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Mechanical interaction between swelling compacted clay and fractured rock, and the leaching of clay colloids

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Abstract

We consider the interaction between a saturated clay buffer layer and a fractured crystalline rock engineered disturbed zone. Once saturated, the clay extrudes into the available rock fractures, behaving as a compressible non-Newtonian fluid. We discuss the modelling implications of published experiments carried out in Sweden and more recently in Japan. Extrusion is halted either when the advancing clay front reaches a narrow enough aperture along the fracture (relative to the yield stress of the gel-like front), or when enough of the clay mass has extruded so as to reduce the density back in the buffer, and hence the swelling pressure (which is exponentially dependent upon density). In the latter case a relatively small reduction in density may be sufficient. As the clay extrudes, the gel-like front may be a source of clay colloids, being sufficiently hydrated so as to allow clay platelets to escape the matrix and diffuse away. We show that such mass loss is limited as a mechanism for leaching away the emplaced barrier, yet may still be significant in mobilising otherwise highly sorbing radionuclides within the buffer. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Clay barrier; Colloids; Fractured rock; Leaching; Mechanical interaction

1. Introduction

The emplacement of clay buffers surrounding canisters and overpack has long been a feature of disposal concepts adopted for high-level waste and spent fuel disposal programmes within a number of countries. Such barriers are constructed from well-defined and quality-controlled clays (or clay sand mixtures). The principal role of this buffer is to provide a hydrological barrier — the extremely low permeability offered by the clay means that, following a breach of the metal canisters and overpacks, any leaching and migration of radionuclides in solution, or in a colloid phase, is limited to a (retarded) diffusion process. The second role of the clay barrier is in providing a reducing chemical environment, buffering the pH, to immobilise certain radionuclides. The additional performance of the buffer may be mechanical — since the clay is a stressed fluid medium, it may provide a tight, though non-rigid, environment, so that applied stresses or displacements within the surrounding host rock can be distributed around the near field, avoiding breaches due to localised high stresses.

A typical emplacement is shown in cross-section in Fig. 1. In this paper, we assume that the host rock is crystalline containing small (sub-millimetre) fractures at the engineered, exposed, rock faces.

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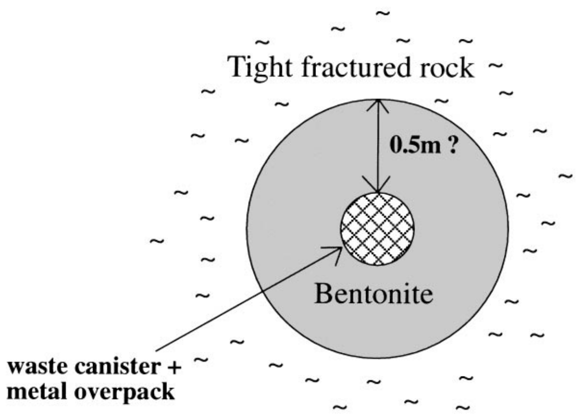

Fig. 1. Horizontal section through a waste emplacement borehole, showing the cylindrical buffer (Fig. 3 shows vertical sections through the emplacement borehole and tunnel).

We also assume that the emplacement boreholes are drilled vertically beneath a tunnel, though almost identical comments are valid for horizontal emplacement strategies.

The clay employed in the buffer depends upon the particular programme, so here we refer to it as bentonite, an (almost) pure sodium montmorillonite clay, though similar considerations would apply to other clays or clay–sand mixtures.

