COLLOIDAL SILICA GROUT SELECTION AND CHARACTERIZATION IN SUPPORT OF THE BARRIER DEPLOYMENT AT BROOKHAVEN NATIONAL LABORATORY

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ABSTRACT

Small and large-scale laboratory testing of colloidal silica grout was performed in support of the Brookhaven National Laboratory (BNL) Linear Accelerator Isotope Producer (BLIP) grouting project. The testing consisted of two phases: the selection of a colloidal silica variant that best reduced the saturated hydraulic conductivity of the native Brookhaven soils in column injection tests, and the injection of the selected colloidal silica into large sand tanks to determine the in situ saturated hydraulic conductivity of the grouted sand. Nine colloidal silica variants, having different colloid particle size ranges, were identified, injected into sand columns, and tested in a flexible wall permeameter. Results indicate that the selected colloidal silica grout was successful at reducing the saturated hydraulic conductivity of the BNL soils by four to five orders of magnitude. Additional soil columns were grouted to replicate the results and to develop soil-water characteristic curves for the grouted material for computer modeling parameters. The soil-water characteristic curves developed for the grouted soils were unique and supported the use of the colloidal silica grout to isolate activated soils in the vadose zone. Further testing was conducted with the selected colloidal silica by injecting it into sand tanks placed in a load cell to simulate injection at depth; and results indicate that the saturated hydraulic conductivity of the grouted sands was reduced by two to six orders of magnitude.

INTRODUCTION

The Brookhaven Linear Accelerator Isotope Producer produces radioisotopes, which are crucial to nuclear medicine for both research and clinical use. During operation, a linear accelerator generated proton beam impinges a target to produce the required medical isotopes. High-energy neutrons and protons created in the process pass through the target cooling water and into the surrounding soils. The particles are then absorbed by the soil creating radioactive isotopes from the naturally occurring elements in the soil surrounding the isotope producer tank. The two principle isotopes of concern are tritium and sodium 22, which are easily transported downward to the groundwater. A relatively small volume of activated soil lies beneath the isotope producer facility, approximately 7 meters below the foundation of the building and 8 meters above the water table. To minimize the transport of contaminants to the aquifer from infiltrating atmospheric water, flow through the activated soils in the vadose zone needed to be reduced.

Surface water management improvements were implemented by BNL, once the contaminants were detected in the groundwater downstream of the isotope producer and further characterization indicated the isotope producer was the source of that contamination. However, the United States Environmental Protection Agency indicated that further actions were necessary to mitigate contaminant migration to the groundwater. Emplacement of a viscous liquid barrier was selected by BNL as the preferred alternative for preventing further groundwater contamination. Emplacement of a viscous liquid barrier involves the injection of a low viscosity grout into the soil matrix where it gels to form a barrier. The grout would be injected to encapsulate the activated soil zone, creating a region with reduced saturated hydraulic conductivity and altered water retention characteristics.

Once the preferred remedial alternative was identified, selection of an acceptable colloidal silica grout variant became important to the success of the project. A series of laboratory tests were conducted to select a colloidal silica that best achieved project objectives. Once a colloidal silica variant was selected, the project proceeded to the field at BNL where the colloidal silica grout was emplaced using downstage permeation grouting. The injection string was driven down in multiple locations to the top of the activation zone where a series of vertically overlapping grout bulbs were injected at predetermined depths into the activated soil. The barrier was designed to deliver the grout into the activation zone ensuring the activated soils were completely encapsulated.

The focus of this paper is on the laboratory testing needed to support the selection of the colloidal silica variant that best reduced the saturated hydraulic conductivity of the native BNL sands. Column injection tests were conducted to select the colloidal silica variant and additional sand tank testing was conducted to compare two injection designs having different diameter grout bulbs.

COLLOIDAL SILICA TES TING

Nine colloidal silica variants were selected for laboratory testing, based on percent solids, particle size, and particle size distribution. Several of the colloidal silica variants had narrow particle size ranges, while others had wider ranges of particle sizes. To provide unbiased results during the initial testing phase, the colloidal silica variants were labeled MSE 1 to MSE 9.

The colloidal silica variant testing involved a series of consecutive tests. The ultimate goal of this testing was to identify a single colloidal silica variant that achieved the highest reduction of saturated hydraulic conductivity in the BNL sandy soils. The following laboratory tests provided the systematic methodology for identifying the optimal colloidal silica variant. The colloidal silica variant tests included:

- colloidal silica variant drain-in tests,
- colloidal silica grout gel time determination,
- colloidal silica grout column injection, and
- colloidal silica grout saturated hydraulic conductivity testing.

