Effect of Natural Organic Matter on Radionuclide Partitioning in Nuclear Fuel Storage Ponds – 17136

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ABSTRACT

One of the nuclear fuel storage ponds at Sellafield is open to the air, and has contained a significant inventory of corroded Magnox fuel and sludge for several decades, with some fission products having been released into solution. The sludge is predominantly composed of brucite ($Mg(OH)_2$), a corrosion product from the magnesium-based fuel cladding, amongst other corrosion products resulting from the uranium metal fuel. Sr-90 is known to constitute a small mass of the radionuclides present in the pond, but due to its solubility and activity, it is a radionuclide that is at risk of challenging effluent discharge limits. Organic molecules are known to be present also due to the ponds being open to the air, with occasional algal blooms restricting visibility. Understanding the chemical interactions of these components is important to inform ongoing sludge retrievals and effluent management. Batch sorption-desorption experiments have been performed with brucite, Sr-90 and natural organic matter (NOM) (namely humic acid (HA) and *Pseudanabaena catenata* cyanobacterial growth supernatant) in both binary and ternary systems at high pH. Ionic strength, pH and order of addition of components were varied. Sr-90 was shown not to interact strongly with the bulk brucite surface in binary or ternary systems under pH conditions relevant to the pond. Humic acid in both binary and ternary systems demonstrated a strong affinity for the brucite surface. In ternary systems HA appeared to enhance sorption of Sr-90, and vice versa, regardless of the order of addition of components. There was no distinct effect of varying ionic strength or pH.

INTRODUCTION

The contents of one of the nuclear fuel storage ponds at Sellafield need to be retrieved imminently for safe storage as a matter of high priority, but the removal and reprocessing of such a complex undefined inventory poses a unique challenge[1]–[3]. Brucite ($Mg(OH)_2$), derived from corrosion of the Magnox cladding, is a major component of the sludge that has accumulated at the bottom of the pond. The pH of the pond is usually between 10.5 and 11.5, and brucite typically buffers solutions towards pH 10.8-11, the value of its pH_{pzc}. As brucite is a major component of the sludge, it is likely that the pH of the sludge is also in this region. Organic molecules are known to be present in the pond, which is an open-air structure that has been in place for several decades. These organics are expected to comprise chemical degradation products of natural organic matter (NOM, e.g. humic substances), and algal or cyanobacterial organisms that cause occasional algal blooms. These blooms can further restrict visibility, as well as complicate the chemical interactions of the pond components. Sr-90 is known to constitute a small mass of the radionuclides present in the pond, but due to its solubility and activity, it is a radionuclide that is most at risk of challenging effluent discharge limits. Retrievals of sludge have already begun, and it is essential to understand the chemistry of these systems (particularly that of Sr-90 and Cs-137) to ensure the safe management of these legacy wastes.

Humic substances (HS) are a significant component of NOM and are formed through chemical degradation of organic material. Humic substances form strong complexes with cations[4] and therefore are an important consideration for understanding the chemistry of Sr-90 within the pond. Complexation of strontium with humic substances has been well researched[5]–[9]. Several studies note increasing strontium concentrations will increase precipitation of strontium-humate complexes, and that Mg^{2+} is a competitor for Sr^{2+}, which may substitute for it in the formation of humate complexes[5], [6], [10]. With increasing pH, association of strontium with humics is also expected to increase as a result of an increased negative charge on the humic acid (HA) molecule due to a greater extent of dissociation of carboxylic acid and phenolic groups[5], [11]. The apparent stability constant for strontium humates has been investigated for the pH range 4-10, and has been shown to increase with pH but level off at higher pH values indicating saturation was reached[12]. The concentrations of HA investigated are at least 10 times higher than those in this work. In the presence of metal ions, humic acid can form bridged aggregates also[13], [14]. Humic-metal interactions are often roughly divided into two fractions; those that bind but are rapidly exchangeable, and those that are slowly exchangeable[15]. Slow metal ion dissociation kinetics have been demonstrated for dissolved organic matter (DOM) at high pH in a number of studies[8], [16]–[18].

There is much discussion in the literature regarding the behaviour and composition of humic substances as they encompass a very heterogeneous group of molecules, with varied molecular weights and functional groups[12]. Some studies categorise the behaviour of humics as supramolecular assemblies of simple molecules, whilst others do so as macromolecules with aromatic or aliphatic character; some recognise behaviour that demonstrates a combination of the two (Figure 1)[13], [14].

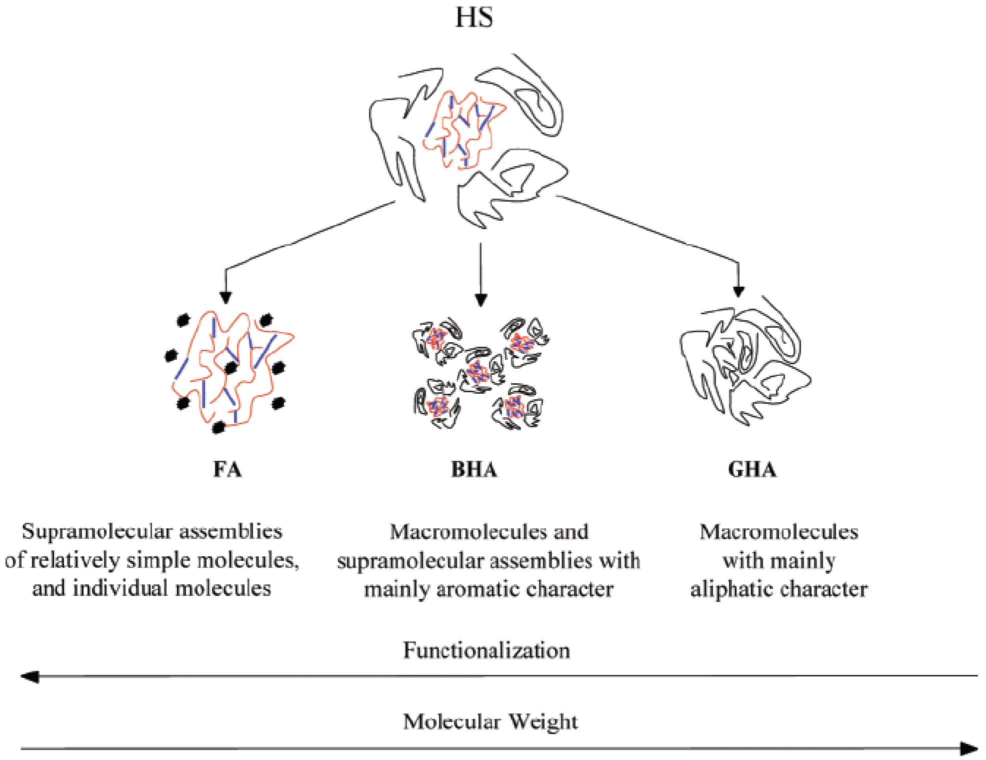

Figure 1: Molecular patterns associated with different fractions of humic substances, where FA = fulvic acid, BHA = brown humic acid and GHA = grey humic acid[13].

