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Natural metamict minerals as analogues of aged nuclear ceramic waste forms

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Natural metamict U-Th-containing minerals are well known analogues of aged nuclear ceramic waste forms, which were affected by self-irradiation and chemical alteration under the conditions of geological environment. Main concern related to the behavior of radioactive ceramics during their long-term storage is the stability of radionuclide solid solution in the crystalline structure of durable host-phases. Self-irradiation may cause not only damage of crystalline structure of host-phase (its amorphization or metamictization) but also destruction of solid-solution and release of radionuclides in separate chemically mobile forms. We have studied two large (1-3 cm in size) and relatively homogeneous samples of natural fully metamict minerals: Y-Fe-niobate, $(Y,Fe,U,Th)_x(Nb,Ti,Ta)_yO_z$ (containing 1.0 wt.% ThO₂ and 4.2 wt.% UO₂) and lovchorrite, $Na(Ca,Na)_2(Ca,Ce)_4TiO_2F_2(Si_2O_7)_2$ (containing 1.1 wt.% ThO₂). No evidences of solid solution destruction as a result of metamictization were observed in these samples. Both samples: untreated and annealed at different temperatures in order to initiate recrystallization were analyzed in comparison by precise X-ray powder diffraction, electron microprobe (EMPA) and scanning electron spectroscopy (SEM). It was observed that annealing caused not only recrystallization of both samples but also destruction of initially homogeneous solid solutions with partial release of U or Th in the forms of separate phases. It was proposed that recrystallization of aged nuclear ceramic waste forms as a result of any possible natural process is not positive phenomenon as it was assumed before. Recovery of crystalline structure can be accompanied by essential release of radionuclides.

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