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## Measuring the Electro-osmotic Permeability Coefficient in Single Bricks

--Manuscript Draft--

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| <b>Abstract:</b>                    | <p>The transport process electroosmosis (EO) has been suggested to be used for dewatering damp brick masonry for decades. Still, it is debated whether EO can be obtained in bricks. By use of an advanced EO laboratory cell, this paper reports that an EO flow can be generated in two types of Danish bricks. The electroosmotic permeability coefficient was shown to be dependent on both intrinsic properties of bricks, such as pore size distribution, and extrinsic properties, such as zeta potential. Results showed that the brick with a higher pore volume but with a lower surface charge the EO permeability coefficient is lower.</p>                                                                                                                                                                                                                                                                 |
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# Measuring the Electro-osmotic Permeability Coefficient in Single Bricks

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## ABSTRACT

The transport process electroosmosis (EO) has been suggested to be used for dewatering damp brick masonry for decades. Still, it is debated whether EO can be obtained in bricks. By use of an advanced EO laboratory cell, this paper reports that an EO flow can be generated in two types of Danish bricks. The electroosmotic permeability coefficient was shown to be dependent on both intrinsic properties of bricks, such as pore size distribution, and extrinsic properties, such as zeta potential. Results showed that the brick with a higher pore volume but with a lower surface charge the EO permeability coefficient is lower.

**Keywords:** Electro-osmosis, EO permeability coefficient, pore size distribution, Fired-clay brick, Moisture

## 24 1. Introduction

25 Moisture is one of the main problems affecting both new and historic masonry buildings in Europe  
26 and around the world (Barbieri et al., 2022). It is responsible, among other factors, for unsuitable  
27 indoor thermal–hygrometric conditions, low thermal insulation efficiency of the external walls,  
28 and a decrease in the mechanical performance of the masonry (Franzoni, 2014). In addition,  
29 moisture is involved in all the major deterioration mechanisms affecting porous building materials  
30 once in service (Barbieri et al., 2022), since the presence of moisture in the pores, in combination  
31 with other factors, may lead to biological attack (Warscheid & Braams, 2000), promote physical  
32 damage caused by salt crystallization (Charola & Bläuer, 2015), chemical attack (by different  
33 processes such as hydrolysis, oxidation or hydration), swelling in clay-bearing stones (Elert &  
34 Rodriguez-Navarro, 2022), frost damage (Winkler, 1968), etc., finally causing material loss and  
35 even structural problems (Fort et al., 2022; Sandrolini & Franzoni, 2006).

36 Rising damp, in particular, is a widespread phenomenon leading to the presence of moisture in  
37 masonry, which is a major cause of deterioration of ancient building materials (Franzoni, 2014).  
38 There are different systems for dealing with rising damp in masonry, and according to (Franzoni,  
39 2018) they are mostly based on reducing water supply from the ground, enhancing water  
40 evaporation, or modifying the materials in the wall. The current available international scientific  
41 literature indicates that the drying effectiveness of the systems for overcoming rising damp still  
42 needs to be fully elucidated, and the data about their actual effectiveness in the field are still quite  
43 limited (Franzoni, 2018). Among the different systems currently used, the methods based on  
44 electroosmosis (EO), i.e., based on the application of a direct current (DC) electric field to move

the water in a preferred direction, appear to be a promising way to keep walls dry without affecting the integrity of the building materials.

EO is the movement of liquid in response to an applied electric field across a conduit such as a membrane, capillary tube, microchannel, or porous material (Wiley, D., Fimbres Weihs, 2016). Different researchers independently discovered EO in the early 1800s (Biscombe, 2017) and the early works on electrokinetic phenomena gave rise to the concept of electrical double layer in colloid science (Wall, 2010). The surfaces of many porous and particulate materials (e.g., clays and cation exchange membranes) are negatively charged in an electrolyte solution. This causes the development of the electric double layer (EDL), which is a surplus of the positively charged cations in the EDL close to the surface. The EDL consists of multiple layers such as the diffuse layer; where the electrical potential decreases exponentially from the surface to the bulk solution, the shear plane; this plane is assumed to have mobile ions and accommodates the electrokinetic phenomena, and the Stern layer where a compact layer of ions are attached to the surface of the solid particle (i.e. the ions in this layer are not mobile). The mechanism of EO is explained at the level of a negatively charged surface in Fig. 1. When an electric DC field is applied to the fluid, the net charge in the EDL is induced to move by the resulting Coulomb force. As these cations move under the influence of the electric field, a net flow of water is induced, as water is carried along the cations. This flow of water is known as EO (Grundl & Michalski, 1996).

