



# Comparative study of Eu and U sorption on cementitious materials in the presence of organic substances

Marta Burešová<sup>1</sup> · Jana Kittnerová<sup>1</sup> · Barbora Drtinová<sup>1</sup>

Received: 27 June 2022 / Accepted: 1 December 2022 / Published online: 25 January 2023  
© The Author(s) 2023

## Abstract

A comparative study of the sorption behavior of europium, a chemical analogue of trivalent actinides, and uranium was performed on hydrated cement paste (HCP) CEM I and calcium-silicate-hydrates (CSH) phase. Kinetic experiments proved that Eu sorption is faster than that of U depending on the phase ratio L/S and the sorption material, whereas EDTA has no significant effect. Same conclusions can be made for equilibrium experiments with HCP and U while with Eu EDTA clearly influenced the distribution ratio  $R_d$  with sorption reduction factor (SRF) below 0.5. Uranium sorption on CSH compared to HCP is higher with SRF above 1.2.

**Keywords** Sorption · Cement · Cementitious materials · HCP · CSH · Europium · Uranium · EDTA

## Introduction

The cementitious materials are widely use in the storage of radioactive waste, mainly of low level waste-intermediate level waste (LLW-ILW). In the Czech Republic, steel barrel of solidified waste is placed in a larger barrel, the space between them is filled with concrete and concrete is also used for pouring the chambers (cements of type CEM II, CEM I and CEM III are used sequentially). Expectedly, the cementitious material forms also structural support of repositories. Similarly, in the planned deep geological repository (following Swedish concept), cementitious materials will generally be part of construction elements and also of engineering barriers at least for decommissioning wastes (backfilling, waste treatment or part of storage containers).

The contact of cement and radionuclides leading to influencing their migration in both types of repositories will occur after possible damage of the packaging [1, 2] unless the cement materials directly serve as solidifying medium already. After their release, radionuclides will interact with engineering barrier materials in various processes, including sorption and diffusion. The uptake rate depends on the

properties of the radionuclides (cation  $\times$  anion, charge size), the type of material (cement  $\times$  bentonite), the conditions (temperature, pH, etc.) and can also be affected by organic substances present in radioactive waste [3–5], e.g. gloves, cables, ion exchange resins, detergents, including EDTA as the most abundant and one of the strongest complexing decontamination agents [6].

Another important parameter influencing the sorption behavior of cement materials is their aging. In Stage III of HCP aging where it is most likely the time concurrence with the possible release of radionuclides to occur, phase CSH considered a main sorption material for (mainly) cations uptake is the main component [7].

For sorption behavior quantification, a distribution ratio,  $R_d$ , is commonly used, that describes the distribution of the radionuclide between the liquid and solid phases whereas does not provide any information on the mechanism of radionuclide uptake [8].

The uptake of europium on cements is not very thoroughly described yet, but according to the comparisons in [9–11] it generally reaches distribution ratio in the order of  $10^5$ – $10^6$  L kg<sup>-1</sup>. For the pure CSH phase with different Ca/Si ratios, the determined  $R_d$  values were higher than  $10^4$ – $10^5$  L kg<sup>-1</sup> [12]. Due to the mechanism of sorption of europium on cements, the values of sorption on HCP and CSH phase are expected to be similar [13], which is confirmed by the results so far.

✉ Marta Burešová  
marta.buresova@jfifi.cvut.cz

<sup>1</sup> Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, Břehová 7, 115 19 Prague 1, Czech Republic

Sorption of U on cementitious materials of various types has been described in several studies (e.g. [14–16]), however the mechanism of U(VI) uptake by HCP is still poorly understood at the atomic level. Under different conditions,  $R_d$  values are in the order of  $10^3$  L kg<sup>-1</sup>.  $R_d$ 's with order of magnitude higher ( $10^4$ – $10^5$  L kg<sup>-1</sup>) are obtained when sorbed onto degraded cements (degradation reduces the Ca/Si ratio in CSH) and pure CSH [17].

The importance of organic matter on sorption behavior is quantified by SRF (sorption reduction factor) that can reach for trivalent lanthanoids (i.e., Eu) and hexavalent actinides (i.e., U) values about 1–10 as reported when exceeding the NEC (No Effect Concentration, i.e., the highest concentration of an organics that does not cause any effect on sorption, the value of NEC tends to be about  $10^{-2}$ – $10^{-4}$  mol L<sup>-1</sup>). Moreover, the increase of SRF with increasing organics concentration occurs [18, 19].

