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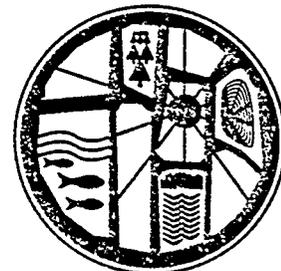
**OAK RIDGE
NATIONAL
LABORATORY**

MARTIN MARIETTA

Demonstrations of Technology for Remediation and Closure of Oak Ridge National Laboratory Waste Disposal Sites

B. P. Spalding
G. K. Jacobs
E. C. Davis

Environmental Sciences Division
Publication No. 3358



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Environmental Sciences Division

DEMONSTRATIONS OF TECHNOLOGY FOR REMEDIATION AND CLOSURE
OF OAK RIDGE NATIONAL LABORATORY WASTE DISPOSAL SITES

B. P. Spalding
G. K. Jacobs
E. C. Davis*

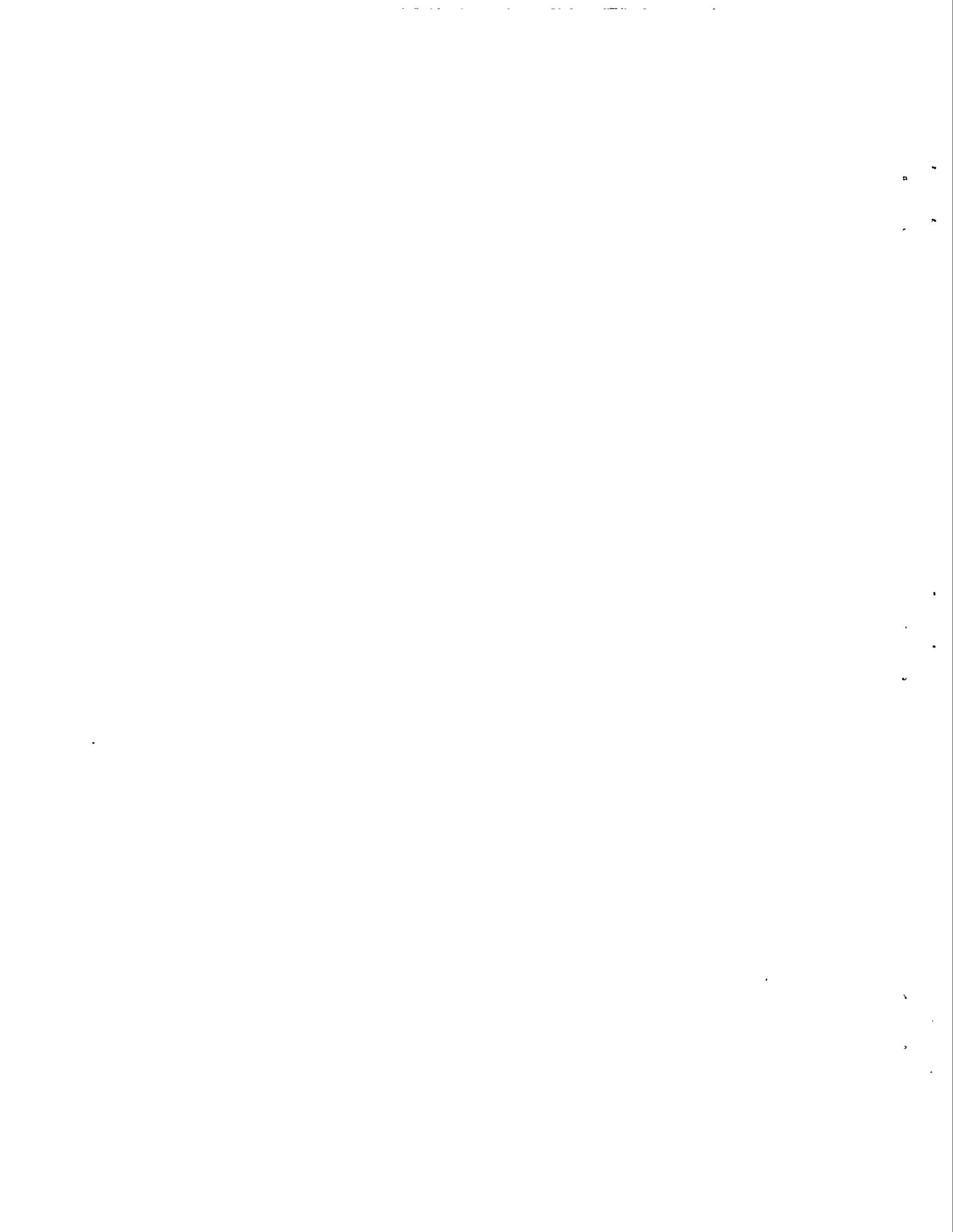
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*East Tennessee State University, College of Medicine, Johnson City,
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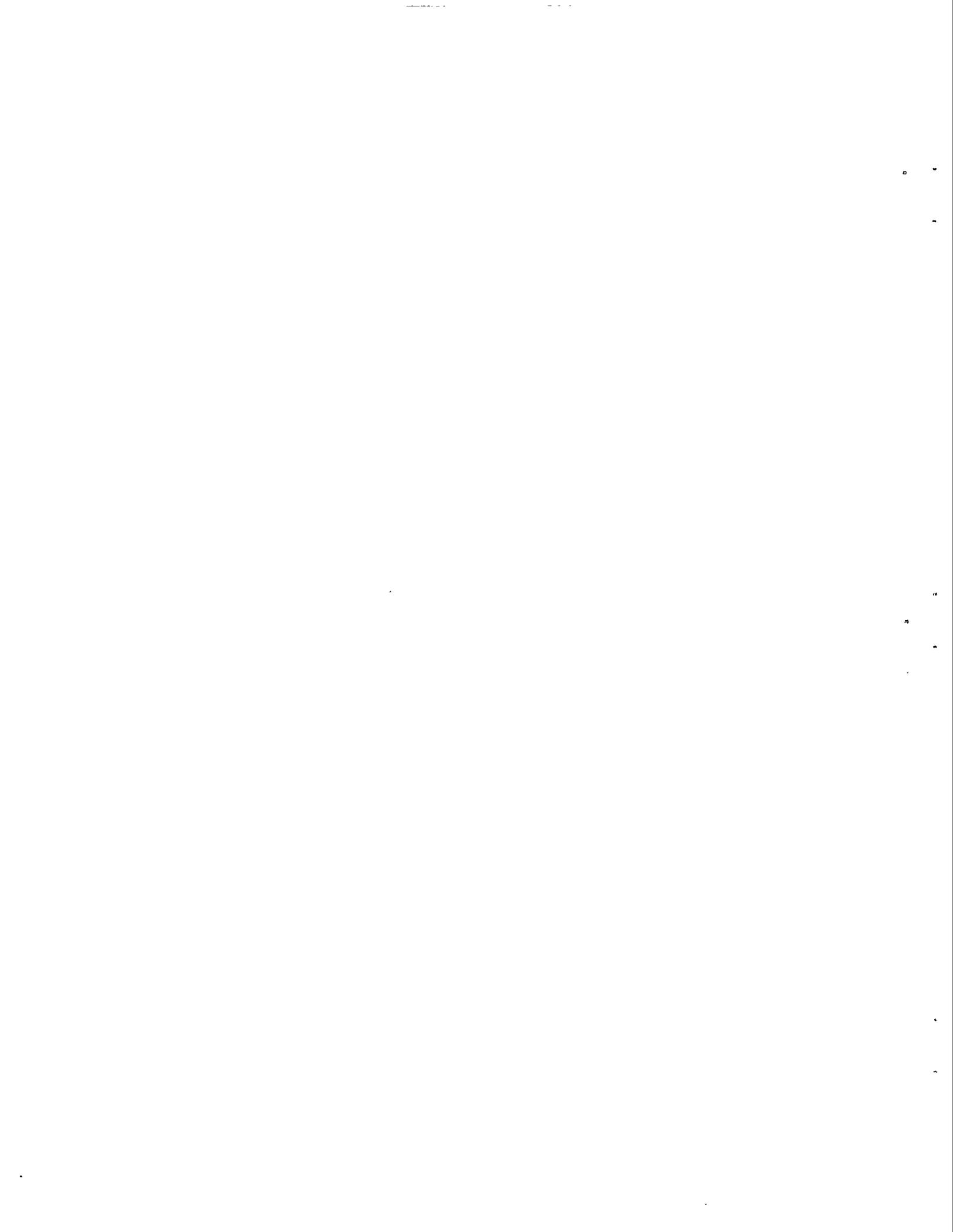
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EXECUTIVE SUMMARY

The environmental restoration of Oak Ridge National Laboratory (ORNL) sites, that have been contaminated by previous waste disposal operations, will require extensive remedial actions to protect human health and the environment from continuing and future releases. However, many of these waste management sites do not have obvious solutions to reduce the contamination of the environment and to provide long-term performance of contaminant measures. Many potential techniques for stabilization and closure of these contaminated sites have unanswered questions about their effectiveness, method of implementation, and costs. Few techniques have established case histories that would provide assurance for their applicability to ORNL sites. Given this present state of evolution of remedial technology, the planners for environmental restoration are faced with the problem of proceeding with the best available or established technology, which inherently assumes a large risk because long-term performance is not assured. Considering the huge potential costs of a second future round of environmental restoration, a compelling justification can be made for the development, demonstration, and assessment of potential yet unproven closure techniques so that their effectiveness can be established before the implementation of environmental restoration projects. Such techniques under evaluation at ORNL include in situ vitrification, in situ grouting, and dynamic compaction.

A field-scale vitrification of a cold (nonradioactive) model of an old seepage trench was completed on July 20, 1987, by personnel of Battelle Pacific Northwest Laboratories in cooperation with ORNL. An estimated 20 tons of soil and crushed limestone backfill were melted. The desired melting depth of 2 m was achieved. The molten mass tended to grow parallel to the long axis of the trench rather than symmetrically around the square array of electrodes. All melt geometry, safety, and process performance objectives were achieved satisfactorily. A 2-month cooling period was required before core sampling of the vitrified product was initiated. Samples of the off-gas scrub solutions have indicated that 99.88% and >99.99% of the nonradioactive Cs and Sr tracers, respectively, were retained in the trench. Samples of the field-produced material were found to be as good or better than two standard high-level nuclear vitreous waste forms when using standard leach tests. Approximately half of the block devitrified or crystallized to pseudowollastonite and anorthite mineral phases during cooling. However, devitrification produced no significant change in the leach properties of the material.

Sodium amendments to mixtures of ORNL soil and limestone have been investigated for their ability to lower melting temperatures required for vitrification in an effort to further reduce the volatilization of ^{137}Cs that would occur during ISV of ORNL seepage trenches. Additions of up to 13.5% Na_2O to 7:3 soil:limestone mixtures reduced the apparent melting range from about 1250°C to 1050°C; greater additions of Na_2CO_3 reduced melting points below 1000°C but were not quantified. It is hoped that this lower melting material would help reduce the volatilization of ^{137}Cs in a planned field demonstration in the summer

of 1990 to levels lower than were experienced in a 1987 nonradioactive field demonstration at ORNL. Additions of either NaOH or Na₂CO₃ to the soil-limestone did significantly enhance volatilization of ¹³⁷Cs during laboratory thermal treatments compared to unamended materials. Although extrapolations to what would be experienced in the field are difficult (5% Cs volatilization was observed in the laboratory for the same melt composition for which only 0.12% volatilization was observed in the field), only a twofold increase in Cs volatilization would be expected from a 10% Na₂O-amended melt assuming identical ISV processing temperatures. However, because ISV processing temperatures would probably be reduced by about 200°C, Cs volatilization might be reduced even below the 0.12% observed in the field demonstration. Leachability of Cs from Na₂O-amended vitrified products increased significantly over unamended materials. This effect would be minimal on a field-produced ISV waste form where the product has such a small surface area per unit weight.

Potassium was found to be identical to sodium on an equivalent weight basis in enhancing the volatilization of ¹³⁷Cs during vitrification. Increasing limestone composition of the soil mixture led to significant increases in Cs volatilization. However, in the proportions of limestone inclusions that are expected in the field, an effect on Cs volatilization was not detected. It appears that the major mechanism for Cs volatilization during vitrification occurs when the material is undergoing a change from an initially calcined or sintered phase to the vitrified material. The high porosity of the sintered phase presumably facilitates Cs volatilization at high temperature until porosity is lost during actual vitrification. Little additional volatilization of Cs was observed once a mixture had vitrified, similar to previous work which had found that holding time after melting had little influence on final Cs volatilization at or below 1600°C. This information, coupled with ongoing work on the effects of Na₂O on melt viscosity and electrical conductivity, should result in an optimal amendment level for the radioactive field test in the summer of 1990.

The proposed in situ grouting of burial trenches with polyacrylamide had raised several concerns about the potential for environmental contamination by acrylamide. To establish the compatibility of the acrylamide grout set reaction with potential interference by buried waste constituents, several tests were performed to verify the ability to predict and control the polymerization or set of the acrylamide grout. Samples of soil from the bottom of 7 trenches and 31 samples of burial trench leachate were collected and added to grout solutions before set. No soil or leachate significantly retarded the grout set time. By chemical analysis of extracts of grout/soil specimens, it was determined that the percent polymerization in the presence of soil or leachate was identical to that achieved with unamended grout (i.e., 99.5%). Five trenches that had been dynamically compacted in 1988 were sampled again for leachate and bottom soil in March 1989; neither the precompaction nor postcompaction samples exhibited any effect on grout set times or degree of polymerization. Ten organic chemicals that had previously been identified in leachates from SWSA 6 trenches were tested for compatibility with acrylamide grout set; only benzene and toluene reduced the percentage polymerization

marginally to 98.8 and 99.3%, respectively. In addition, specimens of burial trench soil were converted from high to unmeasurably low hydraulic conductivities by percolation with reacting grout. Thus, it appears that these burial trench soils and leachates pose no significant interference with acrylamide grout set. Both the degree of polymerization and the impermeable nature of the resulting polyacrylamide grout lead to the assurance that environmental contamination of groundwater by grout injection into these burial trenches is extremely improbable.

Shallow land burial in unlined trenches has been the disposal method used at ORNL for radioactive solid waste. Recent regulatory scrutiny has accelerated the planning for stabilization and closure of many of these waste disposal areas. However, protection of buried waste from infiltrating precipitation via impermeable covers is compromised by the tendency of burial trenches to subside as backfill collapses into voids within the trenches. As part of a low-level radioactive waste burial site stabilization and closure technology demonstration project, a group of five 14-year-old burial trenches in ORNL SWSA 6 was selected for testing trench compaction, grouting, and infiltration barrier design and performance. To obviate the chronic problem of trench subsidence and to provide a stable foundation for the infiltration barrier, the five trenches were dynamically compacted by the repeated dropping of a 4-ton weight with a 1.1-m² base onto each trench from a height of approximately 7 m.

The five trenches were compacted to a maximum depth of 1.2 m, requiring an average of 5.5 drops/m² of trench area, and the site was graded to facilitate surface runoff. Measurements of void reduction within the trenches averaged 77% and were calculated by a comparison of ground surface depression and measured water-accessible voids prior to compaction. Penetration tests were performed on trenches before and after compaction and on the surrounding undisturbed soil formation. The penetration resistance of the trenches was extremely low before compaction and was increased to a level equivalent to that of the undisturbed soil after compaction.

Thus, dynamic compaction was found to be very effective in stabilizing burial trenches to the extent that no differential land surface settlement should be expected to compromise the foundational support of an infiltration barrier. However, the benefit of ground surface stabilization must be weighed against the risks associated with the loss of any ambient containment of incidental amounts of buried liquids that may be present in the waste. Because ambient containment of small but unknown amounts of contained liquids is not a characteristic on which to base a sound strategy for protection from environmental releases, which will occur ultimately for any buried liquid containers, such risks, inherent in dynamic compaction, are quite minimal.

There has been an obvious bias in the selection of technologies for demonstration. In situ methodologies are preferred over any technologies that require exhumation or clean closure. Not only do in situ technologies minimize the risk to remedial action workers but they minimize the potential for inadvertent releases which could occur during field exhumation activities. More importantly, exhumation also creates the new problem of redispersion which, of necessity, would move waste to a

new site for potential contamination. The other strong bias in the strategy for demonstrating these technologies has been to select actual waste sites, or as close a simulation as possible, for the demonstrations. Dynamic compaction and in situ grouting have been carried out on low-level waste burial trenches. Dynamic compaction has been found very effective in providing stabilization of burial trenches for the support of infiltration barriers. However, the risk of breach of any liquid contaminant within the buried waste must be assessed. In situ grouting technology requires further field demonstration of its hydrologic effectiveness. Additional tests of the compatibility of polyacrylamide grout with waste constituents need to be established. In situ vitrification has been performed on a scale model of an ORNL seepage trench but without radioactivity present. Evaluations of the operational performance of in situ vitrification and waste form durability from ORNL earthen materials have been extremely encouraging. The addition of melt-lowering materials, such as sodium hydroxide, has good potential to lower cesium volatilization during in situ vitrification. Future in situ vitrification demonstrations, however, will use actual waste from the ORNL seepage pits and trenches. The cost for development and demonstration of these technologies has been significant but pales in comparison to costs for actual environmental restoration.

1. INTRODUCTION

Beginning in 1985, intense scrutiny by federal and state regulatory agencies has been focused on both present and past waste management operations at the Oak Ridge National Laboratory (ORNL) and other U.S. Department of Energy (DOE) sites. This oversight has resulted in a renewed and vigorous commitment to environmental restoration of many contaminated sites at ORNL. Although much of this effort at environmental restoration to date has been focused on quantifying the extent of contamination and assessing its associated risk to human health and the environment, corrective actions, such as the recent interim corrective measures in Solid Waste Storage Area (SWSA) 6 (Lockwood Greene Engineers 1988), are becoming necessary to meet regulatory deadlines and to abate continuing discharges of contaminants to the environment. However, many of the waste management sites do not have obvious solutions to reduce contamination of the environment and to provide long-term performance. Many potential techniques for stabilization and closure of these contaminated sites have unanswered questions about their effectiveness, method of implementation, and costs (Spalding 1989). Few techniques have established case histories that would provide assurance for their applicability to ORNL sites. Given this present state of evolution of remedial technology, planners for environmental restoration are faced with the problem of proceeding with the best available (or, more likely, the least objectionable) technology, which inherently assumes a large risk because long-term performance is not assured and postpones the ultimate costs for final restoration to the unspecified future. Considering the huge potential costs of a second future round of environmental restoration, a compelling justification can be made for the development, demonstration, and assessment of potential yet unproven closure techniques so that their effectiveness can be established before implementation of environmental restoration projects. The costs for such development and demonstration are significant but pale in comparison to actual costs for environmental restoration.

The diversity of environmental contamination problems at ORNL sites is quite large. Most of the hazards are associated with radioactive waste disposal, but concomitant disposal of nonradioactive hazardous substances complicates the remediation problems. The types of contaminated sites at ORNL include solid waste disposal areas, where radioactive and hazardous wastes were buried primarily in unlined trenches; surface water impoundments, where liquid wastes were stored or disposed of in unlined and leaking ponds; seepage pits and trenches, where liquid radioactive waste was pumped and allowed to seep into the soil formation; hydrofracture grout sheets, where liquid radioactive waste was mixed with cement and pumped about 300 m below ground into induced fractures in the host bedrock; and underground waste storage tanks and transfer lines, where liquid waste was transported, stored, and often leaked. Associated with all of these sites is secondary contamination as substances leached from the disposal points and contaminated surrounding soil, rock, groundwater, vegetation, streams, White Oak Lake, and, ultimately, the Clinch and Tennessee rivers. It

should be emphasized that this environmental contamination occurred over a 40-year period under the deliberate and explicit policy of diluting and dispersing contaminants which, of course, is contrary to the present environmental policy of containment and isolation from the environment. The risks associated with these various sites are quite diverse also. The risks range from minor amounts of a single radioisotope contaminating a remote site to hundreds of thousands of curies of mixed fission and transuranic isotopes, with unknown amounts of nonradioactive hazardous substances, disposed of in close proximity to groundwater and surface streams. Some sites have known and monitored continuing releases to surface waters while others have no apparent present releases. Obviously, a comprehensive risk assessment of all sites is needed to determine their priority and necessity for remedial actions. However, the selection of which specific remedial actions are appropriate for a particular site to improve its performance, such as to reduce continuing releases in the short term, or to assure long term performance against future releases, is a difficult choice.

Potential techniques for remediation of these problem contaminated sites are equally diverse. For sites with high inventories of radionuclides that have not yet been dispersed significantly by groundwater, in situ vitrification (ISV) is potentially applicable. Such sites would include the seven old seepage pits and trenches, which were used between 1951 and 1966 to dispose of over a million curies of fission and transuranic isotopes, and auger holes in the SWSAs which were used for disposal of high-activity shipments of fission products. ISV may also be applicable to underground storage tanks that contain residual sludges with significant inventories of radioactivity. For sites with lower inventories or more dispersed sources of contaminants, other technologies may be more applicable (e.g., covering with an infiltration barrier to prevent further leaching and contaminant migration). However, the burial trenches of low-level solid waste in many of the ORNL SWSAs do not have the stability necessary to support such an infiltration barrier; they were not compacted when filled and, hence, continue to subside. A technique such as dynamic compaction may be appropriate to consolidate burial trenches in situ, making them stable enough to support an infiltration barrier. In situ grouting of buried waste may also provide enough support to construct an infiltration barrier with assurance that it will not subside. In situ grouting would have the additional potential benefit of converting buried waste to a hydrologically impermeable mass. This would have a good application to those burial trenches that experience inundation by groundwater from which, in some cases, an infiltration barrier would offer no protection. Such hydrologic isolation may also be necessary for closure of surface impoundments which, by design, were often situated in low-lying areas with direct connection to the peripheral groundwater.

The following descriptions of technical demonstrations, carried out at ORNL over the last 2 years, attempt to address the objective of evaluating long-term effectiveness. These technologies include ISV, in situ grouting, and dynamic compaction. There is an obvious bias in the selection of technologies for demonstration. In situ methodologies are preferred over any technology that requires exhumation or clean closure.

Not only do in situ technologies minimize the risk to remedial action workers but they also minimize the potential for inadvertent releases which could occur during field exhumation activities. More importantly, exhumation also creates the new problem of redisposal which, of necessity, would move waste to a new site for potential contamination. The other strong bias in the strategy for demonstrating these technologies is to select actual waste sites, or as close a simulation as possible, for the demonstrations. Dynamic compaction and in situ grouting have been carried out on low-level waste burial trenches. ISV, however, has been performed on a scale model of an ORNL seepage trench but without radioactivity present. Future ISV demonstrations, however, will use actual waste from the ORNL seepage pits and trenches.

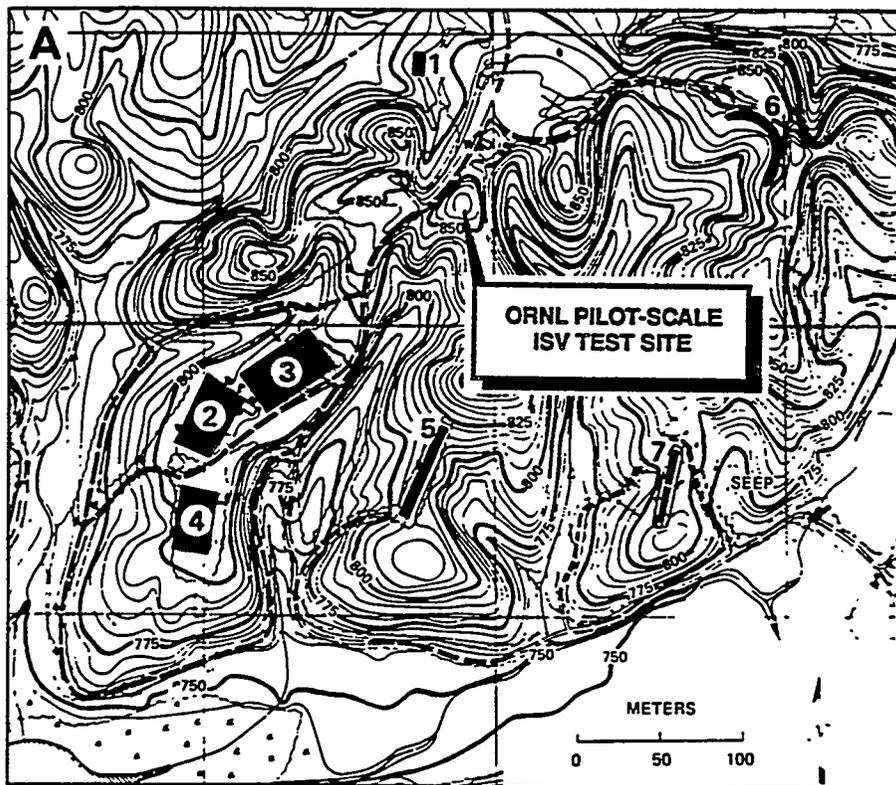
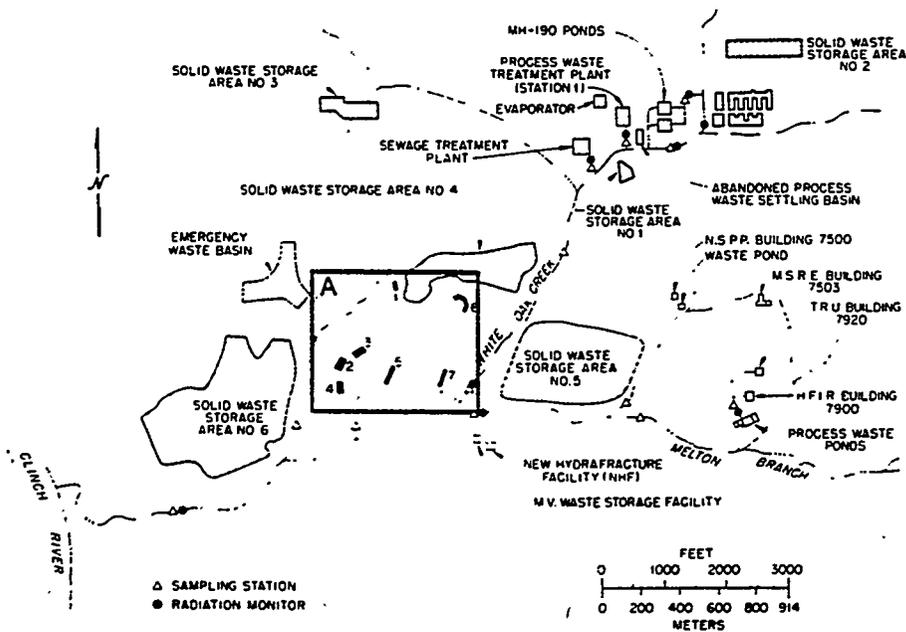
This strategy for performing technology demonstrations on actual field sites has several benefits. Obviously, the technical feasibility and effectiveness are determined on the actual field scale so that little extrapolation is required. Establishing effectiveness, however, necessitates the measurement of critical parameters that would probably not be performed during actual applications. Thus, the technology demonstrations require a good deal of before, during, and after characterization, often with extensive instrumentation. In addition, the technology demonstrations often develop or determine operational techniques. For instance, in dynamic compaction, with what pattern and how often should the weight be dropped, and how should a performance contract be specified? The technology demonstrations also help to identify institutional constraints that would probably be overlooked in planning activities. Interaction with federal and state regulatory agencies allows review of the proposed demonstration for conformance to rules and regulations. This interaction also allows the regulatory agencies to become familiar with the proposed techniques before they are presented in formal procedures. The technology demonstrations help a great deal in the estimation of costs for actual applications, in particular, by identifying required actions and providing a case history for time and personnel requirements. Lastly, the demonstrations help greatly in transferring the technology to other DOE sites. Dynamic compaction is a significant part of the stabilization activities at the Savannah River Plant and is based on the techniques developed at ORNL in 1985. ISV has been developed by Pacific Northwest Laboratory but, ironically, will probably have its first DOE applications at ORNL.

The following sections contain descriptions of the three technologies under demonstration at ORNL. Each section describes a particular technology and is independent of the others. They often represent a summary of previously published information updated with current observations and discussion of related issues. The combination in this report should also serve as a single source for technical information for technology transfer. Additional details on each technology can be found in the referenced information in each section.

2. IN SITU VITRIFICATION TECHNOLOGY DEMONSTRATION

A series of seven seepage pits and trenches (Fig. 1) was used between 1951 and 1966 for the disposal of approximately 2.5×10^7 gal of liquid radioactive wastes at the Oak Ridge National Laboratory (ORNL). Approximately 200,000 Ci of ^{90}Sr and 600,000 Ci of ^{137}Cs , along with smaller quantities of other fission products, uranium, and transuranic elements, were disposed of in this series of pits and trenches. To facilitate the seepage of liquids, the trenches were constructed on the tops of ridges, oriented perpendicular to the strike of the bedding of the formation, and filled with crushed limestone or dolomite (Fig. 2). As the liquids seeped out, the Cs and Sr remained within, or in close proximity to, the trenches. Cesium is generally irreversibly sorbed by the illite-rich soils at ORNL. Strontium, on the other hand, is poorly sorbed. To reduce the mobility of Sr, the pits and trenches were treated with a highly alkaline solution (NaOH) at the time of disposal. All of the pits and trenches are now covered with asphalt caps to reduce the direct flow of precipitation through the waste. Currently, the pits and trenches do not contribute significantly to surface-water contamination, and most of the Cs and Sr remains in close proximity to the bottom of the pits and trenches. However, the large inventory of ^{137}Cs and ^{90}Sr , the close proximity of the wastes to the surface and White Oak Creek, and the potential for significant releases in the future necessitate either a long-term site maintenance and monitoring program or some form of remedial action to allow the sites to be permanently closed.

In situ vitrification (ISV) is one possible technology that could be applied to the pits and trenches (others include grouting and ground densification). ISV, developed and patented for the U.S. Department of Energy (DOE) by Battelle Pacific Northwest Laboratory (PNL), involves placing four electrodes in a square array around the contaminated volume of soil, applying power to the electrodes, and melting the entire mass of soil into a chemically homogeneous and durable glassy-to-microcrystalline waste form. The melting is initiated at the surface of the soil and progresses downward through the contaminated zone (Fig. 3). Gases produced during the high-temperature (1600 to 2000°C) operation are collected through an off-gas hood and scrubbed of possible contaminants in a process trailer. The ISV technology has been extensively tested by PNL at electrode spacings from approximately 0.3 to 6 m. The pits and trenches at ORNL are good candidates for vitrification because of their small areal extent and shallow depth (<6 m). The potential for personnel exposure from the high concentrations of ^{90}Sr and ^{137}Cs in the pits and trenches makes an in situ technology highly desirable compared with one that would require excavation of the contaminated zone.



ILW WASTE PITS AND TRENCHES

Fig. 1. Location of the radioactive liquid-waste disposal pits and trenches (numbered 1-7) at ORNL and the pilot-scale ISV demonstration.