#### **INSERT FIGURE 1**

The theories of Helmholtz, Gouy-Chapman, and Stern (Philip H.Rieger, 1935) and (Melada et al., 2018) link EO to the EDL. The zeta potential ( $\zeta$ ) is the potential at the shear plane in the diffuse layer separating the mobile liquid from the liquid that is attached to the solid surface (Fig. 1). The zeta potential depends on many factors such as the mineral composition of the porous materials,

properties of the fluid, pH, temperature, etc. (Thanh et al., 2021). For clay soils, the  $\zeta$  potential is usually negative, but it is strongly dependent on the pore-fluid chemistry (Kaya & Yukselen, 2005). Several researchers have studied the  $\zeta$  potential in soil materials and the results show that the concentration of electrolyte, type of electrolyte, valence of ions, and pH are important parameters that can affect its value (Asadi et al., 2013). Thus, the  $\zeta$  potential is not a material's characteristic, but it is a property that depends also on several outer factors.

According to Helmholtz and Smoluchowski, the EO velocity is linearly proportional to both  $\zeta$  potential and the electric field (Keisuke Horiuchi, Prashanta Dutta, 2007). Thus, the actual surface charges on the particulate materials are the most predominant factor influencing dewatering performance. Therefore,  $\zeta$  potential of particles is a good indicator of their electrical potentials: the higher the zeta potential, the higher the surface potential of charged clay particles (Yukselen & Kaya, 2003). As the  $\zeta$  potential increases, the thickness of the double layer increases (Asadi et al., 2013).

The model proposed by Helmholtz and Smoluchowski (H-S model), widely used to describe EO processes in a variety of engineering fields, determines the electro-osmotic permeability coefficient ( $k_e$ ) as follows:

$$k_e = -\frac{\zeta \varepsilon}{\eta} n \quad (1)$$

where  $n$  is the porosity,  $\varepsilon$  is the dielectric constant of the pore fluid and  $\eta$  is the viscosity of the fluid.

According to this model, the electro-osmotic permeability coefficient is highly dependent on total porosity and  $\zeta$  potential (Hu et al., 2019), as seen in equation (1). The H-S model has also limitations, since according to this theory,  $k_e$  should be independent of the pore size distribution

of the material and also it does not take an excess of ions over those needed to balance the surface charge into account (Mitchell, 1993). Therefore, according to the H-S model, EO can be effective for water movement in fine-grained soils. In terms of bricks, it might be important to take pore size distribution into account, especially because the 3-D pore network in this material is stiff and does not change during drying as in soils.

At the pore level, bricks are highly heterogeneous materials, and thus the  $\zeta$  potential must be expected to vary significantly, even over distances in the size of  $\mu\text{m}$ . Thus, the correlation between the  $\zeta$  potential and the EO flow in a brick specimen in practice is not straightforward. A more appropriate option may be to use the theory based on phenomenological coefficients, which can give insight into the EO flow in a specified volume of a material specimen, and hereby cover the heterogeneities, e.g. the anisotropic characteristic of a construction material.

Direct relations exist between the flow of, respectively, fluids, electrical charge, dissolved chemicals, and heat through clays and their corresponding driving force. In most cases, these flows are coupled such as described, e.g., in (Heister et al., 2005). EO is a coupled flow in which the water flow is obtained in an applied electric field. In the case of porous materials, it is common to write the fluxes as a linear function of the driving forces. In consistency with this, the EO volume flux ( $J_V$ ) (m/s) and the charge flux (the current density) ( $J_q$ ) (A/m<sup>2</sup>) can be expressed as

