
Model Calculations and Assessment of the Importance of the Colloidal based Radionuclide Transport in a Waste Repository in Granite

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ABSTRACT: Highly compacted bentonite buffers are planned to be used as engineered barriers in the near-field of nuclear waste repositories in crystalline formations. The erosion of these bentonite buffers at the boundary to the surrounding Excavation Disturbed Zone (EDZ) can be a source of colloids leading to a notable concentration of colloids in the EDZ water, which is high above the natural colloid concentration in granitic groundwater. The colloids are transported with the fracture water through the far-field of the repository.

In this work the influence of the bentonite colloids on the radionuclide transport in the far-field and on the resulting dose is studied for the Finnish safety assessment TILA-99 [7]. This work was carried out as part of the Benipa project [1].

The release of radionuclides from the far-field is calculated without the presence of colloids in the reference case and is compared to the release rate with the presence of colloids at different concentrations. This study shows, that the colloids can have an impact on the release rate of some special radionuclides, however these nuclides have shown not to be dose-relevant in this calculations.

1 INTRODUCTION

Colloids are particles of 1 nm to 1 mm in size which are suspended in water. They are ubiquitous in natural groundwater and are known to be able to influence radionuclide transport if found in sufficient concentration since the radionuclides can be sorbed on the colloids, what changes their transport behaviour [2].

Especially in fractured rock media like granite their influence is of interest: Due to their size the colloids cannot penetrate the matrix pores of the granite rock and consequently they are only transported in the fractures. Radionuclides which are sorbed on colloids are therefore also excluded from matrix diffusion, which is generally one of the most important reasons for radionuclide retardation in fractured media. In addition radionuclides being sorbed on colloids cannot be sorbed elsewhere. Both effects can lead to an accelerated radionuclide transport. For natural groundwater colloids these effects are well known and a mathematical description was developed to calculate the influence of colloids on radionuclide transport [6].

Recent experiments from Missana et al. [4] on FEBEX bentonite have shown that the erosion of a bentonite buffer can be an additional source of colloids and lead to a notable

concentration of colloids in the fracture water, which is high above the natural colloid concentration in granitic groundwater.

Since the generation of colloids at the granite / bentonite boundary seems to be likely according to these experiments, GRS investigated their influence on the radionuclide transport for the Benipal reference case for borehole emplacement. The term colloid is used in the following synonymous for colloids generated at the granite / bentonite interface.

2 MODEL AND CODE

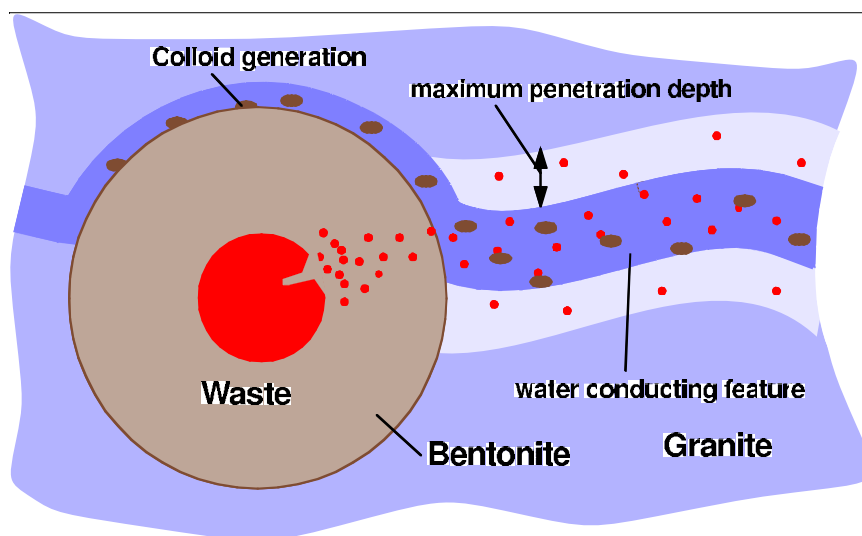


Figure 1: Colloid generation and transport process

A simplified description of the colloid facilitated transport process is shown in figure 1. A fracture intersects the EDZ around a borehole resulting in a water flow at the bentonite / granite boundary, where the colloids are generated. Radionuclides diffuse through the bentonite buffer and are also released to the fracture water. The radionuclides then are partially in solution, diffusing into the granite matrix or are sorbed on the colloids. A mathematical model for the colloid facilitated radionuclide transport under these assumptions was developed by Smith [6]. This model was implemented in the CHETMAD code, which thereupon was used for modelling the influence of the colloid facilitated transport on the Benipal reference case.

