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BNL-64958

In-Situ Stabilization of TRU/Mixed Waste Project at the INEEL

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August 1997

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Prepared for
U.S. Department of Energy
Contract No. DE-AC02-76CH00016

ABSTRACT

Throughout the DOE complex, buried waste poses a threat to the environment by means of contaminant transport. Many of the sites contain buried waste that is untreated, prior to disposal, or insufficiently treated, by today's standards. One option to remedy these disposal problems is to stabilize the waste *in situ.* This project was in support of the Transuranic/Mixed Buried Waste - Arid Soils product line of the Landfill Focus Area, which is managed currently by the Idaho National Engineering Laboratory, through Lockheed Martin Idaho Technologies Company. Brookhaven National Laboratory (BNL) provided the analytical laboratory and technical support for the various stabilization activities that will be performed as part of the *In Situ* Stabilization of TRU/Mixed Waste project at the INEL. More specifically, BNL was involved in laboratory testing that included the evaluation of several grouting materials and their compatibility, interaction, and long-term durability/performance, following the encapsulation of various waste materials. The four grouting materials chosen by INEL were: TECT 1, a two component, high density cementious grout, WAXFIX, a two component, molten wax product, Carbray 100, a two component elastomeric epoxy, and phosphate cement, a two component ceramic. A simulated waste stream comprised of sodium nitrate, Canola oil, and INEL soil was used in this study. Seven performance and durability tests were conducted on grout/waste specimens: compressive strength, wet-dry cycling, thermal analysis, base immersion, solvent immersion, hydraulic conductivity, and accelerated leach testing.

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ACKNOWLEDGMENTS

The authors would like to thank Ernest Carter of Carter Technologies for providing the technical support during the preparation of test specimens. Thanks to Dileep Singh of Argonne National Laboratory for providing the phosphate cement test specimens. We also gratefully acknowledge Laura Ayres for her assistance in the preparation of this report.

This work was funded by the U.S. Department of Energy Office of Science and Technology, EM-50.

1.0 INTRODUCTION

Throughout the DOE complex, buried waste poses a threat to the environment by means of contaminant transport. Many of the sites contain buried waste that was untreated, prior to disposal, or insufficiently treated, by today's standards. Presently, there are two options available to remedy these disposal problems. The first requires exhuming, treatment (or re-treatment), and then the re-disposing of the waste into an engineered vault/disposal site. Dispersion is a primary concern since most of the waste is untreated and consists of friable materials (e.g., salt wastes). Exhumation will result in airborne contamination and additional exposure, safety concerns, and contamination control requirements. The second alternative is to stabilize the waste *in situ*. Treatment/stabilization in situ will not only reduce, but may even eliminate, the majority of worker exposure. Furthermore, there is a high probability that the dispersal of contaminated products will be substantially minimized if exhumation is required/desirable after treatment.

This project is in support of the Transuranic (TRU)/Mixed Buried Waste - Arid Soils product line of the Landfill Focus Area (LFA), which is managed currently by the Idaho National Engineering Laboratory (INEL), through Lockheed Idaho Technologies Company (LITCO). In particular, the primary purpose of this project is for Brookhaven National Laboratory (BNL) to provide analytical laboratory and technical support for the various stabilization activities that will be performed as part of the In Situ Stabilization of TRU/Mixed Waste project at the INEL (TTP no. ID7-6-LF-23).

The technical support involved determining which cementitious and/or polymer materials should be evaluated for long-term stabilization and interim retrieval purposes, as well as the type of analytical tests that will be needed to verify the long-term durability of immobilized products. The task also included project management activities and analytical laboratory testing support. BNL's primary involvement was laboratory testing. The testing included the evaluation of several grouting materials and their compatibility, interaction, and long-term durability/performance, following the encapsulation of various waste materials.

Important criteria in the selection of viable candidates for treatment/stabilization are grout materials properties and buried waste compatibility. Consequently, grouting systems were chosen based on their low viscosity and compatibility with; conventional jet grouting techniques, the soil, environmental, and geotechnical characteristics of the site, and the contaminants and chemistry of the buried waste. Four grouting materials were chosen by INEL for in situ treatment/stabilization of buried waste and interstitial soils. The grout materials included: TECT 1, a two component, high density cementitious grout, WAXFIX, a two component, molten wax product, Carbray 100, a two component elastomeric epoxy, and phosphate cement, a two component ceramic. The surrogate waste stream was comprised of sodium nitrate, Canola oil, and INEL soil. Initial scoping studies were performed to insure that the material and the formulations selected were suitable for in situ stabilization at INEL for long-term stabilization and interim retrieval purposes.

Equally important in selecting a grout material is the final disposition of the stabilized waste. For example, the grout and waste type will determine whether the stabilized waste can be left in place for long term monitoring or retrieved for additional treatment or more secure storage. The requirements of a grout for long interim storage will be different than those of a retrievable grout. Grouts used for in situ stabilization for retrieval of buried waste do not have as rigorous demands on the durability of the waste form as does long term *in situ* storage. The stabilized waste can be repacked or further treated after retrieval. However, the grouts must have the ability to set or cure in place without interference from the buried waste or geochemistry of the site. In addition, a retrievable grout should be easily broken and excavated by conventional construction equipment (e.g., a backhoe). Finally, the cured product should not change over the short term, thus requiring the waste form to exhibit reasonable short term durability in case retrieval operations are delayed.

If long term in situ storage is desirable, the grout must produce a waste form that can withstand the chemistry of the waste and site specific parameters. Furthermore, the grout material must be capable of containing waste contaminants within acceptable levels for long periods of time. BNL prepared laboratory scale test specimens consisting of a grout/surrogate waste stream and grout/INEL soil mixtures using the candidate grouts. These specimens were evaluated for durability and performance.

The following seven performance and durability tests were conducted:

- compressive strength
- wet-dry cycling
- thermal analysis (differential scanning calorimetry)
- base immersion
- solvent immersion
- hydraulic conductivity
- accelerated leach testing

All specimens were prepared using a standardized waste stream (70 wt% INEL soil, 20 wt% sodium nitrate, and 10 wt% Canola oil) and encapsulated in TECT 1, WAXFIX, and Carbray 100 Epoxy grouting materials. In addition, 100% INEL soil encapsulations in TECT 1, WAXFIX, Carbray 100 Epoxy, and Phosphate cement grout materials were tested for compressive strength, hydraulic conductivity, and wet-dry cycle durability.

1.1 Materials

1.1.1 TECT 1 Grout

TECT 1 grout is a high density, low permeability, cementitious grout slurry designed for block encapsulation of buried waste using jet grouting systems.[1] This low viscosity grout can be mixed and delivered in concrete mixing trucks. In comparison, TECT 1 grout remains liquid longer than Portland cement slurries and is significantly less permeable than cement or cement/bentonite materials. Injecting TECT 1 grout into a soil matrix results in a firm product within several hours. Although the grout contains both organic and inorganic components, neither are combustible or hazardous. The vendor describes the product as being highly resistant to degradation caused by freezing-thawing or chemical attack. The vendor also claims that the immobilization of low concentrations of nitrate salts and organic liquids have resulted in an improvement over hydraulic cements. High compressive strengths are achieved following grout curing.

TECT 1 grout is a proprietary two component system consisting of a dark red dry powder and an orange liquid. However, tap water may be added to the mixture if more economy and lower viscosity is desired. The two components are mixed together using low shear at ratios of 100 parts by weight of powder to 35.2 parts by weight of liquid. The mixing of the two grout components may cause the material to form lumps and, therefore, must be removed by using a 40 mesh screen prior to jet-grouting. The vendor states that the slurry mix is jet groutable for at least 12 hours, but can be optimized to 36 hours if necessary. The normal density range for TECT 1 grout is 2.2 - 3.3 g/cm^3. A TECT 1 grout density of 2.09 g/cm^3 was measured when 4.8 wt% tap water was added to the mixture.

1.1.2 WAXFIX Grout

WAXFIX 125 is a low melting point wax, which has been modified with other polymers and a surfactant. The material has been formulated for jet-grouting applications where radioactive/mixed/hazardous waste stabilization/retrieval or close-coupled barrier fabrication is required. The molten wax is miscible with both wet and dry soil and has a greater ability to penetrate lower permeable soils than water. WAXFIX may be mixed with soil at ratios as high as 3 parts soil to 1 part WAXFIX by volume. Upon cooling, the wax produces a solid, low, permeable matrix which remains moderately soft, plastic, and chemically durable in the environment. Pumpable through wet water piping, this wax material can be cleaned by flushing with hot water. Due to the plastic nature of the WAXFIX grout, it is expected to generate very little airborne dust if excavating is required.

This grout consists of two components, WAXFIX 125 and WAXFIX 12. The grout is prepared by heating 100 parts by weight of the WAXFIX 125 component to 71- 77°C (160 to

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[1] Grout material product description and laboratory mixing instruction document received from the vendor, Carter Technologies, Sugar Land, Texas.

170°C), followed by the addition of 12 parts by weight of WAXFIX 12. Blending the two components in the laboratory using a low shear mixer is acceptable. However, the material has been designed for the high energy conditions used during jet grouting. The WAXFIX fluidity is maintained when blended with high concentrations of soil. The vendor MSDS states that the density of the WAXFIX grout is between 0.8 and 0.9 g/cm^3 and the flashpoint is ~207 °C. A molten WAXFIX density of 0.77 g/cm^3 was measured at BNL.

1.1.3 Carbray 100 Epoxy Grout

Carbray 100 is a two component, low viscosity, water miscible, elastomeric epoxy.[1] The vendor specified a diluent to reduce the cost of the grout system. The diluent was a 50% solids lignosulfonate liquid that was provided by the vendor. Lignosulfonate is a sticky black syrup derived from wood pulp, similar to the glue used in plywood fabrication. The vendor cautions that lignosulfonate is water soluble. Unless the material is blended into a very fine water internal/oil external emulsion, the diluent will be removed from the epoxy with repeated water washes. The Carbray 100 Epoxy system was formulated specifically for jet grouting applications when encapsulation and/or buried waste stabilization/retrieval is required. In addition, the vendor prefers that the two or three component system be premixed into a single low viscosity fluid prior to jet grouting. The consistency of the mixed components is similar to pancake syrup or 5W30 weight motor oil. Carbray 100 Epoxy is miscible with water prior to curing. The vendor claims the grout material permeates/mixes/ bonds well to oil and wet or dry soils. A very soft, low-permeability, chemically resistant rubber similar to silicone caulking is produced when the grout is blended with a soil or waste matrix. The mixed components have a pot life of approximately 8 hours. Clean-up of this pumpable material can be performed using warm water flushing. Furthermore, little airborne dust is expected when excavation of jet grouted soil/ stabilized waste products are required.

The three components will be identified by the letters A, B, and C for the purpose of this program. Component "A" is the hardener, component "B" is the epoxy resin, and component "C" is the diluent, a lignosulfonate liquid. The components were mixed in the following ratio: 1 part "A", 3.25 parts "C", and 2 parts "B" by weight. The order of mixing was important. Initially, components "A" and "C" are preblended using high shear mixing. Component "B" is then introduced and all three components are combined and blended using a low shear mixer (for laboratory mixing only). The MSDS provided by the vendor states that the density and flashpoint of the Carbray 100 Epoxy grout material is 1.2 g/cm^3 and >93°C, respectively. A Carbray 100 epoxy grout density of 1.05 g/cm^3 was measured by BNL when the diluent, lignosulfonate was used.

2.0 GROUT MATERIAL ENCAPSULATIONS

Four different grout materials were used to encapsulate two different waste streams. Three of the grouts, TECT 1, WAXFIX, and Carbray 100 Epoxy were provided by Carter Technologies, (Contact - E. Carter, Sugarland, Texas) and samples prepared by BNL. The fourth grouting candidate, phosphate cement was fabricated by Dileep Singh of Argonne National Laboratory and specimens submitted to BNL for performance/durability testing. Preliminary scoping tests were performed on grout/waste materials to insure compatibility prior to final specimen preparation. In addition, grout candidates TECT 1 and Carbray 100 Epoxy were prepared to confirm the vendor formulations. Melt temperature optimization and verification of vendor specifications using the thermoplastic, WAXFIX binder/waste stream were performed also.

Initially, INEL in consultation with Carter Technologies chose a waste form formulation that consisted of 33 wt% grout material and 67 wt% waste. The formulation is given in Table 1. In addition, INEL/Carter Technologies suggested that the waste stream be composed of 60 wt% INEL soil, 20 wt% sodium nitrate, and 20 wt% Canola oil. The ratio of components chosen represents an estimation of the average waste/soil composition that would occur when stabilizing in situ. These formulations were not verified via jet grouting prior to BNL's investigation. The percentages were later changed to 70 wt% INEL soil, 20 wt% sodium nitrate, and 10 wt% Canola oil (Table 1), based on the grout/waste verification studies performed by BNL. As preliminary scoping studies progressed, it became necessary to make additional adjustments to the grout/waste formulation in order to mix/encapsulate each waste stream in the three grouting materials effectively. The evolution and modifications to grout/waste stream compositions are described below.

Table 1. The standardized grout material/waste stream formulation that was expected to be used for all waste form preparations.

Standard Grout/Waste Stream Formulation

Component	*Weight %*
Grout	*33*
Waste Stream	*67*
INEL Soil	*70*
Sodium Nitrate	*20*
Canola Oil	*10*

This laboratory study required constituents to be mixed in a certain order in order to simulate burial conditions, yet provide adequate waste stream component mixing/encapsulation. First, the Canola oil was added to the soil and blended until homogenized, using a planetary mixer. Then, the sodium nitrate was added to the oil/soil mixture and blended thoroughly. The blended waste stream was mixed with each of the grout materials, as specified by the vendor (Carter Technologies).

2.1 TECT 1 Grout

Scoping tests were first performed using the two component (Powder - 74 wt% and Liquid - 26 wt%) TECT 1 grouting system. It was determined during formula verification testing that the addition of 20 wt% Canola oil to 60 wt% INEL soil resulted in soil saturation. This would result in a free standing oil requiring encapsulation by the grout. Earlier testing indicated that the grout had only a small miscibility with oil. Further testing indicated that 10 wt% Canola oil could be mixed with 70 wt% INEL soil, thus avoiding the potential soil saturation/encapsulation problem. Varying the percentage of sodium nitrate (5, 10, 15, 20, 25, 50, and 67 wt%) addition to TECT 1 grout was examined also. When mixture stirring was discontinued, all TECT 1/sodium nitrate compositions setting occurred within 45 to 120 seconds to the extent that inverting the containment cup resulted in no product loss. The temperature decreased by 6 - 7°C during component mixing regardless of the ratio tested. This initial quick set of the TECT 1 grout in the presence of sodium nitrate is indicative of a compatibility problem between the waste and the grout. In addition, the final set of the grout from the gel to a hard, brittle compound was delayed (several days to two weeks), and was dependent on the sodium nitrate loading. All TECT 1/sodium nitrate mixes were solidified and hard after 30 days of curing.

Based on these preliminary results, a 33 wt% TECT 1 grout and a 67 wt% waste stream (INEL soil: 70 wt%, Sodium nitrate: 20 wt%, and Canola oil: 10 wt%) mix was prepared. This resulted in a mixture that was very dry and clumpy. On the recommendation of the vendor, 3.6 wt% tap water was added to the grout material to improve the mixture fluidity. The water addition did not improve the grout/waste viscosity (still a dry mix), even when the percentage of tap water was increased to 4.8 wt%. Consequently, BNL decided to modify the grout/waste ratio. A viable encapsulation was achieved using a 60 wt% TECT 1 grout and a 40 wt% waste (~ 1:1 volume ratio).

The formulation used in the preparation of all TECT 1 waste forms for performance/durability testing is presented in Table 2. The TECT 1 grout powder and liquid components were blended together first, by using a low shear planetary mixer. Then, the tap water was then added and additional mixing was performed until mixture homogeneity was achieved. The production of minor material lumping was minimized with the introduction of tap water. Any remaining clumps were removed by forcing the slurry through a 40 mesh sieve (per vendor recommendation). The waste stream (thoroughly mixed) was then added to the grout and mixed until homogeneous. The grout/waste mixture was transferred to a PVC mold that was placed on a vibrating table to minimize air entrapment. It should be noted that a 6 - 7°C decrease in temperature was measured during grout/waste mixing, which can be attributed to the waste component, sodium nitrate. Preliminary verification studies using TECT 1 grout and sodium nitrate exhibited the same temperature decrease. Note, however, that there was no evidence of decreasing temperatures associated with TECT 1/INEL soil preparations.

The second waste stream considered for encapsulation in TECT 1 grout was 100% INEL soil. Initially, a TECT 1 /INEL soil optimization was performed. A maximum soil loading of 37 wt% was achieved using the TECT 1 grout formulation which included the 4.8 wt% tap water addition. Although the grout/soil mixed well initially, gelling occurred rapidly (90 secs) and made it impossible to mold the samples. This was further evidenced during the preparation of a TECT 1 /INEL soil mixture using the standardized formulation (Table 1), which resulted also in a very dry mix. To rectify this problem, BNL made a slight modification to the grout/waste formulation to 57 wt%

TECT 1 grout and 43 wt% INEL soil (100%). This resulted in a workable laboratory mixture. Note that no modifications to the TECT 1 grout component ratios were necessary (i.e., 70.4 wt% powder, 24.8 wt% liquid and 4.8 wt% tap water were used). This mixture represents a grout to INEL soil volume ratio of ~ 1:1.

Test samples were prepared by mixing the grout/waste streams as described earlier and pouring the mix into 5 cm diameter PVC tubing. The tubes were initially sealed at the bottom and after pouring were sealed at the top to retain moisture. All waste forms were cured in 75 cm length PVC molds for 30 days and then cut to 10.2 cm lengths using a masonry saw and diamond impregnated blade. Following baseline specimen mass and dimension measurements, all the waste forms were placed in 100% humidity storage until performance testing was started.

Table 2. The TECT 1 grout/waste stream formulation used in preparation of performance/durability test specimens.

TECT 1 Grout/Waste Form Formulation

Component	*Weight %*
Grout	***60***
Powder	*70.4*
Liquid	*24.8*
Tap Water	*4.8*
Waste Stream	***40***
INEL Soil	*70*
Sodium Nitrate	*20*
Canola Oil	*10*

2.2 Carbray 100 Epoxy

Verification pretests using the Carbray 100 Epoxy and the standardized waste stream (Table 1) resulted in a viable mixture and encapsulation. The formulation used in the preparation of Carbray 100 Epoxy/waste performance/durability specimens is presented in Table 3. This mixture represents a grout to waste stream volume ratio of ~ 1:1. The polymerization of a small Carbray 100 Epoxy/waste mixture (~200g) resulted in only a minor exothermic reaction ($\Delta T = +2\text{-}3\,^\circ C$).

It was necessary to contact the vendor to obtain workable mixing ratios because the INEL soil encapsulations in Carbray 100 Epoxy grout were not part of the original project test plan, Carter Technologies suggested preparing a 1 part epoxy grout (40 wt%) to 1.5 parts INEL soil (60 wt%). An acceptable mix was achieved by maintaining the grout component proportions that were used for the waste stream preparations (Epoxy resin - 32 wt%, Hardener - 16 wt%, and Lignosulfonate liquid - 52 wt%) and adding 100% INEL soil as the total waste stream. This mixture represents a grout to INEL soil volume ratio of ~ 1:1.5.

Table 3. The Carbray 100 Epoxy grout/waste stream formulation used in the preparation of performance/durability test specimens.

Carbray 100 Epoxy Grout/Waste Form Formulation

Component	***Weight %***
Grout	***33***
Hardener- "A"	*16*
Epoxy Resin- "B"	*32*
Lignosulfonate Liquid - "C"	*52*
Waste Stream	***67***
INEL Soil	*70*
Sodium Nitrate	*20*
Canola Oil	*10*

All Carbray 100 Epoxy grout/waste forms were molded and cured in 5.1 cm diameter x 75 cm lengths of PVC tubing for 14 days. The grout/waste forms were cut to 10.2 cm lengths using a masonry saw and diamond impregnated blade. Attempts to de-mold the specimens were unsuccessful. The adhesion properties of the epoxy grout/waste to the plastic PVC mold were so great that it was impossible to remove the specimen as one monolithic piece. As a consequence, eight additional 75 cm length sleeves of Carbray 100 Epoxy grout/waste were prepared. Prior to preparation, it was determined that the epoxy grout mixtures could be removed from the PVC molds successfully when a thin coating of paraffin wax was applied to the interior wall of the mold. This result pushed the date for the testing of these particular specimens back approximately two weeks. However, this re-preparation of Carbray 100 Epoxy grout/waste forms had only a minor affect on the testing schedule as described in the project test plan. After 14 days, the second batch of grout/waste forms were carefully removed from their PVC molds for 16 days of ambient air curing, as per recommendation of the vendor. Mass and dimensional measurements were performed. The specimens were very soft, sticky to the touch, and had low mechanical strength. Exposure to the air accelerated the curing process as indicated by the specimen firming and exterior surface crusting. However, the specimen mass and volume losses were measured to be 5.4 wt% and 8.1%, respectively between the time the Carbray 100 Epoxy/waste specimens were removed from the PVC mold (14 days) for air-drying (16 days) to the completion of the total 30 day cure period. Same specimen density comparisons revealed slight increases from 1.53 ± 0.005 g/cm^3 to 1.56 ± 0.02 g/cm^3 after air curing. All waste forms were placed in 100% humid storage until performance testing was to be started.

The grout/waste PVC mold adhesion problem also occurred with the Carbray 100 Epoxy grout/INEL soil mixes and, as a consequence, four additional grout/waste mixes were prepared in 75 cm lengths of PVC tubing. The procedure for curing as described above was followed also for these composites. The Carbray 100 Epoxy/INEL soil specimens exhibited similar losses that resulted in considerable mass (>5%) and volume (~10%) decreases after 18 days of ambient air curing. Composites were placed in 100% humidity storage until performance/durability testing was to be conducted.

2.3 WAXFIX

Preliminary compatibility/formulation tests were performed using the WAXFIX grouting material and waste stream components (Table 1). WAXFIX and 10 wt% Canola oil were mixed and heated to 75°C. The Canola oil separated from the WAXFIX grout as the mixture cooled. The immiscibility of this material was quite evident following total product cool-down. For example, the sequestered oil became entrapped in a central cavity within the form as the mixture cooled, while remaining completely encapsulated by WAXFIX grout. As observed with TECT 1/Canola oil encapsulations, the oil needs to be mixed in with the soil before a homogeneous product can be prepared. Also, it was difficult to mix the sodium nitrate and WAXFIX grout effectively in the laboratory. The nitrate quickly settled to the bottom of the container with the termination of mixture stirring and was encapsulated in the rapid product cooling that occurs at the peripheral edges of the mold. Once again, the soil matrix is required to thicken the mix and suspend the waste components. It is assumed that the mixing conditions tested in the laboratory should not be fully encountered at the INEL burial site since soil and/or saturated waste should be major components at the burial site. WAXFIX grout/soil optimizations were performed also. The maximum soil incorporated in WAXFIX grout was 86.9 wt%. However, an optimal soil loading was achieved between 65 - 70 wt% for practicality and efficient mixing (i.e., good homogeneity and soil suspension in grout). It was determined that loading the WAXFIX grout with >70 wt% soil resulted in a mix that was too thick and difficult to pour/transfer in the laboratory. Conversely, using a lower soil ratio (<65 wt%) resulted in a settling problem. Therefore, based on these results the standardized formulation (Table 1) of 33 wt% WAXFIX grout and 67 wt% waste was mixed successfully and used for all performance/durability test specimen preparations. The WAXFIX grout/waste formulation is presented in Table 4. This mix represents a grout to waste stream ratio of 1:1 by volume.

Table 4. The WAXFIX grout/waste stream formulation used in the preparation of performance/durability test specimens.

WAXFIX Grout/Waste Form Formulation

Component	***Weight %***
Grout	***33***
WAXFIX 125	*89.3*
WAXFIX 12	*10.7*
Waste Stream	***67***
INEL Soil	*70*
Sodium Nitrate	*20*
Canola Oil	*10*

WAXFIX grout/INEL soil composites were prepared for limited performance/durability testing although not part of the original project test plan. A 33 wt% WAXFIX grout and a 67 wt% waste stream (100% INEL soil) resulted in a viable laboratory mixture. This mix represents a grout to INEL soil volume ratio of ~ 1:1.

All WAXFIX grout/waste forms were molded in 5.1 cm diameter x 75 cm length PVC tubing. After 30 days of curing, the specimens were cut to 10.2 cm lengths using a masonry saw and diamond impregnated blade and de-molded. Material shrinkage following cool-down was fairly significant for this WAXFIX/waste mixture. Theoretically, a 75 cm length of PVC tubing should provide seven 10.2 cm length cylindrical waste forms per tube. However, material shrinkage during cooling sometimes resulted in only 4 - 5 viable specimens available for testing purposes. The grout/waste cooling-shrinkage started at the top of the molding tube in the shape of a funnel which narrowed to a ~1 cm diameter cylindrical void that migrated off-center down the mixture for ~ 20 to 30 cm. It is possible that lowering the grout/waste processing temperature would minimize material shrinkage during cool-down. Specimen mass and dimensions were measured and they were then stored in 100% humid environment until performance testing began.

WAXFIX/INEL soil specimens were removed from their molds in the same manner as above. However, it was observed while cutting these mold/specimens to their 10.2 cm lengths that a narrow, off-center, void channeled the length of each waste form. Apparently, this void developed during sample cooling. New WAXFIX/INEL soil specimens were prepared by melting the existing forms and molding them at a lower temperature. This procedure proved to minimize both shrinkage and void generation during mixture cool down. But still resulted in only 4 - 5 (40 to 50 cm total) useable specimens from each 75 cm length PVC sleeve. Performance testing included: baseline compressive strength, wet-dry cycling, and hydraulic conductivity measurements.