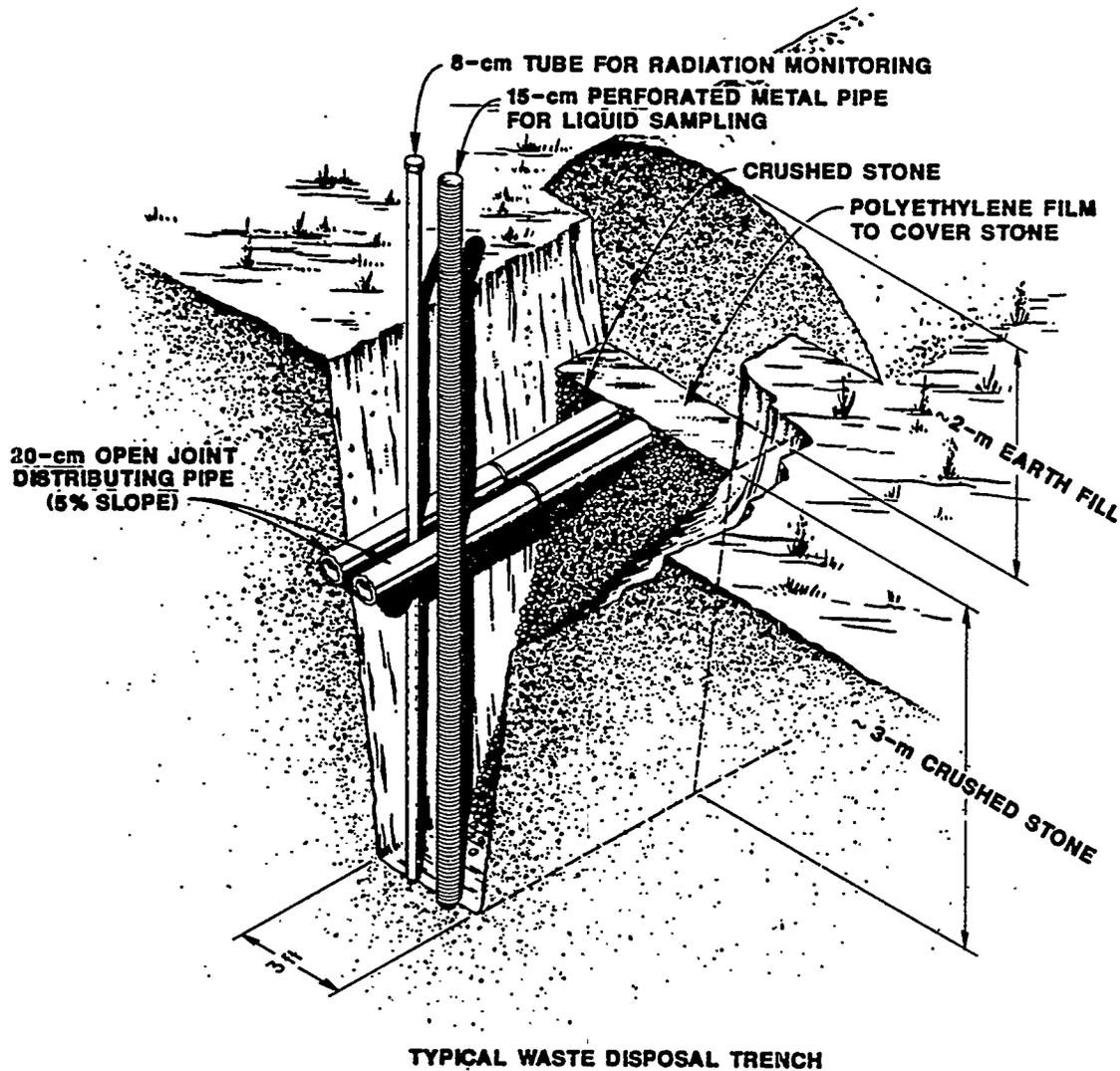


Fig. 2. Construction details of ORNL liquid waste disposal Trench 7.

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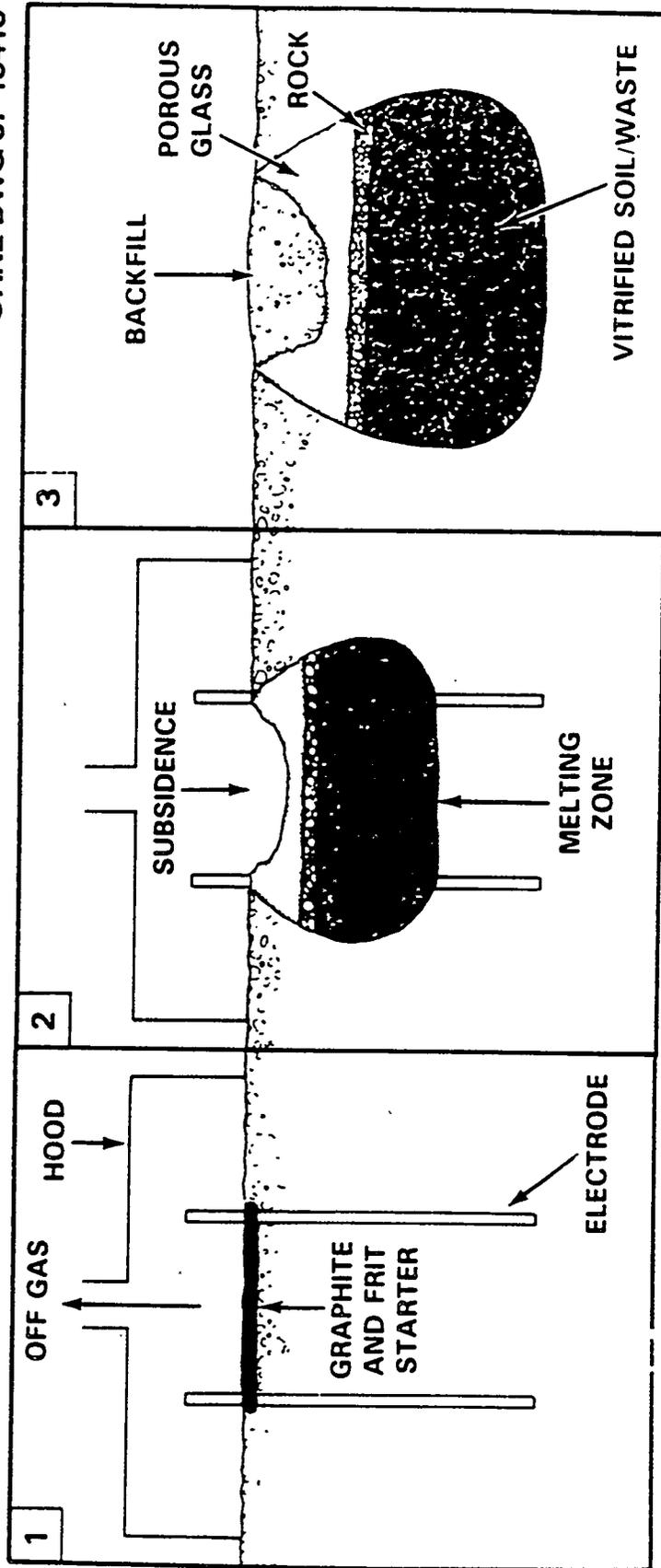


Fig. 3. Schematic of the ISV operating sequence.

2.1 FIELD DEMONSTRATION OF IN SITU VITRIFICATION FOR APPLICATION TO ORNL LIQUID WASTE DISPOSAL TRENCHES

The pilot-scale, cold (nonradioactive) demonstration of ISV technology was performed jointly by ORNL and PNL to achieve four main objectives: (1) complete an application of ISV technology away from the Hanford site to evaluate the feasibility of technology transfer to ORNL, (2) assess the operational performance of ISV for applications in structurally heterogeneous, high-carbonate soils and rocks, (3) determine the retention factors (mass in melted soil divided by mass in off-gas) for Cs and Sr under field conditions, and (4) evaluate the durability of the waste form produced in the ORNL soil system. Objectives 1 and 2 were addressed in a preliminary report last year (Jacobs et al. 1987). Detailed assessments of results relating to objectives 3 and 4 have been published recently (Carter et al. 1988).

The principles of ISV are based on developments from work performed at PNL on joule-heated melters for various nuclear waste immobilization projects (Buelte et al. 1979). The joule-heating principle involves internal resistance heating as electrical current passes through the molten media. In ISV, the resistance decreases as the molten mass increases in size. To maintain a power level high enough to continue melting additional soil, the current must be increased. To accomplish the variable current during ISV processing, a power transformer with multiple voltage taps is used. At startup, the ISV process requires high voltage and low amperage. As the melt progresses and resistance decreases, the lower voltage taps on the power transformer allow increased amperage to be applied to the melt, maintaining a high power level. The process continues until heat losses from the melt approach the energy delivered to the soil via the electrodes or until power is discontinued to the electrodes.

2.1.1 Pilot-Scale Test System

The pilot-scale test system at ORNL uses four electrodes having a 1.2-m separation and consists of a power-control unit and off-gas containment hood over the test site. A cutaway view of the support trailer and off-gas hood is illustrated in Fig. 4. Prior to the ORNL test, this same system had been used on 11 pilot-scale tests at the Hanford site.

2.1.1.1 Power Delivery System

The pilot-scale power system uses a Scott-Tee connection to transform a three-phase input to a two-phase secondary load using diagonally opposed electrodes in a square pattern. The 500-kW power supply may be either voltage or current regulated. The alternating primary current is rated at 480 V, 600 A, 3 phase, and 60 Hz. The three-phase input feeds a Scott-Tee connected transformer (Fig. 5), which provides a two-phase secondary load. The transformer has four separate voltage tap settings of 1000, 650, 430, and 250 V. Each voltage tap has a corresponding amperage rating of 250, 385, 580, and 1000 A per phase, respectively. The amount of three-phase input power

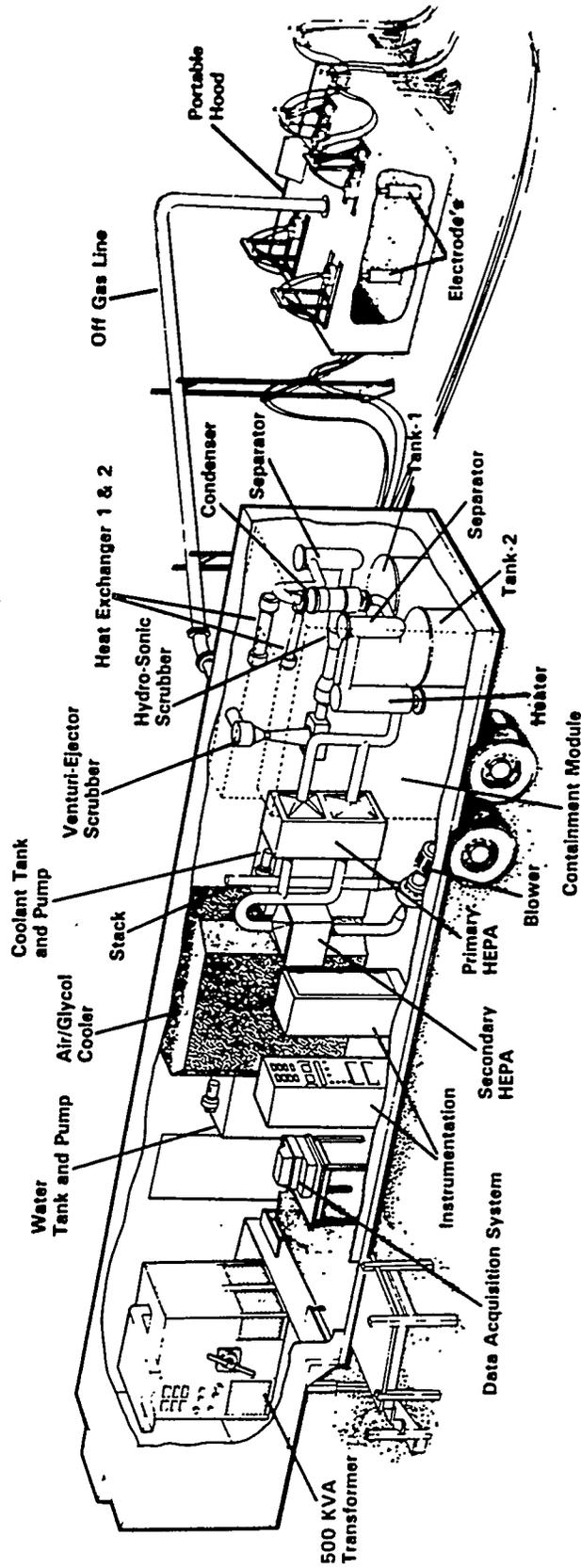


Fig. 4. Cutaway view of the pilot-scale process trailer and off-gas hood.

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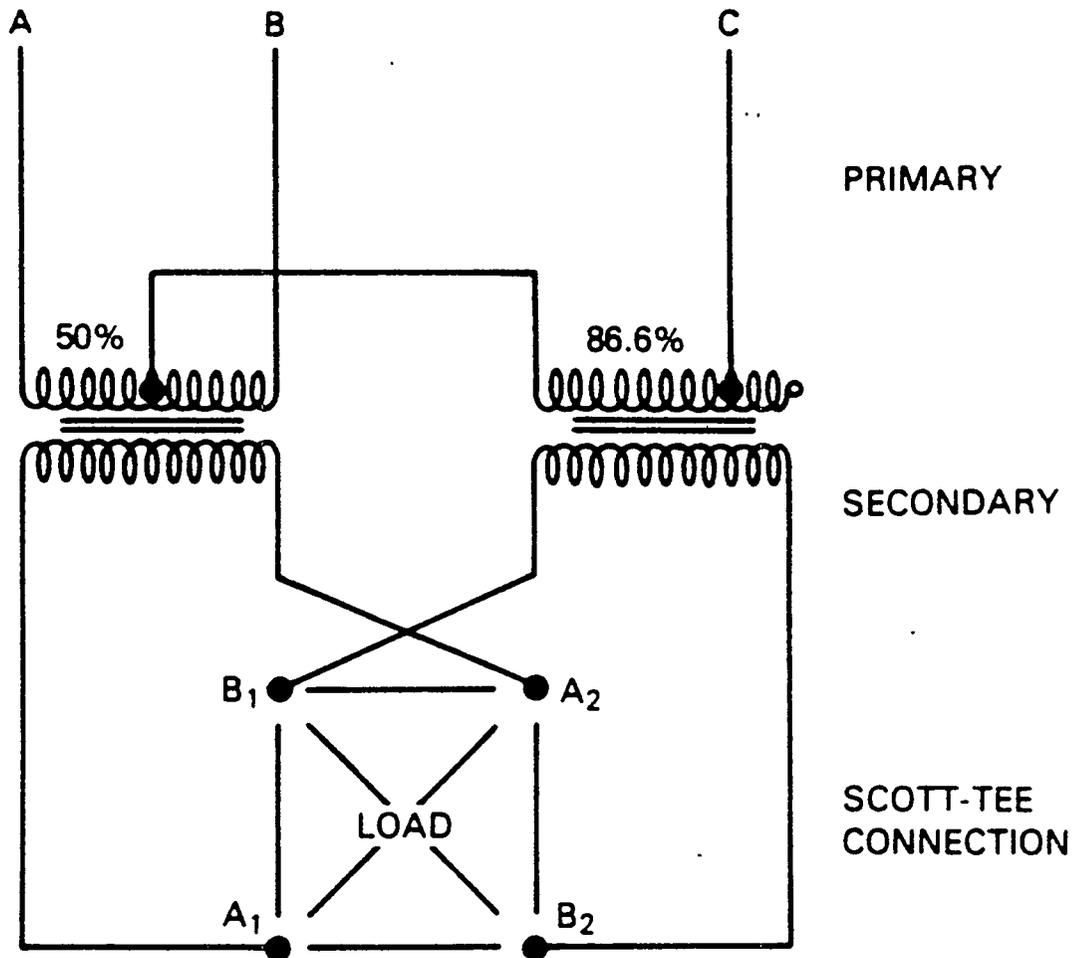


Fig. 5. Scott-Tee electrical connection for the pilot-scale ISV system.

delivered to the transformer is controlled by adjusting the conduction angle of the thyristor switches located in each of the three input lines. These switches, in conjunction with selectable taps on the transformer secondary, regulate the amount of power deliverable to both secondary phases. The Scott-Tee connection provides an even power distribution among the three primary phases when the molten zone approaches a uniform resistance load. During the test at ORNL, this power system proved effective in maintaining a balanced load to the electrodes.

2.1.1.2 Off-Gas Containment and Electrode-Support Hood

The off-gas containment and electrode-support hood, constructed from seven panels of 20-gauge stainless steel bolted together, is 3.1 m wide, 5.5 m long, and 0.9 m high. Four leveling supports are attached to the corners of the hood, which also has a port for viewing (and access to) the surface of the melt. A central off-gas port allows direct coupling of the hood to the processing trailer and off-gas treatment system. The hood is equipped with heat fins installed on the surfaces of panels to help cool the hood to which radiant heat is transferred from the partially molten surface during processing. The hood, designed to withstand a water vacuum of 18 cm, is sealed to the surface of the soil surrounding the molten zone by means of a flexible skirt of tightly-woven, high-temperature-resistant fiber attached to the bottom of all side panels. The skirt extends approximately 0.6 m away from the hood to allow for a hood-to-ground seal when covered with a layer of soil. Electrical bus bars connected to the molybdenum electrodes protrude through the hood; these are surrounded by electrically insulated sleeves that allow adjustment of the electrode positions. The electrodes and bus bars are supported by insulators above the sleeves. The insulators are designed to withstand movement of the molten mass against the electrodes from convective currents and the gravitational and buoyant forces exerted on the electrodes.

2.1.1.3 Off-Gas Treatment System

The off-gas treatment system is shown schematically in Fig. 6. The off-gas passes through a venturi-ejector scrubber and separator, a Hydro-Sonic* scrubber, a separator, a condenser, another separator, a heater, two stages of HEPA filtration, and a blower. Liquid to the two wet scrubbers is supplied by two independent scrub recirculation tanks, each equipped with a pump and heat exchanger. The entire off-gas system is installed in a 13.7-m-long semitrailer to facilitate transportation (see Fig. 4). All of the off-gas components except the second-stage HEPA filter and blower are housed within a removable containment module that has gloved access and is maintained under a slightly negative pressure to protect workers from potential contamination. Heat is removed from the off-gas by a closed-loop cooling system consisting

*Hydro-Sonic scrubber is a product of Hydro Sonic Systems, Dallas, Texas.

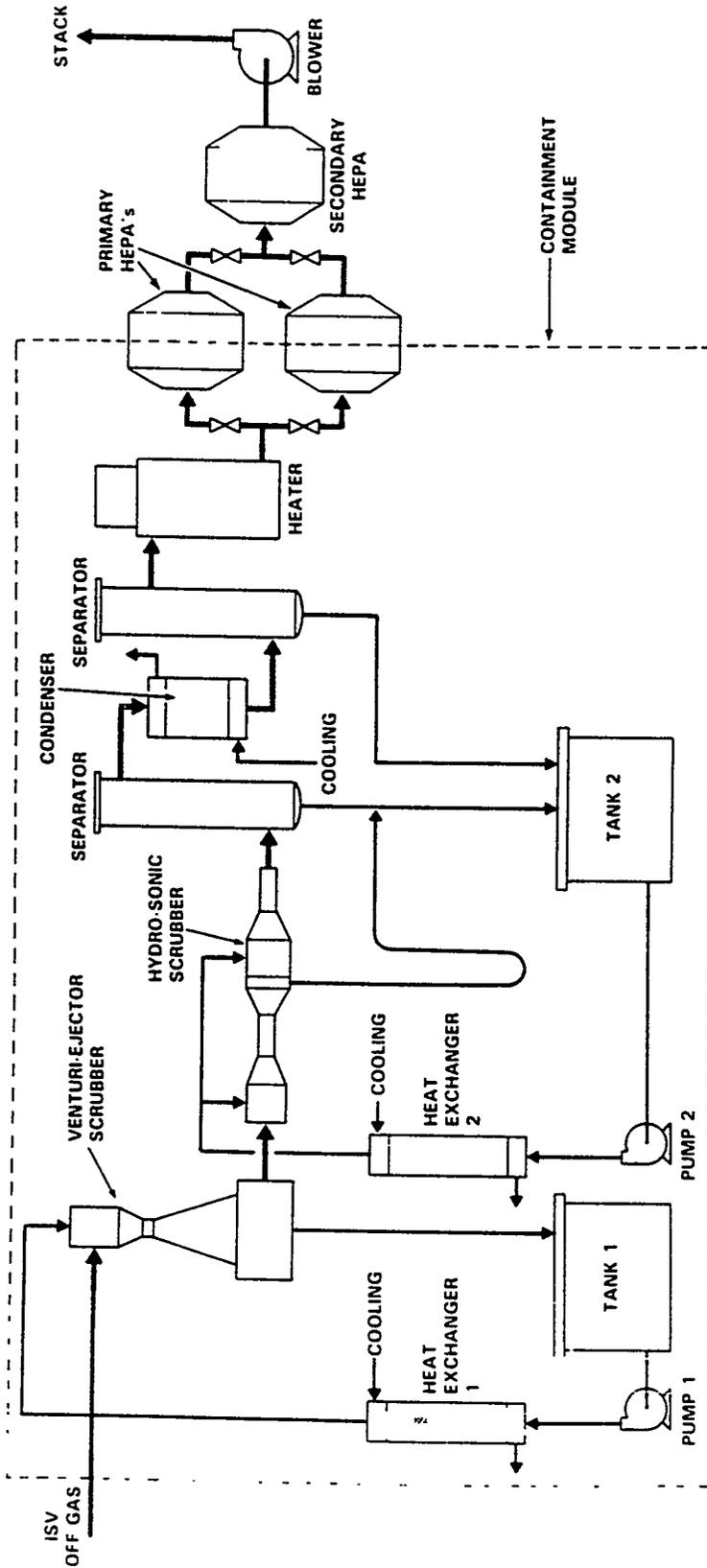


Fig. 6. Schematic drawing of the pilot-scale ISV off-gas treatment system.

of an air/liquid heat exchanger, a coolant storage tank, and a pump. A 1:1 mixture of water and ethylene glycol is pumped first from the storage tank through the shell side of the condenser and the two scrub-solution heat exchangers, and then through the air/liquid exchanger, where heat is removed from the coolant.

The venturi-ejector scrubber serves both as a quencher and high-energy scrubber. The second scrubber is a two-stage Hydro-Sonic scrubber (tandem nozzle scrubber), as illustrated in Fig. 7. The first section condenses vapors, removes large particles, and initiates growth of the finer particles so that they are easily captured in the second stage. Particles are captured when the gas is mixed with fine water droplets produced by spraying water into the exhaust of the subsonic nozzle. Mixing and droplet growth continue down the length of the mixing tube. Large droplets containing the particles are then removed by a vane separator and drained back into the scrub tank. When operated at a differential pressure of 127 cm of water, the unit is designed to remove over 90% of all particles greater than 0.5 μm in diameter. Efficiency of removal increases with an increase in pressure differential.

Additional water is removed from the gas system by a condenser having a heat exchange area of 8.9 m^2 and a final separator. The gases are then reheated to approximately 25°C above the dew point in a 30-kW heater to prevent condensate carry-over to the HEPA filters. The first stage of filtration consists of two 61 x 61 x 29 cm HEPA filters in parallel. During operation, one filter is used and the other remains as a backup in case the operating filter becomes loaded. The primary filter can be replaced during operation. The second-stage HEPA filter acts as a backup in case a first-stage filter fails.

2.1.2 Cold Demonstration Description

The objectives of the pilot-scale demonstration of ISV technology at ORNL were developed to address key differences between the conditions during previous tests of the ISV technology and those anticipated for contaminated sites at ORNL. For example, the waste trenches at ORNL contain large quantities of ^{137}Cs that could volatilize at high temperatures and be carried into the process trailer with the off-gas, resulting in additional operator exposure and wastes. Therefore, it was necessary that the retention factor of $>10,000$ for Cs (Cs in melted soil divided by Cs in off-gas) obtained in engineering-scale tests (Carter et al. 1987) be confirmed at a larger scale under field conditions. Also, the soils at ORNL are more structurally and chemically heterogeneous than those used in previous tests at Hanford. The trench design, with a significant quantity of crushed carbonate rock present, results in a bulk composition of the ISV melt lower in silica and higher in calcium and magnesium than previously studied compositions (Oma et al. 1983). Therefore, the operational performance of the ISV technology as well as the long-term durability of the resulting waste form needed to be addressed specifically for sites at ORNL to provide more confident scale-up predictions. Some of these issues have been investigated during laboratory- and engineering-scale tests conducted at PNL during 1985-1986 (Carter et al. 1987).

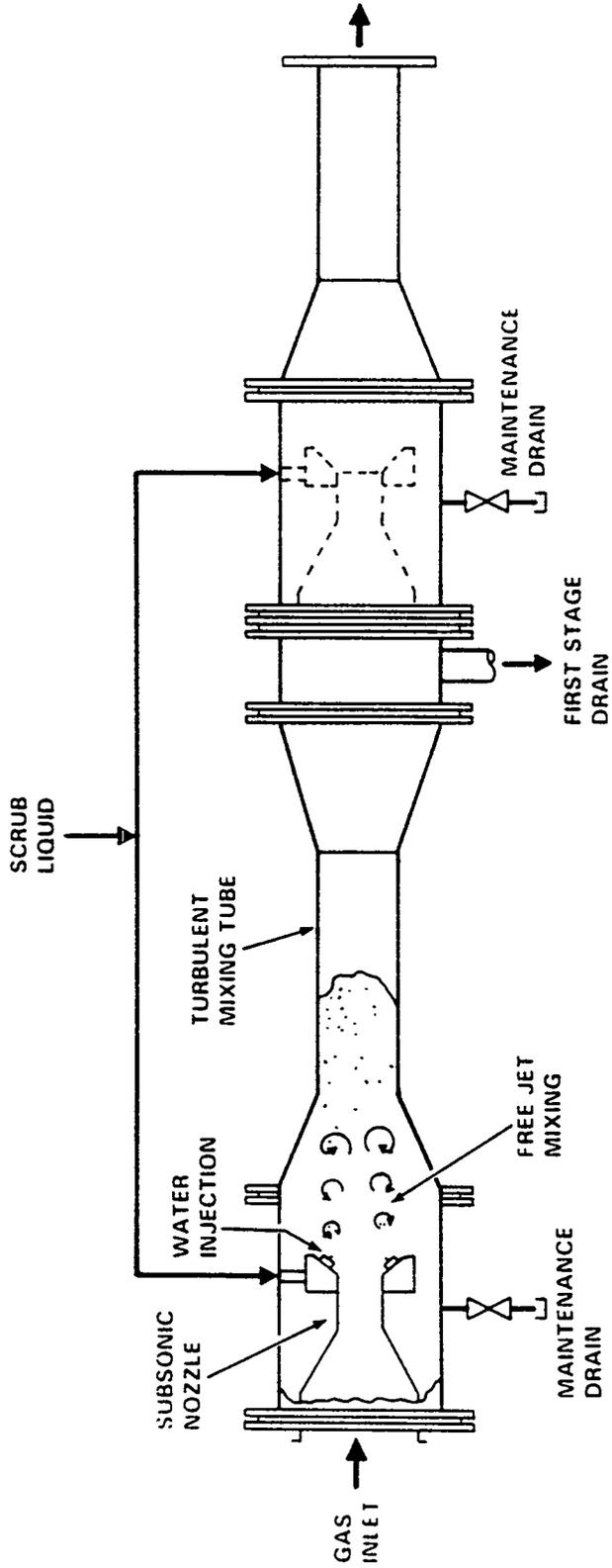


Fig. 7. Tandem nozzle hydro-sonic scrubber (Hydro Sonic Systems, Dallas, Texas).

Trench 7 was chosen as a model because of its size, inventory characteristics, and the fact that some characterization of the trench had already been accomplished (Olsen et al. 1983). To enable the pilot-scale ISV system to be used, a 3/8-scale model of Trench 7 was constructed in an uncontaminated portion of the ORNL site. The site, located on top of a ridge in the Maryville Limestone (an interlayered limestone-shale), was chosen for the similarity of its physical and geological characteristics to the areas used for seepage disposal in the past at ORNL. After preparation of the site (clearing, leveling, electrical service, etc.), the trench was constructed perpendicular to the strike of the bedding. The 9.2-m-long trench was 1 m wide at the top and tapered to 0.4 m at the bottom. A schematic cross section at the midpoint of the trench is shown in Fig. 8 (see Fig. 10 also). The trench was constructed to a depth of 1.5 m, except for the central section, which was excavated to a depth of 2.5 m to allow for the placement of a vertical array of eight type-K (to 1300°C) thermocouples (one thermocouple at every 0.31 m depth) for monitoring the depth of the melt. To simulate the contaminated sludge that is present in Trench 7, 526 kg of a mixture of 18 wt % Cs carbonate and 82 wt % Sr carbonate, both nonradioactive, was placed in the central portion of the trench. These quantities of Cs and Sr were selected to yield a waste form with sufficient concentrations of Cs and Sr that their leach characteristics could be determined. An actual segment of Trench 7 of equivalent size would contain only 114 g of ^{137}Cs (10,000 Ci at 88.1 Ci/g) and 53 g of ^{90}Sr (3300 Ci at 62.4 Ci/g). The entire trench was then filled from the 0.6- to 1.5-m level with crushed carbonate rock. The upper 0.6 m of the trench was backfilled with original soil. In addition to the array of thermocouples in the center of the trench, type-K thermocouples were placed at depths of 0.6, 1.2, and 1.5 m along both sides of the trench at distances of 2.1 and 3.1 m from the center of the trench. Three type-R thermocouples, which have a higher maximum operating temperature (1750°C) than type-K thermocouples, were placed in a vertical array at the center of the trench at depths of 0.9, 1.5, and 2 m in an unsuccessful attempt to monitor melt temperatures. The type-R thermocouples are extremely expensive and cannot function while current is being applied to the melt; they were included in an effort to monitor the temperature of the melt during cooling. Moisture detection cells were placed in the trench at several locations but did not function properly and will not be discussed further. The four molybdenum electrodes (5 cm diam and 3.7 m long) were placed in graphite sleeves (15 cm OD) and placed 1.2 m apart in a square array of augered holes approximately 2.5 m deep. The off-gas hood was then placed over the trench and connected to the off-gas treatment system. The electrodes were connected to the power-delivery system. Run data (temperatures, off-gas flow rates, CO and CO₂ concentrations, electrical parameters, etc.) were recorded every 6 min and logged into a computerized data storage system. Off-gas scrub solution samples were collected approximately every 2 h (duplicate samples were obtained for analysis at both PNL and ORNL).

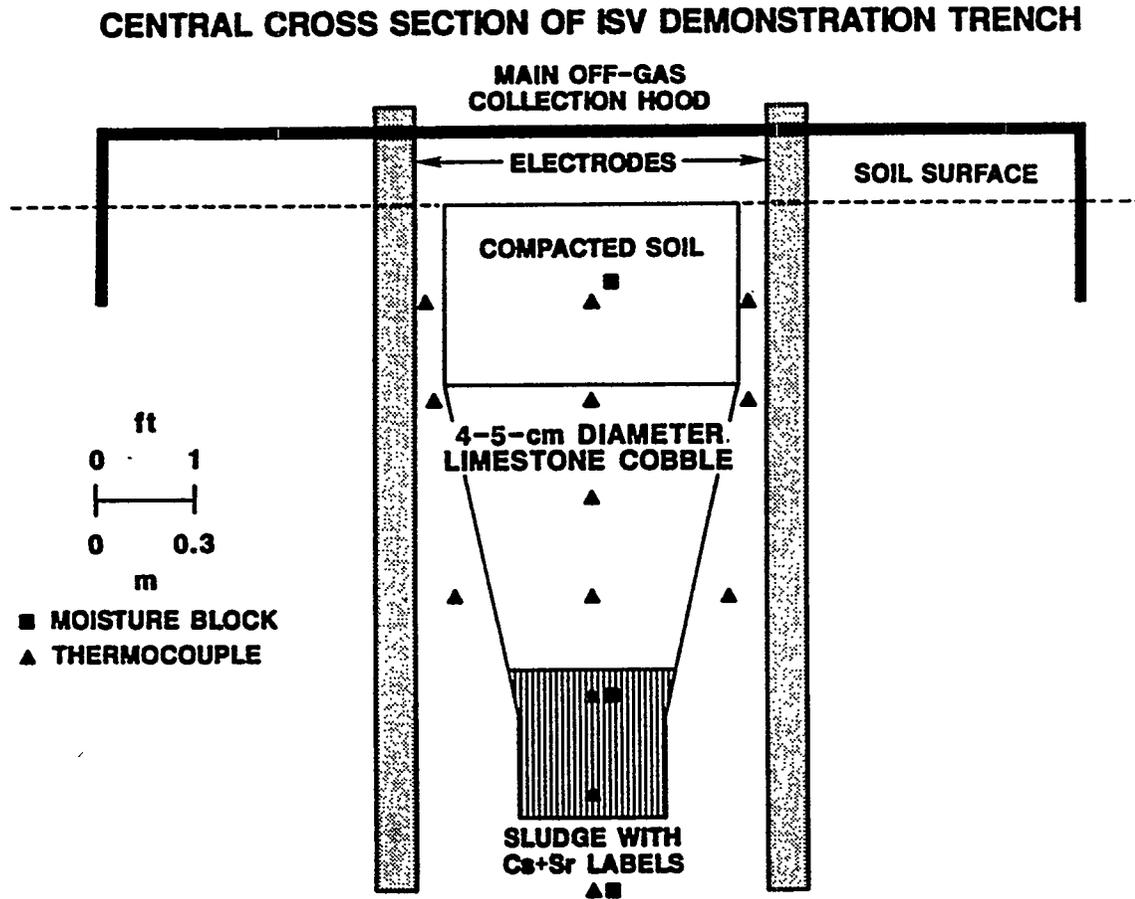


Fig. 8. Central cross section of the pilot-scale ISV demonstration trench (approximate scale).

