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Some aspects on colloids as a means for transporting radio nuclides

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SOME ASPECTS ON COLLOIDS AS A MEANS FOR TRANSPORTING RADIO NUCLIDES

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KTH

SOME ASPECTS ON COLLOIDS AS A MEANS FOR TRANSPORTING RADIONUCLIDES

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Ivars Neretnieks

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1 Summary

The diffusivity of a macromolecule through a compacted clay layer was measured. The results indicate that molecules and thus colloids of this size - M = 24 000 - will not diffuse through the clay barrier in any appreciable amount.

Another set of experiments indicate that the clay will not be a source of colloids as the montmorillonite particles form a stable gel in groundwater with its rather high Ca^{2+} content.

Transport of radionuclides from the repository by adsorption on colloids coming from the clay will thus be small.

The colloid content of groundwater from Finnsjön was measured. This was less than 1 mg/l, so low a colloid content is of little importance.

Experiments designed to observe the adsorption of colloidal particles from the clay on rock surfaces were inconclusive.

2 Background

In a repository for radioactive waste, the consequences of a leak in the capsule must be considered.

The radioactive waste may be transported by the water as ions and as charged and uncharged complexes. In addition to this, many of the nuclides may exist as colloids. Colloids are stable suspensious of very small particles. The particles range in size from about 3 nm to 300 nm. They are aggregates of some 10^3 - 10^9 atoms. There is no definite border between colloids and solutions on one hand and colloids and suspensious on the other hand. Colloidal particles in many ways behave as macromolecules of the same mass. The colloids will not be retarded by the same mechanisms as the solved species and might in some instances travel with the velocity of the groundwater. Many of the important nuclides eg. Pu, U, Am, Np will be strongly retarded when they are in solved form. If some fraction exists in colloidal form this fraction may reach the biosphere very much faster than the solved fraction.

There are two types of radiocolloids. The true colloid can be formed from a supersaturated solution of the radioisotope. The pseudocollid is formed by adsorption of the radioisotope on colloidal particles in the water.

The pseudocolloids may exist in very low concentrations $< 10^{-10}$ mol/l Kepak (1) whereas the true colloids form at higher concentrations 10^{-5} mol/l Davydov (2).

Colloids may adsorb strongly on various surfaces and may also coagulate and precipitate. The latter may be enhanced by addition of electrolytes.

Various sources for radiocolloids are possible.

1 A true colloid may form when the glass or UO_2 matrix dissolves by leaving behind small particles of low solubility.

2 A true colloid may form by precipitation of some nuclide on its way out through the backfill material which consist of bentonite clay in the KBS study.

3 A true colloid may form outside the barrier by precipitation

4 A pseudocolloid may form by adsorption on small clay particles in the clay barrier.

5 A pseudocolloid may form by adsorption on clay or other particles outside the barrier.

Radiocolloids formed on the inside of or in the compacted clay barrier according to mechanisms 1, 2 and 4 will have to wander through the barrier. The water velocity is negligible. Any transport must be due to diffusion. An experiment was therefore designed to measure the diffusivity of particles of colloidal size in compacted clay. This is described below under heading 3.

Colloids formed outside the barrier by mechanisms 3 and 5 or arriving from inside the barrier may be adsorbed on the rock surfaces as the water flows in the fissures. An experiment was designed to investigate this. It is described under heading 5.

Two sources of colloidal carrier particles were identified. The groundwater itself may supply them and they may be supplied by the montmorillonite particles of the day.

A thorough theoretical treatment of montmorillonite behaviour and its tendency to form stable gels was made within the KBS project by LeBel 1978 (4). In this investigation the critical coagulation concentration of Ca^{2+} was investigated by two ex-

perimental methods. They were both based on measurements of escaping particles from a bentonite gel. In our experiments the depletion of particles from a bentonite suspension was measured.

Our experiments are described under heading 4.

The colloid content of groundwater from Finnsjön was measured. This is described under heading 6.

3 Diffusion of colloid particles through compacted bentonite

3.1 Experimental results

A solution of sodiumlignosulfonate (LS) with a mean molecular weight of 24 000 was used to simulate small colloid particles. This corresponds to a molecular diameter of about 6 nm. The experiment is described in detail in (3).

A 3 mm thick compacted bentonite bed was contained between two porous metal plates. On one side a LS solution in synthetic groundwater was circulated and on the other side pure synthetic groundwater was used to collect any LS which diffused through the slab. No LS was found on the pure water side of the barrier after 36 days. The dectection level of the measuring method used -UV- was such that LS should have been detected if the diffusivity would have been larger than $3 \cdot 10^{-14}$ m^2/s. This low or lower values were expected as the channels in compacted bentonite should be very small.