The aims of this study were: (1) to compare the sorption of Eu and U on cementitious material (for this purpose hydrated cement paste of CEM I type (i.e., Ordinary Portland Cement) was chosen as the most homogeneous cementitious material); (2) to study the dependence of equilibrium values of distribution coefficient  $R_d$  on the EDTA concentration; (3) to describe the difficulties, especially for europium, with wall sorption; (4) to compare sorption of U on commercial cement and synthetic phase CSH.

## Experimental

### Materials and radionuclides

A comparative study of Eu and U sorption was performed on HCP CEM I prepared from Portland cement (CEM I 42.5 R). The HCP was crushed and sieved to a fraction <0.4 mm. The second material used was CSH with a Ca/Si ratio of 1.0 prepared following the procedure of [20] as described in detail in [21].

The isotopes of interest (occurring in radioactive waste, i.e., <sup>152</sup>Eu ( $\gamma$ , 13.5 y), <sup>233</sup>U ( $\alpha$ , 1.6·10<sup>5</sup> y) and <sup>238</sup>U ( $\alpha$ , 4.5·10<sup>9</sup> y) were studied to compare their sorption on HCP CEM I (pH 12.5) and CSH (pH 10.7). Specifically, EuCl<sub>3</sub> and UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> · 6H<sub>2</sub>O were used for the experiments as initial compounds.

### Experimental method

The sorption of Eu and U was evaluated using batch type experiments carried out in plastic ampoules (PP), in the case of U-CSH system under inert gas atmosphere in a glove-box (N<sub>2</sub>, < 50 ppm CO<sub>2</sub>), as CSH is more sensitive to atmospheric CO<sub>2</sub>. After the required reaction time and centrifugation (RCF 900 g for HCP and 2500 g for CSH, 10 min),

samples of the liquid phase were analyzed for the remaining radionuclide activity in solution. The influence of addition of organic compound Na<sub>2</sub>EDTA (Ethylene-diamine-tetra-acetic acid disodium salt dihydrate, C<sub>10</sub>H<sub>14</sub>N<sub>2</sub>Na<sub>2</sub>O<sub>8</sub> · 2H<sub>2</sub>O, hereinafter referred to as EDTA) of two concentrations:  $5 \cdot 10^{-5}$  mol L<sup>-1</sup> (referred to as lower conc. EDTA) and  $5 \cdot 10^{-3}$  mol L<sup>-1</sup> (referred to as higher conc. EDTA) was investigated. These concentrations were chosen based on the expected NEC [22] as one working concentration is below this limit and the other exceeds it.

The required equilibration times were determined in a set of kinetic experiments. Experiments of <sup>152</sup>Eu ( $c_{\text{init}} = 2.9 \cdot 10^{-9}$  mol L<sup>-1</sup>) uptake were performed with HCP CEM I in the porlandite water (saturated solution of Ca(OH)<sub>2</sub>, 0.02 mol L<sup>-1</sup>, pH = 12.4–12.6) at liquid to solid (L/S) ratios of 100 L kg<sup>-1</sup> and 500 L kg<sup>-1</sup> (6 mL of the liquid phase) in the presence/absence of EDTA with the duration of 96 h. After the given time, the ampoules were centrifuged, and activities of supernatant (2 mL) were measured on HPGe detector system (type GEM40P4 with Ortec DSPEC jr 2.0, Maestro software version 7.0). The kinetic experiments set the duration of 48 h to be appropriate to achieve equilibrium state. Thus, the equilibrium experiments with Eu were performed for 48 h, in the L/S range 100–800 L kg<sup>-1</sup> with/without EDTA of both concentrations.

