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USE OF SWELLING CLAYS TO REDUCE PERMEABILITY AND ITS POTENTIAL APPLICATION TO NUCLEAR WASTE REPOSITORY SEALING

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Abstract. The injection of swelling-clay slurries into joints or faults at a deep-burial nuclear waste disposal site may result in significant permeability reductions for the effective containment of radioactive wastes. In an experiment conducted to illustrate the permeability change accompanying clay swelling, a coarse stone with interconnected pore spaces was injected with a clay-electrolyte slurry, modelling the pressure-grouting of a fractured repository rock. Subsequently, solutions with lower electrolyte concentrations were driven through the clay-filled stone, corresponding to migration of lower salinity ground-waters through the clay-grouted fracture. The initial injection procedure reduced the permeability of the stone from 1-10 darcies to 700 nanodarcies; the changes in solution composition decreased permeability by more than 2 additional orders of magnitude to 3 nanodarcies. For application at a nuclear waste repository, the electrolyte concentration of the injected clay slurry should be made higher than that of the ground-water in the host rock. Subsequent interaction of the ground-water with the clays would initiate swelling and create the additional, post-injection permeability reductions that may be important in preventing the escape of buried radioactive wastes. The measured permeability of the clay filling is considerably lower than that of cement tested for borehole plugging. Clays also have the advantage over cement and chemical grouts in that they are geologically stable at relatively low temperatures and have a high capacity for radionuclide adsorption.

Introduction

One of the proposed methods of radioactive waste disposal is deep burial in suitable rock formations. Preferred sites are in areas of low tectonic activity situated away from faults. Nevertheless, even the most favored rock units for a repository may contain numerous joints and fractures that could serve as flow paths for radionuclide-bearing fluids. Rock types such as basalt and welded tuff, which otherwise have favorable characteristics for underground repositories, have a major drawback in often being highly fissured.

One way to reduce the permeability of joint/fracture systems near a waste repository site is to fill them by means of slurry injection, using squeeze-grouting techniques [Suman and Ellis, 1977]. However, the cement grouts used in the oil industry [Suman and Ellis, 1977] and by foundation engineers [Bowen, 1975] may not be appropriate for the long-term sealing required at a nuclear waste disposal site. Low-temperature cements solidify to semi-amorphous, metastable phases that gradually recrystallize to thermodynamically stable assemblages. A volume decrease accompanies such reactions, which increases the porosity of the cement and can also lead to the development of new fractures [Roy et al., 1976]. Injection of heated cement slurries creates more favorable reaction kinetics for crystallization of the stable assemblage, thus avoiding the intermediate metastable form. Research is being conducted in this area [Roy et al., 1976, 1979], but as yet the stability and durability of such cements over time and the effects of interaction with the repository rock are not known. A wide variety of chemical grouts have been developed in recent years [Mitchell, 1970; Bowen, 1975]; their use has been limited because of their very high costs relative to cements and clays. As with the heated cements, the long-term stabilities of these chemical grouts are unknown, making it difficult to assess their worth for nuclear waste containment. There is also the problem of compatibility of a chemical grout with the repository rock.

Clay slurries may provide a better alternative to cement and chemicals for the pressure grouting process. In foundation engineering work, clays have frequently been employed either as an additive or an alternative to cement grouts [Kravetz, 1958; Bowen, 1975]; clays are often used where a permeability reduction rather than an increase in foundation strength is the principal goal of the grouting procedure and where the crack or opening size is too small for the coarser cement particles to penetrate. Clay minerals are known from the geologic record to have long-term stabilities at low temperatures and pressures. Natural clays could be chosen for slurry injection that are compatible with the chemistry of the host rock, thus avoiding the possibility of unfavorable chemical reactions. Suitably peptized clay slurries can tolerate relatively high salt concentrations without flocculation or gel formation [van Olphen, 1977]. Because of this, the swelling properties of the clays can be used to cause significantly greater permeability reductions in fractured rock than attained in normal grouting procedures, which may be very important when applied to the problem of nuclear waste isolation.