Pre-equilibration of mineral surfaces with NOM has been investigated by a number of studies, and shown to suppress sorption of the metal in ternary systems[17], [19]–[22]. This may be a result of organics physically blocking metal sorption sites, or a mixture of size fractions of HA present resulting in varied metal-humic interactions in solution[22].

Reported visual observations of the pond have indicated the presence of green algal blooms on the water's surface obscuring water clarity (Figure 15)[23]. Algal samples from pond samples at Sellafield have been found to contain *Pseudanabaena catenata*, a type of cyanobacteria. Cyanobacteria, or blue-green algae, are prokaryotic unicellular or filamentous organisms with no cell sub-structures such as chloroplasts or mitochondria. Their photosynthetic pigments contain mostly chlorophyll-a (green) and phycocyanin (blue), creating the blue-green colour from which they get their name. *P. catenata* is a long narrow or 'single trichome' bacterium in shape with cells that are approximately 1.8-2.2 µm wide and 2-5 µm long (Figure 2)[24]. Information in the literature regarding this particular type of cyanobacteria is extremely limited.

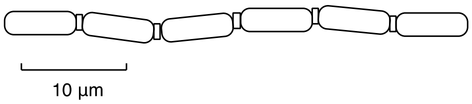

Figure 2. Structure of *P. catenata*.

Sorption of strontium to a range of surfaces including clay minerals and oxides is often described as outer-sphere complexation, although inner sphere complexation is also known[25]–[30].

Investigation of these systems provides insight into, and understanding of, the effect of humic acid and *P. catenata* growth supernatant on Sr-90 partitioning to brucite at high pH. The majority of previous sorption-desorption work for Sr-90 has been conducted at neutral to acidic pH values, and mostly without NOM. These studies are particularly important for the characterisation of legacy wastes at Sellafield, to ensure their safe retrieval and storage.

EXPERIMENTAL

All materials were of analytical reagent grade. $Mg(OH)_2$ (Fluka) was >99% pure. Humic acid was supplied by Sigma Aldrich. Adjustments of initial pH were made using 0.1 mol dm^{-3} NaOH, with NaOH pellets provided by Fisher Scientific. Ionic strength adjustments were made using $NaOCl_4$ stock solutions of 0.01 and 1 mol dm^{-3}. The scintillant cocktail Scintisafe 3 was provided by Fisher Chemical. Deionised water purified in a Millipore system was used throughout the experiments. pH adjustments were made before addition of the solid and the pH was not controlled thereafter. All batch experiments were performed in triplicate in a CO_2-free atmosphere glove box to avoid the presence of unwanted carbonate species in solution. Brucite starting material was analysed by XRD and N_2 adsorption surface area analysis. Humic acid was analysed by a Thermo iCap 6300 Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) for trace metal analysis. Vivaspin ultracentrifugation membranes made from polyethersulfone (PES) with pore cut off sizes of 3 and 100 kDa were used for ultrafiltration of solutions.

Preparation of Sr-90 standards

297 mL deionised water was added to 3 mL of Sr-90 stock solution (1 kBq mL^{-1}) and left to equilibrate for at least 24 hours prior to separation into 3 standards of 10 Bq mL^{-1}.

Preparation of humic acid stock solution

0.1 g of humic acid was dissolved in NaOH (2 mL, 0.1M) and made up to 100 mL with deionised water. The same humic acid stock solution was used for standards and across all batch experiments.

Contents of BG11 medium for growth of cyanobacterial cultures

BG11 medium was used to grow cultures of the cyanobacteria *Pseudanabaena catenata*. BG11 contains a combination of different stock solutions (Table 1), and those stocks are combined form the final BG11 growth medium as shown in Table 2.

TABLE I. Contents of stock solutions used to make BG11 growth medium.

Stock	**Component**	**Concentration**
1	$NaNO_3$	15.0 g per litre
2	K_2HPO_4	2.0 g per 500 mL
3	$MgSO_4.7H_2O$	3.75 g per 500 mL
4	$CaCl_2.2H_2O$	1.80 g per 500 mL
5	Citric acid	0.30 g per 500 mL
6	Ammonium ferric citrate green	0.30 g per 500 mL
7	$EDTANa_2$	0.05 g per 500 mL
8	Na_2CO_3	1.0 g per 500 mL
9	Trace metal solution: H_3BO_3 $MnCl_2.4H_2O$ $ZnSO_4.7H_2O$ $Na_2MoO_4.2H_2O$ $CuSO_4.5H_2O$ $Co(NO_3)_2.6H_2O$	Concentration per litre: 2.86 g 1.81 g 0.22 g 0.39 g 0.08 g 0.05 g

TABLE 2. Volumes of stock solutions combined to create BG11 growth medium.

Medium	per Litre
Stock solution 1	100 mL
Stock solutions 2-8	10 mL each
Stock solution 9	1 mL

Binary sorption-desorption experiments

All experiments were performed in a CO_2-free atmosphere, to exclude unwanted carbonate species in solution, and reduce the possibility of significant pH drift. Aqueous solutions were prepared containing Sr-90 tracer (10 Bq mL^{-1}, 2.17 x 10^{-11} mol dm^{-3}), or humic acid (5 ppm). For pH variance investigations, the initial pH was adjusted to 10.5, 11 and 11.5 (with some conducted at 12.5 also) to cover the range exhibited within the pond. $NaOCl_4$ solution was used to adjust the ionic strength (2.5 x 10^{-3}, 0.01 and 0.1 mol dm^{-3}) and subsequently the initial pH was adjusted to 10.5. These solutions were added to bottles containing pre-weighed brucite powder (0.1 g, 1 g L^{-1}). All experiments were performed in triplicate. Samples were taken at regular intervals and analysed for Sr-90, and humic acid content as appropriate. The content remaining in solution of each component was calculated as a percentage. Error bars in figures depicting the content of each component in solution represent errors of 2σ from the mean.

Desorption experiments were conducted by carefully removing the supernatant from a system (care was taken to ensure minimal disturbance of the bulk solid). The supernatant was then refreshed with the same volume of liquid at the same pH as that of the original. Samples were taken at regular intervals, to determine the Sr-90 and humic acid content released back into solution. This step was only performed for pH variance experiments. Distribution coefficients were calculated for both Sr-90 and HA at the end of periods of sorption and desorption.

