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ORIGINAL PAPER

Effects of organic degradation products on the migration behaviour of radionuclides in cementitious materials

Naila Ait-Mouheb[1] · **Nathalie Macé**[2] · **Pierre Henocq**[3] · **Dirk Bosbach**[1] · **Guido Deissmann**[1]

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Abstract

The impact of phthalates, which might be present in a cementitious repository as degradation products of superplasticisers or PVC additives, on the migration behaviour of *An*(III)/*Ln*(III) was investigated in batch sorption experiments on hardened cement paste. In the absence of organics, *An*(III)/*Ln*(III) sorb strongly on HCP with distribution ratios, $\boldsymbol{R}_{\mathbf{d}}$, of 10^5 to 10^6 L kg^{-1}. Above a no-effect level of ~ 10^{-3} mol L^{-1} phthalate, a distinct decrease of the *An*(III)/*Ln*(III) sorption was observed with sorption reduction factors (i.e., the ratio between the R_d values without and with organic ligand) of 100 to 1000 at phthalate concentrations of 10^{-1} mol L^{-1}. This sorption reduction is attributed to the effect of Ca complexation with phthalate on the stability of calcium silicate hydrate (C–S–H) phases as main sorbing phase. These results and ongoing diffusion experiments indicate an increase in the mobility of *An*(III)/*Ln*(III) in cementitious barriers with increasing phthalate concentrations.

Introduction

The safety concept for deep geological disposal of radioactive wastes is based on the confinement of the radioactivity over long periods of time by a multiple barrier system, combining an engineered barrier system and a suitable host-rock formation. Many disposal concepts for radioactive wastes developed internationally make extensive use of cementitious materials, for example, for solidification and stabilisation of low and intermediate level wastes (L/ILW), or as construction materials, drift and shaft seals, and backfill [1–4]. This is due to the general radionuclide fixation and immobilisation properties of cementitious materials and their low permeability and diffusivity. Although the behaviour of safety-relevant radionuclides in cementitious environments has been investigated extensively in the last decades, the impact of organic degradation products, originating from organic waste components or from superplasticisers in cementitious materials, on the migration of radionuclides under highly alkaline, cementitious conditions is not yet fully understood [5].

Polyvinyl Chloride (PVC) and styrene-based ion-exchange resins (IER) form substantial constituents of L/ILW streams in various European countries such as France, Switzerland, Sweden, and Finland [6]. Thus, the aim of our work, performed within the framework of EURAD WP3 CORI (Cement-Organic-Radionuclide Interaction), was to fill knowledge gaps in the understanding of the impacts of the presence of phthalate species ($C_8H_4O_4{}^{2-}$; degradation product of superplasticizers used as additives to enhance the workability of concrete [7] or from PVC plasticisers such as di(2-ethylhexyl)phthalate (DEHP) or di-isononyl phthalate (DINP) [8]) and tri-methyl-amine (TMA, C_3H_9N; degradation product of basic anion exchange resins [9]) on the migration behaviour of safety relevant radionuclides in cementitious barriers. Here, we focus on the potential effect of phthalates on the solubility and sorption of trivalent actinides and lanthanides such as ^{241}Am(III) and ^{152}Eu(III) on hardened cement pastes. Batch sorption experiments were performed to quantify the uptake of these radionuclides in cementitious materials and assess the effects of phthalates on their sorption behaviour. The role and significance of phthalates on the migration behaviour of ^{241}Am and ^{152}Eu in a cementitious repository are further explored in diffusion experiments using cement monoliths.