2.1.3 Results

After one false start on June 26, 1987 (a result of transformer circuitry malfunctions), the ISV test was started on July 15, 1987. The test ran continuously, except for a few interruptions, for 110 h until power was shut off on July 19. Soil temperatures and manual probing of the melt with a steel rod confirmed that the desired depth (approximately 2.1 m) of melting had been reached.

Figure 9 illustrates key operational parameters as a function of run time. As discussed earlier, the trends of decreasing voltage and increasing amperage during operation are necessary to maintain melting. Tap changes, indicated by the sharp increases in the total power, were made at run times of approximately 10, 20, and 30 h. The total power consumed during the test was approximately 29 MWh. The depths of melting were determined by observing the maximum temperature reached at each thermocouple in the vertical array located at the center of the trench. Upon contact with the melt, type-K thermocouples generally burn out, giving a sharp response on the data-logging system and making depth determinations of the melt relatively easy. Based on visual observations of the surface of the soil and temperatures recorded by the thermocouples throughout the trench, the final dimensions of the melted mass were approximately 4.9 m long x 2.1 m wide x 1.2 m thick (Fig. 10). Based on the uniform SrO and Cs₂O content of samples from the melt (1.5 and 0.39%, respectively) and the known quantities of Sr and Cs placed in the trench, a vitrified mass of 20 Mg was calculated. Based on this vitrified mass, approximately 1.26 kWh of electrical energy was required to produce 1 kg of ISV product. As of August 28, 1987, the edges of the mass had cooled to temperatures <70°C. Core sampling of the block was carried out on October 12-14, 1987, to obtain material for durability testing and chemical analyses. A longitudinal cross section of the mass was exposed by using a backhoe and hydraulic jack hammer on November 6, 1987 (Fig. 11).

Samples of the off-gas scrub solutions have been analyzed so that the Cs and Sr retention factors can be determined. Samples of the crushed carbonate rock and soil used in the trench, wall smears from the off-gas ducting and hood, and HEPA filters have also been analyzed for Cs and Sr so that a mass balance for the system can be calculated. Most of the Cs (86 g) was found in the process scrub solutions (Table 1), with a significant amount on the primary HEPA filter (2.1 g) and the ducting (2.9 g). The only significant amount of Sr (0.7 g) was found in the process scrub solution samples. These small amounts of Cs and Sr, compared to the amounts placed into the trench (73,274 and 236,931 g, respectively), yield retention factors of 99.88 and >99.999%, respectively, within the melted mass.

The overall off-gas retention factors are listed in Table 2. The retention factors (RFs) for Cs, Na, and Li were low compared with the other elements. The low Na and Li RFs were strongly influenced by the soda ash addition early in the run; however, the Cs RF was not influenced by this addition. Increasing the RF for ¹³⁷Cs during ISV represents one of the major challenges in the application of ISV to ORNL seepage pits and trenches. The low RF for Mo is strictly an artifact of its use in the electrode coating and annular packing. The RFs for Si,

PILOT-SCALE ISV OPERATIONAL PARAMETERS

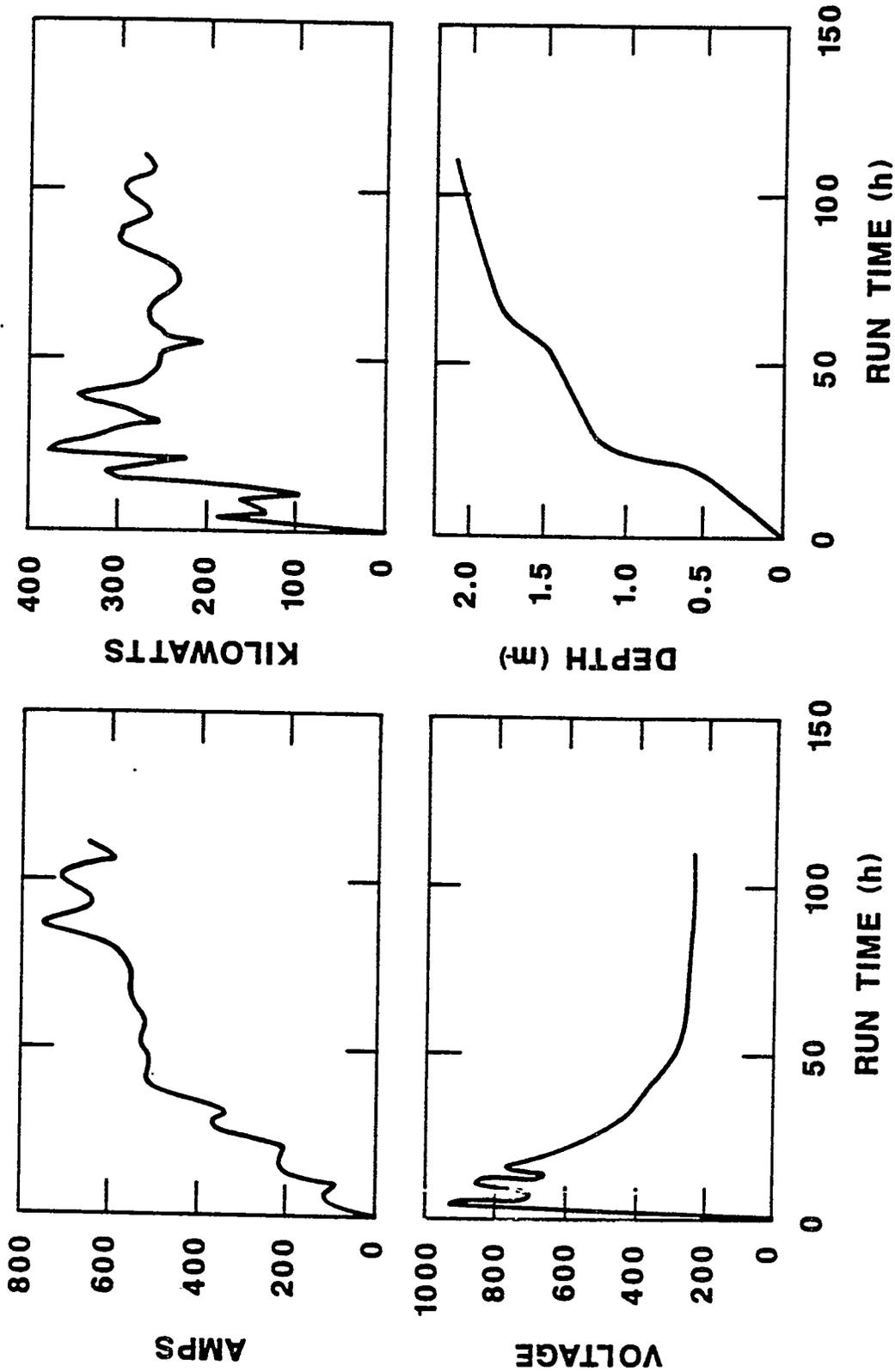


Fig. 9. Voltage, amperage, power, and melt depth as a function of run time. Voltage and amperage are averages of the A and B phases.

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Fig. 11. View of the vitrified mass from the western side after excavation following the ORNL ISV demonstration.

Table 1. Distribution of elements in ISV off-gas system components
(Units are grams, with fraction of total in parenthesis)

Element	Hood	Ducting	Scrub solution	HEPA-1	HEPA-2
Al	3.83 (0.14)	6.35 (0.23)	17.32 (0.63)	0.12 (0.004)	0.06 (0.002)
Ca	1.21 (0.03)	2.00 (0.05)	35.43 (0.90)	0.54 (0.01)	0.25 (0.01)
Cs	0.56 (0.006)	2.94 (0.032)	86.02 (0.94)	2.15 (0.23)	0.06 (0.001)
Fe	6.23 (0.05)	10.33 (0.08)	117.91 (0.88)	0.07 (0.003)	0.01 (0.00)
Li	4.31 (0.05)	7.15 (0.08)	78.97 (0.87)	0.29 (0.003)	0.04 (0.00)
Mg	1.10 (0.04)	1.83 (0.07)	22.13 (0.88)	0.06 (0.002)	0.05 (0.002)
Mo	2.01 (0.14)	3.33 (0.24)	8.05 (0.57)	0.61 (0.04)	0.01 (0.00)
Na	27.93 (0.02)	46.32 (0.03)	1693.42 (0.95)	19.62 (0.01)	4.64 (0.003)
Si	233.22 (0.30)	386.81 (0.50)	159.51 (0.20)	0.95 (0.001)	0.84 (0.001)
Sr	0.04 (0.05)	0.02 (0.03)	0.67 (0.83)	0.06 (0.06)	0.03 (0.03)
Solids	542 (0.03)	899 (0.05)	17138 (0.92)	23.48 (0.001)	0.0 (0.00)

Table 2. ISV off-gas retention factors for elements

Element	Amount in off-gas system (g)	Amount in ISV mass (kg)	Retention factor
Al	27	1472	54,500
Ca	39	2415	61,900
Cs	92	76	826
Fe	135	655	4,850
Li	91	52	571
Mg	25	329	13,200
Mo	14	0.14	10
Na	1,792	236	132
Si	781	5,377	6,890
Sr	0.8	250	312,000
Solids	18,600	20,000	1,075

Al, and Fe are slightly high because the total amounts of these elements, in suspended solids, were not analyzed in the ISV off-gas scrub solutions; more accurate RFs for these elements would be better reflected in the RF calculated for solids in the system. Carter et al. (1988) performed total elemental analyses on selected samples from the ISV scrub solutions; their analytical results were significantly higher for Si, Al, and Fe but quite similar for Ca, Mg, Sr, Na, Li, and Cs.

Chemical analyses of HF-HCl digests of over 30 core samples from various depths and locations within the resulting vitrified block indicated an average composition of 13% Al₂O₃, 18% CaO, 0.39% Cs₂O, 4% Fe₂O₃, 3% K₂O, 2% MgO, 1% Na₂O, 55% SiO₂, and 1.5% SrO. This composition varied only slightly throughout the melted portions of the trench and did not vary between glassy and crystalline (devitrified) samples. Analysis of the crystalline materials by X-ray diffraction indicated that it was composed predominantly of pseudowollastonite (CaO-SiO₂) and anorthite (feldspar, CaO-Al₂O₃-SiO₂). Based on the Si and Ca analyses, it is estimated that the melted mass was formed from about 30 wt % limestone (the major source of its Ca) and 70 wt % from the surrounding and cover soil (the major source of the Si). Standard leach procedures [MCC (Materials Characterization Center)-1 test] were carried out on both the glassy and devitrified phases. MCC-1 test results are usually reported as normalized release (NR) values (grams per square meter). By normalizing the quantity leached to the amount of the element in the sample, one can evaluate whether the release of elements is congruent or incongruent. These values are calculated as follows:

$$NR_i = m_i / (F_i \cdot SA)$$

where

NR_i = normalized release of element i (g/m^2),
 m_i = mass of element i in leachate (g),
 F_i = (wt % of i oxide + 100) (MW of i + MW of i oxide),
 SA = surface area of sample (m^2).

Typically, a 1-cm cube of sample is equilibrated for 28 days at 90°C with a volume of distilled water such that a sample surface area:volume ratio of $10\ m^{-1}$ is maintained. Figure 12 indicates the results of the MCC-1 tests on both phases and compares their performance to two glass waste forms that were developed for high-level waste disposal. The low normalized releases illustrate that the field-produced waste form exhibited excellent durability. The MCC-1 tests illustrated in Fig. 12 were performed at 90°C using deionized water. When the tests were repeated at 22°C using an ORNL streamwater sample, the normalized releases of Cs and Sr were reduced over an order of magnitude to $<0.2\ g/m^2$. These lower values are probably more useful for predicting waste form leaching performance in the field.

2.1.4 Discussion

Perhaps the greatest concern and justification for an actual "hot" (radioactive) test derives from the volatilization of 0.12% of the Cs within the melted mass. Such Cs retention within the melt is an excellent starting point, but Cs retention needs to be improved during an application of ISV to seepage trenches. If ISV were applied to liquid waste seepage trench 7 with process performance similar to that achieved for this cold demonstration, a significant radiological hazard would be encountered due to volatilization of ^{137}Cs . Application of ISV to Trench 7 would probably require about ten settings of hood and electrodes to complete the entire trench volume which contains a total inventory of about 100,000 Ci of ^{137}Cs . Thus, each melt setting would be dealing with about 10,000 Ci. If only 99.88% of this were retained in the melt, then approximately 12 Ci of ^{137}Cs would enter the process off-gas, and most of this would be contained in scrub solutions (Table 3). Without performing any dose calculations, this clearly represents a significant hazard for the process operators and would also produce a significant amount of waste for disposal by other means. It is highly desirable to achieve a net melt retention of 99.999% of the ^{137}Cs . Such a net retention would bring only 0.1 Ci into the process trailer, which could be accommodated without extreme shielding or waste handling precautions.

2.2 VOLATILIZATION AND LEACHABILITY OF CESIUM FROM VITRIFIED SOILS WITH SODIUM AND LIMESTONE AMENDMENTS

There are several possible methods by which such a larger melt retention of ^{137}Cs might be achieved. First, unlike the cold test where elemental Cs analyses in the process off-gas scrub samples were, of necessity, determined after the demonstration, real-time monitoring of

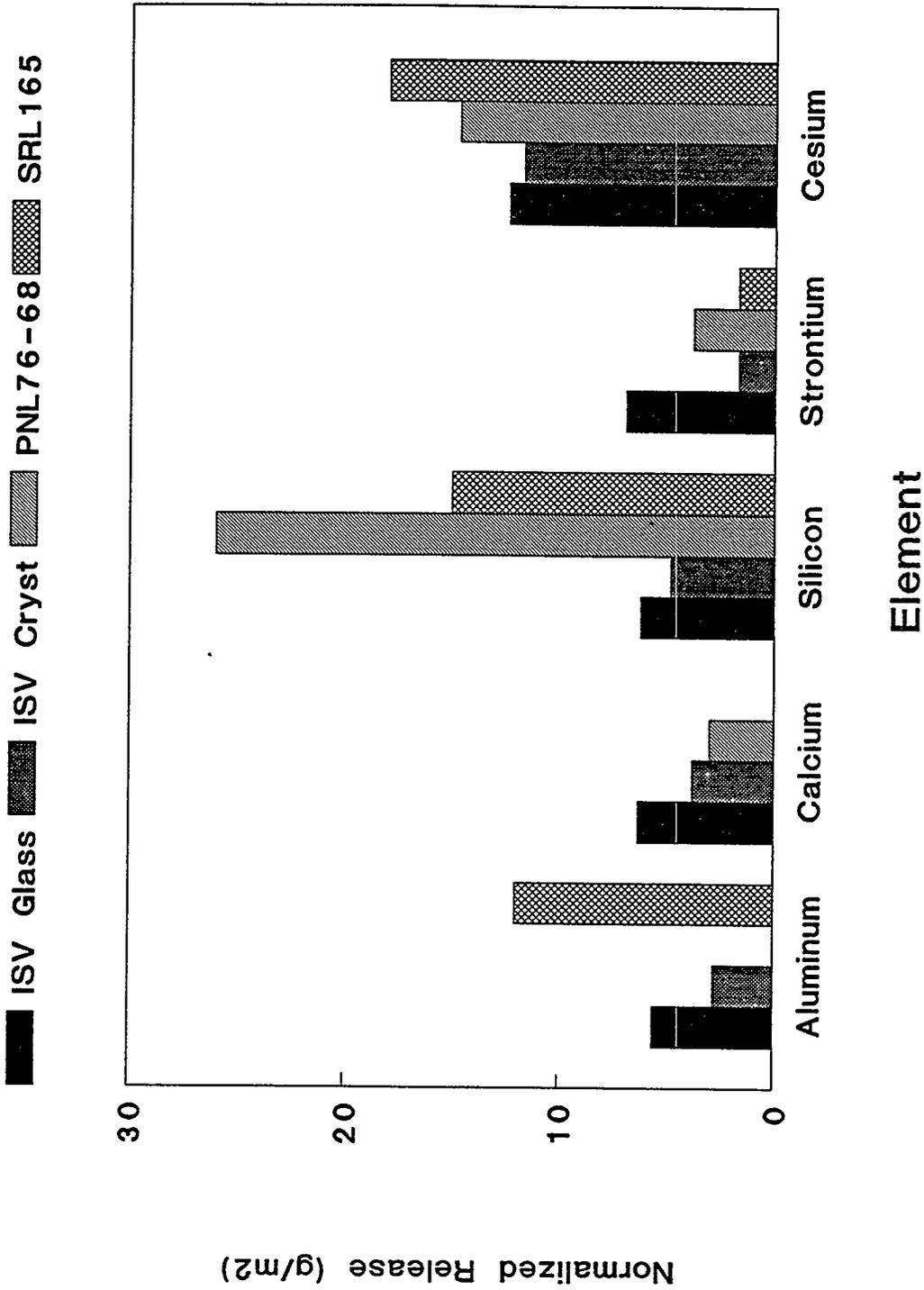


Fig. 12. Comparison of normalized release from MCC-1 leach testing for ORNL pilot-scale ISV products and some high-level nuclear waste glasses (PNL 76-78 and SRL-165).

Table 3. Hypothetical distribution of ^{137}Cs activity in ISV off-gas system after vitrifying 10,000 Ci

Off-gas system component	Amount of ^{137}Cs (Ci)	Amount per unit area ($\mu\text{Ci}/\text{cm}^2$)
Hood	0.079	0.24
Ducting	0.401	5.5
Scrub solutions	11.74	0.5 ($\mu\text{Ci}/\text{mL}$)
Primary HEPA filter	0.293	1.46
Secondary HEPA filter	0.008	0.04

^{137}Cs within the process trailer is available and necessary during a hot test. The process trailer is equipped with several radiation monitors within the off-gas train. If significant volatilization of ^{137}Cs were observed, then power to the melt could be lowered. The volatilization of ^{137}Cs is highly temperature dependent, and power regulation can be employed to regulate melt temperature. Most Cs volatilization probably occurs in hot spots near the electrodes where power density and, hence, temperatures are greatest. Thus, a hot demonstration will provide a test of the feasibility of such a feedback control procedure to minimize ^{137}Cs volatilization while maintaining melt progress. It should be noted that the method used in the cold demonstration did not attempt to minimize Cs volatilization because power delivery to the melt was maximized at all times. Thus, this cold test can be interpreted as measuring the maximum Cs volatilization potential were the ISV process applied to a trench containing actual radioactive waste.

Several possible modifications may also help to minimize Cs volatilization during ISV and could be tested during a hot demonstration. Because the ORNL seepage trenches contain crushed limestone to facilitate liquid seepage, they have significant voids, probably about 47% of the trench volume, into which glass forming or modifying materials might be introduced. Of particular interest would be materials that could produce a glass of significantly lower melting temperature. Sodium hydroxide would be a promising choice as an additive because it could be introduced as a liquid solution (e.g., 50%) and provide enough Na to yield a glass of significantly lower melting temperature. In addition, the introduction of NaOH could provide an electrically conductive path between the electrodes within the trench so that surface melt initiation could be obviated. A subsurface melt initiation has the additional potential benefit of placing a cold cap between the melt and the surface to aid in particulate and ^{137}Cs removal

from the off-gas. Potential benefits from a subsurface melt initiation and a lowering of glass melt temperature will be tested in the laboratory before actually being incorporated into a hot field demonstration. A net reduction in the ambient aboveground inventory of ^{137}Cs during an ISV test could also be attained by recycling scrub solution to the bottom of the trench via an angled well driven into the test trench. Thus, only the ^{137}Cs volatilized during the very last stages of an ISV run would be retained within the process trailer and result in waste. The remainder would be incorporated into the melted mass as it engulfed the well discharge point(s) at the trench bottom. Thus, many techniques may be developed, or applied in combination, to increase the net retention of ^{137}Cs within the melted mass during ISV.

A study was undertaken to quantify the effects of Na_2O amendment of soil-limestone mixtures on the volatilization of Cs and to quantify any changes in the leachability of the resulting waste form. This study also addressed the effects of Na_2O amendment on the melting range of these materials. Additional work is under way to quantify the effects of composition (soil, CaO , and Na_2O) on the viscosity and electrical conductivity of molten materials at various temperatures. All of this work is aimed at devising an optimal Na_2O amendment method and level for the radioactive ISV demonstration planned for the summer of 1990.

2.2.1 Methods and Results

2.2.1.1 Vitrification Materials

The sources of the soil and limestone used throughout this investigation, have been previously described (Spalding and Jacobs 1989b). The elemental compositions of these materials are presented in Table 4, while details of the analytical procedures are available in the original reference. All materials were dried at 80°C to constant weight and sieved to -10 mesh (<2 mm).

2.2.1.2 Effect of Soda Ash on Melting Point of Soil-Limestone Mixture

A suite of six soil-limestone mixtures was prepared in Pt crucibles to which were added increasing amounts of soda ash (Table 5). The 7:3 weight ratio of soil:limestone was selected because this composition was observed in the field demonstration of ISV at ORNL (Spalding and Jacobs 1989b). All crucibles were then heated to 1000°C for 1 h. (Starting from room temperature, the furnace usually required about 2 h to attain this temperature.) The samples were lowered out of the furnace and allowed to cool to room temperature. Samples were then observed for several properties to determine if vitrification had occurred. Manual probing with a spatula determined whether or not the sample had fused depending on whether the particles cohered. Melting was inferred when the appearance no longer exhibited individual grains but rather a vitrified cohesive mass. The completion of melting was judged based on whether or not grainy inclusions were observed in the vitrified material. After making the observations on sample melting, the samples were returned to the furnace for 1 h at 1050°C , removed,

Table 4. Bulk chemical composition of limestone and soils used for preparing vitrified materials

Oxide	ISV limestone	ISV soil	¹³⁷ Cs soil
SiO ₂	5.41	57.34	55.8
Al ₂ O ₃	1.58	18.35	14.1
TiO ₂	0.06	0.81	nd
Fe ₂ O ₃	0.70	6.23	5.7
MgO	3.65	1.83	1.3
MnO	0.02	0.09	0.1
CaO	50.37	2.53	2.5
SrO	0.06	0.01	nd
Na ₂ O	0.04	1.03	0.4
K ₂ O	1.02	3.76	nd
P ₂ O ₅	0.07	0.24	nd
CO ₂	40.06	--	--
H ₂ O	--	7.64	5.5
Total	103.05	99.86	85.5
¹³⁷ Cs (dpm/g)	nd	nd	16280
⁹⁰ Sr (dpm/g)	nd	nd	384

nd = not determined.

Table 5. Sample compositions for soda ash amendment tests

Sample number	Weight of soil (g)	Weight of limestone (g)	Weight of Na ₂ CO ₃ (g)	Calculated Na ₂ O added ^a (%)
1	1.400	0.600	0.000	0
2	1.400	0.600	0.213	7.2
3	1.400	0.600	0.427	13.5
4	1.400	0.600	0.855	23.9
5	1.400	0.600	1.710	38.5
6	1.400	0.600	3.420	55.6

^aFraction Na₂O added = (weight of Na₂CO₃ x 0.5848)/[(weight of Na₂CO₃ x 0.5848) + (weight of soil x 0.90) + (weight of limestone x 0.5603)].

cooled, and observed again. The process was repeated in increasing steps of 50°C until a temperature of 1400°C was reached. Results are given in Table 6.

Even modest additions of Na₂CO₃ (i.e., 13.5%) depressed the apparent melting point of 7:3 soil:limestone from 1250°C to 1050°C, and addition of 23.9% soda ash produced a material that melted below 1000°C. Thus, addition of soda ash can produce a significantly lower melting material and, therefore, an ISV processing temperature significantly below, by about 200°C, that for a melt not amended with Na₂O. However, the concept of melting at a particular temperature is an oversimplification of the vitrification process. Melting occurs over a significant range of temperature and is more accurately described by a temperature at which a given material exhibits a certain viscosity (e.g., the T100P, the temperature of a 100-poise viscosity). Additional work is being performed to determine composition effects on viscosity and electrical conductivity of vitrified soil and limestone mixtures. The T100P of a 7:3 ORNL soil:limestone has been measured at 1325°C (Carter et al. 1988), while the T100P of neat ORNL soil has been estimated to be 1720°C (Carter et al. 1987). Thus, the blend of limestone and soil in the ORNL seepage trenches results in a material that is significantly lower in melting and processing temperature than an unamended soil without limestone inclusions. An additional amendment with soda ash or other sodium source should further reduce the T100P and, therefore, the ISV operating temperatures. Obviously, a great excess of soda ash could be added to a soil, similar to amounts used for soil fusion for total elemental analyses; typically, a 4:1 ratio of soda ash to soil is employed for complete fusion and subsequent dissolution in acid (Lim and Jackson 1982). Although soil fusions with soda ash are typically low melting (1000°C or less), they are relatively soluble and would not represent a particularly durable waste form. The question of how much sodium to add to a soil:limestone mix depends on (1) the significance of the deterioration in waste form durability with increasing Na₂O content and (2) any effect of the added Na₂O on the volatilization of ¹³⁷Cs present in the soil.

2.2.1.3 Assay Procedures for ¹³⁷Cs, ¹³⁴Cs, and ⁸⁵Sr and Extraction Procedures Following Thermal Treatments

Prior to any thermal treatment, a 1-g sample of material (usually 0.700 g of soil and 0.300 g of crushed limestone) was placed into a crucible into which was mixed the desired amount of amendment (usually NaOH solution or Na₂CO₃). If the amendment was a solution, the crucible was dried at 105°C overnight or to constant weight. The activity of the material was then determined as described here. All thermal treatments were carried out in 12-mm-diam by 25-mm-high cylindrical crucibles (2-mL capacity) made of 99.8% pure recrystallized alumina (Coors Porcelain Company). Generally, duplicate samples were run for each material at each temperature. Each tared crucible was weighed with its contents before and after each thermal treatment; blank crucibles were carried through an overnight 1600°C treatment and no detectable weight loss was observed. For temperatures above 1100°C, the crucible was placed inside a larger alumina crucible fitted with a lid for thermal

Table 6. Effects of soda ash on melting of 7:3 soil:limestone mixture

Added Na ₂ CO ₃	Temperature																	
	1000°C			1050°C			1100°C			1150°C			1200°C			1250°C		
	Fu	Mt	Com	Fu	Mt	Com	Fu	Mt	Com	Fu	Mt	Com	Fu	Mt	Com	Fu	Mt	Com
0Z	No	No	No	No	No	No	No	No	Yes	Yes	No	Yes	Yes	No	Yes	Yes	Yes	
7.2Z	Yes	No	No	Yes	No	No	Yes	Yes	No	Yes	Yes	No	Yes	Yes	Yes	Yes	Yes	
13.5Z	Yes	No	No	Yes	Yes	No	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	
23.9Z	Yes	Yes	No	Yes	Yes	Yes	Yes	Yes										
38.5Z	Yes	Yes	No	Yes	Yes	Yes	Yes	Yes										
55.6Z	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	

Fu = fused, Mt = melted, and Com = completely melted (one vitrified phase).

treatment. This secondary containment was necessitated by the tendency of the small crucibles to crack or occasionally shatter during cooling. Most melted materials did not appear to react significantly with the alumina crucible but melted materials did adhere strongly to the crucible walls. Presumably, the differential contraction of the alumina and its adhering vitrified material caused the stresses that resulted in the occasional shattering. After thermal treatment, samples were cooled and recounted for gamma activity as described above. Gamma activity determinations, with and without containment in alumina crucibles, exhibited no difference in counting efficiencies. Thus, shattered crucibles, when all fragments had been contained and collected, were assumed to be counted with the same efficiency as intact specimens. For amendments to soil of CaCO₃, Na₂CO₃, and NaOH, a calcination temperature of 850°C for 4 h was found by trial and error to avoid frothing and spilling of material from the crucible during thermal treatment and to attain the maximal weight loss indicative of near-complete calcination. Early trials with Na₂CO₃ amendments did not exhibit much frothing during melting, and the need for calcination only became apparent in the first trials with NaOH amendments; NaOH-amended samples would often overflow their crucibles without a calcination pretreatment. Routinely, unless noted otherwise, this calcination procedure was applied to all samples prior to their actual thermal treatment at higher temperature. Following this calcination, gamma activities were assayed again to determine if any volatilization had occurred.

Activities of ¹³⁷Cs was determined by NaI-detector gamma ray spectroscopy with 1-g samples contained in 2-mL alumina crucibles contained within 25-mL plastic scintillation vials. Counting times were typically 30 min per sample, and the multichannel analyzer contained a program that subtracted baseline counts in the photopeak for the ¹³⁷Cs region of interest. Standards for ¹³⁷Cs in soil were prepared, and counting efficiencies on a given day ranged between 10 and 12%. Activities of ¹³⁴Cs and ⁸⁵Sr were determined from the net counting rates in photopeak windows for their respective regions of interest. For both ¹³⁴Cs and ⁸⁵Sr, the net counting rates were proportioned to the same standard amounts of radioisotopic solution during each counting period

so that calculations of counting efficiencies and decay corrections were not required.

After final thermal treatment and assay for residual radioactivity, the crucibles were placed in 30-mL "Oak Ridge" polycarbonate centrifuge tubes. Into each tube was placed 20.0 mL of 0.1 N HCl and the contents shaken lengthwise for 16 h at 100 oscillations per min at ambient laboratory temperature. The tubes were then centrifuged at 5000 rpm (3550 relative centrifugal force) and the clear supernatant decanted into polyethylene scintillation vials with polyethylene-lined caps. An additional 20 mL of 0.1 N HCl was added to the centrifuge tube and the contents shaken for an additional 1 h. A second extract of each sample was then prepared as above. Sequential extractions were repeated three more times on each sample, for a total of five 20-mL extracts from each sample. All 20-mL extracts were assayed for levels of ^{137}Cs , ^{134}Cs , or ^{85}Sr as described above. Fractional amounts of radioisotope extracted from each sample were calculated from the summation of the amounts in the five sequential extracts to the amount in the sample prior to extraction but after thermal treatment.

2.2.1.4 Effects of Soda Ash and Caustic Soda Amendments on ^{137}Cs Volatilization

Samples containing 0.3 g of crushed limestone and 0.7 g of the ^{137}Cs soil were amended with 0, 0.106, 0.213, 0.319, 0.427, or 0.533 g of dried Na_2CO_3 ; these correspond to equivalent Na_2O additions of 0, 7.2, 13.5, 19.0, 23.8, and 28.1%, respectively. The percentage of Na_2O additions were calculated on an after vitrification weight basis; the soil dehydrates to 90% of its starting weight, the limestone to 56.03% of its weight, and the soda ash to 58.48% of its weight. Additional samples containing 0.3 g of crushed limestone and 0.7 g of the ^{137}Cs soil were amended with 0, 0.105, 0.211, 0.316, 0.422, or 0.526 mL of a 50% by weight solution of NaOH (specific gravity = 1.525 g/mL); these correspond to equivalent Na_2O additions of 0, 5.6, 11.2, 16.8, 22.5, and 28.0%, respectively. These percentage of Na_2O additions were also calculated on an after-vitrification weight basis where the NaOH dehydrates to 77.48% of its weight. All samples were heated to 850°C for 4 h before final thermal treatment at either 1200°C or 1400°C for 18 to 21 h (overnight). Results are depicted in Fig. 13.