Ternary sorption-desorption experiments

All experiments were performed in a CO_2-free atmosphere, to exclude unwanted carbonate species in solution, and reduce the possibility of significant pH drift. Aqueous solutions were prepared containing Sr-90 tracer (10 Bq mL^{-1}, 2.17 x 10^{-11} mol dm^{-3}), and/or humic acid (5ppm), and the initial pH was adjusted to 10.5, 11 or 11.5. These solutions were added to bottles containing pre-weighed brucite powder (0.1 g, 1 g L^{-1}) in the case of brucite-humic acid or brucite-Sr-90 equilibration. For experiments in which brucite was added later, the powder was carefully added, gently mixed and allowed to settle. Samples were taken at regular intervals and analysed for Sr-90, Mg^{2+}, and humic acid content as appropriate. Desorption experiments were conducted in the same way as for the binary experiments. Sr-90 and HA were analysed in the same way as for binary experiments. Mg^{2+} in solution was presented as concentrations in ppm. In the figures, dashed lines indicate the point at which the third component was added. Error bars in figures depicting the content of each component in solution represent errors of 2σ from the mean. Distribution coefficients for Sr-90 and HA were calculated at the end of the sorption and desorption steps. The distribution coefficient Kd, is defined as;

$$\mathrm{Kd} = \frac{\text{sorbed concentration (ml/g))}}{\text{solution concentration (ml/g))}}$$

Equation 1. Distribution coefficient.

For cyanobacterial supernatant-Sr-90-brucite ternary experiments the cyanobacterial supernatant sample was added in place of the humic acid. All other steps were the same, with only Sr-90 content analysed in solution. The cyanobacterial supernatant sample was taken at day 21 of its growth (approximately 4 days prior to signs of significant cell death), and had a chlorophyll-a concentration of ~1200 μg/L.

Analysis of Sr-90 content

Sr-90 content was analysed using liquid scintillation counting. 0.2 mL aliquots were acidified with 2 mL HCl (2%) and left for 3 weeks to reach secular equilibrium before addition of 5 mL scintillant cocktail. Samples were left to equilibrate with the scintillation fluid for at least 12 hours before counting on a Wallac Quantulus 1220 Ultra Low Level liquid scintillation spectrometer.

Ultrafiltration of supernatant samples

0.5 mL samples of supernatant were centrifuged using Vivaspin ultracentrifugation membranes with membrane filter cut-offs at 3 and 100 kDa. Subsequent analysis of Mg content in the filtrate was carried out as detailed below.

Analysis of magnesium content

Samples were taken from the supernatant and diluted to 5 mL with HNO_3 (2%) to <0.4 Bq mL^{-1}. Magnesium content was analysed using a Perkin Elmer Optima 5300 Dual View ICP-AES.

Analysis of humic acid content

The absorbance of 1 mL samples was measured in 2 mL polymethylmethacrylate (PMMA) cuvettes at 254 nm using a Shimadzu UV-1800 Spectrophotometer[31].

Analysis of chlorophyll-a concentration

In order to measure the photosynthetic biomass in the growth cultures the concentration of chlorophyll-a (Chl-a) was determined. 1 mL samples of *P. catenata* were centrifuged at 14,000 g for 10 minutes, the supernatant discarded, and the cells re-suspended in 1 mL of 70 % ethanol. These suspensions were incubated at room temperature for 2 hours, with the resulting samples centrifuged at 14,000 g for 10 minutes. The supernatant was then removed and analysed using a Jenway 6700 UV/Vis spectrophotometer. The absorbance was measured at 665 nm (Chl-a) and at 750 nm to correct for turbidity[32]. The concentration of Chl-a was then calculated using the formula of Jespersen and Christoffersen (Equation 3)[33].

$$\mathrm{Chl-a}\ \ (\mathrm{\mu g}\ \ \mathrm{L}-1) = \frac{(V_e{\cdot}f{\cdot}A)}{(V_s{\cdot}L)}$$

Equation 2. Determination of the concentration of Chl-a, where V_e = total volume of solvent (ml); A = Absorbance at 665nm - Absorbance at 750nm; V_s= Total volume of sample filtered (litres), L = Cell path length (cm); f= (1/specific extraction coefficient)*1000; where the specific extinction coefficient for Chl-a in 96% (v/v) ethanol is 83.41 g^{-1} cm^{-1}.

RESULTS AND DISCUSSION

Brucite and humic acid characterisation

Brucite starting material was a very fine white powder shown to be consistent with synthetically produced $Mg(OH)_2$ (Figure 3). Surface area analysis using the N_2 gas adsorption BET method calculated the surface area of the brucite at 5.4346 m^2/g. The PHREEQC v.3.3.3 model and LLNL database was used to calculate the surface charge density, σ, versus pH to predict sorption behaviour of the different system components (Figure 4). A site density of 10 sites nm^{-2} was used from Pokrovsky and Schott (2004)[34].

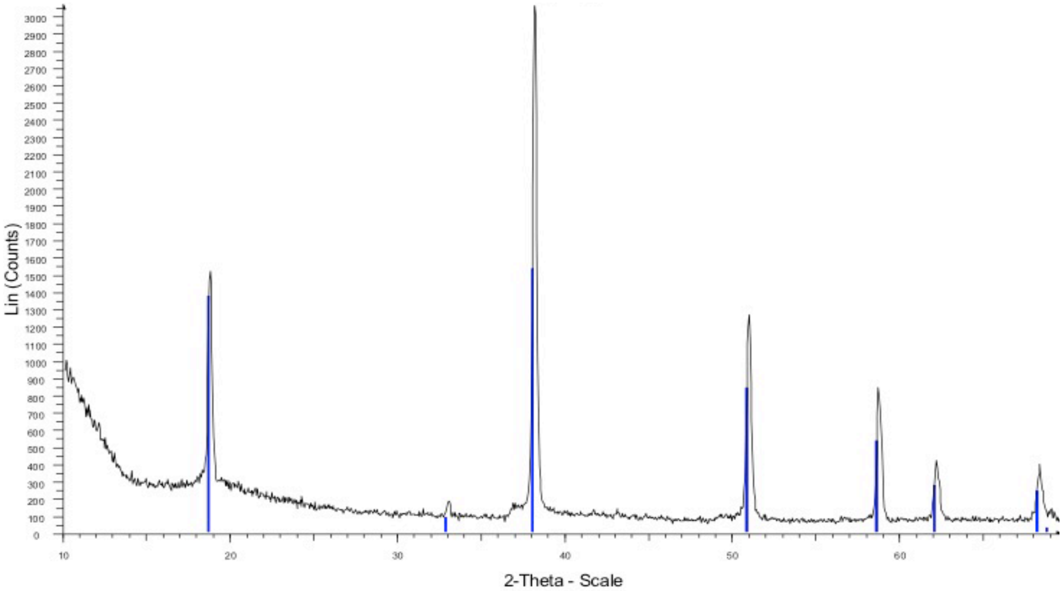

Figure 3. Powder XRD of brucite starting material (black) compared with reference for synthetic brucite (blue).

