Ultrafine Cement Grout For Sealing Underground Nuclear Waste Repositories

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ABSTRACT: Sealing fractures in nuclear waste repositories concerns all programs investigating deep burial as a means of disposal. Because the most likely mechanism for contaminant migration is by dissolution and movement through groundwater, sealing programs are seeking low-viscosity sealants that are chemically, mineralogically, and physically compatible with their host. This paper presents the results of collaborative work between Whitesell Laboratories, operated by Atomic Energy of Canada, Ltd., and Sandia National Laboratories; the work was undertaken in support of the Waste Isolation Pilot Plant (WIPP). This effort addresses the technology associated with long-term isolation of nuclear waste in a natural salt medium. The work presented is part of the plugging and sealing program, specifically the development and optimization of Ultrafine cementitious grout that can be injected to adequately lower excessive, strain-induced permeability in the Distributed Rock Zone (DRZ) surrounding underground excavations. Innovative equipment and procedures employed in the laboratory produced a usable cement-based grout whose particles are 90% smaller than 8 microns and average 4 microns. The process involved simultaneous wet pulverization and mixing. The grout was used for a successful in situ test underground at the WIPP. Injection of grout sealed microfractures as small as 8 microns and lowered the gas permeability of the DRZ by three orders of magnitude. Following the WIPP test, additional work produced an improved version of the grout containing particles 90% smaller than 6 microns and averaging 2 microns. This grout can be produced in the dry form at a competitive cost ready to mix.

1 INTRODUCTION

The Waste Isolation Pilot Plant (WIPP) is a research and development facility being evaluated for its suitability to dispose of approximately 185,000 cubic meters of transuranic, defense-generated wastes. The WIPP site is located in southeastern New Mexico within a vast geologic structure known as the Permian basin. The location of the WIPP relative to the surface is illustrated in Figure 1. The disposal horizon is situated in bedded salt (the Salado Formation) at a depth of 658 m.

One advantage of salt as a host for a waste repository is its ability to creep and ultimately encapsulate the waste. Creep closure begins immediately after excavation; early time closure rates at the WIPP are on the order of several centimeters per year (Munson et al., 1987). The creation of underground openings changes the stress state from lithostatic to one of maximal stress difference in the proximity of the excavated surface, and the stress difference gives rise to fracturing. Fracturing from excavation-related stress redistribution creates a near-field zone of mechanically altered rock within the first few meters of the excavations. Within this disturbed rock zone (DRZ), fractures increase the intrinsic rock permeability by a few orders of magnitude (Stormont, 1989). Fractures in the DRZ provide potential pathways for contaminant transport, especially in higher permeability interbeds (Ahrens et al., 1996). The WIPP sealing program is investigating the use of low-permeability grouts to seal these fractures.

Much of the increased permeability in the DRZ results from formation of microfractures having apertures equal to or less than 100 microns, which the grout must penetrate effectively. A rule of thumb in the grouting industry states that to penetrate a fracture, the maximum particle dimension in the grout should be no more than one third of the fracture aperture, clearly indicating the need for grout with very fine particles. For WIPP seal applications, a cementitious grout was preferred because of its low cost, ready availability, engineering properties, successful history of use, and nontoxic attributes.

Commercially available microfine cement grouts were evaluated, but particle sizes were larger than desired (e.g., 90% smaller than 10 microns) or compressive strengths too low for expected WIPP test conditions. It was therefore decided to develop a new product with smaller particles and higher compres-