$$J_V = L_{11}\nabla(-p) + L_{12}\nabla(-\psi) \quad (2)$$

$$J_q = L_{21}\nabla(-p) + L_{22}\nabla(-\psi) \quad (3)$$

where  $p$  (Pa) is the fluid pressure, and  $\psi$  (V) is the electric potential (Laursen, 1997). The coefficients  $L_{11}$  and  $L_{22}$  are direct coefficients, where the flux is directly linked to the force. The terms direct and cross coefficients refer to the link between flux and the gradient applied. The coefficient  $L_{11}$  is the factor of proportionality between solution flow and pressure gradient in the

case where no electric field is present (Darcy law). Similarly,  $L_{22}$  is the factor between electric current and applied potential gradient (Ohm's law). The cross coefficient  $L_{21}$  describes the proportionality between applied pressure and the resulting current in the case of no electric fields (the streaming current). Finally, the coefficient  $L_{12}$  is the coefficient of proportionality between volume flow and applied electric field in case of no applied pressure gradient, i.e.,  $L_{12}$  is the EO permeability coefficient (often termed  $k_{eo}$ , which is also the term used in this paper). According to Onsager reciprocity,  $L_{12} = L_{21}$ . In case of no applied pressure gradient, this means that:

$$L_{12} = J_V / \nabla(-\psi) \quad (4)$$

$$L_{22} = J_q / \nabla(-\psi) \quad (5)$$

i.e. the electroosmotic coefficient can be calculated from the volume flow of the solution and the applied potential gradient in a known DC potential field under the condition that there is no fluid pressure as demonstrated also by (Bertolini et al., 2009; Laursen, 1997). The main advantage of this empirical, but direct, approach is simplicity. It is particularly useful when only a few of the possible couplings are likely to be important and when some uncertainty in the measured coefficient is acceptable (Mitchell, 1993). The EO cell of the current investigation is based on equation (4).

A significant percentage of the studies related to EO are focused on the theoretical considerations of the water transport mechanism (Ayoubi et al., 2021; O'Brien, 1986; Rastogi et al., 1970; Vijn & Novak, 1997). Experimental studies, on the other hand, have predominantly concentrated on EO in soils (Ling et al., 2017; Lo et al., 1991; Mele et al., 2020). However, limited studies have been conducted on EO in bricks, and the existing literature has contradictory conclusions on the existence of EO in such materials. For instance, some authors (Ottosen & Rörig-Dalgaard, 2006; Thanh et al., 2021) have successfully dried out bricks by EO, whereas others (Melada et al., 2018)

conclude that EO does not exist in their samples. In (Bertolini et al., 2009), an EO flow of water from the anode towards the cathode was observed, obtaining also a linear relationship between the water flow and the applied gradient potential, establishing 30 V/m as the minimum value for moving water. However, the authors show that the effectiveness of this technique for drying masonry depends on many aspects that tend to nullify or diminish its effectiveness, such as the presence of soluble salts, the presence of positively charged materials that make the water flow in the opposite direction (for example lime mortars), the drying of the samples by evaporation or the greater influence of capillary transport on the electroosmotic one. In (Thanh et al., 2021), it is shown that EO flow is occurring in the brick sample and drying measurement has been successfully carried out with a considerable decrease of water content in the vicinity of the positive electrode.

The methods based on EO for drying moist masonry are offered commercially in some countries, e.g., Denmark, Check Republic, and Spain. The systems have theoretically clear operating principles and have already been applied for decades to masonry buildings. However, their actual effectiveness in real masonry is still to be fully understood and still debated among professionals (Franzoni, 2018). The present work aims to deepen the existing knowledge about the EO transport of water in bricks, analyze if EO can be obtained, and determine which parameters of bricks' physical properties influence the process. For this purpose, an experimental setup for measuring EO coefficients in brick specimens is developed and tested for two different types of bricks.

## **2. Material and Methods**

### **2.1. Bricks and properties**

Two brick types were used in this study, namely ordinary yellow and red bricks. They represent bricks that are widely used in buildings in living areas in Denmark. The bricks were new from the

manufacturers. Four cubes (3 cm of side) were cut from each brick to be used in the EO experiments (Fig 2). The rest of the brick was cut into blocks (5 cm long) and they were used to measure different properties of the bricks.