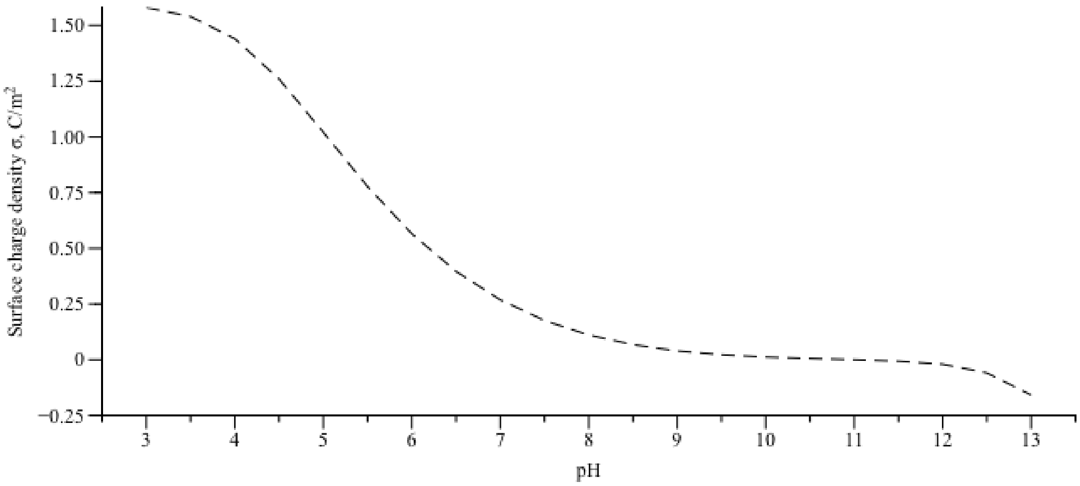

Figure 4. Brucite surface charge density vs. pH modelled with PHREEQC.

Trace metal content of the humic acid starting material was analysed by ICP-OES and is listed in Table 3.

TABLE 3. Trace metal composition of humic acid starting material determined by ICP-OES.

Metal	% Composition
Al	3
Ca	2
Mg	<0.3
Na	4
Fe	1
Cu	<0.3
K	0.7

Brucite-Sr-90 behaviour

Brucite-Sr-90 binary systems were investigated with variance of ionic strength and pH. The content of Sr-90 in solution was monitored over time after addition to brucite, with no sorption seen in either scenario (Figures 5 and 6a). The variation of ionic strength or pH had no impact on the behaviour of Sr-90.

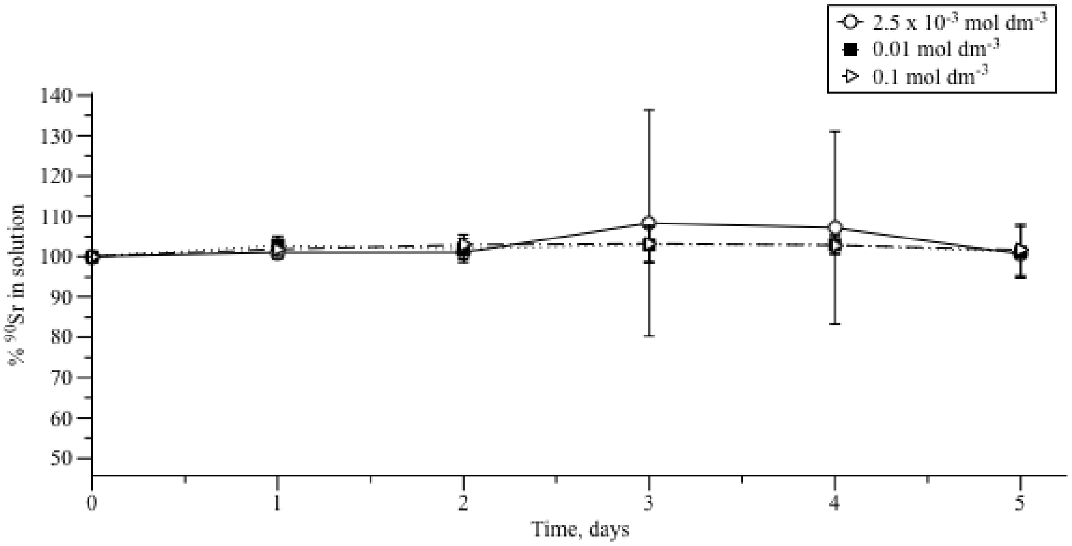

Figure 5. Percentage of Sr-90 in solution after addition to brucite, with variance of ionic strength.

The point of zero charge (pH_{pzc}) of the brucite surface is unusually high at pH 10.8-11[34], [35]. LSC data for pH 10.5 and 11 (not shown) displayed large variations, but no significant sorption of Sr-90 likely due to the proximity of these pH values to the pH_{pzc}. The pH of these two systems also demonstrated buffering towards pH 10.8-10.9 within 7 days. Figure 6a indicates that Sr-90 behaviour at the two different pH values is virtually indistinguishable from one another, despite the model prediction of the surface beginning to accumulate a net negative charge at these pH values. The predicted surface charge across this range however (Figure 4) demonstrates that even at the highest pH of 12.5 this negative charge is still only small (5.90×10^{-2} C m^{-2}).

