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SPECIATION OF SOME ^{235}U FISSION PRODUCTS IN NITRATE SOLUTION AND THEIR SORPTION BEHAVIOR ON THERMALLY TREATED HYDROTALCITES

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ABSTRACT

^{235}U fission products and their daughter radioisotopes sorption behavior on calcined hydrotalcite were studied through batch and dynamic experiments. Electrophoresis showed that ^{99}Mo and $^{99\text{m}}\text{Tc}$ (the decay product of ^{99}Mo) were present in the solution mainly as anions. ^{103}Ru was found as neutral and anionic species and ^{131}I formed an insoluble compound. ^{132}Te was present only as a neutral species. It was found that under the utilized experimental conditions, ^{99}Mo presented the highest sorption value on calcined hydrotalcite, followed by $^{99\text{m}}\text{Tc}$, ^{131}I , ^{103}Ru , and ^{132}Te . Cations such as $^{140}\text{Ba}^{2+}$ and $^{140}\text{La}^{3+}$ were also retained by calcined hydrotalcite although in smaller proportions. It was also found that

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a very high concentration of NO_3^- anions in the solution decreased the sorption of the radioactive anions on calcined hydrotalcite.

INTRODUCTION

High activity radioisotopes can be produced by the ^{235}U fission process. Over 300 different nuclides of about 40 elements have been observed in spent nuclear fuels. The radioactive fission products have half-lives ranging from fractions of a second to a million years. Some of the most important ^{235}U fission products are the following: ^{95}Zr , ^{103}Ru , ^{106}Ru , ^{129}Te , ^{132}Te , ^{131}I , ^{140}Ba , ^{141}Ce , ^{144}Ce , ^{90}Sr , ^{99}Mo , ^{85}Kr , and ^{137}Cs . Some specific radioisotopes can be separated from the burned nuclear fuels as main or by-products, and utilized as radiotracers in a variety of chemical and medical studies. Several methods have been developed to recover radionuclides from spent nuclear fuels (1,2), or even from radioactive wastes by using different sorbent materials, such as organic resins and inorganic exchangers. Sameh and Leidfeld (3) have developed a process to separate radiocesium from acid intermediate level waste solutions by sorption on inorganic adsorbents. Mishra and coworkers (4) have performed an extensive work on inorganic ion exchangers in radioactive waste management and adsorption studies by using radiotracers.

More recently, considerable interest has been directed towards different materials such as hydrotalcites (HT) to sorb radionuclides present in aqueous solutions as anions. Serrano et al. (5) used calcined hydrotalcite (CHT) to sorb ^{99}Mo from aqueous solution and found that $^{99}\text{MoO}_4^{2-}$ ions were strongly retained in the hydrotalcite structure.

Until now, most of the researches on radioisotopes separation dealt with cation separation; however, papers on radioactive anion separation are less common.

Hydrotalcite (HT), which is an anion exchanger (6), and especially the calcined hydrotalcite (CHT), an Al and Mg mixed oxide (7), can be used to separate anions from slightly acidic solutions. In hydrotalcite, $\text{Mg}_6\text{Al}_2(\text{OH})_{16}\text{CO}_3\cdot 4\text{H}_2\text{O}$ (8), the Mg and Al hydroxides are assembled in a lamellar structure which bears an excess of positive electric charge because of a partial replacement of Mg^{2+} ions by Al^{3+} ions in the original structure (brucite mineral) (9–11). This positive charge is compensated by CO_3^{2-} anions, which are located in the interlayer space together with water molecules (12). Calcined hydrotalcite is the resulting compound of a dehydroxilation and decarbonation of HT when the latter is calcined at 500°C . Calcined hydrotalcite regenerates the layer structure of hydrotalcite when contacted with water containing inorganic and/or organic anions (10,13). In the regeneration process, the anions are incorporated into the layer space of the hydrotalcite.



Therefore, HTC can be utilized to eliminate anions from water solutions or recover them as valuable materials. The aim of this work is to study the speciation of some ²³⁵U fission products in nitrate solution and their sorption on CHT as a function of contact time and NO₃⁻ concentration in batch experiments and under dynamic conditions. ²³⁵U fission products were obtained by short irradiation time in a nuclear reactor.