Phosphate-bonded INEL soil specimens were prepared by Dileep Singh of Argonne National Laboratory (ANL) and shipped to BNL after curing. BNL received 21 cylindrical specimens measuring 5.1 cm in diameter with lengths ranging between 7.5 - 10.1 cm. The nominal soil loading in these samples was ~58 wt%. Therefore, the phosphate cement fraction was ~42 wt%. No details describing sample preparation were forwarded to BNL. However, ceramic grouts are usually prepared at room temperature by mixing contaminated solids (e.g., ash, soil, sludge) with either phosphoric acid, or powdered phosphates that dissolve in water. The mixture supposedly thickens immediately and sets in ~2 hours. Only four specimens had a measured length greater than 8.9 cm. This put limitations on the number replicates that could be tested for compressive strength (baseline only), wet-dry cycling, and hydraulic conductivity (shorter length requirements) measurements. After recording Phosphate ceramic/INEL soil dimensions and mass, the specimens were stored in a 100% humid environment. Prior to durability testing, the specimens were removed from humid storage and dimension and mass were measured again. Phosphate cement/INEL soil composites gained 0.6% moisture during this 30 day storage, but no dimensional changes were recorded.

A summary of the grout material and waste stream and soil ratios used in the preparation of durability/performance test specimens are presented in Table 5.

Table 5. A summary of the grout/waste and soil ratios used in the preparation of durability/performance test specimens.

Waste Form Type	**Percent Grout (wt%)**	**Percent Waste (wt%)**			**Volume (cm^3) of Grout per 100 g of Filler**
		Canola Oil	**Sodium Nitrate**	**INEL Soil**	
TECT 1/Waste	60	4	8	28	72
TECT 1/INEL Soil	57	0	0	43	63
Carbray 100/Waste	33	6.7	13.4	46.9	47
Carbray 100/INEL Soil	40	0	0	60	64
WAXFIX/Waste	33	6.7	13.4	46.9	64
WAXFIX/INEL Soil	33	0	0	67	64
Phosphate Cement/INEL Soil (a)	42	0	0	58	n/a

n/a = not applicable

(a) These specimens were prepared by Dileep Singh of Argonne National Laboratory and represent nominal values.

3.0 CHARACTERIZATION

Viscosities were measured on each of the grout materials (TECT 1, Carbray 100 Epoxy, and WAXFIX) prepared at BNL. Also, densities were measured for all 30 day cured (baseline) grout/waste and INEL soil specimens that were to be used in performance/durability testing. In addition, the INEL soil received from the Geotechnical Engineering & Construction Company (AGEC) of Richland, Washington was characterized for particle size distribution, density, and moisture content.

3.1 Grout Material Characterization

3.1.1 Viscosity

Grout material viscosity is an important property that needs consideration prior to jet grouting injection/emplacement. The importance of this property is two fold. A relatively low-viscosity (usually < 300 cP) grout material is required since jet grouted materials are pumped, and then injected through tiny nozzle orifices (2 - 5 mm) at high pressures into the surrounding soil/waste medium. Secondly, the penetration depth of the grouts into soil/waste formations is dependent on material viscosity (formation composition and injection pressure are important also). Therefore, both the grout and grout components of the three materials studied were measured for viscosity using a Brookfield Model DV-II+ viscometer.[1] Grouts were mixed according to vendor specifications using the formulations used to prepare 5.1 cm x 10.2 cm performance and durability specimens. All measurements were performed in a 600 mL, low form, griffin beaker, containing approximately 500 mL of test slurry, as recommended by the viscometer manufacturer. The results of these tests are summarized in Table 6.

The three component TECT 1 grout viscosity measurement presented some difficulties because of the modest settling by the denser powder component. Ordinarily, this would not be a problem during jet grouting applications because the mixture is mechanically agitated or stirred. However, this was not possible during the viscosity measurement. Therefore, prior to performing the measurement, the mixture was stirred vigorously by hand before the spindle was lowered into the TECT 1 grout slurry. Measurements were taken after one minute intervals, to insure both reproducibility, and to minimize the affects of settling by the denser components. The viscosity of the three component TECT 1 grout mix was 145 - 150 cP @ 18.5°C. The viscosity of the liquid portion of the TECT 1 grout measured between 5 - 6 cP @ 18°C.

The two component WAXFIX grouting system (WAXFIX 12 and WAXFIX 125) are mixed and injected at elevated temperatures (55 - 75 °C). The measured viscosities for WAXFIX 12 and WAXFIX 125 were ~1000 cP @ 20°C and 4 - 5 cP @ 70°C respectively. A very low viscosity between 5 - 6 cP @ 65°C was obtained when the two WAXFIX components were mixed together.

The three component Carbray 100 grout system was processed at room temperature. Viscosity measurements were performed on both the individual and the two mixed component combinations. For example, component "A" (hardener) measured 20 - 25 cP @ 20°C and component "C" (lignosulfonate) measured 1100 - 1150 cP @ 19°C. However, a viscosity measurement of ~500 cP @ 22°C was obtained after high shear blending of the "A" (16 wt%) and "C" (52 wt%) components was implemented. A viscosity of 6600 cP @ 21°C was measured for Component "B"(epoxy resin). It should be noted that although, the component "B" is pumpable and injectable, the viscosity is relatively high for jet grouting applications. Additionally, when component "B" (32 wt%) is mixed with blended components "A" and "C", the viscosity of the grout increases to 6650 cP @ 33°C. The relatively high viscosity measurement could be attributed to polymerization and/or the lack of continual mixing (thixotropic properties) of the grout material. Also note, the increase in temperature ($\Delta T \approx +12°C$) associated with the mixing of the three components.

3.1.2 Density

After 30 days of curing, specimens' mass and dimensions were measured. The homogeneity of each grout form was measured by comparing the densities of each of the individual composites. Mean specimens' densities are given in Table 7. The results show that specimens' homogeneity was excellent with 2 sigma errors <1%.

Table 6. Grouting material viscosity measurements.

Grouting Materials	Temperature, °C	Viscosity, cP
TECT 1		
Liquid	18	5 ± 1
Powder + Liquid + Tap Water	18.5	140 ± 10
WAXFIX		
WAXFIX 125	70	4 ± 1
WAXFIX 12	20	1000 ± 50
WAXFIX (125 + 12)	65	5 ± 1
Carbray 100 Epoxy (1)		
Carbray "A"	20	22 ± 5
Carbray "C"	19	1100 ± 50
Carbray (A + C)	22	500 ± 10
Carbray "B"	21	6500 ± 50
Carbray (A + C) + B	33	6600 ± 50

(1) Carbray 100 Epoxy components: "A" = Hardener, "B" = Epoxy resin, and "C" = Diluent.

Table 7. Densities of grouted waste and INEL soil composites.

Grout Material	**Mean Density, g/cm^3**	
	Standard Waste Stream	**INEL Soil**
TECT 1	2.02 ± 0.004	2.08 ± 0.01
WAXFIX	1.36 ± 0.01	1.45 ± 0.02
Carbray 100 Epoxy	1.53 ± 0.005	1.53 ± 0.006
Phosphate Cement	n/a	1.62 ± 0.01
n/a = not applicable		

3.2 INEL Soil Characterization

One, partially filled 55 gallon drum of INEL soil was shipped to BNL by the Applied Geotechnical Engineering & Construction Company (AGEC) of Richland, Washington for this study. Representative samples were removed from the drum and characterized for particle size distribution, density, and moisture content.

3.2.1 Moisture Content

Moisture content of as-received INEL soil was determined using two different methods. The ASTM Standard D-2216, "Laboratory Determination of Water (Moisture) Content of Soil, Rock, and Soil-Aggregate Mixtures," [2] was used to evaluate the soil sample moisture content. The procedure involves taking a representative soil sample, oven drying at a temperature of 110 ± 5°C for a minimum of 18 hours, and then measuring for moisture content. Confirmatory measurements were made using a Sartorius MA30 Automatic Moisture Analyzer. [3]

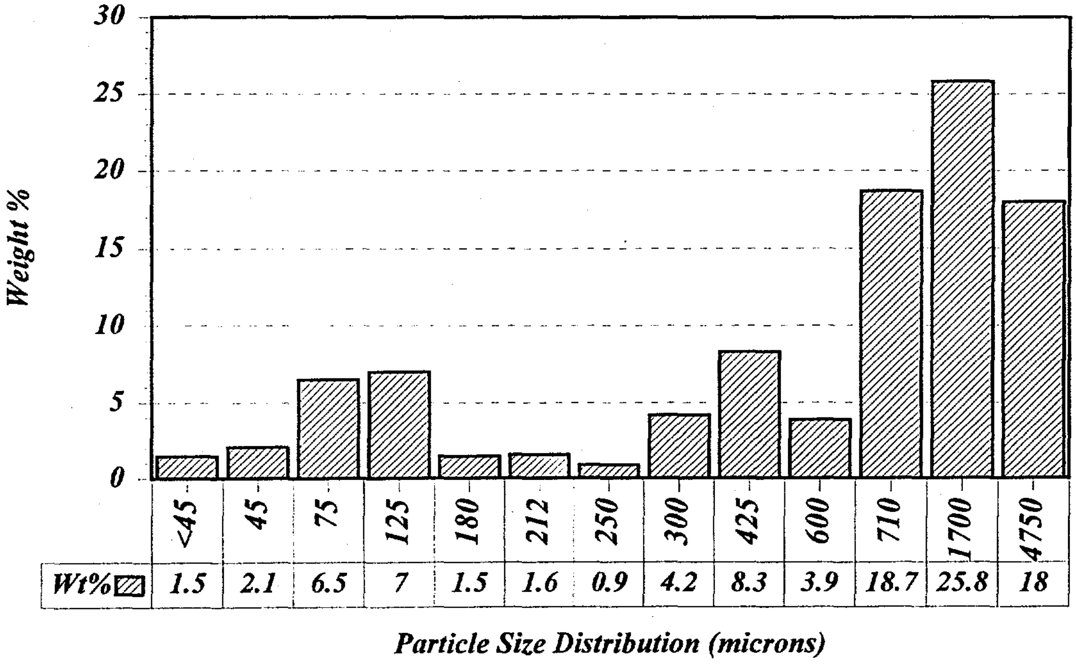
Moisture content results were based on the average of five replicate specimens, which were reported with 2σ errors. The measurements obtained using Method D-2216 and the Sartorius MA30 Automatic Moisture Analyzer were 13.6 ± 0.2 wt% and 10.6 ± 1.3 wt%, respectively. Reasonable agreement between the two moisture analysis methods was indicated, with a mean INEL soil moisture content measurement of ~12 wt%.

3.2.2 Density

Density measurements were performed on the INEL soil using a Quantachrome Multipycnometer, Model MVP-1 [4] to measure the actual volume, and a Mettler electronic balance to weigh the soil. The multipycnometer measures the true volume of solid materials using Archimedes' principle of fluid displacement to determine the volume. The instrument uses an inert gas (helium) as the displacement fluid which is capable of penetrating fine pore spaces. All soil specimens were oven dried initially at 110°C for a minimum of 18 hours prior to performing density measurements. The volumes of four replicate soil samples were measured resulting in an average density of 2.28 ± 0.02 g/cm^3.

3.2.3 Particle Size Distribution

A sieve analysis was performed on the INEL soil according to ASTM Standard C-136, "Sieve Analysis of Fine and Coarse Aggregates." [5] All soil specimens were oven dried initially at 110°C for a minimum of 18 hours prior to the sieve analysis. Six kilograms of dried soil was used to determine the particle size distribution. Twelve different sieve sizes were selected for this analysis, with sieve openings measuring between 45 μm and 4750 μm. A Ro-Tap, Model B, mechanical sieve shaker manufactured by Tyler Industrial Products was used to agitate the sieves and soil samples. Sieving did not exceed 10 minutes for each fraction tested in order to minimize soil specimen degradation as per the ASTM Standard. The INEL soil sieve analyses are shown in Figure 1. Table 8 indicates that 33.6 wt% of the soil particles were <600 μm, while 48.4 wt% of the larger particles measured between 600 and 4750 μm. Approximately 18 wt% of the soil particles were greater than 4750 μm. The proportionately higher percentage of larger particles (>600 μm - 66.4 wt%) may be due, in part, to the soil composition (e.g., moisture and mineral content) and the natural "clumping" observed while performing the test.



	<45	45	75	125	180	212	250	300	425	600	710	1700	4750
Wt%▨	1.5	2.1	6.5	7	1.5	1.6	0.9	4.2	8.3	3.9	18.7	25.8	18

Figure 1. Particle size distribution of INEL soil.

Table 8. Particle size distribution of INEL soil.

Particle Size Distribution (μm)	*Weight Percent of Total INEL Soil*
<125	10.1
<250	20.2
<600	33.6
<4750	82.0
>4750	18.0

3.3 DOT Oxidizer Test

The oxidizing potential of nitrate salt encapsulated in refined paraffin wax was determined as part of the initial scoping studies.

Strong oxidizers, such as nitrate salts have the potential to become hazardous if involved in an accidental fire during shipping or storage. The recommended test procedure for quantifying hazards associated with solid oxidizing materials such as nitrates is identified in the Transportation volume (49) of the Code of Federal Regulations (CFR) under Part 173, "Shippers - General Requirements for Shipments and Packaging." The definition of an oxidizer and the assignment of packing groups is described in 173.127, Class 5, Division 5.1. The actual procedure is located in Appendix F entitled "Guidelines for the Classification and Packing Group Assignment of Division 5.1 Materials" (49 CFR 173.127, Appendix F, Revised October 1, 1995).

The test method for solid oxidizing substances "is designed to measure the potential for a solid substance to increase the burning rate or burning intensity of a combustible substance when the two are thoroughly mixed." [6] This test was used by BNL to determine whether the paraffin wax encapsulated nitrate salt would be classified as a DOT oxidizer. A description of the procedure and the results are summarized below.

The test procedure requires three reference chemicals to be used: ammonium persulfate (fine crystals), potassium perchlorate(fine crystals),and potassium bromate (powder).These chemical substances were passed through a No. 50 (300 μm) sieve. The potassium compounds passed through the sieve without much difficulty. However, the ammonium persulfate was a larger sized crystal and slightly moist, thus causing caking and clogging of the sieve mesh. The chemical compound was placed in a mechanical convection oven for 18 hrs at 65°C and then ground to a smaller particle size using a mortar and pestle in order to resolve these problems. The ammonium persulfate was sieved without difficulty following this treatment It should be noted that this test procedure advises against grinding the reference compounds. However, this was the only form of the reagent available. Thus grinding was necessary. Additionally, all the reference chemicals were placed in a mechanical convection oven overnight (18 hrs) at 65°C and then cooled and stored in a desiccator.

A combustible material, wood sawdust, was also required for this DOT solid oxidizing test. The wood sawdust was obtained from the BNL Carpenters Shop. It was passed through a No. 10 (2.0 mm) and a No. 14 (1.4 mm) sieve. All the sawdust that did not pass through the No. 14 sieve was used in testing. The sawdust was placed on glass pyrex trays, spread to an approximate thickness of 25 mm, dried in a mechanical convection oven at 110°C for 4 hrs, and cooled and stored in a desiccator.

The test materials consisted of the following compositions:

- Refined paraffin wax cut and sieved to a particle size < 2 mm and mixed with sodium nitrate in mass ratio of 1:1.
- Sodium nitrate salt encapsulated in refined paraffin wax in a 1:1 mass ratio,
- Sodium nitrate salt encapsulated in refined paraffin wax in a 1:1 mass ratio, but cut to a sieve mesh size < 9.5 mm following solidification,
- 100% sodium nitrate salt.

The first material to be tested consisted of small particles of refined paraffin wax (< 2 mm) which were mixed with sodium nitrate in a mass ratio of 1:1. The smaller sized paraffin wax particles were obtained by taking the shavings from a larger piece of wax and chopping them into smaller pieces using a single-edge razor. The paraffin wax was sieved to a particle size < 2 mm, and then thoroughly mixed with a granular sodium nitrate in a mass ratio of 1:1. Sodium nitrate salt encapsulated in paraffin wax (1:1 ratio) was the second material to be tested. Preparation of this mixture involved melting the wax in a beaker placed in boiling water. Once completely melted, the sodium nitrate salt was added directly to the wax and thoroughly mixed. However, the salt immediately accumulated on the bottom of the beaker. It was necessary to permit the mixture to cool down and become more viscous to insure wax/sodium nitrated homogeneity before mechanical mixing was once again initiated, to achieve the desired results. The sodium nitrate encapsulated in paraffin wax was tested with two individual pieces of solidified material (weighing 15 g and the other 24 g, respectively). The third composition to be examined was also the 1:1 mixture of encapsulated sodium nitrate in paraffin wax, but now the solidified product was cut to a sieve mesh size < 9.5 mm. The majority of these cut pieces had a cubic or angular shape which just pass through a 9.5 mm sieve. The final material to be tested was 100% sodium nitrate salt. This chemical was used directly from the bottle without any pretreatment (e.g., drying or sieving).

Each reference chemical (15 g) was combined with the wood sawdust (15 g) and mixed thoroughly to form a mass ratio of 1:1. The test materials were mixed thoroughly with sawdust in 1:1 (15 g : 15 g) and 4:1 (24 g : 6 g) mass ratios. Each test performed included the three reference chemicals (1:1 ratio) and the test materials at the two different mixture ratios (1:1 and 4:1). Test piles were placed on an insulating board, over an inert metal heating wire to form conical piles measuring between 80 - 100 mm in diameter and between 35 - 45 mm high. It was impossible to obtain the 70 mm diameter x 60 mm height recommended in the procedure due to the inconsistency of the reference chemicals/sawdust and the various shapes of the test materials The larger, encapsulated test materials (one large piece or < 9.5 mm cubes) were placed on top of a layer of wood sawdust (~5 mm thick) and then covered with the remaining sawdust. An attempt was made to cover all test material with sawdust. However, it was not always possible with the larger solidified materials (#2 and #3).

The heating apparatus was fabricated using a nickel (80%) - chromium (20%) resistance wire (0.6345 ohms/ft), approximately 66 cm long, attached to copper terminal blocks, and anchored to a metal base plate with the thermosetting plastic, Bakelite as the insulators. A thermal insulating fiber board was placed under the wire with a gap measuring 2 - 3 mm between the wire and board. Voltage was applied to the wire using a DC power supply. The power supply settings of 23.5 volt and 16 amps provided the 1000°C wire temperature required to perform the test. The temperature of the wire was calibrated using an optical infrared pyrometer. The temperature varied along the wire by about 20°C. The ignition time of each substance was recorded and the burning time observed until the flame of the last mixture disappeared. All solid oxidizing tests were photographed using a video camera recorder to insure the accuracy of burn time measurements.

A total of six solid oxidizing tests were performed. The refined paraffin wax was cut and sieved to a particle size < 2 mm and mixed with sodium nitrate in mass ratio of 1:1 (#1). The sodium nitrate salt was encapsulated in refined paraffin wax in a 1:1 mass ratio (#2). These two tests were repeated twice. The sodium nitrate salt was encapsulated in refined paraffin wax in a 1:1 mass ratio, but cut to a sieve mesh size < 9.5 mm (#3) and the 100% sodium nitrate salt (#4) were each run once. The mean burn times with 2σ errors for each of the reference chemicals are presented in Table 9. Reference chemicals showed the following mean burn time (sec) order: Potassium Bromate (14.5 ± 2.2 sec.) < Ammonium Persulfate (63.7 ± 13.9 sec.) < Potassium Perchlorate (90.7 ± 19.6 sec.). These mean burn times were used to classify each of the test materials. Table 10 shows the burn times for each of the materials tested at mass ratios of 1:1 and 4:1 (i.e., test material to wood sawdust ratios).

Results indicated that the nitrate salt encapsulated (one large piece or cubes cut to < 9.5 mm size) in refined paraffin wax burned significantly slower (by a factor of 1.5 to 56 times) and less violently than any of the reference oxidizing materials. Also, the nitrate salt mixed with the chopped paraffin wax (particle size <2 mm) resulted in burn times that were ~2 to 13 times slower than the reference oxidizing materials.

Based on these results, sodium nitrate solidified or just mixed with refined paraffin wax is not classified as an oxidizer by the U.S. DOT and need not meet regulations for shipping of oxidizers. The unsolidified nitrate salt, as expected, was classified as an oxidizer by the U.S. DOT.

Table 9. Combustion time (sec) test results for each of the reference chemicals required in the solid oxidizer substance test.

Combustion Time for Reference Chemicals			
Test Number	**Reference Chemical Compounds**		
	Potassium Bromate	**Ammonium Persulfate**	**Potassium Perchlorate**
	Combustion Time (sec)		
1	11	52	84
2	17	59	95
3	17	65	102
4	14	58	119
5	13	92	58
6	15	56	86
Mean (a)	**14.5 ± 2.2**	**63.7 ± 13.9**	**90.7 ± 90.7**

(a) Mean values for 6 replicates with 2σ errors.

Table 10. Combustion time (sec) results for each of the materials tested in the solid oxidizer substance test.

Combustion Time for Test Materials		
Material Composition	**1 : 1 Mass Ratio Test Mixture**	**4 : 1 Mass Ratio Test Mixture**
	Burn Time (sec.)	
50% Chopped Wax (<2 mm)/50% $NaNO_3$	**161 (a)**	**188 (a)**
Encapsulation: 50% Wax/50% $NaNO_3$	**131 (a)**	**819 (a)**
Encapsulation: 50% Wax/50% $NaNO_3$, Cut and Sieve to <9.5 mm	**628 (b)**	**525 (b)**
100% $NaNO_3$	**37 (b)**	**25 (b)**

(a) Mean of two replicates.

(b) Based on one replicate.

4.0 TESTING AND RESULTS

This investigation was conducted to determine the viability of several grouts for *in situ* treatment/stabilization of buried waste and interstitial soils at the INEL. Therefore, the grouting materials had to be compatible with conventional jet grouting techniques, the soil, environmental and geotechnical characteristics of the site, and the contaminants and chemistry of the buried waste.

Seven performance and durability tests (compressive strength, wet-dry cycling, thermal analysis [differential scanning calorimetry], base immersion, solvent immersion, hydraulic conductivity, and accelerated leach testing) were conducted on specimens prepared using the standardized waste stream (70 wt% INEL soil, 20 wt% sodium nitrate, and 10 wt% Canola oil) and encapsulated in TECT 1, WAXFIX, and Carbray 100 Epoxy grouting materials. In addition, 100% INEL soil encapsulations in TECT 1, WAXFIX, Carbray 100 Epoxy, and Phosphate cement grout materials were tested for compressive strength, hydraulic conductivity, and wet-dry cycle durability. Unless otherwise stated, all results were based on the average of five replicate specimens, which were reported with 2σ errors at the 95% confidence level. Furthermore, all samples were cured for 30 days, removed from their molds, baseline mass and dimensional data recorded, followed by immediate storage in a 100% humid environment to replicate conditions expected in the subsurface. In all cases, specimens were placed in 100% humidity for a minimum conditioning period of four days.

4.1 Hydraulic Conductivity

One of the most important performance criteria for buried waste forms is the rate at which liquids will penetrate the product. Hydraulic conductivity is a direct measure of the ability of the solidified grout/waste/soil product to encapsulate effectively, prevent leakage, and exclude percolation waters. The hydraulic conductivity of each grout/waste and grout/INEL soil specimen was measured using a flexible wall permeameter (see Figures 2 and 3) following ASTM method D-5084. [7] The permeant was a de-aired tap water with an electrical conductivity of ~120 μmhos. Test specimens were right cylinders, nominally measuring 3 cm in height by 5.1 cm diameter. All samples were preconditioned by placing them in a vacuum desiccator and evacuating (<1 mm Hg) for 3 hours. De-aired water was then introduced into the desiccator at the end of three hours, without losing the vacuum. The vacuum was applied for an additional hour after the samples were covered with water. Samples were left in the de-aired water for a minimum of one week or until the hydraulic conductivity measurement was to be performed in order to induce saturation. The water in the desiccator was de-aired periodically by re-applying a vacuum to the desiccator for 1 - 2 hours.

After pre-conditioning, the test specimens were placed in the permeameter cell and sealed using a latex membrane and a 35 kPa differential cell pressure. Back pressuring was used to accomplish full saturation of the specimens. Full saturation was determined by measuring the pore pressure parameter B according to ASTM D-4767-(88).[8] The B value was ≥0.95 for all specimens tested except for the Carbray 100 Epoxy/waste which was difficult to back saturate. After full saturation was achieved, the measurement was started. The cell pressure was set to 310 kPa (45 psi), the lower cap to 275 kPa (40 psi), and the upper cap pressure to 70 kPa (10 psi). This resulted in a pressure differential across the specimen of 210 kPa (30 psi). The inflow and outflow of the water was measured at ~24 hour intervals.

