SORPTION OF CESIUM ON COMPACTED BENTONITE

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Abstract—Sorption parameters are important components of models used to predict mass transport through dense or compacted earthen materials. These parameters are, however, generally determined in batch tests with loose, unconsolidated materials. Here we directly measure, using a specially designed cell, the extent of Cs^+ sorption on bentonite compacted to a series of densities ranging from 0.50 to 1.50 Mg/m^3, and compare the results with those obtained from batch tests with loose bentonite. The clay was saturated with a Na-Ca-Cl-dominated solution with an effective ionic strength of 220 mol/m^3. The sorption data were expressed as distribution coefficients, K_d. Over the clay density range examined, K_d values for Cs^+ with compacted clay are about one-half to one-third the value of those with loose clay. The lower sorption on compacted clay is attributed to small and occluded pores that Cs^+ cannot enter; thus it cannot access the entire volume, or all the sorption sites, of compacted clay. The data suggest that reasonable estimates of K_d with compacted clay can be obtained by scaling down the K_d values measured on loose clay by a factor n_a/n, where n_a is the accessible porosity and n the total porosity of compacted clay.

Key Words—Bentonite, Cesium, Sorption.

INTRODUCTION

Sorption parameters are important components of mass transport models used to predict the migration of contaminants through compacted or consolidated earthen materials (Cho *et al* 1993; Yong *et al* 1992; Jury and Ghodrati 1989). In practice, however, these parameters are generally measured on loose unconsolidated materials using either batch or column techniques. To have confidence in mass transport models, it is necessary to ensure that sorption parameters obtained on loose materials do not differ markedly from those on compacted material, or if they do, we must understand how and why they differ.

The literature on the sorption of contaminants on loose or unconsolidated clay and soil is voluminous. To our knowledge, however, Muurinen *et al* (1987) give the only direct determination of the sorption of contaminants on compacted clay. They reported the distribution coefficient, K_d, for Cs^+ was three to four times greater for compacted (clay density = 1.8 Mg/m³) than loose bentonite; the value for Sr^{2+} was the same for compacted and loose bentonite. On the other hand, from indirect measurements, usually involving migration studies, other investigators have suggested that contaminants are sorbed to a lesser extent on compacted than loose clay (Sato *et al* 1992; Miyahara *et al* 1991).

Here we present a technique, using a specially designed cell, to directly measure sorption processes on compacted or consolidated materials. The results of first tests with Cs^+ as a sorbate and compacted bentonite as a sorbent are compared with those obtained from batch tests. Cesium-135 is an important radioisotope in nuclear fuel waste management because of its long half-life of 2.3×10^6 a, and bentonite-based ma-

terials are important barriers in many waste management strategies because of their high sorptivity and low permeability.

MATERIALS AND METHODS

Materials

The Avonlea bentonite is from the Bearpaw Formation of Upper Cretaceous age in southern Saskatchewan, Canada (Oscarson *et al* 1990). It contains approximately 80 wt.% smectite (montmorillonite), 10% illite, 5% quartz, and minor amounts of gypsum, feldspar, and carbonate (Oscarson and Dixon 1989). The clay has a cation-exchange capacity of about 60 cmol$_c$/kg, a specific surface area of 480×10^3 m²/kg, and Na$^+$ is the predominant exchangeable cation. The Avonlea bentonite is a component of the reference buffer material in the disposal concept developed for nuclear fuel waste within the Canadian Nuclear Fuel Waste Management Program (Hancox and Nuttall 1991). The reference buffer material is a 1:1 mix by dry mass of bentonite and sand compacted to a dry density of about 1.7 Mg/m³. With respect to most sorption reactions, clay is the active component of the buffer and sand is essentially inert filler (Gillham *et al* 1984; Sharma and Oscarson 1991). Hence, 100% clay was used in this study.

The composition of the synthetic groundwater (SGW) solution that was used in all experiments is given in Table 1. The solution represents groundwaters found deep in granitic rock in the Canadian Shield (Frape *et al* 1984).

