August 29, 2012

MEMORANDUM TO: Gregory F. Suber, Chief
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FROM: Mathews George, Project Manager ***/RA/***
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SUBJECT: DRAFT LETTER REPORT, FISCAL YEAR 2012 MESO- AND INTERMEDIATE-SCALE GROUT MONOLITH TEST BED EXPERIMENTS

Summary

The U.S. Department of Energy (DOE) may rely on natural and engineered system properties that provide attenuation and retardation of radionuclide migration as part of its waste disposal system performance assessments. The NRC staff use information from independent analyses to support their consultation responsibilities for non-high-level, waste incidental to reprocessing (WIR) determinations and their monitoring responsibilities for subsequent actions taken under the National Defense Authorization Act for FY 2005 (NDAA). To this end, the Center for Nuclear Waste Regulatory Analyses (CNWRA) has been tasked to provide mechanistic information on the physical and chemical degradation of cementitious waste forms that are used for the isolation and containment of radioactive wastes and to evaluate the potential for radionuclide bypass of the engineered barriers via preferential or fast flow pathways. Previous reviews of DOE waste determinations (WD) indicated that potential fast flow pathways going through and bypassing barriers may dominate waste release from large, grout-filled tanks and vaults. Thus, macrocrack density and connectivity may play major roles in the release of radionuclides from *in-situ* tank closures and monolithic waste forms. Review of experimental and observational data on mass transport properties of cementitious materials did not reveal empirical data from which to estimate the likely properties of macrocracks that may develop in large grout monoliths. Most crack characterization measurements to date have been made on small-scale laboratory specimens using construction materials with significantly different formulations than those DOE proposed for radioactive waste disposal at NDAA facilities.

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Additionally, no DOE or third-party data are available to assess the significance of annular gaps that may develop between cementitious grout and internal tank fixtures (e.g., interior tank walls, pipes, and cooling coils). Lack of relevant data represents a key uncertainty when evaluating DOE WDs that rely on cementitious grout integrity to meet the performance objectives for low-level waste found in 10 CFR Part 61, Subpart C.

To establish a base-level understanding of the potential for fast flow cracks and annular gaps to form soon after grout is emplaced in a waste tank, CNWRA staff developed mesoscale grout monolith specimens in fiscal year 2009 at Southwest Research Institute) facilities in San Antonio, Texas, as physical analogs to grouted tanks at NDAA facilities. Twelve mesoscale specimens were constructed in 55-gal drums and also in a 3 m[10 ft]-radius by 30° sector to help inform development of conceptual designs for larger scale grout monolith experiments. The mesoscale grout monolith specimens, which were constructed using grout formulations similar to those being considered for use at Savannah River Site (SRS and INL, were instrumented permitting (i) observation and quantification of the effects of macrocracks, annuli between grout and internal fixtures or container walls, and lift separations that may develop due to thermal contraction cracking, shrinkage, and partial curing of the grout during the interval between lift emplacements, and (ii) the conduct of gas permeability tests.

These experiments led to the design, placement, testing, and analysis of an intermediate-scale grout monolith housed in a steel tank during Fiscal Years (FY) 2010 and 2011. The monolith is 6.1 m [20 ft] in diameter and nominally 0.8 m high, and is composed of a reducing grout that is very similar to the SRS-like Alternative 1 Reducing Grout specimens that Walter, et al. (2009) developed and described. Grout was transferred from the delivery trucks to the intermediate-scale grout monolith tank using a boom pump truck with discharge line typically positioned near the center of the tank. Distribution of the grout within the tank was by gravity and not by vibration. During the current FY12, testing and analysis of mesoscale drum specimens and the intermediate-scale grout monolith specimen continued, including initial pneumatic testing and borescopic observations of FY 2010 drum grout specimens and pneumatic injection retesting of FY 2009 drum grout specimens to characterize pneumatic property evolution with time. The FY 2010 intermediate-scale grout monolith testing consisted of (i) pneumatic injection retesting of four coreholes and three embedded pipes to identify pneumatic property evolution with time, (ii) borescopic observations of all nine corehole, and (iii) three-color fluorescent-dye tracer slug testing to identify interconnected fast pathways and seeps and acquire pressure head data fit for calculating the hydraulic properties of the grout surrounding injection coreholes. Near-surface (tan) and deeper subsurface (gray) grout samples from the intermediate-scale grout monolith were also analyzed using X-ray diffraction to determine whether and how mineralogical compositions status are reflected in the observed depth-dependent matrix color variations.

Conclusion

In response to questions regarding the representativeness of the local San Antonio, Texas-based grout models relative to grouts that DOE has proposed for NDAA tank closures, CNWRA concluded that it is clear that from a pneumatic property perspective, Local Reducing Grout is a good physical analog of South Carolina Reducing Grout because these specimens had nearly identical permeabilities. Although more air was entrained in the Local Reducing Grout with its ADVA 380 high range water reducer, the entrained air did not translate to an interconnected

porosity that would have significantly increased the permeability of the specimen. Instead, the slightly higher permeability of Local Reducing Grout may be due to its coarser sand grain size distribution. Different high range water reducers require unique proportioning to obtain equivalent flow characteristics, but these two sand-only reducing grout specimens were developed with different high range water reducers at the same dose, which likely led to a significant difference in the amount of water required to produce the desired slump flow, and further led to differences in the peak temperature.