Drain-in Tests

The drain-in tests were used to identify colloidal silica variants that gel prematurely in the presence of native BNL soil. In this test, clean BNL soils collected from near the isotope producer were packed into vertical columns, and each colloidal silica variant (without the electrolyte gelling solution) was poured into the top of the sand columns. The volume that flowed through each sand column was collected and measured. If the particular colloidal silica variant did not prematurely gel, the majority of the volume would flow through the sand column. Nearly the entire volume of each colloidal silica variant flowed through the sand columns; therefore all of the variants passed the test.

Gel Time Determination

The colloidal silica variants gelled by adding one part by volume electrolyte solution to five parts of each of the colloidal silica variants. A 90-minute State 2 gel time was determined by viscometer and jar tests for each of the colloidal silica variants. State 2 gel time viscometer readings correspond to a viscosity between 10 and 12 centipoise (cP). A State 2-gelation time is defined as a highly flowing gel that appears to be only slightly more viscous than the initial polymer solution. This gives the lab team 90 minutes to inject the colloidal silica grout before the liquid starts to become more viscous. After an additional 90 minutes, the grout will generally obtain a State 9 gel where it becomes a rigid gel with no gel-surface deformation upon inversion of the sample jar. The process of gelation for the jar tests was recorded by assigning gel states according to gel time states modified by Lawrence Berkeley National Laboratory (1).

Calcium chloride ($CaCl_2$) was used as the initial electrolyte solution for all nine variants. Two variants did not gel with the addition of the $CaCl_2$ electrolyte solution at any molarity used, so other electrolyte solutions with varying molarities were tried, without success. Therefore, these variants were eliminated from further laboratory testing because they did not gel.

Column Injection

Groups of four sand columns were injected with colloidal silica grout made from each of the remaining seven variants. The columns were packed with native BNL sand that was dried and re-wetted to duplicate the actual soil moisture conditions (5% by weight) found at the Brookhaven isotope producer facility. The sand in each of the columns was packed volumetrically to 90% of the Standard Proctor Test for the BNL soil. The column injection apparatus assembly is shown in Figure 1. Grout was injected into each column until 2.5-pore volumes of grout were collected in each of the overflow containers.

The grouted colloidal silica sand columns were visually inspected weekly to determine if the colloidal silica grout had strengthened or cured within the columns. It was noted that the columns grouted with colloidal silica variants with larger colloid particle size did not cure as fast as the sand columns grouted with colloidal silica variants having a smaller colloid particle size. This is because the larger colloid particle size variants produce a more stable silica sol and therefore take longer to cure (1). The columns were allowed to cure for 28 days before the saturated hydraulic conductivity tests were initiated.

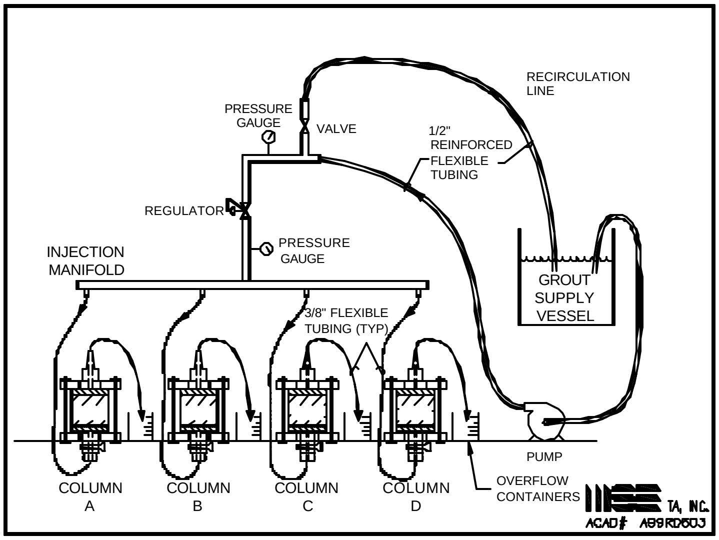

Fig. 1. Schematic of column injection apparatus

Table I. Geometric mean of hydraulic conductivity results

Grout Sample	Geometric Mean of Hydraulic Conductivity (cm/sec)
MSE 1 Summary	3.26E-05
MSE 2 Summary	3.21E-06
MSE 3 Summary	2.29E-06
MSE 4 Summary	9.71E-06
MSE 5 Summary	3.29E-05
MSE 6 Summary	3.20E-07
MSE 7 Summary	1.03E-04

Hydraulic Conductivity Testing

The hydraulic conductivity value for each sample was determined according to *ASTM D5084--Measurement of Hydraulic Conductivity of Saturated Porous Materials Using a Flexible Wall Permeameter.* The tests were performed using *Method C of D5084 (Falling Head--Increasing Tailwater Pressure)*. A summary of the hydraulic conductivity results obtained from flexible wall permeameter testing is shown in Table I. Tests were performed at low effective stresses to mimic a falling head test apparatus. During several of the tests, it was noted that grout flowed out of the samples with the permeameter water.