A diagram of the sorption cell is shown in Figure 1. The outer shell is made of Polysulphone® (Union Carbide). The inner stainless steel ring that contains the clay has an inside diameter of 2.2 cm; ring lengths of
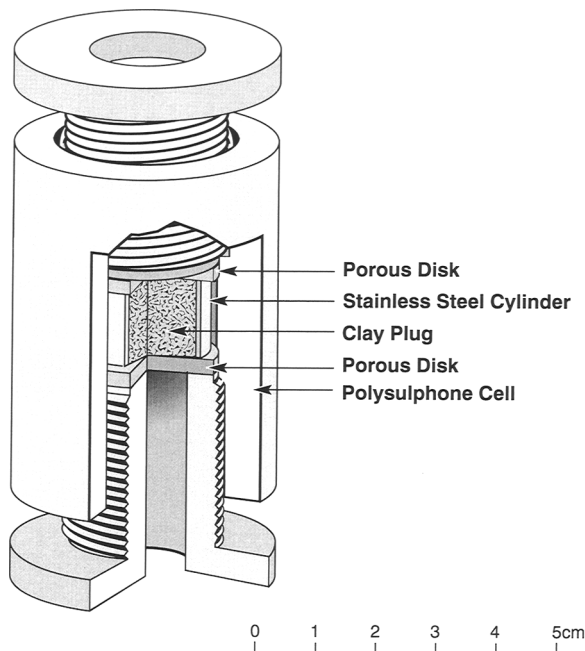
Figure 1.   Diagram of the sorption cell.

Table 1.   Composition of the synthetic groundwater solution.

	(mol/m³)
Na	83
K	0.36
Mg	2.5
Ca	53
HCO₃	0.28
Cl	170
SO₄	11
pH	7.2

solution became constant with time indicating that a steady-state condition had been established. The reaction period ranged from about 350 d when the plug length was 6.35 mm to 600 d at 12.7 mm.

Distribution coefficients, K_d, were calculated from,

$$K_d = \left(\frac{A_o}{A_e} - 1\right)(S - S_a)/\rho_w; \qquad (1)$$

where A_o is the net (total activity minus background activity) initial and A_e the net equilibrium activity of ^{137}Cs in solution (the activities were corrected for the small amount of radioactive decay of ^{137}Cs ($t_{1/2} = 30$ a) that occurred during the experiments); S the solution-to-clay ratio (by mass) at the end of the experiment and S_a the solution-to-clay ratio, or the gravimetric moisture content, of the air-dried clay; and ρ_w the density of the SGW solution.

The use of K_d implies a linear sorption isotherm; this is generally the case at low sorbate concentrations: <1 mmol/m³ or so (Chang *et al* 1993; Meier *et al* 1987; Alberts *et al* 1986). The parameter has been used extensively in modelling the fate of contaminants in aqueous and geological environments (Yong *et al* 1992; Gillham and Cherry 1982). The K_d is, however, merely a means of expressing empirical data and may not contain intrinsic thermodynamic or mechanistic information.

After the sorption experiment, the clay plugs at $\rho \geq$ 0.90 Mg/m³ were extruded from the rings using a specially designed jig (Hume 1993) and sectioned into slices about 2 mm thick. At the lower densities, the clay was too soft to be extruded and sectioned. The longer plugs were used at the higher densities so several slices could be obtained. The slices were dried at 105°C and the amount of ^{137}Cs associated with the clay was measured by gamma spectroscopy using an Ortec high-purity germanium detector (EG & G Ortec, Oak Ridge, Tennessee). From these data, activity profiles for ^{137}Cs within the clay plugs were determined. These profiles provide additional checks on the sorption reaction—a uniform activity of ^{137}Cs throughout the clay plug indicates steady state was reached.

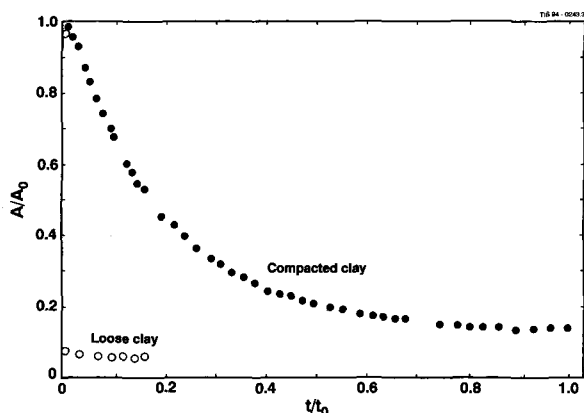*Loose clay.* For comparison with compacted clays, batch experiments were conducted with loose clays at the

6.35 mm and 12.7 mm were used depending on the desired length of the clay plug. When the cell is assembled, the clay plug in the ring is sandwiched between porous Ni discs (3.2 mm thick) and constrained by hollow end pieces. The cell was designed to withstand the swelling pressure generated by bentonite compacted to a density of at least 1.7 Mg/m³.