EXPERIMENTAL

Sorbent and Solution Preparation

Hydrotalcite was prepared as described by Sato et al. (14): 1000 mL of 0.25 mol L⁻¹ AlCl₃ and 0.75 mol L⁻¹ MgCl₂·6H₂O aqueous solution were added dropwise to 1000 mL of 0.5 mol L⁻¹ Na₂CO₃ and 2.5 mol L⁻¹ NaOH under vigorous stirring. Both solutions were heated previously and then maintained at 60°C during the stirring. Once produced, HT was separated by filtration and then washed by dialysis with deionized water until no chloride was detected with the help of silver nitrate aqueous solution. The HT was dried at room temperature in an open dish for 5 days and then at 80°C in air for 3 hr, crushed in an Agatha mortar (Cole-Palmer, Vernor Hills, IL), and finally sieved to obtain HT grain size of 333 μm. The Al³⁺/(Al³⁺+Mg²⁺) molar ratio of the HT sample prepared as described was 0.25. The calcined product CHT was obtained by heating HT at 500°C in air for 18 hr. The obtained HT was characterized by x-ray diffraction measurements, TGA, and IR spectroscopy as specified elsewhere (5).

UO₂(NO₃)₂ was prepared by dissolving U₃O₈ (20% enriched in ²³⁵U) in hot concentrated HNO₃ in excess. The sample was evaporated to dryness and then redissolved in distilled water to obtain a 5 × 10⁻⁴ M uranium solution.

Fission products were produced as follows: 4.5 mL of the UO₂(NO₃)₂ solution was irradiated for 5 hr in a TRIGA MARK III nuclear reactor (General Atomic, La Joya, CA) with a neutron flux of 1.3 × 10¹² cm⁻² seg⁻¹. The concentration of fission products in the solution was about 10⁻¹² mmol/mL. Samples were allowed to decay for 5 days in the nuclear reactor pool. Afterwards, in the radiochemical laboratory, the irradiated sample was diluted with distilled water to 250 mL. In a previous work, it was observed that pH 5 was the optimum to sorb ⁹⁹MoO₄²⁻ on CHT (5), therefore, the pH of the solution in the present work was adjusted to 5 with NaOH. However, the pH increased rapidly along the sorption process, and at equilibrium, the pH value was found to be 9.5 after 9 hr. The increase of pH is due to the consumption of protons in the reconstruction of the layer HT structure (15). Measured concentration of NO₃⁻ ion in this solution was 0.002 M. This solution was utilized to study the sorption behavior of ²³⁵U



fission products on CHT. The studied fission products were ^{99}Mo , $^{99\text{m}}\text{Tc}$, ^{103}Ru , ^{131}I , ^{132}Te , ^{95}Zr , ^{140}Ba , ^{140}La , and ^{141}Ce which have half-lives between 2 and 65 days, and with enough activity to be determined by gamma spectrometry as described below.

Electrophoretic Analysis

Chemical species in the ^{235}U fission products solution were identified by paper electrophoresis and gamma spectrometry. A 1.0 cm wide and 30 cm long Whatman paper was used. The paper was impregnated with the supporting electrolyte and then the sample was applied on the paper. The electrophoregrams were obtained at a tension of 2000 V applied for 30 min. The supporting electrolyte was prepared as follows: 0.01 M HNO_3 solution was adjusted to pH 5 by adding drops of NaOH solution.

Sorption Experiment

Batch sorption experiments were carried out by shaking 10 mL of ^{235}U fission products solution and 0.050 g CHT for 72 hr. Kinetics experiments showed that this shaking time was enough to reach the equilibrium distribution of the above-mentioned ^{235}U fission products. When shaking was over, the solid and liquid phases were separated by centrifugation. A 5 mL liquid aliquot was taken to measure the gamma activity of the residual fission products in the solution by using a gamma spectrometer, which was set up with a Ge/hyperpure solid-state detector and a computerized multichannel analyzer. The radioactivity of the fission products was compared with a standard sample to calculate the residual concentration of the specific fission products, as well as the sorption percentage of the same fission products on CHT. In order to confirm that the utilized fission products solution was homogeneous, it was shaken without CHT in order to see whether precipitates were formed. As expected, no precipitate was found, as the concentration of fission products in the solution is extremely low, about 10^{-12} mmol/mL.

