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Radiation and chemical durability of actinide crystalline matrices

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In order to isolate long-lived radionuclides from the environment it is necessary to immobilize them into highly stable forms (matrices). Crystalline materials of the mineral-like type are considered to be the most promising form of the matrix incorporating high-level waste (HLW). Alpha particles and heavy recoil nuclei lead to the damage of actinide-containing crystalline phase structures of up to their complete amorphization that can effect isolation properties of the HLW matrices.

The objective of the present work was to synthesize and investigate radiation and chemical durability of crystalline matrices based on titanate and titanate –zirconate pyrochlore, ferritic garnet and murataite. The materials obtained contained short-lived alpha- emitting isotopes of plutonium-238 ($T_{1/2} = 87,7$ years) and curium-244 ($T_{1/2} = 18,2$ years).

Synthesis was performed by cold pressing with subsequent sintering of the low calcined oxide mixture. The initial charge was saturated with high-activity curium-244 or plutonium-238 nitrite solutions. Therefore, compositions based on titanate ($\text{Ca}_{0,89}\text{Gd}_{0,1}(\text{U}_{0,44}\text{Hf}_{0,23}\text{Pu}_{0,22}\text{Gd}_{0,11})\text{Ti}_2\text{O}_7$ (mass fraction of 238-Pu is 8,7%), titanate zirconate $\text{Gd}_{1,935}\text{Cm}_{0,065}\text{TiZrO}_7$ (mass fraction of 244-Cm is 2,1 %) pyrochlores, ferritic garnet $\text{Ca}_{1,5}\text{Gd}_{0,908}\text{Cm}_{0,092}\text{Th}_{0,5}\text{ZrFe}_4\text{O}_{12}$ (mass fraction of 244-Cm is 2,0%) and murataite $\text{Ca}_{2,5}\text{Mn}_2\text{Th}_{0,41}\text{Cm}_{0,12}\text{Ti}_{7,5}\text{Zr}_{0,5}\text{AlFeO}_{24,5}$ (mass fraction of 244-Cm is 1,82%). Optimum annealing temperatures for each composition varied from 1300 to 1400 C and were established in the preliminary experiments with samples which did not contain highly active actinides. The phase composition of the ceramics was made up by the basic target phases in the presence of minor phases.

The change of the ceramic crystalline structure during irradiation was studied using the X-ray diffraction. The X-ray diffraction patterns vary with time. At the initial stages the diffraction reflexes change their angular positions that can be referred to the crystalline lattice swelling. Further dose accumulation leads to widening of diffraction reflexes in the large angles. Then the intensity of diffraction peaks sharply weakens (beginning of amorphization) and subsequently all the reflexes disappear (complete X-ray amorphization).

Amorphization doses of the matrix phases were as follows: $2,5 \times 10^{18}$ for titanate pyrochlore, $4,6 \times 10^{18}$ for titanate-zirconate pyrochlore, $1,6 \times 10^{18}$ for ferritic garnet, $2,6 \times 10^{18}$ for sintered murataite and $2,46-2,53 \times 10^{18}$ alpha-decay/g for melted murataite. The doses were obtained during storage of materials in argon at room temperature (293 K).

Chemical durability of crystalline and metamict samples was studied using the MCC-1 test (distilled water, 90 C, solution sampling for analysis on the 3rd, 7th and 14th day). Quantitative analysis of cations in solutions was made using the emission-spectral analysis and alpha-spectrometry. The calculated values of the leaching rates for the basic and potentially dangerous components (actinides) for all studied ceramics are within 10^{-6} to 10^{-7} g/cm²day. The change of the element leaching rate in the metamict ceramics is different for different types of matrices.

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