

## **Dissolution kinetics of pyrochlore ceramics for the disposition of plutonium**

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### **ABSTRACT**

Single-pass flow-through (SPFT) experiments were conducted on a set of non-radioactive Ti-based ceramics at 90 °C and pH = 2 to 12. The specimens contained 27.9 to 35.8 wt% CeO<sub>2</sub> as a surrogate for UO<sub>2</sub> and PuO<sub>2</sub>. Compositions were formulated as TiO<sub>2</sub>-saturated pyrochlore (CeP1) and pyrochlore-rich baseline (CePB1) ceramic waste forms. Pyrochlore + Hf-rutile and pyrochlore + perovskite + Hf-rutile constituted the major phases in the CeP1 and CePB1 ceramics, respectively. Results from dissolution experiments between pH = 2 to 12 indicate a shallow pH-dependence with an ill-defined minimum. Element release rates determined from experiments over a range of sample surface areas (*S*) and flow rates (*q*) indicate that dissolution rates become independent of *q/S* values at 10<sup>-8</sup> to 10<sup>-7</sup> m/s. Dissolution rates dropped sharply at lower values of *q/S*, indicating rates that are subject to solution saturation effects as dissolved constituents become concentrated. Forward dissolution rates were 1.3(0.30) × 10<sup>-3</sup> and 5.5(1.3) × 10<sup>-3</sup> g/m<sup>2</sup>·d for CeP1 and CePB1 ceramics, respectively. Dissolution rates obtained in other laboratories compare well to the findings of this study, once the rate data are placed in the context of solution saturation state. These results make progress toward an evaluation of CeO<sub>2</sub> as a surrogate for UO<sub>2</sub> and PuO<sub>2</sub> as well as establishing a baseline for comparison with radiation-damaged specimens.

**Keywords:** Crystal structure, radiation damage, kinetics, dissolution rate, phase transition, amorphization, XRD data