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The distribution of zeolites and their effects on the performance of a nuclear waste repository at Yucca Mountain, Nevada, U.S.A.

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ABSTRACT

Yucca Mountain, Nevada, has been approved by the U.S. Congress as the site of the first high-level radioactive waste repository in the U.S., paving the way for the Department of Energy to submit a license application to the Nuclear Regulatory Commission. The U.S. concept of a radioactive waste repository involves both engineered (man-made) barriers and geologic barriers to minimize the long-term migration of waste from the repository. The minerals at Yucca Mountain are an integral part of the geologic barrier to the migration of radionuclides, and understanding the distribution of reactive and potentially sorptive minerals is crucial to modeling the long-term performance of the repository. We have determined the three-dimensional distribution of minerals at Yucca Mountain by analyzing a number of drill cores using quantitative X-ray diffraction methods. Clinoptilolite and mordenite are locally abundant in the rhyolitic tuffs in both the unsaturated and saturated zones. They formed by the alteration of glassy rhyolitic tuffs, and their occurrence is stratified, often following the distribution of nonwelded, initially vitric tuffs near and below the water table. Because of their large cation-exchange capacities, these minerals have traditionally been considered to be important in retardation of the migration of cationic radionuclides such as ^{137}Cs and ^{90}Sr. However, analysis of radionuclides that may be contained at Yucca Mountain reveals that, for the most part, the radioactive species of concern are not those that would be readily sorbed by natural zeolites. Although clinoptilolite and mordenite are highly selective for Cs and Sr, the half-lives of these radionuclides (~30 years) are sufficiently short that the man-made containment at any radioactive waste repository is expected to outlive these radionuclides. The most important radionuclides are thus anionic species and long-lived actinides that often form large, complex aqueous species, both of which are not strongly sorbed by cation exchangers such as zeolites. However, zeolites play an important role in a repository at Yucca Mountain for several other reasons. Emplacement of radioactive waste will change the temperature and P_{H_2O} conditions in large volumes of rock, and zeolites will be important sources and sinks of water and thermal energy during heating and cooling in the vadose zone. In addition, dehydration/hydration reactions are accompanied by volumetric changes that can affect macroscopic rock properties. Longer-term reactions, e.g., of clinoptilolite to analcime, would give rise to larger volume reductions and production of water and silica. These studies show that the importance of zeolites extends far beyond cation exchange by zeolites to phenomena affecting the entire thermohydrologic system.