Before the data were analyzed, each data set was tested for outliers using the Extreme Value Test and DataQuest software. No outliers were identified. The geometric mean of each data set was calculated. During previous studies, the distributions of data from grouted sand samples have followed a log normal distribution. While there was not enough data to definitively determine whether the data in Table I were lognormally distributed, this assumption was made for comparing the data sets to determine the grout of choice. Therefore, the geometric mean defined the central tendency of the data for comparison purposes. Table I shows the grout designated as MSE 6 had the lowest geometric mean value for hydraulic conductivity.

After all of the initial laboratory tests were completed, a colloidal silica variant selection matrix was compiled. MSE 6 was selected to advance to the next phase of laboratory testing because it ranked highest of all the variants. MSE 6 or Nyacol NP6010 is a colloidal silica produced by Eka Chemicals Inc. – Paper Division. Two, 300-gallon tote bins of NP 6010 and additional containers of calcium chloride electrolyte solution were shipped to the MSE's testing facility in Butte, Montana to complete the colloidal silica grout optimization testing.

Additional Hydraulic Conductivity Testing

Further testing was performed on samples grouted with NP 6010 to replicate sample results. For the replicate tests, samples were prepared in smaller diameter columns, so they did not require trimming for hydraulic conductivity testing following the curing period. One extra set of the columns was injected with differing volumes of grout. The first column of the set was injected with 0.5 pore volumes of grout, the second with 1 pore volume, the third with 1.5 pore volumes, and the fourth with 2 pore volumes of grout. These columns were sent to the Montana State Soils Physics Laboratory for analysis and used to develop soil-water characteristic curves for computer modeling parameters.

Because more saturated hydraulic conductivity data points were available for grouted NP 6010 samples, a more complete analysis of the data was performed. A histogram of the hydraulic conductivity results is shown in Figure 2. The data appear to be lognormally distributed, as seen with previous data sets. The assumption of log

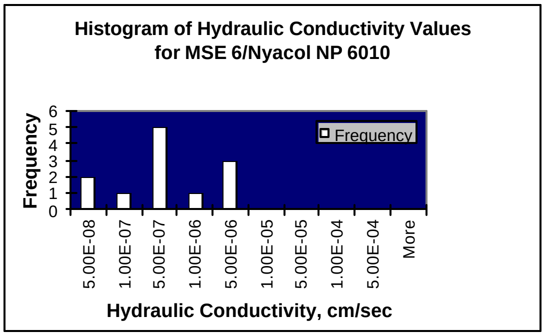

Fig. 2. Histogram of hydraulic conductivity

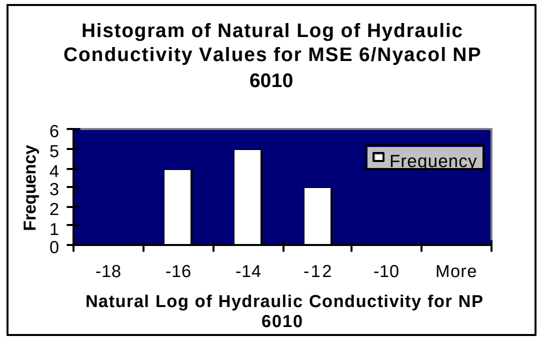

Fig. 3. Histogram of natural log values

normal distribution was verified using the DataQuest software. The data set was then log transformed and all subsequent data analysis was performed on the transformed data. Figure 3 is a histogram of the data following transformation. The high and low data points were analyzed using the Extreme Value Test. Neither the high nor the low values were statistical outliers. Table II is a summary of all flexible wall permeameter data for NP 6010.

As shown in Table II, the geometric mean of the entire data set is $2.9x10^{-7}$ cm/sec. The data set includes data from samples that were pushed out of columns and trimmed and those that were pushed out of columns and placed in the permeameter. Because sample-handling concerns exist, the data were also analyzed as distinct sets. Trimmed samples had a geometric mean of $2.5x10^{-7}$ cm/sec and samples not requiring trimming had a geometric mean of $3.2x10^{-7}$ cm/sec. These results indicate that hydraulic conductivity values were not detrimentally impacted by the trimming techniques used in the laboratory.