The clay buffer is emplaced in a partially saturated form as bricks or rings. Tolerances of a few centimetres allow the cylindrical overpack and canisters to be emplaced, and a further clay cap is added to backfill and seal the borehole excavation. Subsequently, the tunnel is also backfilled. Following this emplacement the clay saturates, drawing moisture from the surrounding crystalline host rock. This effect has been the subject of various studies based on the extreme capillary pressures (many megapascals) generated by partially saturated clays: resaturation times of between 5 and 15 years are typically estimated for 1 m of clay. In another paper presented at this conference (Grindrod et al., 1999), the interplay between a saturation gradient and a thermal gradient and their impact on the clay chemistry are discussed. Resaturation is also the subject of experimental investigations such as FEBEX (Gens et al., 1998), where the heterogeneity of the saturation behaviour of a buffer emplaced within fractured rock is examined. In this paper we shall focus on the post-saturation, and post-thermal, phase of the clay–rock interaction. The emplaced saturated clay possesses a high swelling pressure, and is not initially in mechanical equilibrium with the confining rock and groundwater. Here we focus on two potential problems that might have an impact upon the performance controlling nuclide release and migration. Specifically, we consider how the bentonite may extrude into the host rock, with a resultant loss in the density and the integrity of the barrier; and how the loss of bentonite in the form of colloids may further reduce the buffer mass, or serve as a source of radioactive colloids.

Over the last year or so we have considered a number of related problems, treating the saturated clay as a compressible fluid, under both Newtonian and non-Newtonian assumptions. From the modelling perspective this generated a useful problem field for analysis and discussion at the European Study Group with Industry (held at the University of Southampton, UK, during March 1998) (Bolchover et al., 1998). There, the published experimental behaviour observed in Swedish, Spanish, and Japanese studies was considered by a group of mathematical modellers. The bulk of this paper is based on the outcome and conclusions of these discussions, seeking to integrate them within the context of waste disposal scenarios, rather than in a more abstract mathematical modelling context, and to identify what data is relevant and required in the future.

In Section 2, we discuss the key clay properties that are important from the modelling perspective, making our best estimates of what information should be made available. In Section 3, we discuss some experiments relevant to this paper. In Section 4 we outline some mathematical modelling, of varying sophistication, that can support the conclusion made at the end of this paper. The modelling is discussed more fully elsewhere (Bolchover et al., 1998).

2. Clay properties

From the modelling perspective, we view the saturated clay as a viscoelastic structure made up of clay platelets (of the order of 50 nm × 100 nm × 200 nm) compressed against each other, with water or air occupying the voids/porespace. During saturation all clay surfaces become hydrated (resulting in a release of free energy as clay–air interfaces are replaced by clay–water interfaces). This not only lubricates the clay platelet contacts, but also results in a further compression of the platelets (to allow the water in, using up the free energy). The result is a swelling pressure of the clay, i.e. a clay-to-clay stress measured as the bulk pressure above any water pressure, which may be driving any fluid flow. This is experimentally measurable and satisfies an empirical exponential law as a function of clay density. As we shall see, in extrusion problems there may be a considerable density (and hence swelling pressure) gradient set up, and it is essential to have knowledge of such a curve down to the low densities, and pressures, corresponding to the confining pressure of the goundwater (say 1 MPa, at approximately 100 m depth). Such low density results in an almost gel-like front existing at clay–water interfaces moving into rock fractures. This has been observed experimentally. A density–swelling pressure relationship from data given by Pusch (1983) is shown in Fig. 2.

If different types of clay or clay–sand mixtures

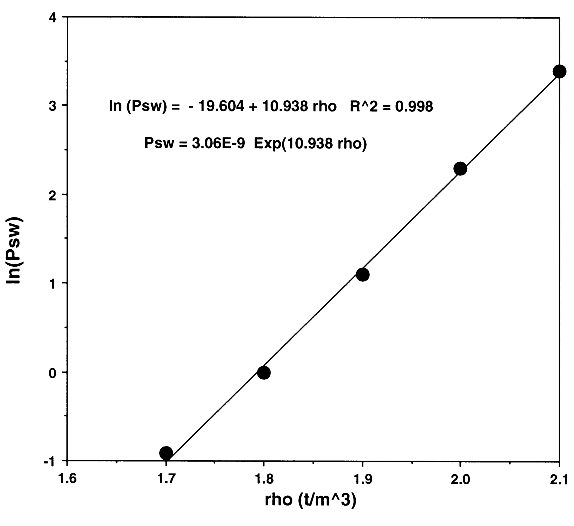

Fig. 2. Swelling pressure as a function of density data from Pusch (1983).