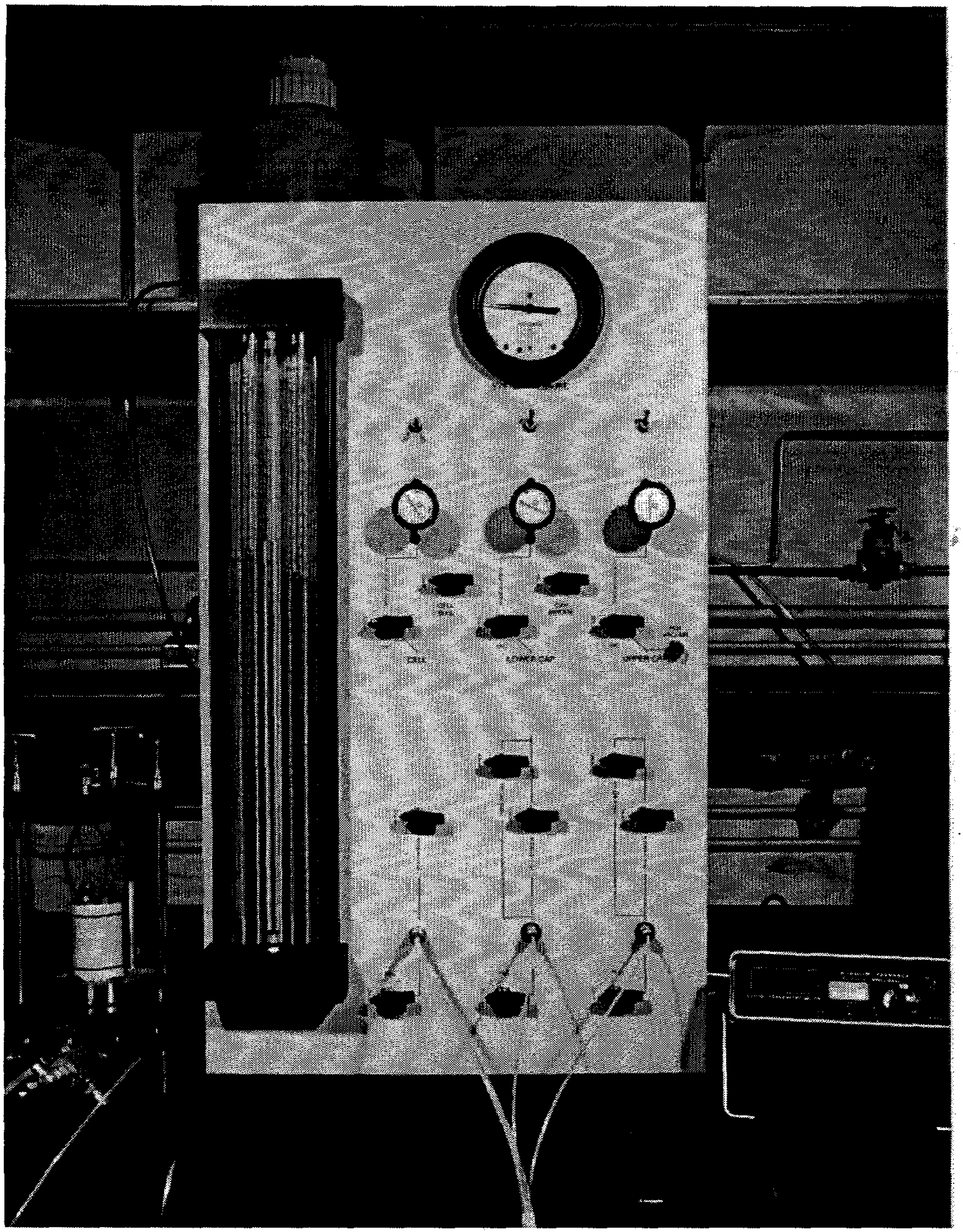


Figure 2. Flexible wall permeameter used to measure hydraulic conductivities.

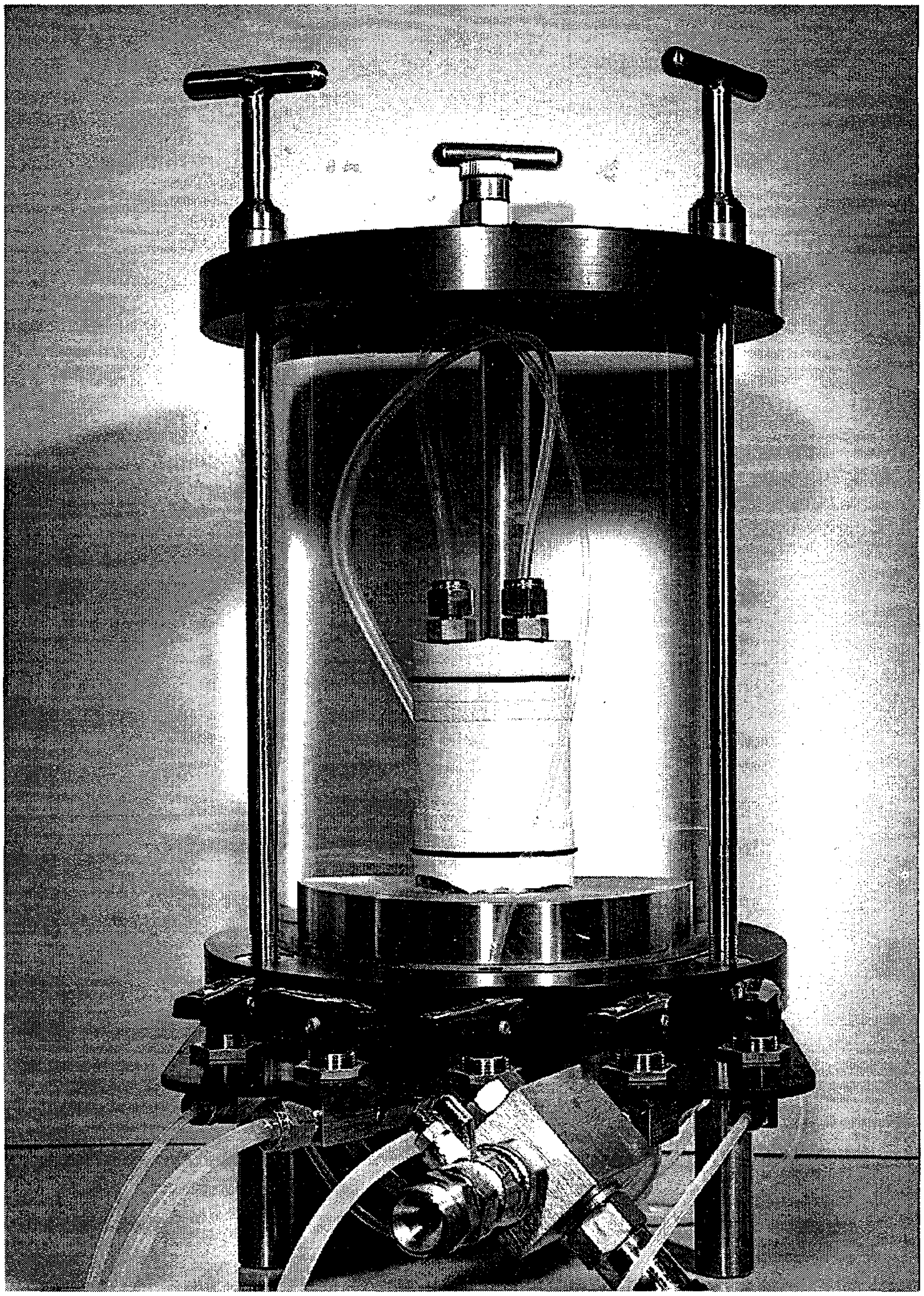


Figure 3. Permeameter cell showing a grout/waste specimen sealed with a latex membrane.

The hydraulic conductivity was calculated according to:

$$k = QL/Ath$$

where:

k = hydraulic conductivity, cm/sec.
Q = water quantity through the specimen, taken as the average of inflow and outflow, cm^3
L = length of the specimen, cm
A = cross sectional area of the specimen, cm^2
t = time interval, s over which Q was measured
h = pressure differential across the specimen

All permeability results were based on one hydraulic conductivity measurement except for the Phosphate cement which had two determinations. Results are presented in Table 11. The instrument limits were determined to be 2×10^{-11} cm/sec. All hydraulic conductivities except the Phosphate cement/INEL soil and Carbray 100 Epoxy/waste composite were below the instrument detection limit. The two measurements performed on Phosphate ceramic/INEL soil composites resulted in a mean hydraulic conductivity of $3.1 \times 10^{-7} \pm 1.5 \times 10^{-8}$ cm/s ($\pm 1\sigma$). For comparison, jet-grouted Portland cement slurry has a permeability of ~1.0×10^{-7} cm/s. The Phosphate cement/INEL soil was found to have approximately the same rate. A permeability value was not obtained for the Carbray 100 Epoxy/waste material because of the inability to de-gas the specimen during the back pressuring procedural step. Full saturation of the specimen could not be achieved because of the continual production of gas bubbles. Bubble generation required purging of the upper and lower cap feed lines every other day for a three week period. After three weeks, BNL decided to terminate back pressuring in order to test other grout composites.

Table 11. Hydraulic conductivities of grout/waste and INEL soil composites.

Grout Type	**Hydraulic Conductivity, cm/sec x 10^{-10} (a)**	
	Standard Waste Stream	INEL Soil
TECT 1	<0.2	<0.2
WAXFIX	<0.2	<0.2
Carbray 100 Epoxy	n/a (b)	<0.2
Phosphate Cement	n/a	3050
Portland Cement Slurry	~1000	

n/a = not applicable

(a) All results are based on one measurement except for Phosphate cement which represents the average of two hydraulic conductivity measurements.

(b) No measurement could be performed on the specimen because of the continual generation of gas bubbles during back saturation in the permeameter. The test was terminated to allow for other grout/waste specimen measurements.

4.2 Unconfined Compressive Strength

Five replicates of each of the 5.1 cm x 10.2 cm cylindrical grout/waste and grout/INEL soil specimens were tested for unconfined compressive strength in accordance with ASTM C-39 or ASTM D-695 in order to determine baseline strength. Usually, the compressive strength of a jet grouted encapsulated waste or soil product would not be an important parameter to test due to the low overburden forces (< 100 psi) present. However, this characteristic may take on greater significance for insuring minimum strength requirements, with the development and availability of many new grouting materials. In addition, the nature of the test makes it most useful when comparative results on the effects of testing (specimen integrity) between different grout/wastes and soil products are required. Therefore, the compressive strength values reported will be used as an indicator of the loss of integrity after resistance and durability testing.

All TECT 1 and Phosphate cement/INEL soil specimens were measured on a Soiltest Versa-Tester compression tester (270 kN or 60,000 lb capacity) in accordance with ASTM C-39.[9] All WAXFIX and Carbray 100 Epoxy grout composites were compression tested using an Instron Universal Testing Instrument, Model 4202 (9 kN or 2,000 lb capacity), following method ASTM D-695. [10] Method D-695 was used to test the WAXFIX and Carbray 100 Epoxy grout encapsulation products because they might have exhibited non-rigid, plastic characteristics, resulting in specimen failure without brittle fracture. Therefore, the test (Method D-695) was slightly modified to include, compressive strength measurements at 10% deformation or at the compressive yield point, whichever occurred first to compare results on an equivalent basis. Results are presented in Table 12. All baseline WAXFIX and Carbray 100 Epoxy specimens failed prior to 10% deformation except the Carbray 100 Epoxy/INEL soil composites. Note that one replicate was measured beyond 10% deformation. A compressive yield strength of 0.69 MPa (100 psi) was achieved with a specimen deformation of 35%. The standardized waste stream encapsulated in Carbray 100 Epoxy exhibited an extremely low value of 0.046 MPa (6.6 psi). TECT 1 encapsulations results were significantly higher. The WAXFIX/waste compressive strengths were moderate. All grout/INEL soil composite compressive strength values were higher than their grout/waste stream counterparts.

Table 12. Unconfined compressive strength for grout/waste and soil composites.

Grout Composite Baseline Compressive Strength		
Grout Type	*Compressive Strength, MPa* (psi)[a]	
	Waste Stream	**INEL Soil**
TECT 1	9.8 ± 2.0 (1424 ± 296)	20.5 ± 2.8 (2966 ± 413)
WAXFIX	0.73 ± 0.03 (106 ± 3.7)	1.8 ± 0.2 (263 ± 22.1)
Carbray 100 Epoxy	0.046 ± 0.008 (6.6 ± 1.2)	0.33 ± 0.03 (48 ± 3.3)
Phosphate Cement	n/a	8.3 ± 0.9 (1204 ± 132)

n/a = not applicable

(a) For consideration, the NRC Technical Position on Waste Form requires a minimum compressive strength for all solidification agents: 0.41 MPa (60 psi) and 3.45 MPa (500 psi) for hydraulic cements.

4.3 Wet-Dry Cycling

The transuranic pits and trenches at the Subsurface Disposal Area at the INEL's Radioactive Waste Management Complex is located in a semi-arid region. The soil moisture content ranges on average between 15 - 20% by mass. However, both lower and higher values have been measured. Occasionally, precipitation percolating into the ground will recharge the aquifer. As a consequence, there will be times when these grout composites will be exposed to moist or saturated conditions. Since wet-dry cycling can have severe impact on construction materials such as hydraulic cements, this test was performed to simulate these intermittent conditions and to evaluate grout product resistivity under these wet/dry cycling circumstances. Five replicate specimens with the following grout compositions were tested: TECT 1/Waste, Carbray 100 Epoxy/Waste, and INEL soil specimens mixed and cured in Phosphate Cement, TECT 1 and Carbray 100 Epoxy grouts. The ASTM standard D-4843, "Wetting and Drying Test of Solid Wastes," was followed.[11] The test involved cycling the specimens 12 times, from 60°C dry to 20°C wet. Although the ASTM test requires evaporation of the water after each wet cycle and weighing of the residue left in the beaker, these samples were wet cycled in the same beaker after each cycle while reusing the same water (additional water was added to replace the loses due to evaporation). At the end of the final cycle, the water was evaporated and the residue weighed. The wet samples were towel dried and weighed after each cycle. The dry cycle samples were weighed following a one hour cool-down period. Additionally, the mechanical integrity of each specimen was determined after the final cycle by compression testing using ASTM C-39 or ASTM D-695. WAXFIX/waste and WAXFIX/INEL soil specimens were wet-dry cycle tested initially using ASTM D-4843 but, as suspected, the test needed to be terminated because the dry cycle (60°C) temperature was just marginally at the lower melt limit for this WAXFIX grout material. After the termination of the test, the glass containers used for both cycles were placed in an oven at 60°C to evaporate the immersion liquid and dry any materials that chipped, flaked, dissolved, or leached from the specimen during testing. The dried residue was weighed and compared to the original specimen mass (Table 13). Table 13 summarizes and compares the compressive strength results obtained for baseline and wet-dry cycled specimen.

During the 12 wet-dry cycles, each of the five grout composites exhibited a variety of physical changes. For example, TECT 1/waste specimens revealed top/side surface hairline cracking following the first dry cycle. This hairline cracking expanded quickly to the entire lateral surface and, to a lesser degree, the end surfaces after the completion of two wet-dry cycles. Hairline cracking continued to progress throughout the test as the number, depth, and width of cracking increased, but never to excess (i.e, cracking never progress to the stage where the specimen split or crumbled).

The cracking appeared to stabilize after 8 cycles were completed. Figure 4 (specimen on right side of figure) shows the type and the extent of cracking associated with the TECT 1/waste specimens following 12 wet-dry cycles and modest specimen dissolution and/or matrix leaching was indicated by the color of the immersion solution (distilled water) at the completion of the test. The distilled water turned a bright yellow color following the first wet cycle and turned progressively a deeper yellow (amber) color by the termination of the test. The resultant dried residue accounted for 2.9% of the initial mean specimen mass (Table 13). A mean specimen volume decrease of 1% was

Table 13. Unconfined compressive strength of baseline and wet-dry cycled grout composites.

Grout Material/ Waste Type	Compressive Strength, MPa [a]		Residue (% of Initial Wt.) [e]
	Baseline	Wet-Dry Cycle	
TECT 1/Waste	9.8 ± 2.0	12.6 ± 1.9	2.9
TECT 1/INEL Soil	20.5 ± 2.8	13.5 ± 2.7	0.1
Carbray/Waste	0.046 ± 0.008	0.16 ± 0.03	14.1
Carbray/INEL Soil [b]	0.33 ± 0.03	0.96 ± 0.06	0.5
Phosphate Cement/INEL Soil [c]	8.3 ± 0.9	3.0 ± 1.2	0.5
Portland Type II [d] Cement Concrete	45.2 ± 13.7	36.1 ± 6.5	n/a

n/a = not applicable

(a) Results based on 5 replicates and 2 sigma errors.

(b) Compressive strength measured at 10% deformation.

(c) Results based on 4 replicates and 2 sigma errors.

(d) These data were taken from reference [12]

(e) These values represent a mean residue that remained in the glassware following 12 wet-dry cycles. Test glassware was placed in an oven at 60°C for a minimum of 48 hours to dry residue.

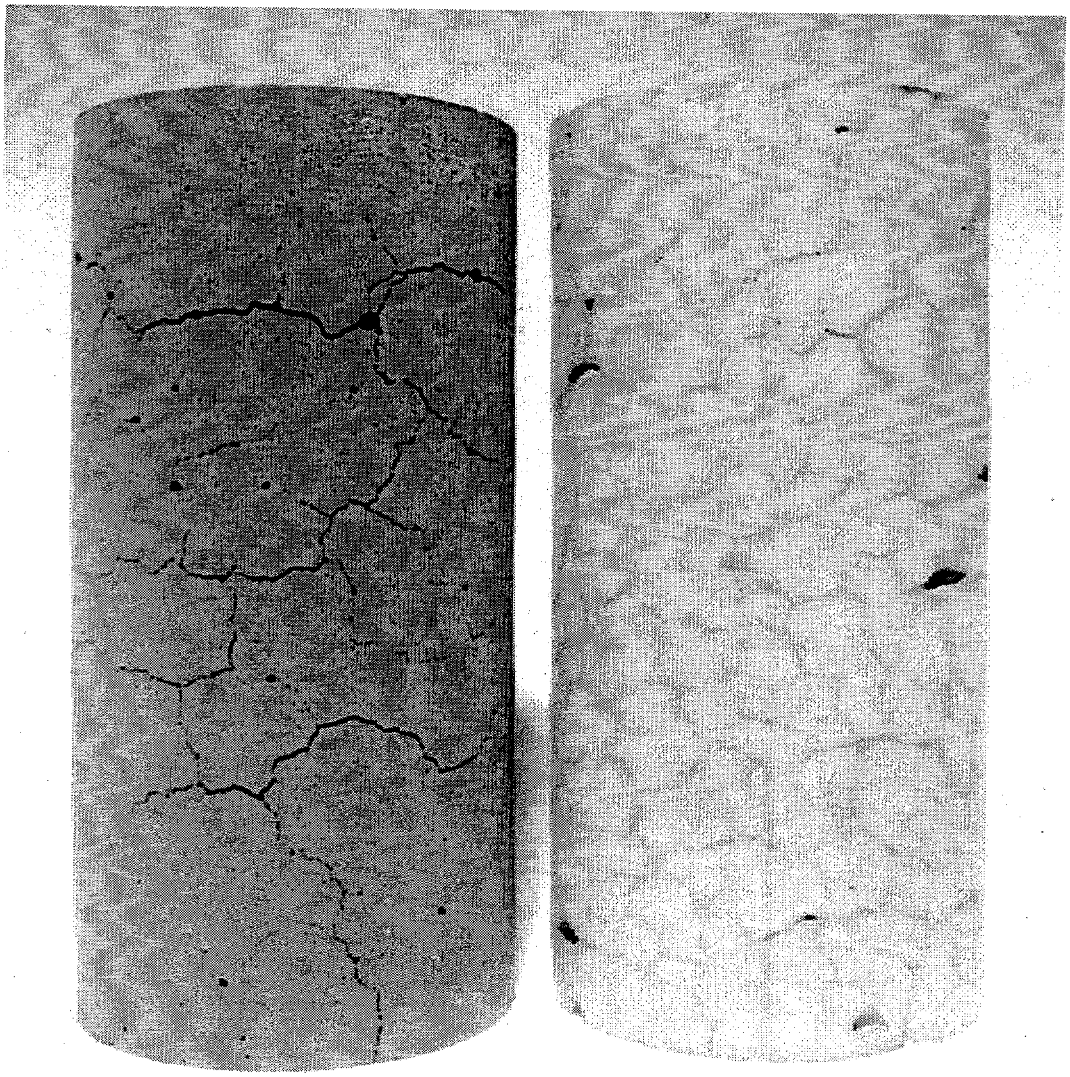


Figure 4. TECT 1/waste (right) and TECT 1/INEL soil (left) composite cracking following 12 wet-dry cycles.

also calculated. However, this slight compositional loss in addition to the moderate hairline cracking did not appear to effect the physical integrity of the specimen adversely since the grout/waste product remained a free standing monolith throughout the entire test. Table 12 shows that the mean specimen compressive strength following wet-dry cycle testing actually increased by nearly 30% (12.6 ± 1.9 MPa) when compared to baseline values (9.8 ± 2.0 MPa). This strength improvement may be attributed to additional curing during the dry cycle (60°C).

The Carbray 100 Epoxy/waste results following 12 wet-dry cycles were somewhat different. Prior to testing, these specimens were characterized as being moderately stiff (surface could be depressed with fingers) and possessed a crust-like layer which was produced during ambient air curing (16 days). The surface of each grout form began to soften following the first wet cycle. However, the specimen rehardened (i.e., surface crusting) during the next dry cycle due to the elevated oven temperature (60°C). This softening/rehardening of the exterior layer of the grout forms was evident following each wet/dry cycle. Note that after only one wet cycle the immersion solution turned opaque/dark brown, and the specimen color was now tan (original color - brown). The sample was dried with a paper towel before weighing after each wet cycle. The paper towel was observed to be heavily stained with a brown residue following specimen drying. The top and bottom surfaces of these Carbray 100 Epoxy/waste forms were cracking by the fourth dry cycle. These cracks increased in size and depth with each successive cycle. Each specimen showed varying degrees of cracking at the completion of the wet-dry test, with the major cracking appearing to be confined to the end surfaces. However, cracking was detected to penetrate the central portion of 3 of 5 Carbray epoxy/waste forms, with the use of a long, thin metal probe. The depth of the obscured internal cracking was difficult to detect by visual inspection alone. The length, width, and penetrating depth of the top/bottom surface cracks measured, at their worse, 3.75 cm, 0.25 cm, and 8 cm, respectively. Apparently, as the end surface cracks widened, so did the length of the inconspicuous central interior cracks. While the exterior of the specimens appeared to show reasonable physical integrity, the interior was deteriorating and eroding with each successive cycle. Figure 5 shows an end view of the crack development of a Carbray 100 Epoxy/waste specimen following test completion.

Another potentially destructive property of these grout composites is significant shrinkage. The mean specimen volume decreased was 6.8% (Note, however, the total sample volume decreased from mold cure to the end of the wet-dry cycle test was >18.5%). Figure 6 shows this specimen contraction following 12 wet-dry cycles (left) in comparison to a baseline Carbray 100 Epoxy/waste specimen (right). Note also, in Figure 6, the narrow vertical crack on the lateral surface of the waste form. This was the only specimen tested that exhibited this lateral surface cracking visibly. Furthermore, note that the original opaque/dark brown immersion solution color never changed through the remaining of the test. However, the consistency appeared to thicken slightly with each successive cycle. At the completion of the test, the wet and dry cycle glass containers were placed in an oven at 60°C to evaporate the liquid and to measure the dried residue losses. Table 13 reveals that 14.1% of the specimen mass (~45 g from a ~300 g total) was lost due to dissolution/leaching and/or matrix evaporation, with the majority of the compositional losses occurring during the wet cycle. Figure 7 shows the dried Carbray 100 Epoxy/waste residue product following wet-dry cycling. Note, the crystals that developed on the glass beaker walls during residue drying. Curiously, with all these apparent destructive processes occurring, the compressive strength of these wet-dry cycled specimens increased ~3.5 times (0.16 ± 0.03 MPa), in comparison to their baseline counterparts. This strength increase may be attributable to continued curing of the epoxy. It was apparent that the epoxy samples slowly "hardened" while air curing. The elevated temperatures may have accelerated slow curing or the curing may simply be a long slow process. The strength may continue to increase for long periods.

The TECT 1/INEL soil specimens subjected to 12 wet-dry cycles behaved in a similar manner to their TECT 1 /waste counterparts. The lateral surface of the specimens were covered with hairline cracks after the first dry cycle. This surface hairline cracking increased with each successive wet-dry cycle. Cracking on both the end surfaces was observed by the fourth cycle. The number of cracks (and cracking pattern) observed were numerically less in comparison to the TECT 1/waste crack development. However, their size appeared to be wider (~ 0.1 mm) and deeper (~ 0.1 mm). An illustration of the distinct pattern and cracking differences associated with each of the TECT 1 grout specimens (waste versus INEL soil encapsulation) are presented in Figure 4. Although these TECT 1/INEL soil composites exhibited extensive surface cracking, the specimens still remained free standing monoliths with only minor mass losses due to dissolution, chipping and flaking (See Table 13 - dried residue measured in test glassware ~ 0.1%). The distilled water immersion liquid used during the wet cycle turned a clear, light brown color after the first wet cycle. The final solution color was clear and bright yellow. It should be noted that some grout composite bubbling occurred each time the specimens were placed in the wet cycle distilled water solution, producing a thin white foam, which initially formed on the solution surface, but dissipated with time. No significant dimensional changes were measured for these composites. However, a slight volume decrease was measured (<1.0%). Table 12 shows that the mean wet-dry compressive strength of TECT 1/INEL soil specimens (13.5 ± 2.7 MPa) decreased by approximately one-third in comparison to mean baseline values (20.5 ± 2.8 MPa). This is opposite to the result that occurred with the TECT 1/waste forms, which exhibited an increase in compressive strength. It is possible that component(s) of the soil matrix interacted with (e.g., hydrate), and/or was partially removed by, the distilled water causing the compressive strength weakness measured with these specimens.