✉ Naila Ait-Mouheb
n.ait.mouheb@fz-juelich.de

[1] Institute of Energy and Climate Research – Nuclear Waste Management (IEK-6), Forschungszentrum Jülich GmbH, 52428 Jülich, Germany

[2] Université Paris-Saclay, CEA, Service de Physico-Chimie, 91191 Gif-Sur-Yvette, France

[3] Research and Development Division, Andra, 92298 Châtenay-Malabry Cedex, France

Materials and methods

Preparation of HCP

The hardened cement paste (HCP) was manufactured by the Commissariat à l'énergie atomique et aux énergies alternatives (CEA) in February 2016 from a CEM V/A cement (CEM V/A (S-V) 42.5N CE PM-ES-CP1 NF, Calcia, Rombas) with a cement/water ratio of 0.40 as described by Macé et al. [10]. The CEM V/A is a ternary blended cement comprising approximately 50 wt% clinker, 25 wt% blast furnace slag and 25 wt% fly ash (mainly silica fume) and is expected to be used in the French nuclear waste disposal program [11, 12]. The main constituents of the cement paste are C–S–H, calcite, ettringite, mullite, portlandite, alite and quartz as well as an amorphous fraction of unreacted slag and fly ash [12]. Further information on the chemical composition of the binder is given in Table S1 in the Supplementary Information. The HCP cylinders were stored in fully saturated conditions at room temperature in a glovebox under controlled atmosphere (Ar) to avoid carbonation. The HCP was mechanically crushed and sieved to < 100 µm for use in batch sorption experiments. For diffusion experiments, the HCP cylinder was sliced with a diamond saw to obtain HCP discs.

Batch sorption and diffusion experiments

Batch sorption studies with both ^{241}Am and ^{152}Eu on CEM V/A HCP were carried out in a glove box under Ar atmosphere (O_2 and CO_2 levels < 5 ppm) at different phthalate concentrations (phthalic acid, 99.5% Sigma-Aldrich; concentration range 10^{-1} to 10^{-5} mol L^{-1}) and in absence of phthalate ions; initial ^{241}Am and ^{152}Eu (Eckert & Ziegler Nuclitec GmbH) concentrations in the experiments were 10^{-8} to 10^{-10} mol L^{-1} and 3.0×10^{-8} to 5.0×10^{-11} mol L^{-1}, respectively. The solution used in the sorption experiments was prepared by immersing crushed CEM V/A HCP in Milli-Q® water (Millipore, 18.2 MΩ cm^{-1} at 25 °C; total organic carbon (TOC) content 2 ppb) at a solid-to-liquid (S/L) ratio of 0.9 g L^{-1} as described by Pointeau et al. [13]. After 15 days of equilibration, the artificial cement water (ACW) is filtered and used in sorption experiment to mimic the degradation state III. Sorption experiments were performed with a fixed S/L ratio of 5×10^{-4} kg L^{-1} prepared in (ultra)centrifuge tubes (polyethylene centrifuge tubes with sealing cap). Before adding aliquots of ^{241}Am and ^{152}Eu, the suspensions were equilibrated for at least 5 months with ACW and—in case of the experiments with phthalate addition—the pH was adjusted to 12.2 using NaOH, where required. The sorption experiments were performed for 7 days, based on literature data on the fast uptake kinetics of *An*(III)/*Ln*(III) on cementitious materials [14]. After one week equilibration time, the suspensions were centrifuged (6000 rpm, 1 h) and analysed for ^{241}Am and ^{152}Eu with an ultra-low-level liquid scintillation (LSC) counter Qantulus 1220 (Perkin Elmer, USA) equipped with alpha–beta discrimination option. Samples for the LSC were prepared by mixing aliquots of the supernatants with Ultima Gold LLT LSC cocktail (Revvity). The counting efficiencies of LSC were 82.6% for ^{152}Eu and nearly 100% for ^{241}Am.

The results of the sorption tests were expressed in form of the distribution ratio R_d [L kg^{-1}] as:

$$R_d = \frac{A_s}{A_l} = \left(\frac{A_i - A_l}{A_l}\right) \cdot \left(\frac{V}{m}\right)$$

where A_s [Bq kg^{-1}] is the tracer activity sorbed on the solid phase, A_l [Bq L^{-1}] the tracer activity in the liquid phase at the end of the experiment, A_i [Bq L^{-1}] the initial tracer activity in solution, V [L] the volume of the liquid phase, and m [kg] the mass of solid phase used in the experiment, respectively. Wall sorption effects and the formation of ^{241}Am and ^{152}Eu colloids were considered in the data evaluation based on the results of solubility tests as described by Tits and Wieland [13].