The chemical basis for producing such added permeability reductions is as follows. The plate surface of a clay particle has an overall negative charge. In a clay suspension, this charge is balanced by the accumulation of positive ions such as Ca^{+2} and Na^{+} in the fluid surrounding the particle [van Olphen, 1977]. This arrangement of surface charge and diffuse

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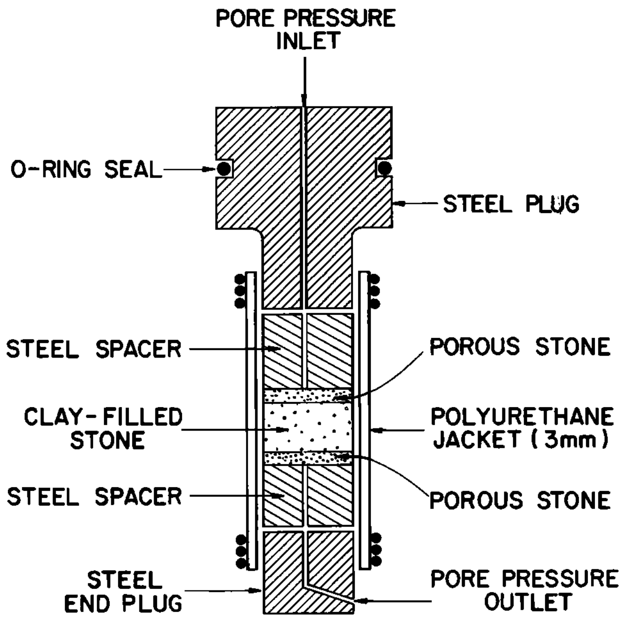

Fig. 1. Experimental sample assembly.

ion cloud of opposite charge is called an electric double layer [van Olphen, 1977]. The fluid and ions in the outer charged layer are loosely adhered to the clay surface and greatly enlarge the effective size of individual clay platelets.

The width of the double layer in an aqueous fluid decreases with increasing electrolyte concentration, owing to a compression of the cloud of ions toward the surface of the clay particle. Thus, the size of the double layer and therefore the degree of swelling or shrinking of the clays can be manipulated by changing the electrolyte concentration. As a result, the permeability of a compacted or sedimented clay or clay-bearing soil will vary with the salt content of the fluids flowing through it [Mesri and Olson, 1971; Hardcastle and Mitchell, 1974].

In the same way, the phenomenon of clay swelling can be used to reduce permeability beyond its initial level on injecting clay into fractures at a nuclear waste disposal site. In practice, a clay slurry would be used that has an electrolyte concentration higher than that of the ground-water of the repository rock. The decrease in the effective size of the clay platelets would allow closer packing of the clays in the fractures than could otherwise be achieved. Then, following injection, percolation of the lower-salinity ground-water into the clay seam would cause swelling and further reduce permeability. The permeability changes that may be expected as a result of clay injection and subsequent clay swelling are illustrated in the experiment described below.

Procedure

A clay-salt slurry was injected into a porous stone and changes in permeability were measured as solutions of varying salinity were passed through the clay-filled stone. The system of interconnected holes in the porous stone serves as a laboratory approximation of an irregularly fractured rock. Such a correlation will not hold in detail; however, the principal object of this experiment is to demonstrate the effects of changing solution chemistry on the permeability of the clay filling. For the experiment, montmorillonite from Chambers, Arizona, was sodium-saturated by repeated washings in 1.0 N NaCl solution. The treated clay was then rinsed in distilled water and dried. A cylinder of synthetic porous stone 2.54 cm in diameter and 5.8 mm in thickness was cut from a coarse grinding wheel. The stone was filled by repeatedly forcing a suspension of the Na-montmorillonite in 0.6 N NaCl solution through it using a hand press, in a simulation of normal pressure-grouting injection practices. Stone cylinders filled to test the injection procedure were cross-sectioned for examination, which showed that when all visible pore spaces contained clay, the interiors of the cylinders also were clay-filled.