are used within different parts of the repository borehole and tunnel backfill, (e.g. buffer, buffer cap, and tunnel backfill), it is important to have such information for each separate material. At a minimum, assuming that little clay is lost into the rock, one can use such information to calculate volumetric changes for each emplaced material, as they become equilibrated at equal swelling pressures. For example, the cap may extrude into the tunnel, as a function of the different material properties and the distribution of individual waste boreholes along the tunnel length. This process can be driven further by the volumetric expansion of the metal overpack and canister due to corrosion of steel and the formation of magnetite. In Fig. 3 we depict such a change.

Returning to our characterisation of the clay, the next important measurable is the stress–strain behaviour of the clay. In simple calculations the clay may be considered as a Newtonian fluid with a constant viscosity of around 200 MPa s. Such figures have been suggested in the literature (Pusch, 1983), and can be used to predict transient displacements of the emplaced clay of a few centimetres towards the drier centre of the waste borehole during the hydration phase. Such displacements are reversed upon full saturation when the clay returns to a uniform density distribution. There is evidence, however, that the clay behaves in a non-Newtonian fashion, perhaps as a Bingham fluid — where any stresses below a threshold value, called the yield stress, result in no shearing. Clearly it is essential to have information supporting this kind of characterisation at the densities encountered within the repository. Moreover, these kinds of measurement are heavily dependent upon the incoming water chemistry. Hence various extremes in fluid behaviour may have to be postulated under the uncertainties inherent in the emplacement site.

An example of the non-Newtonian nature of clay was provided by Pusch (1983) for dilute clay gels, where the resulting shear stress versus rate of shear relationship (linear for Newtonian fluids) is shown in Fig. 4. The corresponding information for more dense saturated clays should also be obtained.

Turning now to the potential generation of colloids at the clay–water–rock interfaces, the

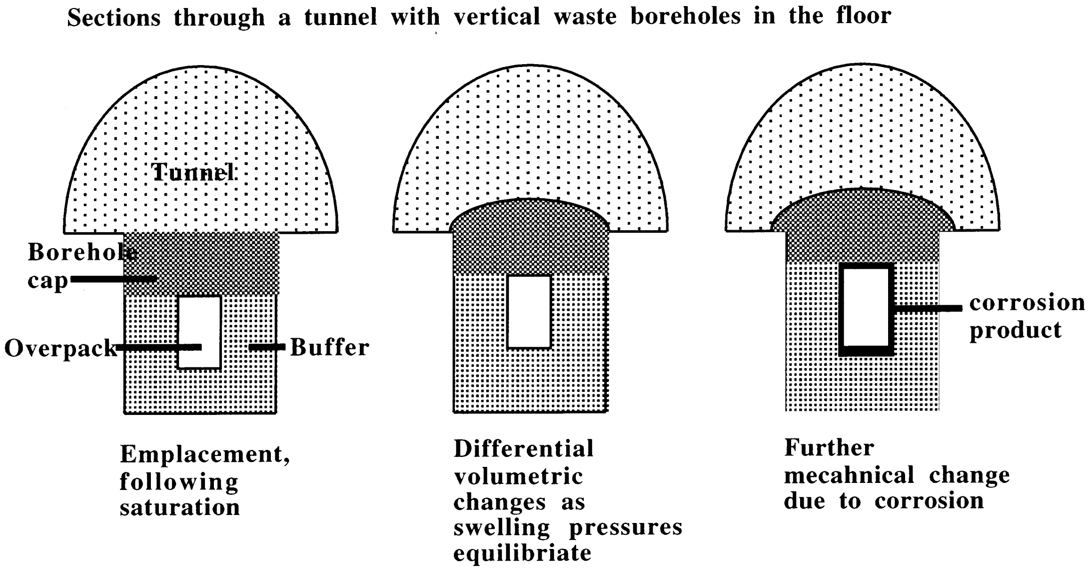

Fig. 3. Possible mechanical/volumetric equilibration of the swelling buffer materials post-saturation.