To carry out dynamic sorption experiments glass columns (0.5 cm diameter and 5 cm length) filled with CHT (1.7 g) were used. The ^{235}U fission products solution was passed through the column at a speed of 0.5 mL/min, and the effluent was fractionated at the bottom of the column. The 5 mL fractions taken from the effluent were analyzed by gamma spectrometry and the measurements were compared with a standard sample, as described earlier. Finally, the column was washed with distilled water.



RESULTS AND DISCUSSION

Speciation

^{99}Mo and $^{99\text{m}}\text{Tc}$ (not a fission product but a β^- decay product of ^{99}Mo) in the fission product solution described before were present mainly as the corresponding anions $^{99}\text{MoO}_4^{2-}$ and $^{99\text{m}}\text{TcO}_4^-$ (16) (Figs. 1 and 2). Electrophoresis technique and the computer program MEDUSA (17) showed that ^{99}Mo was present as a neutral species, very probably non-ionized $\text{H}_2^{99}\text{MoO}_4$, as well as $\text{H}^{99}\text{MoO}_4^-$. As a consequence of the β^- decay of ^{99}Mo in this neutral molecule, $^{99\text{m}}\text{Tc}$ was also detected on the application point of the sample. In addition to this statement, a neutral species of $^{99\text{m}}\text{Tc}$, such as $^{99\text{m}}\text{TcO}_2$ (18) may be present in the fission product solution. In this case, HTcO_4 cannot be the neutral species since the MEDUSA program indicated the formation of this non-dissociated acid only at pH values less than 3. ^{131}I was also detected on the application point of the sample, very probably as a non-dissociated iodate formed with cations such as Ce or Zr (19) (Fig. 3). Considering that the fission products solution contained HNO_3 , ^{103}Ru was probably present as a nitrosyl nitrate complex with low NO_3^- content; for instance, $^{103}\text{RuNONO}_3(\text{OH})_2(\text{H}_2\text{O})_2$ (19–21) as the neutral species, and $[\text{RuNO}(\text{OH})_4\text{H}_2\text{O}]^{1-}$ and $[\text{RuNO}(\text{OH})_5]^{2-}$ as anionic nitrosyl complexes (21)

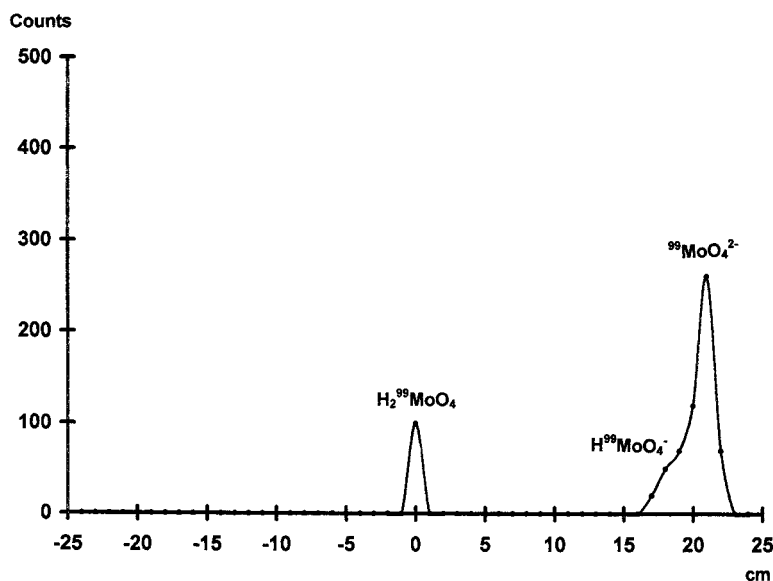


Figure 1. Electrophoregram of ^{99}Mo at pH 5.