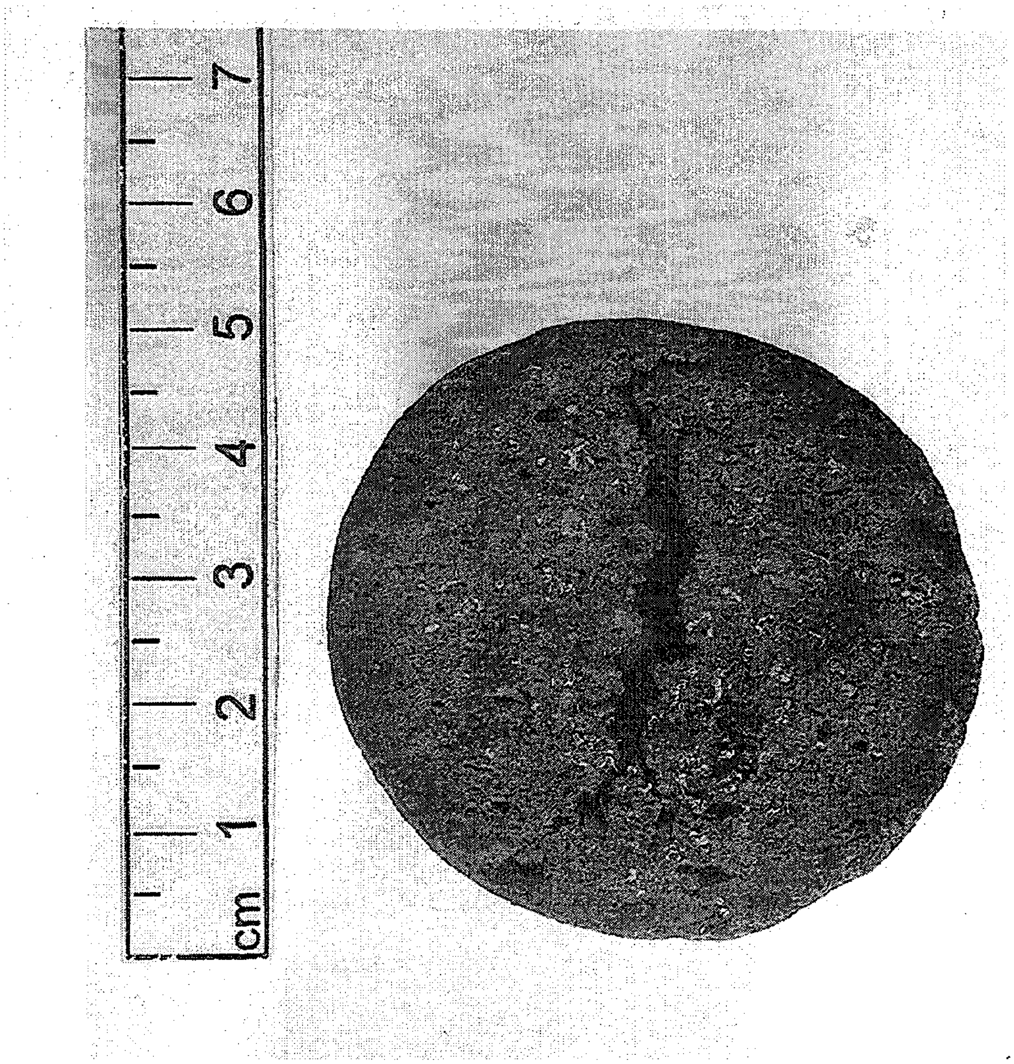


Figure 5. Upper surface cracking of a Carbray 100 Epoxy/waste specimen following 12 wet-dry cycles.

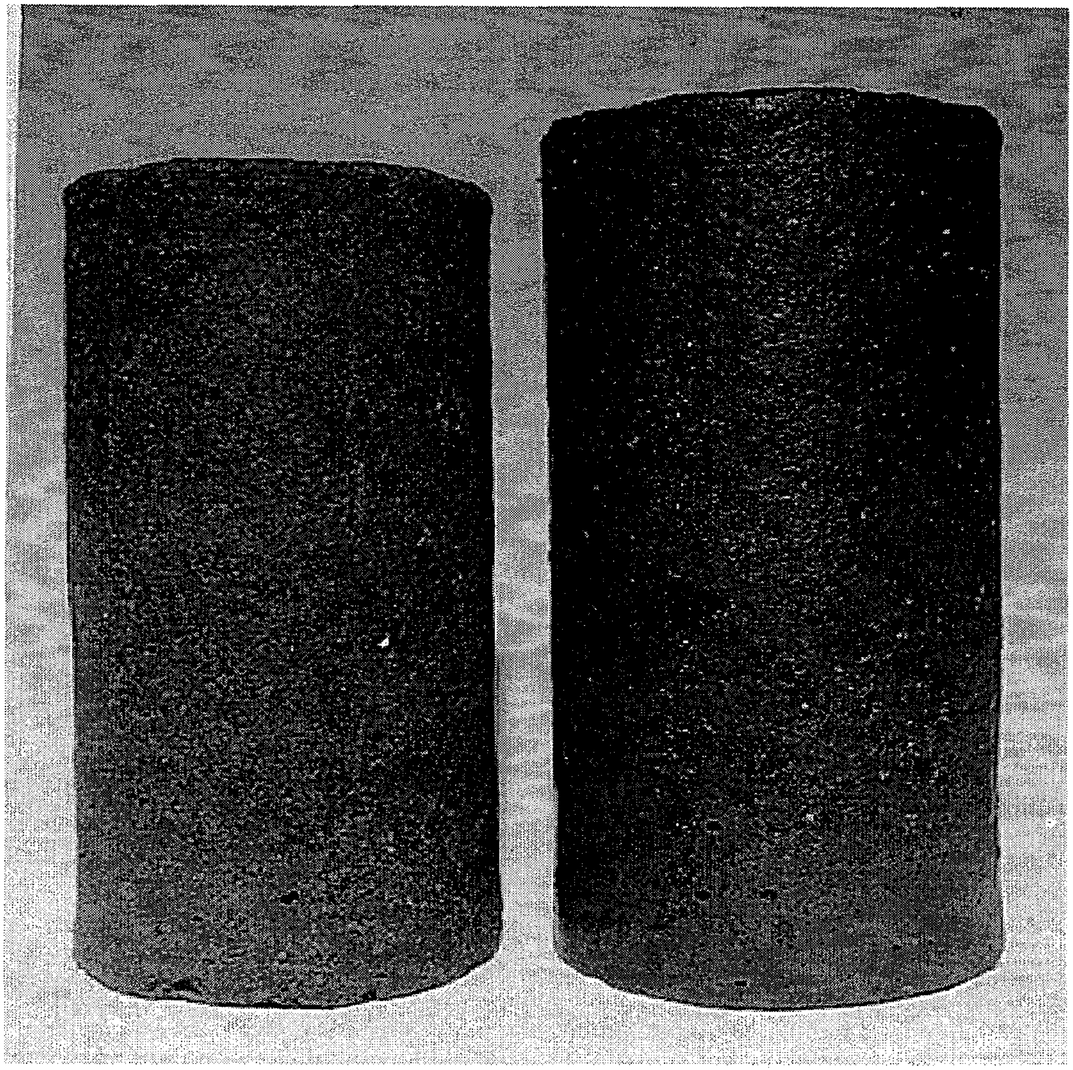


Figure 6. A comparison between a Carbray 100 Epoxy/waste specimen before (right) and after (left) wet-dry testing.

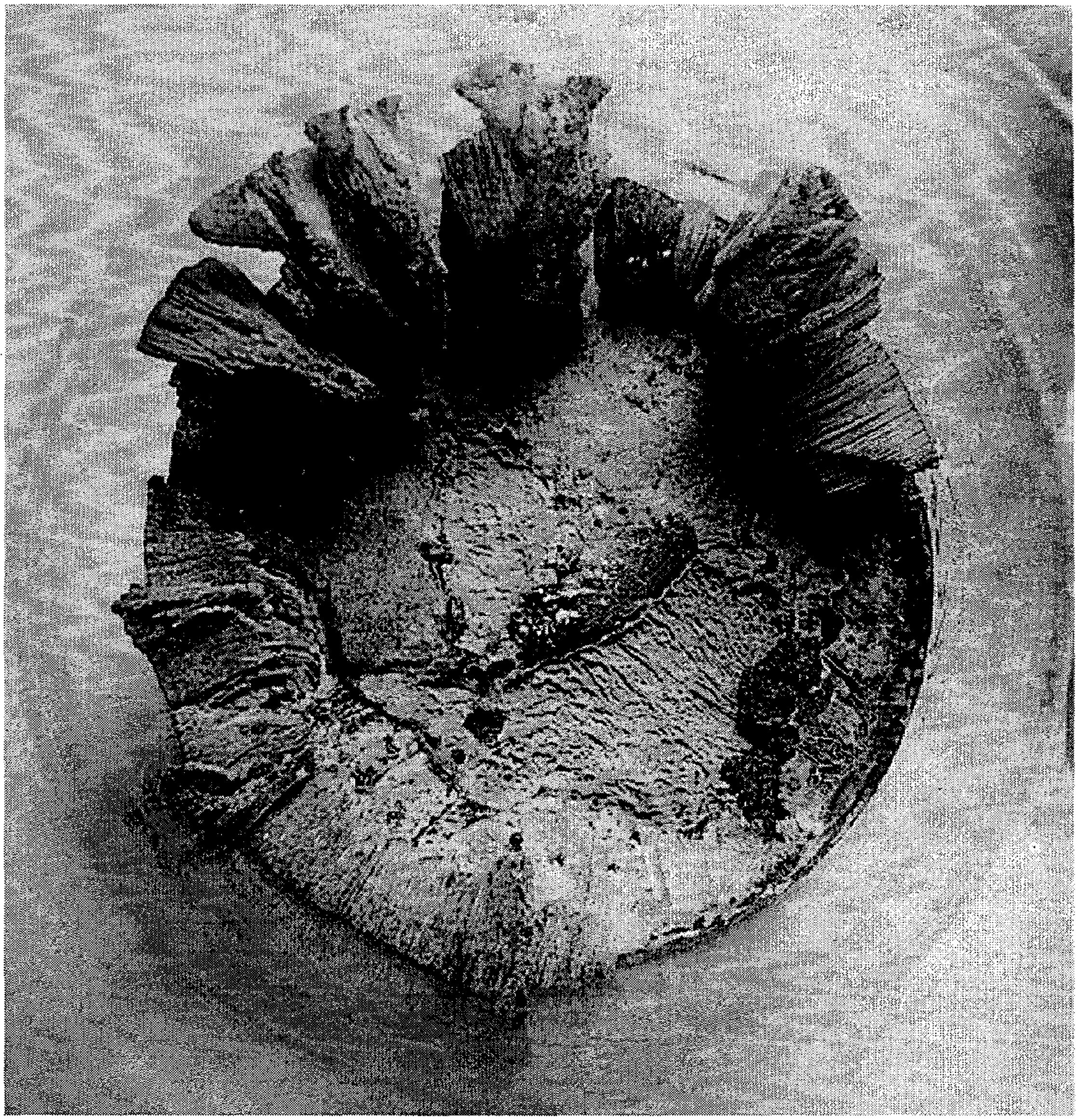


Figure 7. Oven dried Carbray 100 Epoxy/waste residue following 12 wet-dry cycles.

Baseline Carbray 100 Epoxy/INEL soil specimens were characterized as monoliths possessing reasonable hardness (could not be compressed using finger pressure) prior to wet-dry cycle testing. The original specimen firmness did not change throughout the entire wet-dry cycle test period. The specimen size and shape did change although no grout composite cracking occurred. The specimens were observed to possess a slight barrel-like shape (specimen diameter was greater in the middle and thinner at either end) by the fourth cycle. A 6% mean specimen volume decrease was measured by the completion of the wet-dry cycle test (Note, the total sample volume decrease from mold cure to the end of the wet-dry cycle test was ~13%). Furthermore, the specimen lost an average of 5% (17 g)of their original mass from the time the specimens had cured in the molds (18 days) to the time the samples were removed from the molds and ambient air dried for an additional 12 days (total cure time - 30 days). These were similar to the results measured for the specimens used in baseline compressive strength. Figure 8 shows a Carbray 100 Epoxy/INEL soil baseline composite (left) and a Carbray 100 Epoxy/INEL soil (right) after 12 wet-dry cycles. Note the shape and surface texture difference associated with the wet-dry tested specimen. In addition, a difference in the color was observed. The original baseline composite color of dark brown changed to grey- brown following 12 wet-dry cycles. The distilled water immersion solution changed to a deep yellow color after the first wet cycle. A clear, amber color characterized the solution color following the second wet cycle and was maintained until the completion of the test. This immersion solution color was significantly different than that observed with the Carbray 100 Epoxy/waste (dark brown and opaque). The clear-yellow immersion solution associated with the Carbray 100 Epoxy/INEL soil composites revealed minor dissolution/leaching. However, a very small percentage (0.5%) of dried residue was measured in the glassware following the test. Note, the elevated temperatures available during the dry cycle (60°C) were also beneficial in increasing the compressive strength of these Carbray 100 Epoxy/INEL soil composites by almost three times. This is clearly evident when mean baseline values (0.33 ± 0.03 MPa) are compared to specimens compression tested following 12 wet-dry cycles (0.96 ± 0.06 MPa). This result can be attributed to additional curing during the dry cycle. Also note that all compressive strength data for Carbray 100 Epoxy/INEL soil specimens and baseline counterparts were measured at 10% deformation.

Phosphate cement/INEL soil specimens completed the wet-dry cycle testing revealing some minor composite degradation. Specimen deterioration was observed following dry cycle number four and occurred first in the form of hairline cracks located at the upper/lateral surface interface. The cracking was observed occurring at the bottom/lateral surface a few cycles later. The exterior hairline surface cracking progressed with each successive cycle and appeared to migrate from both ends towards the center of the specimen. Figure 9 shows the extent of upper surface hairline cracking of a Phosphate cement/INEL soil specimen following the completion of 12 wet-dry cycles. A comparison between a control specimen (right) and a side view of a specimen that had completed the wet-dry cycle testing (left) is shown in Figure 10. Very little flaking or chipping was observed with these ceramic composites. In addition, no composite dimensional changes were measured following 12 wet-dry cycles. All composites would bubble fairly significantly until all the void space was filled each specimen's introduction to the wet cycle immersion solution. Also note, the sample turned a cement grey color when immersed, but returned to it's original off-white color during the dry cycle. The immersion solution color at the termination of the test had a slight brown tint, thus indicating that only minor compositional losses due to dissolution/leaching occurred during the wet-dry cycle testing. This assumption was substantiated by the drying and quantifying of the residue remaining in the glassware following the test. These measurements revealed that only 0.5% of the original specimen mass was dissolved, leached, or removed during the test (see Table 13). Specimens lost two thirds of their baseline compressive strength (Baseline - 8.3 ± 0.9 MPa versus Wet-Dry Cycle - 3.0 ± 1.2 MPa)despite the relatively benign appearance of these wet-dry tested composites. Note, the statistical variability associated with the wet-dry cycled Phosphate cement compressive strengths.

Figure 11 presents each of the grout/waste and INEL soil compressive strengths as percent of baseline strength.

Grout/waste forms were subjected to 60°C oven temperatures during the dry cycle and to room temperature distilled water saturation conditions during the wet cycle. The ideal test result would have included minimal weight gains/losses and no volume changes. Table 14 summarizes the mean specimen mass gains or losses measured during the first and last wet-dry cycles. Initial composite weight losses after the first dry cycle measured between 1.3 to 6.7%. It would appear based on the results that each grout/waste product lost mass by component volatilization, moisture loss, dissolution or some combination. In addition, the results showed that the greatest percentage losses after the first cycle of 4.0% and 6.7% were associated with the TECT 1/Waste and the TECT 1/INEL soil specimens, respectively. Mass losses for Carbray 100 Epoxy/waste and INEL soil composites were both <1.5%. However, specimen measurements following 12 dry cycles showed considerably different results. Mass losses dramatically increased for both the TECT 1/waste (12.5%)and Carbray 100 Epoxy waste (20%). Apparently, these enhanced grout/waste form compositional losses were the result of significant leaching/dissolution during the wet cycle phase of the test. Wet cycle results were variable. The majority of the initial wet cycle mass values were slightly lower than baseline values, except for Carbray 100 Epoxy/ INEL soil (+1%) and Phosphate cement/INEL soil specimens (+11.7%).

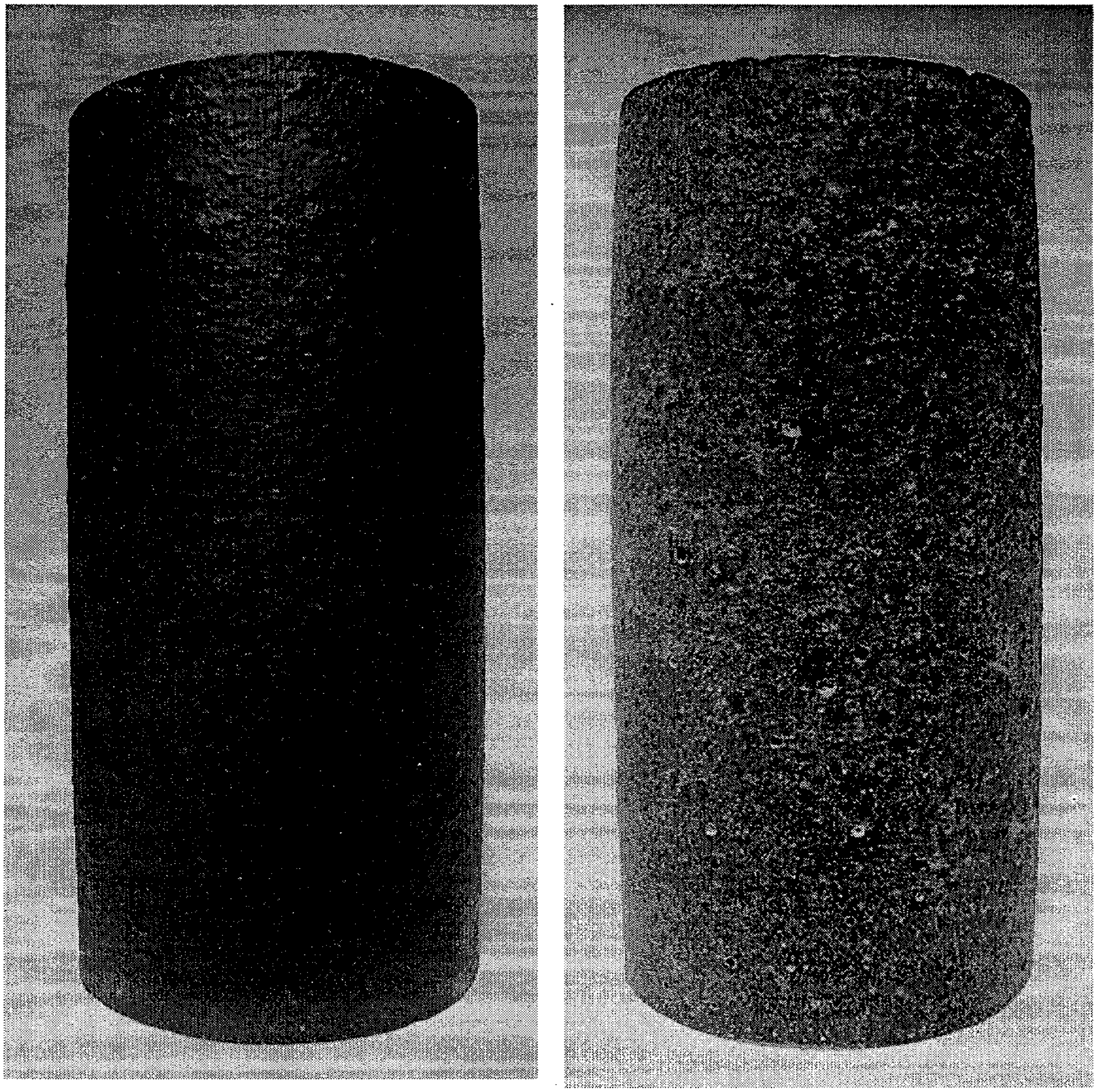


Figure 8. A Carbray 100 Epoxy/INEL soil composite before (baseline sample left) and after wet-dry cycle testing (right).

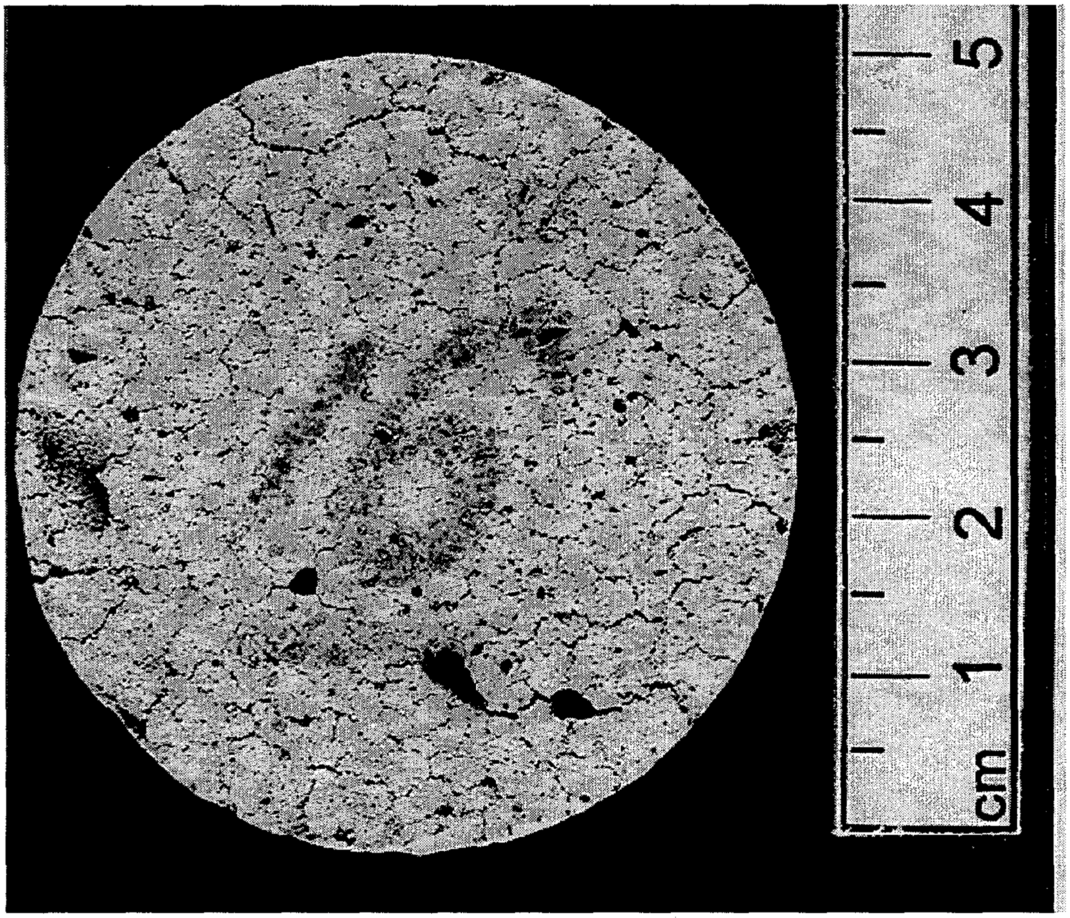


Figure 9. Top view showing the surface hairline cracking of a Phosphate cement/INEL soil specimen following the completion of 12 wet-dry cycles.

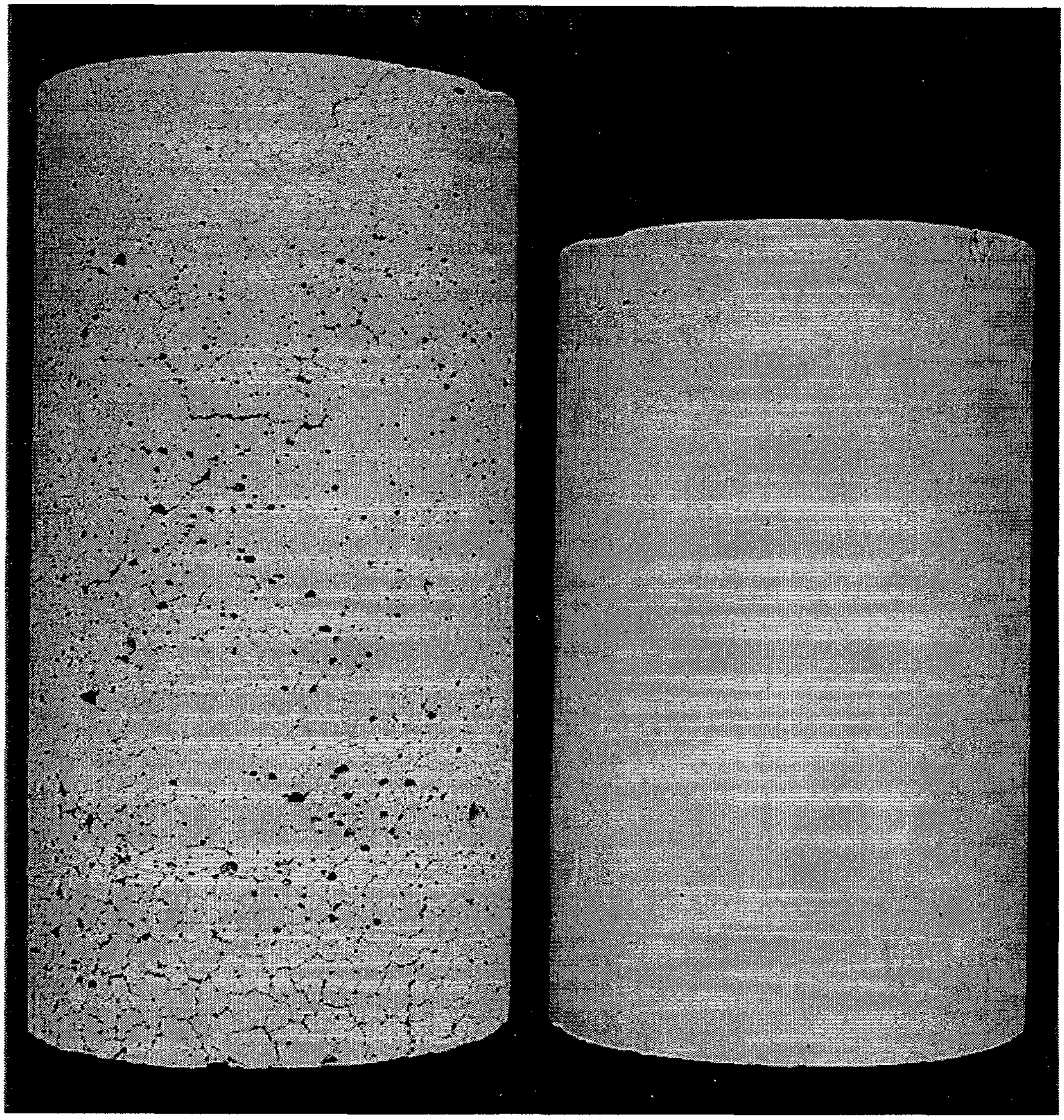


Figure 10. Lateral view of a Phosphate cement/INEL soil specimen showing hairline cracking following the completion of 12 wet-dry cycles.