Sorption experiments

Compacted clay. Air-dried clay was compacted in the stainless steel ring using a hydraulic press to target dry bulk densities, ρ, ranging from 0.50 to 1.50 Mg/m³. The density range encompasses the effective clay dry density of the 1:1 clay/sand buffer material—about 1.2 Mg/m³—in a disposal vault. (Effective clay dry density of a clay/sand mix is the mass of clay divided by the combined volume of clay and voids.) A plug length of 6.35 mm was used for clay at densities of 0.50 and 0.70 Mg/m³, and 12.7 mm for 0.90, 1.25 and 1.50 Mg/m³. The ring containing the compacted clay was placed in the sorption cell and the cell was immersed in 500 cm³ of SGW solution containing 1 μmol/m³ of carrier-free $^{137}Cs^+$ as a sorbate; 2-liter polyethylene containers were used. When the cell is immersed in solution, $^{137}Cs^+$ diffuses through the porous discs of the cell and into the pores of the clay, where some of it is sorbed; hence its activity in the contacting solution decreases. The activity of ^{137}Cs in solution was monitored periodically by liquid scintillation counting (Beckman LS 5801, Beckman Instruments, Inc., Irvine, California). The experiment was continued until the activity of ^{137}Cs in

Figure 2. Relative activity of ^{137}Cs in solution vs normalized time for compacted clay ($\rho = 0.90$ Mg/m³) and loose clay; t_o is the total time of the compacted clay experiment, and in this case is about 600 d.

same initial solution-to-clay ratios and ^{137}Cs activity as used for the compacted clays.

Clay was suspended in SGW solution containing ^{137}Cs in 1-liter polyethylene bottles. The suspensions were gently shaken periodically. The solutions were analyzed for ^{137}Cs as indicated above. When steady state was established, K_d values were calculated from Equation 1.

For both the compacted and loose clay systems, control experiments, conducted identically but without clay, showed that no detectable Cs$^+$ was sorbed on the walls of the polyethylene containers nor any of the components of the sorption cell. All experiments were conducted at least in duplicate at $22 \pm 2°C$. Hereafter, the K_d values determined for the compacted clay are referred to as $K_d(C)$ and those for loose clay, $K_d(L)$.

RESULTS

A typical plot of relative activity versus normalized time for bentonite at $\rho = 0.90$ Mg/m³ and the corresponding loose clay is shown in Figure 2. As expected, it takes much longer for the compacted clay system to reach steady state, due to the slower diffusion of Cs$^+$ through compacted clay. In fact, in the loose clay systems, >90% of Cs$^+$ was sorbed within the first few days. Figure 2 also shows that at steady state less Cs$^+$ is sorbed on compacted clay than on loose clay. This is contrary to the results of Muurinen et al (1987) who reported that more Cs$^+$ is sorbed on compacted bentonite.

Activity profiles for ^{137}Cs in the clay plugs are shown in Figure 3 for $\rho \geq 0.90$ Mg/m³. Although there is some scatter in the data, the amount of ^{137}Cs associated with the clays after the reaction periods was uniform throughout the clay plugs; and it is reasonable to assume a uniform distribution at the lower densities as well. This confirms the systems were at steady state.

Table 2. Distribution coefficients, K_d, for compacted and loose clay.

ρ (Mg/m³)	S/C(C)[1] (m³/Mg)	K_d(C)[2] (m³/Mg)	S/C(L)[1] (m³/Mg)	K_d(L)[2] (m³/Mg)	K_d(C)/ K_d(L)[3]	n_a[4]
0.50	385	1040 920	405	1880 1800	0.53	0.44
0.70	274	790 810	289	1710 1690	0.47	0.35
0.90	82.2	520 520 520	89.1	1380 1410 1380	0.37	0.25
1.25	59.5	490 490 440	60.0	1280 1260 1320	0.37	0.20
1.50	49.5	450 420 430	53.7	1190 1200 1220	0.36	0.16

[1] Solution-to-clay ratio at time steady-state was established; C = compacted clay and L = loose clay.
[2] K_d(C) = K_d for compacted clay and K_d(L) = K_d for loose clay.
[3] Calculated from the mean K_d values for a given system.
[4] Accessible porosity calculated from Eq. 2.