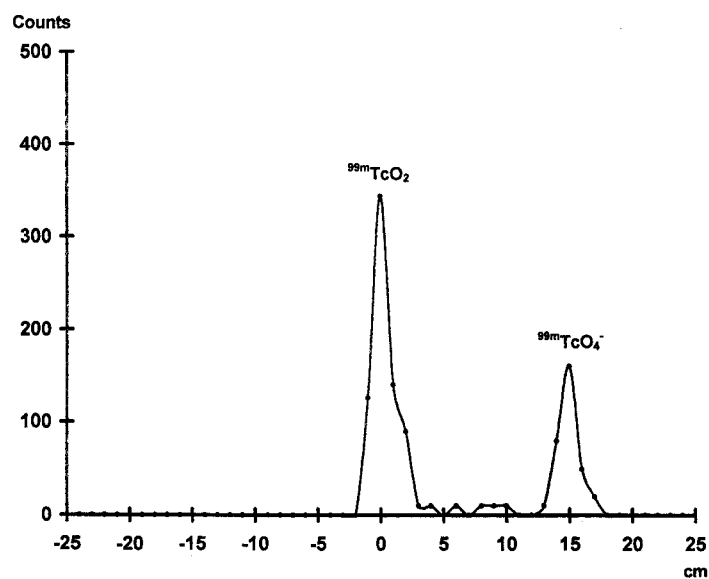


Figure 2. Electrophoregram of ^{99m}Tc at pH 5.

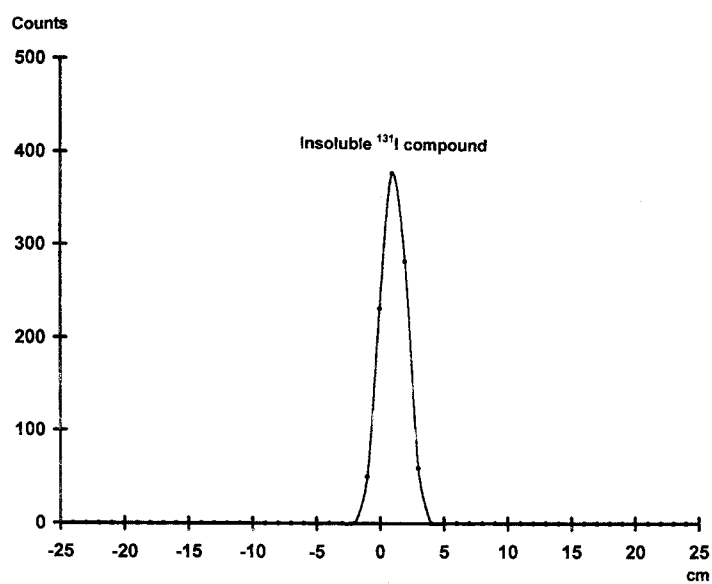


Figure 3. Electrophoregram of ^{131}I at pH 5.



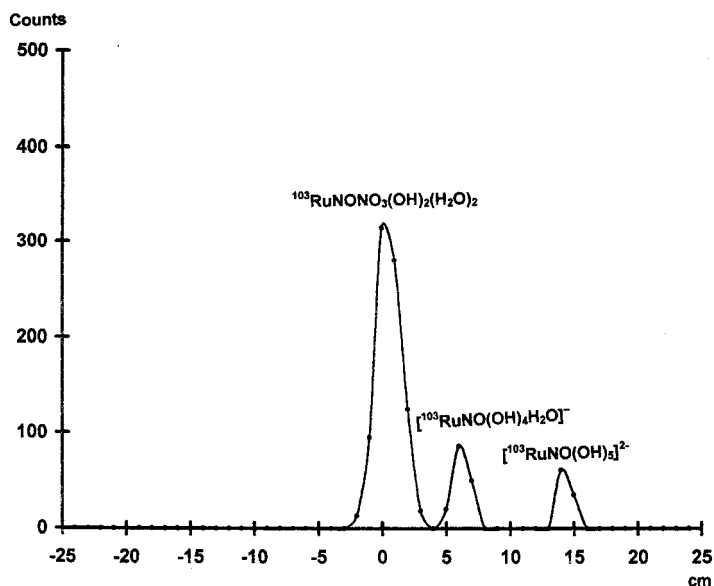


Figure 4. Electrophoregram of ¹⁰³Ru at pH 5.

(Fig. 4). The electrophoresis technique also showed that ¹³²Te was present only as a neutral species (Fig. 5) and MEDUSA indicated non-ionized telluric acid as the most probable species.

¹⁴¹Ce and ⁹⁵Zr were also detected on the electrophoresis paper. They remained on the application point of the sample, probably either as the insoluble iodates as explained before, or as neutral species. The MEDUSA program indicated that these neutral species might be CeO₂ and Zr(OH)₄, respectively. ¹⁴⁰Ba and ¹⁴⁰La (the β⁻ decay product of ¹⁴⁰Ba) were also detected as cationic species.