The experimental assembly for the permeability measurements is shown in Figure 1. The clay-filled stone was sandwiched between 1.2 mm-thick wafers of a much finer grained grinding wheel, to allow free fluid flow but to prevent the possible loss of clays to the pore pressure lines due to particle migration. In effect, the finer grained wafers served as the wall rocks adjoining a clay-packed, breccia-filled fault or joint. The sample was held at 300 bars confining pressure; fluids flowed through the sample in the direction indicated in Figure 1. The pore pressure was 10 bars on the inlet and atmospheric pressure on the outlet side of the sample. Initial permeability measurements of the clay-filled cylinder were made with the 0.6 N NaCl solution used for the injected clay slurry. The solution composition was then changed sequentially to 0.1 N NaCl, 0.01 N NaCl and, finally, back to 0.6 N NaCl.

The times at which a new solution was introduced and the corresponding effects on the permeability of the sample as the pore fluid changed from the old to the new composition are shown in Figure 2. Prior to clay injection, the measured permeability of the porous stone was high, in the range of 1-10 darcies. Filling the

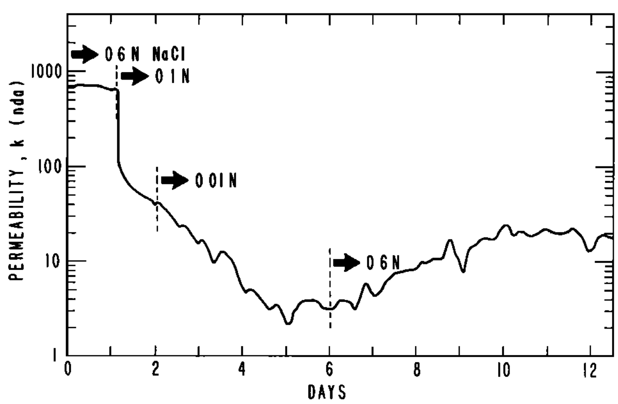

Fig. 2. Changes in permeability of porous stone following clay injection and permeation with electrolyte solutions causing alternate clay swelling and clay shrinking. Permeability of the porous stone prior to clay injection was 1-10 darcies.

interconnected spaces of the stone with clay decreased permeability to approximately 700 nanodarcies. With the first salinity change to 0.1 N NaCl, the permeability was reduced by slightly more than an order of magnitude to 40 nanodarcies; an additional order of magnitude decrease accompanied the change to 0.01 N NaCl solution. At the end of six days, the measured permeability was approximately 3 nanodarcies, which is near the lower measuring capabilities of the experimental apparatus. Thus, although the major permeability reduction was made by physically filling the stone with clays, at least a 200-fold additional decrease was attained by changing the fluid composition. For comparison, the reported permeabilities of sample cement borehole plugs range from 2.1 to 12 microdarcies [Roy et al., 1976, p. 58].

The return to a 0.6 N NaCl solution was made to determine whether or not the permeability decreases were completely reversible. As shown in Figure 2, after 6 days' flow of the more saline solution the permeability increased to a stable value of about 20 nanodarcies, which was considerably less than that of the pre-swelling value for 0.6 N NaCl. Hardcastle and Mitchell (1974) also found that the initial permeability values were not restored to clay-silt soils subjected to a sequence of NaCl solutions causing first swelling and then shrinking of the clays. The lack of complete reversibility may be due, in part, to the rearrangement of clay particles in the stone to a configuration with lower permeability [Hardcastle and Mitchell, 1974]. Interaction of the swelled clay particles may also inhibit shrinking. Mesri and Olson (1971) showed that the shrinking-swelling characteristics of consolidated clays were less than predicted from classical double layer theory, because of irreversible domain formation among the clays.

Discussion

Most of the proposed repository host rocks have low intact permeabilities, so that fluid transport will be concentrated along fractures and joints. Therefore, it is important that these openings be filled with the least permeable, most stable materials available. The very low measured permeability of the swelled-clay filling favors its use as a fracture sealant to reduce the possibility of fluid migration both to and from the canisters, although it should be noted that site conditions and operational procedures may lead to somewhat different amounts of permeability reduction than measured in the experiment. In the possible case of a canister breach, the joint-filling clays also have high radionuclide adsorption coefficients [Komarneni and Roy, 1980]. Aside from the fractured zones, whose locations can be pinpointed using an ultrasonic borehole televiewer [Zoback and Anderson, 1982], the drilled holes could be filled with other materials, which would themselves be protected from fluid-induced alteration by the fracture-filling impermeable clays.

