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The effect of Cs on the structural and hydrothermal stability of barium hollandites

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Hollandite, based on Ba and Ti, has been shown to be an ideal medium by which active ^{135}Cs and ^{137}Cs can be incarcerated. Diffraction measurements have shown that addition of Cs modifies the symmetry of the material, from monoclinic to tetragonal. Experiments carried out here show the effects of Cs on two hollandite systems based on Ba-Al-Ti and Ba-Mg-Ti.

Studies based on the dissolution of hollandite in different solutions under extreme conditions with increasing Cs content are considered from a chemical viewpoint. Structural measurements are reported for neutron diffraction, MAS NMR, and electron microscopy.

1.8.13

Effects of volume swelling in crystalline host phases for nuclear waste immobilization

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The effects of radiation damage in potential crystalline host phases for immobilization of nuclear wastes have been studied using a combination of experimental and theoretical techniques. First principles calculations are used to parametrize interatomic potentials for molecular-dynamic simulations of energetic collision cascades. These empirical large-scale simulations give information on the effects of the damaged region on the volume swelling, and the microstructural properties of the material. The stabilities of the resulting structures are analyzed again with first principles methods, including analysis of the changes in the physical properties (energetics, IR, Raman and NMR spectra) resulting from high concentrations of point defects in the remaining crystalline phase. As a case study, we have examined the role of defects on the lattice and total volume swelling of zircon. This mineral is known to exhibit anisotropic lattice expansion ($\Delta a/a \sim 1.5\%$, $\Delta c/c \sim 2\%$, and $\Delta V/V \sim 5\%$) and total volume swelling of about 18% as a consequence of the crystalline-amorphous transformation. Our results for the crystalline phase indicate that the primary defects responsible for the observed changes might be interstitials of O and Si, and the antisite of Zr in Si. In the amorphous phase, the damage correlates with the increased leaching rate. These, are being experimentally explored for several candidate ceramic forms.