EFFECTS OF ELECTRIC FIELD STRENGTH AND POROUS STRUCTURE ON THE ELECTROOSMOSIS COEFFICIENT

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Experimental results are presented on the electroosmosis coefficient as a function of electric field strength and mean pore size for quartz sand.

Electroosmosis is widely used to consolidate and dry soils, to accelerate impregnation of finely divided systems, and so on, but some systematic studies are required to establish the relationships.

The basic electroosmotic characteristic for a porous material is the electroosmosis coefficient k_e, which is defined as follows [1]:

$$k_e = \frac{\zeta\varepsilon}{4\pi\eta} \cdot \frac{\varkappa_p}{\varkappa_s}. \tag{1}$$

It is found [1] that k_e is dependent on many factors, particularly the concentration of the equilibrium pore solution and the mean pore size. Although (1) does not show any explicit dependence of k_e on pore size, such a relationship exists and makes itself felt via the dependence of $\varkappa_p/\varkappa_s$ on the structural characteristics [2].

The published evidence on this coefficient as a function of grain size is conflicting; for instance, it has been reported [3] that the grain size of a soil has little effect on the electroosmotic behavior, and it has also been claimed [4] that k_e is not dependent on the pore size at all.

In contrast to this, other workers have claimed that the electroosmotic behavior is related to the structural characteristics; there are a few papers on the effects of porous structures in soils on electroosmosis.

Lomize used the method of dimensions to calculate k_e as a function of the porous structure, for which purpose he used characteristics such as the porosity, the hydraulic radius, and the characteristic hydraulic length [5]. The result was checked by experiment on quartz sand of various grain sizes. The curve for $k_e = f(d_{av})$ had a peak for particles of mean size 70 μm.

It has been found [6] that k_e increases as the grain size is reduced for fractions between 500 and 100 μm.

The published evidence thus indicates a need for systematic and thorough studies of the characteristics as regards the electroosmosis coefficient.

The mean particle diameter was used to characterize the porous structure in the above studies, whereas we consider that the mean pore size should be used for this purpose. The porosity of such a medium is not dependent on the particle size and is solely governed by the mode of packing [7], consequently, the pore size in such a system is dependent on the mean particle diameter and on the packing density and is an unambiguous characteristic of the structure.

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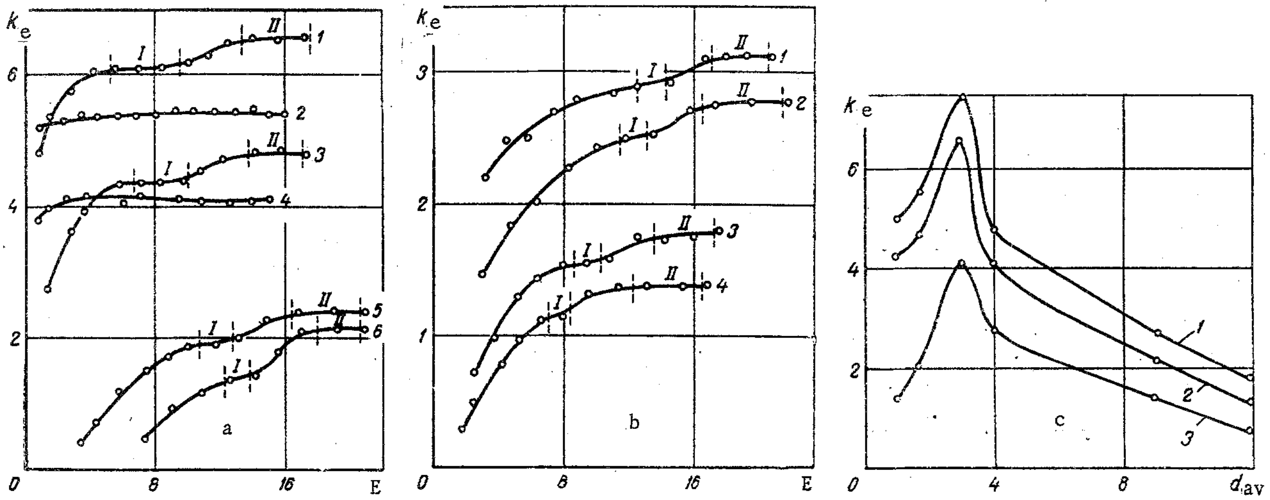

