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OF ACTINIDE COLLOIDS

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SUMMARY

The formation and properties of particle fractions in actinide systems (americium(III), plutonium(IV), neptunium(V) and uranium(VI) have been studied in centrifugation (up to 27000 g), electromigration and diffusion experiments. The influence of physical and chemical parameters such as the storage time (6h - 6 months), temperature ($5^{\circ}C$ - $70^{\circ}C$), ionic strength of the aqueous phase (0.01 M, 0.1 M, 1 M), nuclide concentration (10^{-7} - 10^{-11} M) and pH (3 - 12) was studied.

Particle fractions (radii larger than 20 nm) were formed both for americium and plutonium in the pH-range of interest (7-9). These particle fractions did not exhibit any apparent enhanced mobility. Reduction of the total concentration in solution due to sorption on the vessel walls had maxima in this pH-range. The removal of both americium and plutonium increased with the storage time.

Essentially no particle fractions were obtained in the neptunium systems.

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1. INTRODUCTION

The interaction between radionuclides in solution and exposed geologic media, e.g. in connection with underground storage of radioactive waste, is largely determined by the chemical properties of the system. The sorption and mobility of most radionuclides in geologic systems would largely depend on the chemical state of the element (e.g. degree of hydrolysis, complexformation with organic and inorganic ligands, etc.). A formation of colloidal radionuclide particles would be feasible in the groundwater environment, particularly for the actinides in their lower oxidation states (III, IV). These colloids would be either true radiocolloids, i.e. aggregates of the radionuclide itself, or pseudocolloids, i.e. colloidal material already present in the groundwater, onto which the radionuclide has sorbed (1,2). These colloidal particles may be poorly sorbed on water exposed geologic media in comparison with radionuclides in true solution. The formation and properties of true actinide colloids (Am, Pu, Np, U) are discussed in this report.

2. EXPERIMENTAL

2.1 RADIONUCLIDES AND CHEMICALS

The radionuclides used in this study were ^{241}Am (433y), ^{237}Pu (45.4d) with ^{239}Pu (2.41×10^4y) as a carrier, ^{235}Np (396d) with ^{237}Np (2.14×10^6y) as a carrier and ^{233}U (1.59×10^5y). The short-lived isotopes ^{237}Pu and ^{235}Np were obtained from Harwell and all the others from Amersham Radiochemical Center, Great Britain. All radionuclide stock solutions, which had a pH of less than 1, were filtered through Nuclepore filter with a pore size of 400 nm. All salt solutions were prepared from p.a. $NaClO_4$, filtered through Nuclepore filter and centrifuged at about 27000 g for an hour, in order to remove all impurities from the solutions prior to the experiments. All solutions of acids and bases ($HClO_4$ and NaOH) used for adjustments of pH were treated in the same way.

Sodium perchlorate was used because the perchlorate ion forms very weak complexes with the actinides in solution; thus the complexing effect of the salt medium is negligible in comparison with e.g. hydrolysis in the pH-range of interest. All experiments were performed in aerated systems.

2.2 CENTRIFUGATION EXPERIMENTS

Centrifugation experiments were performed in order to investigate under what conditions a formation of a centrifugable colloidal particle fraction of the various actinides takes place. Solution of $NaClO_4$ of various ionic strengths were poured into 50 ml centrifuge tubes (20 ml per tube). Actinide stock solutions were added to desired total actinide concentrations, and the pH-values were adjusted with $HClO_4$ or NaOH to preselected values.

The solutions were centrifuged for half an hour at about 27000 g (Sorvall Centrifuge SS-3) and in some cases also at about 3300 g and 9200 g. Samples (0.5-1 ml) were taken both before and after the centrifugation and also after agitating the solutions. The samples were measured in a well-type scintillation counter. This separation and sampling procedure, as well as measurements of pH, were repeated after various storage time.

The following parameters were varied:

- pH-value of the solution (2-12)
- storage time (6h, 27h, 1w, 4w, 6w, 6 months)
- ionic strength of the aqueous phase ($NaClO_4$; 0.01 M, 0.1 M, 1.0 M)
- storage temperature (5^oC, 25^oC, 70^oC)
- initial nuclide concentration (10^{-7} M - 10^{-11} M)

All experiments were carried out in polypropylene tubes in order to avoid any releases of e.g. silica from the tube walls, which could serve as sources for pseudocolloids.

2.3 ELECTROMIGRATION EXPERIMENTS

Some electromigration experiments were performed, to determine the sign of the charge of the colloidal particles. Solutions from the centrifugation experiments were used. About 10 µl was put on a wetted filter paper and the migration in an electric field was measured. The apparatus is described elsewhere (3).

2.4 DIFFUSION EXPERIMENTS

Diffusion through a membrane can be used as a method for separating ions and molecules from colloidal particles in solution. The particles have a much slower diffusion rate than ions and molecules (4). Diffusion studies were carried out in order to get some information on the sizes of the colloidal aggregates.

Two 25 ml propylene vessels were put together and Nuclepore filters with pore sizes 15 or 400 nm were placed between them, Fig. 1. Nuclide solutions (0.01 M $NaClO_4$ of various pH) were stored in polypropylene vessels for about one week. In some of these solutions a formation of colloidal particles would be expected, according to the centrifugation experiments. After the storage, the active solution was poured into one of the two compartments of the diffusion vessel. A solution of the same composition, but without the radionuclide, was poured into the other compartment. The solutions were stirred with magnetic stirrers, and samples were taken from both compartments after various times.

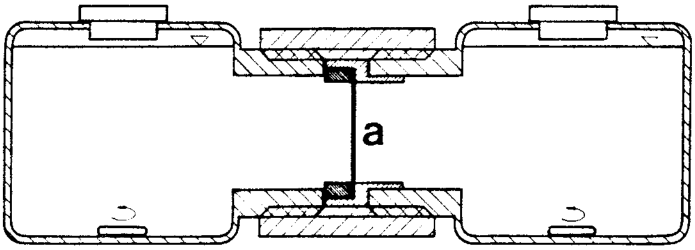


Figure 1. Cell for diffusion measurements. a. Membrane.

2.5 COLUMN EXPERIMENTS

Column experiments were performed to get information on the mobility of colloids in the presence of a solid sorbent.

A small column with a volume of 0.46 cm^3 was packed with crystalline α-Al_2O_3, which was crushed and sieved to a desired grain size (0.090-0.125 mm). Small portions (0.05-0.1 ml) were taken from solutions, where a formation of a colloidal fraction was indicated in the centrifugation experiments, and injected into the column and eluated with inactive solution of the same composition.

3. RESULTS AND DISCUSSION

3.1 CENTRIFUGATION EXPERIMENTS

Since colloid formation of e.g. actinides is strongly related to hydrolysis and hydroxide precipitation in most cases (5), the pH-value of the solution was selected as an independent chemical parameter in all the experiments. The removal of the radionuclide from the solution was studied as a function of the storage time, the ionic strength of the solution, the temperature and the nuclide concentration. (6,7)

3.1.1 Storage time

For all the studied nuclides the removal from the solution by centrifugation (including sorption on the tube walls) seems to increase with increasing storage time as illustrated in Fig. 2-5. For americium, which is trivalent, more than 50% was removed from the solution in the pH-range 6-8 and more than 30% above pH 12 already after 6h, Fig. 2. It also seems as the difference between these two maxima (at pH 6-8 and at pH 12) decreases with increasing storage time. After 6 months more than 80% was removed in the pH-range 6-12.

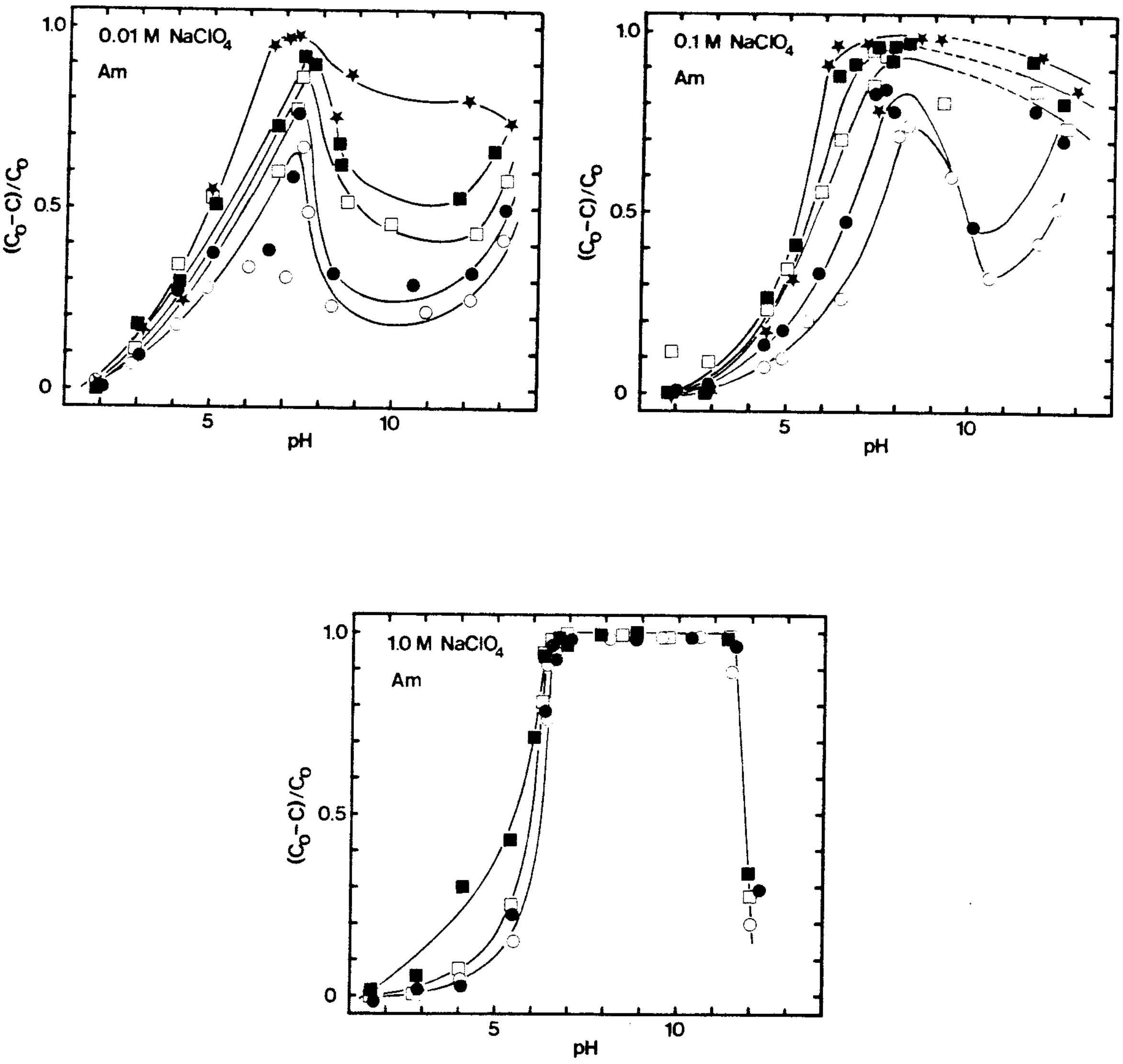


Figure 2. The removal of americium by centrifugation (27000 g) after various storage times and at different ionic strengths (1.0 M, 0.1 M, 0.01 M).
o 6h, ● 27h, □1w, ■6w, ✶6m. Initial nuclide concentration: 2.3×10^{-9} M.