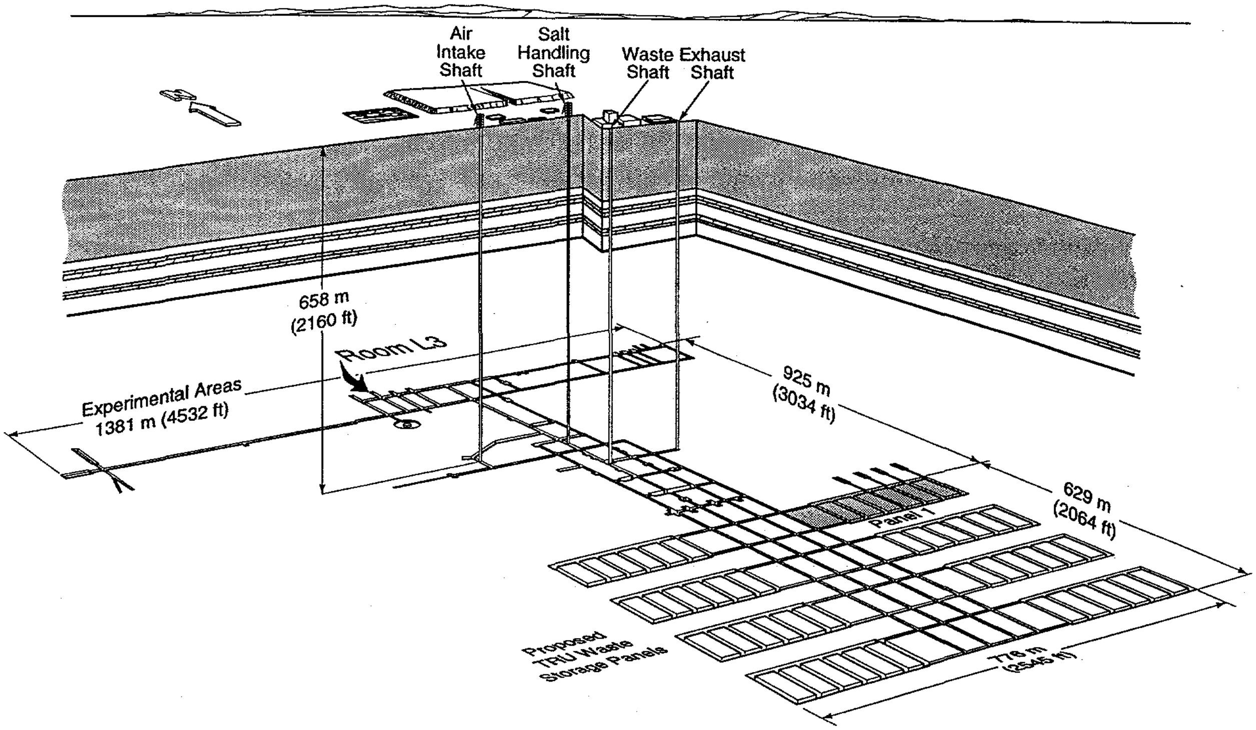

Figure 1. Isometric view of the WIPP, north-central portion.

sive strength. This paper summarizes the successful development and demonstration of this Ultrafine grout.

2 LABORATORY DEVELOPMENT

The desired characteristics of the new Ultrafine grout were:

- Low water content: Minimal water/cement (W/C) ratio and no bleed (i.e., no free water when the grout hardens) because more water than necessary decreases the compressive strength and increases the permeability.
- Injectability: Three hours of injectability without measurable agglomeration.
- Volume stability: Linear shrinkage less than 1% at 50% relative humidity.
- Insoluble: Minimal calcium hydroxide, which is the most soluble phase of the hardened cement paste.
- High compressive strength.
- Low superplasticizer content: Excess superplasticizer increases shrinkage when the grout hardens.

Pulverization equipment was reviewed in depth, resulting in the selection of the Szegvari attritor because of its operational simplicity and ability to pulverize material to an extremely small particle size. Numerous trials in the Szegvari test laboratory in Akron, Ohio indicated that simultaneous wet pulverization and mixing could produce grout with very small particles. Concurrent attempts to pulverize dry were unsuccessful because of an inability to identify a proper dry grinding aid (the grout adhered to the attritor mill and grinding balls).

Whiteshell Laboratories (WL) and Sandia National Laboratories (SNL) conducted the laboratory work at the WL facility. Test equipment consisted of a small batch Attritor, a laser particle size analyzer, and an agitation tank (because the grout is thixotropic, which means it establishes a false set unless agitated). Rheology was determined with a rotary viscometer, and permeability was measured with custom equipment designed and built by WL (Onofrei et al., 1993).

Permeability was determined by measuring the hydraulic conductivity of hollow, cylindrical grout specimens, 150 millimeters long and 75 millimeters in diameter, subjected to a compressive stress in specially designed radial flow permeameters. The apparatus is designed for pressures as high as 10 MPa. The tests were performed at hydraulic pressure differences between 0.4 and 2.1 MPa.

The laboratory variables were:

- Type and weight percent of (1) Portland cement, (2) pozzolan, (3) superplasticizer and (4) W/C ratio.
- Diameter and composition of the pulverization balls (3 mm, 316 stainless steel balls were chosen for simultaneous wet pulverization and mixing).
- Mill rotational speed and duration of pulverization.