The transport processes modelled in CHETMAD are advection, diffusion and dispersion for radionuclides and colloids. Linear sorption is taken into account for the radionuclides on the granite matrix and for the colloids on the fracture walls. The following additional assumptions are made in CHETMAD:

- sorption equilibrium is reached instantaneously,
- the sorption of radionuclides on the fracture walls can be neglected in comparison to the sorption on the granite matrix, i.e. radionuclides do not sorb on the fracture walls,
- colloids do not penetrate the matrix,
- colloid concentration is constant in space and time,
- dispersion length of nuclides in solution and of those sorbed onto colloids is the same,
- distribution coefficient for nuclides on mobile and immobile colloids is the same.

3 DEFINITION AND DATA

Experiments from Missana and also Schäfer et al. [5] showed that even at high concentrations the bentonite colloids are stable over a long period of time under sweet-water conditions, but are becoming unstable in water conditions with an ionic strength higher than 10^{-2} mol/l. Therefore, the influence of the colloids on the radionuclide transport was tested for a case under sweet-water conditions. The near-field and far-field parameters were chosen as for the ns50 case in the TILA-99 safety assessment [7]. The most important parameters of the near-field and the far-field are given in the tables 1 and 2.

The information about colloid generation at the bentonite boundary is still quite poor. Missana et al. [4] at CIEMAT made two types of column experiments simulating the granite / bentonite boundary; in dynamic and quasi static water flow conditions. The dynamic experiments showed that the colloid concentration increases with water flow. At a water flow rate of 175 ml/y a total concentration of more than 200 mg/l of solids with up to 3 mm in size was found including a fraction of particles with less than 450 nm in size of 0.7 mg/l. The water velocity in that experiment can only be estimated from a best guess for the fracture aperture to be about 1mm. The experimental cell geometry is 150 mm in length and 38 mm in width thus the fracture volume results to be 5.7 ml. With the lowest volume flow rate used in the experiment of 61.3 ml/y we obtain that the fracture volume is replaced 11 times a year and the mean water velocity is 1.65 m/y.

However, mechanical erosion does not play the only role in bentonite colloid generation. Colloids can be formed even in static conditions by forming a clay gel. The quasi static experiment lasted one month and resulted in a total concentration of 35 mg/l of solids up with to 3 mm in size including a fraction of particles with less than 450 nm in size of 8 mg/l.

The influence of the bentonite colloids on the radionuclide transport in the far-field and on the resulting dose is studied for the Finnish safety assessment TILA-99. According to the experiments by Missane et al. the colloid concentration was varied in the calculations in four steps to be 0, 1, 10 and 100 ppm. Sorption of colloids on the fracture walls was not taken into account in the calculations neither was an enhancement of the colloid velocity relative to the water flow. The distribution coefficients for the radionuclides on the granite matrix were taken from the TILA-99 study. Data for distribution coefficients for nuclides on the bentonite colloids are not available thus the reference values for nuclides on the bentonite buffer from the SPA study were used [3]. The distribution coefficients for the most important nuclides are given in table 3.

To estimate the influence of the colloids on the radionuclide transport a dose rate was calculated from the release rate of nuclides from the geosphere. The dose conversion factors were derived according the WELL-97 scenario [7]. This scenario is based on the release of the radionuclides into the catchment-area of a deep well for extraction of drinking water.

Table 1: Near-field parameters

Parameter	Value
Inventory	TILA-99
Container Lifetime	10 000 a
Outer diameter of the container	1.05 m
Outer diameter of the bentonite buffer	1.75 m
Bentonite porosity	0,41
Diffusion coefficient in Bentonite	$2.33 \cdot 10^{-10} \text{ m}^2/\text{s}$

Table 2: Far-field parameters

Parameter	Value
Transport pathway length	600 m
Transit time	25 y
Volume flow in the EDZ	$1.2 \cdot 10^{-3} \text{ m}^3/\text{y}$
Diffusion coefficient in the matrix	$6.3 \cdot 10^{-4} \text{ m}^2 \cdot \text{a}^{-1}$
Matrix porosity	$1 \cdot 10^{-3}$
Distribution coefficient for nuclides on matrix	table 3
Distribution coefficient for nuclides on colloids	
Colloid concentration	0; 1 ; 10 ; 100 ppm
Distribution coefficient for colloids on fracture wall	0 m
Factor for enhancement of colloid velocity relative to water velocity	1