INSERT FIGURE 2

Taking into account, as stated by (Zhou et al., 2021), that the EO transport is affected by the water content that a material can absorb, as well as by the thickness of the electrical double layer, and the pore size distribution, the following properties were analyzed.

Properties related to the pore system were measured: (a) Water-accessible porosity (n=5; following UNE-EN 1936:2007), (b) Capillary absorption coefficient (n=5; UNE-EN 1925:1999), and (c) Mercury-accessible porosity and pore size distribution, measured by mercury intrusion porosimeter using Thermo Fisher Scientific Pascal 140 and 240 using duplicate brick samples having size  $1 \times 1 \times 2 \text{ cm}^3$ .

Ultrasonic pulse velocity (UPV) was measured using a Pundit Lab+ instrument fitted with transducers (1 MHz frequency and 11.82 mm diameter). Ten direct measurements were taken in each orthogonal direction in dried brick samples. From these data, the anisotropy index proposed by (Guyader & Denis, 1986) was determined using the following expression:

$$dM(\%) = [ 1 - ( 2V_{pmin} / ( V_{pmean} + V_{pmax} ) ) ] \cdot 100 \quad (6)$$

where  $V_{pmin}$ ,  $V_{pmean}$ , and  $V_{pmax}$  correspond to the lowest, intermediate, and highest velocity values, respectively.

The  $\zeta$  potential was measured on a powdered brick sample at the pH of 6.7, in triplicate, using a Zetasizer Nano Z, from Malvern Instruments. For this purpose, 10 g of each powder was taken and diluted in 25 ml of distilled water.

## 2.2. Electro-osmotic setup

The experimental setup for measuring EO coefficients used in the current work was based on the setup developed in (Laursen, 1997), originally designed for measuring the EO coefficients in clays (Figure 3). To adapt the cell for brick specimens, a new sample holder was designed (Figure 4). The cell was designed so that all the other gradients except the potential gradient are eliminated, i.e., under the application of a potential gradient the movement of water in this cell can only be due to EO. The EO cell had four chambers and was symmetrical around the central vertical axis. It was made of heat-resistant glass.

**INSERT FIGURE 3**

**INSERT FIGURE 4**

The brick sample (a cube with a side of 3 cm) was placed between two compartments, which again were separated into two chambers (upper and lower chamber), by a porous glass plate and glass beads with 3 cm height. The purpose of this barrier of glass beads was to isolate the changes in chemical conditions induced by electrode processes in the lower chambers from the upper chambers of the cell, which was in direct contact with the brick specimen.

The lower chamber contained the working electrodes, which were Cu rods 1.0 mm in diameter, and 1.0 M CuCl<sub>2</sub> solution. In this lower chamber, the electrode processes were as follows:

Anode reaction       $\text{Cu}_{(s)} \rightarrow \text{Cu}^{2+}_{(aq)} + 2e^{-}$

Cathode reaction       $\text{Cu}^{2+}_{(aq)} + 2e^{-} \rightarrow \text{Cu}_{(s)}$

197 The upper chambers contained the measuring electrodes, Ag/AgCl electrodes, which were placed  
198 with the tips outside the brick sample holder, i.e., approximately 7 cm from the brick surface (the  
199 closest possible distance in the cell). They were used for measuring the potential difference over  
200 the brick specimen. The Ag/AgCl electrodes were produced for the purpose from a silver wire  
201 with a thickness of 1 mm. The wire was covered with a solution-resistant cover, so only 1.0 cm  
202 from the tip was uncovered. The wire was then placed in a NaClO solution (5% concentration) for  
203 30 minutes to get a coating of silver chloride (after the procedure from (Tao et al., 2018)). The  
204 solution in the upper chambers was tap water.

