The Future of Radionuclide Retention in Fukushima Soils from First Principles Simulations

K.M. ROSSO[1*], M. SASSI[1], M. OKUMURA[2], S. KERISIT[1], M. MACHIDA[2]

[1]Pacific Northwest National Laboratory, Richland WA, USA

[2]Japan Atomic Energy Agency, Kashiwa, Chiba, Japan (*correspondence: kevin.rosso@pnl.gov)

The fate of accidentally dispersed radionuclides such as ^{137}Cs and ^{90}Sr in the environment, such as at Fukushima, is initially determined by their strong and selective cation exchange into the interlayer spaces of clay minerals such as vermiculite. Contaminant migration is then assumed to be a problem of slow hydrophysical transport of clay particulates through watersheds. However, on decadal time scales the interplay and durability of radioisotope-clay association is not well known. Our team is filling this knowledge gap using first principles simulations to systematically examine the effects of the radioisotope transmutations $^{137}Cs^{+} \rightarrow {}^{137}Ba^{2+}$ and $^{90}Sr^{2+} \rightarrow {}^{90}Y^{3+} \rightarrow {}^{90}Zr^{4+}$ and radiation damage from associated β emissions on the retention of remnant parent isotopes in clay interlayers. For example, density functional theory calculations of threshold displacement energies show that the probability for ^{137}Cs and ^{90}Sr to create Frenkel defects at octahedral Mg sites in vermiculite is about 20%. However, for ^{90}Sr, the follow-on decay of ^{90}Y contributes a further probability of about 85%. The total free energy increase associated with defect accumulation could become significant, ultimately converting the host crystallite locally or globally to phases with lower radioisotope retention. Ongoing modeling will address this long-term evolution of ^{137}Cs and ^{90}Sr cation exchange capacity, an otherwise difficult task for experiment. Our research will build an information database that should be useful for risk assessment, for reducing uncertainties in long-term projections, and for guiding management, disposal and treatment of contaminated soils in the Fukushima area.