Experiments with uranium <sup>233</sup>U were carried out similarly to those with Eu. First, the initial concentration of U  $7 \cdot 10^{-8}$  mol L<sup>-1</sup> was chosen with respect to its limited solubility in strongly alkaline media [23], and then kinetic experiments were performed with HCP CEM I (in saturated Ca(OH)<sub>2</sub>, 6 mL) and CSH 1.0 (in the saturated CSH 1.0 solution, 4 mL, pH 10.7) with the phase ratio L/S of 800 L kg<sup>-1</sup> in the presence/absence of the lower conc. EDTA. Based on the obtained kinetic curves, the duration of the equilibrium experiments was determined to be 3 weeks. Furthermore, a higher initial concentration of uranium ( $1 \cdot 10^{-5}$  mol L<sup>-1</sup> with <sup>238</sup>U as a carrier) was also used for the experiment with CSH 1.0 with/without EDTA at both concentrations. After the centrifugation of the samples, activities of 200  $\mu$ L samples were measured in the 5 mL of scintillation cocktail Ultima Gold AB on the LSC counter (HIDEX 300 SL with MicroWin software) for the period of 12 h.

Moreover, the sorption on the walls of the experimental ampoules was thoroughly determined for both studied elements, especially for europium for L/S 100–800 L kg<sup>-1</sup> in two parallel ampoules in the same way as in the equilibrium sorption experiments except for sampling. After a set time as much liquid over the cement as possible was removed and the rest was allowed to air dry for 3 days. The cement was then moved into the measuring vial and the experimental ampoule was wiped with the dry tampon (subsequently also measured). Desorption solution (1 mol L<sup>-1</sup> HCl, 4.5 mL) was added to the ampoule and shaken (120 rpm) for 2 days.

After that the empty ampoules and desorption solutions were measured separately. Sorption to the walls was taken into account in the  $R_d$  evaluation.

### Evaluation of sorption data

The uptake of the radionuclides by cementitious material is characterized here in terms of the distribution coefficient  $R_d$  according to Eq. (1)

$$R_d = \frac{A_{\text{init}} - A_t}{A_t} \cdot \frac{V}{m}, \quad (1)$$

where  $A_{\text{init}}$  is the initial activity concentration of the radionuclide in solution, and  $A_t$  corresponds to the activity concentration at time  $t$ , respectively, both in the unit of activity (CPM),  $V$  (L) is the volume of the liquid phase, and  $m$  (kg) the mass of solid phase used in the experiment. In the literature most often the term  $K_d$  is used (same relationship), but this term entails that the sorption on the solid has linear dependency on the concentration in the solution and is reversible [8].

The distribution coefficient can be determined by evaluating the shape of the sorption isotherm. For linear isotherm the linear  $K_d$ -model is used according to the Eq. (2):

$$q = K_d \cdot c, \quad (2)$$

where  $q$  (mol kg<sup>-1</sup>) is the equilibrium concentration in the solid phase,  $c$  (mol L<sup>-1</sup>) is equilibrium concentration in the liquid phase and  $K_d$  (L kg<sup>-1</sup>) is the distribution coefficient, dimensionally identical to the distribution ratio  $R_d$ .

The uptake of radionuclides in the presence of organics is usually expected to decrease due to the complexation of the radionuclide with the organic ligand as described by the sorption reduction factor SRF (distribution coefficient in the system without organics divided by distribution coefficient with organics, defined for a given system and conditions).

## Results and discussion

### Sorption on the reaction ampoules

In the wall sorption control experiments (experiments only with the liquid phase in the ampoule), a high sorption of <sup>152</sup>Eu on the ampoule walls was observed. Based on this, a special set of experiments was performed, where the individual parts of the system were measured separately (Table 1), and it was found that the sorption to the walls increases with increasing L/S. One possible explanation is the binding and interaction of <sup>152</sup>Eu with the ampule material. Another option is the formation of colloids with <sup>152</sup>Eu, which during

**Table 1** Determined amount of <sup>152</sup>Eu present in individual parts of the experimental system

L/S (L kg <sup>-1</sup> )	HCP CEM I (%)	Liquid phase (%)	Ampoules (%)
100	92.4	1.1	6.5
250	70.7	1.6	27.7
500	52.9	2.8	44.3
800	43.5	2.7	53.8

The error of determination of the sorption value in the liquid is 0.5%, on CEM I 3.6% and on the walls of the ampoules 3.6%

centrifugation reach the walls of the ampoule where they remain until desorption [15]. All listed below europium sorption results take wall sorption into account.