Fig. 1. Electroosmosis coefficient $k_e \cdot 10^8$ ($m^2/V \cdot sec$) as a function of: a) field $E \cdot 10^{-2}$ (V/m) for pore sizes (μm) of: 1) 3, 2) 1.4; 3) 4; 4) 1.0; 5) 9.1; 6) 12; b) field $E \cdot 10^{-2}$ (V/m) for strengths (M) of KCl solution: 1) 10^{-4}; 2) $5 \cdot 10^{-4}$; 3) 10^{-3}; 4) $5 \cdot 10^{-3}$; c) average pore size d_{av} (μm) for strengths (M) of KCl solution: 1) 10^{-4}; 2) $5 \cdot 10^{-4}$; 3) $5 \cdot 10^{-3}$.

We determine the mean pore size from the electrical resistance and the gravitational infiltration rate as measured by the method described in [2].

The materials were purified finely divided quartz-sand powders, with mean particle sizes from 4 to 80 μm.

The filters were made up from this material using a 40% suspension of the powder, which was allowed to sediment spontaneously with continuous vibration for 60 min. This provided the closest particle packing, and the resulting hydrodynamic resistance was adequate, while the structure did not change during the electroosmotic measurements.

The electrical resistance was measured in 0.1 M KCl solution, which enabled us to neglect the surface conductivity [2]. The measurements were made at 10 kHz with an ac bridge to eliminate electrode polarization effects.

The gravitational filtration rate was also measured with 0.1 M KCl solution with the instrument used for k_e [8]. The instrument had a thermostatic jacket, so all measurements were made under isothermal conditions at 20°C. Three copper–constantan thermocouples monitored the temperature within the specimen.

An important point in electroosmotic measurement is to choose the electric field strength; parts a and b of Fig. 1 show our results on k_e as a function of E. The curves for $k_e = f(E)$ have two parts (plateaux) with constant K_e, which are distinguished by the vertical broken lines in the figure. These plateaux are explained as follows. The actual pore channels arise from contact between nearly spherical particles, and they are therefore complicated in shape and variable in cross section, with narrower and broader parts. The above materials were therefore heteroporous not only in section but also in length.

As a first approximation, we represent a real pore as a system of series-connected cylindrical channels with two different diameters, and in that case it is readily shown that the local variations in field strength have an inverse quadratic relationship to the pores radius.

The liquid in the pores has a limiting shear stress τ_0 [9, 10], so the above shows that the small local E in the wide parts will mean that the shear stress τ set up by the electric field will be insufficient to overcome τ_0; consequently, the liquid in these wide parts will be immobile, but the mass flow in the channels is continuous, so these parts will be excluded from the flow. They will be involved in the electroosmotic transport as E is increased to the point where $\tau \geq \tau_0$.

This effect should be the more pronounced the greater the variation in pore size within the body. If the body is homogeneous in structure, no such effect should be observed. These conclusions agree well with our results. We measured the pore-size distributions for all the specimens and found that the range in pore sizes diminished as the particles became smaller. Specimens made up from the smallest particles were practically uniform in size. The reason is that the finest materials were most uniform in particle size, as was clear from sedimentation analysis.

In that case, there is almost no increase in k_e with E (curves 2 and 4 of Fig. 1a). Conversely, a heterogeneous specimen shows a considerable increase in k_e with E before plateau I is reached, and the latter becomes less apparent as the pores become more uniform. Virtually all the pores are involved in the electroosmosis when the critical value E_{cr} is reached, which corresponds to the start of plateau I. The liquid is at rest only in the channels with the largest diameters. Electroosmosis in the latter begins at E corresponding to the end of plateau I. When some limiting value E_{li} is reached, which corresponds to the start of plateau II, the effective pores of all sizes participate in the transport.

Figure 1a shows that the length of plateau I increases as the pore size is reduced. This is explained by the pore-size distribution. The proportion of identical pore sizes increases as the material becomes more uniform, while the numbers of small and large pores decrease. The result is that the dependence of k_e on E becomes less pronounced, as does the stepwise transition from plateau I to plateau II. Plateau II occurs because the largest pores become active. The distribution curves show that the proportion of the largest pores is small, so they make only a small contribution to the rise in k_e. This contribution diminishes as the pores become more uniform, and it is virtually absent for the essentially uniform-pore case.

Here we may note that the electroosmotic current may bypass the expansions; the pores communicate one with another, and the bypass channels may represent stagnant zones at E insufficient to overcome τ_0. These zones will vanish as E increases, which causes k_e to rise.

