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Influence of temperature on the dissolution kinetics of synthetic $LaPO_4$-monazite in acidic media between 50 and 130°C

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Abstract

Single-phase monazite-type ceramics are discussed as waste forms for the safe disposal of surplus plutonium or separated minor actinides. To gain a deeper insight into the mechanism governing the dissolution of monazite-type compounds, the dissolution kinetics of synthetic $LaPO_4$-monazite was systematically studied by dynamic dissolution experiments carried out in the temperature range from 50 to 130°C in acidic media (0.01M HNO_3). The dissolution rates at far from equilibrium conditions increased from 3.2×10^{-5} g m^{-2} d^{-1} at 50°C to 2.5×10^{-4} g m^{-2} d^{-1} at 130°C. Two different temperature regions were observed, in which the normalised dissolution rates of $LaPO_4$ have a diverging temperature dependence, indicating two different mechanisms of dissolution: namely surface-controlled dissolution (T = 50 – 90°C; E_a = 44 kJ mol^{-1}) and transport-controlled dissolution (T = 90 – 130°C; E_a = 7.5 kJ mol^{-1}). Complementary thermodynamic modelling studies of the dissolution of $LaPO_4$ at the experimental conditions show that La-rhabdophane ($LaPO_4{\cdot}0.667H_2O$) is the thermodynamically favoured phosphate phase in aqueous environments below about 100°C. Apparently, the hydration of monazite and the formation of a thin surface layer consisting of rhabdophane is an intermediate step controlling the dissolution kinetics of the $LaPO_4$ monazite ceramics in low temperature aqueous environments.