Several conclusions can be drawn from this information. First, increasing additions of Na_2O definitely result in increased volatilization of ^{137}Cs . One sample at the highest level of Na addition lost over 40% of its ^{137}Cs . In the unamended condition, all samples exhibited an apparent loss of about 5% of their ^{137}Cs . This latter, no amendment, treatment had the composition of the vitrified material in the field demonstration (Spalding and Jacobs 1989a) in which 0.12% of the Cs_2CO_3 added to the pilot-scale trench was volatilized. A simple extrapolation, based on 0.12% volatilization in the field being equivalent to 5% volatilization in these crucible tests, would lead to an expectation that a soda ash or caustic soda-amended field test would result in 0.96% volatilization at the highest rate of Na_2O amendment. For the 10% Na_2O amendment level (which is a realistic maximum for a

field amendment as discussed below), approximately 10% of the ^{137}Cs volatilized which, by the same extrapolation, would yield a volatilization only twice that observed in the previous field demonstration (Spalding and Jacobs 1989a). This assumes that the ISV operating temperature in the field would be equivalent to that in the 1987 demonstration. However, the addition of Na to the melt would be expected to reduce operating temperatures by decreasing the viscosity of the material. Although increasing Na additions resulted in increasing Cs volatilization, the effect on melt viscosity is not yet known. Producing an ISV product in the field with more than about 10% Na_2O content may be impractical. The expected 47% voids in the gravel backfill of a typical seepage trench (Spalding and Jacobs 1989b) would only be able to hold a finite amount of Na amendment (e.g., 50% NaOH solution). Given the mass of soil and limestone to be vitrified, the amount of Na_2O that could be added to the trench to adjust the final composition, is limited to 7% assuming all limestone gravel porosity, and no soil porosity, was filled with 50% NaOH.

Second, it apparently makes little difference whether the Na amendment is made as the hydroxide or the carbonate. Although the NaOH did produce more Cs volatilization than Na_2CO_3 at 1400°C , this may be due to reasons other than inherent volatility of ^{137}Cs in the presence of the two chemicals. Obviously, both forms result in the same species (Na_2O) once the $1200\text{-}1400^\circ\text{C}$ temperature is reached. However, the appearance of the two treatments after the 850°C calcination was quite different; the NaOH-amended samples tended to creep up the walls of the crucible much more than the Na_2CO_3 -amended samples. This is perhaps due to the lower melting temperature of NaOH (318°C) than Na_2CO_3 (851°C), which also begins to lose CO_2 at about 400°C . If the NaOH has melted before the calcination of the CaCO_3 , the outgassing of the CO_2 could result in the apparent creep up the crucible as the liquid NaOH is spattered. The net result is that the NaOH-amended samples are distributed over a much larger surface area than the Na_2CO_3 -amended samples. When the thermal treatment at 1200 or 1400°C is subsequently imposed, more surface area for volatilization of ^{137}Cs would be available in the NaOH-amended samples. Nevertheless, the effect is relatively small particularly at levels of Na_2O addition below 15% where the two forms of sodium addition are indistinguishable.

Third, the difference in ^{137}Cs volatilization between 1200 and 1400°C is not significantly different at Na_2O amendments below 20%. This implies that Cs volatilization was more dependent on the amount of Na_2O added than on whether the sample was heated to 1200 or 1400°C . This may also imply that Cs volatilization occurred mainly during the melting process rather than during the prolonged period when the molten melt was standing in the crucible. Previous work with neat soils (without either CaCO_3 or Na_2O amendment) has indicated that Cs volatilization at 1600°C proceeded at the slow rate of 0.36% per day over a 13 day period with most of the volatilization occurring in the first day (Fig. 14). Thus, most Cs volatilization is occurring during outgassing of residual CO_2 and H_2O at the time the sample is equilibrating to its final temperature. This outgassing mechanism for Cs volatilization will be discussed further in subsequent tests.

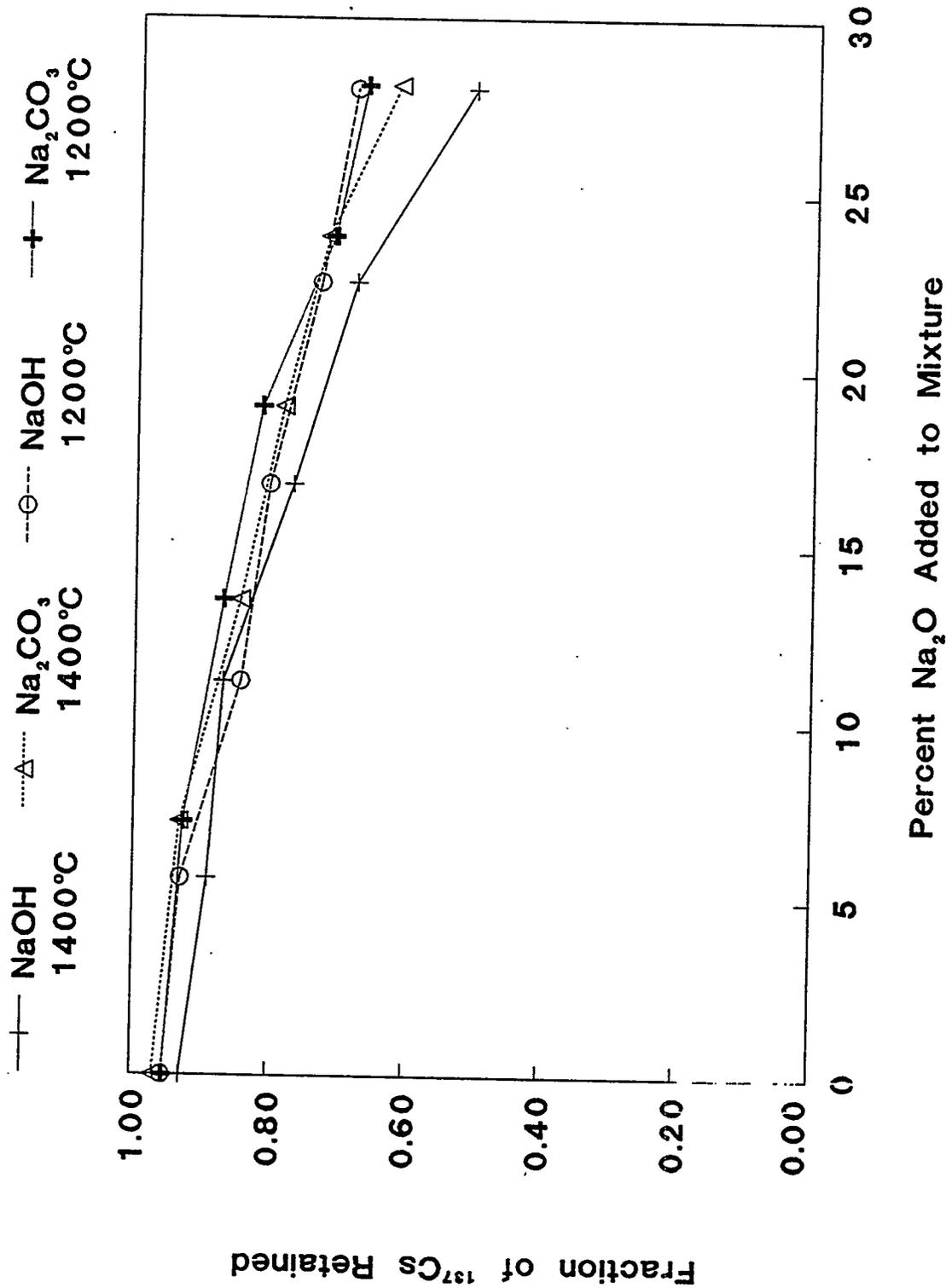


Fig. 13. Retention of ¹³⁷Cs in vitrified soils with additions of NaOH and Na₂CO₃.

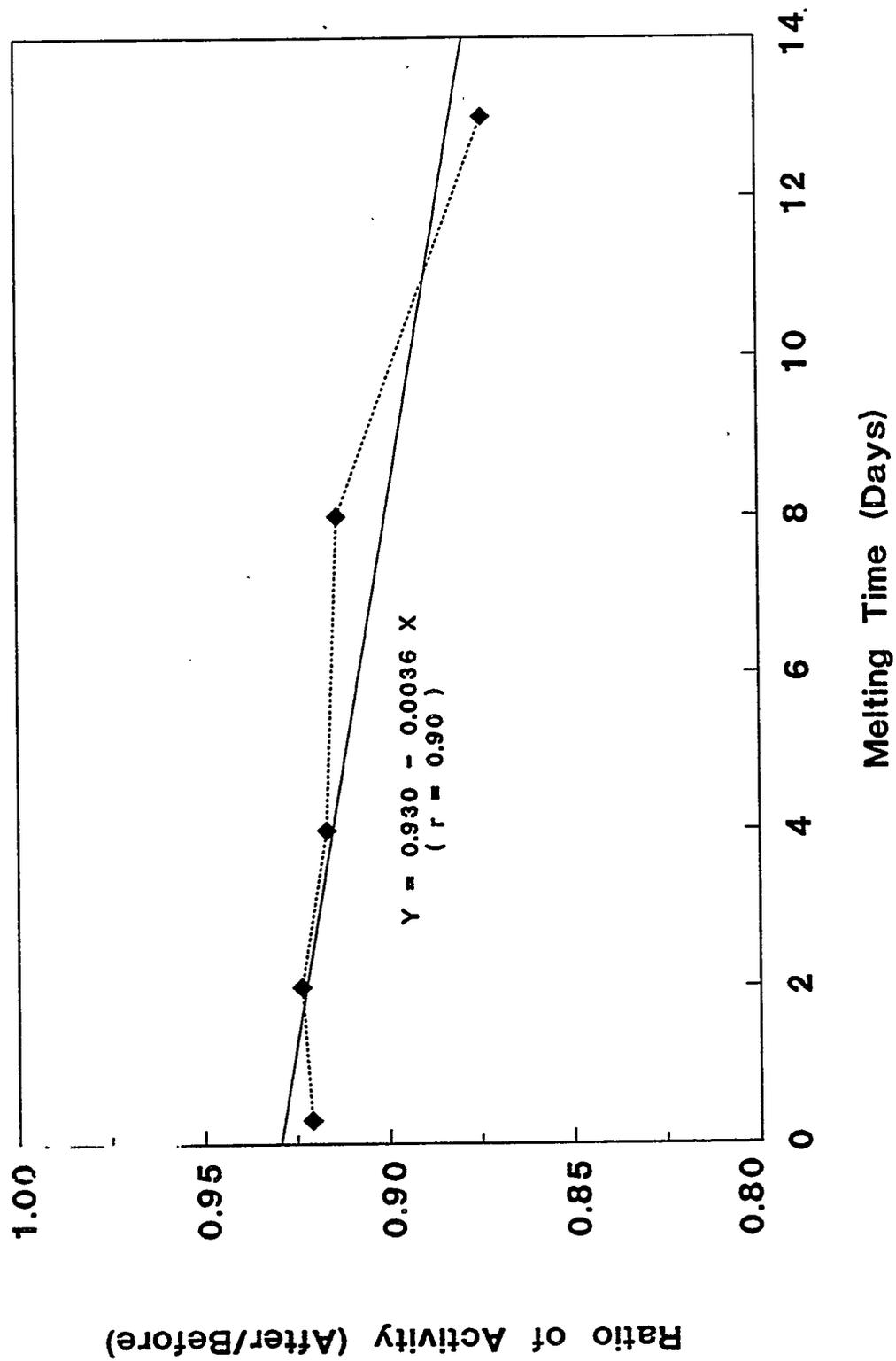


Fig. 14. Retention of ¹³⁷Cs in soil during prolonged melting at 1600°C.

Figure 15 illustrates the reproducibility of the degree of accuracy in the ^{137}Cs volatilization determinations. The standard deviations of triplicate samples for the six levels of Na_2O amendment with NaOH are indicated by the error bars on the graph. In addition, the assays for residual ^{137}Cs in the crucibles following the 850°C calcination treatment indicated that no detectable loss of the radioisotope was observed. Weight loss data after the 850°C treatment indicated that calcination was usually about 90% complete and that the samples were generally fused but not melted. The calcination of CaCO_3 to CaO occurs at about 825°C although the kinetics for completion are not known under the present experimental conditions. The residual calcination of both CO_2 and H_2O from the samples occurs as they are heated to and above their melting points, during which time the ^{137}Cs volatilizes.

The effect of final temperature on ^{137}Cs volatilization is illustrated in Fig. 16. Relatively little difference in Cs volatilization was observed between the 1200 and 1400°C treatments regardless of the amount of Na amendment except at the highest amendment level. This effect can be ascribed to the volatilization of ^{137}Cs as the sintered or calcined porous mixture forms a single-phase homogeneous vitrified product. Whether this occurs at 1200 or 1400°C would not be expected to affect the amount of Cs volatilized because once sample porosity is lost due to homogeneous melting, both surface area and pathways for Cs volatilization are lost. The explanation is also supported by the lower volatilization observed for all samples at 1000°C . After 1000°C treatment, several of the lower Na_2O -amended samples were not melted while the higher Na_2O -amended samples were incompletely melted (i.e., grainy inclusions were still noticeable) unlike completely melted samples which exhibited a homogeneous vitrified phase (Table 5).

The one significant deviation from this explanation of volatilization during melting occurred in the highest Na_2O -amended sample at 1400°C . There was a quite significant increase in the amount of Cs volatilized from this sample and from the sample heated to 1200°C . One explanation for this difference might be that additional outgassing occurred from the sample with the highest Na_2O amendment between 1200 and 1400°C . However, the weight loss data do not show a detectable difference between fractional sample weight losses after the 1200 and 1400°C treatments. An alternate explanation would be that this difference in Cs volatility is due to the difference in temperature at the presumably low viscosity of the melt at this high Na_2O amendment.

Support for the volatilization during melting mechanism can be seen in the behavior of samples undergoing sequential 1200 and 1400°C treatment. (Fig. 17). Initially, this series of samples containing increasing Na_2CO_3 amendments was subjected to a 2-h treatment at 1200°C which produced some ^{137}Cs volatilization. Subsequent treatment at 1200°C for an additional 20 h produced both additional Cs volatilization and weight loss. These samples were not subjected to the 850°C calcination treatment so that the first 2-h treatment at 1200°C was a combined calcination and melting treatment. The samples were next reheated to 1400°C for 2 h during which no significant further volatilization of Cs was observed, particularly at the higher Na_2O

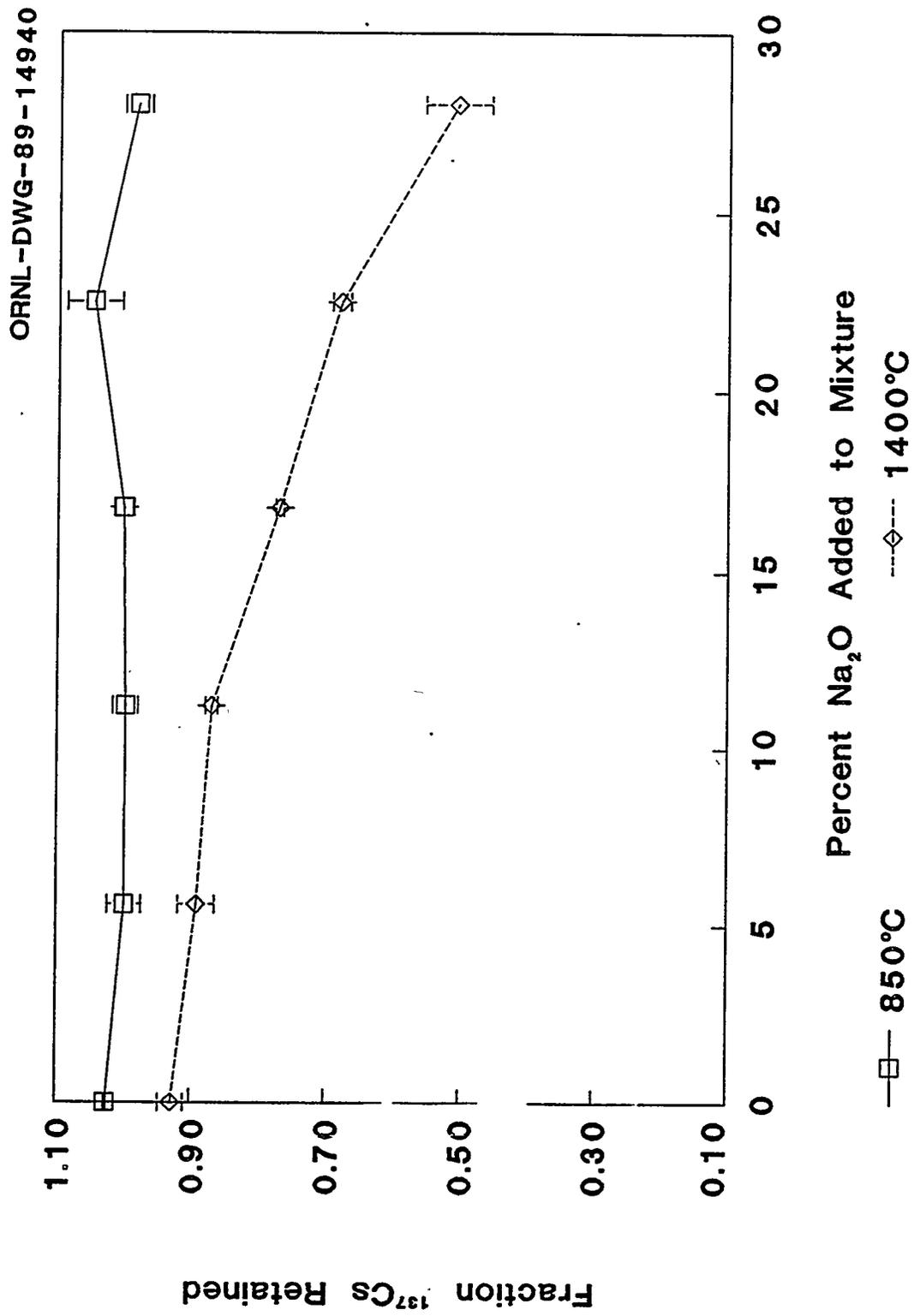


Fig. 15. Retention of ¹³⁷Cs in vitrified soil plus limestone with additions of NaOH.

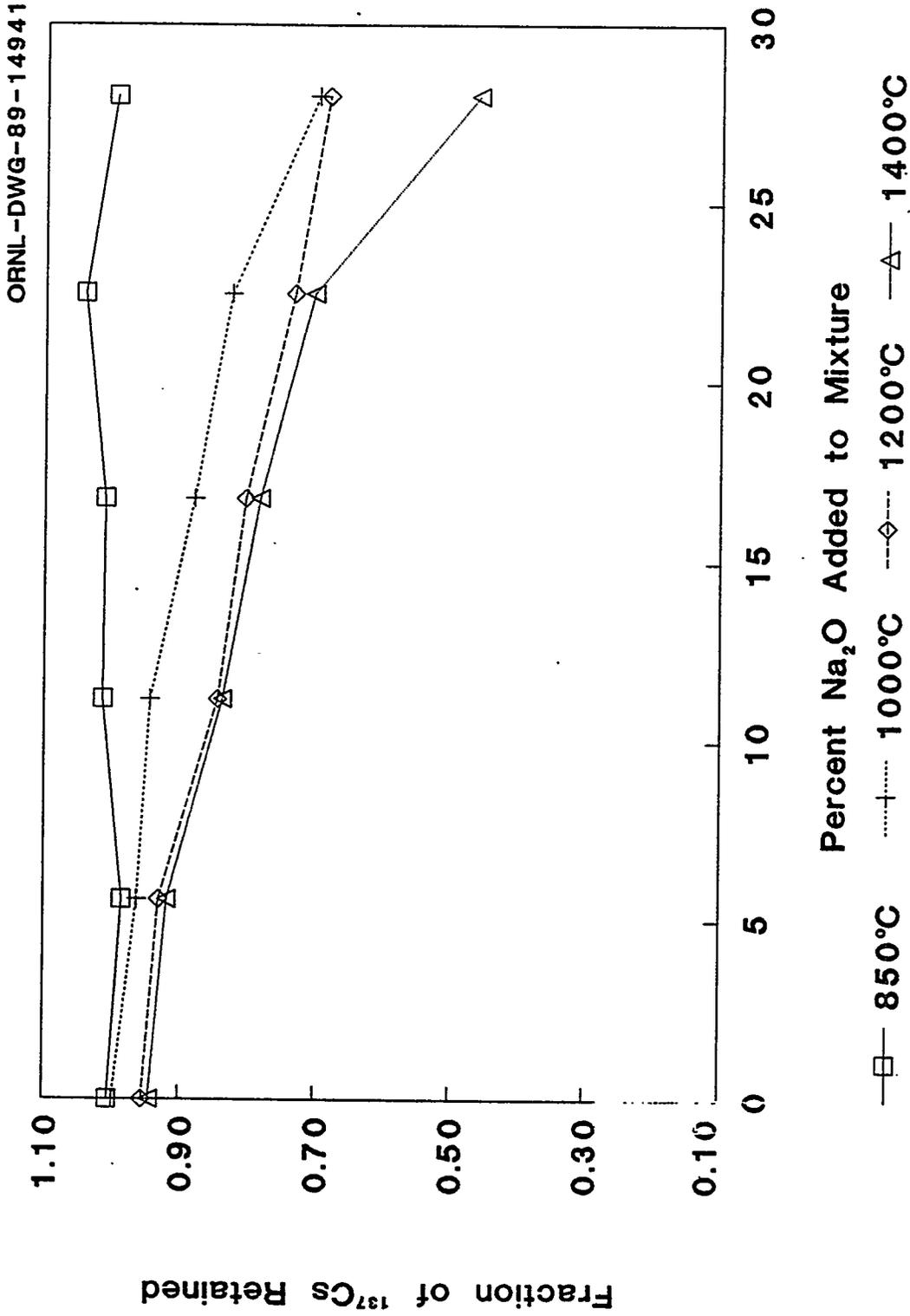


Fig. 16. Retention of ¹³⁷Cs in vitrified soil plus limestone at selected temperatures with additions of NaOH.

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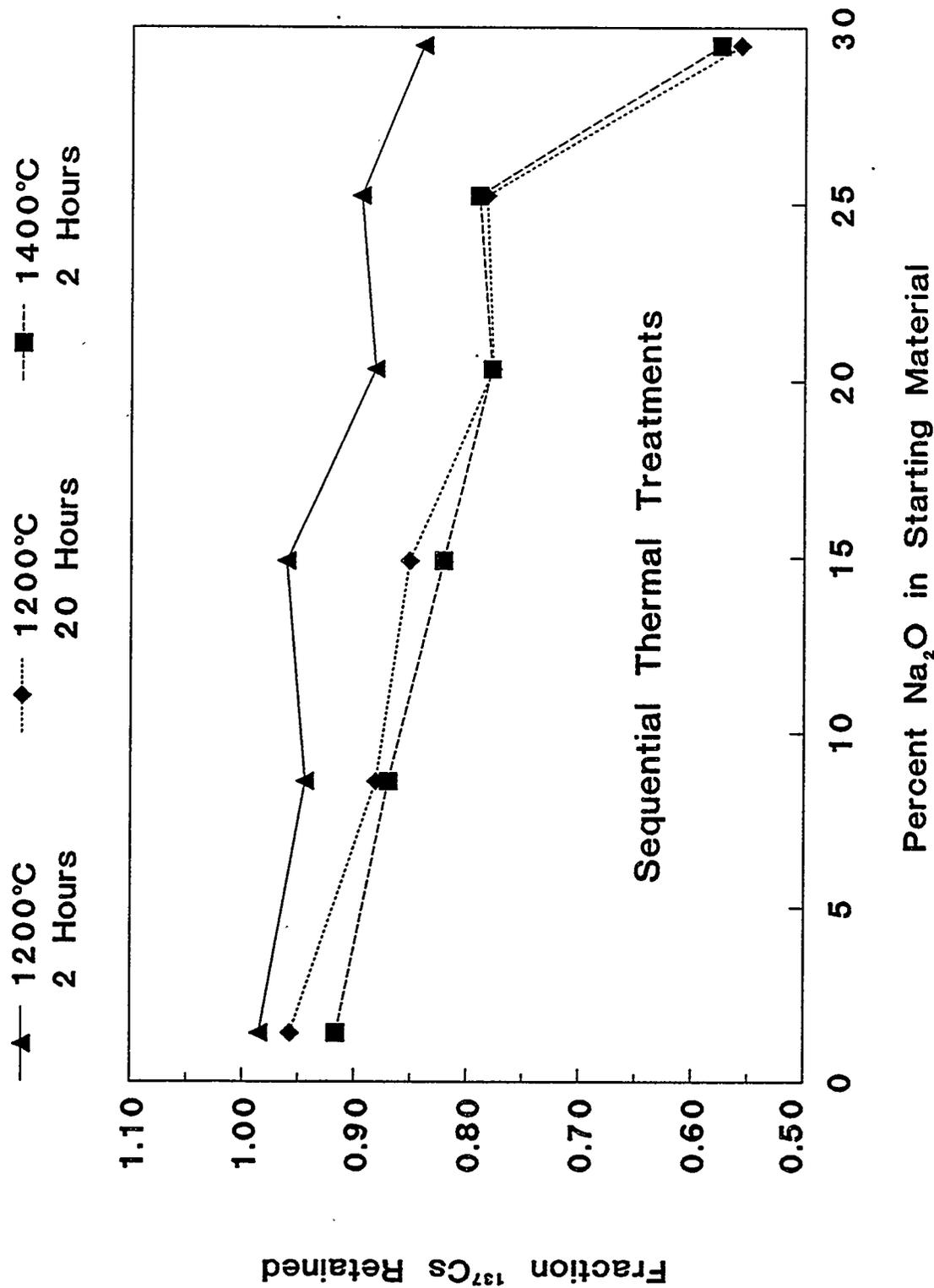


Fig. 17. Retention of ¹³⁷Cs during soil melting with Na₂CO₃ additions.

amendments. The 2-h treatment at 1400°C may not have been adequate for a further volatilization of Cs which might have occurred during an overnight interval. However, the lack of difference between 1200 and 1400°C treatments is consistent with the volatilization during melting mechanism. The volatilization during melting mechanism does not exclude the possibility of a volatilization after melting mechanism also operating. It merely implies that the bulk of the Cs volatilization occurs rapidly with outgassing during melting. A much slower diffusion of Cs out of the molten materials, which is probably affected by the viscosity of the melt, is probably also a concurrent mechanism for Cs volatilization which tends to increase in importance as temperature increases well above the melting point.

The importance of outgassing in the volatilization of ^{137}Cs during melting at 1400°C can be seen when CaO was substituted for CaCO_3 (limestone) in a series of Na_2CO_3 -amended samples (Fig. 18). The amount of CaO used (0.168 g) is equivalent to the 0.300 g of CaCO_3 (the ratio of formula weights of $\text{CaO}/\text{CaCO}_3 = 0.5603$). By substituting CaO for CaCO_3 , less outgassing of CO_2 during calcination and associated effects during melting would be expected. At the higher rates of NaOH amendment this effect becomes significant with a difference of 5% in the amount of ^{137}Cs volatilized at the highest amendment level. Complete elimination of such outgassing effects would not be expected even if completely dehydrated and decarbonated materials were employed. Materials would still have to metamorphose from a porous sintered state to a dense nonporous vitrified state; during this transition, much surface area would be available for Cs volatilization.

2.2.1.5 Effects of Potassium Hydroxide on Cesium Volatilization

To determine if any advantageous effect might be achieved by substituting potassium for sodium as a vitrification amendment, a series of KOH-amended mixtures was prepared. To the usual mixture of 0.7 g of ^{137}Cs soil and 0.3 g of crushed limestone was added 0.052, 0.105, 0.157, 0.211, or 0.263 mL of a 70 wt % solution of KOH (specific gravity = 1.70 g/mL) which was prepared by heating and stirring a mixture of 70 g of KOH with 30 mL of water. The equivalent amounts of K_2O added to the vitrified material calculated to 8.5, 15.0, 21.5, 27.4, and 32.0%, respectively. However, for comparing these K_2O amendments to Na_2O , the amounts were converted to Na_2O equivalent percentages of 5.6, 10.4, 14.4, 18.0, and 21.0%, respectively; the ratio of formula weights of $\text{Na}_2\text{O}:\text{K}_2\text{O}$ is 0.658, and graphical comparisons between K and Na (Fig. 19) should be performed on such an equivalent basis. As is apparent from inspection, K and Na were equivalent in their effects on the volatilization of ^{137}Cs during melting. Considering the greater cost of KOH versus NaOH, both on a unit weight and even greater on an equivalent weight basis, no advantage is apparent.

2.2.1.6 Effect of Na_2O Amendment on Leachability of ^{137}Cs from Vitrified Soil-Limestone Mixtures

After completing the determination of residual ^{137}Cs in the thermally treated soil:limestone mixtures, the samples were subjected to

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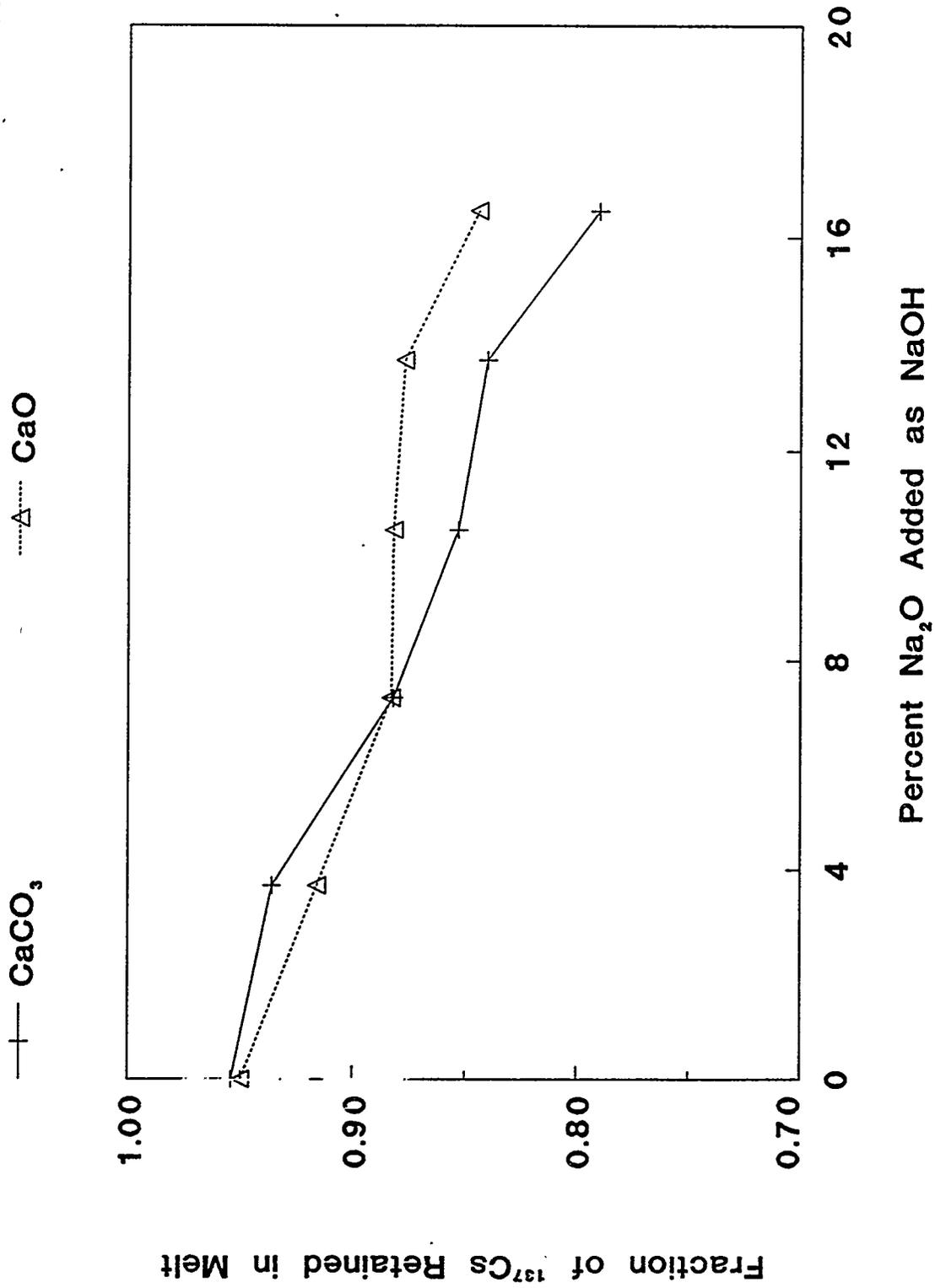


Fig. 18. Retention of ^{137}Cs in 1400°C vitrified soil with NaOH additions, using calcium carbonate and oxide as a source of calcium.