3.2 Transport capacity

Due to the very low diffusion of macromolecules and thus colloids through compacted bentonite, this transport mechanism will be negligible in comparison to other mechanisms. This is clearly seen from the following example.

Assume that the copper capsule has deteriorated entirely and the uranium oxide matrix dissolves and gives off a colloidal solution with a colloid concentration of 1 000 mg/l. This is more than 5 times higher than the highest concentration noted in Kepak's (1) compilation. The colloidal particles diffuse through the bentonite barrier according to Fick's law if no adsorption in the barrier is assumed $N = D \cdot A \cdot \frac{dc}{dz}$

For a copper capsule the area available to transport is about 15 m^2,

$D = 3 \cdot 10^{-14}$ m^2/s and $\frac{dc}{dz} = \frac{1000}{0.375}$ g/m^3, m

This gives $N \approx 3$ mg/year. It will thus take some 400 million years to dissolve the uranium content - 1.4 tons - of one cannister by this mechanism.

4 Montmorillonite as a source for colloids

The montmorillonite clay consists of very small particles which may form colloidal solutions in water with a low salt concentration Le Bel (4). As a complement to Le Bel's investigation we made suspensions of bentonite in waters with various initial calcium concentrations and measured the resulting clay and calcium concentrations. Experiments were also performed with distilled water and synthetic groundwater (5).

4.1 Experimental

Normal 11 cm test tubes were filled with known amounts of clay and water. After vigorous shaking the clay was allowed to settle. After 10 and 40 days samples were taken of the upper centimeter of the water in the test tube. This was analysed for Ca^{2+} by an atomic absorption spectrofotometer and for clay particles by UV-light absorption in a light spectrophotometer.

Due to the ion exchange capability of the montmorillonite the original Ca^{2+} is exchanged for Na^{+} to various degrees.

This may be seen i table 1. The original Ca^{2+} concentration is given in column 1. The amount of clay added is given in column 2. Column 3 and 4 show the clay and Ca^{2+} concentration in the solution after 10 days and columns 5 and 6 after 40 days. In figures 1 and 2 the clay concentration of the water is plotted versus the Ca^{2+} concentration. Figure 2 shows a higher clay concentration than figure 1. This is because the test point was much nearer the gel surface where the concentration was higher.

Both measurements show that at 10 - 20 mg Ca^{2+}/l the particles precipitate from the solution. This is deemed to be a stronger indication on the unwillingness of bentonite to form colloidal solutions at high Ca^{2+} concentrations than experiments which

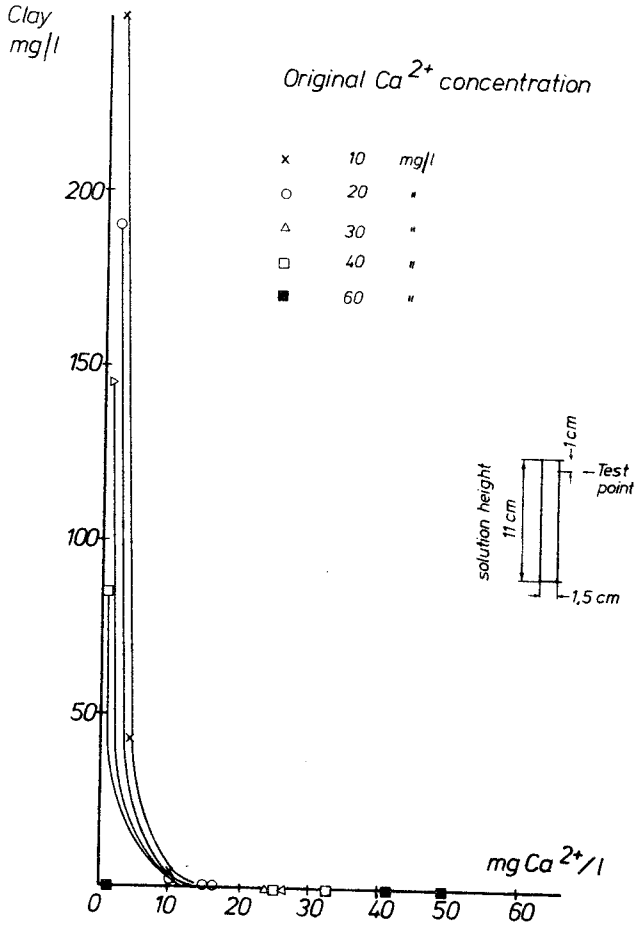

Figure 1. Clay concentration in water as a function of calcium ion concentration. 10 days after mixing.