For plutonium, which is predominantly tetravalent, possibly with some contributions of pentavalent species, more than 60% was removed after 6h in the pH-range 6-11, Fig. 3. After 4 weeks more than 80% was removed even at the low ionic strength.

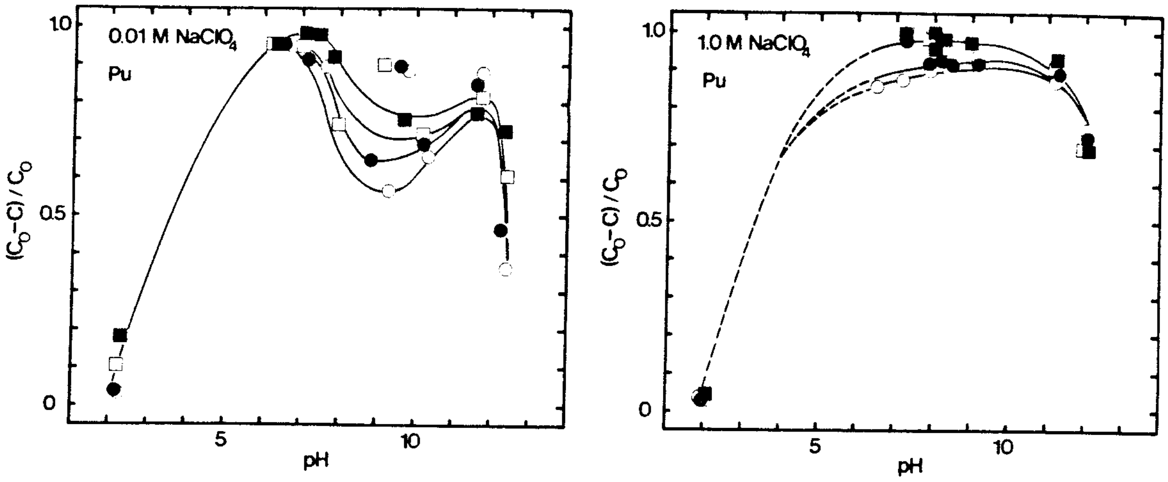


Figure 3. The removal of plutonium by centrifugation (27000g) after various storage times and at different ionic strength (1.0 M, 0.01 M).
o 6h, ● 27h, □1w, ■4w. Initial nuclide concentration: 6.0×10^{-10} M.

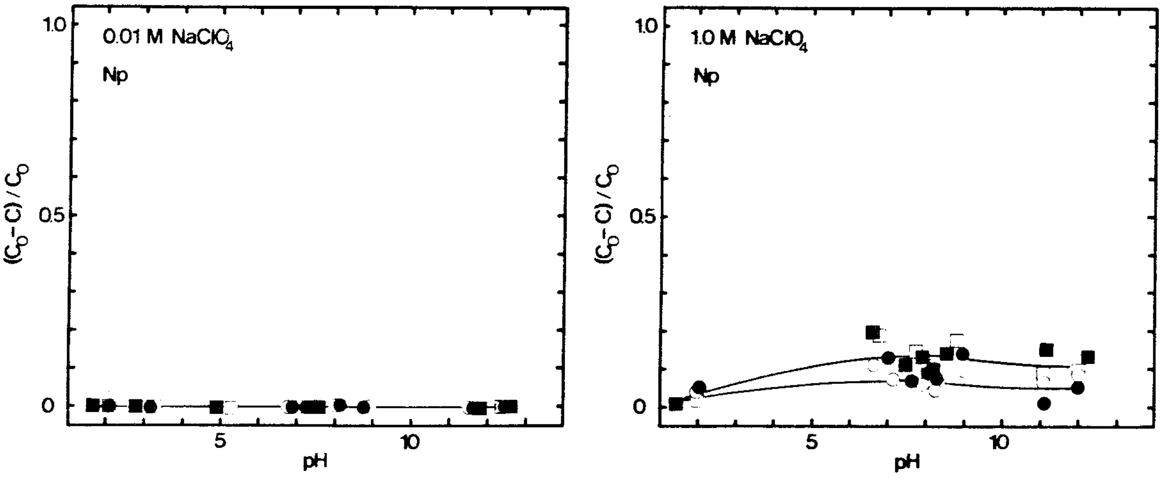


Figure 4. The removal of neptunium by centrifugation (27000g) after various storage times and at different ionic strength (1.0 M, 0.01 M).
o 6h, ● 27h, □1w, ■4w. Initial nuclide concentration: 2.1×10^{-9} M.

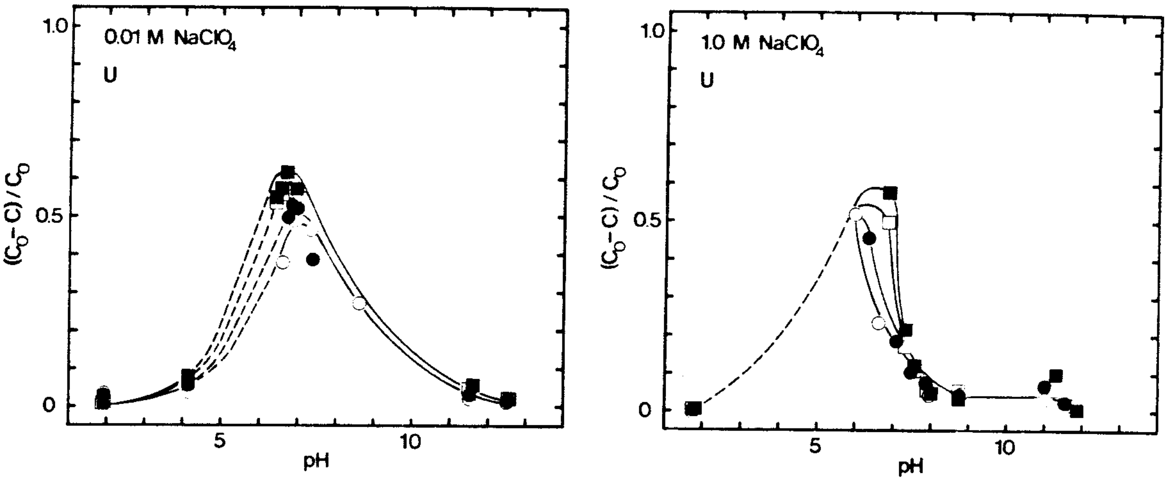


Figure 5. The removal of uranium by centrifugation (27000g) after various storage times and at different ionic strength (1.0 M, 0.01 M).
○ 6h, ● 27h, □1w, ■6w. Initial nuclide concentration: 2.1×10^{-7} M.

Very little of the neptunium, which is pentavalent, was removed from the solution even after a storage time of 4 weeks. Only at the high ionic strength there was a small removal, increasing with the storage time, Fig. 4, just as for americium and plutonium.

About 50% of the uranium, which would be hexavalent, was removed at pH 6-7 after 6 hours. The removed fraction increased to about 60% after 6 weeks, Fig. 5.

3.1.2 Ionic strength

The removal of americium from the solution starts already at pH about 3-4 at all ionic strengths, Fig. 6. At the highest ionic strength americium is almost quantitatively removed at pH between 6 and 11. At pH above 11 there may be a decreasing degree of removal. The rate of removal increases with increasing ionic strength.

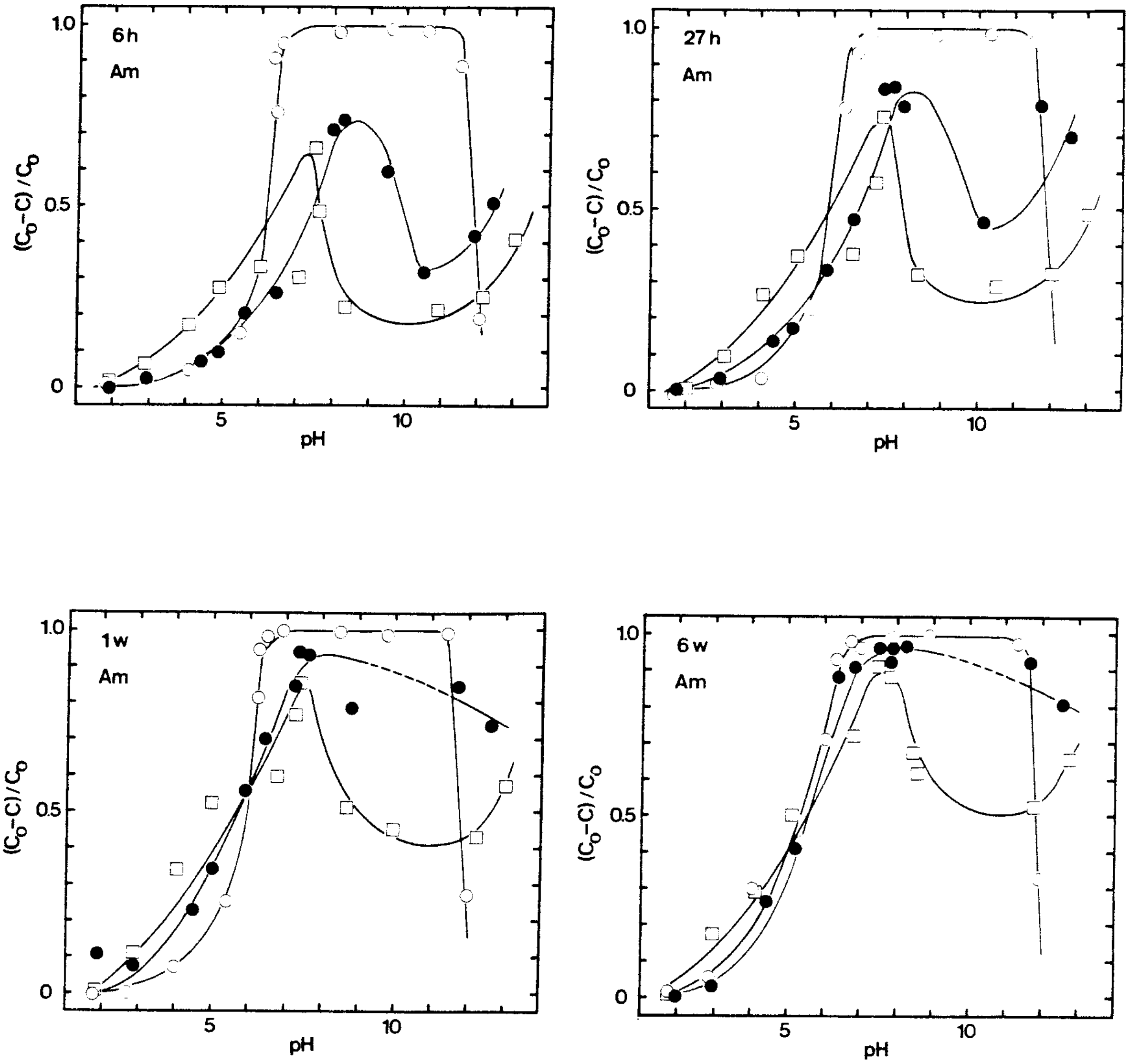


Figure 6. The removal of americium by centrifugation (27000 g) at various ionic strength and storage times (6h, 27h, 1w, 6w).
o 1.0 M $NaClO_4$, ● 0.1 M $NaClO_4$, □ 0.01 M $NaClO_4$;
Initial nuclide concentration: 2.3×10^{-9} M.

