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Impact of phthalates on the migration behaviour of Eu(III) and Am(III) in a cementitious repository environment

NAILA AIT-MOUHEB[1], GUIDO DEISSMANN[1], PIERRE HENOCQ[2], NATHALIE MACÉ[3] AND DIRK BOSBACH[1]

[1]Forschungszentrum Jülich GmbH

[2]Research and Development Division, Andra, 1-7 Rue Jean Monnet, Parc de la Croix Blanche, 92298 Chatenay-Malabry Cedex

[3]Université Paris-Saclay, CEA, Service d'Etude du Comportement des Radionucléides, 91191, Gif-sur-Yvette

Presenting Author: n.ait.mouheb@fz-juelich.de

Cementitious materials are widely used in nuclear waste management, for example as construction materials and backfill in near surface and deep geological disposal facilities. This is in particular due to the radionuclide immobilisation properties of cementitious materials and their low permeability and diffusivity. However, the impact of organic degradation products, derived either from waste constituents or from cement additives, on radionuclide solubility and sorption in cementitious environments is not completely understood. Thus, one aim of EURAD work package CORI is to fill knowledge gaps with respect to the process understanding of interactions between cementitious materials and selected organic degradation products and to assess their impact on radionuclide migration.

In this context, we investigate the effect of phthalates, on the retention behaviour of Am(III) and Eu(III) in cementitious environments, employing hardened cement pastes (HCP) and synthesised hydration phases. HCP provided by CEA were prepared in 2016 from a CEM V/A 42.5N (Calcia, Rombas) with a water/cement ratio of 0.40 as described by [1]. The characterisation (e.g., by X-Ray Diffraction and Scanning Electron Microscopy), supported by complementary hydration modelling using the geochemical code GEMS and the database CEMDATA18, revealed C-(A)-S-H phases and ettringite as main hydration phases, accompanied by AFm, portlandite, mullite, and unreacted clinker, slag and fly ash. In addition, model hydration phases, namely C-S-H and C-A-S-H with Ca:Si ratios between 0.8 and 1.2 and an Al:Si ratio of 0.05, respectively, were synthesised under Ar atmosphere. The uptake of Am(III) and Eu(III) is studied in batch experiments in the presence and absence of phthalates. Preparatory solubility tests showed that phthalates concentrations up to 1 mM have no impact on the dissolved and colloidal fractions of Am(III) and Eu(III) in cementitious water. Thermodynamic modelling suggests that the effect of phthalates on the solubility and speciation of Am(III) and Eu(III) is negligible under alkaline conditions, when assuming amorphous $Am(OH)_3$ and $Eu(OH)_3$, respectively, as solubility limiting phases. The phthalates concentration of 1 mM is likely not sufficient to have a strong impact on the solubility and speciation of Am(III)/Eu(III) and to observe complexing effects.

Reference

[1] Macé et al. (2019). Appl. Geochem. 100, 326–334.