On the Relationship Between Microstructure and Electrical and Hydraulic Properties of Sand-Clay Mixtures

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Abstract. A series of laboratory experiments, including measurements of electrical properties, permeability, and porosity, were performed on saturated sand-clay mixtures. Different mixtures and packing geometries of quartz sand and 0 to 10% Na-montmorillonite clay were investigated using solutions of $CaCl_2$ and deionized water. Two main regions of electrical conduction exist: a region dominated by surface conduction and a region where the ionic strength of the saturating fluid controlled conduction. For low fluid conductivities, the sample geometry was found to greatly affect the magnitude of the surface conductance. The influence of the microstructural properties on the electrical properties was quantified by estimating formation factors, Λ-parameters, and surface conductances. The surface conductances estimated using the theory of *Johnson et al.* [1986] agreed well with measured values. We suggest that high and low bounds on the expected surface and bulk conductances in a natural system can be derived from the measurements on these artificial geometries.

Introduction

Electrical measurements are a useful, nondestructive tool for characterizing porous rocks and soils. Conduction of electricity through porous media occurs primarily by the movement of ions through the bulk-saturating electrolyte, and by movement of adsorbed ions along the surfaces of pores and cracks. Thus, the conductivity of a porous medium is related to microstructural properties such as porosity ϕ, pore geometry, and surface morphology of the mineral grains lining the pores as well as to the dielectric properties of the mineral grains and pore fluid. Electrical conduction is also a function of the prevailing fluid saturation. For brine-saturated porous materials, *Archie* [1942] proposed the following relationship between bulk and fluid conductivities: $\sigma_{\text{bulk}} = \sigma_{\text{fluid}} a\phi^m$, where σ_{bulk} and σ_{fluid} are the electrical conductivities of the porous medium and the saturating fluid, and a and m are constants for a certain type of rock. The ratio $\sigma_{\text{fluid}}/\sigma_{\text{bulk}}$ is the formation factor F. Archie's equation assumes that the contribution of surface conduction to bulk conduction is negligible. If that is not the case, it has been found in theoretical [*Johnson et al.*, 1986] and experimental work [*Waxman and Smits*, 1968; *Sen et al.*, 1988] that Archie's law has to be modified to include a surface-conduction term. *Johnson et al.* [1986] defined a parameter Λ, which is a weighted volume-to-surface-area ratio (a measure of the dynamically interconnected pore size)

defined as

$$\frac{\Lambda}{2} = \frac{\int |E(r)|^2 \, dV_p}{\int |E(r)|^2 \, dS} \qquad (1)$$

$E(r)$ is the electric potential field at point r, V_p is the pore volume, and S is the surface area (i.e., the pore-solid interface). Λ has units of length and it is characteristic of the geometry of the porous medium. The weighting by the electric field $E(r)$ eliminates contributions of isolated regions, which do not participate in transport. This relation is valid for materials where the (insulating) grains are coated with appreciable amounts of clay minerals[*Johnson et al.*, 1986]. When dry clays are saturated with brines, the counter ions, which usually balance charged impurities by bonding to their external surfaces, hydrate and become mobile within a layer. Surface conduction due to these counter ions then acts in parallel with the ionic conduction through the brine-filled pores. For relatively high-salinity pore fluids, a linear relationship was derived[*Johnson et al.*, 1986]:

$$\sigma_{\text{bulk}} = \frac{1}{F}\left(\sigma_{\text{fluid}} + \frac{2\Sigma_s}{\Lambda}\right) \qquad (2)$$

where Σ_s is the surface conductance. When σ_{fluid} and σ_{bulk} are plotted on a linear scale, the slope equals the reciprocal of the formation factor F, and the intercept is $b = 2\Sigma_s/F\Lambda$ (Table 1). As mentioned previously, several investigators have demonstrated the presence of the intercept for clay-bearing rocks. However, the theory of *Johnson et al.* [1986] has not yet been applied to unconsolidated materials. Thus, the objective of the present study was to apply the theory of *Johnson et al.* [1986] to unconsolidated materials (soils) and to investigate the influence of various microstructural properties (such as clay content and configuration) on the electrical properties represented by formation factors, Λ parameters, and surface conductances Σ_s.