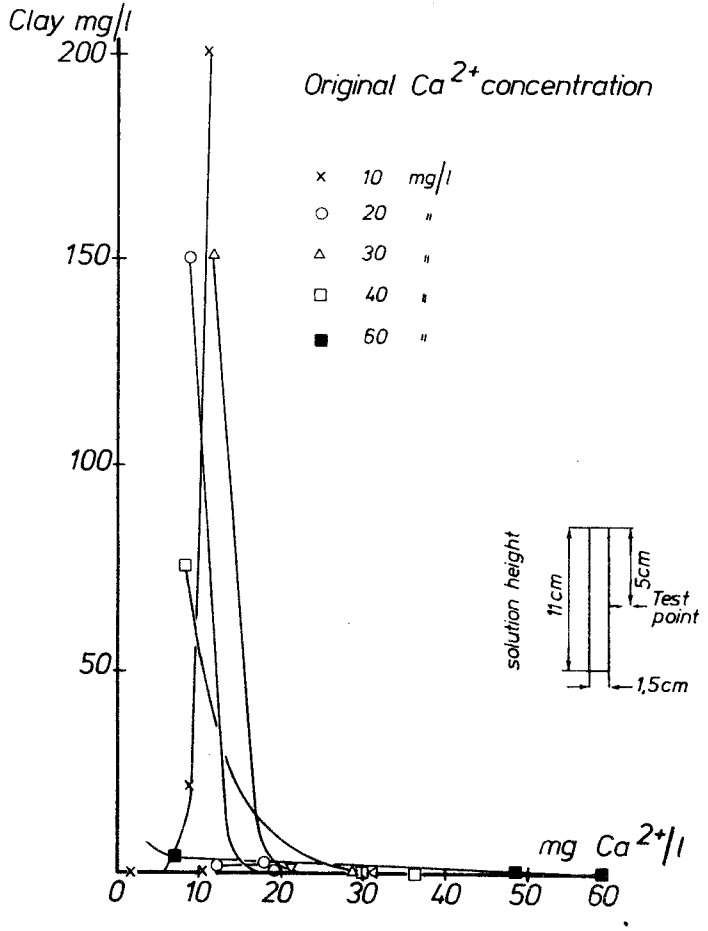

Figure 2. Clay concentration in water as a function of calcium ion concentration. 40 days after mixing.

start with a gel and measure the increase of particles in the water above.

These results are quite in accordance with those of Le Bel (4).

Table 1

	original		10 days		40 days	
	1	2	3	4	5	6
No	Ca^{2+} mg/l	clay mg/l	clay mg/l	Ca^{2+} mg/l	clay mg/l	Ca^{2+} mg/l
1	10	10000	250	2.12	200	12.6
2	10	1000	42	4.54	21	8.7
3	10	100	4	10.34	0.1	11.3
4	10	10	1	4.13	0.5	2.5
5	20	10000	190	1.89	150	8.8
6	20	1000	3	10.18	0.8	12.0
7	20	100	0.8	15.94	3	18.0
8	20	10	0.8	16.50	0.3	18.7
9	30	10000	145	1.64	150	10.6
10	30	1000	< 1	18.05	0.8	21.3
11	30	100	< 1	24.07	0.2	23.6
12	30	10	< 1	25.40	0.3	30.4
13	40	10000	84	1.65	74	8.0
14	40	1000	< 1	25.20	1	30.4
15	40	100	< 1	32.20	<0.1	36.7
16	40	10	< 1	32.40	<0.1	36.8
17	60	10000	< 1	2.73	4.3	6.9
18	60	1000	< 1	41.50	1.2	48.3
19	60	100	< 1	48.90	<0.1	59.6
20	60	10	< 1	49.90	<0.1	58.4

5 Adsorption of montmorillonite particles on granite surfaces

Colloidal particles are known to adsorb on some surfaces. Kepak (1) Davydov (2). An experiment was designed to study if montmorillonite particles are adsorbed on granite surfaces.

Granite from Finnsjön was crushed and sieved. A fraction 0.3-0.7 mm was used to form a bed in a 8 mm i.d. glass tube. The bed length was 100 mm. Two clay suspensions were used. In one experiment clay was suspended in the synthetic groundwater proposed by Rennerfelt (5). This suspension had a concentration of clay particles equal to $2.5 \cdot 10^{-9}$ g/g* due to coagulation and precipitation of the major part of the clay. The suspension was fed through the bed with a linear superficial velocity of 5 m/h. A velocity of this magnitude has been found sufficiently low in other adsorption experiments to allow time for the adsorbing species to settle on the particles.

