U.S. Department of Energy
Office of Civilian Radioactive Waste Management
Office of Science and Technology and International

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10/01/04

OCRWM Science and Technology Program Cementitious Materials Technologies

This potential project will develop and test cost effective cementitious materials for construction of Yucca Mountain (YM) invert, drift liners, and bulkheads. These high-silica cementitious materials will be designed to buffer the pH and Eh of the groundwater, to slow corrosion of waste packages (WP), and to retard radionuclide migration. While being compatible with YM repository systems, these materials are expected to be less expensive to produce, and as strong, and more durable than ordinary Portland Cement (OPC). Therefore, building out the repository with these cementitious materials may significantly reduce these costs and reduce uncertainty in short- (<2,000 yr) and long-term (>10,000 yr) repository performance.

Both laboratory development and natural analog studies are anticipated using a unique combination of expertise at ORNL, UT, UC Berkeley, and Minatom to develop and test high-silica hydraulic, cementitious binders for use at YM. The major tasks of this project are to (1) formulate and make candidate cementitious materials using high-silica hydraulic binders, (2) measure the physical and chemical properties of these materials, (3) expose combinations of these materials and WP materials to static and flowing YM groundwater at temperatures consistent with the expected repository conditions, (4) examine specimens of both the cementitious materials and WP materials periodically for chemical and mineralogical changes to determine reaction mechanisms and kinetics, and (5) predict the long-term performance of the these material by thermodynamic and transport modeling and by comparisons with natural analogs.

High-silica binders have a number of advantages for YM applications. Evidence from archaeological and geological samples shows that high-silica cement binders are extremely durable. These cements, used by the Greeks and Romans, have survived for hundreds of years in hot water and for thousands of years in marine environments. As an additional benefit, high-silica binders are as strong as OPC, while suppressing the formation of free calcium hydroxide that elevates the pH in OPC.

High-silica binders also cost 30-50% less. Furthermore, stoichiometric-excess silica in these binders seals stress cracks in structures by forming alteration-minerals. Leachates from these cementitious binders are nearly saturated in orthosilicates [$\text{SiO}_4\text{H}_2^{+4}$] that passivate metal surfaces. These silicate minerals are also very strong radionuclide absorbers, resulting in very insoluble silicates of actinides, fission products, and uranium.

Candidate concrete binders will include pozzolans, trass, blast furnace slags and other high-silica, hydraulic cements. Static and flow through (dynamic) tests will be used to study interactions between candidate cementitious materials, YM groundwater, and WP materials at temperatures bracketing the expected repository service conditions (30-160°C). These tests will run up to 2 years, during which the chemistry of the solid and liquid phases and the mineralogical changes in the cement will be monitored to determine mechanisms and kinetics of alterations and formation of corrosion products. Petrographic and other analyses and microscopy imaging will be used to detect and characterize the phases that form due to interaction with groundwater. Water will be isotopically labeled, allowing isotopic and elemental imaging using secondary ion mass spectrometry to track the migration of the fluid into the solid samples at the micro- and nanoscale. These techniques will be coupled with inductively-coupled plasma mass spectrometry to monitor the composition and pH of the groundwater. This will determine the mechanisms and rates of elemental transport during the alteration and corrosion of these phases.

To assess the long-term stability of the candidate cementitious materials, the study will characterize effects of weathering and alteration of natural and man-made analogs of the cementitious matrices proposed for YM. Initially, the proposed project will examine ancient Roman pozzolans, which have been in a variety of terrestrial and marine environments for over 2,000 years. Next, natural samples from sanidine-facies metamorphic rocks in Marble Canyon, Texas, and the Hatrurim formation in Israel will be examined. These contain many of the same phases that form in cements. These latter examples have been exposed to weathering for millions of years. For example, the minerals hatrurite, larnite, and their polymorphs exist in these rocks and are natural analogs for the common cement-clinker phases "alite" (Ca_3SiO_5) and "belite" (Ca_2SiO_4). These cementitious analogs and their alteration products provide the opportunity to study transport processes and durabilities on geologic time scales.