The drilled openings at a disposal site will include not only the repository itself, but also other exploratory holes and pre-existing boreholes from oil and gas exploration, mining, water wells, or other uses. Such peripheral sites will be less subject to heating around the canisters, and one of the potential problems of clay sealants --- the dewatering and breakdown of the clays at elevated temperatures [Weaver, 1979] --- would be minimized. According to Pusch (1979), clays held at temperatures below approximately 100°C should remain stable over the active life of the repository. Some poorly crystallized illites show pronounced Cs-adsorption capacities that are either unaffected or only slightly decreased by short-term hydrothermal heating to 400°C [Komarneni and Roy, 1980]. Clays such as these might potentially be useful in filling joints intersecting the actual repository walls. However, additional experiments would need to be conducted to determine the long-term effects of heating on the stability and radionuclide retention of such clays.

In the experiment, 5.5 ml of 0.1 N NaCl and 2.5 ml of 0.01 N NaCl solution were used to produce the observed additional permeability reductions. Hardcastle and Mitchell (1974) estimated that only 2 pore volumes of a low-electrolyte solution are required to achieve the maximum swelling. Because only small amounts of solution were required for the clay swelling process, thorough swelling could be achieved relatively rapidly in a fracture filling even at low rates of ground-water migration. In addition, much of the permeability reduction attending clay swelling was not recovered following the introduction of a solution favoring shrinking of the double layer. In this way the very low permeabilities would be maintained even in the event of a major, unexpected change in ground-water composition, such as that accompanying submergence of the repository site below sea level (seawater salinity is approximately 0.6 N).

To maximize the amount of clay swelling, the most appropriate clay minerals and electrolytes for the specific burial site would be chosen, by means of ground-water and mineralogical studies. Clays with good swelling characteristics that occur as natural alteration minerals in the repository rock would least disturb the chemical conditions at the disposal site. Because of the low volume of injected material, no adverse effects should accompany the addition of small amounts of salt and peptizing agents to the repository rock.

With the addition of peptizers, relatively large concentrations of salt can be added to a clay slurry without causing flocculation. Some drilling muds treated with organic peptizers are unaffected by sea-water salinities [van Olphen, 1977]. However, a limit probably exists to the amount of salt that can be added to a clay slurry without destroying its flow characteristics. Therefore, manipulation of a clay slurry to promote post-injection swelling would not be a viable process for repositories sited in certain rock types, for example brine-bearing marine shales. However, even unswelled clay fillings will lead to lower crack permeabilities than achieved with cements, so that clay-slurry injection could still be of value for such rocks.

Details of the pressure grouting procedure, such as injection pressures and the density and viscosity of the grouting material, will depend on characteristics of the chosen disposal site. Although reduced permeability rather than increased strength is the goal of the proposed injection procedure, some situations may require the addition of some cement to the clay grout to prevent erosion or collapse of the injected clay fillings. Small amounts of cement should not alter appreciably the overall characteristics of the swelling-clay grout [Bowen, 1975]. However, the importance of increased strength must be weighed against the need to maintain flexibility, so that the injected material can withstand ground motion without the creation of new fractures.

Conclusions

The injection of swelling-clay slurries into faults or fractures is a potentially feasible, low-cost process which would create an additional permeability barrier against the escape of radioactive wastes from a disposal site and also help protect the principal borehole-sealing materials. The combined characteristics of the clays, including low permeability, high radionuclide adsorption capacity, longevity, and compatibility with the host rock, favor their use over cement or chemical grouts as fracture sealants. In addition, as shown in the experiment described in this paper, the chemistry of the injected clay slurry can be manipulated with respect to ground-water conditions at the chosen site to create even greater permeability reductions than attained in normal grouting practice. This procedure could remove some of the drawbacks to certain geologic sites proposed for nuclear waste disposal. Additional studies, centering on injection techniques and site-specific requirements, should be made to determine the effectiveness of the injection procedure and its impact on the repository rock.

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