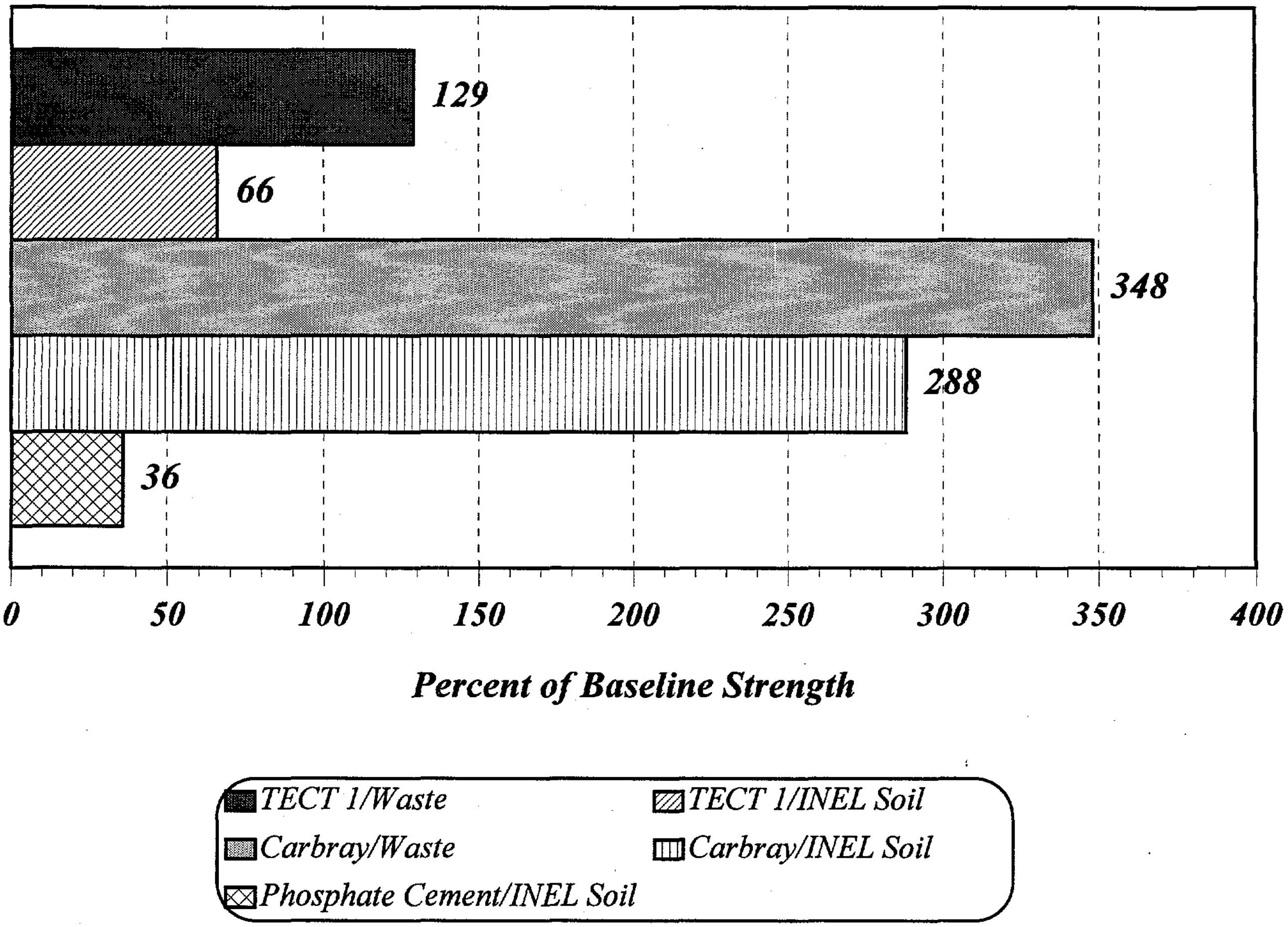


Figure 11. Effect of 12 wet-dry cycles on the compressive strength of grout/waste and INEL soil composites.

Table 14. Specimen weight losses and gains measured after the first and last wet and dry cycles.

Percent Grout Composite Weight Gains & Losses Measured During the Wet-Dry Cycle Test				
Grout Composite	**% Mean Dry Cycle Loss/Gain**		**% Mean Wet Cycle Loss/Gain**	
	Initial	**Last**	**Initial**	**Last**
TECT 1/Waste	**-4.0**	**-12.5**	**-2.3**	**-9.9**
TECT 1/INEL Soil	**-6.7**	**-6.8**	**-0.5**	**-0.4**
Carbray/Waste	**-1.5**	**-20.0**	**-1.6**	**-15.3**
Carbray/INEL Soil	**-1.3**	**-4.8**	**1.0**	**-4.1**
Phosphate Cement/INEL Soil	**-2.1**	**-2.8**	**11.7**	**10.9**

The average weight histories for TECT 1/waste and Carbray 100 Epoxy/waste are presented in Figure 12. Weight changes for both these composites were fairly significant, with considerable mass losses evident in the first three cycles. A downward trend continued throughout the remainder of the test, but the decline was less dramatic and more gradual. The mean specimen mass loss for TECT 1/waste and Carbray 100 Epoxy/waste composites was 12.5 wt% and 20.0 wt%, respectively. These significant composite mass losses were the result of several factors including grout/waste dehydration, dissolution/leaching, and volatilization. The magnitude of the fluctuations between the wet and dry cycles was between 3.5 - 5.0% for the Carbray 100 Epoxy/waste. The TECT 1/waste variations were slightly less at 2.5 - 3.0%, but still relatively high. In the majority of cases, these negative properties (e.g., specimen shrinkage, loss of weight and cracking) would have a deleterious effect on compressive strength normally. However, results cited earlier reveal that just the opposite occurred.

Figure 13 shows the mean wet-dry cycle weight histories for composites prepared with INEL soil and encapsulated in TECT 1, Carbray 100 Epoxy, and Phosphate cement grout materials. The soil encapsulation results were not only variable, but different from their encapsulated waste stream counterparts. The Phosphate cement/INEL soil composites showed the most dramatic fluctuations between cycles. Furthermore, the major changes did not occur during the dry cycle (mean weight decrease was 2.6%), but during the wet cycle (mean weight increase 11.3%). It appears, these particular Phosphate cement/INEL soil composites possessed considerable porosity that accommodated rapid infiltration by low viscosity liquids. The weight loss/gain fluctuations between cycles was fairly reproducible between cycles, with only a slight downward bias through 12 wet-dry

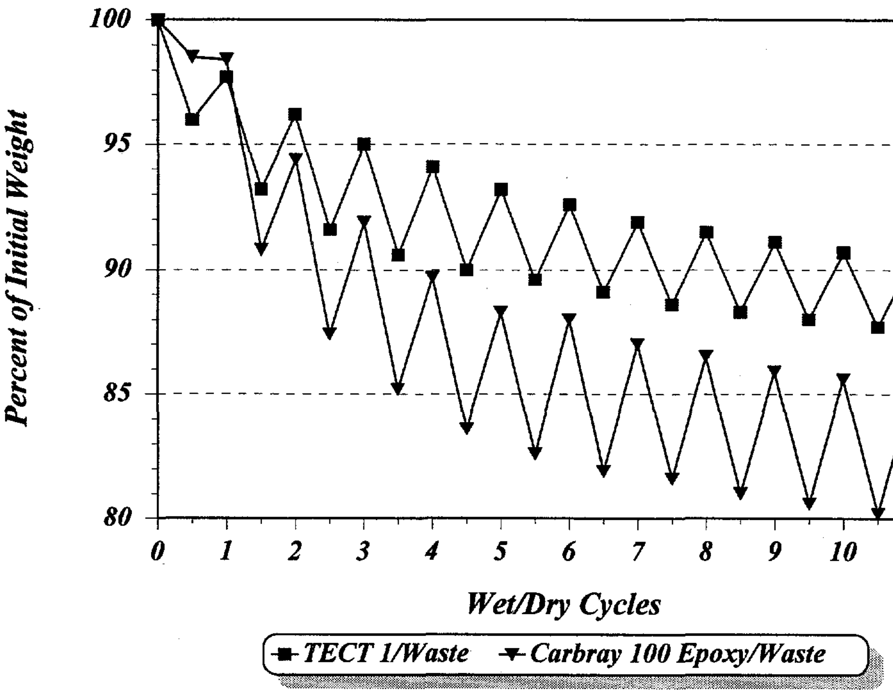


Figure 12. The mean weight changes of TECT 1/waste and Carbray 100 Epoxy/waste composites during wet-dry cycling.

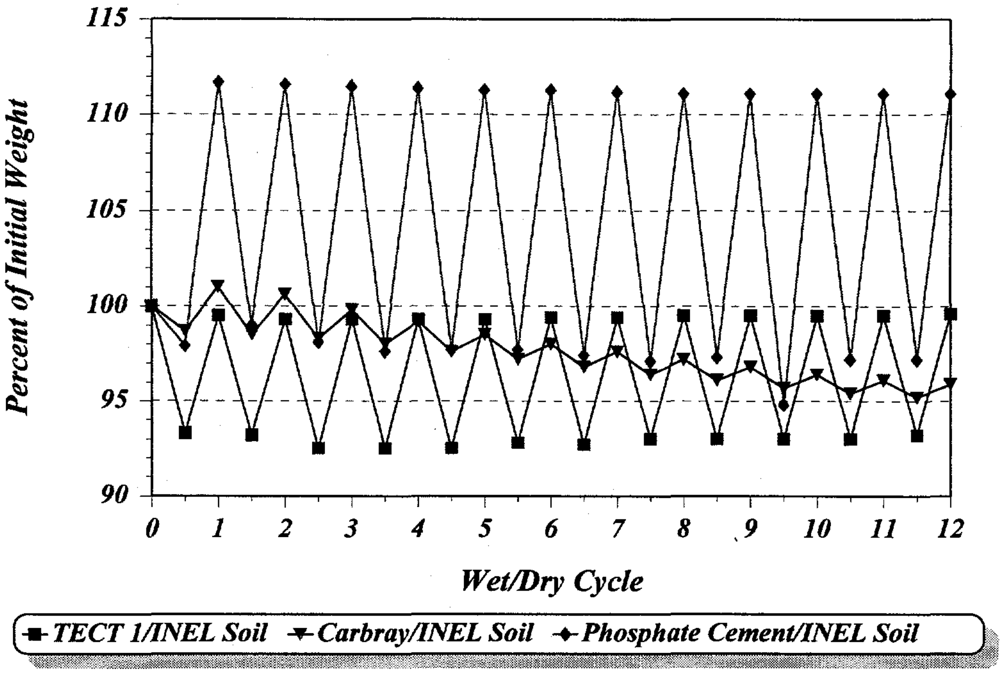


Figure 13. The mean weight changes for INEL soil encapsulated in TECT 1, Carbray 100 Epoxy, and Phosphate cement composites during wet-dry cycling.

cycles. This result could have been attributed to the small compositional losses measured at the completion of the test (dried glassware residue: 0.5%). The magnitude of wet-dry cycle fluctuations for TECT 1/INEL soil was less than that measured for the Phosphate cement/INEL soil specimens. Here also, the weight loss/gain fluctuations remained relatively constant throughout the entire test. The specimens nearly returned to their original pre-test weight after each dry cycle. The mean maximum dry and wet cycle weight losses recorded were 7.5 % (3rd cycle) and 0.7% (2nd cycle), respectively. Carbray 100 Epoxy/INEL soil composites performed quite differently than the Carbray 100 Epoxy/waste specimens during wet-dry cycle testing. The magnitude of the 12 wet-dry cycle weight fluctuations for these specimens was the smallest recorded (Mean value: 1.1%). Furthermore, the mean magnitude of these fluctuations diminished with each successive cycle completion starting at a high of 2.3% (1st cycle) and finishing the test at 0.7% (12th cycle). The overall specimen weight change for Carbray 100 Epoxy/INEL soil drifted slightly downward from the original starting weight (see Figure 13). This result was due also to evaporative losses during the drying cycle and minor compositional losses (0.5 wt%) measured in the glassware at the termination of testing. The maximum specimen weight loss and gain due to water evaporation/absorption (also includes material losses from dissolution/leaching) was 4.8% (12th dry cycle) and 1.0% (1st wet cycle), respectively.

Figure 14 presents a composite of all the wet-dry cycling results for TECT 1 and Carbray 100 Epoxy specimens prepared using the standardized waste formulation and INEL soil as the waste stream. Note that the standardized waste stream encapsulated in both the TECT 1 and Carbray 100 Epoxy were trending downward substantially, while soil encapsulated specimens appeared to be reasonably stable (TECT 1 more so than the Carbray 100 epoxy). This downward trend was indicative of considerable specimen compositional losses.

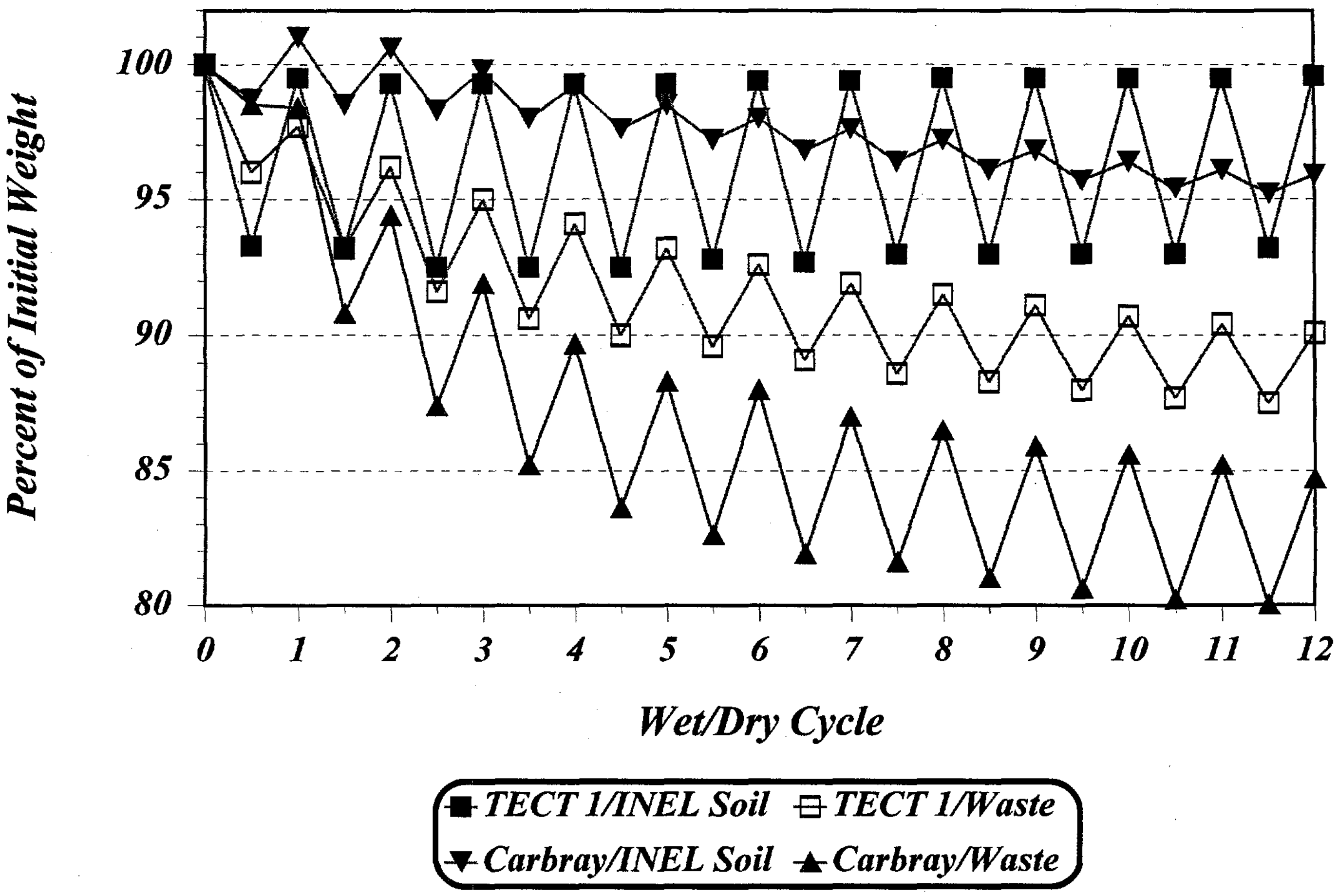


Figure 14. The mean weight changes for all TECT 1 and Carbray 100 Epoxy specimens during wet-dry cycling.

4.4 Base Resistance

Base resistance tests were performed on 15 replicate specimens that were prepared using the standardized waste formulation and encapsulated in TECT 1, WAXFIX, and Carbray 100 Epoxy grouts. These grout products were immersed in 3 liters of aqueous, sodium hydroxide solution at pH 12.5 for 90 days. Sodium hydroxide was chosen as representative of conditions within the DOE complex. The pH value is based upon the EPA characteristic corrosive for hazardous waste. The specimens were inspected visually on a daily basis during the 90 days for cracking, swelling, and/or spalling. The immersion solution pH was checked daily and adjusted back to 12.5 if it changed by more than 0.5 units. Every 30 days, five replicate samples were removed from the media, inspected, placed in a 100% humid environment for a minimum of 4 days to equilibrate, and subjected to compressive strength testing adhering to either ASTM C-39 or ASTM D-695.

The TECT 1/waste forms showed no visual evidence of cracking or dimensional changes after 30 days of base immersion. The sodium hydroxide solution was a clear, orange/yellow color and the pH measured 12.6 at the termination the test. This solution color is an indication that minor compositional losses due to product dissolution/leaching (mean specimen mass loss measured 0.1%) were occurring. However, this minor material loss did not affect the mechanical strength of the immersed TECT 1/waste forms (10.2 ± 0.3 MPa) when compared to the baseline samples (9.8 ± 2.0 MPa). The 60 day immersion performance of specimens in sodium hydroxide solution was similar to the 30 day. However, one of the composites seemed to show evidence of minor hairline cracking at the mid-section of the sample. The surface area involved was very small and the lines so fine that it was difficult to determine whether these cracks were an artifact that existed at the start of the test. There were no specimen dimensional changes, but a 0.45% specimen mass loss was measured. Figure 15 shows the white-brown mottled appearance that characterized the specimen surface following 60 days of base immersion. This very fine white precipitate is an indicator that some minor leaching was occurring. The final immersion solution color was a clear-amber (slightly different than the 30 day color) and the pH measured 12.77 (slightly higher than the 30 day pH). The compressive strength was very reproducible, with a mean measurement of 9.8 ± 1.5 MPa. Ninety day immersion results/observations for TECT 1 grout/waste composites were similar to those cited for 30 and 60 days for the most part. However, some minor physical changes did occur to the specimens. For example, specimen volume increased slightly by 0.5% and the mass decreased by 0.8%. The mass loss was indicative of material removal from the monolith via leaching, dissolution, or particle flaking. This was further confirmed by the evidence of a light tan, crystalline residue/precipitate on the top surface of the specimens following solution removal. The final color of the immersion solution was amber and the pH measured 12.7. The compressive strength was unaffected following 90 days of sodium hydroxide immersion (10.0 ± 0.6 MPa). Figure 16 presents the results as percent of baseline strength.

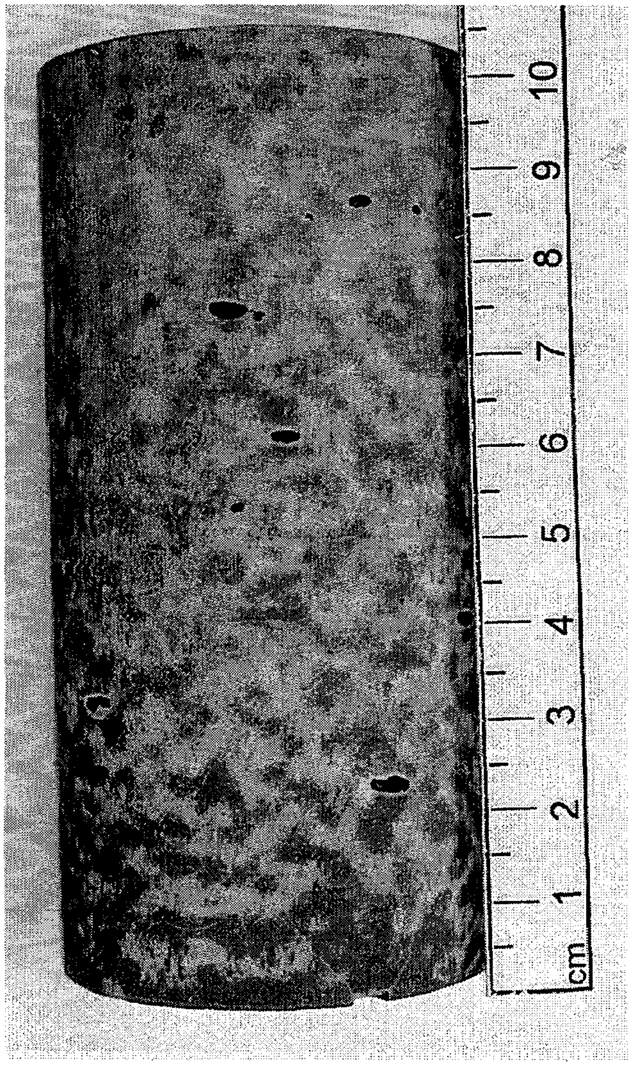


Figure 15. The white-brown mottled surface appearance that characterized the TECT 1/waste specimen following 60 days of base immersion.

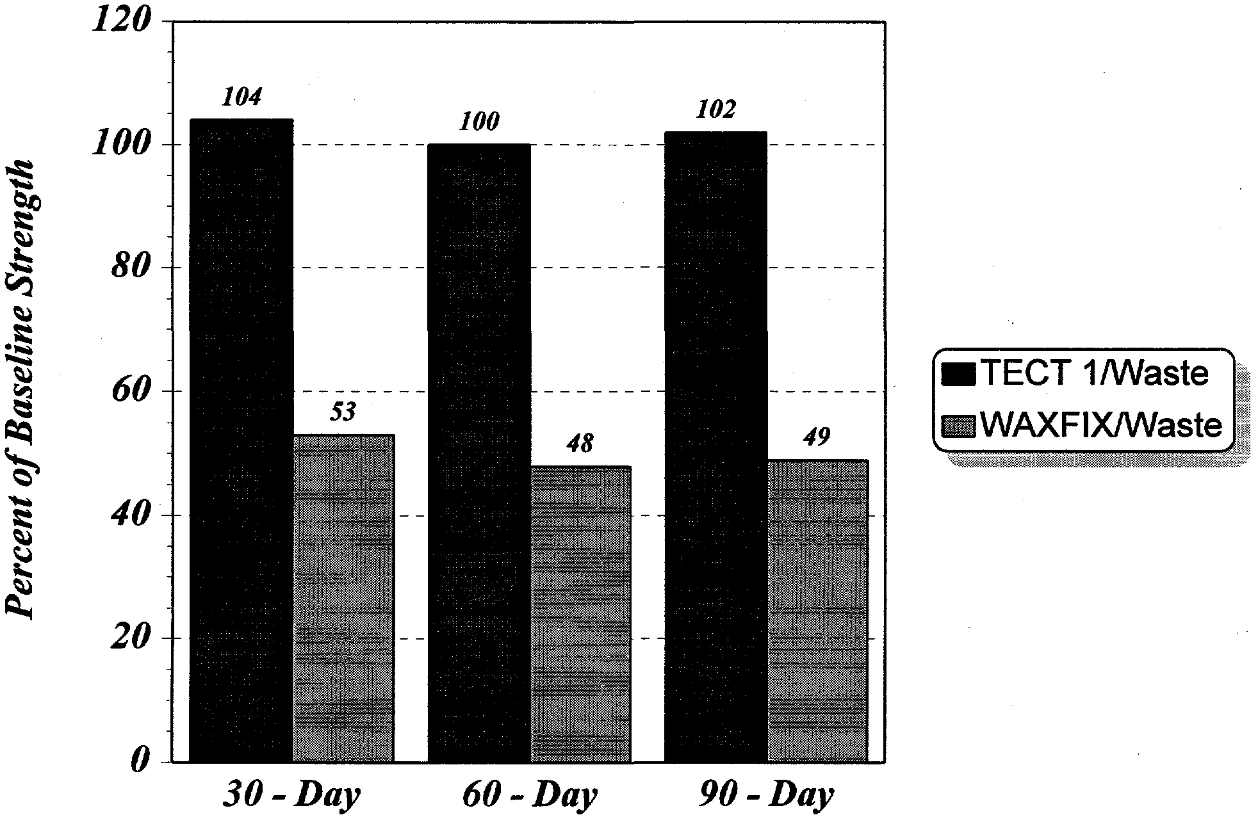


Figure 16. The effect of immersion in sodium hydroxide (pH = 12.5) on the compressive strength of TECT 1/waste and WAXFIX/waste.

