

TEMPERATURE DEPENDENCE OF THE ELECTROOSMOSIS  
COEFFICIENT FOR A POROUS BODY

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The electroosmosis coefficient has been measured as a function of temperature for fractionated quartz sand with various proportions of pore solution.

The solution to many theoretical and practical electroosmosis problems requires a knowledge of the electroosmosis coefficient, which reflects the physico-chemical nature and structure of the porous body, as well as the properties of the pore solution as regards mass transfer rate.

The available evidence does not give a clear conclusion on the temperature dependence of the electroosmosis. It has been reported [1] that the maximum electroosmosis transport of water through clay barriers occurs in the range 35–40°C; Briggs et al. observed an increase in the rate of electroosmosis for cellulose diaphragms as the temperature rose [2]. Measurements have been made [3] of the  $\zeta$ -potential as a function of temperature, which gave a maximum for water and dissolved NaCl, but a linear rise for ethanol. There have also been reports [4, 5] of a slight increase in the electrokinetic potential with temperature.

The available information is conflicting, and we lack a relative coefficient  $k_e$ , so we examined the effects of temperature on electroosmosis coefficient for typical porous bodies.

The results are of some practical interest, because electroosmosis results in heating of the material, and also because it is often necessary to apply electroosmosis under a variety of temperature conditions.

We used a laboratory apparatus (Fig. 1). This was designed partly on the basis of the specific way of performing the experiments and also in accordance with the requirements for accuracy in electroosmosis [6].

We used quartz sand of grain size less than 50  $\mu\text{m}$ ; we chose this because we needed a chemically indifferent surface to the solid, and also because the model should approximate to ones actually used in nature.

To produce a close-packed diaphragm providing adequate and stable hydrodynamic resistance together with reproducibility, we proceeded as follows: a 40% suspension of the quartz sand in double-distilled water was allowed to sediment spontaneously with continuous vibration for 60 min.

We checked the structure for changes via the bulk filtration rate and the effective cross-section of the diaphragm; the maximum discrepancies between successive diaphragms made in this way did not exceed 5%.

The resulting diaphragm was clamped between two cylindrical perforated plates made of plastic with single-layer inserts of ash-free filter paper. The rigid filters prevented change in the sedimentation volume and also changes in the structure. The criterion for constant structure was the filtration rate. The length was chosen to suit the mean particle size and to provide stable laminar flow, which resulted in constancy in the electroosmosis transport [7].

The powders used in the test were made by agitating washed river sand followed by standard treatment; the particle size was determined by sedimentation analysis.

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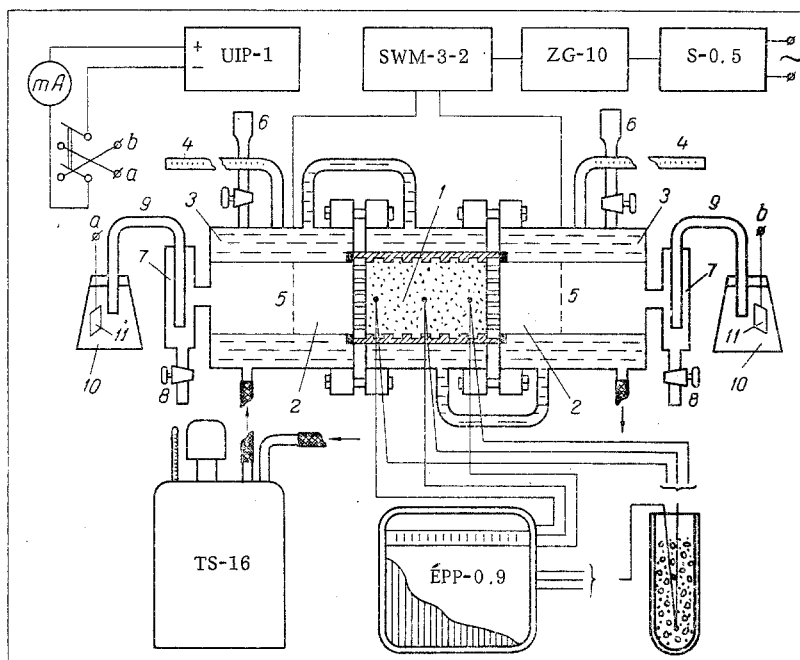


Fig. 1. The apparatus. 1) Sample; 2) equilibrium solution; 3) constant-temperature jacket; 4) reference capillaries; 5) platinum grid electrodes; 6) funnels with taps; 7) traps; 8) taps setting meniscus in capillary; 9) agar-agar bridges; 10) electrode chamber; 11) copper electrodes.

