Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

Thermodynamic stability of radiogenic Ba in $CsAlSi_2O_6$ pollucite[1] JOHN JAFFE, Pacific Northwest National Laboratory (retired), RENÉE VAN GINHOVEN, WEILIN JIANG, Pacific Northwest National Laboratory — Pollucite, a zeolite-like nanoporous aluminosilicate structure with nominal composition $CsAlSi_2O_6$, has been suggested as a nuclear waste storage form for fission-product radioactive isotopes of cesium, especially ^{137}Cs. One factor affecting the long-term stability of this waste form is the valence change associated with the beta decay that converts Cs into barium. We have used first-principles density functional total energy calculations to evaluate the thermodynamic stability of pollucite with Ba replacing Cs at regular lattice sites with respect to the precipitation of Ba, Cs or their oxides. We included small clusters of substitutional Ba_{Cs} as well as localized complexes of Ba_{Cs} with compensating electron donor defects, specifically Cs vacancies and interstitial oxygen. We conclude that Cs-Ba pollucite is thermodynamically stable against precipitation of Cs or its oxide, but that partial precipitation of Ba or BaO may be thermodynamically favored under some conditions. Even this change may be kinetically limited, however.

[1]Fuel Cycle Research and Development, U.S. Department of Energy Waste Form Campaign

John Jaffe
Pacific Northwest National Lab (retired)

Date submitted: 03 Dec 2012 Electronic form version 1.4