WAXFIX/waste samples completed the 30 day base immersion performance testing as free standing monoliths, with no sample cracking evident. However, numerous surface precipitated formations (100 ± 50) ranging in diameter from fractions of a millimeter (small crystalline size) to 5 mm were observed. In some cases, these randomly distributed formations on the lateral surface of each WAXFIX/waste composite measured a maximum height of 0.1 mm. Irregular patterns (blotches) of light/dark shades of grey were quite conspicuous on the exterior surfaces of the composites at the end of 30 days of sodium hydroxide immersion, but not present at the start of the test. Figure 17 shows a portion of this grey discoloration as it wrapped around the waste form. The majority of the precipitated nodules were concentrated in the darker portion of the composite surface, although difficult to observe in a photograph. Note, however, that this precipitation was located on the entire surface, but to a lesser degree. In addition, there were several other indications that the physical integrity of WAXFIX/waste composites might have been deteriorating. For example, a mean specimen volume increase of ~2% was measured. This result was an indicator that the solution had penetrated the subsurface, promoting swelling of encapsulated waste materials. Sample degradation was further verified by the sodium hydroxide solution color changes that occurred as the test progressed. The base solution was a murky amber color by the test completion, thus clearly indicating compositional dissolution/leaching losses. The steady decrease in the immersion solution pH implied specimen decomposition also. The sodium hydroxide solution (pH - 12.5) needed to be replaced once because of dropping below pH - 12 within the 30 day test period. Furthermore, the pH at the completion of the test measured 12. These results suggest the loss of some acidic component within the matrix. The first replacement of 3 liters of base solution occurred after only five days of specimen immersion. The second exchange of base solution was not required until 24 days later (one day prior to the test termination). The degree of specimen degradation was realized during compressive strength measurements. The baseline strengths (0.73 ± 0.05 MPa) declined by nearly 50% (0.39 ± 0.03 MPa) following 30 days of base immersion. It should be noted that these composites were stored in a 100% humid environment for 31 days prior to testing. Specimens gain 0.25% weight during this storage period. The specimens lost a mean mass of 1.2% from the time that the grout composites were removed from humid storage and base immersed tested for 30 days. The four replicate composites immersed for 60 days in aqueous sodium hydroxide maintained their physical integrity throughout the test. The majority of the observations sited for 30 day composites was also representative of the specimens immersed for 60 days. No specimen cracking was observed. It did not appear that the precipitate formations increased in size or number. The two shades of grey surface discoloration did not appear to change either. One additional replacement of aqueous sodium hydroxide solution was necessary after 35 days of immersion because the pH declined below 12 (total replacements in 60 days - 2). These composites gained 0.2% moisture mass during the 30 day 100% humid storage period prior to immersion testing (similar to 30 day samples - 0.25%). The mean specimen volumes increased an average of ~3% from humid storage to completion of the 60 day base immersion test. The mass decreased ~2%. The specimen mass decreased an additional 0.6% during 7 days of post immersion 100% humid storage equilibration. However, the compressive strength (0.35 ± 0.06 MPa) did not change statistically when compared to the 30 day results (0.39 ± 0.03 MPa). The base immersion solution color deepened slightly to a dark amber color during the test. WAXFIX/waste forms maintained their physical integrity after 90 days of base immersion. The specimen surfaces were slimy to the touch. The development of random

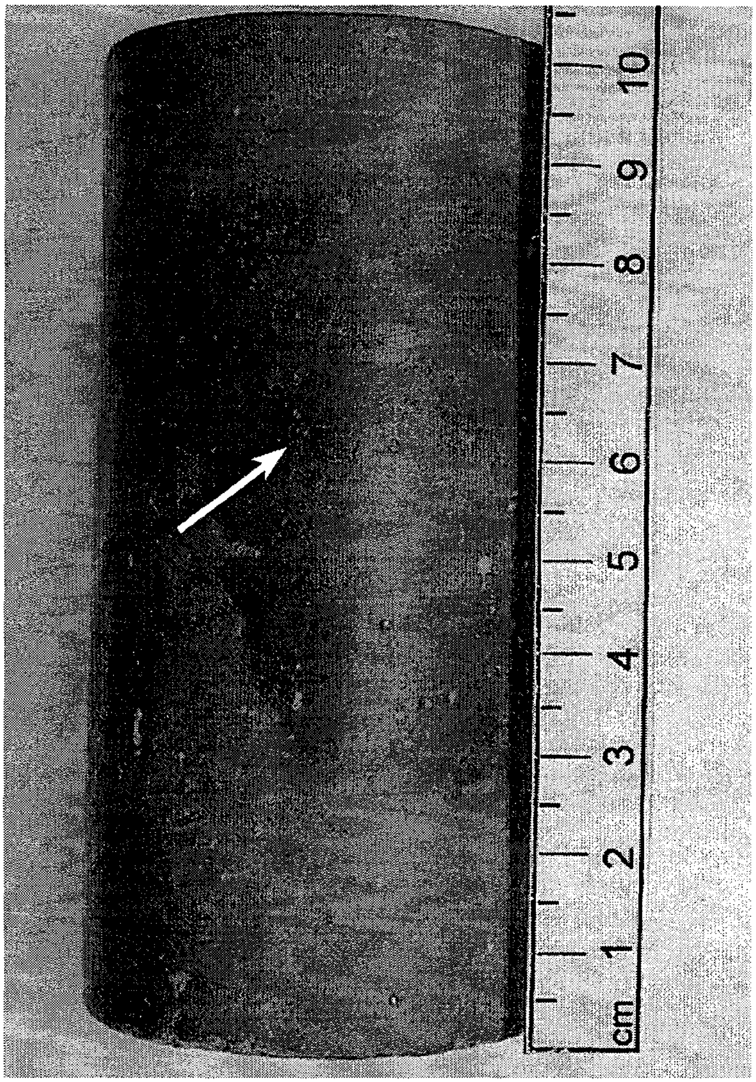


Figure 17. The typical light/dark shaded surface patterns characteristic of each WAXFIX/ waste composite following 30 days of sodium hydroxide immersion.

patches of a blue, green, and brown color formations were evident. This might have been the result of mold and/or bacteria growth. No specimen cracking was observed and the white crystalline precipitate was still present, but did appear to increase dramatically since 60 days. The specimen mass decreased by 1% from the start to the end of the 90 day immersion test and lost an additional 2.1% during 7 days of 100% humid storage equilibration prior to compression testing. Presumably, the mass reduction during the 7 days of storage was attributed to evaporation losses. A mean specimen volume increase of 5% was calculated at the completion of the 90 day base immersion testing, but was followed by 2.7% volume decrease after placement in 100% humid storage for seven days. The 12.5 pH sodium hydroxide solution needed to be replaced three times during the 90 day immersion period. The immersion solution had a murky yellow-green appearance by the end of the test. The apparent modest degradation of the grout/waste form did result in lowering the compressive strength after 90 days of base immersion (0.36 ± 0.07 MPa). Results suggested that specimen strength losses occurred only in the first 30 days of sodium hydroxide immersion. This is indicated in Figure 16 where measured compressive strengths are presented as percent of baseline strengths.

It should be noted, that WAXFIX/waste forms that were stored in 100% humidity (~60 days) and not used in performance/durability testing showed the development of a white, fluffy growth on the surface of the specimens. It is assumed that this is either a bacteria or mold growth. Figure 18 compares the end surface of a baseline sample (right) to that of a WAXFIX/waste form (left) that had been in 100% humid storage for more than 60 days. A view of the lateral surface (Figure 19) of the same WAXFIX/waste form (left) and the baseline sample (right) reveals a similar growth. After 100 days the white growth was observed to have enveloped the entire external grout/waste form surface. A similar growth was exhibited on the WAXFIX/INEL soil composite surface.

Carbray 100 Epoxy/waste composites not only failed the 30 day base immersion performance test, but were too soft and weak to be compressive strength tested. The inability of these grout forms to resist base immersion was observed immediately. The first specimen contact with the sodium hydroxide solution (pH - 12.5) resulted in the release of a medium brown color material which changed the immersion solution instantly to a yellowish-brown tint. Each specimen lost its original surface crustiness, became spongy to the touch, and turned a tannish color after 24 hours of base immersion. During the same time period, the immersion solution turned an opaque, dark brown color, and the pH measured ~12 (the original solution was decanted and replaced with 3 liters of fresh sodium hydroxide solution). However, the sample still maintained its physical integrity exhibiting no cracking or spalling. The immersion solution pH measured 11.75 three days later and was replaced. The Carbray 100 Epoxy/waste forms possessed the following characteristics by the tenth day of base immersion: extremely soft, swollen, weak tensile strength (could not support its own weight when lifted up), and tore easily if not carefully supported by hands. The solution was a dark brown, opaque color, and the pH decreased to ~12.25. Although the base immersion test was taken to completion of the first test period (30 days), it was determined that the specimens' physical integrity failed this particular performance test after approximately 15 days. The samples were extremely soft and fragile and could not be moved without damage, let alone be measured for compressive strength. The solution was never replaced after the 10th day and by the completion of the test the pH measured

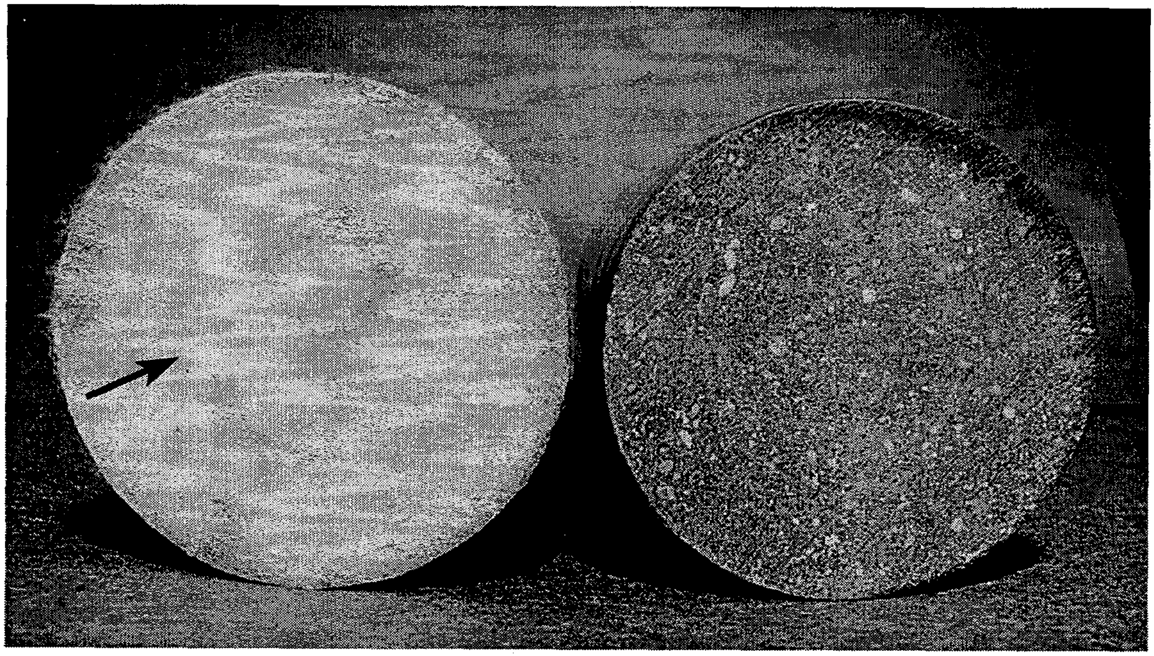


Figure 18. The white, fluffy growth that covered the top surface of each WAXFIX/waste form after 60 days in 100% humid storage.

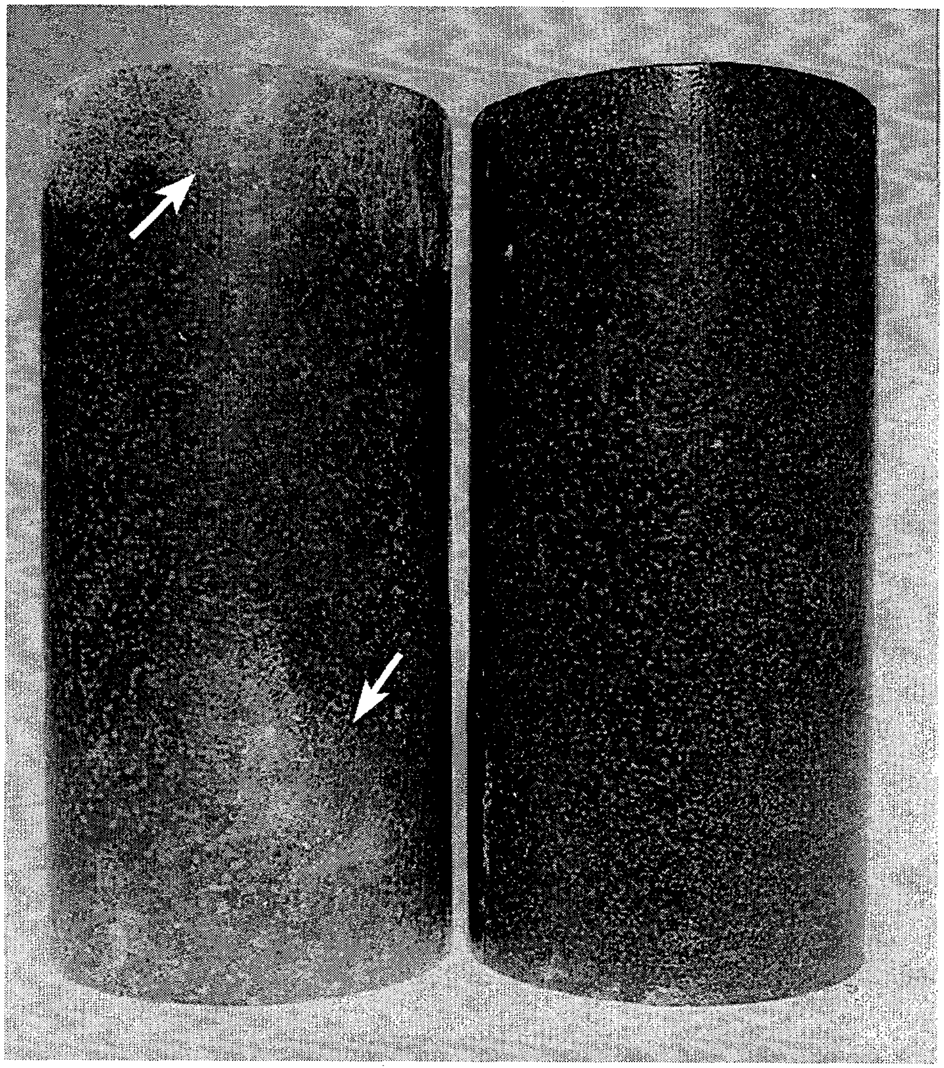


Figure 19. The white, fluffy growth that covered a portion of the lateral surface of each WAXFIX/waste form after 60 days in 100% humid storage.

~11. This decrease in pH indicated a continuous dissolution of compositional components. Figure 20 shows a baseline sample (left) and a swollen specimen that had been base immersed for 30 days (right). The immersed specimen appeared to be a viable composite even in its enlarged and discolored state. However, this particular sample was positioned very carefully and was only one of five specimens that did not disintegrate from immersion in aqueous sodium hydroxide. Figure 21 shows the other four specimens in the immersion container, with the solution removed, and in their final deteriorated state. It should be noted that the 10 replicates associated with the 60 and 90 day immersion tests showed the identical specimen degradation characteristics. Therefore, these tests were also terminated after 15 days.

Note that Carbray 100 Epoxy/ waste forms that had been stored in 100% humidity for more than 30 days began to weep/bleed a brown color liquid. The liquid material appeared to be similar in consistency and color to that of the lignosulfonate component. Figure 22 shows the dark brown liquid that had drained from two Carbray 100 Epoxy/waste forms following ~ 75 days of 100% humid storage. This phenomenon was slow and continuous (it has been observed >120 days). The loss of this dark brown material was not evident, with the Carbray 100 Epoxy/INEL soil specimens.

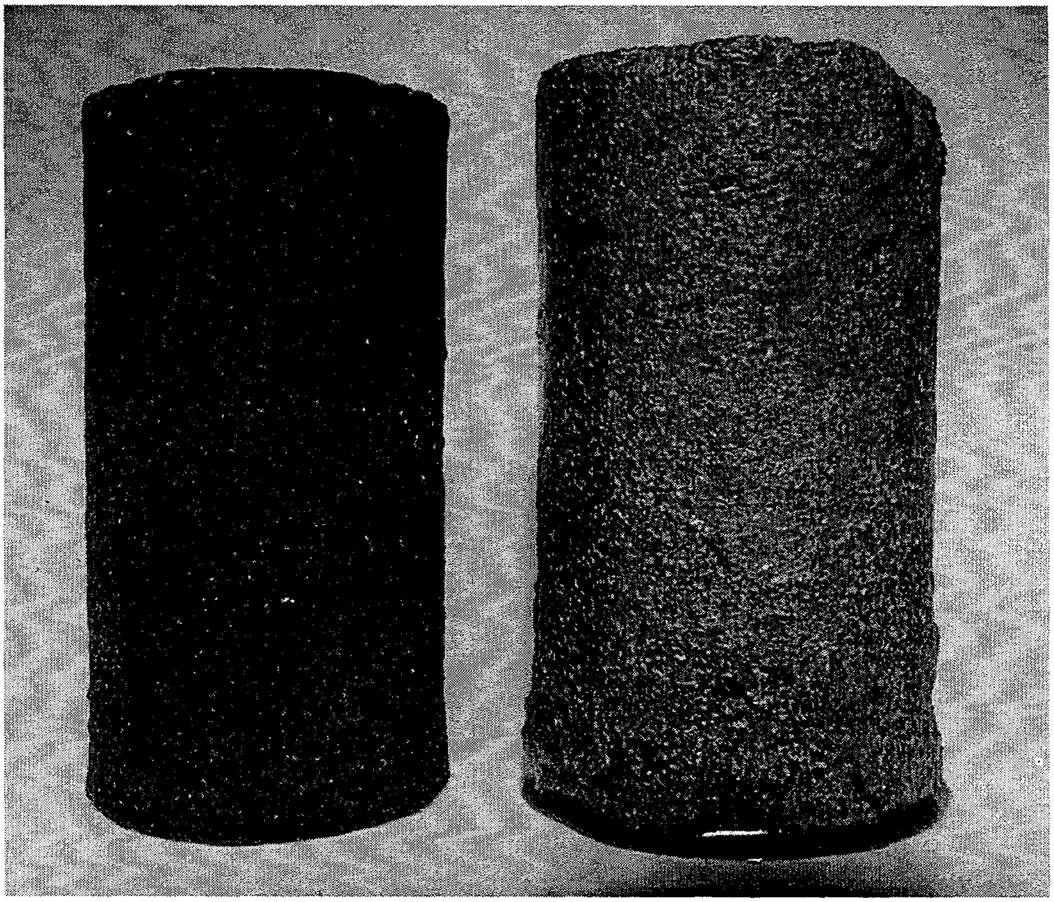


Figure 20. A comparison between a baseline Carbray 100 Epoxy/waste composite (left) and a swollen specimen following 30 days of sodium hydroxide (pH = 12.5) immersion (right).

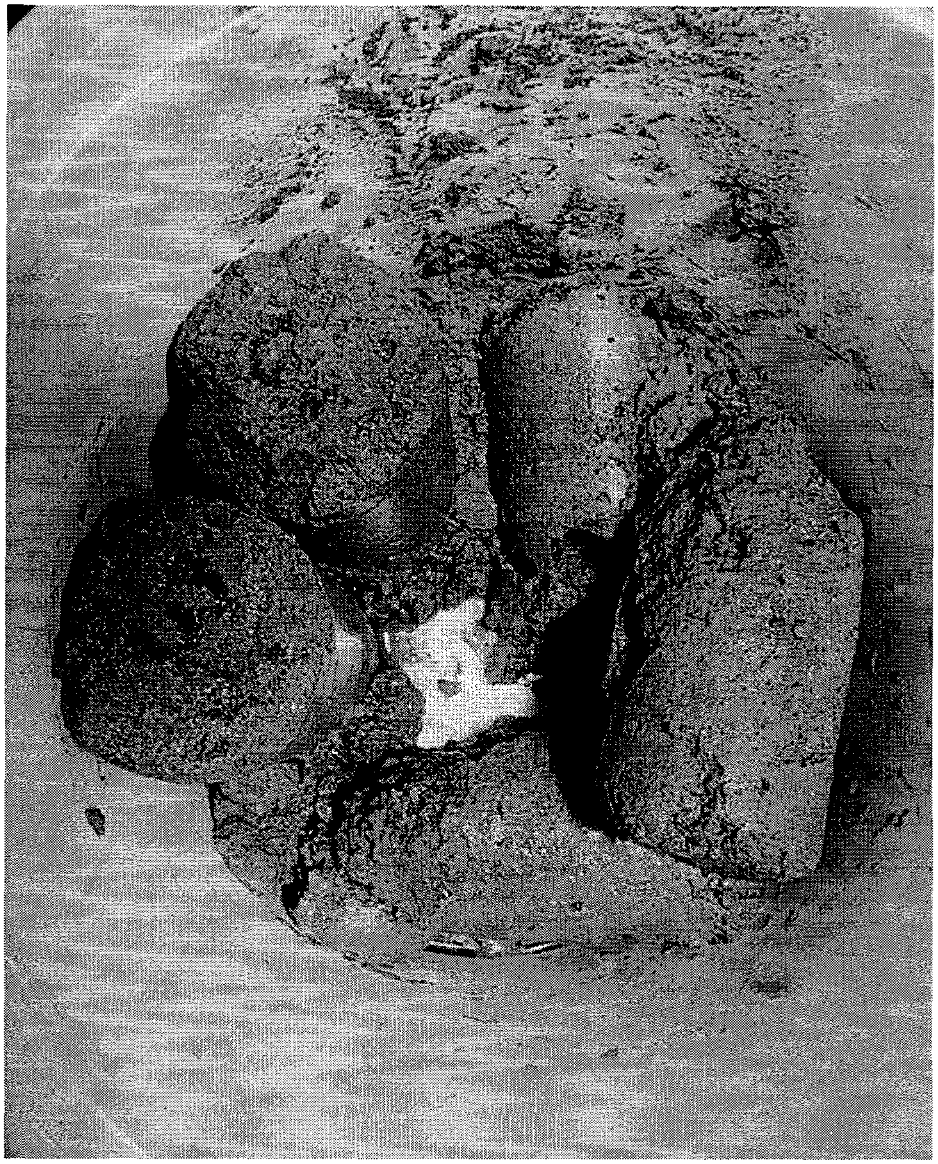


Figure 21. Four Carbray 100 Epoxy/waste compsites that failed sodium hydroxide (pH = 12.5) immersion after 15 days.

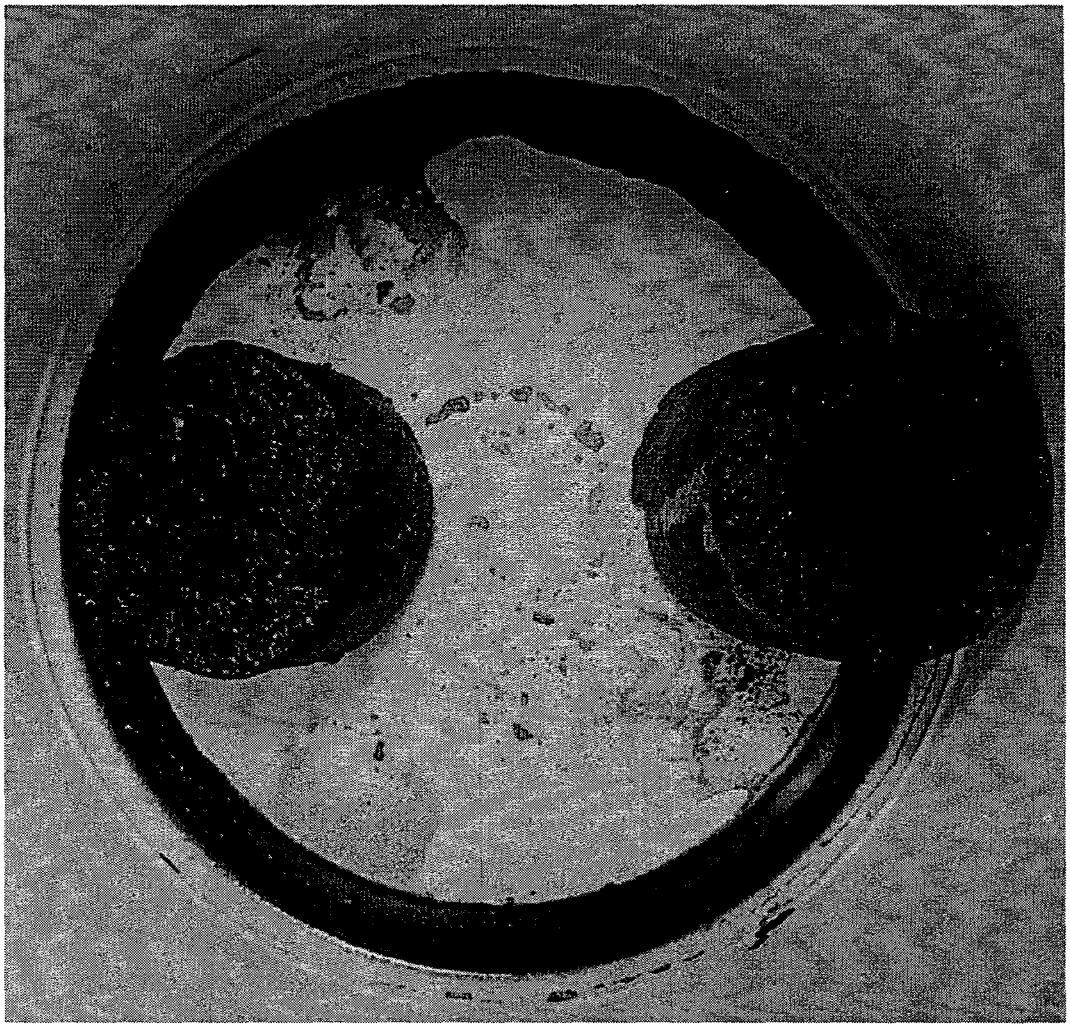


Figure 22. The dark brown liquid material that drained from two Carbray 100 Epoxy/ waste forms after ~75 days of 100% humid storage.

