Pressure- and temperature-induced transformation of natrolite to sequester Cs and Pb

J. IM,[1] D. SEOUNG[2], Y. LEE[1], S. Y. LEE[3], D. A. BLOM[4], T. VOGT[4], C.-C. KAO[2] AND Y. LEE[1]*

[1]Department of Earth System Sciences, Yonsei University, Seoul 03722, Korea (*correspondence: yongjaelee@yonsei.ac.kr)

[2]Stanford Synchrotron Radiaition Lightsource, SLAC National Accelerator, Menlo Park, CA 94025, USA

[3]Division of Radwaste Disposal Research, Korea Atomic Energy Research Institute, Daejeon 34057, Korea

[4]NanoCenter & Department of Chemistry and Biochemistry, University of South Carolina, Columbia, SC 29208, USA

We report here a pressure- and temperature-driven transformation to sequester environmentally toxic cations such as Cesium (Cs^+) and Lead (Pb^{2+}). Ag-exchanged natrolite and Pb-exchanged natrolite are pressurized in an aqueous CsI solution and pure water as a pressure-transmitting medium (PTM), respectively. Ag-NAT with CsI solution PTM results in the exchange of Ag^+ by Cs^+ in the natrolite framework forming Cs-NAT-I and, above 0.5 GPa, its high-pressure polymorph (Cs-NAT-II). During the initial cation exchage, the precipitation of AgI Occurs. Additional pressure and heat at 2 GPa and 160 °C transforms Cs-NAT-II to a pollucite-related, highly dense, and water-free triclinic phase. At ambient temperature after pressure release, the Cs remains sequestered in a monoclinic pollucite phase at close to 40 wt% and favorably low Cs leaching rate under back-exchange conditions [1]. In the case of Pb-NAT in pure water PTM, four high-pressure polymorphs of natrolite (Pb-NAT-I, II, II′, III) and one reconstructive transformation are through the pressure-induced hydration (PIH). Heating at 200 °C and 4.5 GPa, leads to a denser lawsonite phase with orhorhombic space group which is recovered after pressure release. The recovered lawsonite phase has much stable Pb coordination environment in the natolite channel compared to that in the original Pb-NAT and its high-pressure polymorphs [2]. The structures of the recovered Cs- and Pb-sequestered phases are confirmed by HAADF-STEM real spacing imaging.

[1] Im *et al*. (2015) *Environ. Sci, Technol* **49**, 513-519. [2] Im *et al*. (2016) Dalton Trans **45**, 1622-1630