Experimental Details

Sample Preparation and Characterization

F-50 Ottawa pure silica sand and Na-montmorillonite were used to build the samples. The samples were packed in 5.1-cm-diameter heat-shrink tubing with sintered Hasteloy frits at both ends providing support and allowing fluid flow. Sample height was 3.8 cm, and Ag-AgCl wire electrodes (to avoid polarization at the metal-water interface) were positioned in the casing 0.64 cm from either end of the sample. Mixtures of sand and clay were either 1%, 3%, or 10% by weight, and several packing geometries were used: dispersed mixtures, discrete clay clusters, and layers of clay in the sand matrix. The dispersed samples were packed from dry,
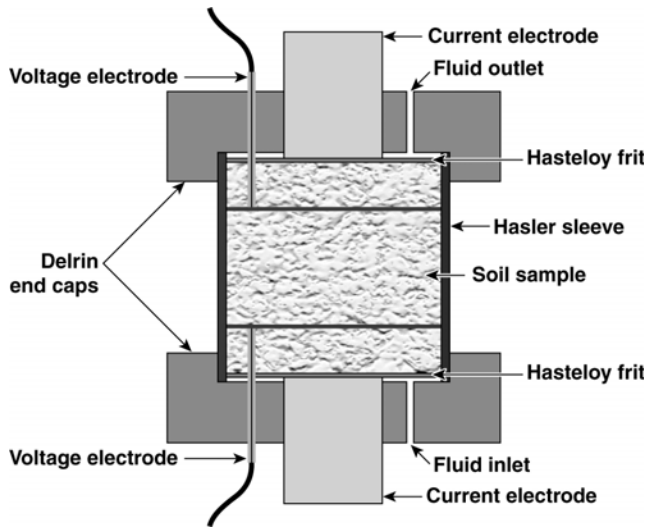
Figure 1. Experimental setup. Cross-section of sample in measurement device.

mixed sand and clay. The samples containing clay clusters were mixed from dry sand and predetermined amounts of dried bits of clay. The clusters varied in size and shape but were ~0.5–1.0 cm diameter when dry. The samples were saturated with saline fluids of varying concentration, ranging from 0.0005 N to 0.75 N of $CaCl_2$ (0.05 to 64 mS/cm) and deionized water. This range of σ_{fluid} encompasses the range of conductivities commonly found in natural groundwater (0.5–1.0 mS/cm).

Apparatus and Measurement Procedures

The experimental setup consists of the sample, the sample holder with pressure controls, the electrical measurement system, and the fluid-flow system. The prepared soil sample was placed in a Hassler sleeve, which enabled pore pressure control (Figure 1). The Hassler sleeve was held in place by end-load and in-load pressures, ensuring that the sample was hydraulically sealed and providing for simulation of shallow burial. The end-load, in-load, and confining pressures were held to 412 kPa during the experiments. Various fluids were pumped through the samples and the electrical properties were measured using an HP4284A LCR-meter, employing the four-electrode method. Following measurement of the electrical properties, we used a constant flow technique to measure hydraulic permeability. A syringe pump provided constant flow rates through the sample, and pressure transducers were used to measure the hydraulic pressure gradient across the sample. Darcy's Law was then applied to obtain the hydraulic conductivity K and permeability k.

Results and Discussion

Hydraulic Permeability Measurements

We performed several tests to confirm that permeability was independent of fluid salinity. Three consecutive measurements were performed on two samples containing 3% clay. On one sample, we measured k using deionized water (DW), then measured it using a 0.1 N $CaCl_2$ solution, and repeated the measurement using DW. The second sample was subjected to permeability measurements with 0.1

N $CaCl_2$, DW, and 0.1 N $CaCl_2$, in that order. We found only minor variation among permeabilities measured using $CaCl_2$ and DW and no consistent change of k with time or total volume of water flowed. Standard deviations for the three measurements, made on the two different samples, were 0.15×10^{-12} m^2 and 0.03×10^{-12} m^2, respectively. The difference in k of the two samples, however, was significant: a factor of ~5 for the two samples with average permeabilities of 1.76×10^{-12} m^2 and 0.36×10^{-12} m^2. We attribute this to variation in packing and compaction, and thus, to variation in bulk density and porosity of the individual samples. All subsequent hydraulic conductivities/permeabilities were measured using DW and are listed in Table 1. These permeabilities varied within two orders of magnitude. The highest was measured for the clean-sand sample (6.1×10^{-12} m^2), and the lowest was found for the 10%-dispersed sample (0.048×10^{-12} m^2). The two other samples containing 10% clay had only slightly lower permeabilities than that of the clean-sand sample. Apparently the 10% clay content did not significantly impede fluid flow when it was arranged as clusters or as a clay layer parallel to the direction of flow.