205 Capillary tubes with an internal diameter of 0.35 mm were placed on the top of the upper chambers.  
206 These were used to measure the volume of flow, which is calculated back to give EO coefficients.  
207 Ground glass sealed the connections between capillary tubes in the upper chambers. The tip of the  
208 capillary tubes was open to the air to let the solution move freely without any disturbing pressure.  
209 The brick specimen was embedded between two sample holders designed for this purpose (Fig.  
210 4). The sample holder was printed in a fast-curing, anti-scratch tough resin for 3D printing called  
211 Value Resin. The ends of the sample holders fitted into the ground glass opening in the upper  
212 chamber and the ends toward the specimen were flat to cover the whole face of the brick specimen.  
213 The free sides of the specimens and the joints between the specimen and sample holders were  
214 coated with epoxy resin, to avoid evaporation, so the only transport of water into and out from the  
215 bricks specimens was through the openings in the sample holders. After coating, the resin was left  
216 to cure for 24 hours, and then the pores of the brick samples were filled with tap water.

### 2.3. Procedure to measure the electro-osmotic coefficient

Before performing a test, the cell parts were washed in distilled water. The capillary tubes were immersed in 1.0 M  $\text{HNO}_3$  for 24 hours for cleaning. After mounting the cell parts, the chambers were filled with the respective solutions. The upper chamber was filled until only some centimeters from the tip of the capillary tubes were empty. This was intentional to see the water movement when an electric potential is applied. All the joints and inputs were checked to avoid leaking, ensuring that the solution movement happens only inside the brick samples. Before applying the electric current, the cell was left for approximately 30 minutes to let the solution and the brick samples reach a pressure balance. The working electrodes were connected to a power supply (Agilent E3612A), which was set to supply a constant current. For each of the brick samples tested, the EO movement was evaluated under two different applied currents: 1 mA and 2 mA. After applying the potential difference, the time needed for the tap water to travel a distance of 5 cm and 10 cm inside the capillary tube were measured. The polarity was changed to make sure that the movement of the solution was due to the electric current (this change in polarity is called iteration). A total of six iterations were performed for each brick cube and each brick type (4 cubes for each condition).

During the test, the potential drop was measured with a multimeter connected to the Ag/AgCl electrodes. From these data and applying Ohm's law, the resistance of each sample to the current flow was determined. Using these data, the electrical resistivity was determined, considering the dimensions of the samples, and its inverse, i.e., the electrical conductivity. The electrical conductivity ( $L_{22}$ ) is the reciprocal of the electrical resistivity, and it is thus a material's property.

Based on the measurements of the flow rate in the capillary tube and the potential difference measured by the reference electrodes (and considering the dimensions of the brick specimen), the EO coefficient ( $L_{12}$ ) can be calculated as seen in Table 1.

## ***INSERT TABLE 1***

At the end of each test, samples were taken from the solution in the upper chambers to measure the pH and conductivity (with electrodes) to document any changes from the initial values of the tap water used (pH 7.3 and conductivity 0.8 mS/cm). The purpose was to confirm no additional factors such as pH and conductivity gradients over the brick sample could have affected the electroosmotic transport of water. Changes in pH and conductivity could also have affected EO by changing the  $\zeta$  potential (caused by a change in the pH), or modification of the EDL thickness caused by the presence of salts.

## **3. Results and discussion**

### **3.1. Bricks Properties**

Table 2 shows the measured values related to the pore system of the bricks and their  $\zeta$ -potential. It can be observed that the Yellow brick has a higher porosity than the Red brick, both in terms of accessible porosity to water and mercury (in both cases approximately 7 % more). It is also observed that capillary porosity is higher in the Yellow brick (approximately 2 % more), although absorption occurs at a slower rate because the capillary coefficient is lower than in the Red brick (Figure 5). This coefficient is the rate of absorption of water due to capillary forces for building

materials and is a measure of the tendency of a material to absorb and transmit water by capillarity (Karagiannis et al., 2016; Steiger et al., 2011).

**INSERT TABLE 2**

***INSERT FIGURE 5***

The water absorption behavior of porous materials can be described through the water absorption coefficient. In this sense, the water absorption coefficients, both under vacuum (16.1% and 19.9% for Red and Yellow bricks, respectively) and by capillarity suction (10.4% and 15.7% for Red and Yellow bricks, respectively), show that there is greater pore connectivity in the Yellow brick, reaching a higher saturation degree in this brick (80% approximately by capillary suction), which is higher than that achieved in the Red brick (60% by capillary suction). The degree of water uptake is mainly related to the pore space itself, the porosity, the type of pores, the pore geometry, and the interconnectivity of the pores (Steiger et al., 2011).