Table 3: Distribution coefficients for selected nuclides

Element	Distribution coefficient [m^3/kg]	
	on granite matrix	on colloid
C	$1 \cdot 10^{-4}$	0.01
Cl	0	0
Ni	0.1	1
Se	$5 \cdot 10^{-4}$	$5 \cdot 10^{-3}$
Sr	$5 \cdot 10^{-3}$	0.01
Zr	0.2	1
Nb	0.02	1
Tc	0.05	0.1
Pd	$1 \cdot 10^{-3}$	1
Sn	$1 \cdot 10^{-3}$	1
Cs	0.05	0.01
Sm	0.02	5
Ra	0.2	0.01
Th	0.2	5
Pa	0.05	1
U	0.1	5
Pu	0.5	5
Np	0.2	5
Am	0.04	5
Cm	0.04	5

4 RESULTS

Figure 2 shows the influence of the colloid facilitated radionuclide transport on the sum dose rate. The black line (0 ppm) denotes for the dose rate obtained in the reference case without any colloids. The other three curves show the influence for different colloid concentrations of 1, 10, and 100 ppm.

It follows that the total effect of the colloid facilitated radionuclide transport on the sum dose rate for the examined case is negligible for times below about $1 \cdot 10^5$ y. There even can be found a small reduction in the dose rate due to the colloid facilitated transport for concentrations of 1 and 10 ppm at about $7 \cdot 10^4$ y. This circumstance is explained below.

For large times greater than $1 \cdot 10^5$ y the dose rate is increasing with colloid concentration. However, the maximum increase for 1 ppm and 10 ppm is only about a factor of 1.5.

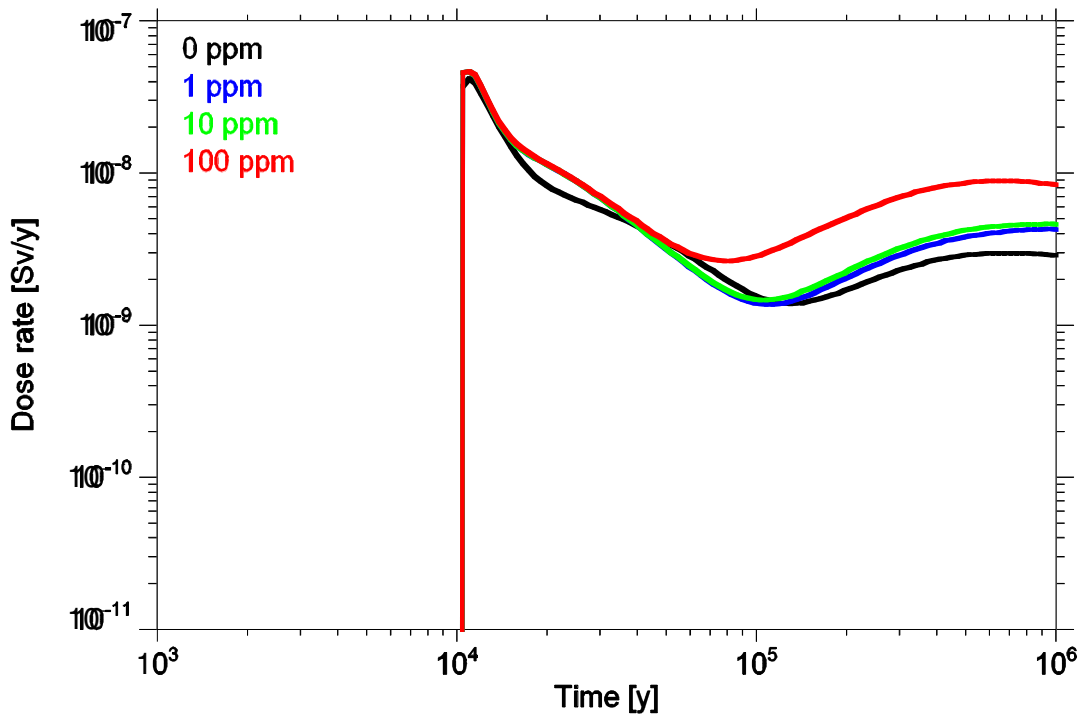


Figure 2: Sum dose rate versus time for 4 different colloid concentrations

The colloid facilitated transport in general has two influences on the time dependence of the radionuclide transport.