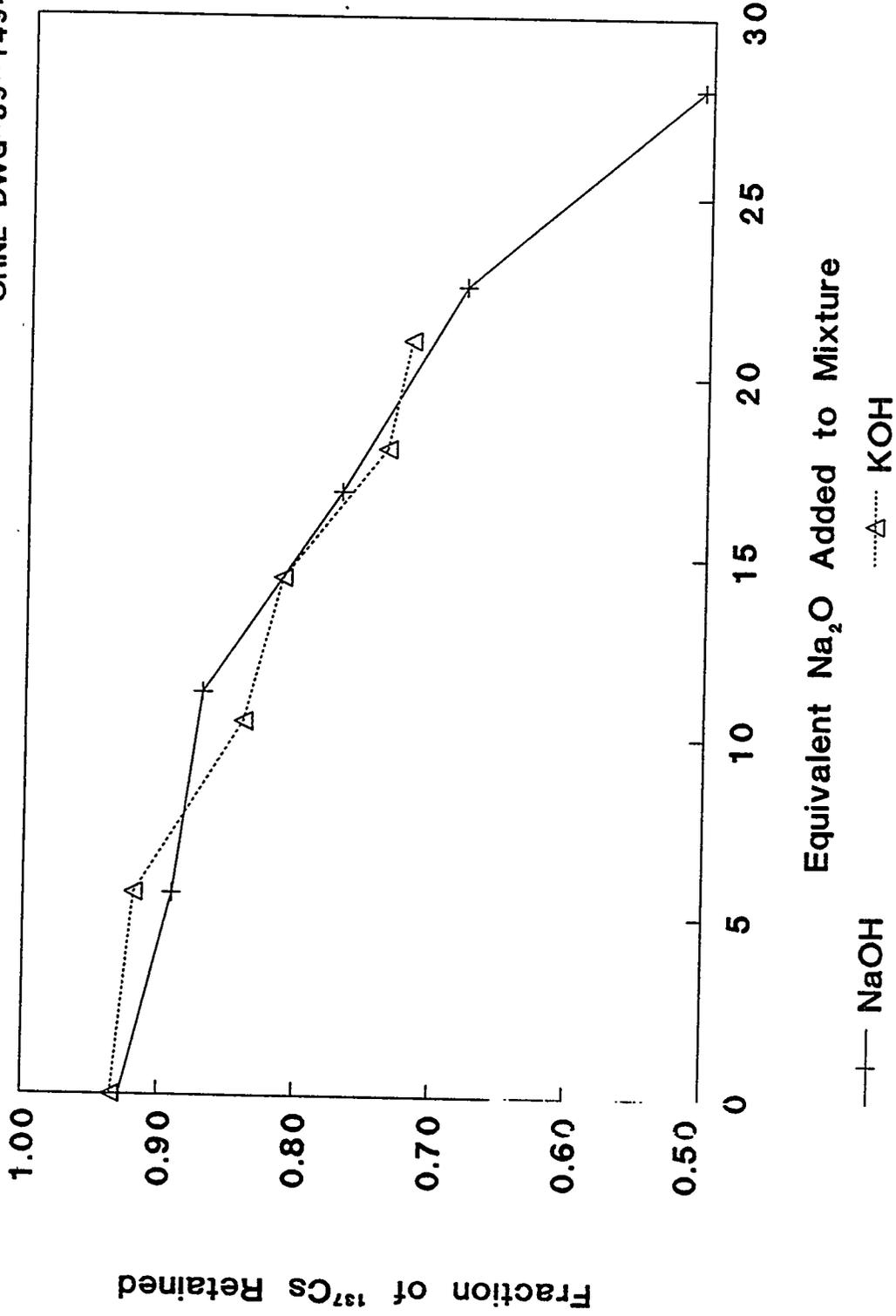


Fig. 19. Effects of KOH and NaOH on the retention of ¹³⁷Cs during vitrification of soil plus limestone at 1400°C.

extraction with 0.1 N HCl as described in the assay procedures (section 2.2.1.3). Results of the percent ^{137}Cs extractable from specimens treated with NaOH are presented in Fig. 20. Regardless of the final temperature of melting, the leachability of Cs was increased significantly by all levels of Na_2O amendment. At the highest Na_2O amendments, leachability was increased by a factor of 20 over the unamended mixture. Some of this increased leachability may be attributed to the higher surface area of samples with higher NaOH amendments as evidenced by the increased creep of the melt up the inside walls of the crucibles. However, this increase in surface area of higher Na_2O -amended samples would, at most, be a factor of 3 and not account for the much larger increase in leachability observed. Quite similar leachabilities were observed with Na_2CO_3 -amended mixtures indicating that the NaOH creep effect must be small (data not presented). Much of scatter in this extractability data can be attributed to the differing amounts of fracturing and cracking of the alumina crucibles; cracked crucibles would expose more sample surface area and, therefore, result in more leaching of the ^{137}Cs into the dilute acid. Nonetheless, the implication of this increase in leachability with increased NaOH amendment is probably not too serious for ISV waste form performance in the field. Standardized leaching tests with groundwater with unamended vitrified material indicated that only 0.01 to 0.04% of the Cs was extractable in 28 days even with a high surface area to leachate volume (Spalding and Jacobs 1989b). In a field situation, where the surface area of ISV product per unit weight is extremely small (i.e., about $10^{-6} \text{ m}^2/\text{g}$), even an increase in leachability of 20 fold would mean very little in terms of releases from the ISV block.

2.2.1.7 Effect of Soil:Limestone Mixture Ratio on Volatilization and Leachability of Cs and Sr During Vitrification

Although all of the previous Na_2O amendment studies were performed on soil:limestone mixtures of 7:3 because this was the ratio found in the field demonstration (Spalding and Jacobs 1989a), it became important to know what the relative volatilization of Cs would be if the ratio of soil:limestone during vitrification should vary. A series of 1-g samples was prepared containing 0 to 100% ISV soil mixed with crushed limestone in increments of 10%. After mixing the contents, a 1.00 mL aliquot of either a ^{134}Cs or ^{85}Sr chloride in tapwater stock solution (10,000 pCi/mL) was added. Samples were then dried at 100°C for 2 h, cooled, reweighed, assayed for gamma activity, calcined at 850°C for 4 h, cooled, reweighed, assayed for gamma activity, vitrified at 1400°C overnight, cooled, reweighed, and assayed for gamma activity. The ^{134}Cs exhibited marked volatilization at limestone compositions above 30% (Fig. 21) with volatilization essentially complete at 100% limestone. Although all samples were fused by the 1400°C treatment, many of the higher limestone compositions were not vitrified by this temperature. Pure CaCO_3 cannot be vitrified near these temperatures; the melting point for CaO is 2572°C , and unless sufficient silicate materials are present with which it can react, it can be considered a recalcitrant material like the alumina crucibles. With insufficient material present

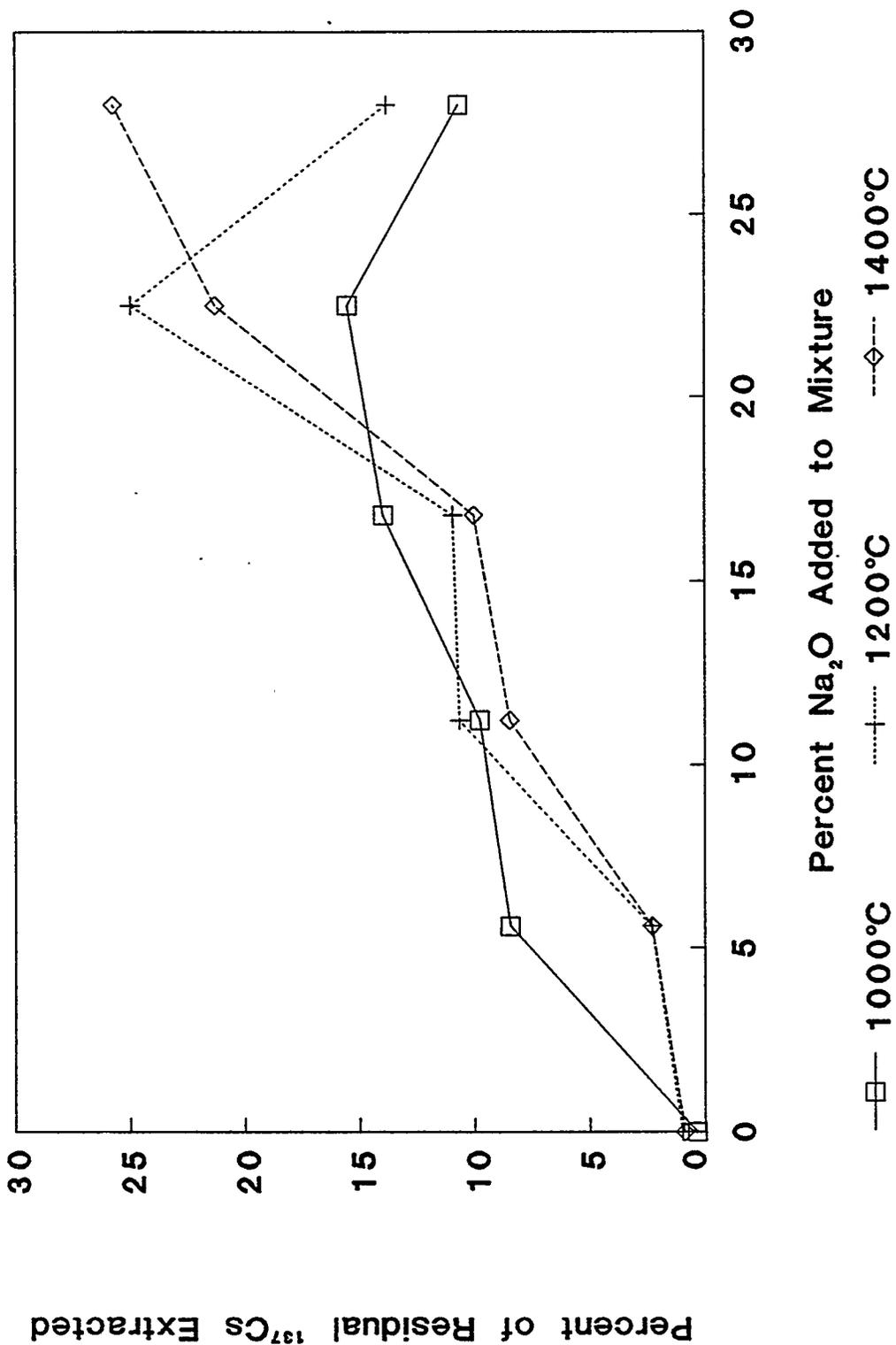


Fig. 20. Extraction of ¹³⁷Cs from vitrified soil and limestone with NaOH amendments.

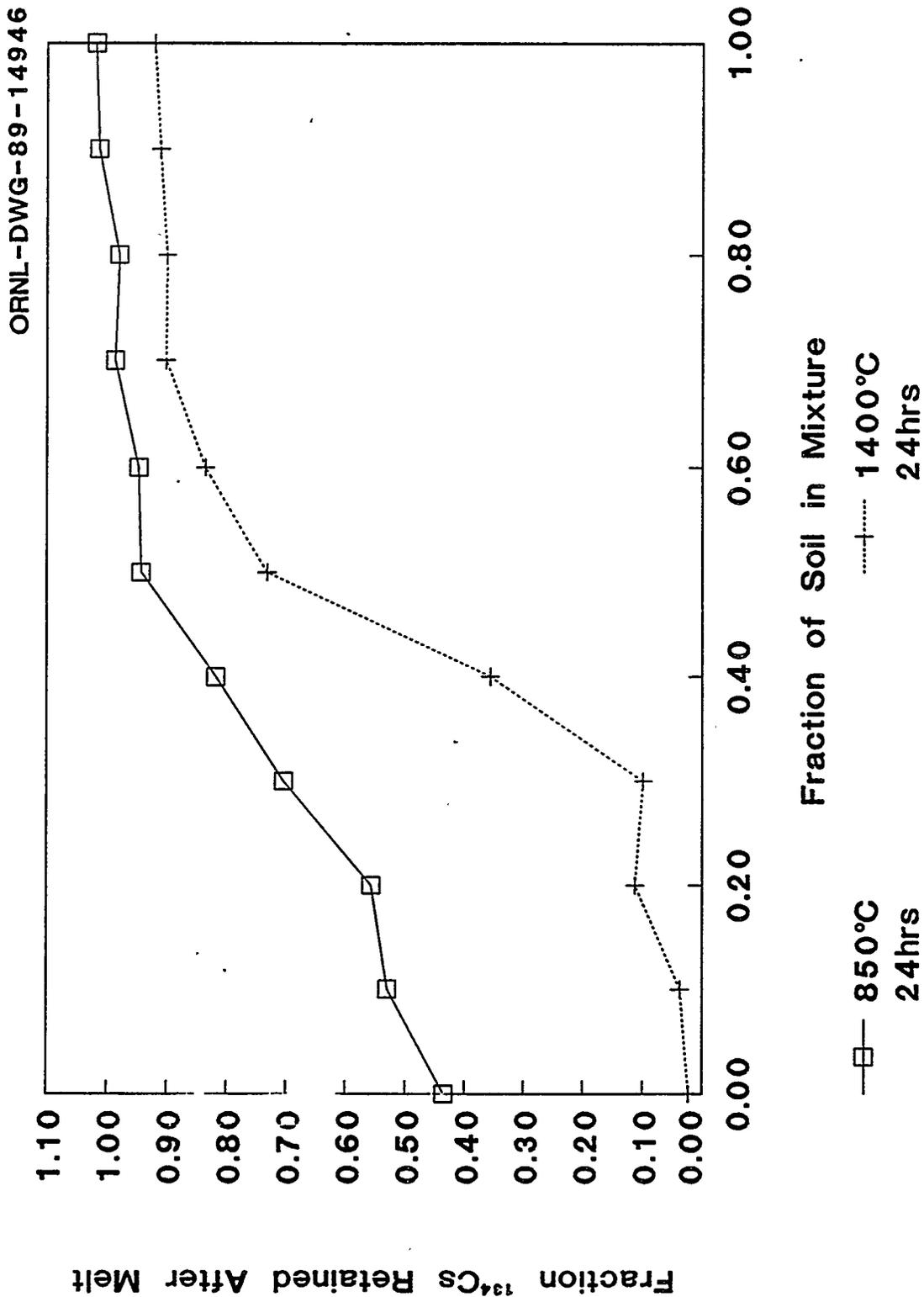


Fig. 21. Effect of limestone-to-soil ratio on the retention of ¹³⁷Cs during vitrification.

material present to form a vitrified product at the high limestone compositions, Cs volatilization from the porous calcined CaCO_3 would not be inhibited by its solubility in the molten liquid. Most Cs salts have low melting and low boiling points (e.g., CsCl melts at 646°C and boils at 1303°C). The volatilization of the ^{134}Cs , even during the 850°C calcination treatment, was quite significant at high limestone compositions. However, as long as limestone composition can be maintained at or below the 30% level, as in the field demonstration, a significant increase in volatilization would not be expected.

In contrast to the behavior of ^{134}Cs , ^{85}Sr exhibited no influence of limestone composition on its volatilization, which was extremely low at all compositions (Fig. 22). This confirms what was observed in the field demonstration: Sr is essentially nonvolatile during vitrification and presents no problems with varying limestone composition. In addition, amendment with Na_2CO_3 up to levels described for the Cs work exhibited no effect on the retention of ^{90}Sr in the vitrified product (data not presented). The leachability of both ^{134}Cs and ^{85}Sr from the various limestone-amended mixtures showed no significant deterioration with limestone compositions below 40% (Fig. 23). The leachabilities of these samples were all significantly lower than Na_2O -amended mixtures (Fig. 19) in which the leachability of Cs approached 25% at the highest Na_2O amendments. Thus, limestone composition variation has significantly less influence on the leachability of Cs or Sr than does variation in Na_2O composition.

2.3 SUMMARY

A field-scale vitrification of a cold (nonradioactive) model of an old seepage trench was completed on July 20, 1987, by personnel of Battelle Pacific Northwest Laboratories in cooperation with ORNL. An estimated 20 tons of soil and crushed limestone backfill were melted. The desired melting depth of 2 m was achieved. The molten mass tended to grow parallel to the long axis of the trench rather than symmetrically around the square array of electrodes. All melt geometry, safety, and process performance objectives were achieved satisfactorily. A 2-month cooling period was required before core sampling of the vitrified product was initiated. Samples of the off-gas scrub solutions have indicated that 99.88% and >99.99% of the nonradioactive Cs and Sr tracers, respectively, were retained in the trench. Samples of the field-produced material were found to be as good or better than two standard high-level nuclear vitreous waste forms when using standard leach tests. Approximately half of the block devitrified or crystallized to pseudowollastonite and anorthite mineral phases during cooling. However, devitrification produced no significant change in the leach properties of the material.

Sodium amendments to mixtures of ORNL soil and limestone have been investigated for their ability to lower melting temperatures required for vitrification in an effort to further reduce the volatilization of ^{137}Cs that would occur during ISV of ORNL seepage trenches. Additions of up to 13.5% Na_2O to 7:3 soil:limestone mixtures reduced the apparent melting range from about 1250°C to 1050°C ; greater additions of Na_2CO_3 reduced melting points below 1000°C but were not quantified. It is

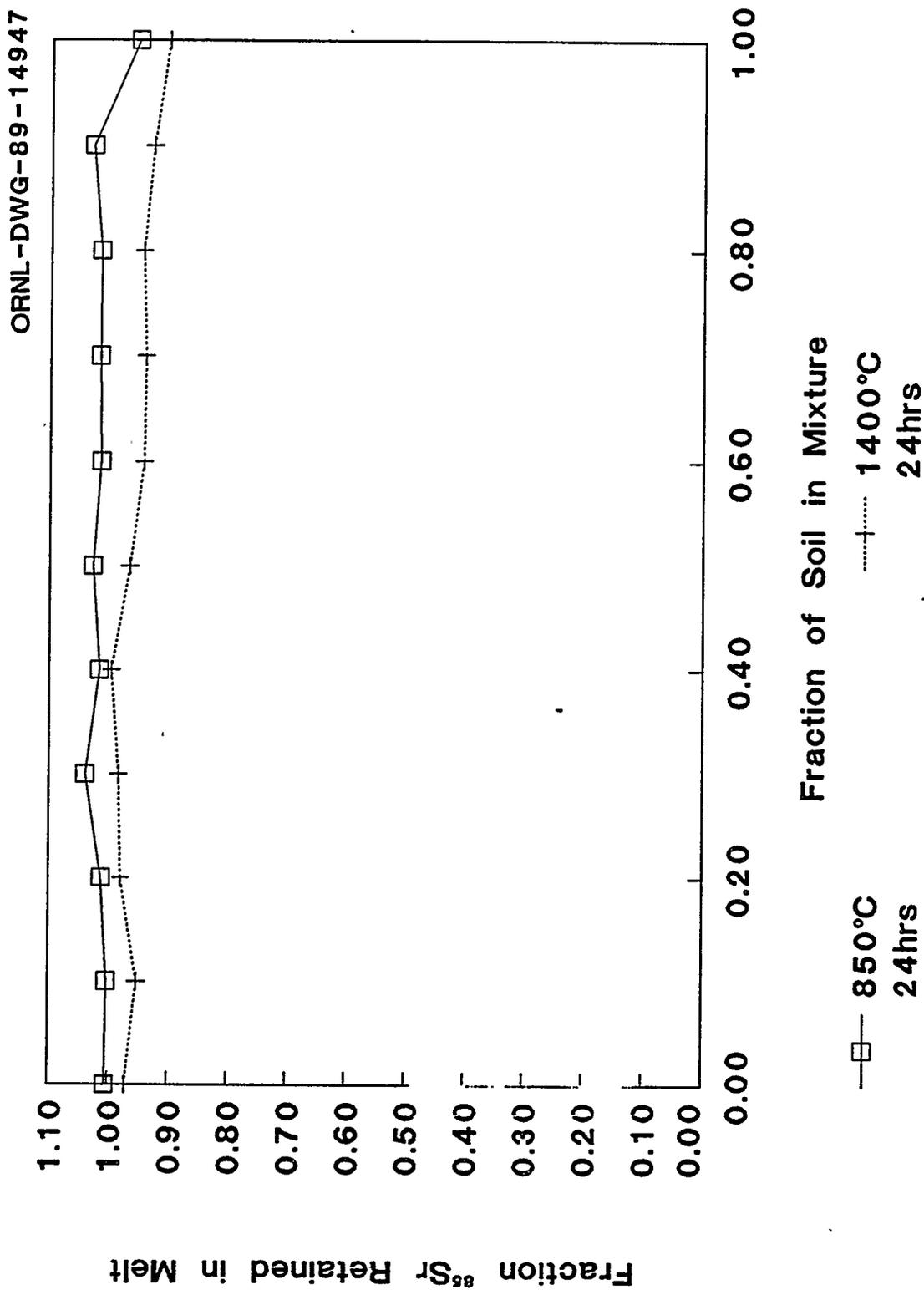


Fig. 22. Effect of soil-to-limestone ratio on the retention of ⁸⁵Sr during vitrification.

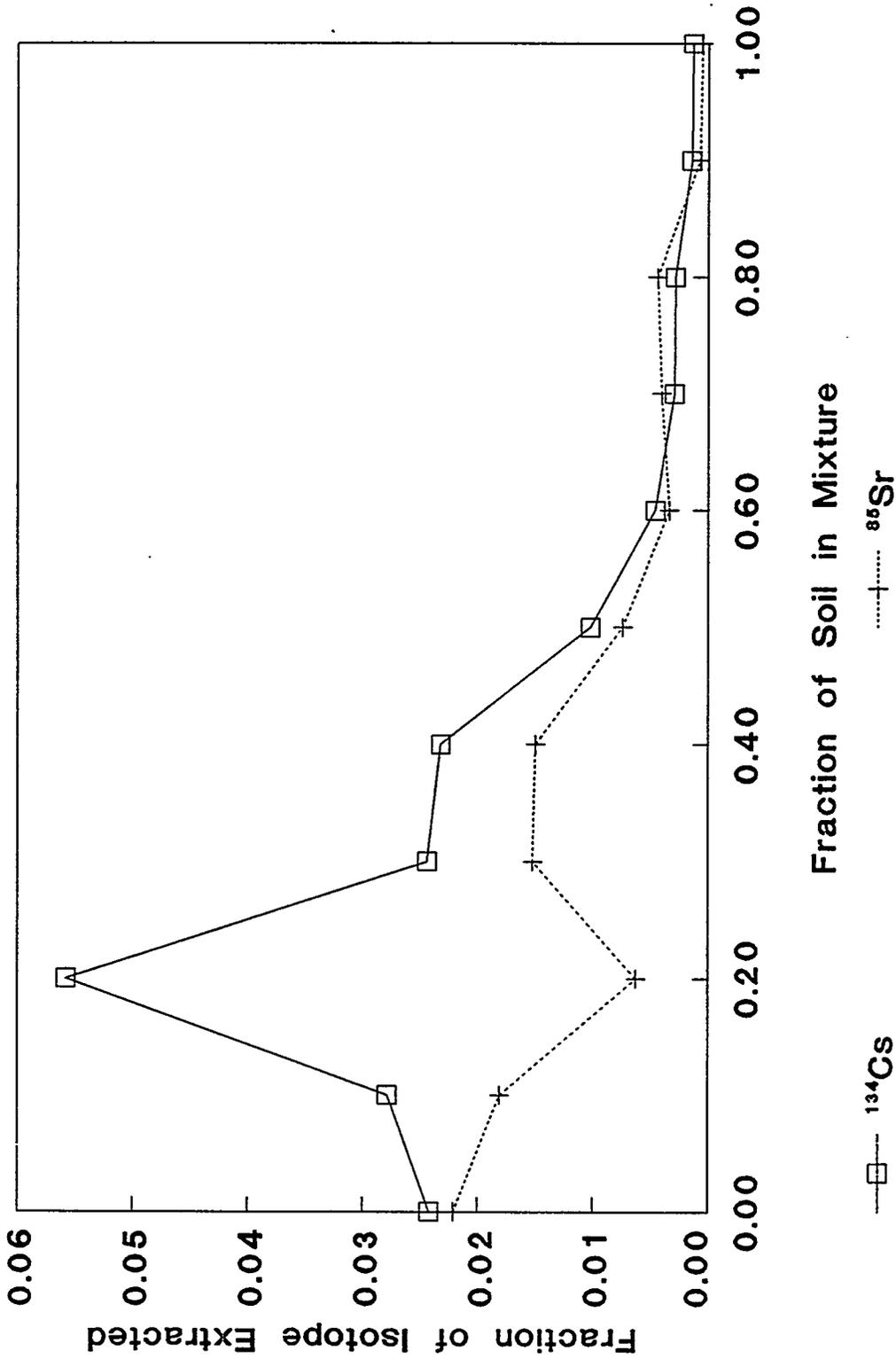


Fig. 23. Extraction of ^{137}Cs and ^{85}Sr from vitrified soil and limestone mixtures.

hoped that this lower melting material would help reduce the volatilization of ^{137}Cs in a planned field demonstration in the summer of 1990 to levels lower than were experienced in a 1987 nonradioactive field demonstration at ORNL. Additions of either NaOH or Na_2CO_3 to the soil-limestone did significantly enhance volatilization of ^{137}Cs during laboratory thermal treatments compared to unamended materials. Although extrapolations to what would be experienced in the field are difficult (5% Cs volatilization was observed in the laboratory for the same melt composition for which only 0.12% volatilization was observed in the field demonstration), only a twofold increase in Cs volatilization would be expected from a 10% Na_2O -amended melt assuming identical ISV processing temperatures. However, because ISV processing temperatures would probably be reduced by about 200°C , Cs volatilization might be reduced even below the 0.12% observed in the field demonstration. Leachability of Cs from Na_2O -amended vitrified products increased significantly over unamended materials. This effect would be minimal on a field-produced ISV waste form where the product has such a small surface area per unit weight.

3. SHALLOW-LAND BURIAL SITE STABILIZATION TECHNOLOGY DEMONSTRATIONS

As part of a burial ground stabilization and closure technology demonstration project, a group of 19 burial trenches in Oak Ridge National Laboratory (ORNL) Solid Waste Storage Area (SWSA) 6 was selected as a demonstration and test area. The purpose of this investigation, known as the Test Area for Remedial Actions (TARA) project, is to (1) identify promising trench stabilization and closure techniques applicable to the ORNL setting, (2) carry out these stabilization and closure techniques on a field scale using actual low-level waste trenches, and (3) collect the necessary data to evaluate each technique as to its feasibility, effectiveness, and cost. In this manner, the ORNL Remedial Action Program will be in a position to select trench stabilization and closure alternatives that have been proven to be successful at the ORNL site.

3.1 SITE SELECTION

The site chosen for the TARA project lies on a small hillock in the northeastern corner of SWSA 6 (Fig. 24). The site was selected based primarily on the following two criteria: (1) it was away from operational areas within SWSA 6 and would, therefore, not interfere with daily waste management activities or vice versa, and (2) it was located entirely on high ground and is thus isolated hydrologically from any peripheral recharge areas which would complicate performing a site water budget and performance monitoring as part of the stabilization and closure evaluation. In addition, the water table at the site is at least 9 m below the bottoms of the trenches, and the trenches in the area are, therefore, unsaturated throughout the majority of the year in contrast to being chronically or seasonally inundated as is the case with other trench areas within SWSA 6. The site also contains some of the first burial trenches used in SWSA 6 which contain higher-level (>200-mrem/h) low-level solid waste.

Initially, plans were made to compact 19 waste trenches located at the TARA site. As planning progressed and cost estimates were made for quantities of grout and construction of a cover that included the entire hillock, the idea of compacting 19 trenches was abandoned and the easternmost 5 of the 19 trenches were selected for compaction, grouting, and covering.