The ionic strength seems to have similar influence on the removal of plutonium, as on the removal of americium, Fig. 7, but starts at lower pH-values. The ionic strength dependence is similar.

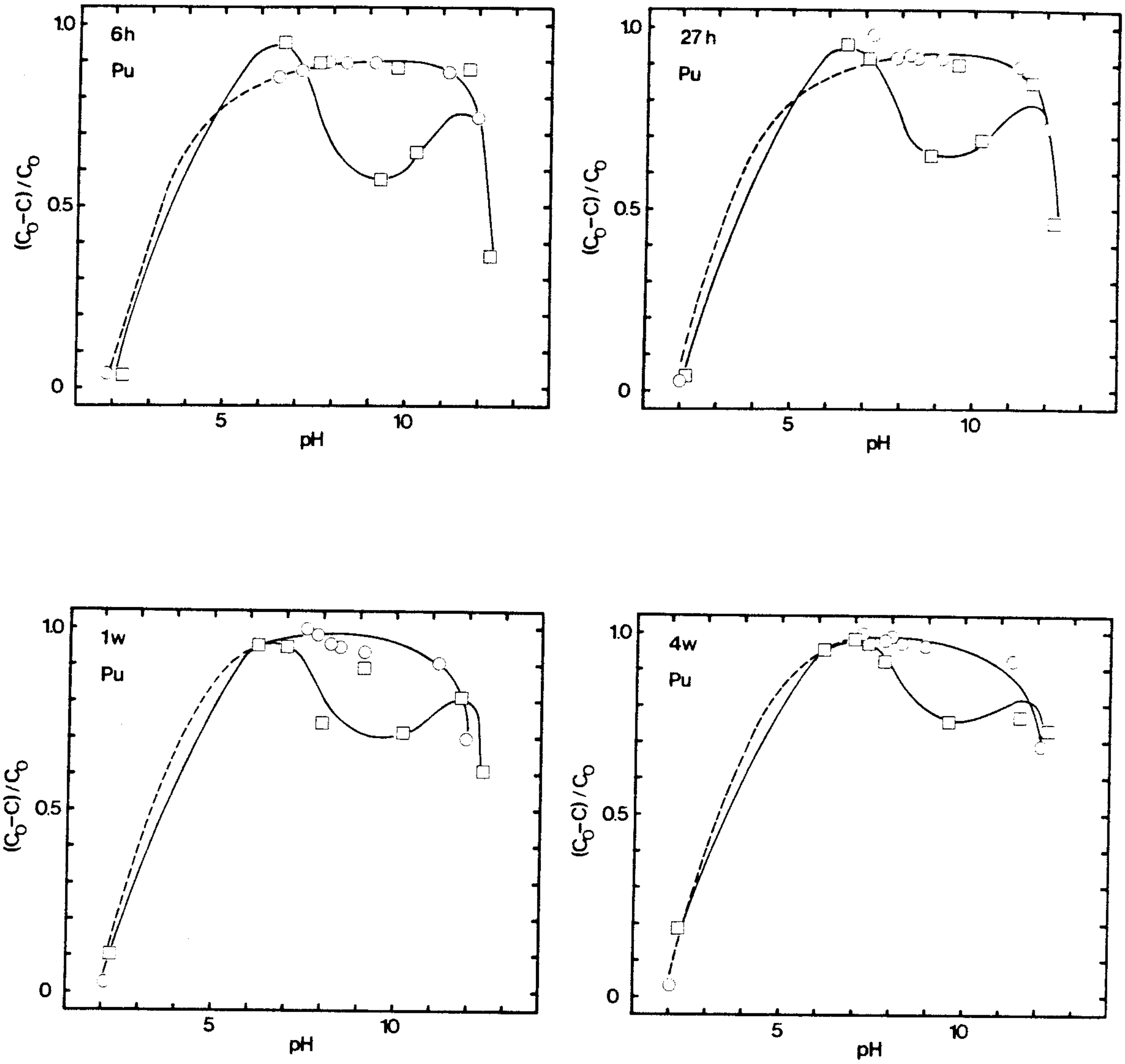


Figure 7. The removal of plutonium by centrifugation (27000 g) at various ionic strength and storage times (6h, 27h, 1w, 4w).
o 1.0 M $NaClO_4$, □ 0.01 M $NaClO_4$; Initial nuclide concentration: 6.0×10^{-10} M.

For neptunium there is no significant removal at low ionic strength, Fig. 8, but at the higher ionic strength about 10% of the neptunium seems to be removed from the solution at pH above 6.

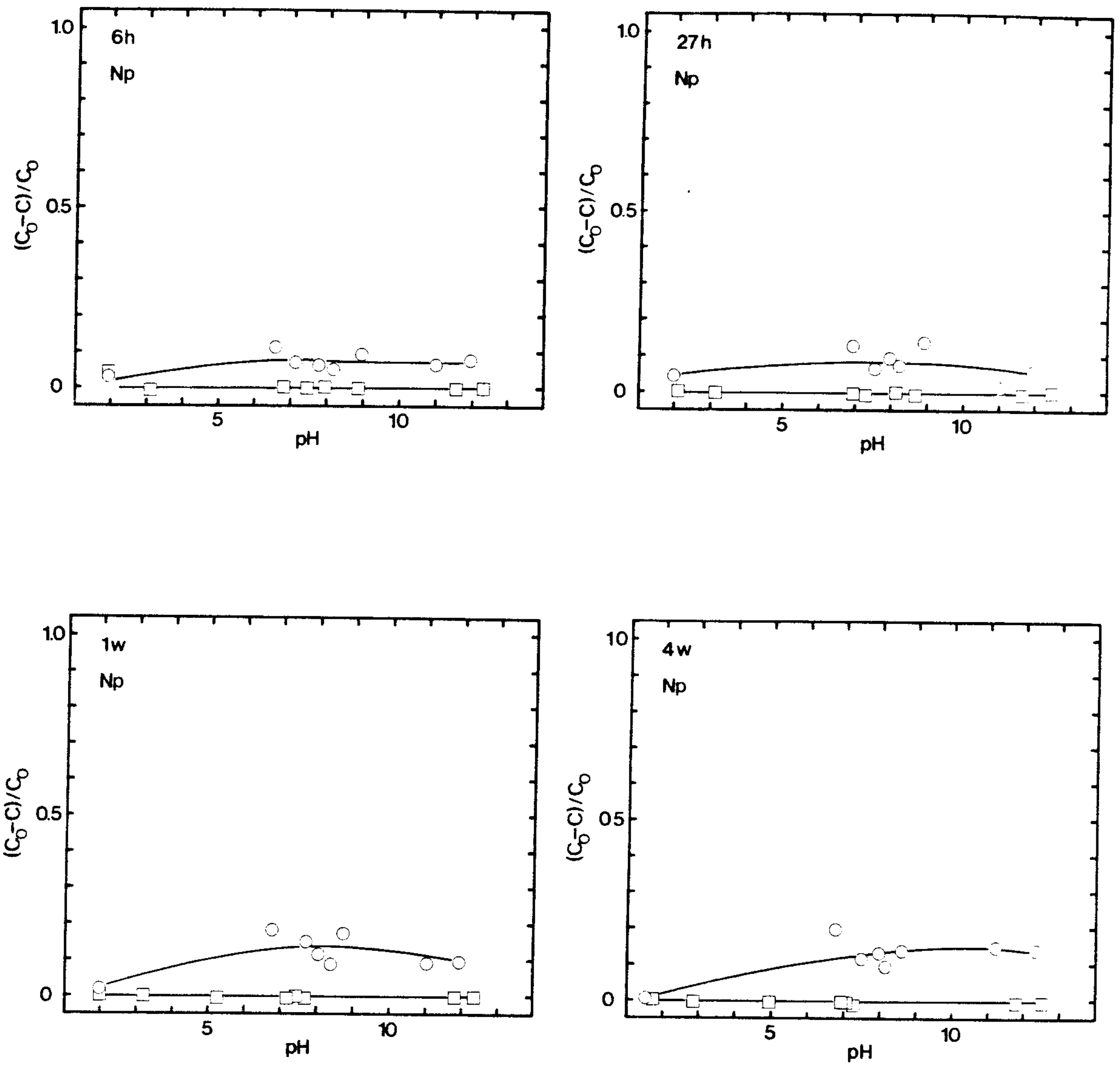


Figure 8. The removal of neptunium by centrifugation (27000 g) at various ionic strength and storage times (6h, 27h, 1w, 4w).
o 1.0 M $NaClO_4$, □ 0.01 M $NaClO_4$; Initial nuclide concentration: 2.1×10^{-9} M.

For uranium, the removal starts at pH about 4, Fig. 9, and reaches a maximum at pH 6-7 and then decreases again with increasing pH. Possibly the removal vs pH is shifted towards a lower pH at the higher ionic strength.