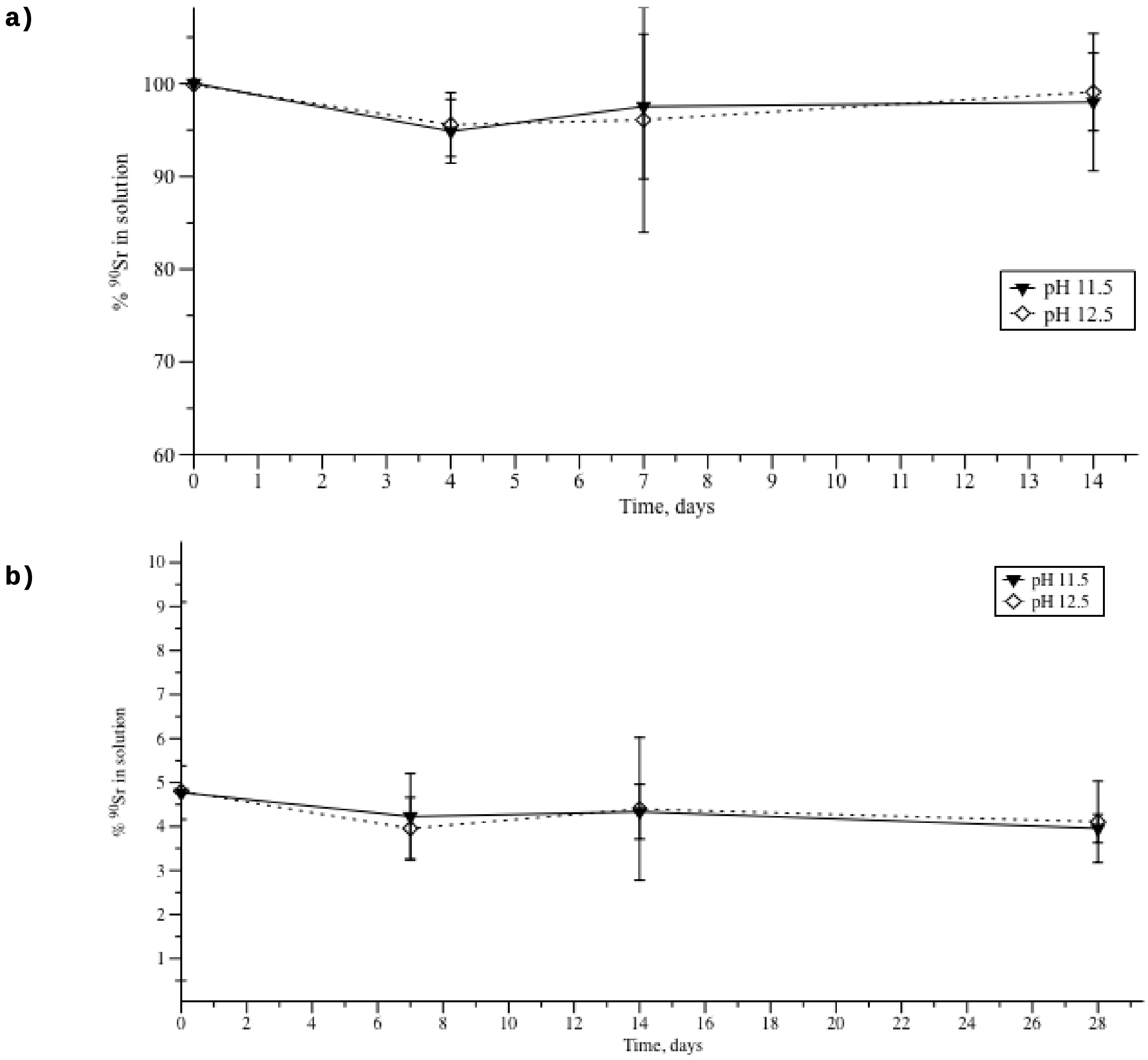

Figure 6. Sr-90 in solution, as a percentage of total initial Sr-90 for **a)** brucite-Sr-90 sorption binary system at pH 11.5 and 12.5; and **b)** brucite-Sr-90 desorption binary system at pH 11.5 and 12.5.

Desorption behaviour of Sr-90 (Figure 6b) suggests that no Sr-90 remains on the brucite surface. In this instance, the Sr-90 seen in solution can be attributed to the few millilitres of residual supernatant that could not be removed without disturbance of the bulk solid.

Brucite-HA behaviour

Brucite-HA binary systems were investigated with variance of ionic strength and pH. The content of HA in solution was monitored over time after addition to brucite, with significant sorption seen in both scenarios (Figures 7 and 8). The variation of ionic strength or pH had no impact on the behaviour on HA.

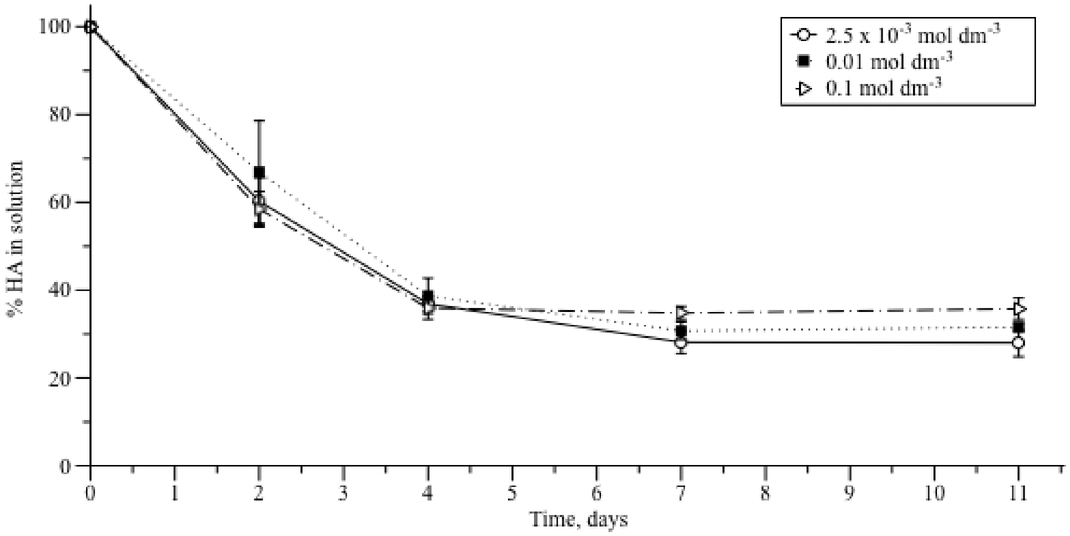

Figure 7. Concentration of HA in solution after addition to brucite, with variance of ionic strength.

Figure 8 also shows comparison between the binary brucite-HA system, and the ternary system where brucite and HA are pre-equilibrated prior to addition of Sr-90 at day 7. There was no immediate effect on HA, however there may be a kinetic component to the behaviour, with some evidence for greater sorption past day 14 in the ternary system. At day 28 there was a significant difference in the percentage of HA remaining in solution, compared with the binary system suggesting that Sr-90 may enhance the sorption of HA. Cation bridging has been suggested as a potential way in which HA sorption is enabled[36].