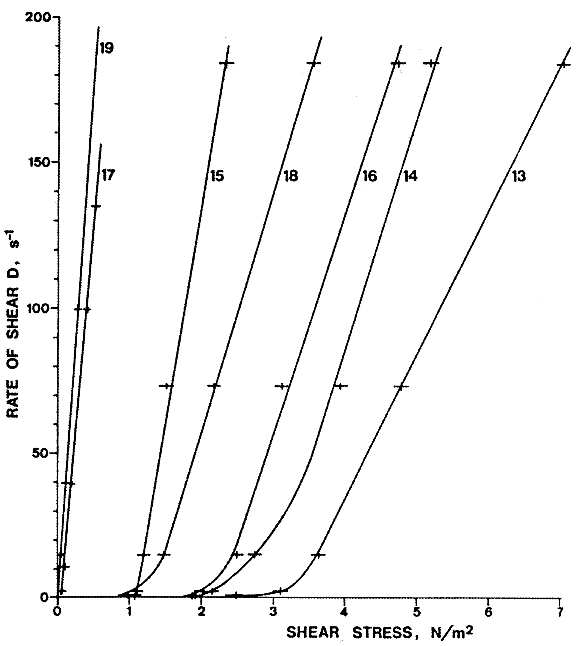

Fig. 4. Relationship between stress and rate of shear for dilute gels. The different numbered curves are for different chemical conditions and different dilutions; from Pusch (1983).

following points are relevant. If the density of the clay becomes very low, the highly hydrated clay platelets become disassociated, and the macroscopic behaviour becomes gel-like. The individual platelets may become free and either be scoured away from the surface by water moving tangentially, or else simply diffuse away as free particles. The size of these platelets puts us firmly in the colloid regime. The diffusivity of such particles, free in water, is given by the Stokes–Einstein relationship, and is two or three orders of magnitude lower than that for solutes. Given the low flow conditions expected to exist around an emplaced nearfield, here we focus on the clay as a diffusive source of colloids. This can impact the buffer performance in two distinct ways. First, the extrusion of clay fingers into fractures may be limited by the aperture variation (see below) or the drag of the clay; thus any erosion of clay at the fingertips may keep alive the extrusion process, resulting in perceptible total mass loss from the buffer. Second, since the clay offers a highly sorbing barrier to some relatively immobile nuclides, any clay colloids leaving the buffer at later (post-failure) times, may be associated with such nuclides. This may provide a limited source, though may well be significant in terms of the overall mobility of some nuclides. Thus the rate at which such colloids are released may be an important extra input to the geosphere calculations, given the standard or reference case assumptions often made of only soluble radionuclides sources.

3. Clay–rock interaction experiments

The experiments summarised in Pusch (1983) and Kanno and Wakamatsu (1991) are our main starting point. In both experiments a clay of known density was emplaced within a cylindrical chamber giving access to water filled 'fractures' (horizontal slots) at a distribution of known apertures. In both sets of experiments there was a similar story — the clay extruded into the slots over a period of around 1000 h. The extruding clay fronts grew at early times (after an initial saturation phase) according to a square-root law in time, but at longer times became self-limiting. In the Kanno and Wakamatsu (1991) experiments, the evolution of the swelling pressure within the core was also monitored and this formed an initial peak after 2 h or so, of 1–4 MPa for 100% pure bentonite, before dropping, during extrusion, down to much lower values. In these experiments there was a single slot with a known aperture in each trial, and the fall in swelling pressure, and hence core density, was the greatest for the widest aperture (falling to below 0.1 MPa). This result is commensurate with the calculation of total mass lost from the core due to extrusion into the slot. Hence the behaviour shown in Fig. 2 is critical in understanding this result. In the Pusch (1983) experiments the cell was surrounded by a number of slots of different sizes. Thus these extruding fingers shared the same cell and hence had a common driving swelling pressure. All extruding fingers became arrested at the same time, providing more evidence that it is not aperture-dependent drag factors, but the reduction in core swelling pressure that is the limiting process. Hence for slots of at least some minimum size, related to the yield stress [see Eq. (2)], the extrusion appears to continue until the density of the core is close to equilibrium with the confining pore water. Examination of Fig. 2 indicates that for an emplaced density of around 1.9 t/m^3 we have around 5 MPa swelling pressure, yet a reduction of total mass by only 5% is sufficient to lower the swelling pressure to 1 MPa.