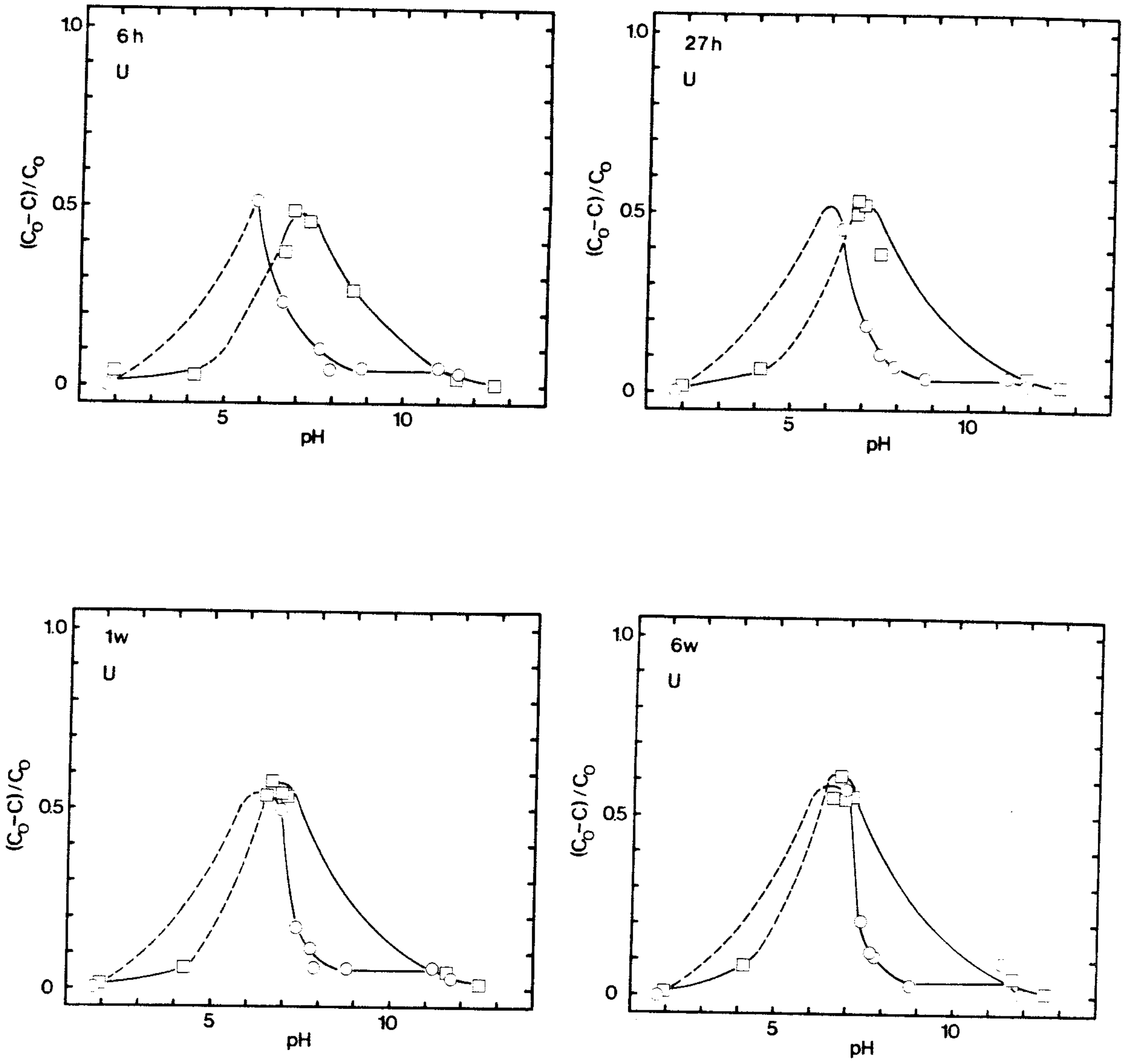


Figure 9. The removal of uranium by centrifugation (27000 g) at various ionic strength and storage times (6h, 27h, 1w, 6w).
o 1.0 M $NaClO_4$, □ 0.01 M $NaClO_4$; Initial nuclide concentration: 2.1×10^{-7} M.

3.1.3 Temperature

Separation of americium by centrifugation was studied at three temperatures: 5°C, 25°C and 70°C. Fig. 10 indicates that the removal of americium possibly increases slightly with increasing temperature after long storage time.

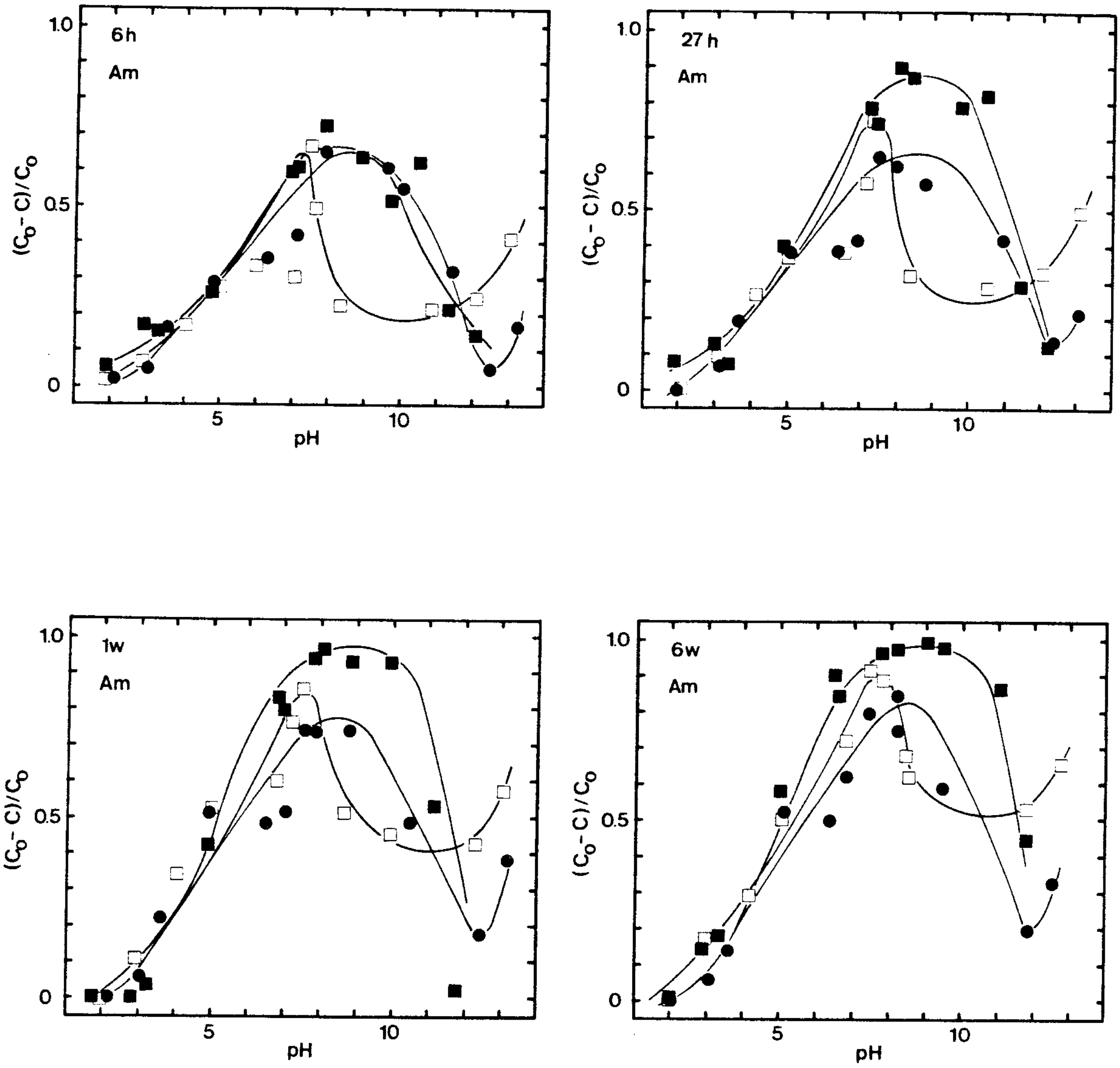


Figure 10. The removal of americium at various temperatures and storage times (6h, 27h, 1w, 6w; ionic strength 0.01 M). ● 5°C, □ 25°C, ■ 70°C. Initial nuclide concentration: 2.3x10^{-9} M.

3.1.4 Nuclide concentration

The removal of americium started at pH about 3 and reached a maximum at pH about 7-8 and had another maximum at pH above 11-12, fig. 11.

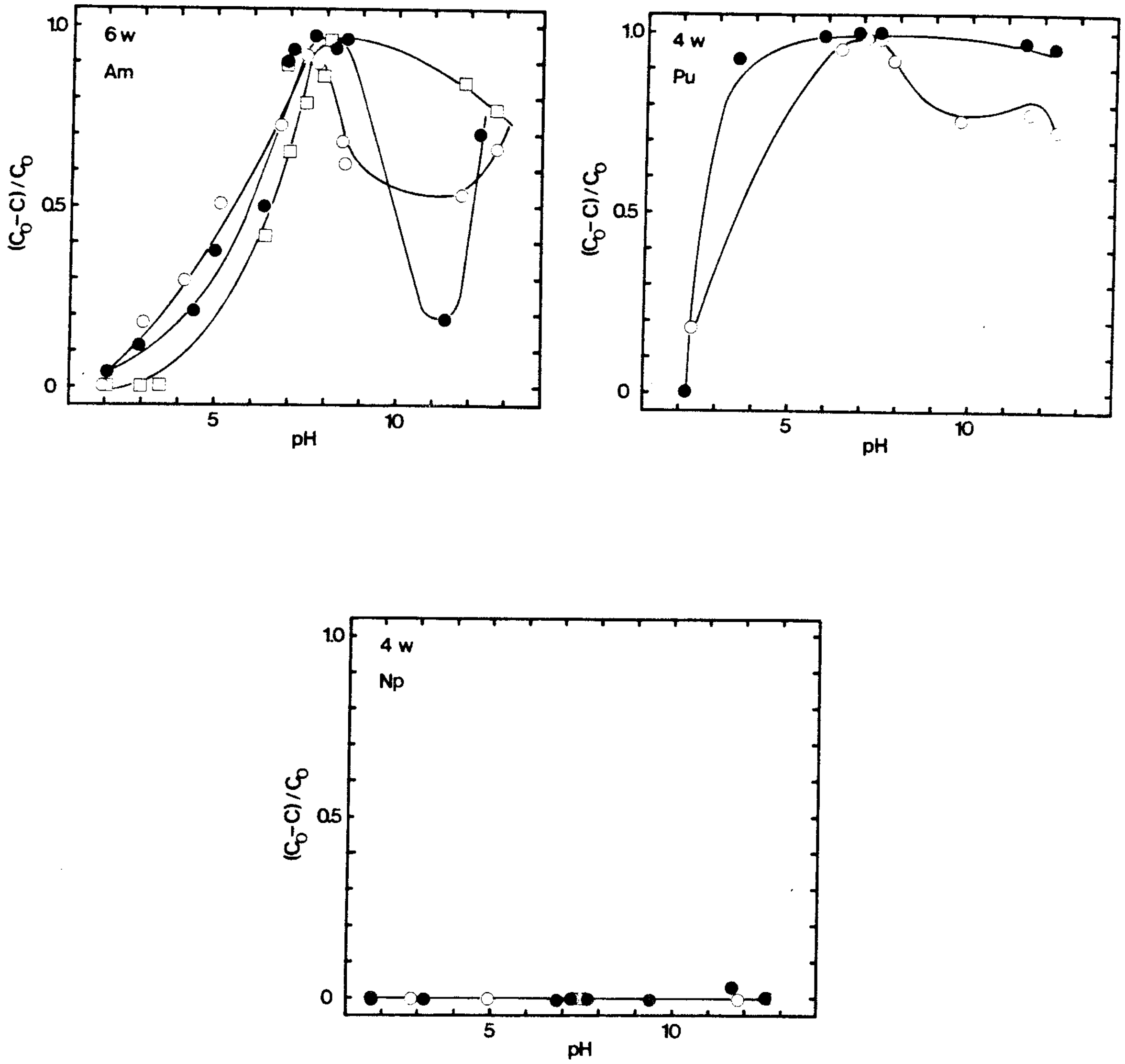


Figure 11. The removal of americium, plutonium and neptunium by centrifugation (27000 g) at various initial nuclide concentrations (ionic strength 0.01 M, storage time 4-6w).