Diffusion experiments with ^{152}Eu on CEM V/A HCP monoliths were carried out inside an Ar filled glove box in absence and presence of phthalate ions ($[\text{phthalate}]_{tot} = 10^{-2}$ mol L^{-1}), using diffusion cells adapted from the work of Yuan et al. [15]. Cylindrical HCP samples (thickness: 10 ± 0.1 mm; cross-sectional area: 19.6 cm^2) were embedded in an epoxy resin (Epofix, Struers GmbH) forming a sample holder. The sample holders were mounted in a cylinder made of polymethylmethacrylate (PMMA), creating an "inlet reservoir" on one side and an "outlet reservoir" on the other. The total volumes of the inlet and outlet reservoirs were 126 mL and 7 mL, respectively. A filtered portlandite-saturated solution, representative of cement degradation state II (pH 12.5; $[\text{Ca}]_{tot} = 22$ mM) is used in the diffusion experiments. After equilibration, diffusion was initiated by spiking the solution inside the inlet reservoir with ^{152}Eu to reach an initial concentration of 2×10^{-9} mol L^{-1} below the solubility limit in degradation state II. The ^{152}Eu tracer solution used was carrier free. Openings in both reservoirs allow regular sampling of the solutions; the ^{152}Eu activities of the samples were measured by LSC as described above. In addition to ^{152}Eu, the solutions in the inlet reservoir were spiked with HTO in tracer concentrations ($[\text{HTO}]_{initial} = 5 \times 10^{-10}$ mol L^{-1}) to determine the effective diffusivity and the accessible porosity of the HCP monoliths for a (assumingly) non-sorbing tracer. After each 2 mL sampling, the solution in the outlet reservoir is fully replaced by a fresh portlandite-equilibrated filtered water. Data evaluation is performed by inverse modelling of the diffusion data using the finite element code COMSOL Multiphysics (Ver. 6.1).

Thermodynamic modelling

To aid interpreting the batch sorption and diffusion experiments in presence and absence of organics, the aqueous speciation of the components in solution and saturation indices of relevant solid phases were calculated using the geochemical codes PhreeqC Ver. 3.5.0 [16] and GeoChemist's Workbench (GWB) Ver. 11.0.8 [17]. The ThermoChimie v.10d thermodynamic database (Consortium Andra—Ondraf/Niras—RWM [18, 19]) was used for the thermodynamic modelling. The activities of aqueous species were calculated using the specific ion interaction (SIT) approach [20–22] in PhreeqC and the B-dot equation [17] in GWB, respectively.

Results and discussion

The results of the ^{241}Am and ^{152}Eu batch sorption experiments with CEM V/A HCP are shown in Fig. 1 and given in Supplementary Material (data publication [23]). In the absence of phthalate, the strong sorption of ^{241}Am and ^{152}Eu on cementitious materials is re illustrated by R_d values in the range 10^5 L kg^{-1} < R_d < 10^6 L kg^{-1}. These results are in good agreement with those obtained for degradation state I and state III in the literature [14, 24–26], considering that in absence of organics, no distinct effect of the cement degradation state on *An*(III)/*Ln*(III) sorption is observed. Comparison between the R_d values obtained for ^{241}Am and those for ^{152}Eu indicates slightly stronger uptake of ^{152}Eu by the HCP CEM V/A state III for the highest concentrations investigated. The retention of ^{241}Am and ^{152}Eu by cementitious materials is mainly due to uptake by calcium-silicate-hydrates (C–S–H) and is thought to be associated with a two-step process [13, 27]: ^{241}Am and ^{152}Eu form first an inner sphere complex on the C–S–H phases. This is followed by a second step in which ^{241}Am and ^{152}Eu become incorporated into the C–S–H structure, substituting Ca in the C–S–H interlayer and in the Ca-octahedral layer.