3.2 COMPATIBILITY OF ACRYLAMIDE GROUT WITH BURIAL TRENCH CONTENTS

Because of the large inventory of radioactivity in some burial trenches in SWSA 6, additional assurance of hydrologic isolation of trench contents from groundwater intrusion and/or percolation, beyond that provided by an infiltration barrier, may be warranted for particular trenches. In situ grouting is a technique for providing assurance of hydrologic isolation of burial trench contents, regardless of assurance of infiltration barrier integrity or performance. Two in

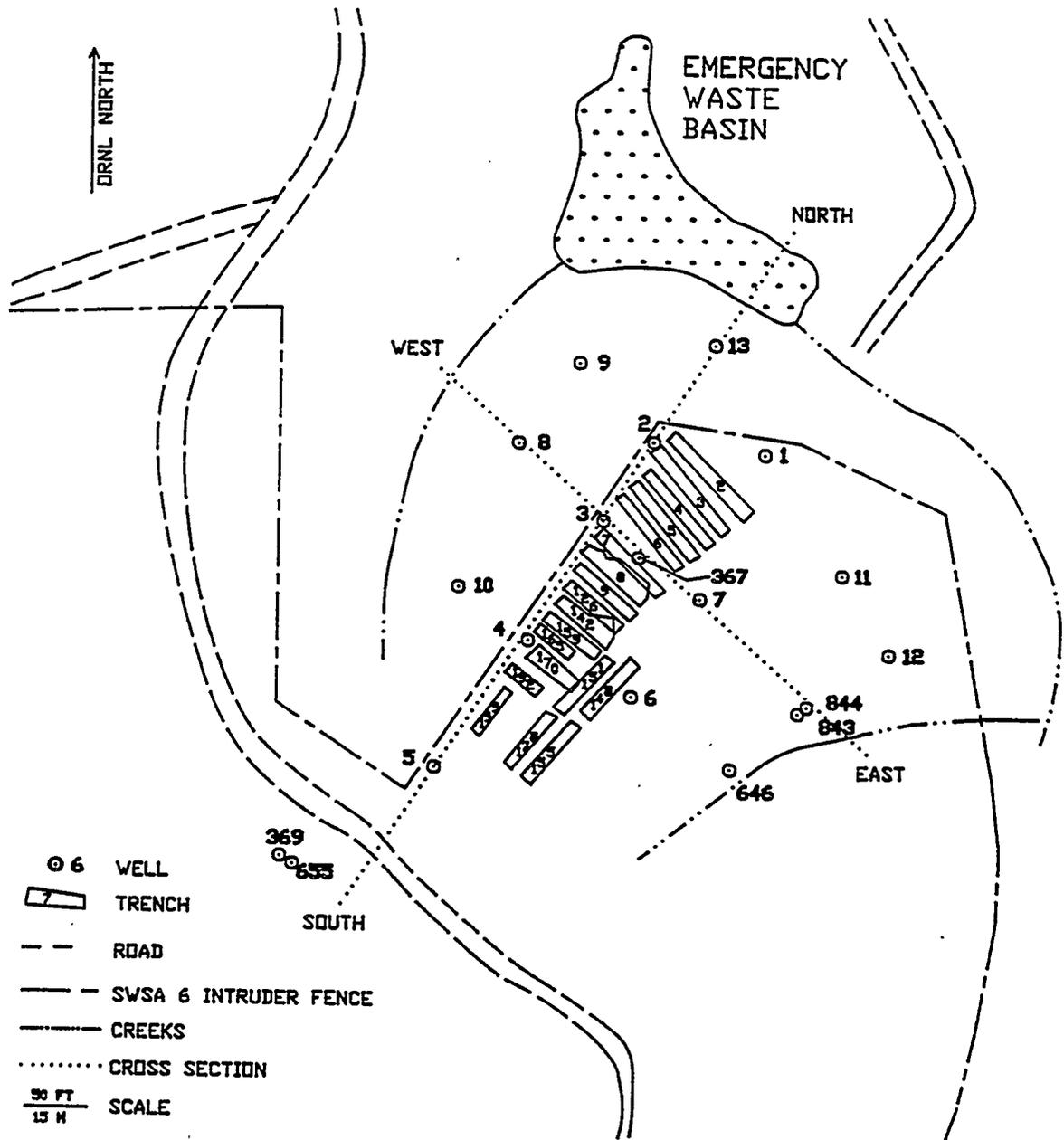


Fig. 24. Plan view of the TARA site in ORNL SWSA 6.

situ grouting field demonstrations have been completed at ORNL (Spalding et al. 1985 and 1987). Both of these demonstrations involved placing acrylamide grout into uncompacted burial trenches. Both were successful in changing the burial trenches from a condition of extreme permeability and facile water intrusion to a condition of unmeasurable permeability. However, based on these demonstrations, it became apparent that the costs of in situ grouting with polyacrylamide were quite high (i.e., about \$50,000 per typical burial trench) due to the large volume of voids per trench and the cost of the acrylamide grout materials (about \$2/gal). Dynamic compaction, on the other hand, is a relatively inexpensive method for eliminating many of the large voids within a trench, thus reducing void volume to be grouted. An additional benefit of dynamic compaction would be its ability to collapse voids within waste packages which may not otherwise be accessible during grout injection into an uncompacted trench. One of the major goals of the proposed in situ grouting demonstration is to establish that dynamically compacted trenches can be grouted with acrylamide and achieve the same degree of hydrologic isolation that has been experienced with unperturbed burial trenches. If this compatibility of in situ grouting with dynamic compaction can be demonstrated, then not only will in situ grouting costs be considerably lower (probably by about 80% due to the trench void reduction by dynamic compaction) but an effective and safe method for stabilizing the burial trench ground surface to support an infiltration barrier cap will be established to a degree that acrylamide grouting alone cannot achieve.

In seeking advice from environmental regulatory agencies and other scientists and engineers, however, a number of questions were raised concerning the hazards and risks of using acrylamide in the proposed demonstration. Acrylamide is a toxic substance and, therefore, poses certain risks not only for those who prepare and inject the grout but also for potential groundwater contamination in the area where the grout is being injected. Once the acrylamide polymerizes (sets), it becomes nontoxic and nonhazardous as are the majority of polymers in the contemporary materials of commerce. Most of the concerns raised center on the ability to control the polymerization process during injection into burial trenches and provide assurance that the grout will set to an impermeable state and not result in any contamination of groundwater at the demonstration site. The setting of polyacrylamide has been shown to be inhibited by strong acids or bases and by concentrated (10%) solutions of reducing agents (Larson and May 1983). What assurances exist that similar substances are not present in the proposed grouting demonstration trenches, or other trenches in SWSA 6, which may potentially inhibit the grout set? What assurance exists that a grout formulation of a given design set time will not reach the water table below the trenches before it sets?

3.2.1 Testing Rationale

One approach to test compatibility of acrylamide grout with the burial trench contents would be to perform chemical analyses of the inorganic and organic constituents in each of the burial trenches and, then, to perform grout compatibility tests with each detected species at

the measured or greater concentrations. This approach is enormously costly and technically difficult. Characterization of trench contents is quite difficult due to the heterogeneous nature of the buried waste. As an alternative, samples of trench leachate, where available, and trench bottom soil could be obtained and tested for compatibility with the acrylamide grout. In addition, specific chemical species that have been identified in previous trench leachate sampling in SWSA 6 (Solomon et al. 1988) could be tested for compatibility with acrylamide grout set at elevated concentrations limited only by their water solubilities. Organic species that have been identified, above regulatory guidelines include chloroform, benzene, tetrachloroethylene, toluene, 1,1-dichloroethylene, and 1,2-dichloroethane. In addition, recent groundwater monitoring has identified acetone in one well (unpublished data). The samples of trench leachate and bottom soil would be characterized in a screening method for gross chemical and radiochemical properties, and, where sample quantities permit, a detailed examination and analysis for specific hazardous constituents could be performed. Trench leachate samples would include randomly selected trenches from throughout SWSA 6 in addition to the specific target trenches for the field grouting demonstrations. Such general compatibility survey information would support the general applicability of polyacrylamide grouting throughout SWSA 6 rather than solely support the applicability to the specific demonstration trenches. Not only could grout set time be measured in the presence of trench leachate, soil, and specific chemical species, but quantitative measurements of the degree of polymerization could be determined by assaying the various grouts for residual concentrations of the acrylamide monomer. In addition, the hydraulic conductivity of trench soil specimens could be determined before and after grouting to verify the degree of permeability expected during actual trench grouting. If it can be demonstrated that (1) grout set time is not affected adversely by the trench leachates, trench soils, or the specific chemicals; (2) the degree of polymerization achieved is equivalent to what can be achieved with the neat grout; and (3) grouted trench soil can be converted to an impermeable state by the grout; then reasonable confidence can be attained that grout set can be controlled in the field demonstration and in other areas of SWSA 6.

3.2.2 Trench Sampling and Characterization

Prior to dynamic compaction, samples of trench bottom soil were collected on July 1, 1988, from the five burial trenches (SWSA 6 Trenches 2 through 6) by inserting a hand auger into residual penetration testing boreholes and/or bottomless well casings in each trench. The penetration test holes were made in February and March 1987 during stability tests on the burial trenches prior to dynamic compaction. After dynamic compaction was completed in late July and August 1988, additional soil samples were collected on March 28, 1989 from the five compacted trenches and from two adjoining uncompacted trenches (8 and 9). Access to these trench bottoms was gained through the postcompaction penetration test holes which were completed between January 16 and 26, 1989. Composites of all samples from an individual trench were made in the field in seal-lock polyethylene bags.

Typically, about 100-200 g of field-moist soil could be obtained from each trench. Samples were screened as collected for gross beta/gamma radioactivity with a field survey Geiger-Mueller meter; several read about 10 mR/h at contact with the sample bag. To reduce the soil moisture content to a level at which the samples could be sieved and homogenized, the samples were dried in the opened bags at 80°C for 2 h. Care was taken that the samples did not completely dry. Samples were then sieved to <2 mm by hand and the coarse fragments (mostly gravels) were discarded as radioactive waste. Moisture contents of the sieved and homogenized soil were determined by drying subsamples at 105°C for 2 h. These samples of oven-dried soil were used for simultaneous determination of ^{137}Cs , ^{60}Co , and ^{90}Sr via gamma-ray spectroscopy and Bremsstrahlung detection by using a NaI detector with a multiple channel analyzer.

Samples of precompaction trench leachate were collected between October 27 and November 20, 1987, during trench drainage following water filling for void determination. Since January 1985, when the first access wells were placed during initial penetration tests on these five trenches, and prior to compaction in 1988, a seasonally ephemeral level of standing water has been observed in only one of these trenches following a sustained rainy period in February 1987. Thus, "natural" trench leachate samples were not available for this grout compatibility study. However, the induced leachate samples were collected at the time near the emptying of the trench and, therefore, likely represent types of leachate with maximal (about 1 week) exposure to trench waste contents. Due to the relatively wet winter in 1988-89, measurable levels of perched leachate were observed in wells within Trenches 2, 3, 5, 7, 8, and 9. These leachates were sampled on March 29, 1989, using suction applied to a precleaned glass jug fitted with a length of Teflon tubing adequate to reach the liquid level. Because only limited sample volumes which could be recovered, these leachate samples were characterized only for their gross chemical and radiochemical properties by measuring pH, electrical conductivity, dissolved solids, hardness, alkalinity, gross alpha, and gross beta activities (APHA 1980). In addition, a water sample from the emergency waste holding pond (used as the source of water for the trench filling tests) was included in this characterization as a point of reference for water quality before contacting the buried waste trenches. Also included for comparison are the mean values for these same water characteristics for samples of groundwater collected 3 weeks after the completion of the last trench filling test and for samples collected after completion of dynamic compaction activities from the 15 groundwater monitoring wells within and around the test area.

As part of a continuing trench leachate monitoring and characterization project supporting both the remedial investigation and the interim corrective measures projects in SWSA 6, sampling of 23 additional trench leachates from SWSA 6 was completed between April 5 and 10, 1989. Sufficient volumes from 14 of these 23 samples were available for a detailed chemical and radiochemical characterization, and the analytical results, when they become available, should augment the limited water quality information on SWSA 6 trench leachates (Solomon et al. 1988). Leachate samples were taken from PVC access

wells (installed between October 10 and 20, 1988) as described above for the trench wells at TARA. Sufficient sample volumes were available from all 23 wells to perform both the grout compatibility tests and the suite of gross chemical and radiochemical characteristics described above.

3.2.3 Grout Compatibility Test Procedures

To measure grout set time in the presence of trench leachate, 10.0 mL of a 20% (weight/volume) solution of Q-1 (Cues, Inc., Orlando, Florida) grout in water was placed in a 50-mL polypropylene beaker. The Q-1 grout is a mixture of 95% acrylamide and 5% methylenebisacrylamide. The precompaction tests of soil and leachate used a batch of grout purchased in 1986, while the postcompaction compatibility tests used samples from a grout batch procured in 1988. The later material will be used in the proposed field demonstrations, and these compatibility tests also function as quality assurance tests on the actual grout to be injected. The 20% grout solution also contained 1% (volume/volume) triethanolamine, the grout free-radical transfer agent, and 0.02% (weight/volume) potassium ferricyanide, the grout set retarder. After adding the grout to the beaker, 5 mL of either trench leachate or water was added. Then, 5 mL of a 4% (weight/volume) solution of ammonium persulfate, the grout catalyst, was added and swirled gently to mix the beaker contents. Time of grout set was measured from the time of catalyst addition until grout would no longer flow within the beaker. The setting of acrylamide grout is a relatively abrupt change in viscosity occurring within a few seconds. Thus, unlike set time measurement with cements or silicate grouts, no elaborate test method or apparatus is required. Insufficient amounts of the precompaction leachate of Trench 4 were available for study.

To measure grout set in the presence of soil, the procedure was identical to that described above except that 5.00 g of fresh (undried) sieved soil was used instead of the 5 mL of leachate or water. The beaker contents were swirled at frequent intervals to keep the soil in suspension to maintain an intimate contact between soil and grout. This method maximizes the potential for observing possible interference of grout set by the soil or its contaminants. Otherwise, the soil would settle to the bottom of the beaker, and the overlying grout could set without interference from the burial trench soil or any possible adsorbed constituents. The observed set times of the grout were recorded to the nearest minute.

For the determination of the effect of specific organic chemicals on acrylamide grout set, polymerization was carried out as described above: 10 mL of 20% Q-1 grout, 5 mL of 4% ammonium persulfate, 5 mL of water, and 1 mL of the organic liquid (except that 1 g of naphthalene solids was used). The organic compounds tested were acetone, chloroform, toluene, trichloroethane, xylene, methylene chloride, trichloroethylene, benzene, naphthalene, and tetrachloroethylene. In the case of tetrachloroethylene, only 0.5 mL was employed in the tests because only a limited quantity was available. The amounts of all these added organic compounds, except acetone, were in great excess of their water solubilities, resulting in two immiscible fluids in the test beakers. All organic species tests were run in duplicate and were

agitated intermittently before grout set to ensure that maximum concentrations were attained in the aqueous phase. Extraction of the set grout specimens was performed as described below.

After storing the 20-mL grouted soil and leachate specimens in a sealed container overnight (16 h), the samples were extracted with water to determine concentrations of residual monomer and, hence, calculate the degree of polymerization achieved in the presence and absence of burial trench soil and leachate. The grouted specimen was placed in a 250-mL stainless steel container with 100 mL of distilled water and macerated for 15 s with a commercial rotating knife blender at 21,000 rpm. The resulting viscous homogenate was filtered through a borosilicate glass microfiber filter (1 μm particle size retention) overlain by a 1-cm-thick sand layer as a prefilter to prevent clogging by the filtrate. The sand was acid-washed and ignited quartz. The resulting clear filtrate was stored in capped polyethylene containers at 4°C until analysis. Samples of the unpolymerized grout were also prepared by substituting 5 mL of water for the ammonium persulfate catalyst solution or 5 mL of a 4% ammonium sulfate solution.

Analysis for acrylamide, methylenebisacrylamide, and acrylic acid was carried out by direct injection on a high-pressure liquid chromatograph (HPLC). A 4.6-mm-diam by 25-cm-long stainless steel column was employed containing octadecylsilane reversed-phase spherical silica absorbent (10 μm particle size) with 20% methanol:water (v:v) as the eluant. An injection volume of either 0.25 or 0.50 mL was employed, using a fixed-volume sample loop attached to the injection valve of a Perkin-Elmer Model 15B or a Glenco System I Liquid Chromatograph equipped with a 254-nm wavelength ultraviolet detector and a flow rate of 1 mL/min. Separation between the three components was good and a detection limit of about 1 mg/L of acrylamide was achieved. Standards for acrylamide in the range of 20 to 100 mg/L were prepared and dilutions of the grout extracts were made to be within this range. Elution peak height was used as a measure of acrylamide concentration and was found to be linearly correlated ($r > \text{or} = 0.9982$) with acrylamide concentration within the range of standards. Generally, an additional peak appeared in each liquid chromatogram for each solvent, but because none of these peaks overlapped with the acrylamide peak, no interference with the acrylamide analyses was observed. Percentage of polymerization was calculated from the difference between the amount of acrylamide in the unpolymerized specimens and that measured in the grouted specimens.

The effect of grouting on the permeability of precompaction burial trench soil was determined by measuring the hydraulic conductivity of small columns of burial trench soil before and after permeating them with grout. Five grams of moist soil, overlying 3 g of quartz sand, was placed into a 1.0-cm-diam by 10-cm-long glass column equipped with polypropylene fittings to regulate flow. A 20-mL aliquot of grout solution, prepared identically to that described above, was carefully poured onto the top of the column. A constant head was maintained manually by keeping the level of grout fixed above the column. From the measured volume of excess grout eluting from the soil column in a measured time interval, a hydraulic conductivity for the soil sample was calculated for the geometric configuration of liquid level and length of

soil column (Klute and Dirksen 1986). The bulk density of the soil in the columns was calculated from the known fresh weights of soil added, their measured moisture contents, and the volume occupied by the soil in the column. Several columns required that their flow be interrupted to prevent grout from running out of the soil, while others attained grout set before grout solution had percolated the soil. After grout set, any set grout on top of the column was removed with a spatula. The glass surface near the column bottom was scored and broken off to allow the removal of any grouted sand from the column. Otherwise, grouted sand at the column bottom may have reduced the apparent hydraulic conductivity of the sample, the effect of which could not be determined independently of any grouting of the soil. The postgrouting permeability measurements were, therefore, performed on columns containing only soil. A head of water was placed on top of each column, and a collecting tube, with only a pinhole opening to the atmosphere to prevent evaporation of any eluant, was placed on each column. The collection tubes were observed periodically over the following 48 h for the presence of eluant, but none was observed. A minimum postgrouting hydraulic conductivity was calculated for each column based on the minimum detectable amount of eluant (one drop or 0.1 mL) in 48 h and the column geometry as in the pregrouting hydraulic conductivity calculation.

3.2.4 Results

The chemical and radiochemical characteristics of the samples of TARA burial trench leachate are presented in Table 7 along with the characteristics of the pond water used to fill these trenches and the average characteristics of groundwater beneath and surrounding them. Several conclusions about the precompaction characteristics of waste buried in the trenches can be deduced from the information about these leachates. First, the trenches do not contain significant leachable forms of contaminants such as acids, bases, or soluble ionic species. The placement of pond water into the trenches did not significantly alter the pH, alkalinity, hardness, dissolved solids, or the electrical conductivity of the water. Nor were there measurable amounts of nonvolatile organic species which would have perturbed the dissolved solids level in the water samples. Most of the modest increases in the chemical properties of the pond water as it evolved into trench leachate can be explained as an interaction between the limestone-containing soil at the site. Because of this interaction, groundwaters tend to be rather hard with readily measurable levels of dissolved constituents. The average dissolved solids in the fifteen samples of groundwater were greater than the dissolved solids in any of the precompaction trench leachates. Thus, the chemical environment that the grout will encounter when placed into the burial trenches at the site will not be much different than that anticipated for areas unperturbed by waste burial. Thus, the reported interferences in acrylamide grout setting by strong acids, bases, and reducing agents (Larson and May 1983) would not be expected in these trenches.

Evidence that the pond water, which filled the trenches, did have significant interaction with the buried waste contents can be seen in the dramatic increase in dissolved radioactivity, particularly the gross

Table 7. Gross chemical and radiochemical characteristics of TARA burial trench leachate and groundwater samples

Sample	Date collected	pH	Electrical conductivity (dS/m)	Hardness (mg CaCO ₃ /L)	Alkalinity (mg CaCO ₃ /L)	Dissolved solids (mg/L)	Gross Alpha (dpm/mL)	Gross Beta (dpm/mL)
Precompaction								
T-2	10-30-87	6.2	170	86	54	60	7	12
T-3	10-27-87	7.2	760	242	166	480	<1	103
T-4	11-03-87	6.4	1100	168	446	200	3	20800
T-5	10-27-87	6.7	510	184	68	440	187	43
T-6	10-30-87	6.1	740	298	158	420	nd	257
Pond	11-03-87	6.2	240	136	78	120	nd	nd
Ground-water ^a	11-20-87	6.8	590	481	381	544	nd	0.3
S.D. ^a		0.2	217	185	124	324	-	0.4
Postcompaction								
T-2	03-29-89	7.6	247	186	169	200	<1	6
T-3	03-29-89	7.9	477	336	294	320	<1	27
T-5	03-29-89	8.1	617	332	276	520	<1	31
Uncompacted								
T-8	03-29-89	8.1	6490	5220	6511	6360	19	856
T-9	03-29-89	6.1	88	16	21	20	<1	nd
Pond	04-27-89	7.8	247	92	44	80	nd	nd
Ground-water	04-20-89	7.6	527	420	338	477	0.07	0.9
S.D. ^a		0.1	223	179	73	267	0.13	0.9

^aGroundwater characteristics are the average and standard deviation (S>D>) for 15 samples collected from the permanent monitoring wells in and surrounding the test area.

nd = not detected.

beta activity. The significant level of gross alpha activity in the sample from Trench 5 is caused by the presence of ^{233}U , which has been identified following alpha spectroscopic assay of this sample. It should be emphasized that this area was used for disposal of higher activity (>200-mR/h) low-level radioactive solid waste. Thus, it is less likely to contain the directly handled laboratory waste which is the probable source of hazardous organic materials (e.g., toluene and/or benzene contained in scintillation fluids) probably contained in other areas of SWSA 6 used for disposal of lower activity low-level solid waste.

Sufficient quantities of leachate from only three of the five compacted trenches were available for characterization. For samples from trenches 2, 3, and 5, no detectable influence of dynamic compaction was observed in either the gross chemical or radiochemical characteristics. Trench 8, which was one of the two uncompacted trenches from which leachate was sampled in anticipation of its potential grouting, did exhibit water qualities that were obviously influenced by its buried waste; dissolved solids, electrical conductivity, hardness, and alkalinity were elevated by an order of magnitude compared with other trench leachates or groundwater at the site.

The moisture content and radionuclide content of the six precompaction burial trench soil samples are listed in Table 8. The presence of such significant activities of radionuclides in these precompaction trench bottom soils indicates that they have experienced significant interaction with the contents of the trenches during the 16 years of burial. Thus, trace levels of inorganic and organic contaminants may also be present in these samples and they represent a good material for a fair testing of compatibility with acrylamide grouts. It should be noted that the sample of cap material from Trench 3 was taken from a depth of 2 to 3 ft below the ground surface and does not represent surface contamination at the site. It represents a soil that would potentially interact with grout at the last stages of trench filling. Samples of trench bottom soil taken after compaction also exhibited significant waste interaction as evidenced by their radionuclide contents (Table 8).

The chemical and radiochemical characteristics of the 23 other samples of SWSA 6 trench leachate are presented in Table 9. Only three samples (T-112, T-395, and T-453) exhibited obvious perturbations of chemical water quality, presumably due to interaction with their buried waste. All samples manifested detectable levels of gross beta activity, which demonstrates their interaction with the buried waste. These leachate quality samples were taken from a suite of 50 intratrench monitoring wells that had been constructed to monitor progress in groundwater suppression within the 10 acres of SWSA 6 that had been covered by a high-density polyethylene liner as part of the Interim Corrective Measures (ICM) Project (Lockwood Greene Engineers, Inc. 1988). Sampling was attempted from every well although only 23 had measurable levels of leachate during the wet spring season when sampling was initiated. Samples came from trenches in cap areas 1, 2, 5, 6, and 8, and the specific location of these trenches can be found in the ICM Plan drawings (Lockwood Greene Engineers, Inc. 1988).

Table 8. Radionuclide and moisture contents of burial trench soil samples

Sample	Moisture (%)	¹³⁷ Cs (dpm/g)	⁶⁰ Co (dpm/g)	⁹⁰ Sr (dpm/g)
Precompaction				
Trench 2	17.9	1060	nd	3,590
Trench 3	18.3	3,350	nd	72,800
Trench 4	16.4	128	nd	410
Trench 5	5.9	622	995	nd
Trench 6	13.7	9,580	nd	15,800
Trench 3				
Cap	15.2	17	nd	nd
Postcompaction				
Trench 2	--	16	<2	nd
Trench 3	--	14,300	3	1,960
Trench 4	--	74,500	nd	90,400
Trench 5	--	284	2	357
Trench 6	--	14,900	nd	4,730
Uncompacted				
Trench 8	--	115	nd	nd
Trench 9	--	135	nd	5,220

nd = not detected. Detection limits were 1.4 dpm/g, 2 dpm/g, and 6 dpm/g for ¹³⁷Cs, ⁶⁰Co, and ⁹⁰Sr, respectively.

Table 9. Gross chemical and radiochemical characteristics of burial trench leachate samples from SWSA 6

Trench number	Date collected	pH	Electrical conductivity (dS/m)	Hardness (mgCaCO ₃ /L)	Alkalinity (mgCaCO ₃ /L)	Dissolved solids (mg/L)	Gross Alpha (dpm/mL)	Gross Beta (dpm/mL)
T-44	04-10-89	7.8	371	172	272	80	nd	0.9
T-57	04-10-89	8.0	420	318	282	340	nd	0.8
T-60	04-10-89	7.7	459	330	301	340	nd	0.9
T-63	04-10-89	7.9	464	366	322	400	nd	0.8
T-69	04-05-89	8.1	478	356	340	360	1.5	14
T-82	04-05-89	7.5	160	96	79	120	2.2	84
T-85	04-05-89	7.7	260	126	171	160	nd	2.2
T-92-1	04-05-89	7.7	253	146	143	180	nd	0.7
T-92-2	04-05-89	7.7	200	134	119	120	0.1	1.1
T-101	04-06-89	8.0	356	290	231	320	0.4	1.5
T-105	04-06-89	6.9	774	400	291	880	0.2	1.5
T-110	04-05-89	7.8	416	244	274	320	nd	6.2
T-112	04-05-89	7.6	4620	168	2020	780	0.1	2.7
T-180	04-10-89	7.9	369	274	277	300	0.1	0.2
T-225	04-06-89	7.4	613	306	219	740	nd	1.4
T-237	04-06-89	8.0	554	372	330	600	0.1	1.2
T-318	04-06-89	7.9	335	208	236	260	nd	0.8
T-329	04-06-89	7.7	323	184	186	300	nd	1.6
T-363	04-05-89	7.8	252	136	171	120	1.2	53
T-367	04-06-89	7.8	55	208	234	360	0.2	0.5
T-395	04-06-89	8.2	440	604	1070	960	0.2	1.4
T-414	04-06-89	7.5	226	154	83	200	0.2	0.3
T-453	04-06-89	8.6	8580	300	2844	1360	0.1	5.3

nd = not detected.

None of the TARA trench leachate samples posed any interference with the setting time of the acrylamide grout formulation (Table 10). None of the TARA soil samples caused any significant delay or inhibition of grout setting time either. Precompaction soil samples from Trenches 3 and 4 did, however, cause a detectable acceleration of grout setting time. The causes of this effect are not known, but even if the same effect were manifested in actual field operations, no increase in hazard risk would result. Postcompaction soil and leachate samples likewise exhibited no significant effect on the polyacrylamide grout set time. In addition to the compacted trenches, samples of leachate from Trenches 7 through 9 and soil samples from trenches 8 and 9 were also found to pose no interference with grout set times. These uncompacted trenches are also candidates for in situ grouting demonstration, and their compatibility with acrylamide polymerization needed to be established. Only limited amounts of leachate from Trench 7 were available, and priority was given to testing grout compatibility over chemical and radiochemical characteristics which, therefore, were not obtained.

As discussed previously, grout set alone is not a quantitative indication of the degree of polymerization achieved by the grout. The HPLC analyses of the extracts of the grouted specimens for residual monomer, acrylamide, and cross-linking agent (methylenebisacrylamide) does provide a quantitative method to calculate the degree of polymerization and, thus, to observe any interference with the polymerization reaction by either soil or leachate. Table 11 summarizes the measured residual acrylamide concentrations within the grout specimens produced for the setting time determinations above. Also included are samples of the unpolymerized grout prepared with either water or ammonium sulfate to replace the ammonium persulfate catalyst, which function to provide the concentration of monomer that would be expected without any polymerization.

None of the TARA samples of burial trench soil or leachate samples, either precompaction or postcompaction, showed any effect on the degree of polymerization of the acrylamide as all samples exhibited $\geq 99\%$ polymerization. The uncatalyzed grout samples were formulated to contain 10% acrylamide, and it is reassuring that 100,800 mg/L were found by HPLC analysis. This verified that the bulk grout solution (procured in 1986) had not deteriorated in over 2 years of storage in the laboratory. All specimens from the 1986 grout batch contained a residuum of about 500 mg/L of acrylamide within their polymerized volume. Polymerization of the 1988 batch of grout was noticeably more complete, resulting in a typical residual acrylamide concentration of about 200 mg/L. The extremely low permeability of the set grout provides the mechanism to retain this residual level within the grouted mass preventing it from leaching and possibly contaminating groundwater. This would be true not only for this application for radioactive waste stabilization but also for the more common non-waste-engineering applications such as dam seepage sealing or ground stabilization for improving bearing strength. In addition, low levels of acrylamide are readily biodegradable in soil (Lande et al. 1979). Thus, both the hydrologic isolation afforded by the polymer and the facile biodegradation of small amounts of acrylamide that might leach from the

Table 10. Setting times for acrylamide grout in the presence of TARA burial trench leachates and soils

Sample	Type	Setting time (min)
Precompaction		
Water	Water	9
Trench 2	Leachate	9
Trench 3	Leachate	9
Trench 5	Leachate	9
Trench 6	Leachate	9
Trench 2	Soil	11
Trench 3	Soil	7
Trench 4	Soil	5
Trench 5	Soil	10
Trench 6	Soil	10
Trench 3	Soil-Cap	11
Postcompaction		
Water	Water	11
Trench 2	Leachate	12
Trench 3	Leachate	12
Trench 5	Leachate	13
Trench 7	Leachate	14
Trench 8	Leachate	11
Trench 9	Leachate	12
Trench 2	Soil	10
Trench 3	Soil	12
Trench 4	Soil	11
Trench 5	Soil	10
Trench 6	Soil	12
Trench 8	Soil	12
Trench 9	Soil	13

Table 11. Residual concentrations of acrylamide and methylenebisacrylamide in specimens of grout prepared in the presence and absence of TARA burial trench soil and leachate

Sample	Acrylamide (mg/L)	Methylenebis- acrylamide (mg/L)	Degree of Polymerization (%) ^a
Precompaction			
Uncatalyzed grout (four samples)	100,800	5360	None
Neat grout A (no soil)	450	0.7	99.55
Neat grout B (no soil)	516	0.6	99.56
Neat grout C (no soil)	462	0.9	99.49
Trench 2 leachate	480	0.9	99.52
Trench 3 leachate	462	0.7	99.54
Trench 5 leachate	486	0.7	99.51
Trench 6 leachate	510	0.9	99.49
Trench 2 soil	498	3.0	99.50
Trench 3 soil	342	2.1	99.66
Trench 4 soil	348	<10	99.65
Trench 5 soil	570	3.4	99.43
Trench 6 soil	516	2.8	99.49
Trench 3 cap soil	456	2.4	99.54
Postcompaction			
Uncatalyzed grout (two samples)	101,200	5340	None
Neat grout (six samples)	221	4.7	99.78
Trench 2 leachate	219	4.3	99.78
Trench 3 leachate	200	4.3	99.80
Trench 5 leachate	203	4.3	99.80
Trench 7 leachate	135	5.3	99.87
Trench 8 leachate	95	5.3	99.91
Trench 9 leachate	222	4.3	99.78
Trench 2 soil	80	5.2	99.92
Trench 3 soil	76	4.1	99.92
Trench 4 soil	40	2.1	99.96
Trench 5 soil	83	9.2	99.92
Trench 6 soil	112	7.2	99.89
Trench 8 soil	137	7.2	99.86
Trench 9 soil	162	10.2	99.84

^aDegree of polymerization = (starting concentration of acrylamide-- sample residual concentration of acrylamide)/starting concentration of acrylamide, where the starting concentration of acrylamide = 100,800 or 101,200 mg/L.

surface of the grouted mass provide adequate assurance that acrylamide will not reach groundwater at the proposed demonstration site.

None of the 23 samples of leachate from randomly selected burial trenches in SWSA 6 exhibited any detectable effect on the degree of acrylamide polymerization (Table 12). One sample (T-453) did accelerate grout set but, as discussed previously, this effect would not be considered a problem.

The effects of selected organic chemicals on acrylamide grout polymerization are presented in Table 13. Only benzene and toluene showed a detectable effect on the degree of polymerization: 98.8 and 99.3%, respectively, versus 99.9% for neat grout. In addition, none of the chemicals had a significant effect on the grout set time. This observation contrasts with the findings of Larson and May (1983), who found that benzene and toluene increased acrylamide grout set times from 0.5 min to 7 and 85 min, respectively, with an additional note that only a partial set was observed in the presence of a saturated solution of toluene. Specific chemical compositions of the acrylamide grout formulation, used by Larson and May (1983) are not described, other than that the manufacturer's recommendations were followed. However, the present information is much more quantitative in that both set time and residual monomer concentrations were determined. The present compatibility tests are also more rigorous in that the grout sets were carried out in the presence of excess organic solvent rather than employing water-saturated solutions of each species. Because no concentration of these organic species would be possible in aqueous SWSA 6 leachates beyond their water solubilities, the present tests provide confidence that interferences with the acrylamide grout polymerization will not occur due to known waste constituents in SWSA 6 leachate (Solomon et al. 1988). The list of compatible waste contaminants will probably grow when analyses of chemical characteristics of the 14 leachate samples are completed. However, the identity and concentrations of contaminants in these additional leachate specimens would not be necessary to assess the applicability of acrylamide grouting to SWSA 6 because their compatibility with the acrylamide grout has already been established (Table 13).