● $C_0=2.9\times10^{-7}$ M (Am), 6.0×10^{-8} M (Pu), 2.1×10^{-7} M (Np)

○ $C_0=2.3\times10^{-9}$ M (Am), 6.0×10^{-10} M (Pu), 2.1×10^{-9} M (Np)

□ $C_0=1.8\times10^{-11}$ M (Am).

For plutonium the removal started at somewhat lower pH-value and reached a maximum at pH 5-6.

For neptunium there was very little removal for both studied nuclide concentrations.

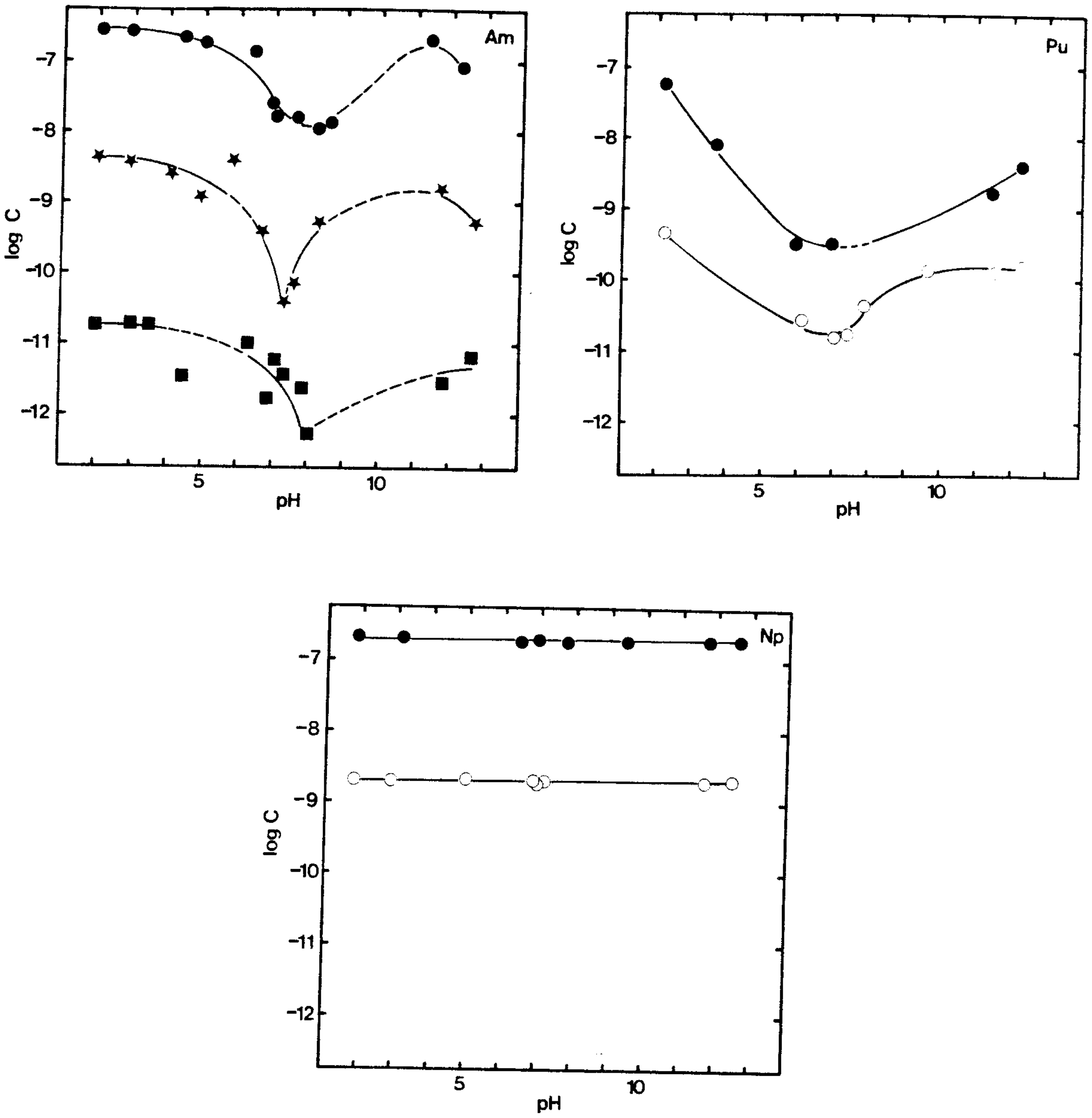


Figure 12. The concentrations of americium, plutonium and neptunium in solution at various initial concentrations.

● $C_0=2.9\times10^{-7}$ M (Am), 6.0×10^{-8} M (Pu), 2.1×10^{-7} M (Np)

✶ $C_0=2.3\times10^{-9}$ M (Am), ○6.0×10^{-10} M (Pu), 2.1×10^{-9} M (Np)

□ $C_0=1.8\times10^{-11}$ M (Am).

Fig. 12 shows that the concentration of americium, plutonium and neptunium in solutions did not decrease by more than two orders of magnitude. The removed fractions were about the same for all nuclide concentrations and evidently largely concentration inde-

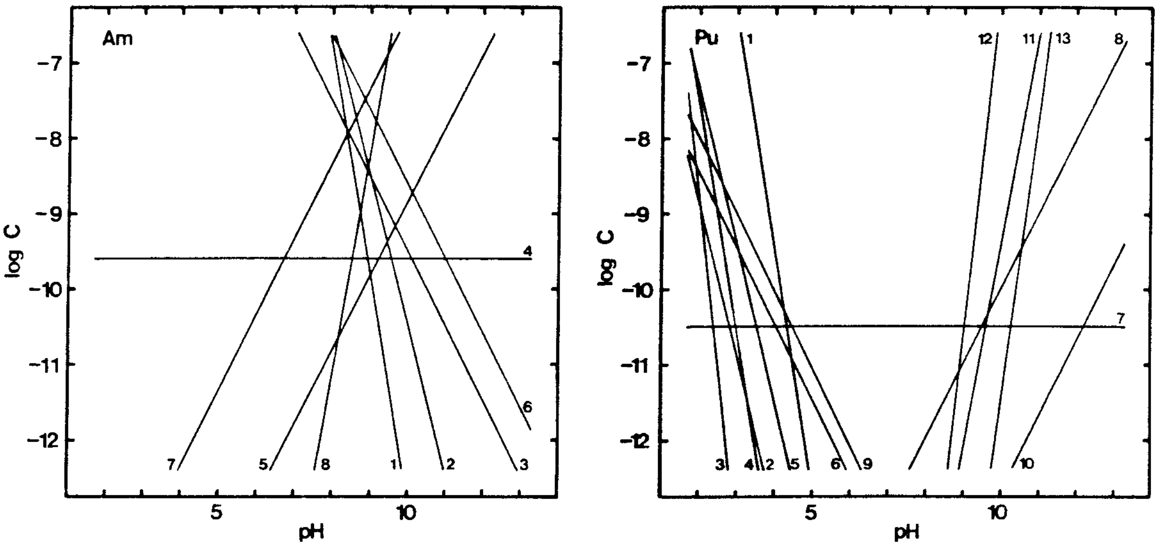


Figure 13. Speciation in saturated americium and plutonium solutions.

(Open system, log $[CO_3^{2-}]$ = 2pH-21.6 (8); Eh = 0.8-0.06 pH; formation constants from ref. 9; Solid solubility limiting phases: $Am_2(CO_3)_3(s)$ and $PuO_2(s)$, respectively.)

Americium: 1 - Am^{3+}, 2 - $AmOH^{2+}$, 3 - $Am(OH)_2^+$, 4 - $Am(OH)_3$, 5 - $Am(OH_4^-$, 6 - $AmCO_3^+$, 7 - $Am(CO_3)_2^-$, 8 - $Am(CO_3)_3^{3-}$.

Plutonium: 1 - Pu^{3+}, 2 - $PuOH^{2+}$, 3 - Pu^{4+}, 4 - $PuOH^{3+}$, 5 - $Pu(OH)_2^{2+}$, 6 - $Pu(OH)_3^+$, 7 - $Pu(OH)_4$, 8 - $Pu(OH)_5^-$, 9 - PuO_2^+, 10 - $PuO_2CO_3^-$, 11 - $PuO_2(CO_3)_2^{3-}$, 12 - $PuO_2(CO_3)_3^{5-}$, 13 - $PuO_2(CO_3)_3^{4-}$.

pendent. However, both for americium and plutonium the pH of maximum removal was possibly shifted slightly towards lower pH with increasing nuclide concentration.

The removal of a significant fraction of americium cannot be due to precipitation of hydroxides or carbonates, when the initial concentrations are as low as 10^{-8} - 10^{-11} M. (8) For plutonium, however, the curve representing log C vs pH in Fig. 12 may in fact correspond to a solubility curve, assuming a solubility product of 10^{-55} for the solubility limiting hydroxide $Pu(OH)_4(s)$ (c.f. Fig. 13).

3.1.5 Centrifugable fraction

The removal of the radionuclide from the solution can be caused either by sorption on the tube walls or by formation of small particles, which can be centrifuged out of the solution. To determine the particle fraction, samples were taken both after centrifuging at 27000 g for 30 min and after agitating the solutions. The difference between these samples is here called the centrifugable fraction.

The radius of centrifugable particles can be determined by the following equation (1)

$$r^2 = \frac{9\eta \ln(x_2/x_1)}{2(\rho - \rho_0)w^2 t}$$

where η and ρ_0 are the viscosity and density of the dispersing medium, respectively, w is the angular velocity, ρ is the particle density, x_1 and x_2 the initial and final distance of the particle from the axis of rotation and t is the centrifugation time. An angular velocity of 17000 rpm (corresponding to 27000 g) and a centrifugation time of 30 min in the present experiments, allowed a separation of particles with radii greater than about 20 nm. This means that the centrifuged samples would represent soluble nuclide species and particles with radii below 20 nm, while the agitated samples also would contain larger particles. Thus, the centrifugable fraction contains particles with radii above 20 nm.