In the investigated systems, ^{241}Am and ^{152}Eu uptake by HCP CEM V/A was found to be strongly affected by the aqueous phthalate concentration. The results in Fig. 1 show a plateau of the uptake at low phthalate concentrations (< 10^{-3} mol L^{-1}), where the R_d values are rather similar to those in absence of phthalate, followed by a distinct decrease in the R_d values above a so-called "concentration edge" [28]. Above the "concentration edge", the R_d values of ^{241}Am and ^{152}Eu decrease by several orders of magnitude to values of about 10^3 L to 10^4 L kg^{-1} at phthalate concentrations of 10^{-1} mol L^{-1}. These results indicate that the sorption reduction factors (SRF, i.e. the ratio of the R_d value in absence of organics to the one in presence of the organic ligand) for *An*(III)/*Ln*(III) are about 100 to 1000 at this phthalate concentration.

At phthalate concentrations of 10^{-3} mol L^{-1} and below, R_d values of ^{241}Am are generally somewhat lower than those of ^{152}Eu, suggesting a slight difference in the sorption behaviour of ^{241}Am compared to ^{152}Eu in cementitious materials, with stronger retention of the latter. However, at the highest phthalate concentrations, the sorption behaviour of both radionuclides is rather similar.

Under near neutral conditions, phthalates are strong complexing ligands for *An*(III)/*Ln*(III) [7], leading to a strong decrease in the sorption of these radionuclides to clay rocks [29]. However, the geochemical speciation calculations showed that, under cementitious conditions, their impact on the speciation and solubility of ^{241}Am or ^{152}Eu is negligible (cf. Fig. 2). The contributions of aqueous Am- and Eu-phthalate complexes to the total radionuclide concentrations were less than 10^{-4}% even at the highest phthalate addition. Thus, the observed "concentration edge" and the decrease in *An*(III)/*Ln*(III) sorption with increasing phthalate concentrations cannot be explained by complexation with the organic compound but rather suggests an effect of the phthalate addition on the cementitious material used in the sorption experiments.

In Fig. 3, the calculated aqueous speciation of Ca in a solution equilibrated with HCP CEM V/A is shown as function of the phthalate concentration. At phthalate concentrations above around 10^{-3} mol L^{-1}, the contribution of the aqueous Ca-phthalate complex ($(Ca(C_8H_4O_4)^0)$) to the total Ca concentration increases distinctly, becoming the dominant Ca-complex at phthalate concentrations above about 10^{-2} mol L^{-1}. The increasing complexation of Ca in solution causes the dissolution/destabilisation of Ca-bearing phases in the cementitious material, leading to decalcification of the C–S–H phases, which are the main sorbing phases for *An*(III)/*Ln*(III) in cementitious materials. This destabilisation of C–S–H phases is also indicated by the calculated saturation indices. In the absence of the organic complexant or at low phthalate concentrations (i.e., ≤ 10^{-3} mol L^{-1}), respectively, the solution equilibrated with HCP CEM V/A is virtually in equilibrium with a C–S–H phase with low Ca/Si ratio (CSH0.8; saturation index SI ≈ 0). With increasing phthalate addition, the saturation index of this phase decreases to about – 0.8 (at 10^{-1} mol L^{-1} phthalate), indicating the increasing destabilisation of the C–S–H.

The diffusion experiments and the interpretation of the diffusion data are still ongoing, since the HTO flux has not reached yet the diffusive steady state. In Fig. 4, the concentration of ^{152}Eu in the inlet of the diffusion cells in the absence and presence of phthalate is illustrated as a function of time. Based on the sorption results, an increase in the mobility of