All $K_d(C)$ and $K_d(L)$ values are given in Table 2. The two or three values shown for a given system are replicates. In all cases, $K_d(C) < K_d(L)$. Both $K_d(C)$ and $K_d(L)$ decrease with decreasing solution-to-clay ratio. This phenomenon has been observed for many different sorbate/sorbent systems (Meier et al 1987; Alberts et al 1986; O'Connor and Connolly 1980), but has yet to be satisfactorily explained. Given that the solution-to-clay ratios used in batch tests are generally higher than those in the environment of interest, batch tests will tend to overestimate K_d values. Therefore, experiments should be conducted under conditions that are as close as possible to those in situ. The final solution-to-clay ratios of the compacted clay systems are somewhat lower than those for the corresponding loose clay—the experiments with compacted clay took longer to complete and more solution was removed for analysis. These small differences in solution-to-clay ratios, however, would not contribute significantly to the large differences between $K_d(C)$ and $K_d(L)$ values.

DISCUSSION

The lower $K_d(C)$ than $K_d(L)$ values suggest the accessible surface area, and hence number of sorption sites, available to Cs$^+$ is less in compacted clay. This is likely due to small and occluded pores in compacted clay that Cs$^+$ cannot enter; thus, it does not have access to the entire clay volume or all sorption sites. On the other hand, in the loose clay system the bentonite (montmorillonite) layers suspended in the SGW would be present as particles composed of either hydrated single-unit-layers or as quasicrystals—several hydrated unit layers stacked along the crystallographic c axis
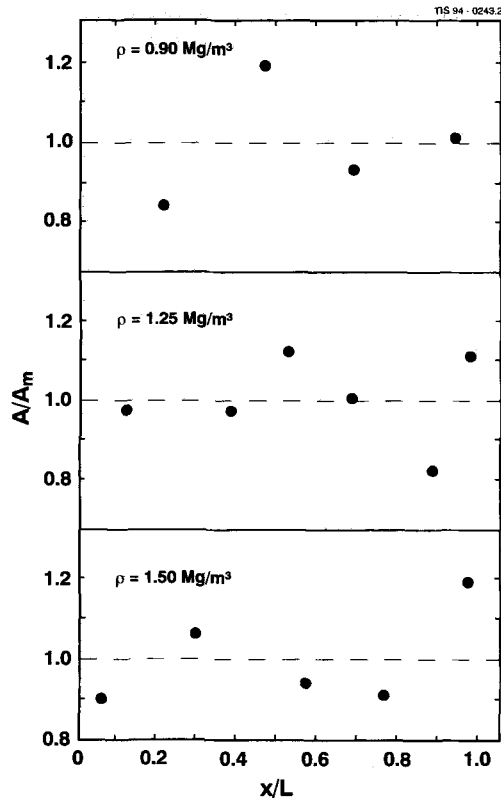
Figure 3. Activity profiles for ^{137}Cs within the clay plugs; A is the measured activity of ^{137}Cs in the clay slices and A_m the arithmetic mean of the activities in all slices for a given clay density; L is the length of the clay plugs and x is the midpoint of a given slice within the plugs; one end of the plugs was arbitrarily taken as zero.

Figure 4. Accessible porosity, n_a, of compacted bentonite versus reciprocal of clay dry density, $1/\rho$.

(Sposito 1984); in either case, several molecular layers of water surround each layer, and Cs^+ should have access to all, or nearly all, of the surfaces (and sorption sites) of the clay.

If the lower $K_d(C)$ values are largely due to small and occluded pores, a measure of the accessible porosity, n_a, of compacted clay can be obtained from,

$$\frac{n_a}{n} = \frac{K_d(C)}{K_d(L)}, \qquad (2)$$

where n is the total porosity, which is readily determined from the dry bulk density and the particle density (taken to be 2.7 Mg/m^3) of the clay. The relationship expressed in Equation 2 assumes that $K_d(L)$ is a measure of the total porosity of compacted clay; i.e., if Cs^+ could access the total porosity, $K_d(C)$ would equal $K_d(L)$.