Both bricks have an unimodal pore distribution (Figure 6), Yellow brick shows a pore range between 0.4 and 2  $\mu\text{m}$  (peaking at 1.3  $\mu\text{m}$ ), while the Red brick shows a pore range between 0.3 and 4  $\mu\text{m}$  (peaking at 3  $\mu\text{m}$ ). Within the range of pore sizes in the capillary access diameter, between 0.1 and 100  $\mu\text{m}$ , the Yellow brick has a pore volume of 98.3%, while in the case of Red this value is slightly lower (97.6%), which is consistent with the different capillary behavior observed above.

In the Yellow brick, the 95.13% lies in the range between 0.1 and 10  $\mu\text{m}$  (32.0% in 0.1 – 1  $\mu\text{m}$  and 63.13% in 1 -10  $\mu\text{m}$ ) (Table 3). Only 1.4% lies in the nanometer range, and the rest of the pore's

volume is higher than 100  $\mu\text{m}$  (about 0.3%). In the Red brick, the 87.66% lies in the range between 1 and 100  $\mu\text{m}$ . Only 2.2% lies in the nanometer range, and as occurs in Yellow brick around the 0.3% of the pore volume is higher than 100  $\mu\text{m}$ .

**INSERT FIGURE 6**

***INSERT TABLE 3***

In summary, it is observed that the Yellow brick is characterized by a greater open porosity, significantly higher than the Red brick, and is also characterized by much thinner pores than the Red one, which indicates that the effect of porosimetry on electroosmotic transport in Yellow brick should be greater according to (Zhou et al., 2021).

The  $\zeta$ -potential of both bricks is negative, suggesting that electroosmotic transport occurs from anode to cathode and it is slightly higher in the Red brick (-36.0 mV and -32.4 mV for Red and Yellow brick, respectively), indicating that the thickness of the EDL when the brick has a similar electrolyte is greater in Red brick.

### **3.2. Electrical conductivity of the brick specimens**

From the literature on measurement techniques of moisture in building materials, it is known that the electrical resistance is, in a simplified description, primarily governed by the following: the amount of water present in the pores, the electrical conductance of the pore fluid, the porosity and pore connection, the presence of salts and finally temperature (Phillipson et al., 2007). The general

trend observed is that the resistance of a material decreases with increasing moisture and salt content (Cacciotti et al., 2018; Feijoo et al., 2013).

Figure 7 shows the average conductivity measurements calculated from the potential drop measured during the test. In general, it can be seen that 1) the average conductivity value, for each applied intensity, is slightly higher in the Yellow brick, although the differences are not statistically significant. This could be due to the higher accessible porosity and the higher water content of this brick when it was saturated with tap water under vacuum. 2) The conductivity increases as higher intensity is applied, especially in the Yellow brick. When 2 mA was applied, the conductivity values ranged between 9.4 and 7.6  $\mu\text{S}/\text{cm}$  for the Yellow brick and from 8.2 to 7.5  $\mu\text{S}/\text{cm}$  for the Red brick, while when 1 mA was applied, these values were lower and ranged between 6.1 to 6.4  $\mu\text{S}/\text{cm}$  for Yellow brick and 5.8 to 6.4  $\mu\text{S}/\text{cm}$  for Red brick. Figure 7 shows that the current applied was outside the ohmic region because the conductivity (and hereby resistivity) was different at the two applied currents. In ion-exchange membranes, it is well known that a limiting current can be reached, at which the current does not increase regardless of the voltage is increased (Długołęcki et al., 2010). Since the bricks had negatively charged surfaces (negative  $\zeta$ -potentials Table 2) a similar limiting current density as in cation-exchange membranes may exist, which would result in lower resistance at this point at higher currents, i.e., it would show as higher conductivity at higher currents, just as seen in Figure 7, however, the possible effect of over-limiting currents in bricks needs further investigations.