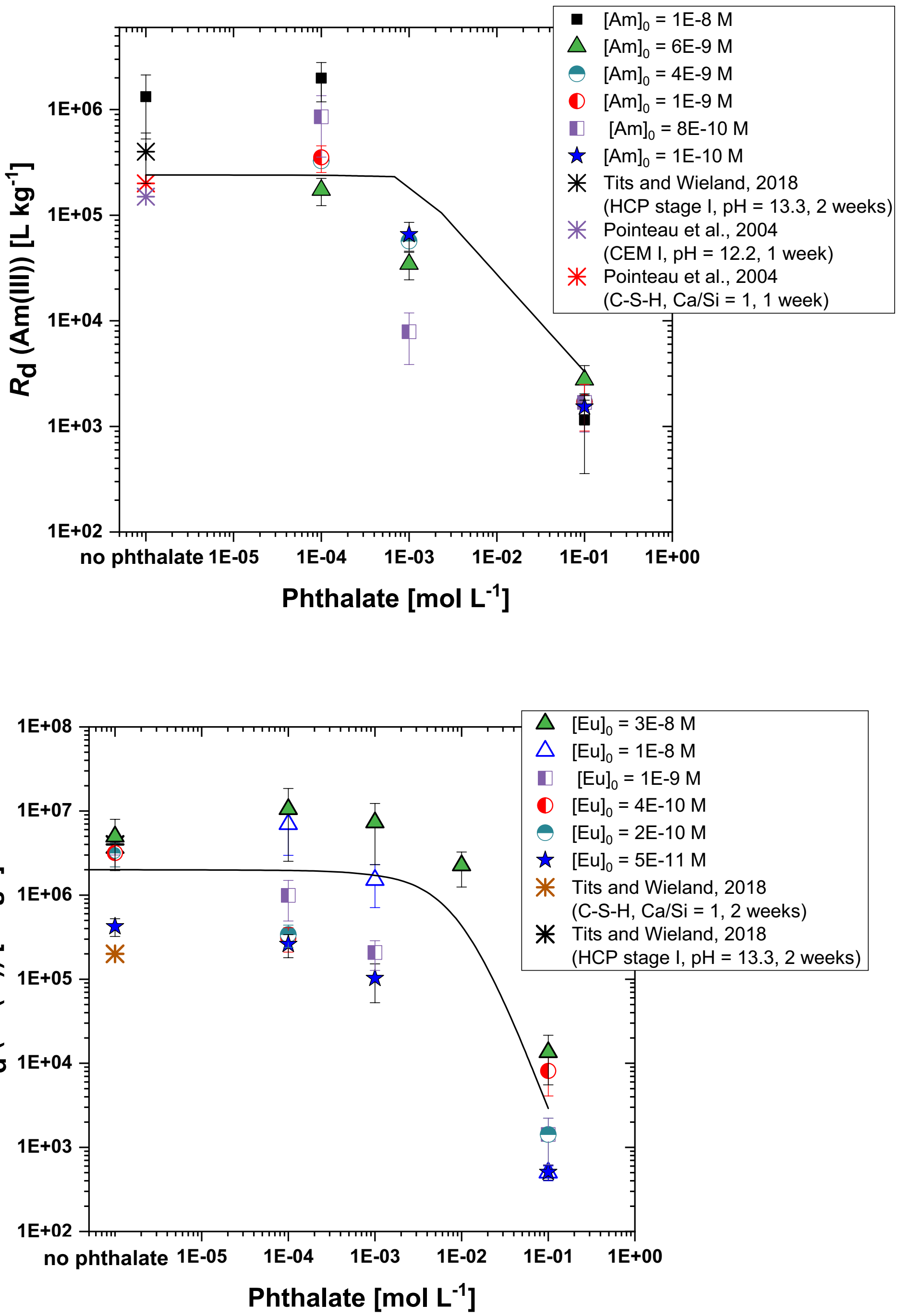