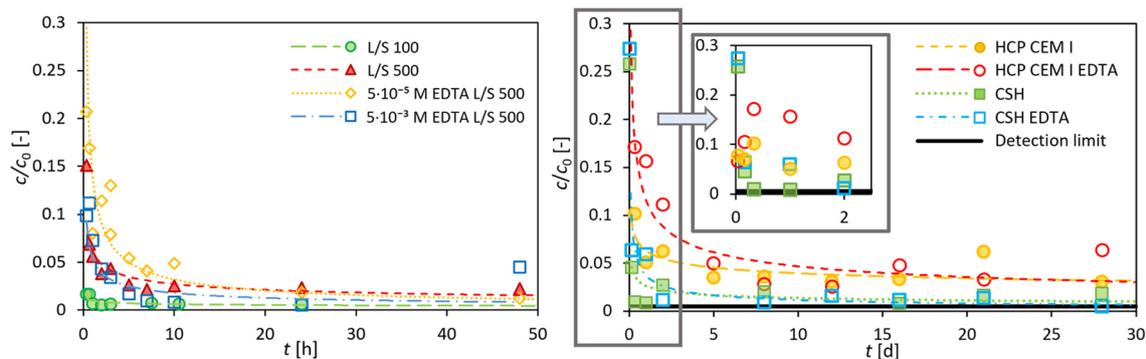
Experiments with uranium are also affected by the sorption on the walls of experimental ampoules. Specifically, the sorption of uranium on the walls of the ampoules in the CSH system was determined to reach 5.4%, independently of the U or EDTA concentration. The sorption on the walls in the system with U-HCP without organics reached 36 ± 12%, that is significantly higher than expected and without a comparable trend as in the case of Eu. The result can be affected by the precipitation of carbonates coming from CO<sub>2</sub> in the air, as the experiment with HCP was not hold in inert atmosphere.

From the results so far, it is clear that sorption on the walls of the reaction ampoules is a significant problem for the system containing Eu and in the system with U-HCP, while for the system U-CSH 1.0 probably due to work in protective atmosphere this phenomenon can be neglected.

### Kinetic experiments

Regarding the kinetics of <sup>152</sup>Eu sorption on HCP CEM I, no significant effect of the addition of EDTA (in both concentration) on the time required to reach equilibrium was observed (Fig. 1 left). For all experiments equilibrium was reached within 24 h. The results show that at the lower phase ratio (100 L kg<sup>-1</sup>) equilibrium occurs significantly faster.

Kinetic experiments with <sup>233</sup>U on HCP CEM I and CSH 1.0 showed influence of lower conc. EDTA that slightly slows down the kinetics (Fig. 1 right). However, equilibrium is reached for both arrangements (with/without EDTA) at the same time, earlier in the CSH system. In the given concentration range, the determination of uranium is more problematic (mainly with regard to the alkalinity of the solution) and burdened with a relatively large error. In this experiment, the detection limit of the method used was verified and it was found that the uptake of <sup>233</sup>U at a given concentration is measurable, but in CSH system the concentration in the liquid phase after the end of the experiment is close to the detection limit (Fig. 1 right). A power trendlines are added



**Fig. 1** Kinetics of  $^{152}\text{Eu}$  sorption on HCP CEM I with L/S ratios 100 and 500  $\text{L kg}^{-1}$  without EDTA and in the presence of EDTA ( $5 \cdot 10^{-5}$  and  $5 \cdot 10^{-3}$   $\text{mol L}^{-1}$ )—left; kinetics of  $^{233}\text{U}$  sorption on HCP CEM

I and CSH 1.0 in the presence/absence of EDTA ( $5 \cdot 10^{-5}$   $\text{mol L}^{-1}$ )—right. (Color figure online)

**Table 2** Average values of distribution ratios  $R_d$ , distribution coefficients  $K_d$  (determined using  $K_d$ -model) and sorption reduction factor values (SRF) for  $^{152}\text{Eu}$  and  $^{233}\text{U}$  sorption on HCP CEM I in presence of EDTA

System	Average $R_d$ ( $\text{L kg}^{-1}$ )	$K_d$ ( $\text{L kg}^{-1}$ )	SRF
$^{152}\text{Eu}$	$(11.9 \pm 2.2) \cdot 10^3$	$(9.8 \pm 0.7) \cdot 10^3$	×
$^{152}\text{Eu}$ EDTA $5 \cdot 10^{-5}$ $\text{mol L}^{-1}$	$(31.9 \pm 6.0) \cdot 10^3$	$(16.0 \pm 2.9) \cdot 10^3$	$0.37 \pm 0.10$
$^{152}\text{Eu}$ EDTA $5 \cdot 10^{-3}$ $\text{mol L}^{-1}$	$(25.7 \pm 4.8) \cdot 10^3$	$(26.0 \pm 2.9) \cdot 10^3$	$0.47 \pm 0.12$
$^{233}\text{U}$	$(2.7\text{--}12.5) \cdot 10^{3*}$	×	×
$^{233}\text{U}$ EDTA $5 \cdot 10^{-5}$ $\text{mol L}^{-1}$	$(2.0\text{--}12.9) \cdot 10^{3*}$	×	$1.05 \pm 0.40$