In addition to the assurance of predictable set times and a high degree of polymerization in the presence of burial trench soil and leachate, the acrylamide also resulted in unmeasurable hydraulic conductivities in columns of precompaction burial trench soil after percolation and grout set (Table 14). The postgrouting hydraulic conductivities, listed in Table 14 may be quite lower than the calculated detection limit based on the minimum detectable volume of water at the small gravity head maintained on these columns. Thus, the burial trench soil is not expected to pose any deleterious effect on the permeability of the grout. Acrylamide grouts have measured hydraulic conductivities of about 10^{-10} cm/s (Avanti International 1985). This extremely low permeability is adequate to retain the low residual concentrations of monomeric acrylamide within the polymeric grout by a diffusion-controlled process.

Table 12. Residual concentrations of acrylamide and methylenebisacrylamide in specimens of grout prepared in the presence and absence of SWSA 6 burial trench leachate

Trench number	Set time (min)	Acrylamide (mg/L)	Methylenebisacrylamide (mg/L)	Degree of polymerization (%) ^a
T-44	14	188	4.3	99.81
T-57	13	169	4.3	99.83
T-60	13	210	4.3	99.79
T-63	13	194	4.3	99.81
T-69	13	188	4.3	99.81
T-82	12	218	4.3	99.78
T-85	11	210	4.3	99.79
T-92-1	12	207	3.4	99.80
T-92-2	13	219	4.3	99.78
T-101	12	210	3.4	99.80
T-105	14	197	3.4	99.81
T-110	14	191	4.3	99.81
T-112	14	197	5.3	99.81
T-180	14	191	3.4	99.81
T-225	10	222	4.3	99.78
T-237	13	213	5.3	99.79
T-381	13	197	4.3	99.81
T-329	13	194	3.4	99.81
T-363	13	200	3.4	99.80
T-367	14	247	5.3	99.76
T-395	14	138	4.3	99.86
T-414	13	197	4.3	99.81
T-453	6	166	5.3	99.84
None (water)	11	221	4.7	99.78

^aDegree of polymerization = (starting concentration of acrylamide-- sample residual concentration of acrylamide)/starting concentration of acrylamide, where the starting concentration of acrylamide = 101,200 mg/L.

Table 13. Residual concentrations of acrylamide and methylenebisacrylamide in specimens of grout prepared in the presence and absence selected organic chemicals

Chemical species	Set time (min)	Acrylamide (mg/L)	Methylenebisacrylamide (mg/L)	Degree of polymerization (%) ^a
Acetone, rep 1	14	191	3.7	99.81
Acetone, rep 2	14	204	3.7	99.80
Chloroform, rep 1	11	162	1.9	99.84
Chloroform, rep 2	11	188	2.8	99.81
Toluene, rep 1	13	748	9.8	99.26
Toluene, rep 2	13	748	9.8	99.26
Trichloroethane:				
rep 1	12	165	1.9	99.84
rep 2	12	178	3.7	99.82
Xylene, rep 1	11	398	4.6	99.61
Xylene, rep 2	11	450	4.6	99.56
Methylenechloride:				
rep 1	12	182	1.9	99.82
rep 2	11	211	1.9	99.79
Trichloroethylene:				
rep 1	12	395	2.8	99.61
rep 2	12	440	2.8	99.56
Benzene, rep 1	14	1200	9.8	98.81
Benzene, rep 2	14	1298	9.8	98.72
Naphthalene, rep 1	12	224	3.7	99.78
Naphthalene, rep 2	12	275	5.6	99.73
Tetrachloroethylene:				
rep 1	12	201	2.8	99.80
rep 2	13	182	1.9	99.82
Water, rep 1	12	143	1.9	99.86
Water, rep 2	12	143	1.9	99.86

^aDegree of polymerization = (starting concentration of acrylamide - sample residual concentration of acrylamide) / starting concentration of acrylamide, where the starting concentration of acrylamide = 101,200 mg/L.

Table 14. Hydraulic conductivities of TARA burial trench soil columns before and after grouting with polyacrylamide

Soil column	Bulk density (g/cm ³)	Hydraulic conductivity (cm/s).	
		Before	After
Trench 2	0.83	1.2×10^{-2}	$<2.4 \times 10^{-7}$
Trench 3	0.80	4.8×10^{-2}	$<2.5 \times 10^{-7}$
Trench 4	0.78	4.0×10^{-2}	$<2.6 \times 10^{-7}$
Trench 6	0.95	3.2×10^{-3}	$<2.2 \times 10^{-7}$
Trench 3 cap	0.80	2.5×10^{-2}	$<2.5 \times 10^{-7}$

3.2.5 Field Grouting Testing

In 1982 a demonstration of grouting of low-level radioactive solid waste was completed at ORNL (Spalding et al. 1985). Seven grout formulations were tested in the laboratory for their ability to penetrate and to reduce the hydraulic conductivities of soils used as backfills for shallow land burial trenches. Three soluble grout formulations (sodium silicate, polyacrylamide, and resorcinol-formaldehyde) were able to penetrate soil and sand columns and to reduce their hydraulic conductivities from initial values of about 10^{-4} m/s to less than 10^{-8} m/s. Three particulate-containing grouts (lime-fly ash, fly ash-cement-bentonite, and bentonite slurry) could not penetrate the soil or sand columns; such formulations, therefore, would be difficult to distribute within a trench for assured hydrologic isolation. Two field demonstrations with both sodium silicate and polyacrylamide showed that these grouts could be distributed throughout 1:10-scale burial trenches and that waste-backfill hydraulic conductivity could be reduced several orders of magnitude. Field grouting with the polyacrylamide reduced the mean hydraulic conductivity of nine intra-trench monitoring wells from 10^{-4} to $<10^{-8}$ m/s while sodium silicate reduced the mean hydraulic conductivity from 10^{-4} to 10^{-6} m/s.

The levels of grout in the nine monitoring wells in each of the two grouted trenches have been measured periodically up to the present. These observations were carried out to establish the persistence of the grout material in the fluctuating groundwater environment of burial trenches in SWSA 6. The results of this monitoring reported in 1985 (Spalding et al. 1985) covered the first 154 days after grouting and indicated that both the sodium silicate and polyacrylamide grouts were shrinking (Fig. 25). However, repeated observations 3 and 7 years after grouting indicated that the levels of polyacrylamide grout have remained

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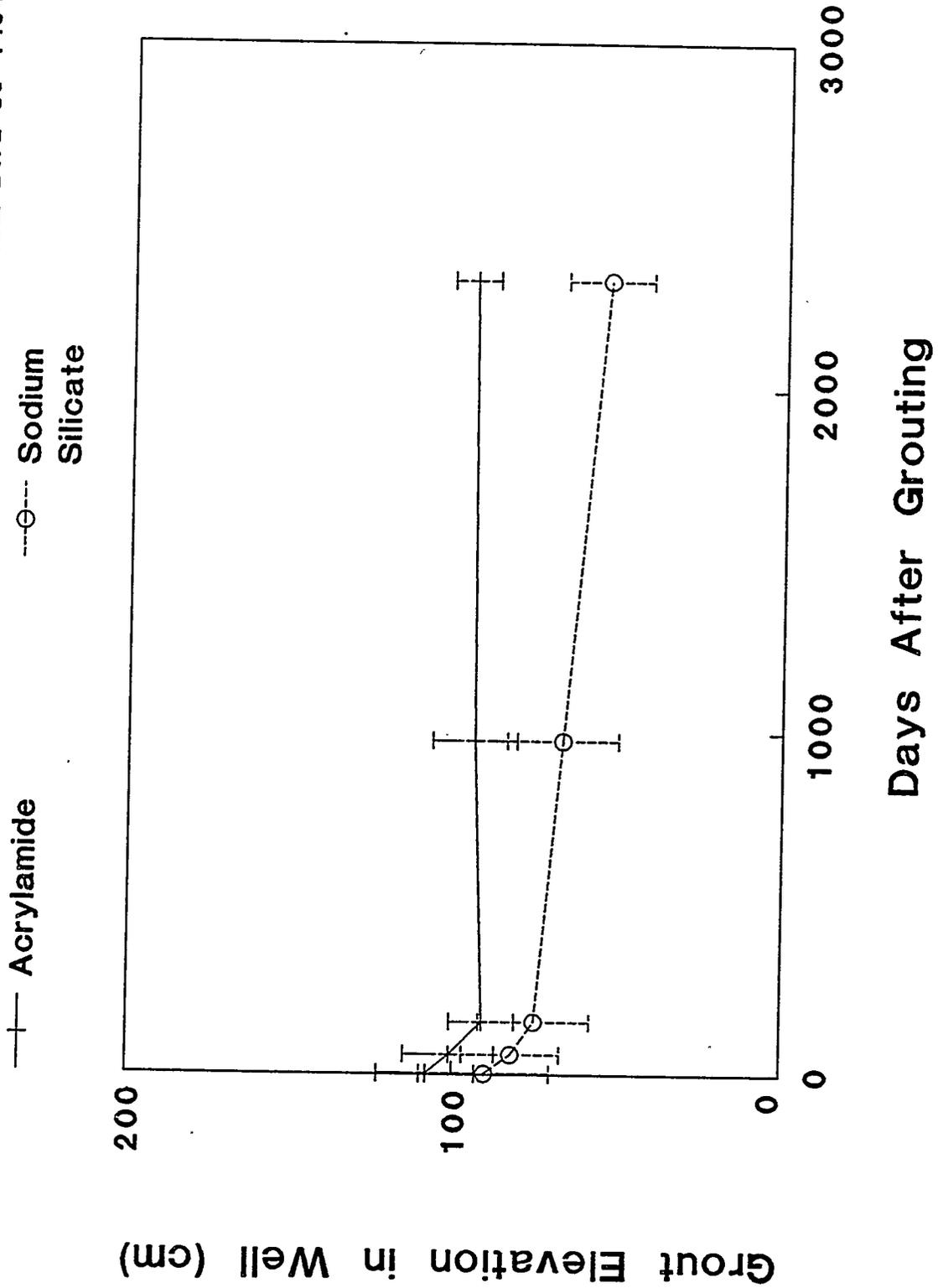


Fig. 25. Solidified grout remaining in monitoring wells in the period after grouting with sodium silicate and polyacrylamide.

stable after that initial shrinkage, while the levels of sodium silicate grout have continued to decline. The stability of the polyacrylamide grout, after initial shrinkage, is supported by these observations.

The average hydraulic conductivities reported previously for these monitoring wells were for the 5-month period after grouting. The measurements were repeated again after 3 years (Table 15). The sodium silicate grout had continued to increase in hydraulic conductivity to the point that the conductivity of the trench's waste backfill became greater than the host soil formation. However, the polyacrylamide remained near field detection limits during this period indicating that the grout had remained impermeable and unchanged during this interval.

In an effort to observe the distribution of polyacrylamide within the grouted trench (which contained solid waste without radioactivity), two downslope edges of the 3- by 3-m trench were excavated to a depth of 1.2 m, resulting in an L-shaped observation trench (Fig. 26). Although the photograph does not offer enough contrast between grouted regions and waste backfill, visual and tactile observations revealed that the polyacrylamide grout had completely permeated voids within the burial trench. Practically all of the exposed grout was solidified although a small amount of highly viscous slime was found near the trench bottom. Clods of soil backfill were observed to be permeated with the polyacrylamide, and fractures within the soil formation at the edge of the trench were filled with grout with visually detectable grout up to 5 cm into the formation. Grout colors included colorless, black, orange, and yellow depending on the type of waste with which the grout was in contact (i.e., black for rubber tires and yellow and orange for iron drums). Ten samples of the grout, approximately 500 mL each, were taken representing the different colors and textures. They were subjected to a drying and ashing procedure (Spalding et al. 1987) to estimate their organic matter content (Table 16).

All of the grout specimens contained a significant amount of organic matter (polymer). The formulation placed into Trench 351 was a nominal 10% grout solids. Whether the grout had dried out somewhat during its 7-year residence in the burial trench or whether the drying out occurred during the 1-day interval when the trench was open prior to sampling cannot be determined. Notably, the two samples of grout slime were below the 3-4% solids level needed for solidification of polyacrylamide (Avanti International 1985). Regardless, the grout appeared to polymerize but did not solidify at this low monomer concentration. Presumably, this slime resulted from dilution of grout with residual standing water in the trench during grouting. As noted previously, the amount of slime in the burial trench was quite small. Its presence does support the safe containment of grout even if diluted below solidifiable concentrations. Analyses for concentrations of residual acrylamide monomer in all these specimens are being performed. However, the absence of residual monomer will not provide an explanation of how it disappeared. The residual monomer could be degraded microbiologically (Lande et al. 1979) or be leached from the grout by percolating groundwater. However, given the low hydraulic conductivity of the polyacrylamide, the leaching mechanism would seem less plausible than a microbiological degradation within the grout.

Table 15. Hydraulic conductivity of monitoring wells within grouted trenches in the interval after grouting

Treatment or area	Before grouting (cm/s)	Five months after grouting (cm/s)	Three years after grouting (cm/s)
Polyacrylamide grouted trench	$10^{-1.8}$	$10^{-6.1}$	$10^{-6.1}$
Sodium silicate grouted trench	$10^{-1.6}$	$10^{-4.1}$	$10^{-2.3}$
Soil formation	$10^{-3.4}$		
Field detection limit	$10^{-6.2}$		

3.2.6 Summary

The proposed in situ grouting of burial trenches with polyacrylamide had raised several concerns about the potential for environmental contamination by acrylamide. To establish the compatibility of the acrylamide grout set reaction with potential interference by buried waste constituents, several tests were performed to verify the ability to predict and control the polymerization or set of the acrylamide grout. Samples of soil from the bottom of 7 trenches and 31 samples of burial trench leachate were collected and added to grout solutions before set. No soil or leachate significantly retarded the grout set time. By chemical analysis of extracts of grout/soil specimens, it was determined that the percent polymerization in the presence of soil or leachate was identical to that achieved with unamended grout (i.e., 99.5%). Five trenches that had been dynamically compacted in 1988 were sampled again for leachate and bottom soil in March 1989; neither the precompaction nor postcompaction samples exhibited any effect on grout set times or degree of polymerization. Ten organic chemicals that had previously been identified in leachates from SWSA 6 trenches were tested for compatibility with acrylamide grout set; only benzene and toluene reduced the percentage polymerization marginally to 98.8 and 99.3%, respectively. In addition, specimens of burial trench soil were converted from high to unmeasurably low hydraulic conductivities by percolation with reacting grout. Thus, it appears that these burial trench soils and leachates pose no significant interference with acrylamide grout set. Both the degree of polymerization and the impermeable nature of the resulting polyacrylamide grout lead to the assurance that environmental contamination of groundwater by grout injection into these burial trenches is extremely improbable.

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Fig. 26. The distribution of polyacrylamide grout in waste and backfill of Trench 351 after excavation to trench edges in May 1989 after grouting in 1982.

Table 16. Properties of polyacrylamide grout samples taken from Trench 351, SWSA 6, 7 years after completed in situ grouting

Sample	Consistency	Color	Ash-free organic matter (%)	Ash-free moisture (%)	Ash (%)
A	Slime	Black	3.6	96.3	0.3
B	Solid	Brown	12.3	87.7	2.9
C	Solid	Clear	11.7	88.3	0.6
D	Solid	Clear	17.2	82.8	72.2
E	Solid	Clear	10.7	89.3	45.4
F	Solid	Orange	11.8	88.2	0.3
G	Slime	Green	2.7	97.3	1.9
H	Solid	Brown	15.2	84.8	61.2
I	Solid	Clear	12.1	87.9	0.0
J	Solid	Clear	10.8	89.2	0.0

3.3 STABILIZATION OF LOW-LEVEL WASTE BURIAL TRENCHES BY DYNAMIC COMPACTION

Many of the problems with burial trench performance stem from the inherent instability of the buried waste and backfill. Dynamic compaction was selected for demonstration as a remedial technique to address this primary fault. If dynamic compaction can establish a structurally sound ground surface, then various caps and moisture barriers can be constructed over burial trenches with assurance that they will not subside.

3.3.1 Precompaction Penetration Tests

The rationale for compacting the TARA trenches as a part of the closure demonstration was to reduce trench void volume and stabilize the ground surface prior to constructing a permanent cover. Water-facilitated trench cover subsidence, which takes place with time, would be undesirable and could cause differential settling, resulting in shear forces that could compromise the performance of an impermeable membrane or other cover components. To quantify the benefits resulting from trench dynamic compaction, soil penetration tests were carried out at 5 positions over each of the 19 waste trenches, as well as at 34 locations over the surrounding soil and saprolite weathered from the Conasauga formation. A nonstandard test was developed for use over the trenches in order to avoid augering [which is required in the standard soil penetration test (ASTM TEST D 1586-84. Penetration test and split barrel sampling of soils)] which would bring contaminated waste to the surface.

The nonstandard test consisted of using a 63.5-kg (140-lb) drill-rig-mounted drop hammer to drive a 4.4-cm diam AW-size drill rod into the soil. The end of the drill rod was fitted with a 5.1-cm-diam 60°-cone point, and the rod was marked off in 30-cm lengths. At each nonstandard penetration site, the rod was hammered into the soil and the number of hammer blows for each 30-cm depth was recorded. The deepest penetration hole in each trench was used for installation of a monitoring well. Either a 5.1-cm-diam stainless steel wire-wound well screen section (1.5 m long) with a tapered well point coupled with threaded flush-joint casing (Johnson Company) or a 3.8-cm-diam slotted PVC screen of variable length coupled with flush-joint pipe (Timco Manufacturing Company) was hammered into the hole. This nonstandard penetration is similar to the Dutch cone penetration test (DCPT) except that the test borehole was not augered out every 46 cm (18 in.) to relieve drill stem friction. Blow count values (penetration resistance) obtained by the DCPT have often been found equivalent to blow count values obtained with the standard penetration test (SPT) (Solymar and Reed 1986). The DCPT, like the herein reported nonstandard penetration test, is not employed frequently in geotechnical work because it does not produce a soil sample for subsequent geotechnical or chemical tests.

A summary of the nonstandard penetration data (Fig. 27) shows that there is a significantly lower resistance to penetration within the trenches as compared to the undisturbed Conasauga Shale. In some cases, large voids were encountered and one or two blows with the hammer would send the drill rod to the bottom of the trench. A marked increase in the number of blows per foot of penetration occurred as a function of depth for setups outside the trenches, whereas within the trenches, this increase was not as apparent. The large voids within trenches apparently resulted in little frictional resistance to drill rod penetration in comparison to the relatively dense soil formation.

In addition to the nonstandard penetration tests discussed above, standard engineering methods (ASTM TEST D 1586-84) of determining soil penetration resistance and collecting undisturbed soil cores were used at various locations. Fifteen locations were above waste trenches where shallow cores into the trench covers were collected (to 91.4 cm only), whereas 34 locations were outside the trenches where cores were collected to a maximum depth of 4.6 m (15 ft).

The standard penetration tests and core retrieval served three purposes: (1) to collect standard penetration resistance data that can be correlated to the nonstandard data, (2) to retrieve soil cores from between and around the five TARA trenches for physical analyses (bulk density and moisture) and chemical and radiochemical analyses prior to trench compaction, and (3) to obtain an estimate of the depth to auger refusal at the TARA site. Figure 28 shows the relationship between penetration resistance and bulk density. There was an expected positive correlation ($r = 0.66$) between penetration resistance and soil bulk density; the denser the material, the more resistance to penetration would be expected. The negative correlation ($r = -0.49$) between penetration resistance and moisture content probably results from the inverse correlation between soil moisture content and bulk density: the less dense a soil or saprolite layer, the more water it would be expected to hold against a given moisture tension or drying stress.

Additional soil and saprolite variables such as texture, structure, and lithologic properties likely contribute to the additional variation in penetration resistance.

3.3.2 Measurement of Trench Void Volumes

An important quantitative indicator of the benefits of trench compaction is the reduction of trench voids that can be achieved. The method selected to measure these voids prior to trench compaction was the filling of the five trenches with a known volume of water, taking into consideration the losses due to seepage into the surrounding soil that takes place during filling (Spalding 1986). Water was pumped from the emergency waste holding basin to the north of the TARA site into one of the five trenches. Trench 2 was filled between October 26 and 28, 1987, and received 55 m³ (14,555 gal) of water. Trench 3 was filled between October 19 and 20, 1987, and received 53 m³ (14,071 gal) of water. Trench 4 was filled between July 27 and August 3, 1987, and received 100 m³ (26,422 gal) of water. Trench 5 was filled between October 22 and 23, 1987, and received 77 m³ (20,367 gal) of water. Trench 6 was filled between October 28 and November 2, 1987, and received 55 m³ (14,440 gal) of water.

Water was delivered to the trenches via a 5-cm (2-in.) rubber hose run to the bottom of one of the intratrench wells. The water delivery was allowed to continue overnight under continuous surveillance until water level monitoring in additional intratrench wells indicated that the water was within approximately 0.5 m of the ground surface. At this point, the flow rate was either adjusted downward to determine the minimum flow required to maintain the existing head or turned off. When the pump was turned off, the head in the observation wells was monitored periodically until the trench was completely drained. This made possible the determination of seepage rates required to correct for seepage during the water delivery interval and indicated when it would be possible to begin the next water pump-in test in the adjacent trench. Water levels, or more frequently the lack of standing water, in neighboring trenches were also monitored during the filling and draining intervals to identify any hydrologic connection between the burial trenches. Among the five trenches at the eastern end of the TARA site, no interconnection of burial trenches was observed during the filling tests. Water levels in the 15 groundwater monitoring wells were measured periodically during the trench-filling process to determine which, if any, of the wells would respond to trench filling.

3.3.3 Method of Dynamic Compaction

The method selected for compacting the contents of TARA trenches 2 through 6 was the repeated dropping of a 4-metric-ton, cylindrical, concrete weight at positions or settings along the trench cover. The method had been tested previously and found successful during the 1985 experimental compaction of trench 271 in SWSA 6 (Spalding 1986). A track-mounted crane with a maximum lifting capacity of 60 tons was used to straddle the trench and drop the weight from a height of approximately 7 m. As the weight was lifted and positioned over the

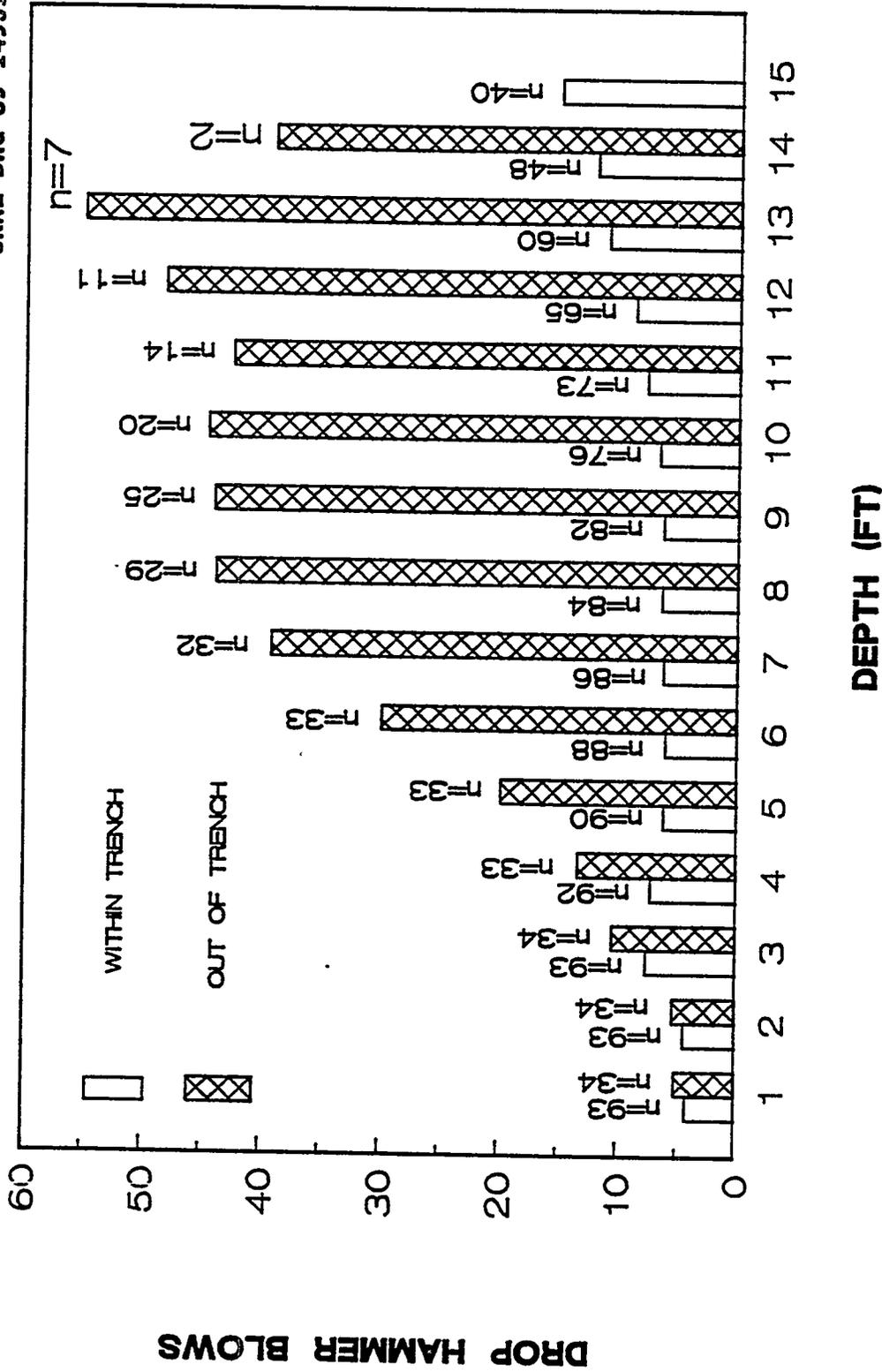


Fig. 27. Comparison of within-trench and out-of-trench nonstandard penetration tests (n is the number of observations on which the average is based).

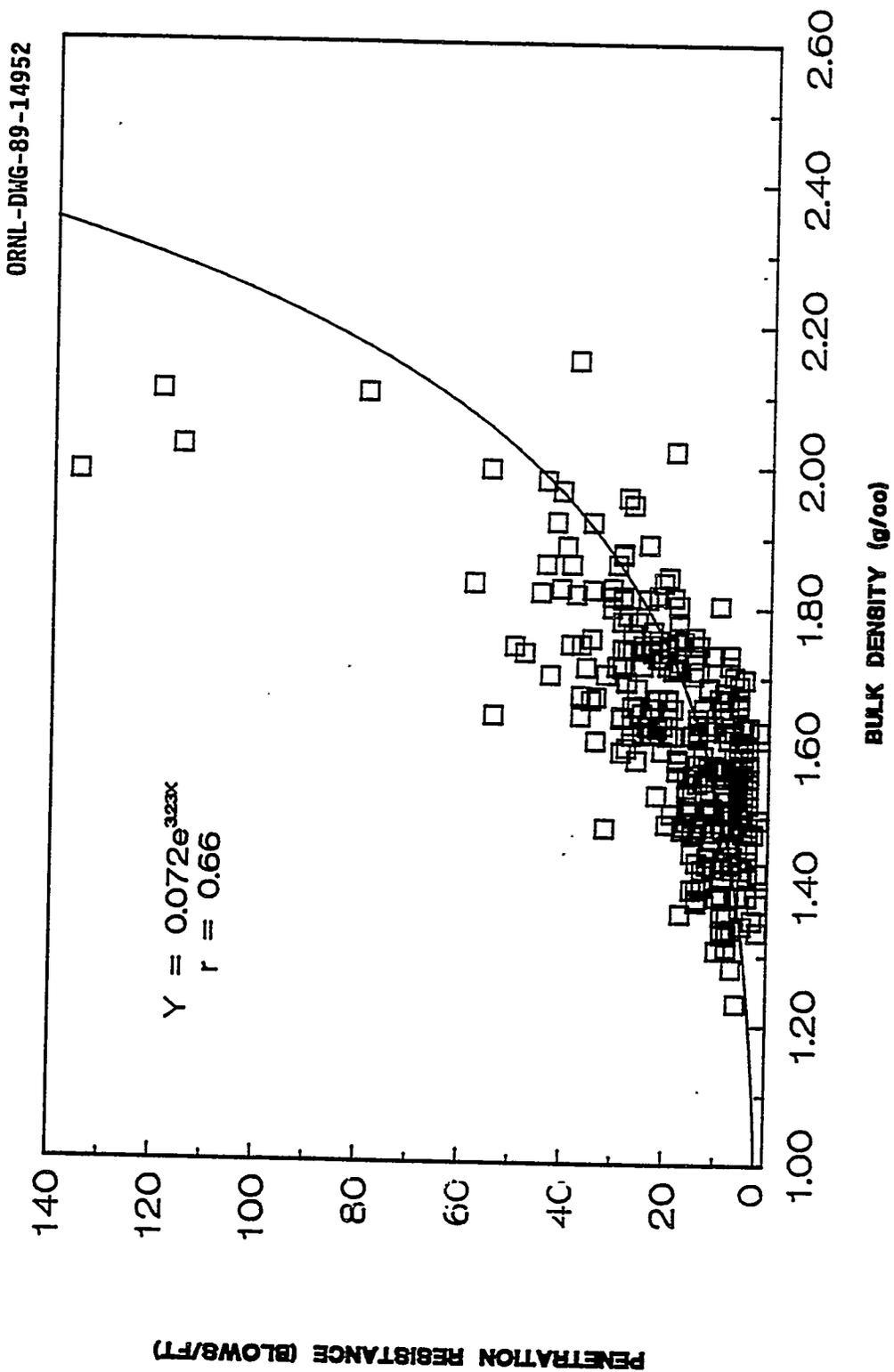


Fig. 28. Relationship between soil penetration resistance and soil bulk density at the TARA site.

trench cap, the break on the cable holding the weight was released and the weight was allowed to fall. Upon impact, the cable break was quickly engaged to prevent excess cable from unrolling from the drum due to the downward momentum of the cable. A steel-belted truck tire was used as a connection between the crane cable and weight to act as a shock absorber. On several occasions during the trench compaction operation, the tire was replaced because of excessive wear. Figure 29 shows the weight being dropped over the cover of one of the TARA trenches.

The method found most successful in compacting the trenches involved beginning at one end of the trench and laying out an approximate 3-m by 3-m square section. The four corners of this section were compacted first to a depth of 1.2 m (4 ft), followed by compaction of the center portion of the section to the same depth. After the entire trench section had been lowered to a depth of 1.2 m, the bottom of the resulting crater was leveled by numerous short drops of the weight. When leveling of the section was completed, the crane was repositioned and an adjoining 3-m by 3-m section of the trench was compacted. In total, 39 trench sections were laid out over the five TARA trenches, and 211 compaction positions were used.

3.3.4 Results of Dynamic Compaction

As a part of the compaction evaluation, the number of drops of the weight required for each 30 cm (1 ft) of depth at each compaction position was counted, along with the number of short drops to level out each section, and the radiation reading in milliroentgens per hour at each compaction position. Table 17 summarizes the average drop counts per foot of compaction for the five TARA trenches and a 3- by 3-m control trench that was laid out immediately to the southeast of Trench 3.

As expected, the average number of drops increased with each 30 cm of depth; 1.2 drops were required for the first 30 cm, 2 drops for the next 30 cm, 2.8 drops for the next 30 cm, and 3.8 drops for the last 30 cm. This increase was due to the compaction of soil and waste that was taking place immediately below the point of impact. Also summarized in Table 17 are the corresponding numbers of drops required for the 3- by 3-m control section. This section, which is similar in all respects to the trenches except that there was no underlying waste trench, required an average of 3.2 drops for the first 30 cm, 5.5 drops for the second 30 cm, 7.3 drops for the third 30 cm, and 6.7 drops for the fourth 30 cm. In addition to the greater number of drops required to achieve a desired depth, a considerable amount of soil upheaval was noted around the control trench, extending to a distance of approximately 2 m from the trench. This upheaved soil volume was estimated by using precompaction and postcompaction elevation surveys, which allowed an actual (corrected for upheaved soil) depth of compaction to be determined for the control section.