Electrical Measurements

The electrical measurements are reported at 1 kHz. Measurements were made using $CaCl_2$ as the saturating fluid. Figure 2 is a log-log plot of σ_{bulk} versus σ_{fluid} for the various samples.

There are two different regions visible in Figure 2. At high fluid concentrations (> 1 S/m), σ_{bulk} has a power-law dependence on σ_{fluid}, and the gradient of the line is approximately unity. Movement of ions through the bulk-fluid phase is the dominant mechanism of conduction in this region. However, at low fluid concentrations (< 1 S/m), σ_{bulk} is no longer solely dependent on σ_{fluid}. In this region, surface conductance due to mobile ions in the electrical double layer of the clay becomes more important; the curves tend to a constant, which represents the value of the surface con-
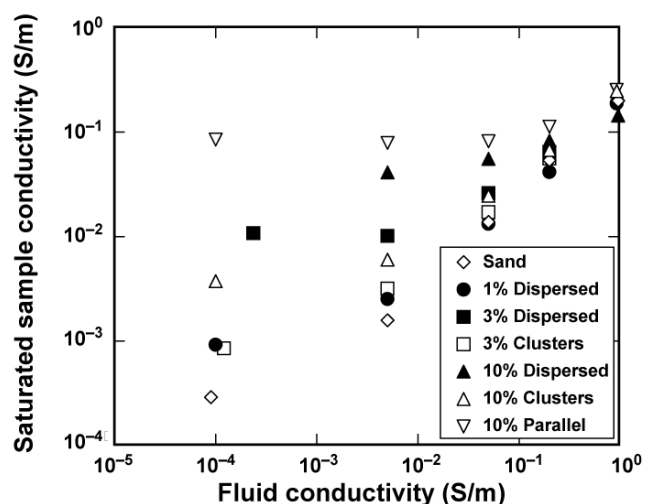

Figure 2. Log-log plot of σ_{bulk} vs. σ_{fluid} for different sand-clay mixtures and distributions. For σ_{fluid} below ~0.2 S/m surface conduction becomes a dominant factor in the total conductivity. Above this value the σ_{bulk} is relatively insensitive to clay content and distribution.

Table 1. Hydraulic and electrical properties measured on the various samples

		F50 clean sand	1% clay dispersed	3% clay dispersed	3% clay clusters	10% clay dispersed	10% clay clusters	10% clay parallel layer
Conductivity (DW)	K (cm/min)	0.360	0.102	0.097	0.135	0.003	0.156	0.228
Permeability (DW)	k (10^{-12} m^2)	6.14	1.74	1.65	2.30	0.05	2.73	3.92
Porosity	ϕ	0.344	0.365	0.365	0.402	0.357	0.306	0.383
Slope	m	0.206	0.191	0.197	0.228	0.076	0.225	0.196
Intercept	b (S/m)	0.0054	0.0055	0.0188	0.0118	0.0576	0.0167	0.0757
Formation factor	F (=1/m)	4.9	5.2	5.1	4.4	13.2	4.4	5.1
Λ parameter	Λ (μm)	15.4	8.5	8.2	9.0	2.3	9.8	12.7
Surface conductance	Σ_s (μS)	0.20	0.12	0.39	0.23	0.85	0.36	2.45
Archie's exponent	m	1.48	1.64	1.61	1.62	2.51	1.26	1.70

ductivity [*Revil et al.*, 1998]. These mechanisms are readily demonstrated in Figure 2 and Table 1. The clean-sand sample has a straight-line dependence of σ_{bulk} on σ_{fluid} and a low intercept of 0.0054 S/m (i.e., a low Σ_s). As the clay content increases, we see an increasing effect of Σ_s on the value of the intercept. With a 3% dispersed-clay content, the intercept is at 0.0188 S/m, increasing to 0.0576 S/m for 10% dispersed clay. A significant change is seen when the distribution of the clay is altered; 10% clay distributed as clusters gives a lower intercept (0.0167 S/m) (i.e., lower surface conductivity) than does a 3% dispersed distribution, and a 10% layered distribution results in the highest intercept (0.0757 S/m) overall. These variations in the contribution of the Σ_s to the σ_{bulk} are discussed in the following section.

