

Solubility of Ln-phosphate waste forms and aqueous Ln-speciation under conditions relevant to geological disposal

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In the last decades, various single- and polyphase ceramic materials have been proposed as potential waste forms for the immobilisation of specific nuclear waste streams, such as separated plutonium from civilian or military sources unsuitable for further use, or separated minor actinides [1, 2]. Among them, monazites (monoclinic LnPO_4) emerged as promising candidates for ceramic nuclear waste forms due to their specific physico-chemical properties including high structural flexibility allowing for significant waste loadings, high chemical durability, and high resilience against radiation damage [3-5].

Within the frame of a collaborative research project on the immobilisation of long-lived radionuclides in ceramic matrices funded by the German Federal Ministry of Education and Research (Support code 02NUK021A), we employed geochemical modelling tools to address the durability and solubility of monazite waste forms under conditions relevant to geological disposal. The calculation cases addressed ground waters occurring in crystalline rocks and in clay rock environments at potential repository sites and pore waters typically encountered in bentonite buffers and under cementitious near-field conditions. The simulations show that the long-term performance of phosphate waste forms is significantly affected by the hydrogeochemical conditions at the repository site and the materials employed in the engineered barrier system. Under static conditions at circumneutral pH, only a small fraction of the waste form will dissolve, with the Ln-concentration limited by Ln-rhabdophane. In contrast, the modelling exercise indicates that monazite-based waste forms may be incompatible with a cementitious repository near-field.

[1] Lumpkin (2006) *Elements* **2**, 365–372. [2] Ewing (2007) *Prog. Nucl. Energy* **49**, 635–643. [3] Ewing & Wang (2002) *Rev. Min. Geochem.* **48**, 673–699. [4] Oelkers & Montel (2008) *Elements* **4**, 113–116. [5] Dacheux et al. (2013) *Am. Min.* **98**, 833–847.