Table II. Summary of hydraulic conductivity sample results for NP 6010.

MSE 6 / NP 6010 Summary	Hydraulic conductivity (cm/sec)
Geometric Mean of Trimmed Samples	2.5E-07
Geometric Mean of Pushed Samples	3.2E-07
Overall Geometric Mean	2.9E-07

SAND TANK TESTING

After selecting the optimum colloidal silica variant, larger scale testing was conducted using specially constructed sand tanks. Three-dimensional sand tank testing was conducted to compare the hydraulic conductivity of the standard engineering design used during the viscous liquid barrier Cold Demonstration and a computer optimization-based design provided by Lawrence Berkeley National Laboratory (LBNL) after the completion of the viscous liquid barrier Cold Demonstration in 1997. The standard design specifies a 0.76-meter diameter grout bulb, while the computer optimized design specifies a 1.22-meter diameter grout bulb. A 1.22-meter tall by 1.22-meter diameter round steel tank was used for the injection of the smaller diameter grout bulb, while a 1.5-meter tall by 1.5-meter diameter round steel tank was used for emplacement of the larger diameter grout bulb. The LBNL model showed that a larger grout bulb would reportedly create a core area within the grout bulb that would achieve a saturated hydraulic conductivity of $1x10^{-7}$ cm/sec or less.

The bottom and sides of the tanks were lined with drain material, designed to intercept and direct grout in contact with the tank wall to drainage ports, therefore preventing artificial boundary conditions in the sand tanks. The tanks were then filled with BNL sand adjusted to 5% soil moisture by weight and compacted to within 90% of the Standard Proctor for the soil to simulate subsurface conditions.

Once the tanks were filled with soil, an injection lance was driven into the sand so that the middle of the injection ports was located in the center of each tank. At this point, the tanks were placed into a specially designed load cell and pressure was applied to the load cell plate to simulate down-hole conditions of 25 psi. Approximately 68 liters and 341 liters of grout were injected into the 1.22-meter and 1.5-meter diameter tanks, respectively. Figure 4 shows the 1.22-meter tank during placement of the load cell plate.

Standard Engineering Design

During the injection of the 1.22-meter tank, two standard laboratory samples were prepared using the colloidal silica grout; one sample was comprised of neat grout and the other was made by pouring sand into neat grout. Both samples were prepared and allowed to cure in a flexible wall permeameter membrane held in place by a sample mold. These samples were allowed to gel and form a mixture of what should represent the lowest limit of hydraulic conductivity for the NP 6010 colloidal silica grout. The standard laboratory samples were tested in the flexible wall permeameter according to *ASTM D5084, Method C*. As expected, the hydraulic conductivity of the neat grout sample was lower than the hydraulic conductivity of the sample with sand poured into the neat grout, 2.94×10^{-8} cm/sec and 5.33×10^{-8} cm/sec, respectively. These numbers are approximate since only one sample of each type was prepared.

Fig. 4. Placement of load cell plate on the 1.22-meter sand tank.

After injection of the 1.22-meter tank, the load and load cell plate were removed from the tank, the tank was moved out of the load cell, and the injection rod was removed from the tank. Shelby tube sampling was attempted but refusal was encountered at all of the sample locations.

Five Guelph permeameter tests were conducted on the 1.22-meter tank to measure the saturated hydraulic conductivity of the grouted material (Table III). As stated previously, a 76-cm diameter, or 38-cm radius, grout bulb was injected into the small sand tank. As shown in Table III, from the center of the tank, approximately 34 cm radially outward, there appears to be a core of well grouted sand with a saturated hydraulic conductivity ranging from 1.02×10^{-6} to 4.70×10^{-8} cm/sec. From 34 cm to 54 cm there appears to be a grout halo, where the grout mixes with the in situ pore water to form an area that is not completely grouted.

Table III. Saturated hydraulic conductivity results for the 1.22-meter test tank.