- The presence of colloids accelerates the radionuclide transport process. Because of the radioactive decay of the radionuclides a smaller travel-time can also result in a smaller fraction of decayed nuclides and thus in a higher dose rate.
- The nuclides are not only transported faster, but many nuclides are transported with the same velocity namely the one of the colloids. This leads to more nuclides to arrive at the biosphere at the same time i.e. the tailing of the dose rate versus time curve is less. The peak of the dose rate versus time curve is sharper and has an increased maximum. Thus the effect also results in an enhanced dose rate.

The detailed effects of the colloids on radionuclide transport are exemplarily discussed in the following for the nuclides Sn-126 and Pu-240. The dose rate resulting from Sn-126 is shown in fig. 3. The maximum of the dose rate resulting from the nuclide Sn-126 is increased by the colloid facilitated transport by a factor of about 2. This increase is nearly independent of the three colloid concentrations used for modelling, what indicates a saturation effect; the maximum amount of Sn-126 is already sorbed at a colloid concentration of 1 ppm. This is due to a low distribution coefficient of Sn-126 in the granite matrix compared to the distribution coefficient on the colloids.

The increase in dose rate of Sn-126 is mainly a consequence of a decreasing tailing of the dose curve due to the presence of colloids. More nuclides arriving at earlier times result in less nuclides arriving at later times and thus in a decrease in the dose rate at times later than the arrival of the maximum, at about $7 \cdot 10^4$ y. Since Sn-126 is the dominating nuclide for the dose at this point in time, the sum dose is reduced, too.