No adsorption of clay was noted. The effluent had essentially the same concentration as the feed during the first hour the experiment was run. Thereafter a decrease in concentration to about 15 % of the inlet concentration was noted. The result is shown in figure 3.

The decrease in particle concentration can be due to adsorption or precipitation or filtering effects due to the increase in size of the particles. The experiment thus is inconclusive except that no major adsorption occurs.

The experiment was repeated using a clay dispersed in distilled water. The clay concentration initially was $67 \cdot 10^{-9}$ g/g water. No adsorption was observed. The result is shown in figure 4.

It was therefore concluded that no major adsorption of the montmorillonite particles on granite surfaces can be expected.

* Analysis were performed by Lars Ödberg and co-workers at Lindköping technical university using a light scattering technique. This is described in (4).

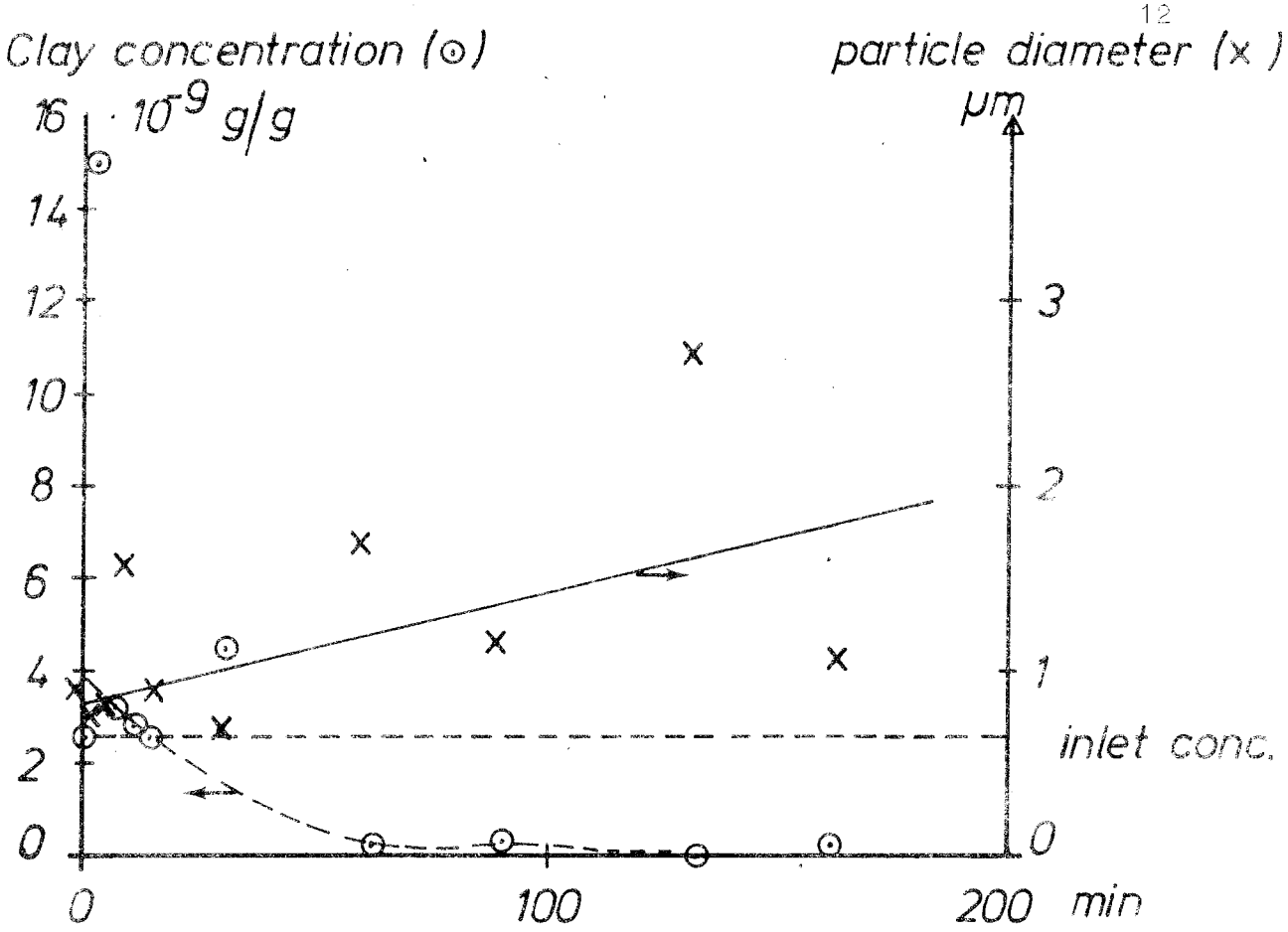

Figure 3. Clay concentration and particle diameter in effluent from bed of granite particles. Synthetic groundwater.