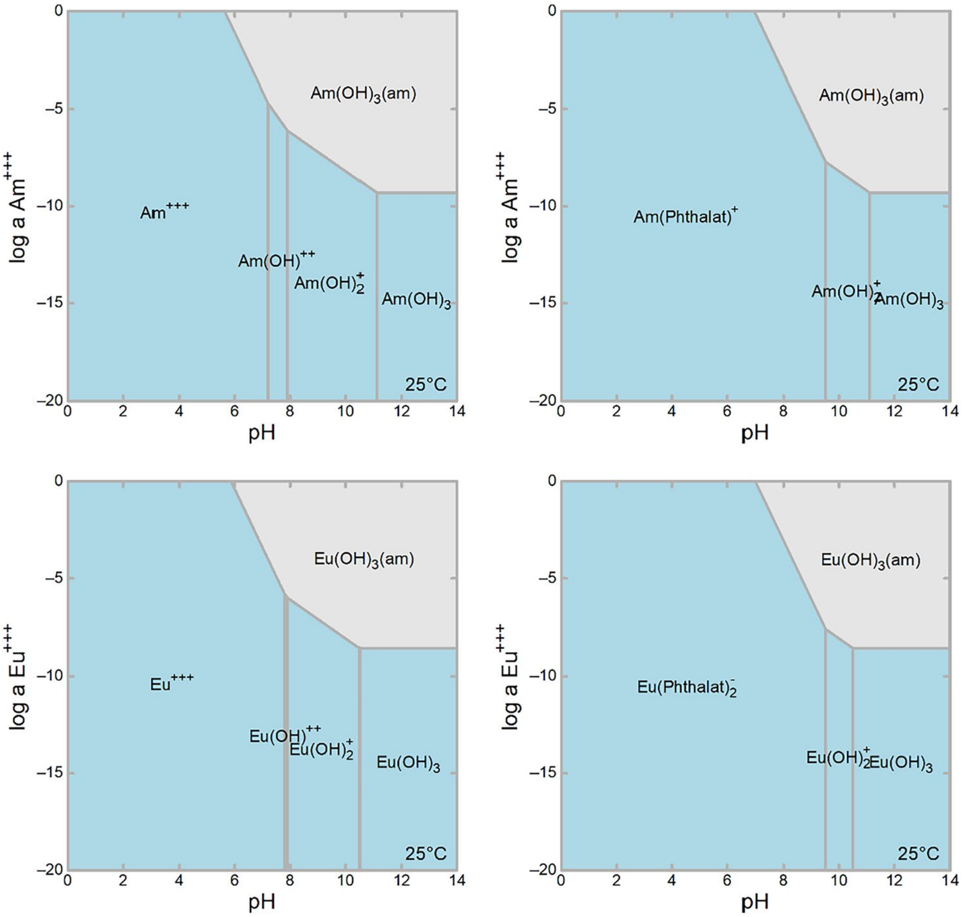


