

Retention of the anions existing in nuclear waste by AFm phases

BIN MA¹, ALEJANDRO FERNANDEZ-MARTINEZ^{1,2*}, SYLVAIN GRANGEON³ AND LAURENT CHARLET¹

¹ISTerre, University of Grenoble, P.O. Box 53, F 38041, Grenoble, France (*correspondence: alex.fernandez-martinez@ujf-grenoble.fr)

²ISTerre, CNRS, P.O. Box 53, F 38041, Grenoble, France

³BRGM, 3, Avenue Claude Guillemin, 45060 Orléans Cedex 2, France

The cementitious structure in geologic nuclear waste repositories contains AFm phases, a family of hydrated calcium aluminates with double layered structure[1], with potential to retard the migration of anionic radionuclides[2]. In this project, we propose to evaluate the capacity of AFm-SO₄ and AFm-Cl₂ to retard the mobility of SeO₃²⁻, MoO₄²⁻, I⁻ and SO₄²⁻.

Anionic exchange in the AFm interlayer was studied using wet chemistry methods. For both AFm phases we observed anion exchange and AFm dissolution over 48 h with the corresponding release of Ca²⁺, Al³⁺, and SO₄²⁻/Cl⁻ into the solution. Total aqueous concentrations of Se and Mo indicate that the removal rates of anions by AFm-Cl₂ from the solution are higher than for AFm-SO₄. On the contrary, I⁻ was found to be little uptaken by any AFm. Although interlayer sulfate is not easily exchangeable, the double-layer structure of AFm-SO₄ was found to be most unstable, leading to dissolution-reprecipitation reactions and formation of gypsum.

Regarding the interaction of SeO₃²⁻ and MoO₄²⁻ with AFm phases, the two anions show a strong affinity with Ca²⁺, that is translated in a co-precipitation at high loadings. When SeO₃²⁻ and MoO₄²⁻ were over stoichiometric amounts of interlayer anions, the AFm phase dissolved and CaSeO₃ and CaMoO₄ precipitates were observed by extended X-ray absorption fine structure spectroscopy and X-ray diffraction analysis.

Both the anion exchange and saturated precipitation give us an insight that affinity of AFm phases for studied anions was found to be in the order of SO₄²⁻ > SeO₃²⁻ > MoO₄²⁻ > I⁻.

[1] Matschei *et al.* (2007) *Cem. Concr. Res.* **37**, 118-130.

[2] Aimoz *et al.* (2012) *ES&T* **46**, 3874-3881.