Laboratory work included evaluation of 90 mix combinations, including mechanical factors such as particle size, rheology, compressive strength, initial and final set times, linear shrinkage, and permeability. Type 5, sulfate-resistant Portland cement was chosen for its low heat of hydration and because WIPP brines are locally high in sulfate.

Based on laboratory investigations, a grout with the following characteristics was selected for field demonstrations:

- W/C ratio of 0.5/1.
- Stable (no water separation when the grout hardens).
- Particles 90% smaller than 10 microns and averaging 4 microns.
- Rheology suitable for three hours of injectability subsequent to pulverization/mixing with no measurable agglomeration.
- Compressive strength of 39.7 MPa at 28 days.
- Permeability of 1×10^{-14} m/s in 63 days decreasing to 1×10^{-16}.
- Linear shrinkage less than 1% in 50% relative humidity (slight expansion if wet).
- Three hours of injectability.
- Final Vicat needle set in 14.5 hours.

3 IN SITU GROUTING TEST AT THE WIPP

After laboratory development, a full-scale demonstration in the WIPP underground was undertaken (Ahrens et al., 1996). The test was conducted in Room L3 (location shown in Figure 1), which is 10.1 meters wide × 47.1 meters long × 3.7 meters high. A marker bed (MB139), approximately 1 meter thick and situated 2 meters below and parallel to the floor of L3, was selected for the grouting demonstration. MB139 is a multilithologic unit consisting of laterally discontinuous pods of anhydrite, halite, polyhalite and gypsum. When stress-relieved near excavations, depositional interfaces in MB139 open, increasing permeability. Thus MB139 is an ideal entity for evaluating the effectiveness of grouting.

A reinforced concrete slab measuring 7.6 × 7.6 × 0.4 meters was poured on leveled halite in the selected test location. The position of the slab relative to Room L3 is shown in Figure 2. The slab served as a staging platform that could be grouted through and against. Upward movement of the slab, resulting from the subjacent injection of pressurized grout, was resisted by four large hydraulic floor jacks (mechanically locked after placement) braced against the ceiling. The slab remained crack-free for the duration of the test.

Gas flow testing was conducted near but exterior to the proposed test location to obtain baseline gas transmissivity data. Twelve 15-centimeter-diameter vertical boreholes were drilled 3.7 meters deep; six were north and six south of the test location. After drilling, these holes were examined using a downhole color video camera

A series of short-duration, instrument-air flow tests was then conducted using a four-packer test tool with two end packers inflated and instrument air injected from the end port. The downhole configuration tested the air transmissivity of the fractured rock by utilizing an air-pressurized test interval between the bottom of the hole and the lower packer, which was located 1.3 meters below the room floor. At a constant pressure, the air flow rate into the test interval was a function of the air transmissivity of the rock into which the borehole was drilled.

Initial test zone target pressures of 0.14 MPa were used. Generally, in halite near the room walls or in the far field, instrument grade air transmissivity is sufficiently small for accurate pressure-decay estimates using one isolated test interval. However, when the maximum air flow rate from the four-packer test tool could not pressurize the test region because of excessive formation transmissivity, specific borehole areas were isolated to locate the more permeable region. Four of the preliminary test holes were subjected to complete isolated-interval testing as part of a baseline permeability study. Air transmissivity of the DRZ below the floor of Room L3 was determined to be approximately 1×10^{-12} m^2.

Eight primary and eight secondary grout holes were drilled (see Figure 2); thus the demonstration required 16 separate grout batches. The holes were drilled with custom-built, reverse-circulation diamond drill equipment, which prevented fine-grained drill cuttings from plugging the microfractures in the formation (Ahrens et al., 1996).

The ultrafine grout developed for the demonstration was to be injected into the fractured rock at pressures as high as 6.9 MPa. Because the grout was thixotropic, a circulating grout-injection system was used. Distinctively colored grout for each hole was simultaneously pulverized and mixed in a 50S Szegvari batch attritor. Particle size and rheology were determined every 15 minutes from the initiation of pulverization throughout the grouting period. No agglomeration and less than 1-micron variation in particle size were noted throughout the 16 batches.