4.5 Solvent Resistance

Fifteen replicate samples prepared using the standardized waste formulation and encapsulated in TECT 1, WAXFIX, and Carbray 100 Epoxy grouting materials were immersed in deionized water saturated with trichloroethylene (TCE) at room temperature for immersion periods of 30, 60, and 90 days. TCE was chosen as the candidate media for VOC solvent testing because it appeared to be a predominate contaminant found at many DOE facilities. Five replicates of each WAXFIX/waste and TECT 1/waste grout composites were placed in the same three containers. Carbray 100 Epoxy/waste replicates (5) were placed in three separate immersion containers to avoid potential cross-contamination by compositional losses (dissolution/leaching) evidenced during the base immersion resistance testing. The WAXFIX/waste and TECT 1/waste products exhibited some minimal compositional losses during base immersion performance testing also. Therefore they solvent tested together in the same container. In each case, grout/waste composites were placed in their respective containers and sealed to zero head space in order to avoid evaporative losses. In addition, each container had a reservoir of excess TCE to keep the solution saturated, and a clear lexan lid to allow visual inspection of the samples. At each sampling interval, five samples were removed from one of the containers, rinsed, dried, measured, visually inspected, and placed in a 100% humidity environment for a minimum of four days. Then, each specimen was subjected to compressive strength testing in accordance with ASTM Method C-39 or D-695. Results are given in Table 15.

All TECT 1/ waste replicates completed the 30 day TCE immersion test as free standing monoliths. Composites showed no signs of cracking or flaking. However, a very fine/thin white residue was observed covering the specimens following solution removal and a short ambient air drying period (wet specimen - dark brown color). There were no specimen dimensional changes and only a small decrease in the mass (<0.1 wt%) was measured. The compressive strength of these 30 day TCE immersed specimens was 10.6 ± 0.6 MPa. The immersion solution at the completion of the test was a murky amber color. Sixty day TCE immersion results were similar to those cited at 30 days. However, a mean specimen mass loss and volume increase of 0.45%, and 0.3%, respectively were measured. The severity of the matrix attack was minimal that stage of the test since their was no specimen cracking and the mean compressive strength (10.6 ± 1.7 MPa) was comparable to the 30 day measurement. The solution remained a murky, greenish-amber color following 60 days of TECT 1/waste immersion (similar to the 30 day solution color). The TECT 1/waste composites 90 day TCE immersion tested resulted in no significant strength or structural changes. The compressive strength measured 10.0 ± 0.6 MPa. The specimen mass decreased by 0.6% and the volume increased by 0.5% at the completion of the 90 day immersion test. These results are similar to the 60 day immersion values. Compressive strengths for all TECT 1/waste forms immersed in TCE are presented in Figure 23 as percent of the baseline strength.

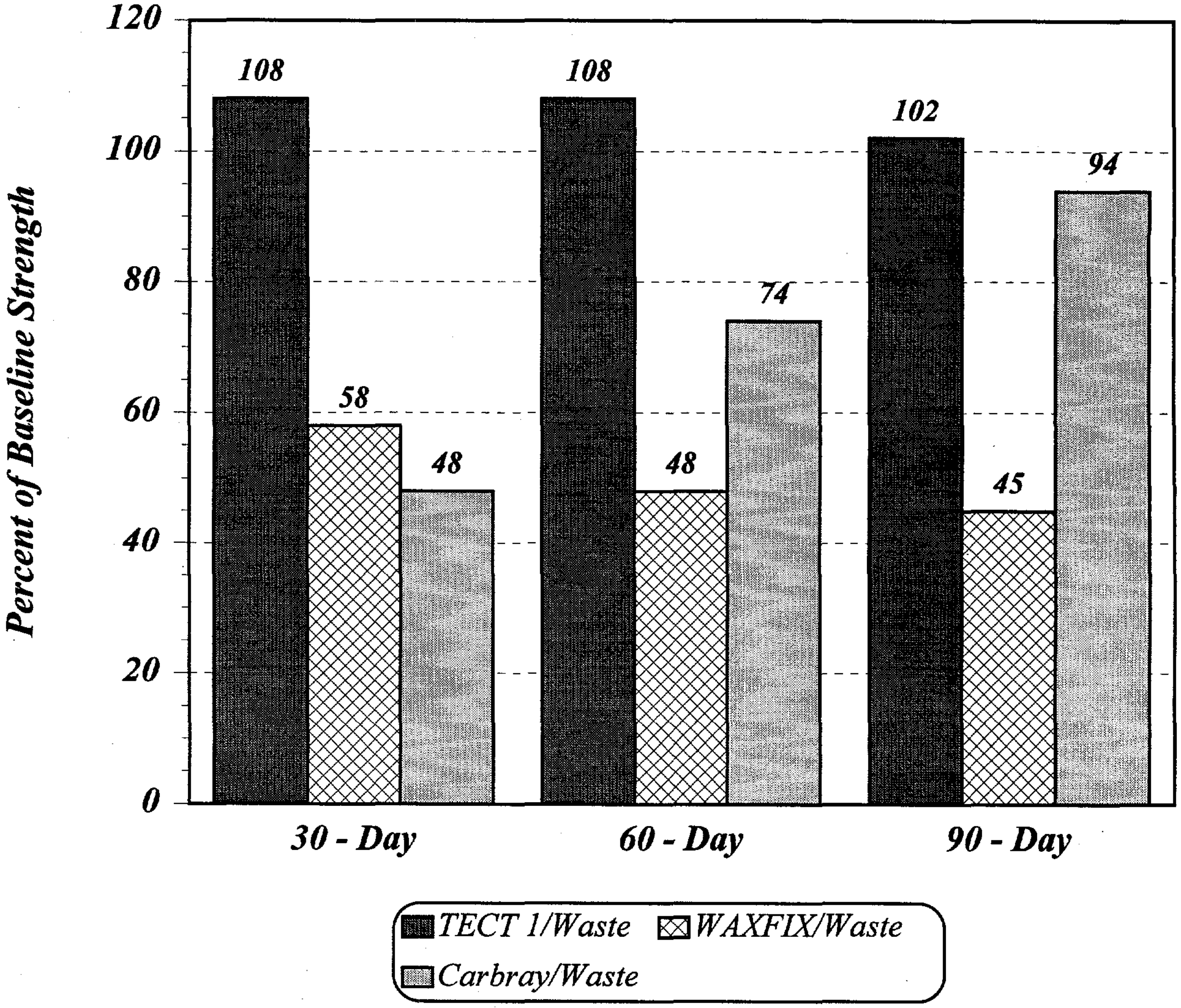


Figure 23. Effect of immersion in a saturated solution of trichlorethylene on the compressive strength of grout/waste composites.

Table 15. Unconfined compressive strengths of grout/waste forms following base and solvent immersion.

Grout/ Waste Stream	Compressive Strength, MPa [a]					
	Base Resistance			Solvent Resistance		
	30 days	60 days	90 days	30 days	60 days	90 days
TECT 1	10.2 ± 0.3	9.8 ± 1.5	10.0 ± 0.6	10.6 ± 0.6	10.6 ± 1.7	10.0 ± 0.6
WAXFIX	0.39± 0.03	0.35 ± 0.06	0.36 ± 0.07	0.42 ± 0.05	0.35 ± 0.05 (c)	0.33 ± 0.05 (b)
Carbray 100	Failed	Failed	Failed	0.022 ± 0.004	0.034 ± 0.005	0.043 ± 0.003
Portland II Cement Concrete (c)	39.4 ± 7.5	39.6 ± 3.5	26.6 ± 3.8	30.1 ± 3.8	41.7 ± 2.9	43.6 ± 2.3

(a) Results based on 5 replicates and 2 sigma errors.
(b) Results based on 4 replicates and 2 sigma errors.
(c) These data were taken from reference 12.

The WAXFIX/waste specimens felt "slimy" (greenish-black color) to the touch following removal (30-day) from the solvent solution, but this film (possible mold or bacteria growth) was easily removed when the sample was rinsed and towel dried for weighing. No composite chipping, cracking, and (unlike the base immersion WAXFIX/waste forms) no surface precipitation was observed. The mean sample gained 0.28% water mass during 31 days of storage in a 100% humidity environment. This same result was measured for base immersed samples. In addition, a 1.3% decrease in specimen mass was measured from humid storage removal to the completion of the 30 day immersion test. Conversely, the sample volume increased by a mean value of 2.4%. These factors in conjunction with nearly a 50% decline in the compressive strength (Baseline: 0.73 ± 0.03 MPa; 30 day TCE Immersed: 0.42 ± 0.05 MPa) indicate strongly the attack of solvent on the physical integrity of the WAXFIX/waste forms. WAXFIX/waste results for 60 and 90 day TCE immersion tests were comparable. Grout/waste forms were still "slimy" to the touch when removed from the saturated TCE solution and the random grey discoloration of the surface was evident, but not to the degree exhibited by the base immersed specimens. Specimen mass losses for 60 and 90 day resistance tests were 1.7% and 1.2%, respectively. Mean specimen volumes increased slightly with each successive immersion interval (30 day: 2.4%< 60 day: 3.4%< 90 day: 4%). Table 14 shows that the compressive strengths were unaffected by the solvent immersion residence time as the 60 day (0.35 ± 0.05 MPa) and 90 day (0.33 ± 0.05 MPa) results did not change statistically (see also Figure 23).

The Carbray 100 Epoxy/waste products were soft and spongy to the touch after 30 days of TCE immersion, but more rigid than their base immersed counterparts. A light finger pressure caused the samples to ooze liquid. These samples could be characterized as free standing monoliths with some minor sample flaking and crumbling observed in the immersion container. The solvent immersion solution turned an opaque, dark brown color, thus once again indicating substantial loss due to composite dissolution/leaching. Furthermore, there is a strong correlation between the color of the composite at the start of the test (dark brown) and tan at the completion of the test, to the dark brown immersion solution color. Additional evidence of specimen performance failure included a decrease in the average specimen mass by 3.3% and a increase in composite volume by 7%. These observations and factors suggested composite deterioration, and consequently, was reflected in the specimen compressive strength. Carbray 100 Epoxy/waste baseline compressive strengths measured an extremely weak 0.05 MPa, which further declined to 0.022 MPa following 30 days of TCE immersion. Solvent resistance tests conducted on Carbray 100 Epoxy/waste forms for 60 and 90 days resulted in no further mechanical strength changes. However, 3 of the 5 specimens tested following 60 days of immersion were observed to develop one relatively large crack per composite. Two of these lateral-vertical cracks were located on the upper third of the specimen, while the third was on the top surface. The lateral-vertical cracks were 2 - 2.5 cm in length, and <0.5 mm wide. The narrow cracking penetrated >90% of the specimen diameter. The top surface crack measured a length, width, and depth of 2.75 cm, 0.75 mm, and 3 cm, respectively. It was impossible to determine whether this cracking affected the mechanical strength of the specimens because all values were extremely low. Furthermore, it is possible that internal cracking had occurred within the two non-cracked specimens, but was undetected. No specimen cracking was observed with composites immersed for 90 days. The specimen/solution properties for both the 60 and 90 day solvent immersion samples include: rigid, but soft (finger pressure leaves impression), matrix surface oozing even after paper towel drying (apparent liquid saturation of specimen). The immersion solution was a opaque-dark brown color. No crystalline precipitates were observed on tested specimens. Table 16 shows that specimen mass losses and volume increases appeared to be stabilized after 30 days of immersion testing. Also note that an additional specimen mass (0.5%) and volume (0.7%) decrease was measured after a 7 day of equilibration period in 100% humidity and just prior to compressive strength testing. The 60 day (0.034 ± 0.005 MPa) and 90 day (0.043 ± 0.003 MPa) Carbray 100 Epoxy/waste form compressive strengths were measured at 10% deformation (Table 15). Extremely low mechanical strength continued to characterize these grout/waste composites.

The compressive strengths of all TCE immersed grout/waste forms are presented in Figure 20 as percent of baseline strength. Note that the compressive strengths for TECT 1/waste composites were unchanged essentially. The strength losses for WAXFIX/waste form, stabilized at 30 days of resistance testing at ~50% of baseline strength. Carbray 100 Epoxy/waste strengths appeared to increase with each successive immersion interval, but the low compressive strength values made trend determinations difficult.

Table 16. Mass and volume changes recorded for Carbray 100 Epoxy/waste forms following 30, 60, and, 90 days of solvent immersion.

Solvent Immersion Time (days)	% Mass Change	% Volume Change
30	-3.3	+7.0
60	-3.3	+4.3
90	-3.2	+7.2

4.6 Accelerated Leach Testing

Grout/waste forms were tested in accordance with the Accelerated Leach Test (ALT). This ASTM standard method (ASTM C-1308) was developed at BNL [13] to determine leach rates. Leaching may be accelerated in this model by preheating the leachant solutions above ambient temperature. (In theory, the effective diffusion coefficient (D_e) is temperature dependent according to the Arrhenius equation: $D_e = A\exp(-E_a/RT)$, where E_a is the activation energy.) All leach testing was performed at room temperature due to the sensitivity of certain grout materials to elevated temperatures.

Accelerated Leach Test samples were prepared by dissolving Lead (II) nitrate [$Pb(NO_3)_2$] and Chromium (III) nitrate [$Cr(NO_3)_3 \cdot 9H_2O$] in six liters of distilled water. One kilogram of INEL soil was added and mechanically mixed for 18 hours. The soil/liquid/dissolved metals were placed in two glass Pyrex trays and dried in an oven for five days at 110°C. The dried soil samples were ground using a mortar and pestle, weighed, and rehydrated to 12% moisture content using distilled water. Note that the final concentration of 1000 ppm for each metal (chromium and lead) was calculated based on the entire waste stream although the metals were loaded on the soil. The metals spiked soil was stored in a covered plastic container until specimen preparation was initiated.

Three grout materials were evaluated: TECT 1, WAXFIX, and Carbray 100 Epoxy. Mixing procedures for these grout/waste encapsulations were similar to the 5.1 cm x 10.2 cm products prepared for performance testing. TECT 1 grout samples were prepared at 60 wt% grout/40 wt% waste. WAXFIX and Carbray 100 Epoxy samples were prepared at 33 wt% grout/67 wt% waste. The standardized waste stream was used for each encapsulation, which was comprised of 70 wt% INEL soil, 20 wt% $NaNO_3$, and 10 wt% Canola oil. Batch mixes between 150 and 200 g were prepared and molded in 23 cm long by 2.5 cm diameter I.D. PVC tubing. All ALT specimens were cut to form nominal 2.5 cm x 2.5 cm right cylinders. As described previously, Carbray 100 Epoxy specimens were cut and removed from their molds after 14 days in order to cure the remainder of the 30 days in ambient air (as per vendor recommendations). Similarly, when the specimen mold cured mass/volumes (15 days) were compared to the air cured mass/volumes (15 days), there were mass and volume losses of 8 wt% and 4.4% respectively. Density calculations were also subject to change when the two curing environments were compared. The 15 day mold cure specimen density (1.56 ± 0.03 g/cm^3) decreased by ~4% after 15 days of air curing. (1.50 ± 0.02 g/cm^3). TECT 1/waste ALT specimen mean densities (1.96 ± 0.03 g/cm^3) compared favorably to the larger cylindrical grout/waste forms that were prepared for performance/durability testing (2.02 ± 0.004 g/cm^3). WAXFIX/waste showed moderate variations, with mean densities (1.57 ± 0.06 g/cm^3) ~15% higher than the cylindrical 5.1 cm x 10.2 cm test specimens counterparts (1.36 ± 0.01 g/cm^3). The reason for this discrepancy is not really known. However, it is possible that the heavier waste components (e.g., soil) settled prematurely. Three replicates of each grout/waste type were tested.

A pretest was performed on a representative sample of each grout/waste type to determine probable leachate metal concentrations. Following this, it was determined that all samples would be leached in 300 mL of leachate. (Normally the test specifies a 3000 mL leach volume to prevent saturation of the leach solution. Lower leach volumes can be used to effectively increase concentration of any metals which are leached when pretests prove this will not occur.) Specimens were suspended in the approximate center of each solution using nylon cable ties. Thirteen (13) leachant changes were made over an 11 day period, two the first day, then once daily for the remainder of the test. Approximately 125 mL of sample was collected at each interval in a polyethylene bottle which was then acidified with 0.1 ml of concentrated nitric acid (analytical reagent grade).

Leachates were analyzed by inductively coupled plasma (ICP) spectroscopy for lead and chromium metal concentrations. Results of the metals analyses were evaluated using the ALT computer program which calculates the Incremental Fraction Leached (IFL), Cumulative Fraction Leached (CFL), and the diffusion coefficient that best fit the leaching data. The mean cumulative and incremental fractional releases for replicate samples are given in Table 17. No chromium or lead releases were detected (i.e., below instrument detection limits, chromium: <0.04 μg/mL and lead: <0.14 μg/mL) for the TECT 1/waste and WAXFIX/waste forms following this short-term leach test. Conversely, Carbray 100 Epoxy/waste composite measurements resulted in mean cumulative fractional lead and chromium releases of 16.5% and 10.1%, respectively. The calculated mean diffusion coefficient was $2.6 \times 10^{-9} \pm 1.0 \times 10^{-9}$ cm^2/sec for lead and was $1.0 \times 10^{-9} \pm 1.5 \times 10^{-10}$ cm^2/sec for chromium.

4.7 Thermal Analysis - Differential Scanning Calorimetry

Differential Scanning Calorimetry (DSC) is a thermal analysis technique used to characterize enthalpy, or heat flux, of a sample relative to a reference material. In heat flux DSC, both sample and reference substance are heated in a furnace with a uniform temperature distribution. Temperatures of the two materials are monitored. The difference is proportional to the heat absorbed or evolved by the sample. Measurement sensitivity is very high due to the fact that temperature fluctuations in the furnace and changes in convection are extremely small. Testing was performed in accordance to ASTM method E-537, "Assessing the Thermal Stability of Chemicals by Methods of Differential Thermal Analysis." [14] DSC was used to access interactions between the organic components of the WAXFIX and Carbray 100 Epoxy grouts and the sodium nitrate of the waste stream. Sodium nitrate is an oxidizer and could potentially interact with the organics.

Table 17. Mean chromium and lead incremental and cumulative fractional releases from Carbray 100 Epoxy/waste forms.

Interval	**Incremental Time (days)**	**Lead**	**Chromium**	**Cumulative Time (days)**	**Lead**	**Chromium**
		IFL x 10^{-2}			**CFL**	
1	0.08	3.42	2.06	0.08	0.034	0.0206
2	0.21	3.00	1.65	0.29	0.064	0.0371
3	0.71	4.52	2.33	1	0.109	0.0604
4	1	2.53	1.52	2	0.135	0.0757
5	1	1.37	0.831	3	0.148	0.0841
6	1	0.776	0.525	4	0.156	0.0893
7	1	0.092	0.460	5	0.157	0.0937
8	1	0.215	0.281	6	0.160	0.0965
9	1	0.012	0.175	7	0.160	0.0983
10	1	0.141	0.152	8	0.161	0.097
11	1	0.087	0.090	9	0.162	.101
12	1	0.077	0.037	10	0.162	.101
13	1	0.293	0.036	11	0.165	.101

DSC runs were performed separately for each WAXFIX (12 and 125) and Carbray 100 Epoxy ("A", "B", and "C") grout component. A sample of pure $NaNO_3$ was tested also, representative of a waste surrogate material. Several grout/waste mixtures were prepared subsequently. A 50 wt% mixture with $NaNO_3$ was tested for each individual WAXFIX and Carbray 100 Epoxy component. In addition, mixtures of the two component WAXFIX (10.7 wt% WAXFIX 25 to 89.3 wt% WAXFIX 125) with $NaNO_3$ were prepared, at WAXFIX: $NaNO_3$ ratios of 25:75, 50:50, and 75:25. For Carbray 100 Epoxy, a 50:50 (wt%) grout to nitrate mixture was prepared, with the three component grout mixture consisting of 16 wt% hardener ("A"), 32 wt% epoxy resin ("B"), and 52 wt% lignosulfonate liquid ("C").

Between 5 and 30 mg of each material was loaded into individual 6 mm diameter aluminum pans which were sealed with aluminum covers subsequently. Duplicate samples of each material were prepared. DSC testing was done using a Shimadzu DSC-50. Test runs were performed from ambient temperature (20 - 23°C) to 350°C at a heating rate of 5°C/min. The furnace atmosphere was nitrogen gas flowing at approximately 30 cc/sec. A pinhole was drilled in the cover of each cell using a #80 (0.0135") drill bit when cells were discovered to be bursting during heating (these sealed cells were rated for use up to 3 atm) resulting in a spike or glitch in the DSC signal. The reference material for each run was an identically prepared empty aluminum cell.

Thermal analysis results for Carbray 100 Epoxy were varied. Three separate DSC scans were performed on the Carbray "A" (hardener) component, resulting in three different shaped thermograms. The different results were attributed, in part, to the relatively high vapor pressure associated with ethyl alcohol (43 mm Hg @ 20°C), a primary constituent of the hardener. The boiling point of ethyl alcohol is ~78°C. It is possible that rapid evaporation of the ethyl alcohol occurred prior to, or during, the early stages of material heating. No melting/boiling or decomposition peaks were produced for the Carbray "B" component (epoxy resin) up to the maximum test temperature of 350°C. The lignosulfonate liquid (component "C") showed multiple endothermic peaks between 100 - 120°C. However, no further peaks were observed during the remainder of the 350°C scan. The results were varied when a 50 wt% mix of sodium nitrate was added to each Carbray 100 Epoxy component. Sodium nitrate/components "A" and "B" mixtures exhibited no endothermic (melting/boiling) or exothermic (decomposition) reactions (i.e., a flat thermogram) until the melting point of the sodium nitrate (308°C) was approached. Carbray 100 Epoxy component "C" (lignosulfonate) and 50 wt% $NaNO_3$ results showed what appears to be a endothermic reaction between 100 and 140°C, but the results are vague and inconclusive. Figure 24 shows the thermogram (two different samples) for the three component, Carbray 100 Epoxy and 50 wt% sodium nitrate mixture. Note that there were no exothermic reactions or polymerization peaks recorded. Furthermore, there did not appear to be any rigorous reaction between the Carbray 100 Epoxy grout and the 50 wt% sodium nitrate until the melting point of the sodium nitrate was attained.