The predicted negative charges on both the HA and the brucite surface at this elevated pH indicates that electrostatic interactions are not involved in the sorption observed. HA must be binding to the positive sites remaining on the brucite surface, due to proximity of the pH_{pzc}. However, as the sorption is significant, it may only be relative to the concentration of HA used in these studies. Large fractions of sorbed HA have been observed at high pH in a number of previous studies, but increasing the concentrations of HA at constant pH has significantly reduced the sorbed fraction[36], [37]. Precipitation of Mg-humates was ruled out as no brown precipitate was seen after spiking a saturated magnesium solution with HA, and without bulk brucite solid present.

No further investigations with variance of ionic strength were taken forward into ternary systems. Variance of pH was regarded as more likely to significantly impact the chemistry of the real system, and so was taken forward for ternary sorption-desorption experiments.

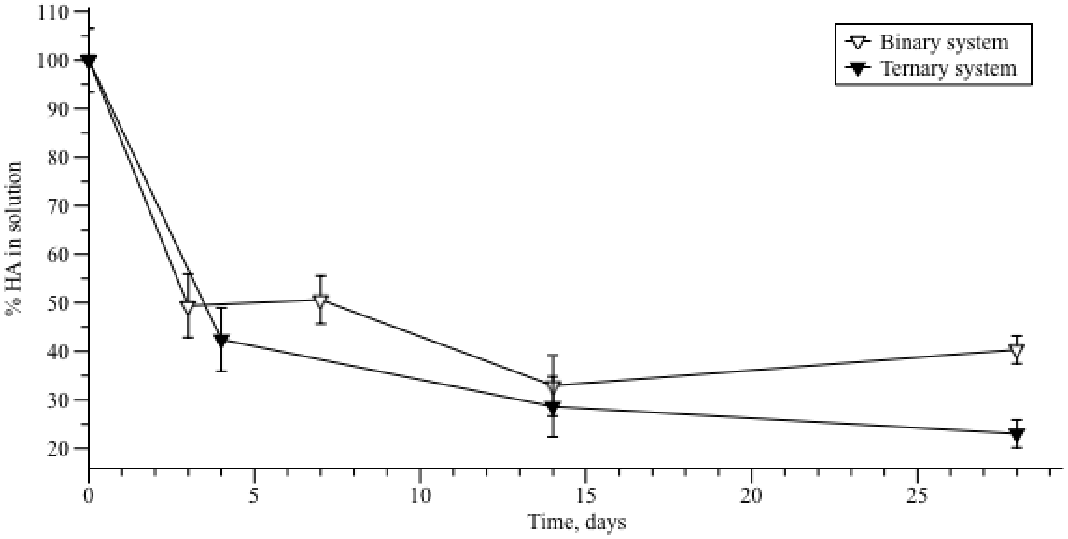

Figure 8. Concentration of HA in solution for both the brucite-HA-Sr-90 ternary system where Sr-90 was added at day 7, and the brucite-HA binary system at pH 11.5.

HA-Sr-90-Brucite ternary system

Pre-equilibration of HA and Sr-90 for 7 days prior to addition of brucite indicated some removal of both HA and Sr-90 from solution (Figure 9a). Since there is no observable removal of Sr-90 by brucite in a binary system (Figures 5 and 6a), this removal in the presence of HA is likely due to the formation of strontium-humate complexes, which are widely reported in the literature[5], [6], [10]. Beyond day 7, a greater proportion of Sr-90 is removed from solution in the pH 11.5 system than the pH 10.5 system. However, the opposite effect is seen for the removal of HA, which is unexpected if the removal of Sr-90 is via strontium-humate complexes. Despite this, there was an enhanced rate of sorption of HA after day 7, with only 5-10% remaining in solution by day 28 across the pH range.