Batch Sorption

The percentage of equilibrium sorption of the ²³⁵U fission radioisotopes retained on the CHT in the batch experiments is shown in Table 1. As can be seen, the radionuclides with high sorption on CHT were ⁹⁹Mo and ^{99m}Tc, followed by ¹³¹I, ¹⁰³Ru, and ¹³²Te. Calcined hydrotalcite, as an adsorbent of anions, retained the anionic species of ⁹⁹Mo, ^{99m}Tc, and ¹⁰³Ru, incorporating them into the interlamellar space of the HT structure. In relation to neutral species present in



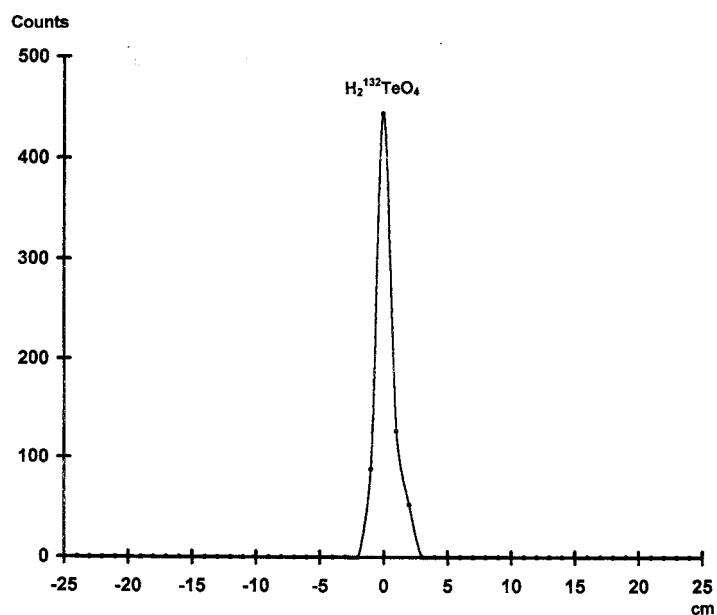


Figure 5. Electrophoregram of ^{132}Te at pH 5.

Table 1. Sorption from Nitrate Ion Solution (0.002 M) of Fission Products with Half-Lives >2 and <70 d on CHT. Batch Experiments

Radionuclide	Sorption (%)	Half-Life	Detected Species in Fission Products Solution
^{99}Mo	99.9	2.75 d	Anionic + neutral
$^{99\text{m}}\text{Tc}$	97.0	6.04 hr	Anionic + neutral
^{103}Ru	89.4	38.9 d	Anionic + neutral
^{131}I	95.3	8.08 d	Neutral
^{132}Te	87.8	3.21 d	Neutral
^{95}Zr	68.2	65.0 d	Neutral
^{140}Ba	76.4	12.8 d	Cationic
^{140}La	76.8	1.68 d	Cationic
^{141}Ce	71.8	32.5 d	Neutral



the fission product solution, they could be retained through an adsorption process on hydrotalcite grains surface.

On the other hand, unexpectedly some cationic fission products such as ¹⁴⁰Ba²⁺ and its β⁻ decay product ¹⁴⁰La³⁺, were also retained by CHT, although in lower proportion, probably by electrostatic attractions on the outer surface of hydrotalcite grains, as the surface charge is not homogeneous. Besides positive charge points, there are also -OH groups which can form surface complexes with Ba and La. Therefore, the immobilization of Ba and La is understandable.

Effect of NO₃⁻ Ions Concentration on ²³⁵U Fission Products Sorption

Under the solution conditions, during the fission products separation process, the NO₃⁻ ions would compete with the radioactive ions for the active sites of CHT. Thus, it was necessary to know the effect of NO₃⁻ anion concentration on the process. For this purpose, batch experiments were carried out as described above. An amount of CHT (0.05 g) was shaken with 10 mL of ²³⁵U fission products solution with various NO₃⁻ ion concentrations (0.25, 1.5, and 3.0 M) for 45 hr to reach the fission products equilibrium sorption. The corresponding solutions were obtained by using the calculated amounts of HNO₃ and then adjusting the pH to 5 with NaOH. Table 2 shows the sorption percentage of ²³⁵U fission products in the presence of NO₃⁻ anion. In general, radioactive

Table 2. Fission and Decay Products Sorption on CHT as a Function of NO₃⁻ Ion Molarity. CHT Mass: 0.05 g