Electrical Parameters

We estimate various electrical parameters based on the theory of *Johnson et al.* [1986]. From the slope and intercept of the linear portion of the curves in Figure 2, we calculate the formation factors as the reciprocal of the slope. All the calculated parameters are listed in Table 1. For most of the samples, the formation factors are ~5, except for the 10% dispersed sample, which has a formation factor of ~13.

Under certain conditions, Λ can be correlated with k and F [*Johnson et al.*, 1986; *Kostek et al.*, 1992]:

$$\Lambda = \sqrt{k8F} \qquad (3)$$

Equation 3 is based on the assumption that the tortuosity for both fluid and electrical permeation is the same and it is also assumed that $2\Sigma_s/\Lambda \ll \sigma_{\mathrm{fluid}}$. Equation 3 is similar to the Kozeny-Carman relation ($k_{KC} = (2V_p/S)^2/8F$, three dimensions) in that $2V_p/S$ is replaced with Λ. According to *Revil and Cathles* [1999] the length scale Λ represents a weighted analog of the length $2V_p/S$ in the same sense that $1/F$ represents a weighted analog of the porosity. We use equation 3 to calculate the Λ-parameters listed in Table

1. A high Λ-parameter of 15.4 μm is found for the clean-sand sample, while the 10%-dispersed clay sample has a Λ-parameter of 2.3 μm. We expect a higher Λ-parameter for a sample with a lower surface area (and thus a lower clay content) and vice versa because $\Lambda \approx V_p/S$. Because the sample volume is practically identical for all samples, the Λ-parameter is proportional to $1/S$.

To check our Λ-calculations we used an alternative expression of *Revil and Cathles* [1999] to calculate Λ:

$$\Lambda = \frac{R}{m(F-1)} \qquad (4)$$

where R is the grain radius and m is Archie's (cementation) exponent. Using an average grain diameter of 237.8 μm (from the grain size distribution of our Ottawa sand) we obtain a Λ-value of 20.9 μm, less than a factor of 1.5 different from our estimated value of 15.4 μm. *Bernabe and Revil* [1995] pointed out that Λ is not easy to determine from electrical conductivity measurements; however, the agreement for the one sample we checked seems reasonable.

The surface conductances listed in Table 1 are calculated from the intercept b as (see equation 2):

$$\Sigma_s = \frac{b\Lambda F}{2} \qquad (5)$$

Σ_s varies between 0.20 μS for the clean-sand sample and 2.45 μS for the sample containing 10% layered clay. In comparison, *Nettelblad et al.* [1995] found Σ_s in the range 0.05–0.15 μS for artificially made clay-free sandstones. Our estimated values of Σ_s follow the trends observed in Figure 2. Σ_s increases with increasing clay content for the dispersed samples; once the distribution changes to clustered or parallel, that is no longer the case. The 10% clustered sample has the lowest Σ_s of the clay-bearing samples (0.36 μm), even lower than the sample containing 3% dispersed clay; the 10% layered sample, on the other hand, has by far the highest Σ_s. This is intuitively acceptable because the artificially constructed layered-clay configuration provides

a preferential path for electrical transfer between the frits, whereas clay in clusters is rarely interconnected and has a comparatively low surface area. The measurements on the two extreme configurations of the clustered and the parallel samples may provide bounds on the expected bulk conductivity of clay-bearing sandy soils and thereby help predict conditions in natural systems. The more natural configuration of the 10% dispersed sample falls within these bounds with $\Sigma_s = 0.85$ μm. Electrical measurements might be used to estimate hydraulic conductivities, however, caution must be used. In some clay distributions, such as the parallel layer, the clay provides an electrical short circuit that is different than the path that fluid must flow. In such a case one should not expect a single model to accurately describe both the flow of current and fluid.

Conclusion

We have investigated the influence of various microstructural properties on the electrical properties represented by formation factors, Λ-parameters, and surface conductances. It appears that the Λ-theory of *Johnson et al.* [1986] is also valid for unconsolidated media. We found fairly similar formation factors and Λ-parameters for all the clay-bearing samples, except for the sample containing 10% dispersed clay. For the clay mineralogies studied here, Σ_s varied significantly with clay content and distribution. An increasing surface conduction was found as the samples contained increasingly larger amounts of dispersed clay. The configurations of the clay also played a major role, with the clustered and parallel configurations having the lowest and highest surface conductances, respectively. Apparent high and low bounds on the expected surface and bulk conductance in a natural system may be derived from the measurements on these more artificial configurations and may thereby provide valuable information for inverse modeling of conditions in natural systems.

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