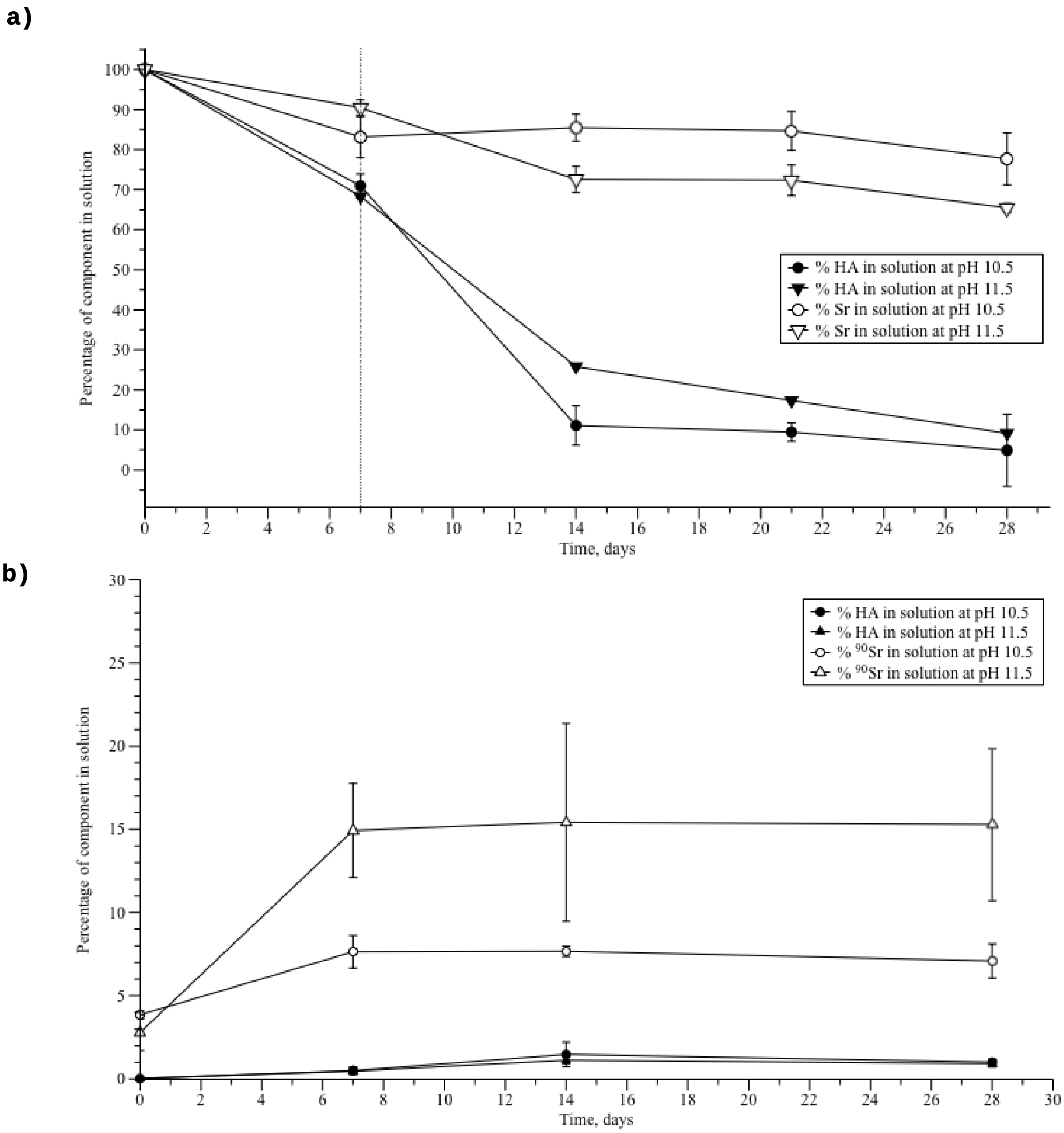

Figure 9. Percentage of total initial HA and Sr-90 remaining in solution in **a)** ternary HA-Sr-90-brucite sorption system at pH 10.5 and 11.5; and **b)** ternary HA-Sr-90-brucite system at pH 10.5 and 11.5 during the desorption step. The dashed line indicates addition of brucite after the sample was taken for that time point.

From desorption at day 28, there was a small release of HA from the brucite surface (Figure 9b). The majority of Sr-90 was removed with the old supernatant, and the Sr-90 seen in solution at the start of desorption can mostly be attributed to the residual supernatant. A significant increase of Sr-90 in solution is seen at pH 11.5, perhaps due to dissociation of some humate complexes as a small amount of HA is released. As no Sr-90 sorbs to the brucite surface in the absence of HA (Figure 5), HA appears to be controlling both the sorption and desorption behaviour of the Sr-90 in this ternary system, particularly at pH 11.5.

No free Mg^{2+} was seen in solution for the duration of the sorption step, after addition of brucite at day 7 for pH 11.5 (Figure 10a). The concentration of Mg^{2+} in solution decreased significantly with each increase in pH, a trend previously reported by Pokrovsky and Schott[34]. Upon ultrafiltration of the supernatant at day 28 (Figure 10b) there was no significant difference between Mg concentrations that passed through either the 3 or the 100 kDa filters, suggesting that no fine colloidal Mg was produced in this system.

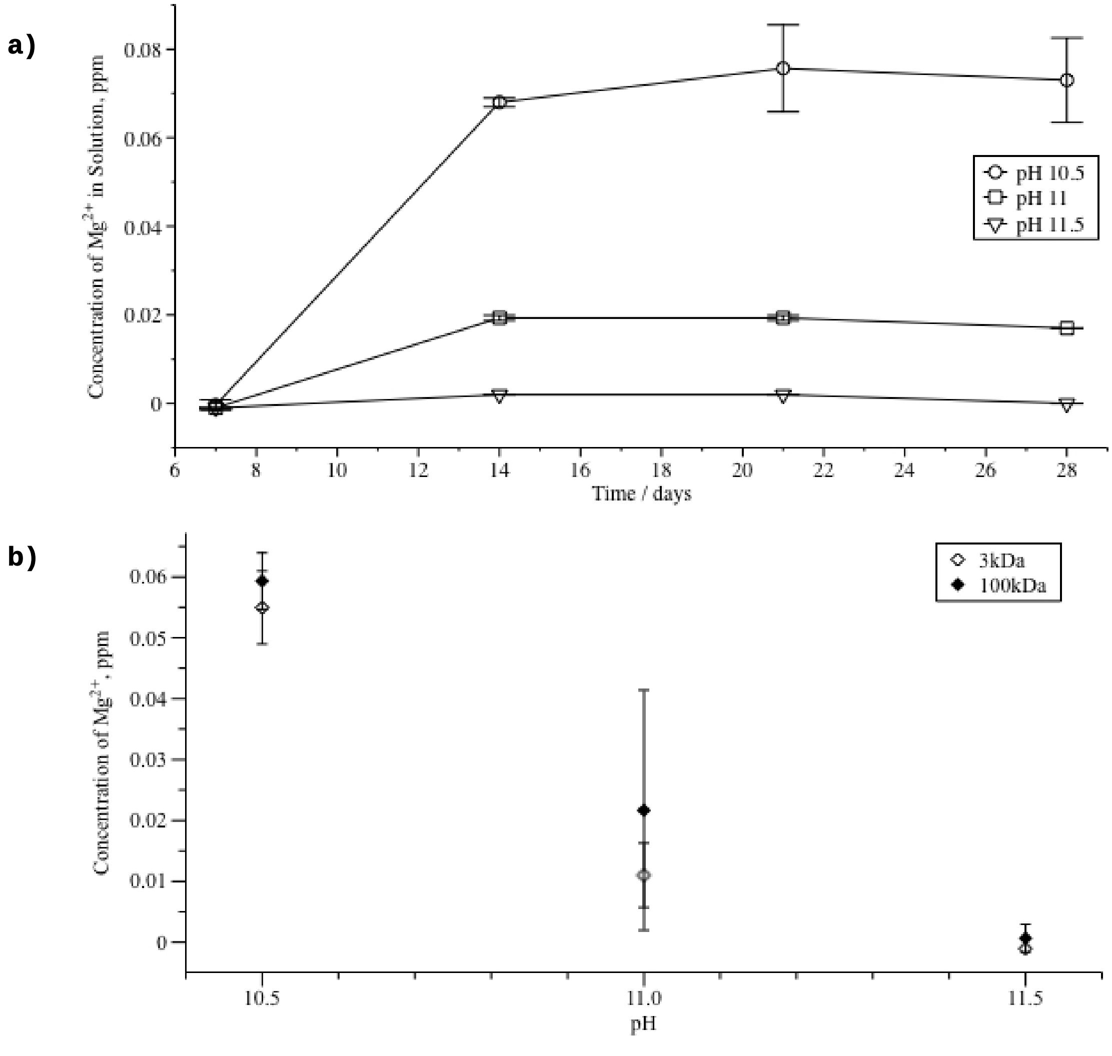

Figure 10. Mg^{2+} concentration in **a)** HA-Sr-90-brucite ternary system solution after addition of brucite at day 7; and **b)** HA-Sr-90-brucite ternary system after ultrafiltration of the supernatant at day 28.

