Hydrothermal Reactions of Clay Minerals and Shales with Cesium Phases from Spent Fuel Elements

Sridhar Komarneni and William B. White[1]

Materials Research Laboratory, The Pennsylvania State University, University Park, Pennsylvania 16802

[1] Also associated with the Department of Geosciences, The Pennsylvania State University, University Park, Pennsylvania 16802.

Abstract: The immobilization of soluble Cs from spent fuel elements by ion exchange and direct chemical reaction with clay minerals or shales was investigated under hydrothermal conditions. Various clay minerals or shales were reacted with likely Cs sources and water at 300 bars pressure and 100 ° , 200 ° , and 300 ° C for 4, 2, and 1 months, respectively. Pollucite was the principal product, but $CsAlSiO_4$ was also observed, along with unreacted or hydrothermally altered aluminosilicates. From Cs concentrations of the product solutions partition of Cs between liquid and solids was found to vary depending on the Cs source, the clay or shale phase, temperature, and run duration. For example, illite-Cs_2MoO_4 interactions resulted in 19, 32, and 95% fixation of added Cs at 100 ° , 200 ° , and 300 ° C respectively. Fixation of as much as 97% of the Cs in some solids was observed. In addition to Cs-aluminosilicates, Cs was fixed on cation-exchange sites by interlayer collapse in montmorillonite. Reactions with Cs_2MoO_4 also produced powellite because Ca was available in the reaction mixture. The U^{6+} from β-$Cs_2U_2O_7$ was reduced to form uraninite by sulfide- and/or organic-rich shales. $(Cs,Na)_2(UO_2)(Si_2O_5)_3 \cdot 4H_20$, an analog of weeksite, was produced in reactions with β-$Cs_2U_2O_7$. The reaction products pollucite and uraninite can immobilize much of the Cs and U from spent fuel elements because Cs in pollucite is extremely difficult to exchange and U in uraninite is insoluble.

Key Words: Cesium • Hydrothermal • Illite • Nuclear waste • Pollucite • Shale • Uranium

Clays and Clay Minerals; August 1981 v. 29; no. 4; p. 299-308; DOI: 10.1346/CCMN.1981.0290408

Clay Minerals Society (www.clays.org)