Test #	Vertical Location, cm below the sand surface	Horizontal Location, cm outward from the injection rod	Radial Distance, cm from injection point	Saturated Hydraulic Conductivity, cm/sec
1	58	38 North	38	2.30E-04
2	38	15 East	27	1.02E-06
3	61	25 North East	25	4.70E-08
4	56	53 West	54	6.92E-04
5	69	33 East	34	3.73E-07
Geometric Mean				4.91E-06

Computer Optimization-Based Design

Four Shelby tube samples were collected from the 1.5-meter test tank at different locations and depths. The Shelby tubes were pushed into the grouted sand immediately after the load cell plate was removed from the tank after injection. The Shelby tubes were collected during the excavation of the 1.5-meter tank and sent to Montana State University Soils Physics Lab for computer tomography (CT) scans. Areas of porosity reduction could be detected, as well as areas disturbed during the sampling process; however since the CT scans are qualitative not quantitative in nature, no numerical data was provided.

Two of the holes created by the Shelby tubes were utilized for in situ Guelph permeameter testing. Nine additional locations were tested for measurements of saturated hydraulic conductivity using Guelph permeameters. Table IV summarizes the in situ saturated hydraulic conductivity values obtained from the testing. As shown in Table IV, from the center of the 1.5-meter tank to approximately 30 cm radially outward there appears to be a core of well grouted sand with saturated hydraulic conductivities ranging from 4.80 x10^{-7} to 8.20 x 10^{-8} cm/sec. From 30 cm to 64 cm there is an area with saturated hydraulic conductivities ranging from 1.74 x 10^{-4} to 2.77 x 10^{-6} cm/sec, not including results from Test # 11. Results from Test # 11, located 33 cm directly below the injection tip, suggest that during downstage permeation grouting, lower areas beneath the injection tip do not get grouted until the rod string advances to the next injection interval.

Table IV. Saturated hydraulic conductivity results for the 1.5-meter test tank.

Test #	Vertical Location, cm below the sand surface	Horizontal Location, cm outward from the injection rod	Radial Distance, cm from injection point	Saturated Hydraulic Conductivity, cm/sec
1	78	53 West	54	3.28E-05
2	102	15 North	22	3.70E-07
3	91	30 South	31	4.80E-07
4	105	41 East	45	1.74E-04
5	71	56 North West	40	1.04E-05
6	112	56 South East	61	2.77E-06
7	104	46 North	58	3.18E-05
8	61	36 South West	44	3.29E-06
9	104	61 South	64	2.99E-06
10	61	16 North West	30	8.20E-08
11	119	0	33	1.44E-02
Geometric Mean				3.87E-06

This is implied by the saturated hydraulic conductivity value obtained during the test, since the saturated hydraulic conductivity of the ungrouted BNL soils ranges from 1 x 10^{-2} to 1 x 10^{-3} cm/sec. Because grouted sand was not encountered Test # 11, the test was not included in the geometric mean in Table IV.

CONCLUSIONS

Test Columns

The colloidal silica, NP 6010, selected after laboratory testing does reduce the saturated hydraulic conductivity of the sandy soil found beneath the Brookhaven Linear Accelerator Isotope Producer. The saturated hydraulic conductivity for the sand columns grouted with NP 6010 was reduced four to five orders of magnitude. In addition to reducing the saturated hydraulic conductivity of the BNL soils, NP 6010 proved to be a very stable silica that produced repeatable results in all phases of the laboratory testing. The consistency of this colloidal silica grout's behavior demonstrated during the laboratory testing would be a valuable asset during field emplacement.

Sand Tanks

The three dimension sand tank testing was conducted to compare the the saturated hydraulic conductivities of sand grouted according to the standard engineering design with sand grouted per the computer optimized based design. The larger grout bulbs specified by the computer based design were simulated by LBNL to create a grout bulb with a core having a saturated hydraulic conductivity of 1 x 10^{-7} cm/sec.

The Guelph permeameter tests conducted on the larger sand tank showed that this design did create a core approximately 60 cm in diameter with saturated hydraulic conductivities in the desired range. This 60 cm diameter corresponds to half of the diameter of the grout bulb size specified in the computer optimized design. The Guelph permeameter tests conducted on the smaller sand tank also demonstrated that the grout bulb had a core volume of reduced saturated hydraulic conductivity. The smaller grout bulb, with a design diameter of 76 cm, produced a core area within the bulb of approximately 68 cm in diameter. The geometric mean of saturated hydraulic conductivity for the 1.5-meter tank was 3.87 x 10^{-6} cm/sec, and the geometric mean of the saturated hydraulic conductivity for the smaller tank was 4.91 x 10^{-6} cm/sec.

Based on these results, it appears that either design achieves a reduction in saturated hydraulic conductivities. Both designs also created similar core areas. Although the larger computer optimized based design had more pore volumes pass through the core area, it did not appear to significantly lower the saturated hydraulic conductivity of the core area compared to the standard engineering design.

ACKNOWLEDGEMENT

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