

Synthesis and study of Pu-doped zircon

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Zircon, ZrSiO_4 , is under consideration as a prospective durable host-phase for the immobilization of waste actinides, in particular, weapons-grade Pu. It was supposed that Pu can substitute significant part of Zr in zircon crystalline structure forming solid solution, $(\text{Zr,Pu})\text{SiO}_4$. Two samples of Pu-doped polycrystalline zircon were synthesized through sintering in air of sol-gel precursors containing 10 and 5-6 wt.% Pu-239. Homogeneous methanol-aqueous solutions of $\text{Si}(\text{OC}_2\text{H}_5)_4$, Pu-nitrate and zirconil-oxynitrate were prepared with excess of Zr to provide final yield of zircon phase not less than 95 wt.% and the rest as zirconia $(\text{Zr,Pu})\text{O}_2$. Solidified from solutions gels were dried, calcined in air at 700°C for 1 hour, ball milled and cold pressed into pellets of 10 mm diameter. Obtained pellets were sintered in air at 1450°C for 4 hours and then at 1500°C for 3 hours. Samples of zircon were studied by XRD and SEM methods. It was found that zircon obtained from precursor containing 10 wt.% Pu incorporated only 6.9 wt.% Pu into its structure and the rest of Pu was converted to separate PuO_2 inclusions. This zircon was characterized of the following cell parameters: $a=6.636$; $c=5.994$. Zircon doped with 6.1 wt.% Pu was determined in the second sample. It was not accompanied by separate PuO_2 phase. Lattice cell parameters of that zircon were: $a=6.616$; $c=5.986$. Zircon lattice capacity to Pu incorporation is discussed. The preliminary conclusion was made that zircon forms limited solid solutions with PuSiO_4 and Pu incorporation into ZrSiO_4 lattice does not exceed 7 wt.%.