As part of the EU 'CARESS' colloid experimental and modelling programme (Gardiner, 1997), researchers at Ciemat have examined the extrusion of bentonite into gaps within crushed granite. There the bentonite consolidated rapidly with the granite, and colloids were generated by fluting water parallel to the interface, primarily on the granitic side. Clay colloids have also been observed in extrusion experiments such as those described above.

4. Modelling

In this section, we discuss some of the modelling from the workshop (Bolchover et al., 1998).

We focus on a highly simplified model, which has enough of the problem to indicate the relevance of different concepts available to the modeller. The outward movement of clay reduces the density of the buffer and causes water to flow in from the fractures. Let V_0 be the initial volume of the clay buffer. Suppose all fractures in the rock that are above threshold, and able to admit extruding clay, are broadly similar and are all penetrated to a distance $s(t)$ by the clay front. Then at time t the volume occupied by clay is

$$V(t) = V_0 + v[s(t)],$$

where for simplicity we take

$$v(s) = As,$$

and A is the approximate total area of the apertures of all suitable exposed 'one-dimensional' radial fractures (A must be a fraction of the exposed porespace of the engineered disturbed zone).

The mass at time t is given by the initial clay mass plus the extra water drawn in. We have

$$M(t) = M_0 + v[s(t)]\rho_{\mathrm{wat}},$$

where $\rho_{\mathrm{wat}} = 1$ t/m^3. Assuming that the clay density equilibrates rapidly relative to the motion of interfaces, the density of the wet clay is simply

$$\rho(t)=M(t)/V(t).$$

Assuming for the moment a constant clay viscosity μ and Poisseuille-type flow of bentonite in the fractures, the time dependence of the extrusion is governed by the swelling pressure $P_{sw}[\rho(t)]$, from Fig. 2, via

$$\frac{ds}{dt}=\frac{d^2}{12\mu}\frac{P_{sw}[\rho(t)]}{s(t)}. \quad (1)$$

Here d is the aperture of the exposed slot fractures. Whether the extrusion process arrests or not within this model thus far depends upon the gradient term, and thus $P_{sw}[\rho(t)]$ becoming zero at some low density, when the resulting clay gel becomes so dilute that individual platelets are not fully connected up and can support no stress. In the experiments though, the gel layer was observed to be confined to the tip of the extrusions, indicating that the assumption of density equilibration was invalid there, at least on the time scales of the experiment. However, the mechanism for much larger reductions in the density and swelling pressure was certainly valid, since the core swelling pressure was observed to decrease by Kanno and Wakamatsu (1991). Although the relation in Fig. 2 is as yet unknown at the extreme of low density, and hence the above model is uncertain, for the full range of the pressures seen within the experiment this model is remarkably good at predicting the extent of extrusion, suggesting though that the fronts may not be entirely arrested on extremely long time scales (i.e. many more thousands of hours).

In practice, the assumption of a uniform viscosity may also be misleading, given its potential non-Newtonian behaviour. However, this affects the rate of extrusion rather than the extent. More important is the possibility that the clay has a finite positive yield stress τ_c. In such cases, a Bingham fluid may only propagate along cracks for which

$$d\frac{P_{sw}[\rho(t)]}{s(t)}>2\tau_c. \quad (2)$$

This new stopping criterion is clearly useful in making scoping calculations: $s(t)$ grows according to Eq. (1), whilst $P_{sw}[\rho(t)]$ shows a corresponding decrease, until Eq. (2) fails, at which point the front(s) stop. Furthermore, this model could easily be generalised to allow for propagation down a continuum, or bundle, of flow paths with a given distribution of apertures, all coupled through the clay density and swelling pressure term behind both the advancing and the previously arrested fronts. Then the fronts within cracks with different apertures would arrest at different extents, the smallest first. Similar types of calculation are routinely made for gas displacing water in tight media such as clay. This avenue or modelling, using Eq. (2) for a coupled distribution of potential flow paths, looks very promising.