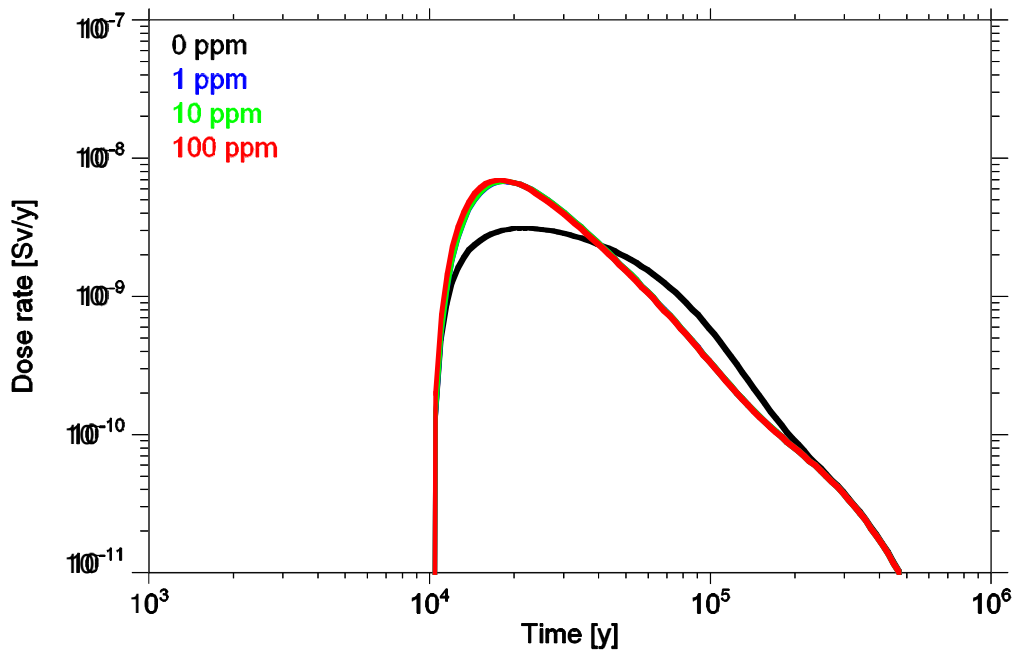


Figure 3: Dose rate of Sn-126 versus time for 4 different colloid concentrations

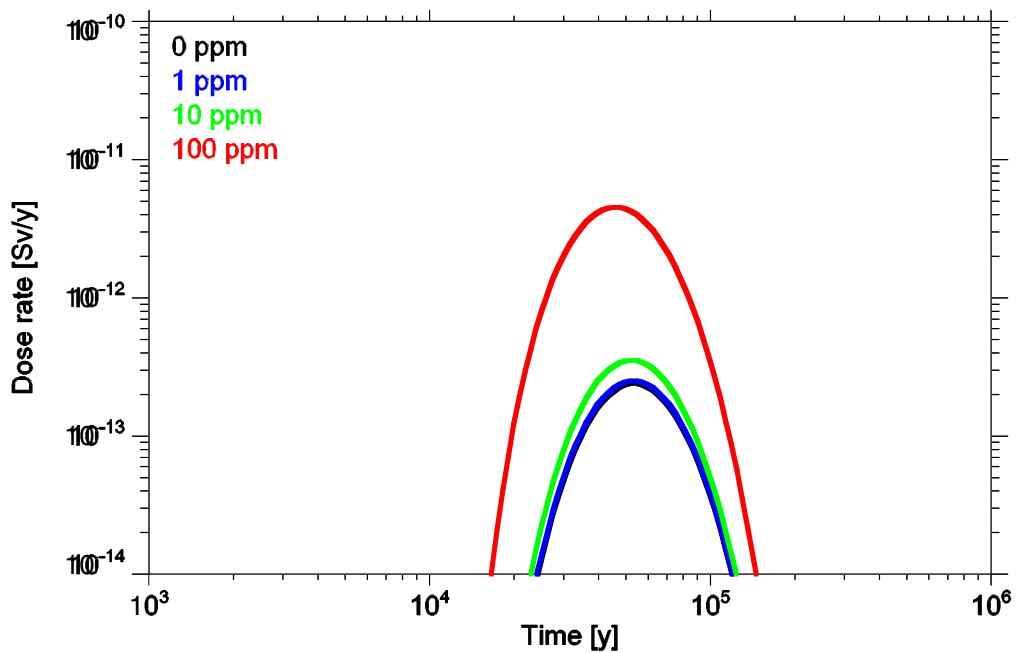


Figure 4: Dose rate of Pu-240 versus time for 4 different colloid concentrations

The dose rate resulting from Pu-240 is shown in fig. 4. This nuclide is not relevant for the sum dose, but it is of interest since the influence of colloid facilitated transport has a different behaviour than for Sn-126. There is a negligible influence on Pu-240 at a colloid concentration of 1 ppm, a moderate increase of about a factor 1.5 at a concentration of 10 ppm and a high increase of nearly a factor of 20 at a colloid concentration of 100 ppm. This behaviour is linked to the half-life and travel-time of Pu-240.

A significant increase of the dose rate due to colloid facilitated transport can be expected whenever the mean travel-time of a nuclide is about twice up to six times its half-life. After

a travel-time of two to six times the half-life the radioactive decay has lead to an intense decrease of the release rate from the far-field. Even a small reduction in the travel-time due to colloid facilitated transport can than lead to a significantly less amount of decayed nuclides and thus to a notable impact on the release rate from the far-field.

The half-live of Pu-240 is 6 542 y. At the time of arrival in the biosphere most of the Pu-240 has already decayed since the travel-time without colloids is about 8 times its half-life. A colloid concentration of 100 ppm leads to a reduction in travel-time of about 9 100 y, which is about 1.4 times the half-life of Pu-240 resulting in a significant less fraction of Pu-240 to decay.

This example shows that colloids generated at the bentonite buffer can have a significant influence on the transport of some nuclides and could also have on the dose rate if these nuclides are dose-relevant.

5 CONCLUSIONS

From the modelling of the colloid facilitated radionuclide transport the following conclusions can be drawn:

- The colloids can have an effect on the radionuclide transport. Since the generation of colloids at the bentonite/granite interface cannot be excluded, the colloid facilitated radionuclide transport from these colloids should be investigated.
- The effect of colloid facilitated transport depends on the distribution coefficient for the nuclide on the colloids. The highest distribution coefficients have the actinides as well as Ni, Zr, Nb and Sn.
- For the examined case the impact of the colloid facilitated radionuclide transport on the sum dose rate is rather low. A colloid concentration of 10 ppm leads to an enhancement in the dose rate by a factor of about 1.6.
- There can be a higher impact on the dose rate from some special nuclides depending on their half-life and mean travel-time in the geosphere. Although these nuclides are not relevant for the dose rate in the examined case, the colloid facilitated radionuclide transport can have a higher impact for a different type of waste or far-field parameters. Thus the colloid facilitated radionuclide transport should be considered in the long-term safety analyses.

6 REFERENCES

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