

Supplementary information

Table S1 OPA¹⁰ and COx¹¹ pore water compositions and corresponding pH, and E_h at 25 °C used for the solubility and speciation calculations.

| Clay rock | OPA | COx |
|---------------------------------|-----------------------|-----------------------|
| ϑ / °C | 25 | 25 |
| pH | 7.5 | 7.2 |
| E_h / mV | -185 | -200 |
| Element concentration / (mol/L) | | |
| Na | 1.64×10^{-1} | 5.24×10^{-2} |
| K | 2.60×10^{-3} | 1.10×10^{-3} |
| Mg | 9.60×10^{-3} | 5.10×10^{-3} |
| Ca | 1.24×10^{-2} | 6.30×10^{-3} |
| Sr | 2.00×10^{-4} | 1.00×10^{-4} |
| Fe | 5.10×10^{-2} | – |
| Cl | 1.60×10^{-1} | 4.05×10^{-2} |
| F | 1.50×10^{-4} | – |
| S | 2.47×10^{-2} | 1.58×10^{-2} |
| C | 1.20×10^{-3} | 3.10×10^{-3} |
| Si | 2.00×10^{-4} | 4.00×10^{-4} |

Table S2 Calculated dominant solid phases of U, Np, Pu, Am, and Cm and their respective dominant aquatic species in OPA and COx pore water in equilibrium.

| Actinide | Solid phase | Dominant aquatic species (COx %; OPA %) |
|----------|--|---|
| U | UO ₂ ·2H ₂ O (am) | Ca ₂ UO ₂ (CO ₃) ₃ (aq) (67; 68) [CaUO ₂ (CO ₃) ₃] ²⁻ (31; 8) |
| Np | NpO ₂ ·2H ₂ O (am) | Np(OH) ₄ (aq) (83; 92) [Np(CO ₃)(OH) ₃] ⁻ (14; 6) |
| Pu | PuO ₂ ·2H ₂ O (am) | [Pu(CO ₃)] ⁺ (49; 35) [Pu(SO ₄)] ⁺ (28; 30) |
| Am | NaAm(CO ₃) ₂ ·5H ₂ O (s) | [Am(CO ₃)] ⁺ (53; 41) [AmOSi(OH) ₃] ²⁺ (30; 37) |
| Cm | NaCm(CO ₃) ₂ ·5H ₂ O(s) | [Cm(CO ₃)] ⁺ (52; 40) [CmOSi(OH) ₃] ²⁺ (31; 39) |