Adsorption and desorption distribution coefficients were calculated for both Sr-90 and HA at the end of each of the sorption and desorption periods (Figure 11). The Sr-90 adsorption coefficients were very small and significantly different compared with desorption coefficients across the pH range. This could suggest that a small amount of Sr-90 is bound irreversibly, however as virtually 100% of Sr-90 remains in solution over the course of the experiment, it may not be calculated with great certainty. In contrast, those for HA indicate that the sorbed HA fraction becomes exchangeable (desorption Kd increases) as pH increases. The exchangeable nature of HA coupled with the increase of Sr-90 in solution seen experimentally (Figure 9b), may suggest that as the HA exchanges some of the strontium-humate complexes are dissociating at pH 11.5.

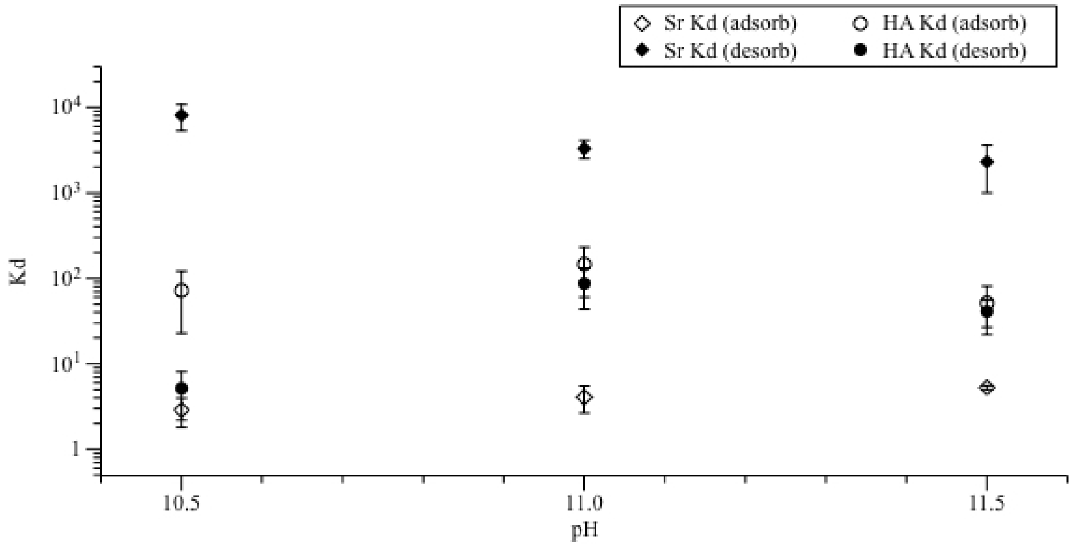

Figure 11. Distribution coefficient values for Sr-90 and HA at the end of the adsorption and desorption steps in ternary HA-Sr-90-brucite system.

Cyanobacterial Growth Supernatant-Sr-90-Brucite Ternary System

Cyanobacterial growth supernatant removed from a *Pseudanabaena catenata* culture was equilibrated with Sr-90 for 7 days. There was no significant removal of Sr-90 from solution during this period, and the addition of brucite at 7 days did not show any alteration in the behaviour of Sr-90 (Figure 12). Bacterial solutions such as these may represent those in pond liquors, but there is no evidence to suggest they have any influence on sorption behaviour of Sr-90 with brucite.

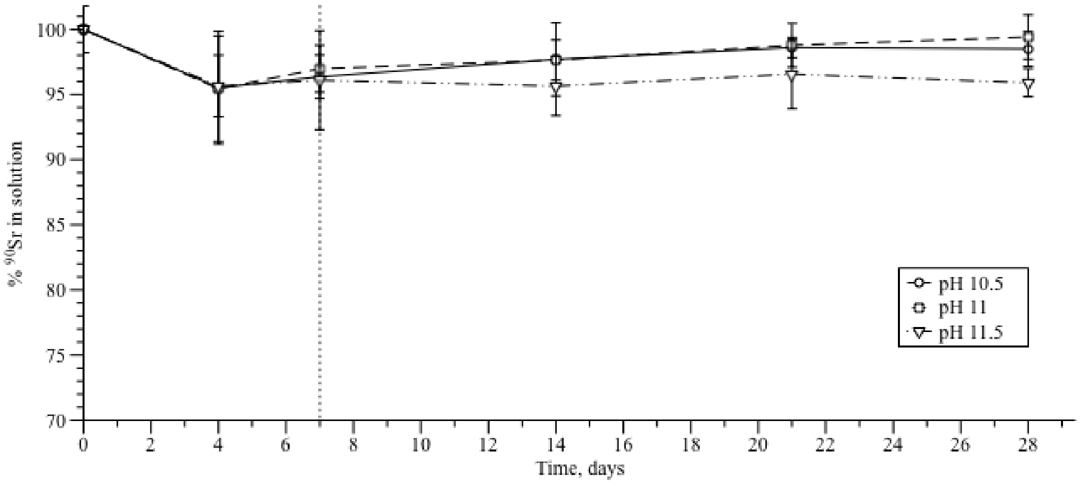

Figure 12. Percentage of initial Sr-90 in solution in cyanobacterial growth supernatant-Sr-90-brucite ternary system, where the growth supernatant and Sr-90 were equilibrated for 7 days prior to the addition of brucite. The dashed line indicates addition of brucite after the sample was taken for that time point.

CONCLUSION

Brucite does not have a significant impact on removing Sr-90 from solution under conditions relevant to the fuel storage pond, but brucite surfaces in the presence of humic acid are liable to have an organic coating. Variance of ionic strength or pH do not cause significant differences in the behaviour of HA or Sr-90 in brucite in binary systems. Sr-90 in the presence of HA forms humate complexes that remain in solution, but in the presence of brucite as a ternary system, the humate complexes will sorb as ternary complexes. As some Sr-90 clearly remains in solution, it could be that only a small fraction of the humate complexes sorb, with the rest remaining in solution. Concentrations of HA in these investigations were small, but perhaps with a higher concentration of HA, more Sr-90 could be removed from solution.

No fine colloidal Mg was present in the ternary system after 28 days suggesting that Sr-90 is not seen in solution as a result of its sorption to a colloidal fraction, but as free Sr^{2+}. Colloidal phases are important to consider for effluent management, particularly if radionuclides associate with them. In the context of the pond, Sr-90 sorption-desorption behaviour should be investigated with other relevant phases such as uranium corrosion products to identify those that are significant in controlling its distribution.

The ternary investigation of cyanobacterial growth supernatant, Sr-90 and brucite showed no evidence of any impact on the behaviour of Sr-90 either to aid or suppress sorption. Further work is needed to assess a range of scenarios including experiments with samples of growth supernatant after cell death, and the effect of the cyanobacterial mass on Sr-90 removal with and without the presence of brucite.

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