Radioisotope	NO ₃ ⁻ Ions Molarity		
	Sorption (%)		
	0.25	1.5	3.0
⁹⁹ Mo	57.14	29.73	10.25
^{99m} Tc	61.88	24.52	9.72
¹⁰³ Ru	96.17	80.92	75.00
¹³¹ I	31.56	7.80	5.58
¹³² Te	100.00	94.30	100.00
⁹⁵ Zr	88.98	91.50	100.00
¹⁴⁰ Ba	10.33	10.54	8.23
¹⁴⁰ La	91.99	90.34	89.60
¹⁴¹ Ce	95.13	95.96	100.00



anions sorption decreases as the anion NO_3^- concentration in the solution increases from 0.25 to 3.0 *M*. Although Chatelet et al. (12) observed that the divalent anions are preferentially sorbed on CHT in the presence of monovalent anions, in our case the sorption of the divalent $^{99}\text{MoO}_4^{2-}$ anions on CHT was very much affected by the presence of NO_3^- ion in the solution because of its high concentration (approximately 10^8 times higher than that of the radioactive ions), since all anions compete for the active sites of hydrotalcite. $^{99\text{m}}\text{TcO}_4^-$ sorption on CHT was also affected, as expected, by the high concentration of NO_3^- ion. ^{132}Te sorption on CHT was not influenced by the high concentration of NO_3^- ion because it is probably adsorbed on the HT grains as a neutral molecule, as mentioned above. These results show that only anions occupy the active sites of the HT. The sorption of ^{131}I was also affected by the high molarity of NO_3^- ion since by using different concentrations of oxidative nitric salt, the concentration of redox iodine sensitive species may change, as well as their sorption on CHT. With respect to the fission products, which are present as cations, it can be seen that increasing NO_3^- ion concentration does not affect their sorption since they do not compete with the NO_3^- ion for the active sites of the hydrotalcite.

Dynamic Sorption

The results of the dynamic sorption of ^{235}U fission products in a 0.002 *M* NO_3^- solution onto CHT are shown in Table 3. ^{99}Mo has the highest dynamic sorption onto CHT, indicating that the low concentration of NO_3^- ions in this case, and the traces of radioactive anions do not interfere with its sorption

Table 3. Dynamic Sorption of ^{235}U Fission Products onto CHT ($^{99\text{m}}\text{Tc}$, Decay Product of ^{99}Mo ; and ^{140}La , Decay Product of ^{140}Ba Are Also Included)

Fission Products	Sorption (%)	Main Gamma Rays Energy (keV)
^{99}Mo	95.25	740
$^{99\text{m}}\text{Tc}$	93.45	141
^{103}Ru	61.32	497
^{131}I	67.07	364
^{132}Te	60.30	228
^{95}Zr	62.04	724
^{140}Ba	32.85	162
^{140}La	47.69	487
^{141}Ce	64.33	145



behavior (5). However, when the molarity of NO_3^- ion is raised to 3, the dynamic sorption of ^{99}Mo onto CHT falls down to about 11%. This very low dynamic sorption is due to the very high concentration of NO_3^- ion, which prevents the high sorption of ^{99}Mo onto CHT. Table 3 shows that ^{131}I , ^{103}Ru , ^{132}Te , ^{140}Ba , ^{140}La , ^{95}Zr , and ^{141}Ce are also sorbed onto CHT. The dynamic sorption values are lower than the ones observed in batch experiments.

CONCLUSIONS

The best-sorbed anionic fission product on CHT was ^{99}Mo . ^{103}Ru and ^{131}I were also retained in high percentage. $^{99\text{m}}\text{Tc}$, the β^- decay product of ^{99}Mo , also showed a high retention on CHT. Anionic fission products occupy the active sites of the interlayer space in hydrotalcites. ^{132}Te , ^{141}Ce , and ^{95}Zr were present in the fission product solution as neutral species and were retained probably by adsorption on hydrotalcite grains, as well as ^{131}I , which was present as a non-dissociated compound. ^{140}Ba and its β^- decay product ^{140}La , as cations in nitric acid solution, were unexpectedly retained by hydrotalcites, probably attracted electrostatically on the outer surface of hydrotalcite grains. Finally, a high concentration of NO_3^- ions in the fission product solution decreases the sorption of the radioactive divalent and monovalent anions on calcined hydrotalcites.

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