\*Range of  $R_d$  for non-linear dependencies of  $R_d$  on L/S

for improving clarity. The average determination uncertainty for U-HCP is 13%, for U-CSH 12%, and for Eu-HCP 18% (also applies to equilibrium experiments).

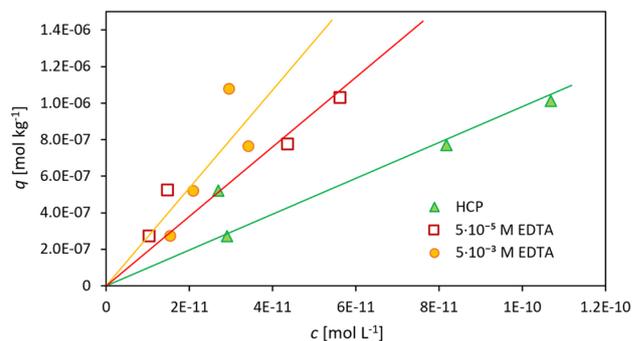
### Equilibrium experiments with HCP CEM I

In all performed europium sorption experiments on HCP CEM I, the distribution ratios were in the tens of thousands ( $\text{L kg}^{-1}$ ) (Table 2) which is slightly less than the values reported in the literature [9–11]. Different observed results may be caused due to the different L/S applied (in the literature ranging from 4000 to 100,000  $\text{L kg}^{-1}$ ) and other input conditions such as europium concentration. Equilibrium experiments clearly showed the effect of the presence of EDTA of both concentrations on the  $R_d$  value, as the obtained SRF are less than 0.5 (Table 2). For all studied systems with Eu, the dependence of  $R_d$  on L/S was not observed, therefore these equilibrium experiments were evaluated using the  $K_d$ -model (Fig. 2). The determined values of  $K_d$  are in good agreement with the average  $R_d$  values (Table 2) especially for the system without organics and with higher conc. EDTA.

Regarding  $^{233}\text{U}$  sorption on HCP CEM I, the  $R_d$  values (mostly in the order of  $10^3$   $\text{L kg}^{-1}$  (Table 2)) are in good agreement with the published data [5, 15, 16]. The presence of low conc. EDTA does not affect the system behavior, as the distribution ratios are comparable and the obtained SRF

is close to 1 (Table 2), which confirms the expectation that the concentration of EDTA ( $5 \cdot 10^{-5}$   $\text{mol L}^{-1}$ ) is under the NEC. The dependence of  $R_d$  on L/S shows the increase of  $R_d$  with a higher phase ratio. By changing L/S, the equilibrium composition of the liquid phase changes. At high solid phase content (low L/S ratio) there is a little specific uptake, which could be explained by the high concentration of competing cations that enter the liquid phase by dissolution (Table 2).

The obtained distribution ratios are approximately an order of magnitude lower for  $^{233}\text{U}$  than for  $^{152}\text{Eu}$ . Comparing the sorption behavior of Eu and U on HCP CEM I, it is clear that  $R_d$  of europium does not depend on L/S (i.e., linear isotherm) while U exhibits a nonlinear shape of isotherms. It



**Fig. 2** Linear sorption of  $^{152}\text{Eu}$  on HCP CEM I under different conditions fitted with  $K_d$ -model. (Color figure online)

can be caused by the inhomogeneity of the cement material, which may be different for different L/S ratio, in particular a compound which is rather inert for sorption of U may be present in the sample while it may behave as a sorbent for Eu due to its affinity for many materials. Another possible cause is the stability of the CSH phase due to its equilibrium with the aqueous phase and, conversely, HCP may be partially dissolved in an aqueous environment, with which it is not in equilibrium.