DSC runs on the WAXFIX 12 and 125 were performed. The WAXFIX 12 (a blue dye and proprietary ingredients) showed only minor reactions (essentially a flat thermogram) up to 350°C. WAXFIX 125 is basically a paraffin wax. It showed two endothermic reactions, with a small peak at ~38°C and a larger peak at ~58°C. The larger peak (~58°C), is presumed to be the melting point of the wax, and falls within the melting point of a typical paraffin wax (47 - 65°C). The vendor suggests jet-grouting the WAXFIX grout between 71 - 77°C. The solidification of the encapsulated wax grout/waste stream product occurred below 55 - 60°C, although waste stream dependent. The WAXFIX thermogram showed no other reactions (flat curve) occurring after the large peak, endotherm at ~58°C. WAXFIX 12 and the 50 wt% sodium nitrate results generated a broad, shallow peak between the temperatures of 50 - 60°C (wax melting point). Two small endothermic

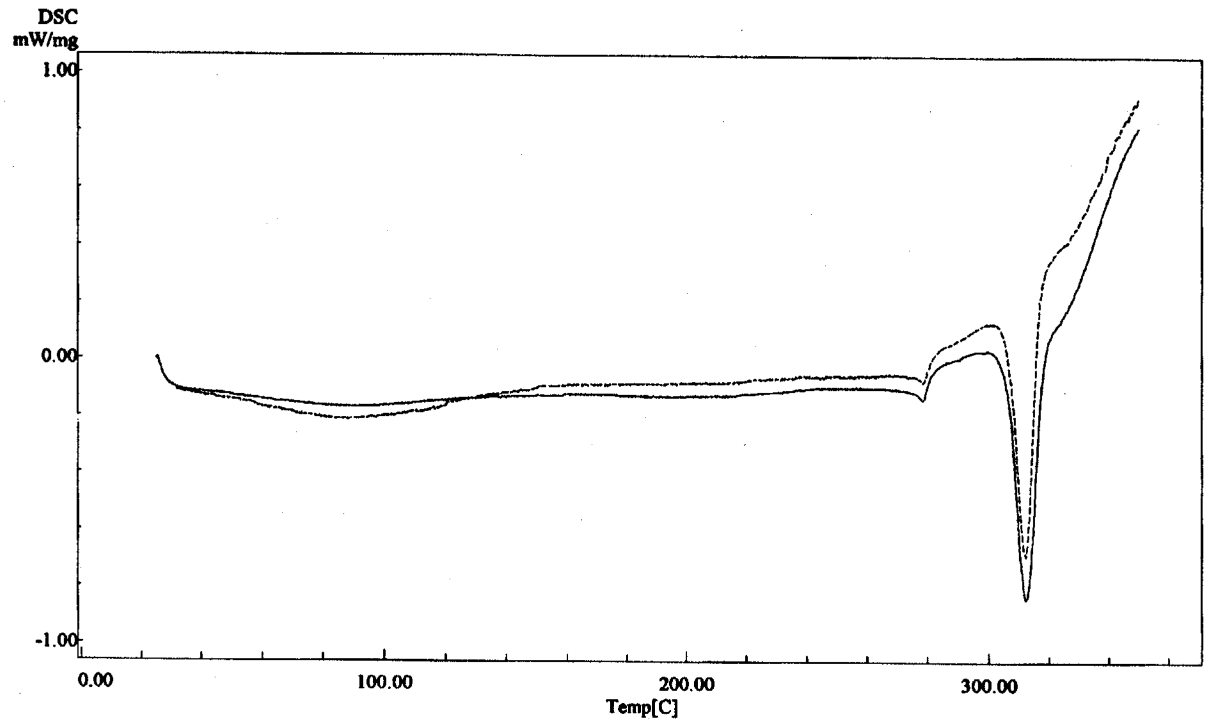


Figure 24. DSC thermogram of the three component, Carbray 100 Epoxy grout and 50 wt% sodium nitrate.

reactions were observed at a temperature of ~250°C. A larger reaction and peak were observed at the melting point of sodium nitrate. Figure 25 shows two WAXFIX 125 and 50 wt% $NaNO_3$ sample thermograms. Note, the small peak (endotherm) generated at the melting point of the wax and the larger peak (endotherm) produced at the melting point of the sodium nitrate. The endothermic reaction causing the production of a smaller peak just prior to the melting of sodium nitrate is unknown, but may be from impurities. Figure 26 shows a composite of the two component WAXFIX grout mixed with 25, 50, and 75 wt% sodium nitrate loadings. Note, as the percentage of the sodium nitrate increases, the size of the endothermic peak at ~300°C (melting point) increases. Conversely, the WAXFIX melting point peak at 58°C decreases in size simultaneously. Based on these results, there does not appear to be any major extraneous reactions occurring between the WAXFIX grout and the sodium nitrate up to a temperature of 350°C.

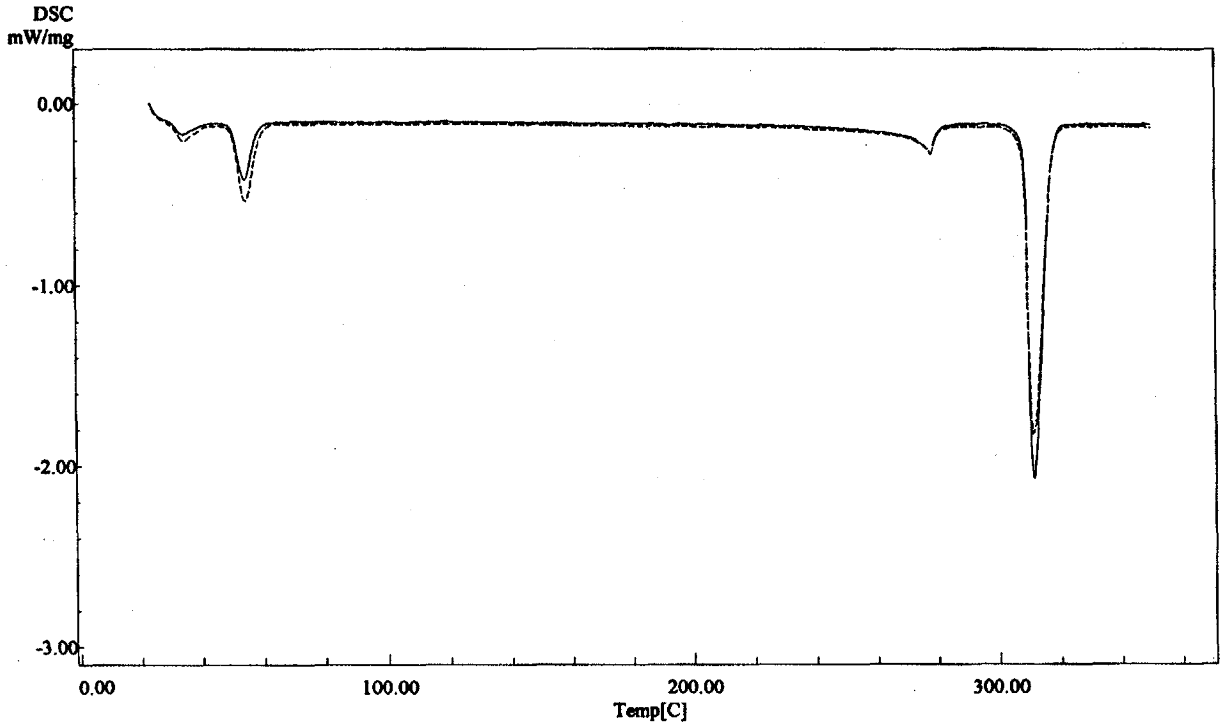


Figure 25. DSC thermogram of the two component, WAXFIX grout and 50 wt% sodium nitrate.

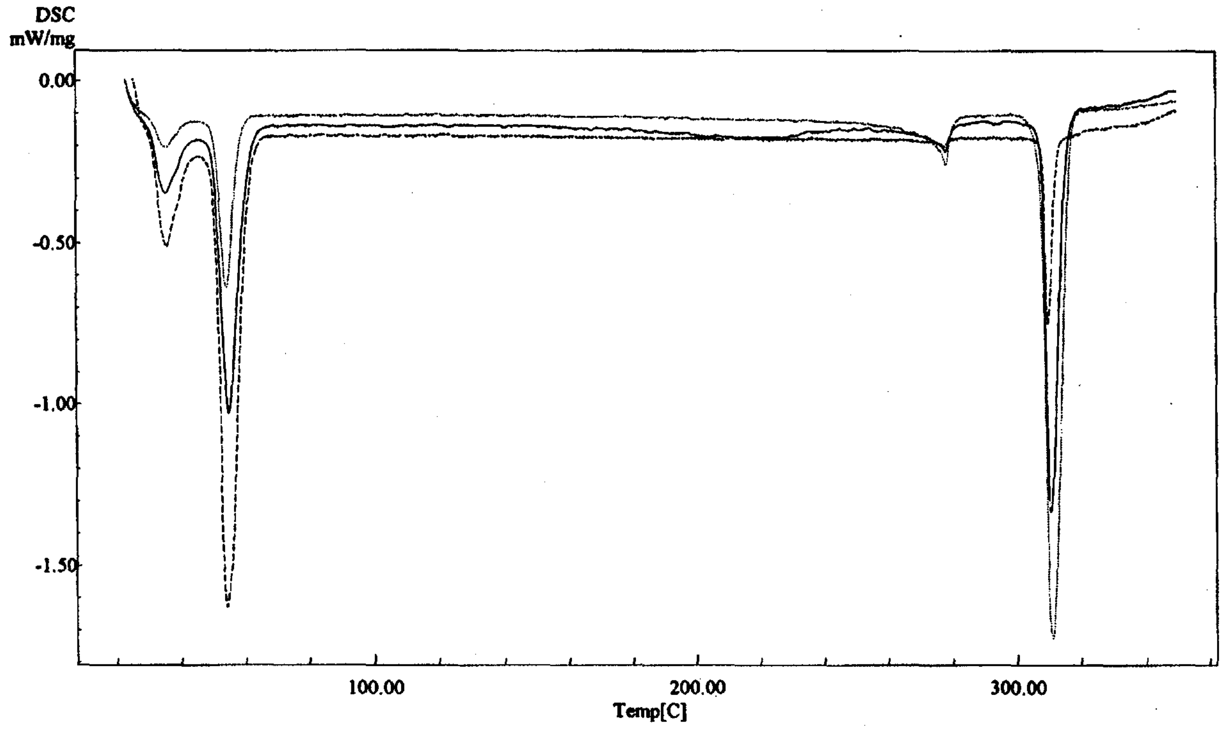


Figure 26. DSC thermogram of the two component, WAXFIX grout mixed with sodium nitrate at loadings of 25, 50, and 75 wt%.

5.0 SUMMARY

Four grout materials were investigated to evaluate long-term durability and compatibility with two different waste streams that require treatment/stabilization *in situ* at INEL burial sites. The surrogate waste stream consisted of sodium nitrate, Canola oil, and INEL soil. The second was comprised exclusively of INEL soil. Preliminary scoping studies necessitated formulation changes. Any possibility of using a universal mixing formulation for all of the grout materials considered was negated. A limited number of phosphate cement/INEL soil specimens were prepared and shipped from ANL, and therefore testing was limited also. Encapsulated grout/waste forms were subjected to seven performance/durability tests. Grout/INEL soil composites were limited to three tests: permeability, unconfined compressive strength and wet-dry cycling. All results were variable being grout type and test dependent.

Carbray 100 Epoxy waste and INEL soil specimens exhibited significant volume and weight losses during the 30 day curing period. Removal of the Carbray 100 Epoxy/waste form mold and placement in ambient air for approximately two weeks resulted in specimen mass and volume losses between 5 - 5.5% and 7.5 - 11.5%, respectively. Slightly lower results were measured for Carbray 100 Epoxy /INEL soil specimens (Mass: ~5% and Volume: ~7%). These same specimens lost considerable mass during the wet-dry testing also. WAXFIX/waste forms (between 0.2 - 0.28%) and Phosphate cement/INEL soil composites (~0.5%) moderately absorbed moisture during 100% humid storage (30 day). TECT 1/waste and INEL soil composites exhibited negligible volume and mass changes.

Hydraulic conductivities were low for all grout/waste and grout/INEL soil composites, except for the Phosphate/INEL soil forms. All the lower permeabilities measured were at the instrument detection limit of 2×10^{-11} cm/sec. The average permeability measured (2 determinations) for Phosphate/INEL soil composites was 3.1×10^{-7} cm/sec. A comparison between the permeabilites of a Phosphate/INEL soil (3.1×10^{-7} cm/sec) and a typical jet-grouted Portland cement slurry (~ 1×10^{-7} cm/sec) suggested a high composite void space and porosity.

Unconfined compressive strengths of grout/waste composites followed the order: TECT 1> WAXFIX>Carbray 100 Epoxy. The order for unconfined compressive strengths for grout/INEL soil encapsulations was TECT 1 > Phosphate cement >WAXFIX >Carbray 100 Epoxy.

Variable results were obtained following wet-dry cycle testing. All TECT 1 grout forms exhibited hairline cracking after just one wet-dry cycle. However, this initial cracking never increased and did not effect the specimen strength. Significant cracking was observed with Carbray 100 Epoxy/waste forms. However, it did not effect the compressive strength adversely, which increased by ~3.5 times over baseline values (although still below NRC regulatory requirements). No cracking developed with Carbray 100 Epoxy/INEL soil waste forms. Phosphate cement/INEL soil composites exhibited modest hairline cracking after four wet-dry cycles. This significantly reduced the compressive strength by more than 60%. After wet-dry cycling, the dried residue losses for each of the grout/waste composites were 2.9% for TECT 1 and 14.1% for Carbray 100 Epoxy. The mean dried residues losses for each grout/INEL soil composites were 0.1% for TECT 1, 0.5% for Carbray 100 Epoxy, and 0.5% for Phosphate cement. Volume changes were measured for all grout waste forms also. They include: TECT 1 (1%), and Carbray 100 Epoxy (6.8%) waste encapsulations and immobilized INEL soil in TECT 1(<1%), and Carbray 100 Epoxy (6%). No volume changes were measured for Phosphate cement/INEL soil composites. Note that both the Carbray 100 Epoxy/waste and Carbray 100 Epoxy/INEL soil composites decreased in volume from the time the specimens were removed from the mold (included air curing) to the completion of the wet-dry cycle testing of >18.5% and >8%, respectively. Compressive strength results were extremely variable. TECT 1/waste, Carbray 100 Epoxy/ waste and INEL soil encapsulations increased over their baseline strengths by 29%, 248%, and 190%, respectively. These increases are presumed to be the result of further curing/polymerization during the dry phase of the wet-dry cycle. Note that the substantial strength increases measured for Carbray 100 Epoxy grout forms are based on very low baseline values. TECT 1/INEL soil and Phosphate cement/INEL soil composites had strength losses of 34% and 64%, respectively.

All grout/waste composites completed 90 days of sodium hydroxide solution resistance testing except the Carbray 100 Epoxy/waste forms. The inability of these grout/waste forms to resist base immersion was exhibited immediately. The structural integrity of these composites was so poor that immersion testing was terminated after 15 days. Specimen deterioration included: swelling, softness, and extremely weak tensile strength (the specimens would tear if not supported carefully). The base immersion solution turned a opaque, dark brown color after 24 hours, indicating that compositional losses (dissolution/leaching) were fairly significant. Carbray 100 Epoxy/waste composites were too fragile to test for compressive strength. TECT 1/waste forms completed the 90-day test with only minor specimen deterioration. The compressive strengths of these specimens were unchanged statistically from their baseline counterparts. Slight compositional losses were identified as very fine white precipitates and by the amber color of the immersion solution. WAXFIX/waste composites demonstrated specimen volume increases and were attacked moderately by the sodium hydroxide solution. Precipitates on the specimen surface and immersion solution color changes (murky, amber) were evident within several weeks of resistance testing. Both specimen weight losses (<2%) and volume increases (<3.25%) indicated base solution attack. WAXFIX/waste composites lost 50% of their compressive strength after 30 days of sodium hydroxide solution immersion and did not deteriorate further during the remainder of the test. Random patches of slimy blue, green, and brown growths (possible mold and/or bacteria growth) were evident after 90-days of resistance testing.

All grout/waste composites completed 90 days of solvent resistance testing. However, the results were variable. The TECT 1/waste composites completed the solvent immersion test with only minor physical changes as observed with the base immersed samples. The mass decreased 0.6% and the volume increased a modest 0.5%. Unconfined compressive strengths were unchanged statistically from baseline values. WAXFIX/waste specimens showed mean volume increases in the following order: (30 day) 2.4% < (60 day) 3.4% < (90 day) < 4%. The WAXFIX/waste specimens were attacked by the solvent in the same manner as the base immersed samples. This conclusion is based on the specimen mass loss (<1.7%), murky amber immersion solution color (attributed mostly to the

WAXFIX/waste specimens dissolution/leaching), and the compressive strength declined by ~50% following 30 days of solvent immersion. After 90 days of TCE immersion, WAXFIX/waste specimens were slimy to the touch, indicating possible mold and/or bacteria growth. The solvent immersion of Carbray 100 Epoxy/waste forms resulted in products that were soft and spongy to the touch, but more rigid than their base immersed counterparts. However, liquid did ooze from the sample with light finger pressure (solution saturated) only. Solvent attack was evident as compositional losses (<3.3%) and volume increases (<7.2%) appeared to maximize in the first 30-days of TCE immersion. Specimen cracking was evident after 90-days of solvent immersion. Compressive strengths were extremely low (<0.043 MPa).

No chromium or lead releases were measured for TECT 1/waste and WAXFIX/waste forms following accelerated leach testing. Carbray 100 Epoxy/waste composite measurements resulted in mean cumulative fractional lead and chromium releases of 16.5% and 10.1%, respectively. The calculated mean diffusion coefficient for lead was $2.6 \times 10^{-9} \pm 1.0 \times 10^{-9}$ cm^2/sec and chromium was $1.0 \times 10^{-9} \pm 1.5 \times 10^{-10}$ cm^2/sec.

Differential Scanning Calorimetry of Carbray 100 Epoxy and WAXFIX grouts and different percentages of sodium nitrate mixtures were performed up to a temperature of 350°C. There did not appear to be any major extraneous reactions between the two component WAXFIX grout and sodium nitrate. A similar result for the three component Carbray 100 Epoxy and the sodium nitrate was demonstrated also.

6.0 CONCLUSIONS

The above-stated summary of the performance/durability testing performed at BNL indicates the strengths and weaknesses of each grout encapsulated waste stream as tested in the laboratory. Below is a brief description with the final conclusions concerning the performance of each grout/waste stream.

TECT 1/Waste

- *Unconfined Compressive Strength* - the 9.8 ± 2.0 MPa compressive strength measured was sufficiently high to be used in the treatment/stabilization of this waste stream.
- *Hydraulic Conductivity* - was below instrument detection limits (2×10^{-11} cm/sec).
- *Wet/Dry Cycle* - the modest specimen exterior surface hairline cracking and compositional losses (2.9%) did not effect the mechanical strength (12.6 ± 1.9 MPa). The compressive strength actually increased by nearly 30% (possible acceleration in curing during the dry cycle at 60°C), thus enhancing an already acceptable product.

- *Base Resistance* - after 90 days of sodium hydroxide immersion (pH=12.5), specimens exhibited minor volume increases (0.5%) and mass losses (0.8%). However, these physical changes did not adversely effect the compressive strength (10.0 ± 0.6 MPa) of these grout/waste forms, which were similar to their baseline counterparts. Although minor compositional losses due to dissolution/leaching from these specimens occurred, test results were still deemed satisfactory.

- *Solvent Resistance* - after 90 days of trichloroethylene immersion the compressive strength values (10.0 ± 0.6 MPa) were statistically unchanged when compared to baseline values. The specimen mass decreased by 0.6% and the volume increased by 0.5%. Based on these results, visual observations, and the compressive strength values achieved, these grout/waste products successfully passed this durability test.

- *Accelerated Leach Test* - no chromium or lead releases were detected following 11 days of leaching.

<u>TECT 1/INEL Soil</u>

- *Unconfined Compressive Strength* - the 20.5 ± 2.8 MPa compressive strength was the highest value measured for all grout/INEL soil composites tested.

- *Hydraulic Conductivity* - was below instrument detection limits (2 x 10^{-11} cm/sec).

- *Wet/Dry Cycle* - modest hairline cracking characterized the external surfaces of these specimens and the mean compressive strength decreased by approximately one-third (13.5 ± 2.7 MPa) in comparison to mean baseline values (20.5 ± 2.8 MPa). This is a considerable loss in mechanical strength, and the result is a caution to what could potentially occur in similar environmental conditions over longer periods.

<u>WAXFIX/Waste</u>

- *Unconfined Compressive Strength* - the measured compressive strength of 0.73 ± 0.03 MPa was low, but the strength appears to be sufficient for the treatment/stabilization of this waste stream (the value is greater than the NRC waste form criteria of 60 psi (0.41 MPa).

- *Hydraulic Conductivity* - was below instrument detection limits (2 x 10^{-11} cm/sec).

- *Wet/Dry Cycle* - this particular test could not be completed because the dry cycle temperature (60 °C) was sufficient to allow the specimens to creep and slump.

- *Base Resistance* - after 90 days of sodium hydroxide immersion (pH=12.5), specimens increased in volume by 5%, revealed various forms of deterioration (e.g., precipitate formations, surface discoloration, immersion solution pH decrease and discoloration) and lost ~1% of their original mass. The compressive strength decreased by a significant ~50% to 0.36 ± 0.007 MPa.

- *Solvent Resistance* - after 90 days of trichloroethylene immersion, specimen degradation was indicated by a mass loss of 1.2%, a volume increase (swelling) of 4%, and compressive strength decreased by >50% (0.33 ± 0.05 MPa).

- *Accelerated Leach Test* - no chromium or lead releases were detected following 11 days of leaching.

- Thermal Analysis (DSC) - thermograms of both WAXFIX components mixed with 50 wt% sodium nitrate showed a small peak (endotherm) generated at the melting point of the wax and a larger peak (endotherm) produced at the melting point of sodium nitrate. Based on all the results, there does not appear to be any major extraneous reactions occurring between the WAXFIX grout and the sodium nitrate up to a temperature of ~300°C.

WAXFIX/INEL Soil

- *Unconfined Compressive Strength* - the measured compressive strength of 1.8 ± 0.2 MPa was low, but greater than the NRC waste form criteria.

- *Hydraulic Conductivity* - was below instrument detection limits (2 x 10^{-11} cm/sec).

- *Wet/Dry Cycle* - this particular test could not be completed because the dry cycle temperature (60°C)was sufficient to allow the specimens to creep and slump.

Carbray 100 Epoxy/Waste

- *Unconfined Compressive Strength* - the compressive strength of 0.046 ± 0.008 MPa was the lowest value measured for all grout products tested and well below the NRC waste form criteria. This may not preclude this material from *in situ* stabilization since it is unclear whether NRC strength requirements apply to *in situ* stabilization.

- *Hydraulic Conductivity* - no hydraulic conductivity measurements were performed because of the inability to de-gas the specimen during the back pressuring procedural step.

- *Wet/Dry Cycle* - both the top and bottom exterior surfaces of the grout forms were observed to increase in cracking throughout the test. Simultaneously, the interiors of 3 of 5 specimens were observed to crack with each successive cycle. The mean specimen volume decreased 6.8%, while 14.1% of the specimen mass was lost due to dissolution/leaching and/or evaporation of the matrix. The wet cycle immersion solution changed to an opaque/dark brown color and the original dark brown color of the sample turned a light tan color at the completion of the test. Nevertheless, the compressive strength of these wet-dry cycled specimens increased ~3.5 times over baseline values to 0.16 ± 0.03 MPa (still very weak). However, due to the many unfavorable results (e.g., the large size of the internal/external cracking, the low compressive strengths, and the severe dissolution/leaching of the matrix which could potentially deteriorate further), these results were deemed unacceptable.

- *Base Resistance* - all grout/waste composites failed the immersion test after 15 days because the specimens were too soft to handle, too weak to compression test, and the majority of the composites disintegrated in the immersion container.

- *Solvent Resistance* - after 90 days of trichloroethylene immersion, specimen degradation was indicated by a mass loss of 3.2% and a volume increase (swelling) of 7.2%. The compressive strength measurements were extremely weak 0.043 ± 0.003 MPa (similar to baseline values). Furthermore, the specimens were observed oozing liquid after paper towel drying and the immersion solution was a dark/opaque color indicating substantial compositional losses due to dissolution/leaching. In addition, some sample cracking was observed. Because of the many destructive results cited, these grout forms failed the solvent immersion test.

- *Accelerated Leach Test* - mean cumulative fractional releases of 10.1% chromium and 16.5% lead releases were detected following 11 days of leaching. These releases along with the results of the other durability testing strongly indicate a problem with the grout/waste stability. Therefore, this formulation should not be considered for *in situ* treatment/stabilization applications.

- Thermal Analysis (DSC) - a thermogram of Carbray 100 Epoxy and 50 wt% sodium nitrate mixture showed no exothermic reactions or polymerization peaks. Also, there did not appear to be any rigorous reaction between the Carbray 100 Epoxy grout and the 50 wt% sodium nitrate until the melting point of the sodium nitrate was (308°C) reached. Since no extraneous reactions occurred, it can be assumed that the mixing of the two materials together results in a reasonably stable product up to ~300°C.

Carbray 100 Epoxy/INEL Soil

- Unconfined Compressive Strength - even though a low mean compressive strength of 0.33 ± 0.02 MPa was measured for these grout products, the composites were free standing monoliths, which effectively maintain their structural stability.

- *Hydraulic Conductivity* - was below instrument detection limits (2 x 10^{-11} cm/sec).

- *Wet/Dry Cycle* - minor specimen size and shape changes occurred during the testing. The specimen volume decreased by 6% and, with time, a slight barrel-like shape was observed. The final clear-yellow immersion solution color revealed minor compositional losses that were verified by the remaining residue (0.5%) dried at the termination of the test. The compressive strength of the grout/INEL soil composite (0.96 ± 0.06 MPa) increased by nearly 3x in comparison to the baseline values. This beneficial result was probably due to increased curing induced by the elevated temperatures associated with the dry cycle (60°C). Overall these grout/soil composites performed well in this test.

Phosphate Cement/INEL Soil

- Unconfined Compressive Strength - a reasonably high mean compressive strength of 8.3 ± 0.9 MPa was measured for these grout products.

- *Hydraulic Conductivity* - a mean permeability of $3.1 \times 10^{-7} \pm 0.1 \times 10^{-7}$ cm/s was measured for two specimens. This higher rate, is indicative of high porosity and void space, which is comparable to a jet-grouted Portland cement slurry ($\sim 1.0 \times 10^{-7}$ cm/s). Any modification to reduce the permeability rate of this grout/soil product would be an enhancement.

- *Wet/Dry Cycle* - by the termination of the test, hairline cracking was observed on the majority of the exterior surface of the specimen. No specimen dimensional changes and only minor compositional losses (0.5%) were measured. However, the wet-dry tested specimens (3.0 ± 1.2 MPa) lost nearly two-thirds of their original baseline compressive strength (8.3 ± 0.9 MPa). It is unclear whether the NRC criteria for post-immersion mean compressive strengths (e.g., the immersion test requires that the post-immersion compressive strength be equal to or greater than 3.45 MPa (500 psi) and not less than 75% of the pre-immersed compressive strength) would apply to the *in situ* stabilization waste forms or if EPA or state agencies would apply similar standards.

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