The solutions were of KCl at various concentrations corresponding to maximal electroosmosis effect; the solutions were prepared from chemically pure KCl (twice recrystallized) and double-distilled water having a specific conductivity not greater than  $2 \cdot 10^{-8}$  ohm $^{-1}$  · meter $^{-1}$ . The concentrations were retained by suitable dilution of a 1 N KCl solution, while the specific electrical conductivity was measured at 10 kHz.

The electroosmosis coefficient is defined by

$$k_e = Qx_p / I\tau. \quad (1)$$

The resistance was measured with an SWM-3-2 bridge at 10 kHz; the supply was taken from a ZG-10 audio oscillator fed from an S-0.5 stabilizer. The error of measurement did not exceed  $\pm 1\%$ .

The measurements were made at 10–60°C, which is the range of greatest practical and scientific interest, because it includes the characteristic temperature for aqueous electrolyte solutions [8].

The specimen was kept at a temperature constant to  $\pm 0.1^\circ\text{C}$  by a TS-16 thermostat; the temperature was monitored by three thermocouples and recorded by an ÉPP-0.9 recorder. The three thermocouples made it possible to establish that there was no thermal-gradient potential and to be sure that one was observing purely electroosmotic transport.

The measurements were begun with dilute solutions; higher concentrations were produced by continuous solution flow through the specimen for an hour. The onset of equilibrium was monitored from the specific electrical conductivity of the pore solution.

Checks showed that the replacement of dilute solutions by more concentrated ones without diaphragm repacking had little effect on the structure, as did the electroosmotic measurements themselves; consequently, the material can be considered as rigidly held, and analogous to a rigid diaphragm.

In numerous experiments on various fractions of quartz sand and various solution concentrations we observed no maximum in the electroosmosis; the peak reported in [1] must have been due to structural change in the diaphragm or to imperfect methods of experiment. In those published experiments, the current was kept constant, and the potential gradient therefore fell as the temperature rose, which might have reduced the electroosmotic transport.

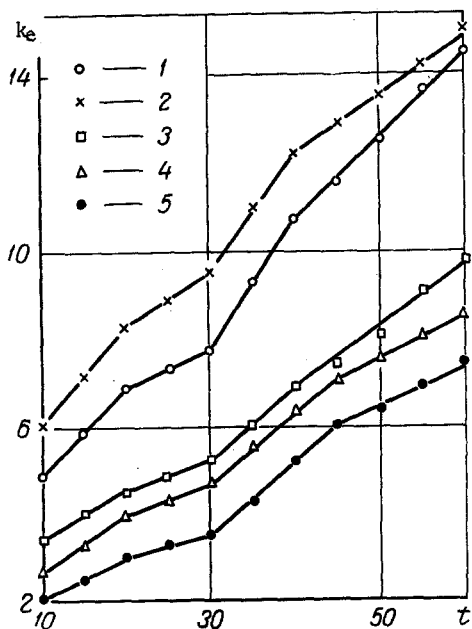


Fig. 2. Electroosmosis coefficient  $k_e \cdot 10^{-8}$  ( $m^2/V \cdot sec$ ) versus temperature  $t$  ( $^{\circ}C$ ) for KCl solutions (N): 1)  $5 \cdot 10^{-5}$ ; 2)  $1 \cdot 10^{-4}$ ; 3)  $5 \cdot 10^{-4}$ ; 4)  $1 \cdot 10^{-3}$ ; 5)  $5 \cdot 10^{-3}$ .