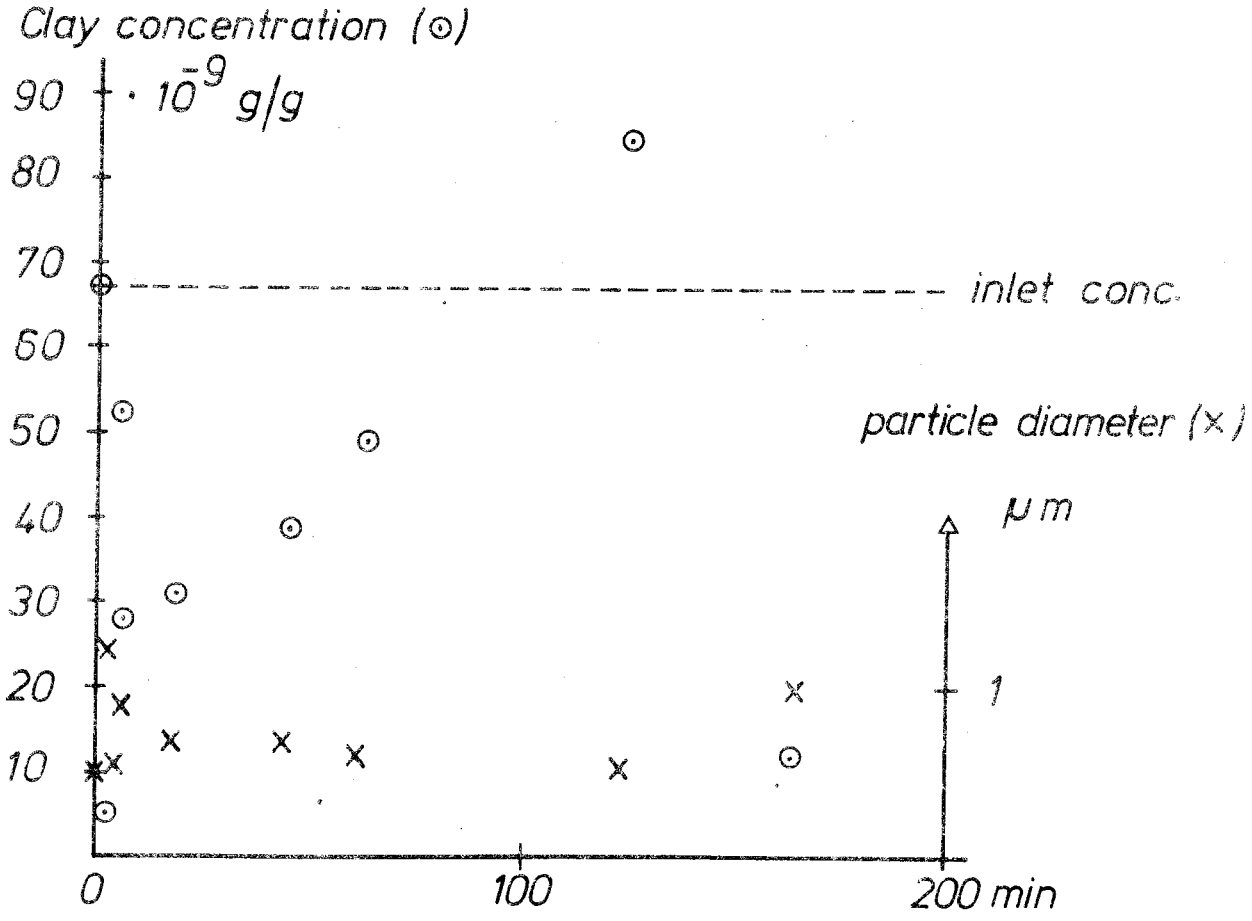

Figure 4. Clay concentration and particle diameter in effluent from bed of granite particles. Distilled water.

6 Colloid content of groundwater at Finnsjön

All groundwaters contain colloidal particles (6). No data have been found in the literature on the colloid content of groundwaters from deep wells.

Measurements were performed on water from Finnsjön. The water is taken from a bore hole at a depth of about 500 m. The measurements were made under the original nonoxidizing conditions. The previously described (Le Bel) light scattering technique was used.

The radius of the largest particles was 0.6 μm. The particle content was $0.6 \cdot 10^{-9}$ g/g.

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