

EROSION OF BENTONITE BUFFER IN A KBS-3 REPOSITORY

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Been directed to understanding the behaviour of expansive gel when the pore water is below the CCC. In the latter case most work has been made on the behaviour of *sodium dominated smectite* because if most There are concerns that the bentonite buffer surrounding the canisters with spent nuclear fuel may erode when non-saline groundwaters seep past the buffer during long periods of time. The non-saline waters may solubilise the smectite particles in the bentonite and carry them away as colloidal particles. This is known to happen if the water content of ions is below the critical coagulation concentration CCC. Above the CCC the smectite forms a coherent gel, which does not release particles. It was also found in the present investigation that the gel thus formed will not be sheared off by the shear forces of the seeping water.

This paper reports on a study that has been directed to assess under which conditions the pore water composition of the gel at the gel/water interface could be lower than the CCC. Another main effort has of the counterions in the smectite are calcium such smectites do not readily release colloidal particles. The reason is that the smectite particles in a calcium dominated gel hold together in larger and less mobile particles consisting of many individual smectite sheets. Such particles are too heavy to be much influenced by Brownian motion. The particles are also bulkier and are much easier strained in narrow pores and passages. Strongly dominated calcium gels are therefore not expected to cause erosion problems.

Sodium dominated gels in waters below the CCC are therefore the main concern because such gels will expand “forever” and release colloidal smectite particles to the seeping water. In addition, as the gel grows increasingly dilute, it becomes less viscous and can *flow* downstream.

We have developed a Dynamic model for sodium gel expansion in fractures where the gel soaks up non-saline water as it expands. The model is based on a force balance between and on smectite particles, which move in the water. Attractive van der Waals forces, vdW, repulsive electric diffuse layer forces, DDL, gravity and buoyancy forces and forces caused by the gradient of chemical potential of the particles act to move the particle in the water. Friction between particles and water restrain the movement. The DDL forces strongly depend on the type of ion and concentration in the water surrounding the particles. The Dynamic model of gel expansion showing the evolution in time and space of a gel was successfully tested against expansion experiments in test tubes. The expansion was measured with high resolution and in great detail over many months by Magnetic Resonance Imaging, MRI. The model also predicted the gel expansion through filters with very narrow pores well. In addition the model predicts the CCC fairly well, order to orders of magnitude better than the conventional DLVO theory.

A gel viscosity model of dilute gels was derived, which accounts for ion concentration influence as well as the volume fraction of smectite in the gel. The model accounts for the presence of the DDL, which seemingly makes the particles larger so that they interact at lower particle densities. The viscosity model uses experimental data to obtain the necessary fitting parameters but is otherwise based on established theories of suspension viscosity.

These models form the core of the erosion model. Both show a strong dependence on the ionic strength of the pore water. Simulations were performed for a case where the gel expands outward into the fracture that intersects the deposition hole. Fresh groundwater approaches and passes the gel/water interface. Smectite colloids move out into the water due to the repulsive forces between the particle and by Brownian

motion (effect included in the Dynamic model). The dilute gel/sol is mobilised and flows downstream in a thin region where the viscosity is low enough to permit flow. Sodium diffuses from the compacted bentonite into and through the expanding gel towards the gel/water interface and further out into the seeping water. Mass transfer resistance for ions as well as smectite particles in the seeping water is accounted for in the simulations. The sodium concentration profile in the gel influences the repulsive forces between the particles as well as the viscosity of the expanding gel. Under the most unfavourable circumstances, i.e. at high flowrates and large fracture apertures, considerable loss of smectite can be expected for a buffer that consists of *only smectite*

Other calculations have been made to assess under which conditions of flowrate, water compositions and initial bentonite chemical compositions the water composition at the gel/water interface could become larger than the CCC. At the same time the proportion of calcium and sodium as counter ions in the smectite at the gel/water interface was studied. This was done because should the calcium make up more than about 90 % of the counterions, the smectite behaves very differently from than smectite with less calcium. There are indications that such gel will not release colloids readily. In a number of studied cases such stabilizing conditions could not be achieved. In these calculations we have accounted for ion exchange in the expanding gel, for diffusion of ions in the gel, for transport to and from the seeping groundwater, and of the dissolution of soluble minerals that may supply the gel with ions.

We conclude that with our present understanding of the processes it is not possible to affirmatively state that erosion of *pure* smectite gels cannot occur to a considerable extent.

However, the commercial bentonites that have been extensively investigated in earlier and present SKB investigations contain tens of percent of non-smectic accessory minerals. These materials do not exhibit the strong repulsive forces as the smectites do because the surface charge density is essentially negligible at the circum neutral pH expected. Furthermore the particle size of these materials is one to two orders of magnitude larger than those of the smectite particles, so they are much less mobile and they could not be lost as colloids in the water. It is highly likely that such particles will penetrate some distance into the fracture and form a porous network through which the smectite particles must pass. We have investigated the idea that the accessory particles left behind could form a porous bed with small enough pores to slow down smectite loss after a short initial period when some smectite is lost leaving the accessory mineral particles behind to form the filter bed. Already a few cm long region of detritus material in the fracture has a strong restraining effect on the rate of smectite loss by colloidal diffusion through the pores.

A set of experiment where highly compacted beds of smectite between metal filters flushed by water on the outside show that sodium smectite readily penetrates filters with 2 µm pore size or larger but are stopped by 0.5 µm filters. The smectite particles are one to a few µm large.

Furthermore, theoretical deliberations and modelling suggest that the particle size distributions are favourable for smectite *straining* in already a thin bed of accessory minerals. The porous bed can have so small pores that the smectite particles are prevented from passing so narrow pores. A set of experiments where mm thick beds have been used to filter a suspension of smectite particles show very promising results.

Our overall conclusion is that a buffer with practically only smectite cannot be proved not to erode significantly with the flowrates and water compositions.

On the other hand, we strongly believe that the accessory minerals will effectively hinder major loss of buffer. There is also the possibility to add material with optimally chosen particle sizes to further strengthen the filtering and straining of smectite colloids.