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Radiation-induced decomposition of U(VI) alteration phases of UO_2

Satoshi Utsunomiya and Rodney C. Ewing

[a]Department of Geological Sciences, University of Michigan, Ann Arbor, MI 48109-1063, USA

Abstract

U^{6+}-phases are common alteration products of spent nuclear fuel under oxidizing conditions, and they may potentially incorporate actinides, such as long-lived ^{239}Pu and ^{237}Np, delaying their transport to the biosphere. In order to evaluate the ballistic effects of α-decay events on the stability of the U^{6+}-phases, we report, for the first time, the results of ion beam irradiations (1.0 MeV Kr^{2+}) for six different structures of U^{6+}-phases: uranophane, kasolite, boltwoodite, saleeite, carnotite, and liebigite. The target uranyl-minerals were characterized by powder X-ray diffraction and identification confirmed by SAED in TEM. The TEM observation revealed no initial contamination of uraninite in these U^{6+} phases. All of the samples were irradiated with *in situ* TEM observation using 1.0 MeV Kr^{2+} in the IVEM (intermediate-voltage electron microscope) at the IVEM-Tandem Facility of Argonne National Laboratory. The ion flux was 6.3×10^{11} ions/cm^2/sec. The specimen temperatures during irradiation were 298 and 673 K, respectively. The Kr^{2+}-irradiation decomposed the U^{6+}-phases to nanocrystals of UO_2 at doses as low as 0.006 dpa. The cumulative doses for the pure U^{6+}-phases, e.g., uranophane, at 0.1 and 1 m.y. are calculated to be 0.009 and 0.09 dpa using SRIM2003. However, with the incorporation of 1 wt.% ^{239}Pu, the calculated doses reach 0.27 and ~1.00 dpa in ten thousand and one hundred thousand years, respectively.

Under oxidizing conditions, multiple cycles of radiation-induced decomposition to UO_2 followed by alteration to U^{6+}-phases should be further investigated to determine the fate of trace elements that may have been incorporated in the U^{6+}-phases.

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