Gas flow testing was conducted after grouting the primary holes and again after grouting the secondary holes. These tests consisted of injecting instrument grade air into the central hole until constant flow was attained, and then injecting a 200-mg/kg spike of isobutene tracer gas. This was followed by a two-hour pulse of instrument grade air to displace the tracer. If the tracer gas was detected in the outer

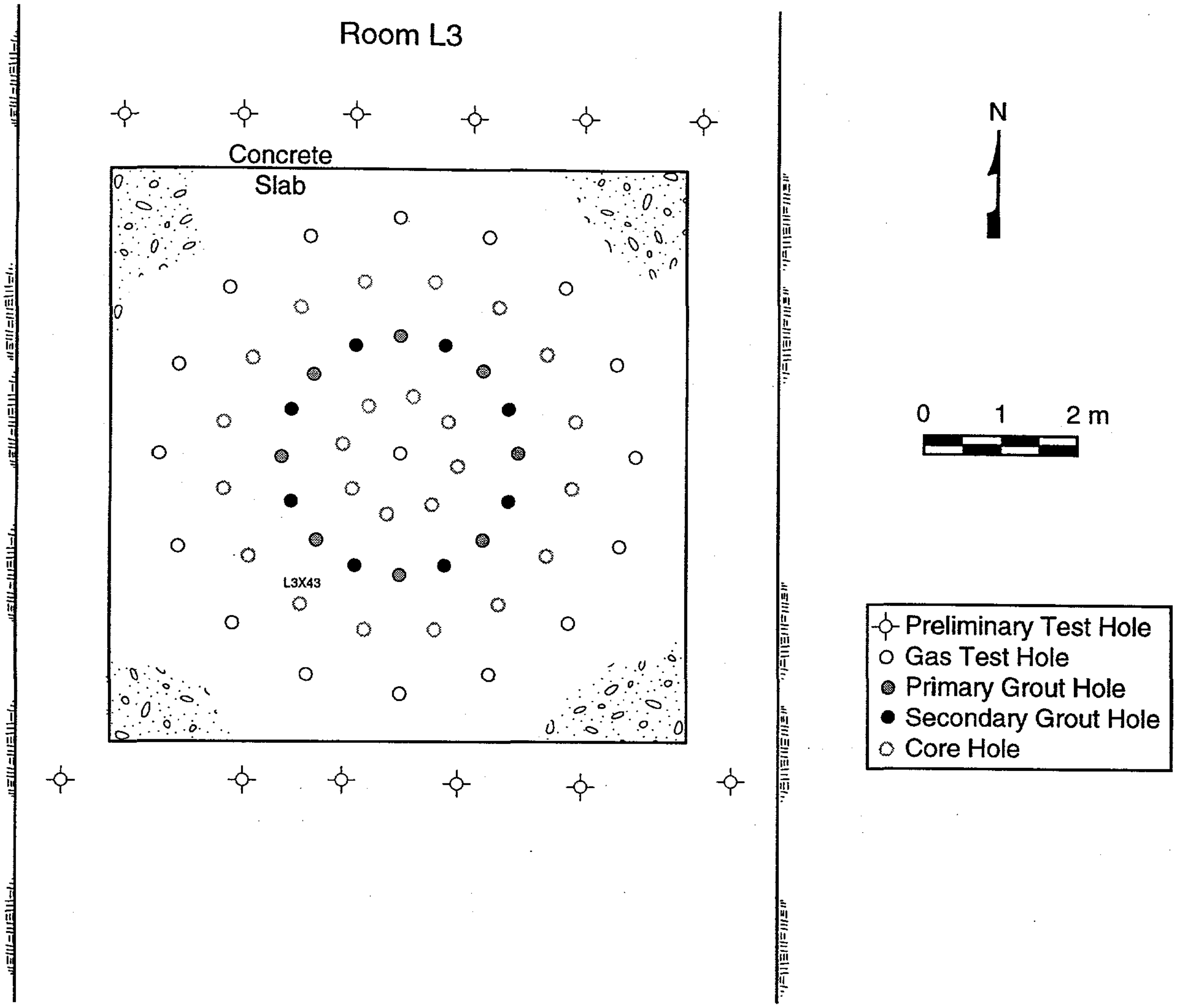

Figure 2. Plan view of Room L3 showing location of concrete slab and test holes.

"sniffer" holes, the breakthrough time and the shape of the breakthrough curve were recorded. Results indicated that primary grouting, at pressures less than 3.4 MPa, decreased air transmissivity of the DRZ by approximately three orders of magnitude. Secondary grouting at pressures as high as high as 6.9 MPa accomplished little additional reduction.