Grout volumetrically contracts and its porosity and permeability evolve during curing. CNWRA observed cracks on mesoscale drum grout surfaces that were not apparent shortly after grout placement, and also observed that grout lobe seams are now actively conducting gas in some specimens. Mesoscale grout monolith pneumatic data suggest that injection pipe and drum wall annulus apertures and grout permeability generally increase with time. Annulus aperture expansion with cure time and the presence of conductive grout lobe seams suggest that poor bonding will occur between grout and NDAA steel tank liners and any internal fixtures such as cooling coils, as well as between grout lifts, lobes, and lobe seams. CNWRA indicated that such shrinkage could be mitigated by development of a shrinkage compensation grout formulation, but DOE has postponed efforts to address this issue.

Development of cracks within a grout monolith is a complex process that is not easy to resolve, but our knowledge base is continuing to improve as a result of these test bed experiments. Based on the latest set of experiments, CNWRA concluded that crack growth is generally progressive, meaning a crack is not necessarily one specific age but may have a spectrum of ages along its length and width. Crack development can vary from continuous lengthening to episodic growth (abrupt lengthening events separated by periods of no growth) depending on the evolution of the stress field, and the final crack geometries do not necessarily allow a unique development history to be determined. When shear offset or faulting occurs, dating the relative age of cracks based upon their termination against other cracks becomes even more uncertain. Cracks may or may not continue to develop in the long term.

CNWRA also performed mineralogical and elemental analysis to study a matrix color change from tan in the near surface of the intermediate-scale grout monolith to gray at greater depths. This color transition was very distinctive when the cores were first removed, but grew less distinctive with time. The causes of the matrix color change and the observed time-dependent fading remain unclear (i.e., analyses did not reveal any obvious mineralogical variation that could account for the color change).

Path Forward

The behavior and characteristics of the intermediate-scale grout monolith indicate that relatively large grout specimens are required to understand the flow behavior and cured properties of the grout that may be placed in the NDAA tanks. Further characterization of the existing intermediate-scale grout monolith and development and testing of a new intermediate-scale grout monolith that is consistent with DOE's final grout formulation for SRS Tanks 18-F and 19-F are recommended next. Methods include the utilizing the following approach:

- Computer-Aided Drafting Model of Intermediate-Scale Grout Monolith.
- Concrete Radar Imaging of Internal Lift Topography and Structures.
- Modeling and Analysis of Slug Tests in the Intermediate-scale Grout Monolith.
- Horizontal Coring of the Intermediate-Scale Grout Monolith.
- Core Description and Thin Section Analysis.
- Numerical Modeling to Understand Crack Development.
- Dissection of the Intermediate-Scale Grout Monolith.
- Pipe Grout Pneumatic Injection to Determine Annulus Apertures within Grouted Pipes.
- Acoustic Emission Monitoring of Crack Initiation in Grout Monoliths.
- Grout Adhesion and Bonding.

Looking ahead, staff recommends NRC consider developing a new intermediate-scale grout monolith by in the upcoming FYs to consider the following:

- Investigate the potential for cracking, shrinkage, and maximum temperatures attained in a large physical model comprising materials and material quantities consistent with the actual NDAA grout formula that is being used to stabilize F-Area waste tanks.
- Investigate the effect of minimized diurnal heating and cooling (as expected in an NDAA waste tank) on grout–tank wall bonds, perhaps by constructing the new monolith in an indoor facility.
- Investigate the effect of humidity evolution on the timing of early plastic shrinkage and later hydration and drying shrinkage crack formation (where the timing of formation is diagnostic of formation mechanism) by monitoring the in-tank environment with humidity, optical and acoustic emission sensors.
- Monitor for crack formation using acoustic emission and surface-observing cameras, given that timing is diagnostic of crack formation mechanisms.

Finally, when additional experimental grout monoliths are constructed under this program, NRC staff recommends that separate pours within each lift be dyed different colors to aid identification, because some lift separations and hiatuses between pours are difficult to identify in cores. CNWRA staff does not currently have enough information to indicate whether thermal cracking of grout should be expected in an NDAA tank. CNWRA staff recommend NRC consider a scope of work that include: (i) measuring the evolution of grout thermal conductivity; (ii) measuring the heat generation rate of grout, and performing numerical modeling and model

calibration of a new intermediate-scale grout monolith to achieve the measured temperature distribution; and finally (iii) upscaling the model to an NDAA-tank scale to ascertain whether sufficient thermal-cracking-magnitude temperature gradients could be realized in an NDAA tank.

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