The n_a values, calculated from Equation 2, are given in Table 2. And a plot of n_a versus $1/\rho$ is shown in Figure 4; the equation of the best-fit line (correlation coefficient, r = 0.993) is,
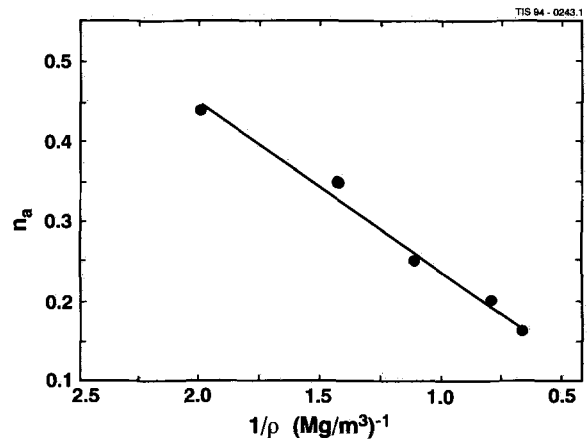
$$n_a = 0.212/\rho + 0.025. \qquad (3)$$

This equation can be used to estimate n_a for a given clay density over the range examined.

The relationship given in Equation 3 is supported by indirect determinations of $K_d(C)$ (a measure of n_a) for sorbates on compacted bentonite from migration studies: Oscarson and Hume (1994) observed that $K_d(C)$ for carbonate decreased with increasing clay density, and Miyahara *et al* (1991) reported the same phenomenon for Cs^+. (In these studies, apparent diffusion coefficients in compacted bentonite were measured and then $K_d(C)$ values were back calculated from transport equations similar to Equation 4.) In fact, from the data of Miyahara *et al* (1991) it can be shown that $K_d(C)$ for Cs^+ on compacted bentonite is a linear function of $1/\rho$ over the clay density range of 0.2 to 1.6 Mg/m^3 (Figure 5). Unfortunately, n_a values cannot be obtained
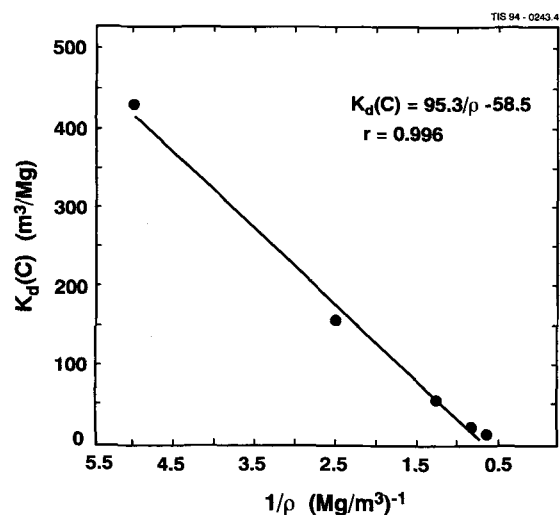

Figure 5. $K_d(C)$ for Cs^+ with bentonite versus reciprocal of clay dry density, $1/\rho$; adapted from Miyahara *et al* (1991).

from their data because appropriate $K_d(L)$ values are not available.

If $K_d(L)$ values are used in transport models to predict migration rates through compacted materials, they should first be determined at as low a solution-to-clay ratio as possible, and then scaled down by the factor n_a/n to obtain $K_d(C)$ (Eq. 2). The solution composition used in the experiments should also be similar to that in the environment of interest. The data of Cho et al (1993) can be used as an illustration. They measured a mean apparent diffusion coefficient, D_a, of 0.54 $\mu m^2/s$ for Cs^+ in bentonite at $\rho \sim 1.35$ Mg/m³ and saturated with the SGW solution, and a $K_d(L)$ value of 100 m³/Mg at a solution-to-clay ratio of 2 m³/Mg (this is about the lowest practicable ratio with this clay and solution). They also calculated a D_a value from the following model,