Other angular velocities (5900 rpm and 10000 rpm, corresponding to about 3300 g and 9200 g respectively) were used, in order to obtain a fractionation of particles of different sizes. However, there were usually no significant differences between samples, centrifuged at different speeds. This can possibly be due to the very low nuclide concentrations.

Fig. 14-17 indicate that most of the removal is due to sorption on the vessel walls for all the studied nuclides.

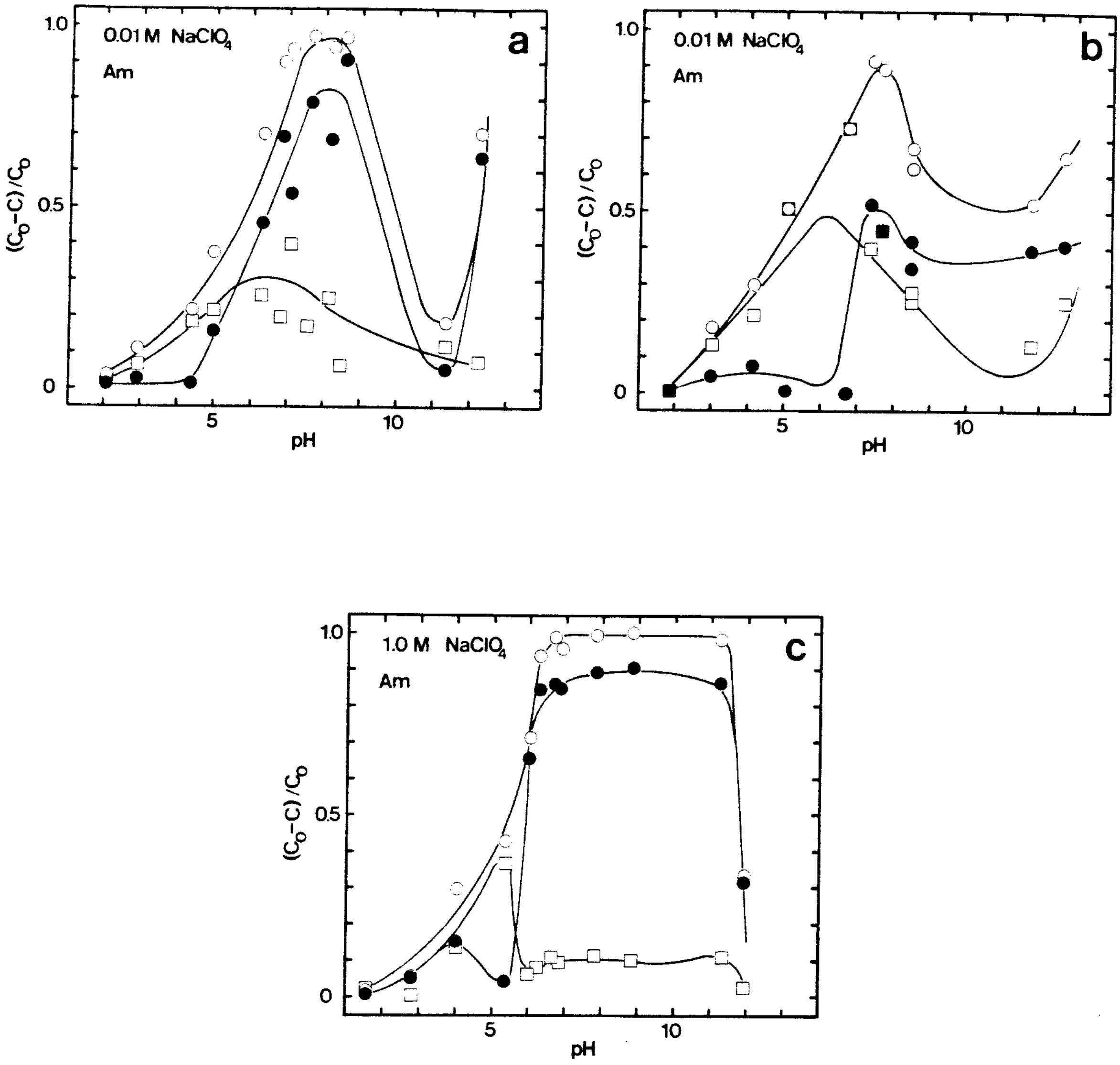


Figure 14. Centrifugable americium fraction at 27000 g. (Ionic strength 0.01 and 1.0, Storage time 6w, Am-concentration 2.9×10^{-7} M, a, and 2.3×10^{-9} M, b and c). o after centrifugation at 27000 g, ● after agitation, □ centrifugable fraction.

For americium, Fig. 14, the centrifugable fraction had maxima at pH about 5-6 and possibly at pH above 12. However, for the highest ionic strength there was no maximum above pH 12. The centrifugable fraction at pH around 5-6 was almost independent of the ionic strength. Thus, this particle fraction is probably not

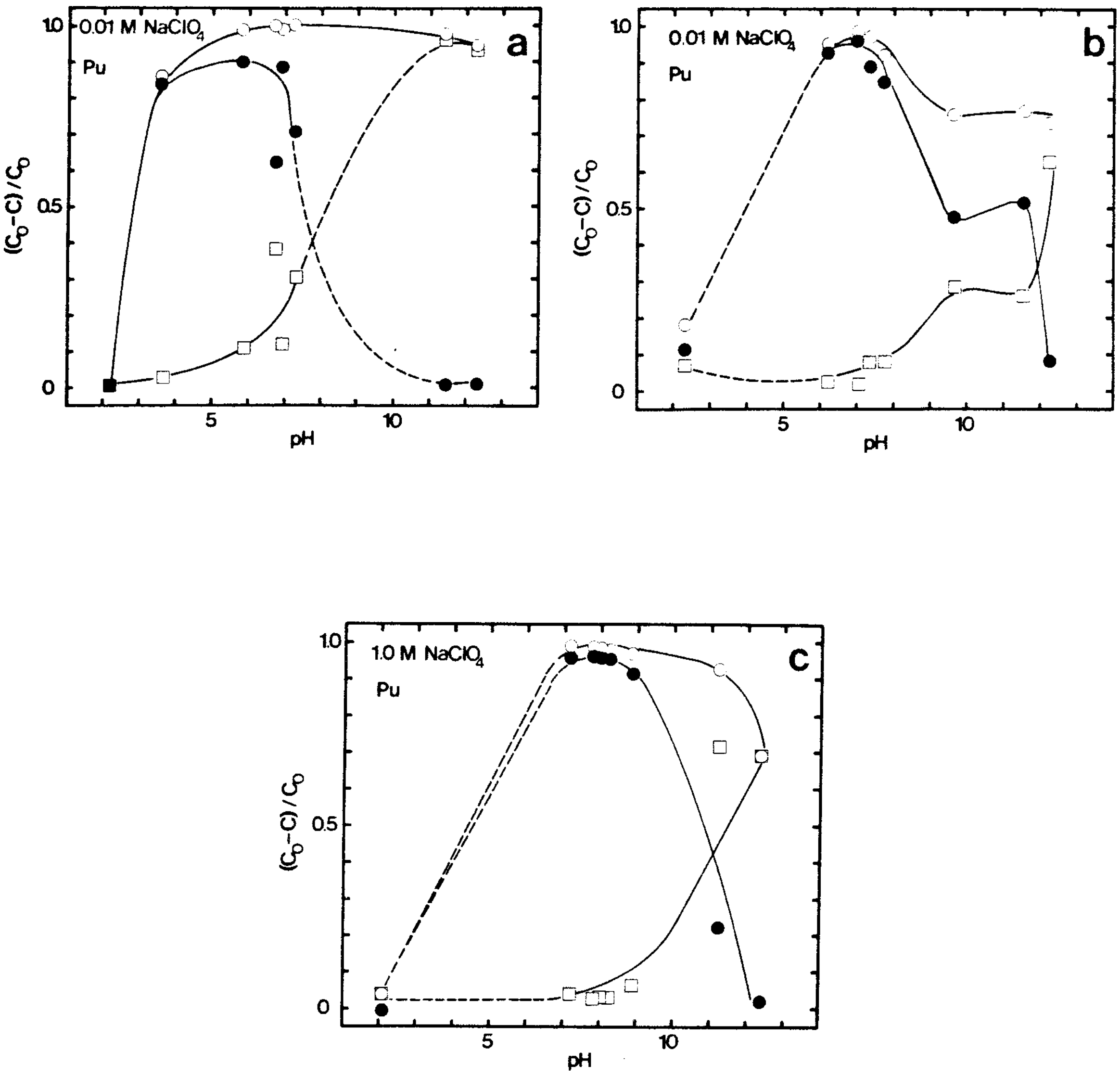


Figure 15. Centrifugable plutonium fraction at 27000 g. (Ionic strength 0.01 M and 1.0 M, Storage time 4w, Pu-concentration 6.0×10^{-8} M, a, and 6.0×10^{-10} M, b and c). o after centrifugation at 27000 g, ● after agitation, □ centrifugable fraction.

caused by impurities in the salt. However, impurities in the acidic ^{241}Am-solutions (e.g. iron, which could exist in true solutions at low pH, but precipitate as $Fe(OH)_3$ in the sample solution) cannot be excluded.