[3] was probably due to swelling of the cellulose, which reduces the flow speed through the diaphragm and therefore reduces the  $\zeta$  potential, which is related to the latter [7]. Evidence for this is that for alcohol, which is less viscous than NaCl solution, there is a straight-line relationship between  $\zeta$  potential and temperature [3].

Figure 2 shows our results for quartz sand with particle size of 30-50  $\mu m$  at various concentrations of the KCl solution; similar results were obtained from 15-30  $\mu m$  sand fraction.

The modern theory of electrokinetic phenomena gives [10] the following expression for  $k_e$ :

$$k_e = \frac{\zeta D}{4\pi\eta} \cdot \frac{\kappa_p}{\kappa_s} \quad (2)$$

The temperature dependence of  $k_e$  is due to temperature changes in the  $\zeta$  potential, the dielectric constant, the viscosity, and the specific conductivity of the pore solution.

It has been shown [4, 5] that the  $\zeta$  potential is only slightly dependent on temperature; our results show, on the other hand, that  $k_e$  increases by a factor 2.5-3 in the temperature range 10-60 $^{\circ}C$ .

As there is only a slight change in dielectric constant with temperature, we are inclined to consider that the viscosity and electrical conductivity are responsible for the variation in  $k_e$ ; then the  $k_e = f(t)$  curves with special points must arise from changes in the properties of the solutions.

Many of the physico-chemical properties of aqueous solutions have numerous anomalies due to their specific structures; recently, some new anomalies related to temperature have been discovered for water [11].

Drost-Hansen [12] has summarized the evidence from numerous studies and has detected four characteristic temperatures: 15, 30, 45, and 60 $^{\circ}C$ , near which there are sharp changes in the properties of water; he himself observed stepwise changes in the viscosity of water at 15 and 30 $^{\circ}C$  in biological processes.

In our case, the singular points occurred at 20, 30, and 40-45 $^{\circ}C$ ; they are seen as kinks on the curves for  $k_e = f(t)$  at 20 and 30 $^{\circ}C$ , which are ascribed to viscosity change in the solution, which is confirmed by the results of [13], which shows that the viscosity of KCl and NaCl solutions as a function of temperature as two parts with different values of the temperature coefficient of viscosity, the second one commencing in the range 10-20 $^{\circ}C$ .

The electroosmosis mechanism [7] indicates that there is a critical potential gradient above which the liquid begins to be transported in the capillaries, this gradient being proportional to the square of the capillary radius [9].

When heteroporous specimens are used, the concept of critical gradient must be applied to the start of transport in the pores of all sizes; if the pore structure is stable and the values correspond to participation of the whole effective cross-section in electroosmosis, one can employ the constancy of the ratio of the bulk transport rate to the current.

The coefficient  $k_e$  is a coefficient of proportionality in the equation for electroosmotic transport and should be dependent on the field gradient. Our results showed that this is correct above a certain potential gradient, in which the effective pores of all sizes are participating in the transport. We call this the limiting gradient, which provides complete participation of all the pores.

The limiting gradients can be calculated theoretically from the differential pore-size distribution for the diaphragm.

One can therefore obtain reliable results for electroosmotic processes in heteroporous specimens if the potential gradient is greater than or equal to the limiting value.

The peak in  $\zeta$  plotted against  $t$  that has been reported

The special point in the range 40-45°C is probably related to the characteristic Mendeleev temperature, which has been considered in papers by Rutskov [14]. There is a kink on the plot of  $\ln \kappa - 1/t$  at this temperature.

Then the continuous variation in  $k_e$  over the range 10-60°C is ascribed principally to temperature variation in the viscosity and specific electrical conductivity of the solution.

#### NOTATION

$k_e$	is the electroosmosis coefficient;
$\zeta$	is the electrokinetic potential;
$t$	is the temperature, °C;
$Q$	is the unit volume transferred by electroosmosis;
$\tau$	is the time for unit volume transfer;
$\kappa_p$	is the specific electrical conductivity of pore solution;
$\kappa_g$	is the specific electrical conductivity of equilibrium solution;
$I$	is the current through sample;
$D$	is the dielectric constant;
$\eta$	is the viscosity.

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