**INSERT FIGURE 7**

### 3.3. Electroosmotic flow

An EO flow was obtained in both brick types when applying the potential difference to the working electrodes, showing that EO can be obtained in these brick types. The water flow by EO was from the anode towards the cathode, as expected from the measured negative  $\zeta$  – potential (Table 2). Movement of water was observed in both capillary tubes, on one side the water was moving toward the outer part of the capillary tube, while on the other side, the water movement was in the opposite direction, toward the cell. In each iteration, when the polarity was changed at the current electrodes, the direction of the water movement in the capillary tubes consequently changed.

#### INSERT FIGURE 8

It was found that the EO water flow in the Red brick was faster than in the Yellow brick (Figure 8), which could indicate the influence of different pore sizes or different  $\zeta$ -potentials on the water transport. For instance, it took approximately 132 s (average) for water flow in the Red brick to reach 10cm in the capillary tube under the current of 1 mA, while it took around 307 s (average) in the Yellow brick under the same current. When the working current was increased to 2 mA it took 62.9 s (average) and 110.2 s (average) to reach 10 cm for the Red and Yellow bricks respectively, in this case, this time-difference between the two bricks (40.3 s) was much lower than that registered for 1 mA (175 s). This is related to the considerable increase in the electrical conductivity of the Yellow brick when the applied current was increased (Figure 7). This result (Figure 8) confirms what was seen in previous studies about the relationship between the applied electric current and the EO flow of water so that as the electric current increases, the speed of water

movement increases (Bertolini et al., 2009). This is related again to the conductivity observed in the brick samples (Figure 7) since it was observed that the higher the current density, the lower the resistivity of the samples to current flow.

The water flux, observed using this setup, can be attributed only to EO since the fluid pressure was removed, no water flow was observed when no current was applied, and the direction of the flow was reversed when reversing the direction of the current. EO flux ( $J_v$ ), also called EO flow rate, was calculated through the volume of water in 10 cm of capillary tube per time of water movement multiplied by the area of the brick (Table 1).

Figure 9 shows the EO flow rate in the Red and Yellow bricks at different electric currents. It was found that (I) the higher the current intensity applied, the higher the EO flow rate, regardless of the brick, a similar result to what has been obtained in clays (Zhou et al., 2021). In the Red brick, at 2 mA, the EO flux was  $1.70 \cdot 10^{-7}$  m/s, while it dropped to  $8.36 \cdot 10^{-8}$  m/s at 1 mA. The same behavior was observed in the Yellow brick,  $1 \cdot 10^{-7}$  m/s at 2 mA, which decreased to  $4.97 \cdot 10^{-8}$  m/s at 1 mA. (II) The EO flux was higher in the Red brick compared to the Yellow brick under the same conditions. These results show a difference in the EO flow between Red and Yellow bricks, which is according to their different pore structures. As mentioned in section 3.1, the Yellow brick has a lower EDL thickness and higher micropore content than Red brick, which makes the effect of pore size more significant in the EO process (Zhou et al., 2021). Furthermore, the capillary coefficient obtained indicates that the capillary pores of the Red brick allow water to move faster than in the Yellow brick, an aspect that seems to have an impact on the effectiveness and rate of electroosmotic transport.

**INSERT FIGURE 9**

### 3.4. The EO coefficients

The EO permeability coefficient was measured using equation (4) in the two bricks (Red and Yellow) with four replicas (overall 8 brick specimens) based on 10 cm of water movement in the capillary tube under two different electric currents (2 mA and 1 mA).

Figure 10 shows the EO permeability coefficient measured in all the replicas. It can be seen that: 1) in general terms, the Red bricks had a higher EO coefficient compared to the Yellow bricks regardless of the electric current applied. 2) The electroosmotic coefficient, regardless of the brick, was higher at higher current intensity. However, this difference was small in the red brick, where both coefficients were very close. The coefficient values for R1 were  $7.33 \times 10^{-11}$  and  $6.86 \times 10^{-11}$   $\text{m}^2/\text{V} \cdot \text{s}$  for 2 and 1mA, respectively, and for R2 were  $7.62 \times 10^{-11}$  and  $6.94 \times 10^{-11}$   $\text{m}^2/\text{V} \cdot \text{s}$  for 2 and 1mA, respectively. 3) The variability between brick samples, as well as the dispersion between iterations (i.e., polarity changes), was higher in the Yellow brick. This variability between samples of the same brick is likely related to the anisotropy of the bricks, which causes that although the cubes were cut from the same piece of brick, the EO water flow rate was not stable. As shown in Table 2, the anisotropy index of Yellow brick is higher than that of Red brick (7.78 and 3.68 respectively).