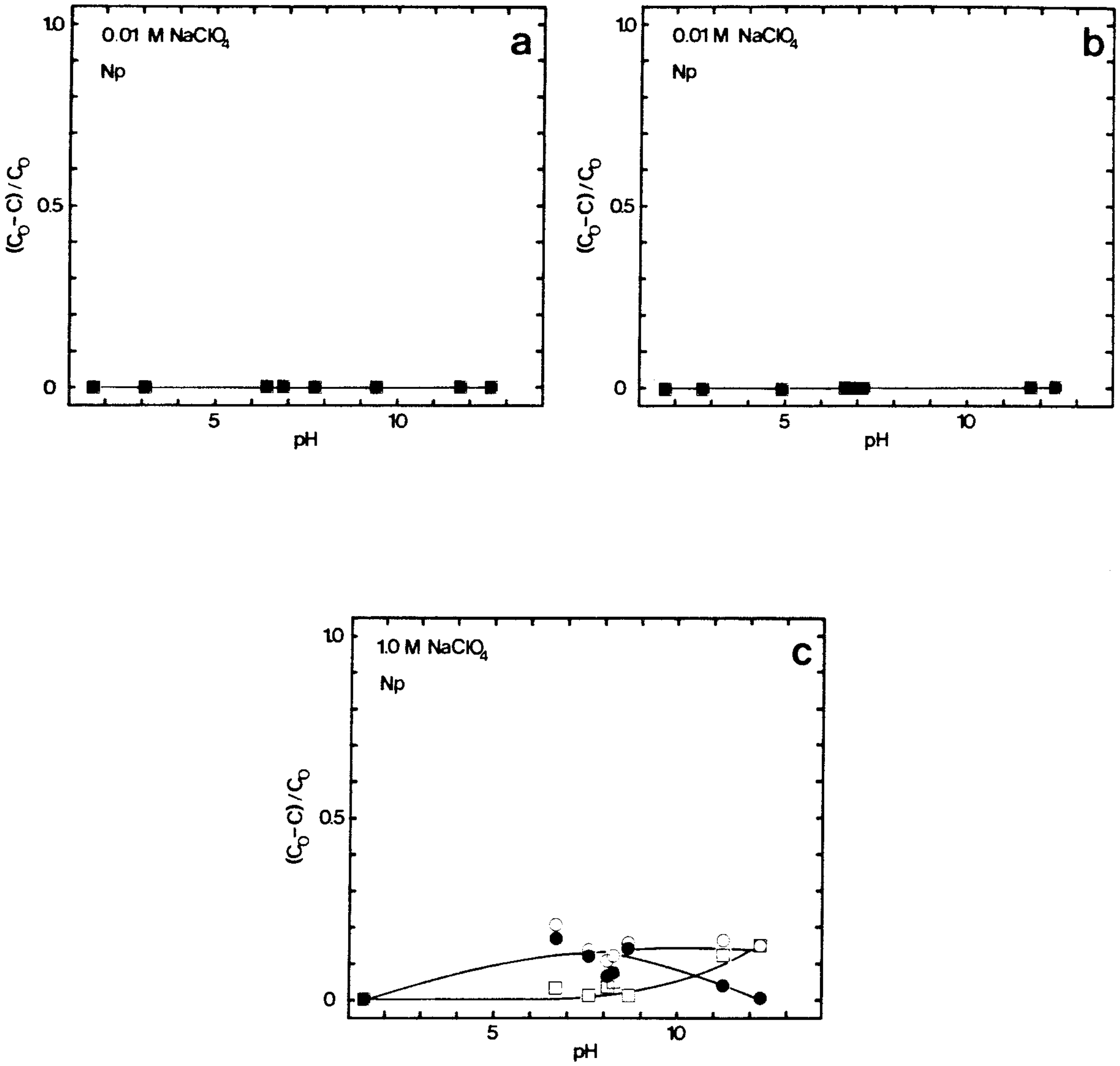


Figure 16. Centrifugable neptunium fraction at 27000 g. (Ionic strength 0.01 M and 1.0 M, Storage time 4w, Np-concentration $2.1\text{x}10^{-7}$ M, a, and $2.1\text{x}10^{-9}$ M, b and c). o after centrifugation at 27000 g, ● after agitation, □ centrifugable fraction.

For plutonium, Fig. 15, there is no centrifugable fraction below pH 8. At higher pH-values, however, the centrifugable fraction increases with increasing pH, and at pH about 12, most of the removed plutonium seems to be in particulate form, which can be centrifuged out of the solution.

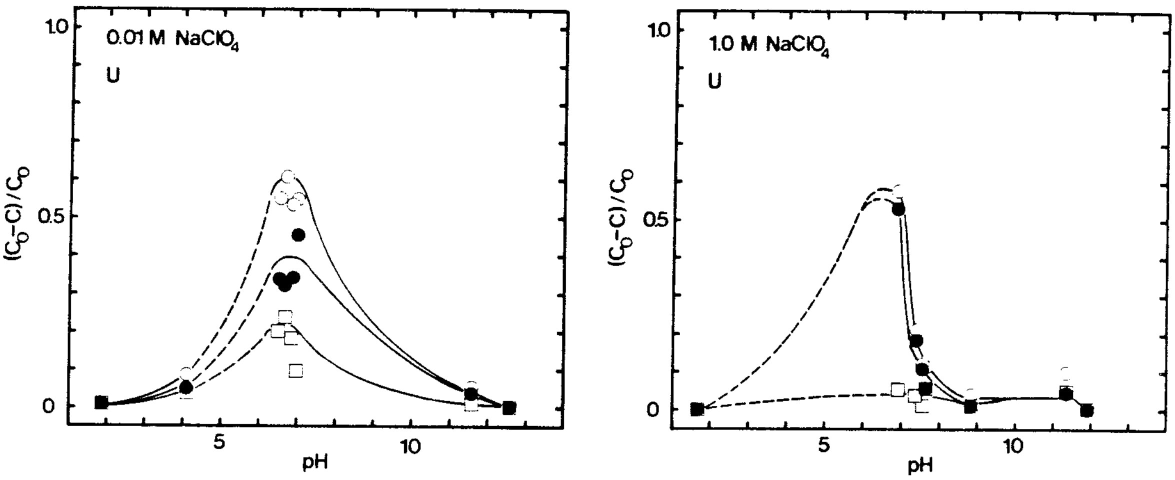


Figure 17. Centrifugable uranium fraction at 27000 g. (Ionic strength 0.01 M and 1.0 M, Storage time 6w, U-concentration 2.1×10^{-7} M).
o after centrifugation at 27000 g, ● after agitation, □ centrifugable fraction.

For neptunium, Fig. 16, there are no indications of a particle fraction at low ionic strength. At higher ionic strength and at pH above 10, however, there may be a small centrifugable particle fraction.

For uranium, Fig. 17, there is essentially no centrifugable fraction at high ionic strength, but at low ionic strength there can possibly be a small particle fraction at pH about 6-7.

In general the centrifugable fractions at high pH may very likely represent true colloids of hydroxides.

3.2 ELECTROMIGRATION EXPERIMENTS

Results from the electromigration studies are given in Table 1.

Table 1. Direction of migration of americium (in 0.01 M $NaClO_4$, 25°C, initial concentration 2.3×10^{-9} M)

pH	Migration of ^{241}Am (%)	
	to the cathode	to the anode
2.0	100 fast	0
3.0	96 fast	4
4.7		
6.0	very slow to both anode and cathode	
7.8		
10.0	50	50
11.0	30	70 fast
12.0	40	60 fast
13.0	30	70 fast

At low pH the americium species are positively charged and migrate to the cathode. At about pH 5-8 there is a very slow transfer to both the cathode and anode coinciding with the first maximum of the centrifugable fraction. At pH above 12, where the second maximum of the centrifugable fraction is obtained, the americium species migrate relatively fast to the anode. These observations are similar to earlier estimations. (5)

Experiments were also done for plutonium and neptunium. The results from these systems were very scattered, due to the low nuclide concentrations, and not conclusive.

3.3 DIFFUSION EXPERIMENTS

For both americium and plutonium the diffusion rates through the membrane was rather fast at low pH, Fig. 18. Above a certain pH-level the rates were much slower which indicates a formation of particles or increased sorption.

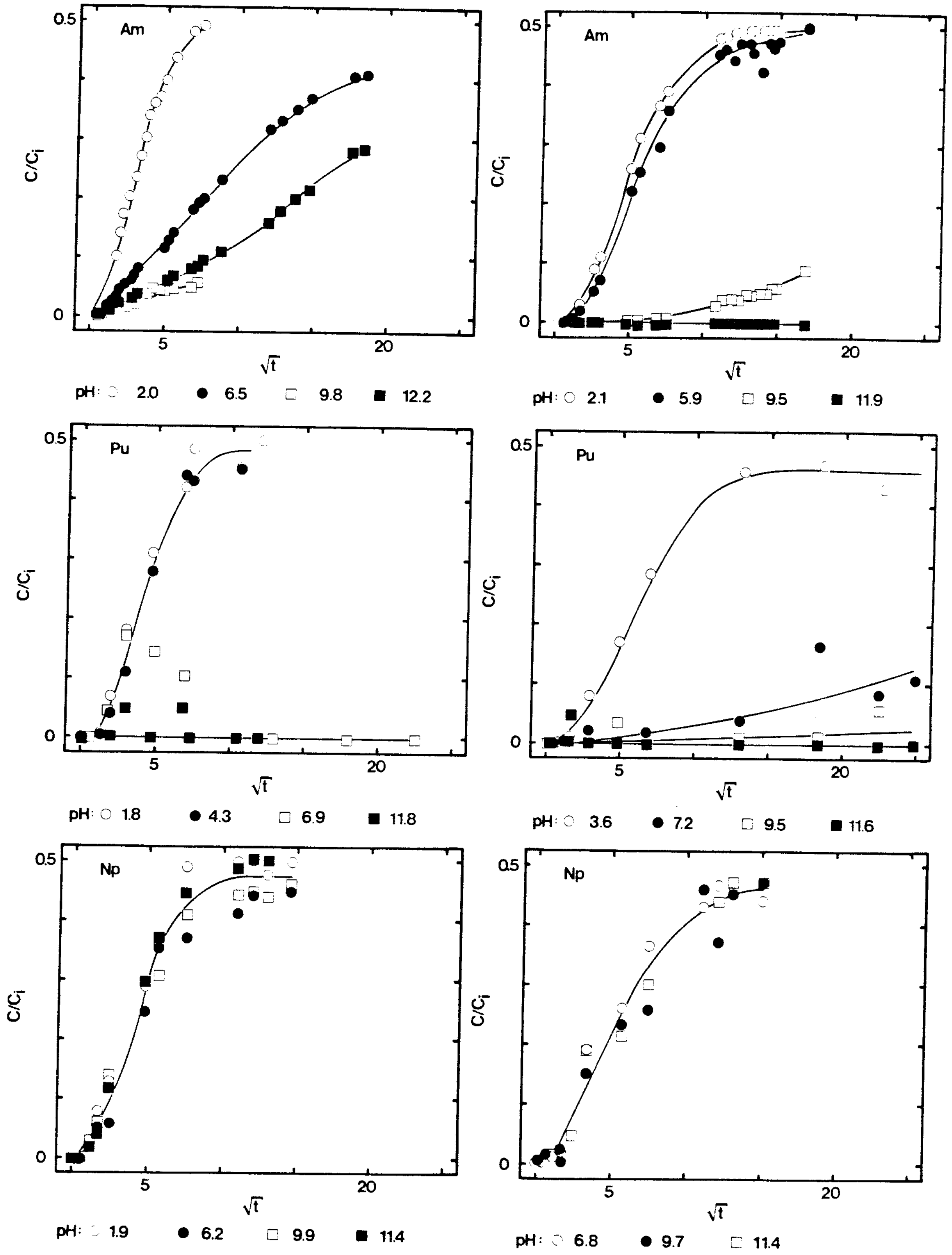


Figure 18. Diffusion of americium, plutonium and neptunium through membranes of pore size 0.400 µm (left) and 0.015 µm (right) at various pH (given in the Figure). Initial nuclide concentrations: 2.9×10^{-7} M (Am), 6.0×10^{-8} M (Pu), 2.1×10^{-7} M (Np).