In the Eu system with a higher conc. EDTA, the dependence of  $R_d$  on L/S is observed, so EDTA was evidently above the NEC in this case. For europium there was a significant increase in sorption by adding organic compound (Table 2) as EDTA in a concentration comparable with the dominant Ca probably changes the speciation of Eu (and Ca) in the system. The uranium system with lower conc. EDTA did not show a significant change in  $R_d$  in the presence of organic matter as SRF is approaching 1.

### Equilibrium experiments with CSH 1.0

The CSH experiments were carried out, but the sorption of  $^{152}\text{Eu}$  on the walls of the reaction ampoules is even higher in the CSH system (almost 100%) than in the HCP CEM I system and evaluation was not possible due to measurement under the detection limit.

The obtained  $R_d$  values for the system  $^{223}\text{U}$  ( $c = 7 \cdot 10^{-8} \text{ mol L}^{-1}$ ) on CSH 1.0 are in the order of  $10^4 \text{ L kg}^{-1}$  (Table 3) as expected (c.f. [24, 25]), they do not show a dependence on the phase ratio and therefore they can be evaluated using the  $K_d$ -model. In contrast with the uptake of  $^{233}\text{U}$  on HCP CEM I, the sorption on CSH 1.0 is linear, which is probably related to its homogeneity and stability.

The results obtained from the  $K_d$ -model are in accordance, within the uncertainty, with the determined average  $R_d$  (Table 3). The  $R_d$  of the system with presence/absence of the EDTA differs, but it can be considered as comparable

**Table 3** Average values of distribution ratios  $R_d$ , distribution coefficients  $K_d$  (determined using  $K_d$ -model) and sorption reduction factor values (SRF) for U in system with EDTA and CSH 1.0

System	Average $R_d$ (L $\text{kg}^{-1}$ )	$K_d$ (L $\text{kg}^{-1}$ )	SRF
$^{233}\text{U}$ ( $7 \cdot 10^{-8} \text{ mol L}^{-1}$ )	$(85.8 \pm 23.2) \cdot 10^3$	$(83.9 \pm 7.6) \cdot 10^3$	×
$^{233}\text{U}$ ( $7 \cdot 10^{-8} \text{ mol L}^{-1}$ ) EDTA $5 \cdot 10^{-5} \text{ mol L}^{-1}$	$(72.4 \pm 19.6) \cdot 10^3$	$(59.1 \pm 3.4) \cdot 10^3$	$1.2 \pm 0.3$
U ( $1 \cdot 10^{-5} \text{ mol L}^{-1}$ ) EDTA $5 \cdot 10^{-3} \text{ mol L}^{-1}$	$(13.9\text{--}128.6) \cdot 10^{3*}$	×	$\geq 1.7$

\*Range of  $R_d$  for non-linear dependencies of  $R_d$  on L/S

due to the growing uncertainty when approaching the detection limit and the value of SRF about 1.2 is close to 1 - the system is almost unaffected by the presence of EDTA of lower conc. (Table 3).

In supplementary experiments with higher uranium concentration ( $^{233}\text{U} + ^{238}\text{U}$ ,  $1 \cdot 10^{-5} \text{ mol L}^{-1}$ ) made possible by the higher solubility of uranium in a system with a pH of 10.7 uptake increased, which for the system without organic and with lower conc. EDTA led to exceeding the detection limit, so it was not possible to determine  $R_d$ 's and SRF. However, the experiment with EDTA at higher conc. showed, that in this case the sorption is reduced with the SRF to be 1.7 or higher. The obtained  $R_d$  values increase with higher L/S. Due to the considerable unreliability of the values near to the detection limit, these were not used to determine the shape of the isotherm.

### Conclusions

Experimental studies of the kinetics of Eu ( $c_{\text{init}} = 2.9 \cdot 10^{-9} \text{ mol L}^{-1}$ ) and U ( $c_{\text{init}} = 7 \cdot 10^{-8} \text{ mol L}^{-1}$  for  $^{233}\text{U}$  and  $1 \cdot 10^{-5} \text{ mol L}^{-1}$   $^{233}\text{U}$  with  $^{238}\text{U}$  as a carrier) uptake on cementitious materials showed a significantly faster uptake of Eu. For the uranium the significant difference was evident between the time required to establish equilibrium in the system with HCP CEM I and CSH 1.0, as the system containing pure cementitious phase reached equilibrium approximately three times faster. Presence of EDTA ( $5 \cdot 10^{-5} \text{ mol L}^{-1}$  and  $5 \cdot 10^{-3} \text{ mol L}^{-1}$ ) did not significantly affect the sorption rate of both studied radionuclides in all studied arrangements.