$$D_a = D_o\tau/R. \qquad (4)$$

The retardation factor, R, is given by,

$$R = 1 + \rho K_d/n_e, \qquad (5)$$

where D_o is the diffusion coefficient in pure bulk water (taken to be 2000 $\mu m^2/s$ for Cs^+), τ the tortuosity factor (0.1), and n_e the effective porosity of the clay (0.1) or the porosity that contributes to mass transport. [See Cho et al (1993) for a discussion of these parameters and their values in compacted clay.] Using a $K_d(L)$ value of 100 m³/Mg, D_a for Cs^+, calculated from Equation 4, is 0.15 $\mu m^2/s$, which is lower than the measured value of 0.54 $\mu m^2/s$. The $K_d(C)$ value is 36 m³/Mg for this system, calculated from $K_d(L)$, n, and n_a [0.18 at $\rho = 1.35$ Mg/m³ (Equation 3)]. Using $K_d(C)$ in Equation 4, gives a D_a value of 0.41 $\mu m^2/s$—much closer to the measured D_a value. If the $K_d(L)$ value could be measured at a lower solution-to-clay ratio, it and the corresponding $K_d(C)$ value would have been lower; this in turn means the calculated D_a value would be greater and closer still to the measured value.

Given that the effective hydrated radii, r_H, of most inorganic sorbates are about the same—ranging from 0.33 to 0.45 nm (Nightingale 1959)—n_a for other sorbates in compacted bentonite should be similar to that found for Cs^+ ($r_H = 0.33$ nm) other factors being equal. The salinity of the pore solution, for example, could influence n_a. Salinity affects the thickness of the electrical double layer of charged surfaces: increasing salinity decreases the double layer thickness, and, hence, a corresponding increase in n_a might be expected depending on the sorbate charge. The magnitude of the change in n_a with salinity, however, can only be determined experimentally.

Mass transport through porous media depends on both the transport or effective porosity, n_e, and the storage porosity, n_s; n_e is the fraction of the total porosity that contributes to mass transport, and n_s is that fraction that is accessible to a solute, but does not

contribute to its transport, and could include, for example, dead-end pores. The storage porosity is often invoked as a retarding mechanism for mass transport through porous media (Oscarson 1994). The porosity we have termed accessible porosity would be equal to the sum of n_e and n_s. From through-diffusion experiments with I^- and Cs^+ in Avonlea bentonite at $\rho \sim 1.35$ Mg/m³, n_e has been estimated to be about 0.1 — or 20% of n (Oscarson et al 1992; Cho et al 1993). This is approximately half the n_a value in this clay at a similar density (Table 2), and, thus, n_s would also be about 0.1 at this clay density.

In addition to inaccessible sorption sites on compacted bentonite, a difference in the composition of the solution in contact with the compacted and loose clays could account for the observed differences in K_d values for the two systems. If, for example, the K^+ concentration was lower for some reason in the solution in contact with the loose clay, $K_d(L)$ for Cs^+ would likely be greater than $K_d(C)$ other factors being equal (Oscarson et al 1987). However, given that the same clay, solution and solution-to-clay ratio were used for both the compacted and loose clay systems, the composition of the solution in contact with the clays should be the same. In similar experiments with the Avonlea bentonite and the same sorption cell used here, Ryan and King (1994) reported that the solutions in contact with loose and compacted bentonite were virtually the same after an 18-month reaction period. The only way K^+ (or other species that compete with Cs^+ for sorption sites on the clay) could influence the determination of n_a as obtained from the ratio $K_d(C)/K_d(L)$ (Equation 2), is if Cs^+ enters pores in compacted clay that K^+ cannot. This is unlikely since the r_H values of the two ions (and most other common inorganic species) are similar (Nightingale 1959). We conclude that the difference in K_d values for compacted and loose clay is due largely to inaccessible sorption sites on compacted clay and not to differences in solution composition.

SUMMARY AND CONCLUSIONS

Over a clay density range of 0.50 to 1.50 Mg/m³ $K_d(C)$ values for Cs^+ are about one-half to one-third $K_d(L)$ values. The lower sorption on compacted clay is attributed to small and occluded pores that Cs^+ cannot enter; thus it does not have access to the entire volume, or all the sorption sites, of the clay. The data suggest that reasonable estimates of $K_d(C)$ can be obtained by: (1) measuring $K_d(L)$ in batch tests at the lowest possible solution-to-clay ratio, and (2) then scaling down the measured $K_d(L)$ value by a factor, n_a/n, where n_a is the accessible porosity and n the total porosity of compacted bentonite. Similar experiments with other sorbates and clays need to be conducted to establish the general applicability of the results presented here.

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