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The effect of ionizing radiation on uranophane

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ABSTRACT

The susceptibility of uranophane, a uranyl sheet silicate, ideally $Ca(UO_2)_2(SiO_3OH)_2(H_2O)_5$, to ionizing irradiation has been evaluated by systematic irradiations with 200 keV electrons over the temperature range 94 to 573 K. High-resolution transmission electron microscopy revealed that amorphous domains formed locally, concurrently with a gradual disordering of the entire structure. Amorphization doses at room temperature were 1.1×10^{10} Gy for uranophane, 1.3×10^{10} Gy for Sr-substituted uranophane, and 1.9×10^{10} Gy for Eu-substituted uranophane; thus, there was an increase in amorphization dose with increasing average atomic mass. At 573 K, the amorphization dose of uranophane was 2.0×10^{11} Gy. The temperature dependence of the amorphization dose of uranophane has two stages; ≤413 K and >413 K. Based on a defect accumulation model, the effective activation energies for amorphization at each stage are 0.0440 eV and 0.869 eV, respectively. This suggests that the presence of H_2O (and OH^-) reduce the energy deposition required to cause amorphization. Above 413 K, the amorphization dose increased due to the absence of H_2O and OH^- and the absence of radiolytic decomposition of H_2O and OH^-.