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The effect of ionizing radiation on U^{6+}-phases

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U^{6+}-minerals commonly form during the alteration of uraninite and spent nuclear fuel under oxidizing conditions. By the incorporation of actinides and fissiogenic elements into their structures, U^{6+}-minerals may be important in retarding the migration of radionuclides released during corrosion of spent nuclear fuel. Thus, the stability and the structural transformation of the U^{6+}-minerals in radiation fields are of great interest. In this study, in order to examine the effects of ionizing radiation on U^{6+}-minerals, electron irradiations (~8-33 x 10^{17} ($e^-/cm^2/sec$) at the room temperature) were completed on:

boltwoodite, $K[(UO_2)(SiO_3OH)](H_2O)_{1.5}$
kasolite, $Pb[(UO_2)(SiO_4)](H_2O)$
saléeite, $Mg[(UO_2)(PO_4)]_2(H_2O)_{10}$
carnotite, $K_2(UO_2)_2(V_2O_8)(H_2O)_3$
liebigite, $Ca_2[(UO_2)(CO_3)_3](H_2O)_{11}$
schoepite, $[(UO_2)_8O_2(OH)_{12}](H_2O)_{12}$.

During the irradiation, boltwoodite, saléeite, and carnotite became amorphous at doses of 1-4 x 10^{10} gray (Gy), while the amorphization dose (D_c) of kasolite, $50x10^{10}$ Gy, was about an order of magnitude higher than that of boltwoodite. This high D_c for kasolite is consistent with the hypothesis that the D_c increases as the mass of the inter-layer cation increases. Only amorphization, rather than chemical decomposition, occurred in boltwoodite, saléeite and carnotite, even at doses as high as $80x10^{10}$ Gy. In contrast, uraninite nanocrystallites began to form with a random orientation at ~$43x10^{10}$ Gy in liebigite that had already become amorphous prior to irradiation in the vacuum of the TEM. For schoepite, the D_c was only $0.51x10^{10}$ Gy, and randomly oriented uraninite nanocrystallites formed at $7.8x10^{10}$ Gy, which is approximately the same dose as compared with the D_c of the other U^{6+}-phases. Because the predicted cumulative dose by the ionizing radiation in spent nuclear fuel is ~10^7-10^8 Gy during the first 10^{2-3} years after discharge, an additional contribution of ionizing radiation dose by α-decay events is needed in order to induce amorphization in these U^{6+}-phases.