Post-grouting examinations of coreholes by downhole color television, and of drill core by scanning electron microscopy (SEM) and energy dispersive x-ray (EDX), showed that the grout routinely penetrated and sealed fractures as small as 8 microns. In one rare instance a 3-micron fracture was sealed. Figure 3 shows an SEM photomicrograph of a completely grout-filled 3-micron fracture in a core sample taken from hole L3X43 (see Figure 2).

4 DEVELOPMENT AND OPTIMIZATION OF DRY PULVERIZED ULTRAFINE GROUT

Simultaneous pulverization and mixing were suitable for the in situ test, but volume limitations

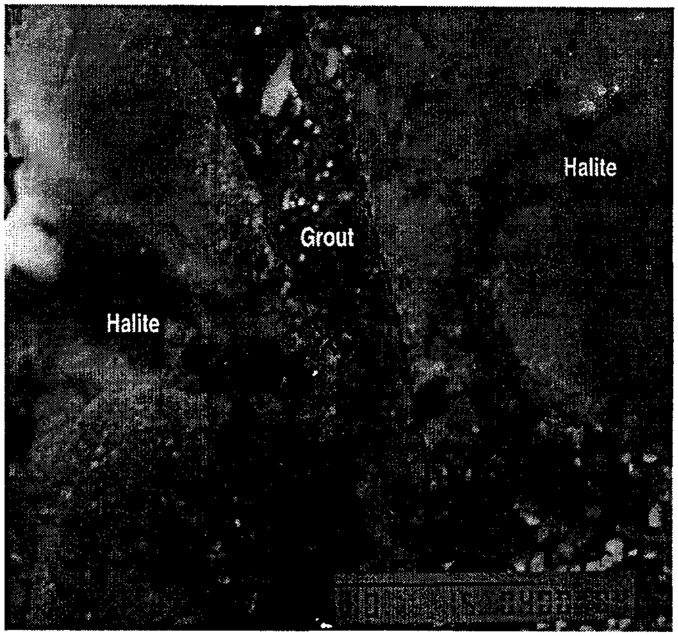

Figure 3. Electron microphotograph showing a grout-filled fracture 3 microns wide.

(approximately 27 gallons prepared every 105 minutes) preclude use of this method in normal industrial grouting. A need for preparation of the grout in dry form, ready for mixing, was evident. Subsequent to the in situ test, problems previously encountered with dry pulverization were solved, permitting the production of a dry ultrafine grout. Dry pulverization was even more efficient in particle size reduction than the wet process, resulting in particles 90% smaller than 6 microns and averaging 2 microns.

The attendant increase in surface area of the fine dry powder necessitated laboratory optimization. Twenty-three mixes were evaluated in order to minimize the W/C ratio and superplasticizer content. These efforts resulted in development of a grout possessing improved characteristics over the wet-pulverized grout that had been demonstrated in situ. Optimization of the dry-pulverized Ultrafine cementitious grout was conducted at WL (Onofrei et al., 1995).

Mechanical and rheological characteristics of the dry pulverized grout are comparable to the wet pulverized material, as shown in Table 1. Laboratory tests determined that the dry pulverized grout, injected at 0.7 kPa, penetrated interstitial spaces in sand as small as 3 microns in sand. The dry pulverized grout, which has been produced at a competitive cost, has been approved for use at the WIPP.

5 CONCLUSIONS

Wet and dry pulverization methods for preparation of ultrafine cementitious grouts have been developed. A process for simultaneous wet pulverization/mixing was initially developed in the laboratory. This grout was found to exceed most of the desired characteristics available with commercial grouts. Its deployment in the field at the WIPP site demonstrated adequate quality control, volume preparation, and performance. Building on the success of the wet process, dry-pulverized grout was developed with characteristics superior to the wet-pulverized grout. These grouts possess great promise for sealing applications at the WIPP as well as similar uses in environmental remediation, mining, construction, and concrete repair.

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Table 1. Grout Characteristics

Parameter	Pulverization	
	Wet	Dry
Water/Cement Ratio	0.5/1	0.6/1
Particle Size <10 microns	90%	——
Particle Size <6 microns	——	90%
Average Particle Size (microns)	4	2
Injectability (hours)	3	2
Set Time (hours)	14.5	6.75
Shrinkage (% in 50% relative humidity)	<1	<1
Compressive Strength (MPa at 28 days)	39.7	47.2
Hydrational Heat	ND*	30°C
Permeability (meters/second)	1×10^{-16}	1×10^{-16}

* ND = not determined.

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