For the system with HCP without addition of the EDTA, observed values of the distribution ratios for U were in the range  $(2.7\text{--}12.5) \cdot 10^3 \text{ L kg}^{-1}$  due to non-linear behavior while for Eu linear sorption occurred with  $R_d = (11.9 \pm 2.2) \cdot 10^3 \text{ L kg}^{-1}$ .

The presence of EDTA in the U-HCP system had no significant effect on sorption. In contrast to uranium experiments, a significant increase in  $R_d$  values was observed for europium sorption in the presence of organic matter (the highest determined  $R_d = (31.9 \pm 6.0) \cdot 10^3 \text{ L kg}^{-1}$  was found for a lower concentration ( $5 \cdot 10^{-5} \text{ mol L}^{-1}$ ) of EDTA.

Sorption experiments with a CSH phase and cementitious material HCP CEM I within several conditions confirmed the dominant role of CSH for sorption of U. With increasing concentration of U sorption on CSH increased. The effect of EDTA was more pronounced for sorption on CSH with SRF higher than 1.2.

Europium wall sorption increased significantly with increasing L/S. Based on the results of a separate set of experiments, the effect of Eu sorption on the walls of the reaction ampoules was corrected depending on L/S. On

contrary, uranium wall sorption was found to be independent on the phase ratio and this effect was corrected appropriately.

**Acknowledgements** The research leading to these results has received funding from the European Union's Horizon 2020 Innovation Programme under grant agreement n° 847593 (EURAD – CORI). The output was created with the financial participation of SÚRAO (Czech Radioactive Waste Repository Authority) (SO2020 017). This contribution is also partially a result of European Regional Development Fund-Project “Centre for Advanced Applied Sciences” (Grant No. CZ.02.1.01/0.0/0.0/16\_019/0000778) and the grant of the CTU Student Grant Scheme No. SGS22/187/OHK4/3T/14.