Also, an electroosmotic pressure is set up at the narrow points that can displace liquid into the expansions; consequently, the stagnant zones may become activated as the field is increased on account of this electroosmotic pressure acting in conjunction with the electric field. This pressure is inversely proportional to the square of the pore radius under otherwise equal conditions, and this can be one of the reasons why E_{cr} and E_{li} shift to smaller values as the pores become smaller.

The rise in k_e with E, and also the effect of the mean pore size, would indicate that there are many stagnant zones in a heteroporous specimen at low E; the number of these decreases as E rises. The largest stagnant zones are activated only at E_{li}, which corresponds to the start of plateau II.

The concepts used in this paper are due in part to N. V. Churaev.

Figure 2b provides some confirmation for this explanation, which shows $k_e = f(E)$ for one particular specimen with various concentrations for the solution. It is clear that E_{cr} and E_{li} shift to lower values as the electrolyte concentration increases, which is due to the increased shear stress on account of the higher density in the diffusion layer.

The constant k_e at $E \geq E_{li}$ indicates that all the liquid is involved in the electroosmosis within all the pores.

Our results indicate that one can take as reliable and comparable results from electroosmotic measurements obtained at field strengths providing $E \geq E_{li}$.

Failure to meet this condition may be one of the reasons for the existing conflict over k_e as obtained on identical materials [1, 11]. This may also explain why our values for k_e for the different fractions of quartz sand are higher than those given in the literature. Previous studies would appear to have been made without considering the $k_e = f(E)$ relationship. Evidence for this is that k_e for a quartz-sand fraction of $\sim 100\ \mu m$ given in [11] coincides with our result only for $E \ll E_{li}$.

Figure 1c shows results for k_e for quartz-sand fractions in relation to mean pore size for different salt concentrations. There is a peak at a mean pore size of 3 μm, which corresponds to a mean particle diameter of 52 μm. The values of k_e are for $E \geq E_{li}$.

It has been found [12] that the electrokinetic potential calculated from the electroosmotic transport decreases as the mean pore radius increases; this was explained in terms of the heteroporous structure.

The effects of heteroporosity were eliminated in our measurements, since we made sure that all the capillaries were involved in the transport.

Bondarenko [13] has shown by calculation that the relationship of [12] is due to effects from the hydrodynamic resistance in the capillaries. We have calculated the error arising from this as regards k_e, and it is about 15% for the coarsest fraction we used, which cannot balance out the fourfold reduction in k_e as the mean pore size increases. This error becomes considerable in a capillary medium with low hydrodynamic resistance, as has been observed [12].

The results might also be influenced by the resistance of the meniscus, so we did checks on k_e with capillaries of various cross sections. However, the dependence of k_e on the mean pore size remained as shown in Fig. 1c.

The fall in k_e as the mean pore size increases appears to be due to the presence of currents of two types in a capillary filled with liquid, which arise from charge displacement in the bulk liquid and in the double electrical layer [14]. The ions produce a galvanic current in the central part of the pore, while the ions in the diffusion part of the double layer produce a surface or electroosmotic current. The two currents coexist, but transport arises only from the second. This view is taken also in [15].

The contribution of the electroosmotic current to the total as recorded by the instruments decreases as the pore size rises; the result is that k_e falls. This also fits in with the observation that calculations of k_e via the available formulas are calculated in terms of the total current from both causes.

The observed fall in k_e for small pores (Fig. 1c) is due to the fall in ζ, which occurs for thin capillaries, as has been pointed out [2, 16]. The complex structure of the pore channels leads us to suppose that the fall in k_e in narrow pores is due to overlap between the diffuse ion layers at the narrow points. A theory has been given for this effect [17].

The experimental results therefore indicate that the electroosmosis coefficient is dependent on the structural and geometrical characteristics of the system. The results give the necessary information for choosing the optimal conditions for applying electroosmosis to intensify filtration processes.

NOTATION

k_e	is the electroosmosis coefficient;
ζ	is the electrokinetic potential;
ε	is the dielectric constant;
η	is the viscosity;
$\varkappa_p$	is the specific electrical conductivity of pore solution;
$\varkappa_s$	is the specific electrical conductivity of equilibrium solution;
d_{av}	is the mean diameter of particles;
E	is the electric field;
E_{li}	is the limiting field;
E_{cr}	is the critical field;
τ	is the shear stress due to electric field;
τ_0	is the limiting shear stress.

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