In summary, the dynamic compaction of the 5 TARA trenches required 2141 drops of the weight from a height of 7 m and approximately 2445 drops of the weight from a lower height (1-2 m) for leveling purposes.

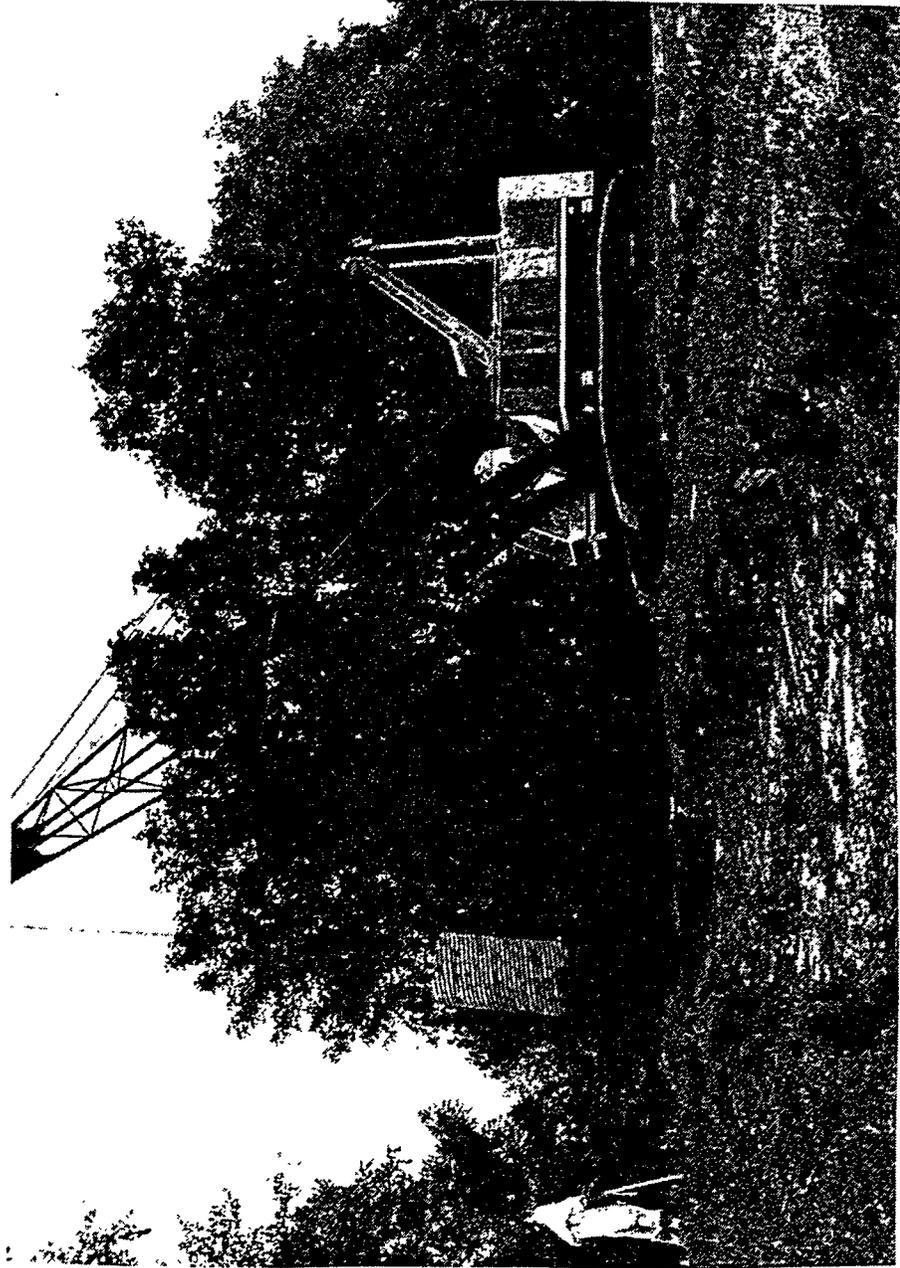


Fig. 29. Dynamic compaction of TARA burial trenches using a 3.6-Mg weight dropped from height of 7 m.

Table 17. Summary of the number of weight drops required for each foot of trench compaction

Depth (ft)		Trench 2	Trench 3	Trench 4	Trench 5	Trench 6	Total	Control trench
1	AVG ^a -	1.79	1.11	0.85	1.02	1.13	1.20	3.17
	STD ^b -	±1.27	±0.80	±0.58	±0.84	±0.93	±0.98	±2.11
2	AVG -	2.51	1.98	1.72	1.68	2.03	2.00	5.50
	STD -	±1.50	±1.12	±0.78	±1.00	±1.19	±1.20	±2.75
3	AVG -	3.53	3.36	2.23	2.29	2.48	2.82	7.33
	STD -	±2.18	±1.58	±0.95	±1.17	±1.40	±1.64	±3.40
4	AVG -	4.74	3.86	3.26	2.93	3.88	3.76	6.75
	STD -	±2.46	±2.04	±1.50	±1.47	±2.03	±2.05	±2.68

^aAVG - Average.

^bSTD - Standard Deviation.

Knowing that the total area of the 5 trenches is 387 m², these total drop numbers can be expressed as 5.5 drops/m² of trench for compaction and 6.3 drops/m² for leveling purposes.

The depth of compaction of each trench was determined by comparison of a trench surface elevation survey made before compaction with that of a postcompaction elevation survey. In contrast to the control section, an insignificant amount of soil upheaval was noticed adjacent to each of the five trenches; thus, depth corrections for upheaval did not have to be made for the waste trenches as they were for the control section. Table 18 summarizes the precompaction and postcompaction elevation surveys and indicates that an average depth of compaction of 0.46 m, 0.49 m, 0.60 m, 0.63 m, and 0.54 m was achieved for Trenches 2, 3, 4, 5, and 6, respectively. Knowing the length and width of each trench allows one to calculate the total volume of compaction that took place for each trench.

The depths and volumes of trench compaction reported in the previous paragraph are uncorrected [i.e., the values have not been corrected for the degree of compaction that could be achieved in the trench cover alone (without an underlying trench)]. To obtain this correction value, a 3- by 3-m control section was laid out adjacent to Trench 3 and compacted with the concrete weight in a manner identical to the waste trench compaction. Elevation survey points were established at 41 locations around the perimeter of the trench out to a distance of 1.5 m. By comparing the elevations at the survey points before and after compaction, the depth of the control section crater could be determined (0.75 m) and the total volume of soil upheaval that occurred could be estimated. Dividing the perimeter of the control section into

Table 18. Percentage reduction in trench water-accessible void space achieved by dynamic compaction

Trench number	Average crater width (m)	Average crater length (m)	Average crater depth (m)	Corrected crater depth (m)	Corrected crater volume (m ³)	Water void (m ³)	Void reduction (%)
2	3.43	28.65	0.46	0.30	29.48	30.42	96.9
3	3.22	27.74	0.49	0.33	29.48	25.72	115
4	2.78	24.08	0.60	0.44	29.45	61.04	48.2
5	2.99	23.77	0.63	0.47	33.40	58.79	56.8
6	2.61	23.47	0.54	0.38	23.28	35.18	66.2
Control	2.74	2.74	0.16	-	-	-	-

a set of concentric octagonal elements and summing element volumes resulted in a calculated upheaved soil volume of 4.44 m³. The corrected control crater volume (i.e., the actual volume reduction achieved at the control section had the soil not been able to be pushed up) was calculated as the difference between total volume (5.65 m³) and upheaved volume, yielding a corrected control crater depth of 0.16 m of the total 0.75 m. In other words, the approximate 3- by 3-m control section could actually be compacted to a depth of 0.16 m, yielding a total compacted volume of 1.21 m³.

The control section depth of 0.16 m was then subtracted from the average trench crater depths, resulting in corrected trench crater depths. These depths are summarized in Table 18 and range from a minimum of 0.30 m in Trench 2 to a maximum of 0.47 m in Trench 5. The corrected trench crater volume was then calculated from the product of trench width, length, and corrected depth. The corrected trench crater volumes were then compared with the volume of water-accessible void space (measured via the water pump-in tests described above) and expressed as a percentage of this value. For example, Table 18 shows that a compaction volume of 29.48 m³ was achieved over Trench 2, accounting for 96.9% of the measured water-accessible void space of 30.42 m³. These water-accessible void space reductions ranged from a low of 48.2% to a high of 115%, with an average value of 77%.

Several sources of error could account for the variations in trench void reduction that were observed. First, the water pump-in tests could underestimate the actual trench void volume if all observation wells along the trench did not maintain equal elevations during the filling process. In other words, only one side of the trench would have actually been filled with water while the other side of the trench would have been partially filled. This could lead to an overestimate in the trench void reduction such as was the case with Trench 3 (115% reduction). Another source of error could be void volume within waste packages themselves that may not have been measured in the water pump-in

tests but would be accounted for in the trench crater volume after the waste packages were crushed by dynamic compaction. This could again lead to an overestimate of the trench void reduction such as was the case for Trench 3. One last point to consider is that the total void volume of the TARA trenches was not measured by the water pump-in tests, only the water-accessible void volume, which is smaller. This is because the trenches contain a certain volume that cannot be unsaturated completely during the relatively short drainage period following water pump-in tests and also because of the formation of air pockets in and around the waste and soil backfill. This results in a somewhat lower reduction of total trench void space than of water-accessible void space depending on the actual ratio of total void space to water-accessible voids. Considering these sources of uncertainty, the best estimate of water-accessible void reduction achieved by compaction of the five TARA trenches would be the average of the five calculated values, or 77%. This estimate can be verified in the future by the independent measurement of residual void volume in selected trenches as determined by the volumes of grout accepted by compacted trenches.

3.3.5 Postcompaction Penetration Tests

Between January 16 and 26, 1989, the nonstandard penetration tests were repeated on the five dynamically compacted burial trenches. In addition, penetration tests were repeated on uncompacted Trenches 7, 8, and 9 as a check on the reproducibility of the precompaction penetration test results approximately one year after the initial tests. As in the previous tests, five locations were selected on each burial trench. Three control tests were also repeated in the area just south of Trenches 4 through 6 to check reproducibility of test results on the undisturbed soil formation. The test results on the three uncompacted trenches were identical to the initial test results on these same trenches 1 year previous. Test results on the compacted burial trenches, before and after compaction, are presented in Fig. 30. The penetration resistance of these trenches was significantly increased by dynamic compaction to a degree that was not significantly different from the undisturbed soil formation. Thus, no differential subsidence of the burial trench surface would be expected following dynamic compaction, and a stable surface was created for the support of an infiltration and biointrusion barrier.

The three control sites (undisturbed soil formation) exhibited significantly lower penetration resistance in the 1989 tests than in the 1988 tests (Fig. 27 vs Fig. 30). It is assumed that this effect was not caused by the dynamic compaction of the trenches that were located approximately 3 m from these test points. If the soil profile were wetter in January 1989 than in 1988, lower penetration might be expected in these areas because moisture content was found to be negatively correlated with penetration resistance. However, standard penetration tests, which produce core material for moisture determination, were not carried out in 1989. Thus, the increased moisture effect is only a plausible explanation.

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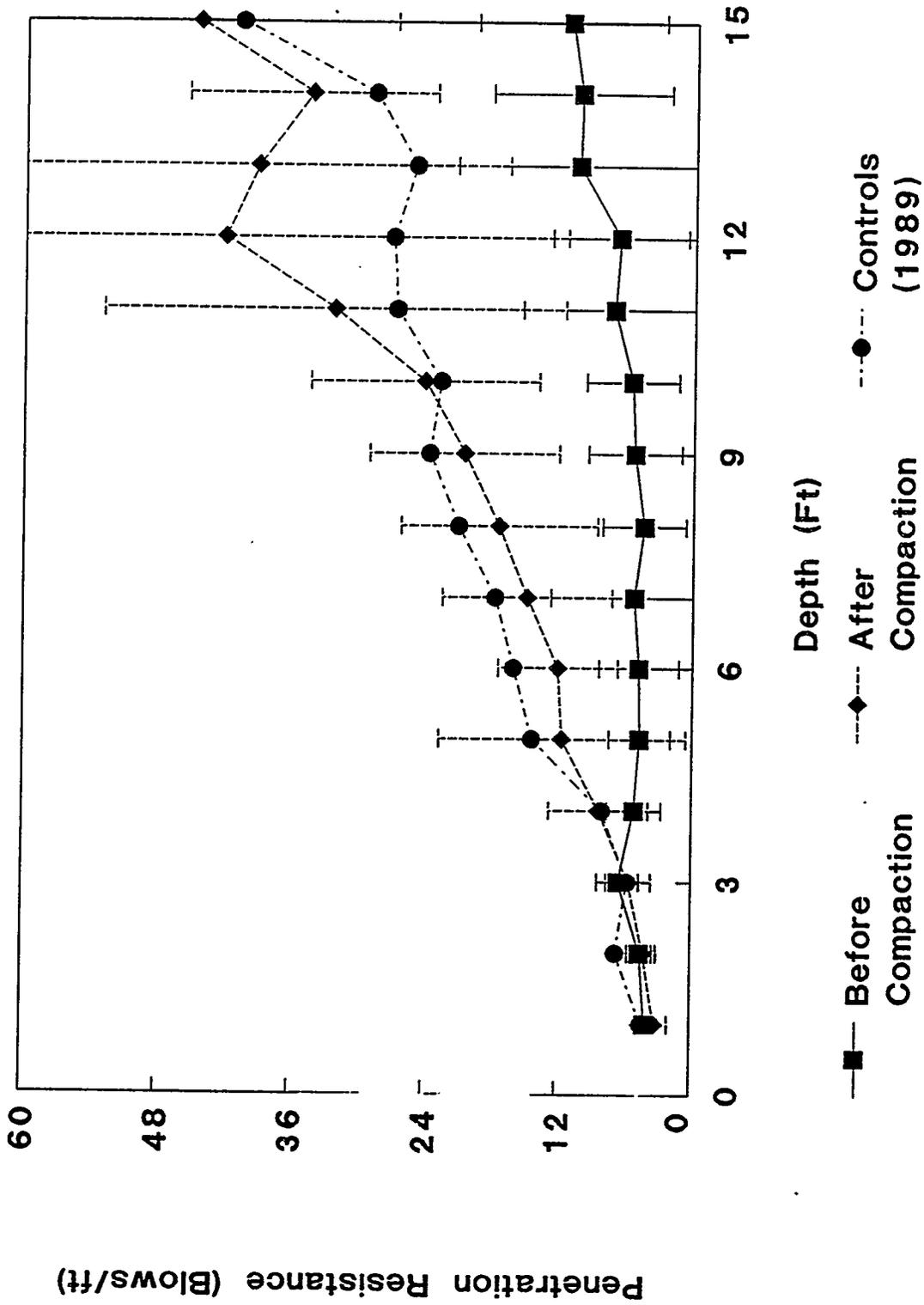


Fig. 30. Penetration resistance of five burial trenches at TARA before and after dynamic compaction.

3.3.6 Time and Personnel

Compaction of the five trenches began the morning of July 27, 1988, at the north end of Trench 2. Trench 6 was completed on August 4, 1988. Of the 7 working days during this time period, a total of 48 h was spent actually dropping the weight over the five trenches. An additional 1 h was spent compacting the control section. During the 7-day working period, the cable breaking mechanism of the crane had to be adjusted periodically, and additional time was spent for crane lubrication, fueling, and cable inspection. In total, the crane was operating 85% of the time (48 h), with the additional 15% downtime due to maintenance activities. It was estimated that approximately 504 employee hours were spent during the 7-day working period compacting the trenches. Site grading, seeding, and mulching, which followed compaction, required 2 days to complete. Additional time was spent in relocating a section of the SWSA 6 fence, which was required for future cap construction purposes.

3.3.7 Static Equivalent of Dynamic Compaction

Another technique used in civil and geotechnical engineering to achieve densification of soil is static loading. In this technique, a specified weight is applied to the surface of the ground and the ground allowed to subside for a length of time until the desired amount of consolidation is attained. The weight is usually applied as a surcharge of soil, rock, or rubble, but sometimes reinforced concrete is used. Because this static technique could, in theory, deliver an equivalent pressure to a burial trench, but with lower potential for airborne releases or penetration which exposes waste packages to the surface, the following calculations were performed to compute what the static equivalent of dynamic compaction, as performed in this demonstration, would be. Rather simple Newtonian mechanics are all that is required to calculate the static equivalent weight. The force exerted by given mass is given by

$$f = m \cdot a \quad [1]$$

where

- f = force (dynes or poundals),
- m = mass (grams or pounds),
- a = acceleration or deceleration.

The deceleration of the falling mass can be calculated from its velocity and the distance in which it comes to rest, i.e., the depth of penetration of the weight into the antecedent ground surface. Typically, the depth of penetration ranged from 30 cm to as little as 2 cm during the final drops on the trench at a given location. The deceleration, a, is calculated as

$$a = v/t \quad [2]$$

where

v = velocity of the falling weight (centimeters per second),
t = time (seconds) required for the weight to decelerate to v = 0.

The time is calculated from

$$t = 2 s/v \quad [3]$$

where

s = penetration distance (2 to 30 cm).

The velocity at impact (v) is calculated from equation for a free-falling body

$$v = (2 dg)^{1/2} \quad [4]$$

where

d = drop height, 700 cm (23 ft),
g = acceleration due to gravity, 980 cm/s² (32 ft/s²),
v = 1171 cm/s (38 ft/s or 26 mile/h).

The resulting forces applied by the 3.6-Mg (4-ton) mass traveling at 1171 cm/s is equivalent to 350 times its weight for a 2-cm penetration and 23 times its weight for a 30-cm penetration. These are equivalent to static loads of 1260 Mg (1400 tons) and 83 Mg (92 tons). Because the weight of dynamic compaction is exerted on a limited area (8,938 cm² for this demonstration), the equivalent weights can be converted to the corresponding pressures of 142,100 g/cm² (2,020 psi) and 9,338 g/cm² (133 psi), respectively. Thus, putting this in perspective, the maximum force exerted by the dynamic compaction weight is equivalent to a half-mile-high concrete cylinder placed over the ground surface. Clearly, static loading cannot practically achieve the equivalent of dynamic compaction. It is also clear that if a given static load is desired or can be specified, dynamic compaction can be used to deliver it at less cost. The weight could be dropped from much lower heights to achieve less densification such as might be specified in a typical surcharging technique.

3.3.8 Risks and Benefits of Dynamic Compaction

The major benefit of dynamic compaction of buried waste trenches is that it provides a stable surface from which an infiltration barrier can be supported without the threat of future subsidence to compromise barrier performance. We base this conclusion on the results of penetration tests that were performed on the five burial trenches before and after their dynamic compaction. On each of the five trenches, five positions were tested for penetration resistance, and Fig. 30 presents the average number of blows required to penetrate each foot of trench depth. In addition, the average penetration resistance of the

unperturbed soil formation around these test trenches is also presented. As can be seen from the Fig. 30, dynamic compaction increased the penetration resistance of the burial trench soil and contents to a degree that its behavior was equivalent to the host soil formation. The research effort required to obtain this information was quite significant, both in money and time, but allows us to conclude that differential settlement or subsidence of the land surface would not be expected after dynamic compaction. Thus, it would be expected that dynamically compacted trenches would provide long-term support for an infiltration barrier free from future collapse. Without stabilization of the burial trenches, one would expect land surface subsidence at some unpredictable future time in the range of 2 to 6 ft (i.e., the range of compaction depths observed). Ignoring this ultimate subsidence would be irresponsible on our part. Without trench stabilization, rather than planning for the closure of SWSA 6, we would really be planning another interim covering and delaying final closure to some unknown time in the future.

There are other potential, although undemonstrated, methods to stabilize burial trenches against subsidence. However, the critical issue regarding dynamic compaction centers on the risk of loss of any existing waste containment which definitely will result from dynamic compaction. The risk associated with loss of containment centers on four attributes of the waste in SWSA 6: How much hazardous waste, liquid or solid, was disposed of? What was its degree of containment at the time of disposal? What is its degree of containment now or at the time when trenches will be stabilized? What will be the integrity of containment in the future? These questions are difficult to answer quantitatively, but they would have to be answered at least qualitatively no matter what method is finally employed for the stabilization of SWSA 6 burial trenches.

First, consider the nature of the waste disposed of in SWSA 6 burial trenches. It is composed largely of solid waste. The waste did contain incidental quantities of liquids (e.g., scintillation vials) which, of course, were containerized only to a degree necessary to transport them to SWSA 6. But these contained liquids were only a minor fraction, on either a volume or weight basis, of the waste disposed of in SWSA 6. Unfortunately, the waste disposal records are not particularly helpful in quantifying the amount of contained liquids. These records do not have the required information on liquid content or containment or sufficient detail to make even a crude estimate of contained liquids. What is helpful, however, are pictures of the waste in trenches which lead to the qualitative conclusion that the waste did not contain significant amounts of contained liquids. Pictures of all waste trenches used in a 1-year interval between 1984 and 1985 (Davis et al. 1986) can be considered typical of waste disposed of in SWSA 6 since it was opened in 1972. What can be inferred from these pictures is that the waste in SWSA 6 is largely solid in nature and that its level of containment from hydrologic intrusion is not adequate for ORNL to merit any credit for environmental isolation. Whatever containment existed in a particular waste shipment was often breached when it was dumped into the trench; the ubiquitous plastic sheeting is almost always torn. The very heterogeneous nature of this waste and its loose incidental packing

within the trench is precisely the reason why we observe and expect to continue to observe significant subsidence over burial trenches.

Second, considering the level of waste containment likely existing in the burial trenches when they were filled, what is the degree of waste containment (the oldest trench was filled 17 years ago) and what are the prospects for this containment to continue into the future? Present waste containment is likely something worse than when the waste was shipped to SWSA 6 but something better than total loss of containment. However, it must be realized that whatever containment currently exists in SWSA 6 waste will ultimately disappear. Waste containers will corrode, break, and/or biodegrade. If this loss of containment occurs after capping, then releases will not be detected until perimeter or other monitoring wells intercept the contaminant migration. Would it not be better for whatever limited containment may still exist in SWSA 6 trenches to be lost during closure activities when effective and economic corrective action could be taken? Site closure with dynamic compaction would obviously need to include a plan to monitor and collect significant releases if necessary. ORNL is not planning or proposing to ignore this potential impact of site stabilization. Were SWSA 6 waste to consist mostly of 55-gallon drums of spent solvent, it would be irresponsible for ORNL to propose dynamic compaction as a trench stabilization method; ORNL would, in that case, be proposing solvent extraction and recovery.

In summary, the risk for liquid release resulting from dynamic compaction is small because SWSA 6 burial trenches contain only small amounts of contained liquids. The benefit of long-term infiltration barrier performance far outweighs this short-term risk. More importantly, the small but finite risk of liquid release during dynamic compaction can be controlled by monitoring and collecting groundwater if necessary. The shallow aquifer in SWSA 6 has quite a low yield. Measured storage coefficients range between 0.0005 and 0.01 with an effective porosity of 0.03. Saturated hydraulic conductivity averages 6.3×10^{-5} cm/s. Thus, groundwater collection would be feasible based on the small anticipated collection volumes.

Dynamic compaction, however, does need the additional development and demonstration which is planned for early next fiscal year in SWSA 6; this is the TARA Phase II demonstration as outlined in the SWSA 6 closure plan. Another two trenches at the TARA II site will be dynamically compacted to address several concerns with the effectiveness and range of applicability of dynamic compaction. For this demonstration two burial trenches that are chronically inundated with groundwater will be selected. It is anticipated that this condition will exist in a significant number (10-20%) of the total burial trenches in SWSA 6. Obviously, to apply dynamic compaction, these trenches will have to be dewatered prior to compaction. First, it must be learned and demonstrated how effectively burial trenches can be dewatered before and even during dynamic compaction because seepage into a dewatered trench will likely occur during the interval of compaction. Second, the effectiveness of dynamic compaction (again via penetration tests) needs to be demonstrated when carried out on a trench still quite wet after removal of its free water. Third, because compaction of a wet trench is the most likely scenario for groundwater contamination due to liquid

containment release, the ability to deal with the consequences of a liquid release in or near the water table needs to be demonstrated. Rather than rely on the fortuitous selection of demonstration burial trenches that contain an unknown quantity of hazardous liquid, it is proposed to simulate a liquid release from a trench during its dynamic compaction by injecting 55 gal of ethanol. Ethanol is proposed because it is not a hazardous substance and is rapidly, but not too rapidly, degraded by soil microorganisms. Its use would allow the quantitative demonstration of the ability of intratrench dewatering and groundwater monitoring wells to collect a known quantity of released liquid. The overall TARA II demonstration would provide the currently missing information to plan the stabilization of the chronically inundated burial trenches in SWSA 6 and should also provide the EPA with additional assurance regarding the environmental impact of dynamic compaction.

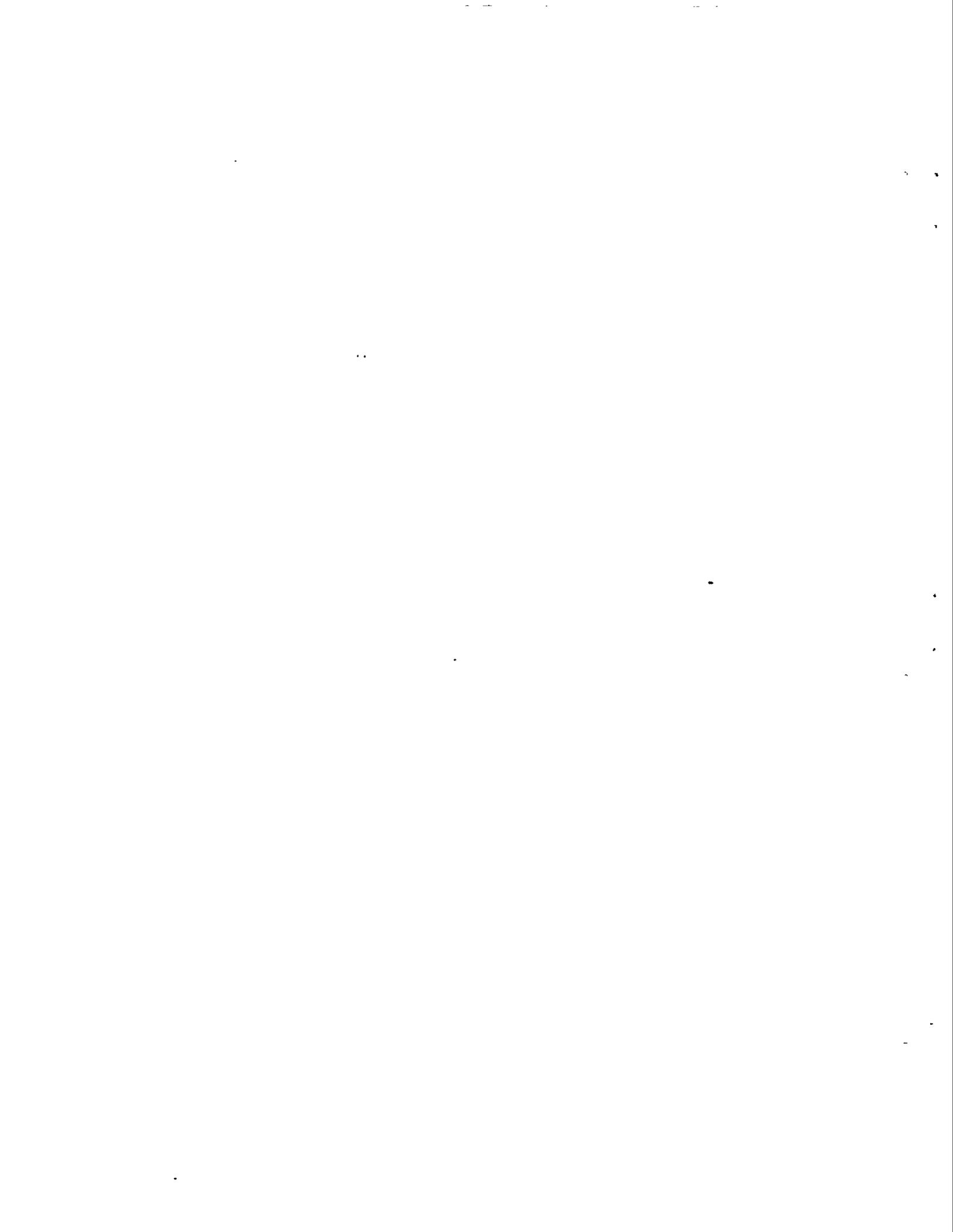
3.3.9 Summary

Shallow land burial in unlined trenches has been the disposal method used at ORNL for radioactive solid waste. Recent regulatory scrutiny has accelerated the planning for stabilization and closure of many of these waste disposal areas. However, protection of buried waste from infiltrating precipitation via impermeable covers is compromised by the tendency of burial trenches to subside as backfill collapses into voids within the trenches. As part of a low-level radioactive waste burial site stabilization and closure technology demonstration project, a group of five 14-year-old burial trenches in ORNL SWSA 6 was selected for testing trench compaction, grouting, and infiltration barrier design and performance. To obviate the chronic problem of trench subsidence and to provide a stable foundation for the infiltration barrier, the five trenches were dynamically compacted by the repeated dropping of a 4-ton weight with a 1.1-m² base onto each trench from a height of approximately 7 m.

The five trenches were compacted to a maximum depth of 1.2 m, requiring an average of 5.5 drops/m² of trench area, and the site was graded to facilitate surface runoff. Measurements of void reduction within the trenches averaged 77% and were calculated by a comparison of ground surface depression and measured water-accessible voids prior to compaction. Penetration tests were performed on trenches before and after compaction and on the surrounding undisturbed soil formation. The penetration resistance of the trenches was extremely low before compaction and was increased to a level equivalent to that of the undisturbed soil after compaction.

Thus, dynamic compaction was found to be very effective in stabilizing burial trenches to the extent that no differential land surface settlement should be expected to compromise the foundational support of an infiltration barrier. However, the benefit of ground surface stabilization must be weighed against the risks associated with the loss of any ambient containment of incidental amounts of buried liquids that may be present in the waste. Because ambient containment of small but unknown amounts of contained liquids is not a characteristic on which to base a sound strategy for protection from environmental

releases, which will occur ultimately for any buried liquid containers, such risks, inherent in dynamic compaction, are quite minimal.



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93. S. Riddle, U.S. Environmental Protection Agency, Region IV, 345 Courtland St., NE, Atlanta, GA 30365
94. R. J. Starmer, HLW Technical Development Branch, Office of Nuclear Material Safety and Safeguards, Nuclear Regulatory Commission, 5E4 (OWFN), Washington, DC 20555
95. L. A. Thomas, U.S. Environmental Protection Agency, Region IV, 345 Courtland St, NE, Atlanta, GA 30365
96. David H. Thompson, Argonne National Laboratory, Bldg. 206, 9700 S. Cass Avenue, Arlington, IL 60439
97. Douglas Underwood, U.S. Department of Energy, Oak Ridge Operations, P.O. Box 2001, Oak Ridge, TN 37831
98. T. J. Wheeler, U.S. Department of Energy, Oak Ridge Operations, P.O. Box 2001, Oak Ridge, TN 37831
99. Frank J. Wobber, Ecological Research Division, Office of Health and Environmental Research, Office of Energy Research, ER-75, U.S. Department of Energy, Washington, DC 20545
100. J. G. Yates, Office of Energy Research, U.S. Department of Energy, ER-42, Washington, DC 20585
101. Office of Assistant Manager for Energy Research and Development, Oak Ridge Operations, P.O. Box 2001, U.S. Department of Energy, Oak Ridge, TN 37831-8600
- 102-111. Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831