Variability of the aperture along the pathway is also important given Eq. (2) and this could also be incorporated in a stochastic modelling approach. In particular, the extrusion along any fracture or crack may eventually encounter a neck narrow enough to arrest further motion. Hence uniform slot fractures may be extremely conservative.

Now, comparing this model with the experiments of Kanno and Wakamatsu (1991). Assuming a guestimated yield stress $\tau_c=500$ Pa (based on that of other reference materials), and employing the cylindrical experimental geometry explicitly, from Eq. (2) we would obtain an extension of 11 mm for a crack of 1.5 mm, compared with 20 mm observed in the experiment. Obviously, experimental measurements of τ_c are an extremely important requirement at relatively high clay densities, rather than for the gels shown in Fig. 4. If the clay, which is possibly entering a fracture or moving along a narrowing fracture, is dense enough, with a high enough yield stress, then it will not flow.

Further modelling by Bolchover et al. (1998) employed a lubrication theory approximation, taking a closer look at the dynamics of extrusion and the gradients in density set up between the interface and the core. This kind of modelling (nonlinear density-dependent diffusion equations) is becoming fairly standard, though it has not to date, to our knowledge, been widely employed in these kinds of problem.

Next we turn to the possible loss of clay as a colloidal phase at the extruded gel-tip. There we imagine that there is a small layer of dilute gel where the structure of the platelets has become partly disconnected. Clay platelets are assumed to diffuse away. The diffusion rate D should be around 5×10^{-5} m^2/year (from the Stokes–Einstein relation). If we suppose that the buffer lies in a cylindrical hole of known radius r (approximately 1 m) and height h, and that 10% of the exposed rock consists of fracture porespace into which colloids may escape, then the possible mass lost after time T can be calculated, assuming free colloid material is available at the clay interface at a dilute density of $c_0=0.01$ t/m^3. We have:

$$\text{lost mass (tonnes)} = 2c_0(DT/\pi)^{1/2}0.10(2\pi rh)$$

$$\text{emplaced mass (tonnes)} \approx 1.8(\pi r^2 h).$$

The lost fraction is extremely small, even after 100 000 years, so this mechanism is not significant compared with the extrusion process. However, as noted above, this kind of calculation may be important in scoping the effect of a (pseudo) colloid source term for otherwise highly sorbing, and hence immobile radionuclides.

5. Conclusions

It is recommended that, besides an overall plan of the geometry at emplacement, for each backfill/buffer material used within the emplaced barriers the following information should be made available:

- the range of the proposed emplacement densities;
- the density at which the saturated medium becomes fragmented, and is so gel-like as to support zero stress, yet may act as a source of colloids;
- the density versus swelling pressure curve (as in Fig. 2), for fully saturated medium, from low densities (where the gel/matrix fragments) up to a factor of say 50% above the emplaced density;
- the yield stress (if nonzero) as a function of density and, if possible, the pore water chemistry;
- the non-Newtonian behaviour of the saturated medium at relevant densities (shear thinning).

In this paper we have set the experimental data and the modelling considerations within the context of calculations likely to be useful within the assessment of nearfield performance scenarios. Given the availability of both scoping models (such as those discussed here) and full multi-dimensional simulation codes for mechanical deformation problems, it is certainly past the time when considerations such as those presented here should have become an integral part of any assessment of barrier performance.

Moreover, we feel strongly that by making such calculations available within the disposal programmes and the literature, it will be easier to understand how engineering controls and options (such as the choice of emplacement barrier geometry, the selection of different materials for their mechanical properties, the possible pre-treatment of exposed rock cracks, the thickness of corrosive layers, emplacement strategy, etc.) could be cost effective in limiting the possibilities for highly undesirable barrier performance and the consequent high releases of radioelements to the geosphere.

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