Fig. 1 Effect of phthalate on the uptake of ^{241}Am and ^{152}Eu by HCP prepared from CEM V/A. The solid line represents the best fit obtained from the data using a Freundlich isotherm. The experimental data of ^{241}Am and ^{152}Eu uptake parameters are in the Supplementary Material (data publication [23])

An(III)/*Ln*(III) in cementitious barriers in the presence of high phthalate concentrations is expected. However, the preliminary diffusion data shown here are probably mainly associated with sorption processes occurring at the surface of the monoliths. Even in the presence of phthalate, *An*(III)/*Ln*(III) are strongly sorbing radionuclides on cementitious materials (log R_d about 3.5 {L kg^{-1}} at 10^{-1} mol L^{-1} phthalate); thus, a break-through of such radionuclides in diffusion experiments may require years. Therefore, the transport parameters of ^{152}Eu will be evaluated by fitting the experimental tracer distribution profiles in the HCP, using laser ablation coupled to ICP-MS for providing the concentration profiles in the HCP disks. Based on initial transport simulations, the diffusion profiles will be determined after 120 days of diffusion.

Fig. 2 Solubility of Am (top) and Eu (bottom) in the absence (left) and presence (right) of 10^{-2} mol L^{-1} phthalate in a solution containing 5×10^{-3} mol L^{-1} of Ca as function of pH



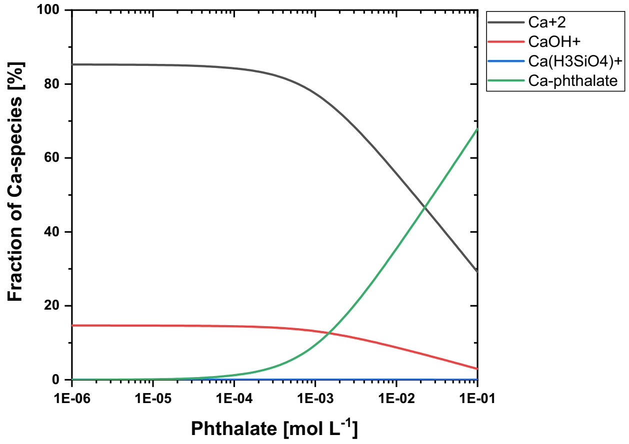


Fig. 3 Calculated Ca-speciation in a solution equilibrated with HCP CEM V/A as function of the phthalate concentration ($[\mathrm{Ca}]_{\mathrm{tot}} = 6.5 \times 10^{-3}$ mol L^{-1}; $[\mathrm{Si}]_{\mathrm{tot}} = 9.2 \times 10^{-5}$ mol L^{-1}; pH fixed at 12.2)

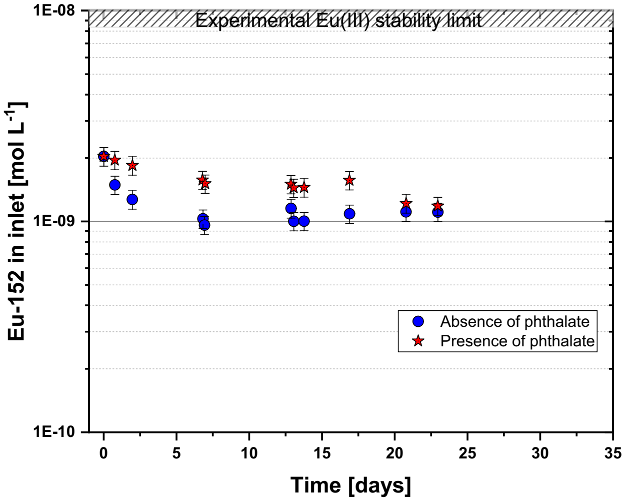


Fig. 4 ^{152}Eu concentration in the inlet of the diffusion cells under absence and presence of phthalate ($[\mathrm{phthalate}] = 10^{-2}$ mol L^{-1})

Conclusions

As expected, in absence of phthalate ions, ^{241}Am and ^{152}Eu sorb strongly on HCP CEM V/A as illustrated by R_d values in the range of 10^5 to 10^6 L kg^{-1}. Above a no-effect level of about 10^{-3} mol L^{-1} phthalate, the presence of phthalate species leads to a strong decrease in the sorption of *An*(III)/*Ln*(III) in this system by several orders of magnitude to R_d values of about 10^3 to 10^4 L kg^{-1}, resulting in sorption reduction factors of about 100 to 1000 at 10^{-1} mol L^{-1} phthalate. This sorption reduction is not caused by the complexation of *An*(III)/*Ln*(III) by the organic ligand under the prevailing highly alkaline conditions. However, it is rather due to the decalcification of C–S–H as the main sorbing phases for *An*(III)/*Ln*(III) in consequence of the increasing formation of Ca-phthalate complexes in solution. Further effects might be due to the sorption of phthalate ions by the C–S–H phases. The results indicate that the presence of elevated phthalate concentrations will enhance the mobility of *An*(III)/*Ln*(III) in the cementitious barriers of a repository system, even without complex formation with the radionuclides. This effect of phthalates on the migration behaviour of *An*(III)/*Ln*(III) will be further evaluated in the ongoing diffusion experiments using HCP monoliths.

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Data availability The data generated during the current study are available at https://doi.org/https://doi.org/10.26165/JUELICH-DATA/PCZCKP.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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