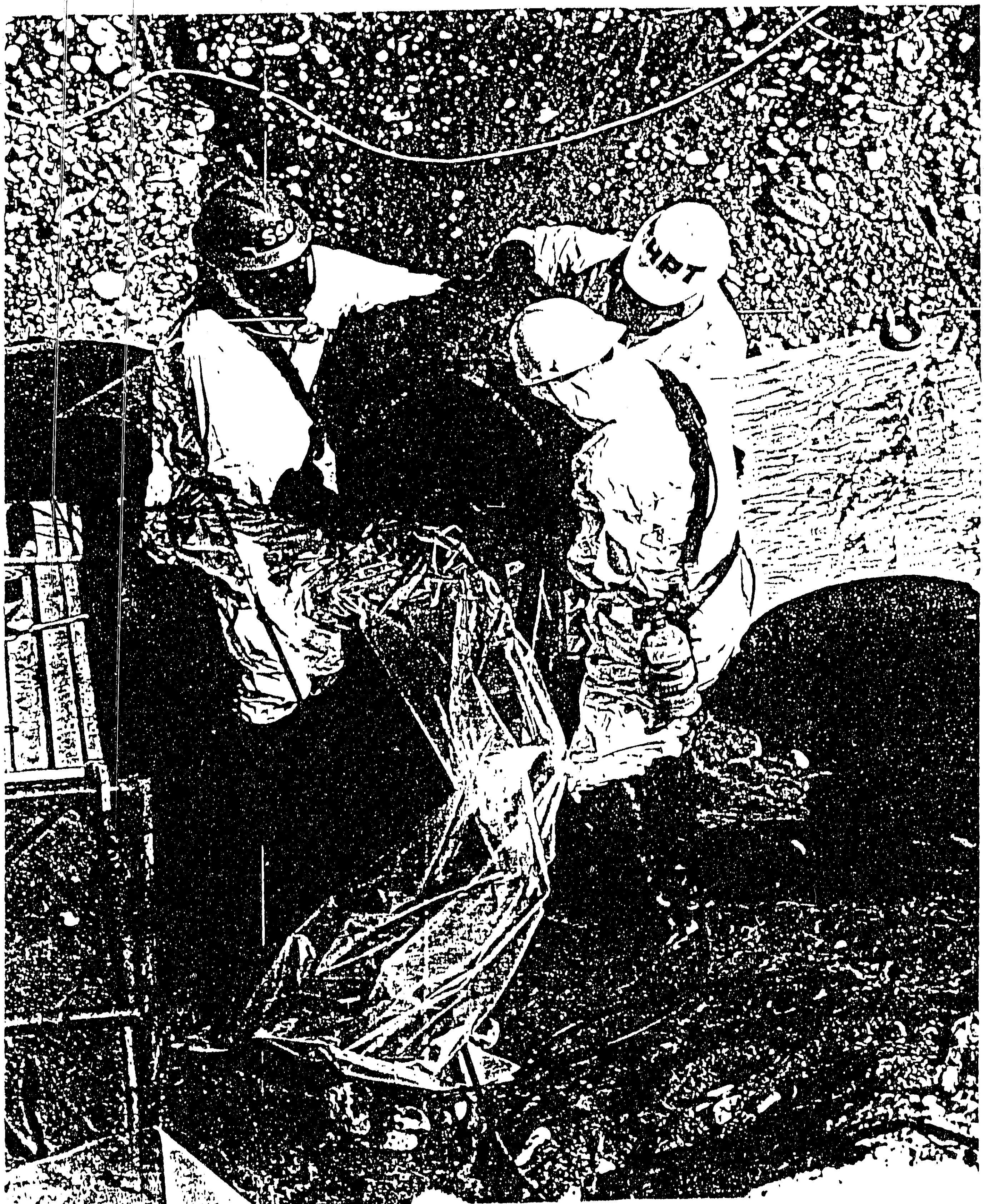


Figure 4. During this buried waste retrieval operation, workers load remains of a corroded drum into a plastic bag. ISV potentially offers a much safer and more cost-effective alternative waste remediation method.

REFERENCES

1. R.A. Brouns, J.L. Buelt and W.F. Bonner, (Battelle Memorial Institute), U.S. Patent 4,376,598 (March 1983).

2. J.L. Buelt, C.L. Timmerman and J.H. Westsik, Jr., In Situ Vitrification: Test Results for a Contaminated Soil-Melting Process, PNL-SA-15767, Supplement 1, Pacific Northwest Laboratory, Richland, 1989.

3. S.S. Koegler, In Situ Vitrification of a Mixed Radioactive and Hazardous Waste Site, PNL-SA-17753S, Pacific Northwest Laboratory, Richland, 1990.

4. R.A. Callow, L.E. Thompson, J.R. Weidner et al., In Situ Vitrification to Buried Waste: Final Report of Intermediate Field Tests at Idaho National Engineering Laboratory, EGG-WTD-9807, EG&G Idaho, Inc., Idaho Falls, 1991.

5. R.D. Mutch, Jr. and J.L. Sutherland, "Estimating the Life of Landfilled Drums," Hazmat World, July:60 (1990).

6. L.E. Thompson, Underground Storage Tank Remediation by Use of In Situ Vitrification, PNL-SA-19889, Pacific Northwest Laboratory, Richland, 1991.

7. S.S. Koegler, R.D. Gibby and L. E. Thompson, In Situ Vitrification of Radioactive Underground Tanks, PNL-SA-19225, Pacific Northwest Laboratory, Richland, 1991.

8. J.S. Tixier, J. A. Stottlemyre and M. T. Murhpy, Vitrified Underground Barrier, PNL-SA-19014, Pacific Northwest Laboratory, Richland, 1991.

9. M.R. Garnich, Thermal Stress Modeling of In Situ Vitrified Barriers for Hazardous Waste Containment, PNL-SA-19347S, Pacific Northwest Laboratory, Richland, 1991.

10. J.L. Mayberry, W.J. Quapp, R. Feizollahi, et al., Preliminary Systems Design Study Assessment Rep. Vol. 1 through 8, EGG-WTD-9594, EG&G Idaho, Inc., Idaho Falls, 1991.

11. K.D. Boomer et al., Systems Engineering Study for the Closure of Single-Shell Tanks, WHC-EPP-0405 (A, Vol. 1), Westinghouse Hanford Company, Richland, 1991.

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