**Open Access** This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

## References

- Jantzen C, Johnson A, Read D, Stegemann JA (2010) Cements in waste management. *Adv Cem Res* 22:225–231. <https://doi.org/10.1680/adcr.2010.22.4.225>
- Lagerblad B (2001) Leaching performance of concrete based on studies of samples from old concrete constructions. SKB Tech Rep 01–27 1–80
- IAEA (2009) IAEA Safety Standards Classification of Radioactive Waste for Protecting People and the Environment. Gen Saf Guid No GSG-1. <https://doi.org/10.1097/00004032-197407000-00015>
- Park JB, Kim CW, Kim SH, Kim JY (2012) Determination of a representative composition of dry active waste from Korean nuclear power plants. *Ann Nucl Energy* 42:89–95. <https://doi.org/10.1016/j.anucene.2011.12.004>
- Wieland E, Kosakowski G, Lothenbach B, Kulik DA (2020) Geochemical modelling of the effect of waste degradation processes on the long-term performance of waste forms. *Appl Geochem* 115:104539. <https://doi.org/10.1016/j.apgeochem.2020.104539>
- Bradbury MH, Van Loon LR (1998) Cementitious Near-Field Sorption Data Bases for Performance Assessment of a LLILW Repository in a Palfris Marl Host Rock. CEM-94: UPDATE I, June 1997. PSI Bericht Nr. 98 – 01.
- Ochs M, Mallants D, Wang L (2016). Sorption Values for Palladium and Lead. In: Radionuclide and Metal Sorption on Cement and Concrete. Topics in Safety, Risk, Reliability and Quality, vol 29. Springer, Cham. <https://doi.org/10.1007/978-3-319-23651>
- Maes N, Glaus M, Baeyens B, Marques Fernandes M, Churakov S, Dähn R, Grangeon S, Tournassat C, Geckeis H, Charlet L, Brandt F, Poonosamy J, Hoving A, Havlova V, Fisher C, Scheinost A, Noseck U, Britz S, Siitari-Kauppi M, Missana T (2021) State-of-the-Art report on the understanding of radionuclide retention and transport in clay and crystalline rocks. Final version as of 30.04.2021 of deliverable D5.1 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593
- Glaus MA, Laube A, Van Loon LR (2004) A generic procedure for the assessment of the effect of concrete admixtures on the sorption of radionuclides on cement: Concept and selected results. *Mater Res Soc Symp Proc* 807:365–370. <https://doi.org/10.1557/proc-807-365>
- Wieland E, Tits J, Spieler P, Dobler JP (1998) Interaction of Eu(III) and Th(IV) with sulphate-resisting Portland cement. *Mater Res Soc Symp - Proc* 506. <https://doi.org/10.1557/proc-506-573>
- Tits J, Bradbury MH, Wieland E, Mantovani M (1998) The Uptake of Cs, Sr, Ni, Eu and Th by CSH Phases under high pH Cement Pore Water Conditions.67
- Pointeau I, Piriou B, Fedoroff M, Barthes MG, Marmier N, Fromage F (2001) Sorption mechanisms of Eu<sup>3+</sup> on CSH phases of hydrated cements. *J Colloid Interface Sci* 236:252–259. <https://doi.org/10.1006/jcis.2000.7411>
- Wieland E, Loon LR Van (2003) Cementitious near-field sorption data base for performance assessment of an ILW repository in Opalinus Clay. *PSI Bericht* 03–06 87
- Wieland E, Macé N, Dähn R, Kunz D, Tits J (2010) Macro- and micro-scale studies on U(VI) immobilization in hardened cement paste. *J Radioanal Nucl Chem* 286:793–800. <https://doi.org/10.1007/s10967-010-0742-y>
- Tits J, Wieland E (2018) Actinide Sorption by Cementitious Materials. *PSI Rep* 18 – 02 1–88
- Allard B, Eliasson L, Höglund S, Andersson K (1984) Sorption of Cs, I and actinides in concrete systems. SKB Tech Rep 84 – 15 1–30
- Pointeau I, Landesman C, Giffaut E, Reiller PE (2004) Reproducibility of the uptake of U (VI) onto degraded cement pastes and calcium silicate hydrate phases. *Radiochim Acta* 92:645–650
- Ochs M, Dolder F, Tachi Y (2022) Decrease of radionuclide sorption in hydrated cement systems by organic ligands: comparative evaluation using experimental data and thermodynamic calculations for ISA/EDTA-actinide-cement systems. *Appl Geochem* 136:105161. <https://doi.org/10.1016/j.apgeochem.2021.105161>
- Ochs M, Colàs E, Grivé M, Olmeda J, Campos I, Bruno J (2014) Reduction of radionuclide uptake in hydrated cement systems by organic complexing agents: Selection of reduction factors and speciation calculations. Rep R-14-22 1–58
- Atkins M, Glasser FP, Kindness A (1992) Cement hydrate phase: solubility at 25°C. *Cem Concr Res* 22:241–246. [https://doi.org/10.1016/0008-8846\(92\)90062-Z](https://doi.org/10.1016/0008-8846(92)90062-Z)
- Lange S, Kowalski PM, Pšenička M, Klinkenberg M, Rohmen S, Bosbach D, Deissmann G (2018) Uptake of <sup>226</sup>Ra in cementitious systems: A complementary solution chemistry and atomistic simulation study. *Appl Geochem* 96:204–216. <https://doi.org/10.1016/j.apgeochem.2018.06.015>
- Hummel W (1993) Organic complexation of radionuclides in cement pore water: A case study. *PSI Intern Rep* 1–33
- Brownsword M, Buchan AB, Ewart FT, McCrohon R, Ormerod GJ, Smith-Briggs JL, Thomason HP (1990) The solubility and sorption of uranium(VI) in a cementitious repository, *MRS Proceedings*, 176:577. <https://doi.org/10.1557/PROC-176-577>
- Häußler V, Amayri S, Beck A, Platte T, Stern TA, Vitova T, Reich T (2018) Uptake of actinides by calcium silicate hydrate (C-S-H) phases. *Appl Geochem* 98:426–434. <https://doi.org/10.1016/j.apgeochem.2018.08.021>
- Tits J, Fujita T, Harfouche M, Dähn R, Tsukamoto M, Wieland E (2014) Radionuclide uptake by calcium silicate hydrates: case studies with th (IV) and U (VI) Nuclear Energy and Safety Research Department. *PSI Bericht Nr* 14 – 03 1–104

**Publisher's Note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.