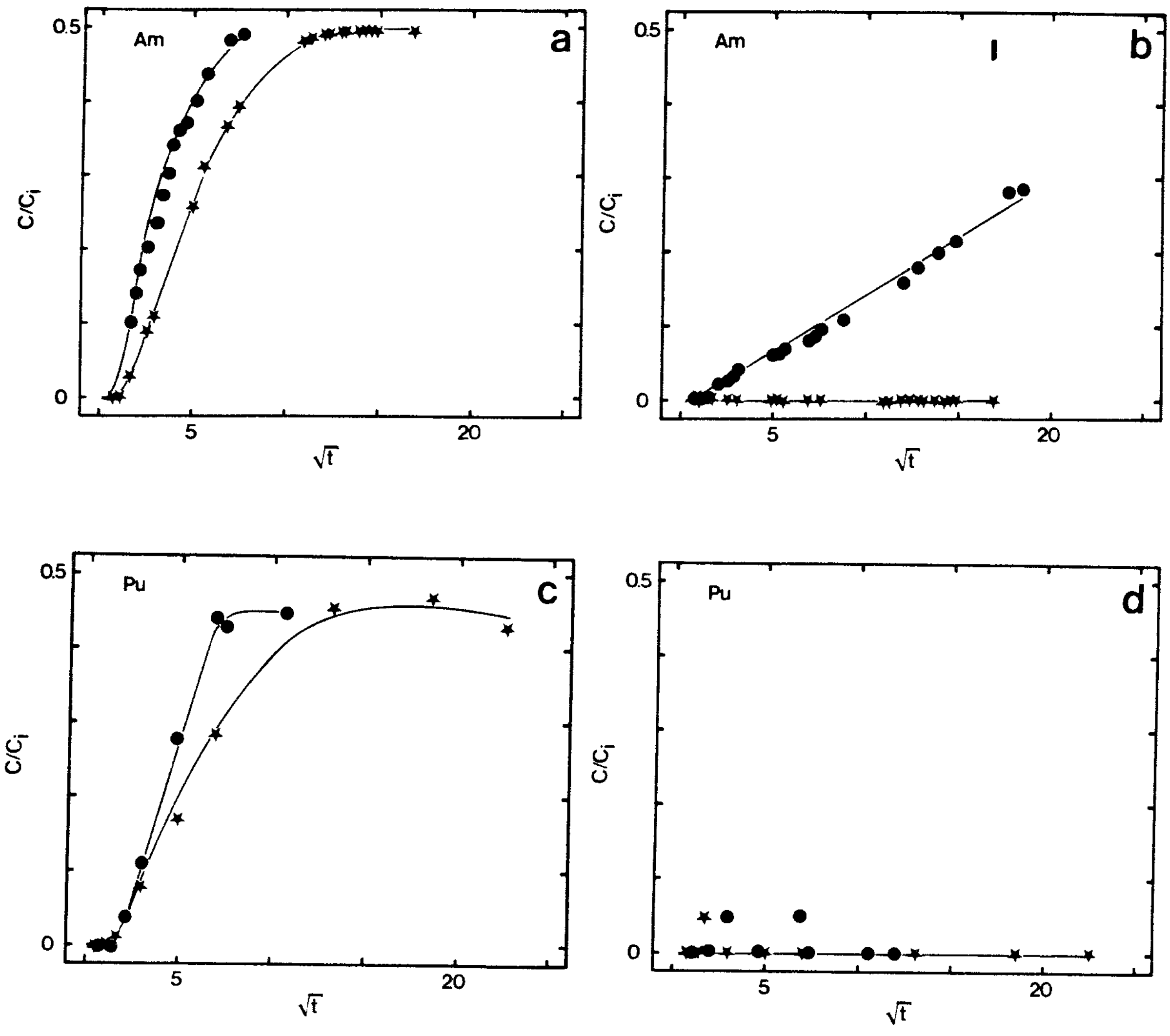


Figure 19. Diffusion of americium and plutonium through membranes at various pH.
pH=2.0-2.1 (a), 11.9-12.2 (b), 3.6-4.3 (c), 11.6-11.8 (d).
● pore size 0.400 µm ✶ pore size 0.015 µm

For neptunium pH did not seem to affect the diffusion rate, indicating that there was no formation of particles within the studied pH-range.

Fig. 19 shows the diffusion of americium and plutonium through filters with pore sizes 400 nm and 15 nm at constant pH. At pH about 2 the americium diffusion rates are rather similar for both filters, possibly only a little slower for the filter with the small pore size. This indicates that there are no particles in the solution. At pH about 12, however, there is no diffusion at all through the 15 nm filter, but a slow diffusion through the 400 nm filter. This indicates the existence of a fraction of particles with sizes between 15 nm and 400 nm, as well as particles with sizes larger than 400 nm.

For plutonium at pH about 4 there seems to be essentially no particles with sizes larger than 15 nm, but at pH about 12 there is no significant diffusion through the filters at all, possibly indicating a predominant particle size larger than 400 nm.

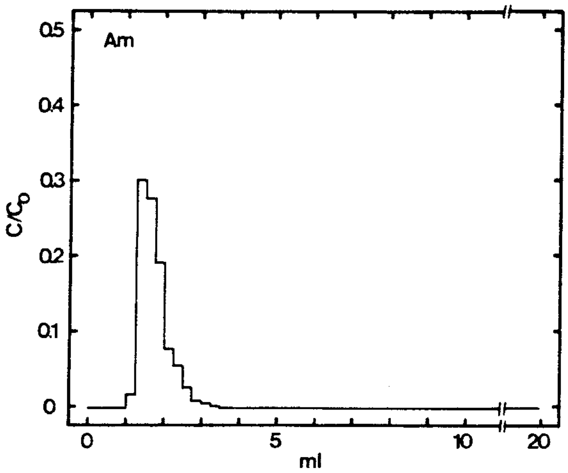
3.4. COLUMN EXPERIMENTS

Table 2 shows some results from transport studies in the α-Al_2O_3-packed columns.

Table 2. Mobility of americium, plutonium and neptunium in an Al_2O_3-packed column.

Element	Initial pH	Eluation, % [a]
Am	13	70
"	12	60
"	8	0.1
"	6	0
"	4.5	0
"	2	100
Pu	12	0
"	8	0
"	2	100
Np	12	0
"	8	100
"	2	100

[a] After more than 20 column volumes.



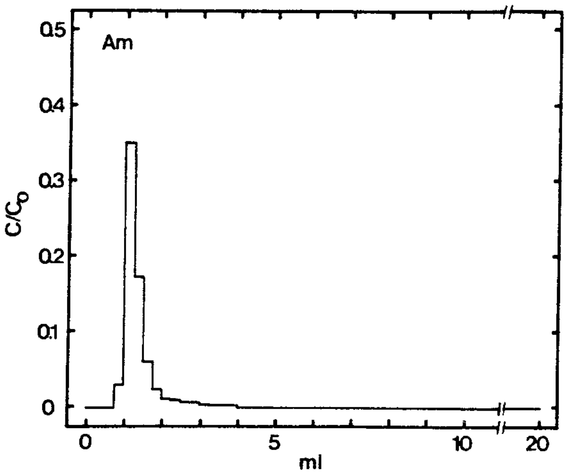


Figure 20. Eluation curves for americium at pH 2 (top) and pH 12 (bottom). (Ionic strength 0.01, flow-rate 0.05 ml/min, column filling Al_2O_3, 90-125 μm).

For americium it appears that most of the particle fraction in the high pH-range (12-13) are very poorly adsorbed by the column filling, but at lower pH (6-8) the particles are retained. The particles from the pH-range 12-13 were eluated after only 4-5 column volumes, Fig. 20.

For plutonium there is a particle fraction at pH above 8 which, however, does not pass through the column under the present conditions.

For neptunium no break-through was observed for the systems at pH 12.

4. CONCLUSIONS

The observed reduction of the initial actinide concentration in solution and the indications of a formation of colloidal fractions can be summarized as follows:

- A partial removal (after centrifugation only) of americium (trivalent) from an aqueous solution was observed in the pH-range 5-12, of plutonium (largely tetravalent) in the pH-range 3-12 and of uranium (hexavalent) in the pH-range 5-10 in the absence of any sorbing surfaces except for the vessel walls (polypropylene). The removal was significant already after six hours and was increasing with the storage time. Up to 60-70% of the total initial concentration had disappeared from the solution after four weeks (for americium and plutonium). Similar reductions of the concentrations in the solution phase were not observed for neptunium (pentavalent).

- The removal was increasing with the ionic strength of the aqueous phase. After long storage time (4 w) at high ionic strength (1 M) the removed fraction was almost 100% of the total initial amount, both for americium and plutonium, still in the absence of any sorbing surfaces but the vessel walls. Impurities in the salt solutions proportional to the salt concentrations can not account for the observed effects.

- The rate of americium removal increased somewhat with increasing temperature ($5^{o}C-70^{o}C$)

- The remaining concentrations in solution vs pH resembled the corresponding solubility curves in the americium and plutonium systems, with concentration minima at the pH of calculated minimum solubilities. Similar pH-dependencies were also obtained in systems were the initial total concentrations were several orders of magnitude below the solubility limit. The concentration reduction appeared to start at a somewhat lower pH in systems with the highest initial concentrations. The solubility product was probably reached in the plutonium systems of the highest concentrations, leading to the possible formation of a centrifugable precipitate.

- The processes behind the concentration reductions appeared to be fairly independent of the total nuclide concentration (below the solubility product).

- Most of the reduced fractions were sorbed on the vessel walls.

- Particle fractions were obtained both for americium and plutonium (radii above 20 nm); at pH 5-6 and above 12 for americium and at pH above 7-8 for plutonium. For plutonium the particle fractions dominated at pH above 12. The high-pH particle fractions may represent true hydroxy colloids, while the particle fractions at pH 5-6 in the americium system probably represent pseudo colloids related to impurities in the systems.

- For neptunium and uranium, which would exist predominantly as carbonate complexes of the type $MO_2(CO_3)_x$, x=1-3, there were essentially no significant formation of particle fractions, except for uranium at low ionic strength and pH around 6-7.

- The americium particle fraction at pH above 12 was anionic.

For both americium and plutonium, which appeared to exhibit colloidal properties in certain pH-ranges, the removal from the solution phase had maxima in the pH-range of prime importance in connection with underground storage of radioactive waste (pH 7-9). The particle fractions observed within this pH-range did not exhibit any enhanced mobility in the column and diffusion experiments. Thus, the formation of a non-sorbing highly mobile particle fraction of true colloidal species of americium or plutonium at pH 7-9 appears to be highly unlikely.

Studies of the formation and properties of colloidal fractions in the thorium system, as well as extended transport studies (column experiments, diffusion measurements) are in progress and will be presented in subsequent reports, where also more thorough discussions on the chemical nature of the observed particle fractions will be given.

5. ACKNOWLEDGEMENTS

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