### INSERT FIGURE 10

Analyzing all the data together, the EO permeability coefficients were in the ranges from  $1.49 \times 10^{-11}$  to  $6.64 \times 10^{-11}$   $\text{m}^2/\text{V} \cdot \text{s}$  under the current of 1 mA, and from  $2.93 \times 10^{-11}$  to  $7.92 \times 10^{-11}$   $\text{m}^2/\text{V} \cdot \text{s}$  under the current of 2 mA. These values are lower than those usually found in fine-grained soils, which ranges between  $1.10^{-8}$  and  $1.10^{-10}$   $\text{m}^2/\text{V} \cdot \text{s}$  (Mitchell, 1993), and even in clays, for example, in

(Laursen, 1997) the EO permeability coefficient for clays ranges from  $7.94 \times 10^{-9}$  to  $6.30 \times 10^{-10}$   $\text{m}^2/\text{V} \cdot \text{s}$ .

Figure 11 shows the EO coefficient results obtained from the average values for both Red and Yellow bricks under the applied electric current of 2 and 1 mA. As shown in the previous figure, it was found again that the Red brick has a higher EO coefficient value than the Yellow brick in both applied currents. The results showed that in the Red brick the EO permeability coefficient decreases by decreasing the applied electric current, i.e., from  $7.46 \times 10^{-11} \text{ m}^2/\text{V} \cdot \text{s}$  to  $6.19 \times 10^{-11} \text{ m}^2/\text{V} \cdot \text{s}$  passing from 2 mA to 1 mA. Regarding the Yellow brick, the same behavior was observed, as the EO coefficient decreases from  $4.80 \times 10^{-11} \text{ m}^2/\text{V} \cdot \text{s}$  to  $3.23 \times 10^{-11} \text{ m}^2/\text{V} \cdot \text{s}$  by decreasing the applied electric current from 2 mA to 1 mA. These results show that both the  $\zeta$ -potential (higher in the Red brick) and the physical properties related to water transport, especially the capillary coefficient and the shape and size of the pores, have a direct influence on the electroosmotic flow. The presence of a capillary pore volume with larger pore size (between 10-100  $\mu\text{m}$ ) seems to favor the electroosmotic flow, while the presence of a higher micro-porosity between 0.1-1  $\mu\text{m}$ , despite being within the capillary range, seems to hinder it, as observed also in (Haijun Zhou, Yingguang, Minghua Chen, Rengou Gu, 2021) and in the Yellow brick.

#### **INSERT FIGURE 11**

During the experiments, it was observed that sometimes the movement of water inside the capillary tube was not continuous. The EO flow slowed down until the pressure caused by the EO inside the chamber was large enough to accelerate the movement again. This implies that the measurement of the EO permeability coefficient is more effective at longer distances from the capillary tube,

such as 10 cm, than at short distances, since the rate of water movement varies along the capillary tube.

According to the results, what was previously indicated by the Helmholtz and Smoluchowski model seems to be contradicted. In fact, in the case of building materials, such as bricks, it seems that the pore size distribution has a strong influence on EO permeability. Meanwhile, can be a suitable method to measure the EO coefficient in bricks since the bricks are heterogeneous materials and the phenomenological method covers better these heterogeneities, such as the variations of the pore sizes.

The results obtained in this study show that the pore size distribution greatly influences the EO permeability of bricks, so the values obtained from the Helmholtz and Smoluchowski model, widely used in soils, do not correspond to those obtained using the phenomenological approach. In fact, using this H-S model (equation 1) it would be expected that the EO permeability would be higher in the Yellow brick, obtaining an electroosmotic coefficient of  $7.75 \times 10^{-7} \text{ m}^2/\text{V}\cdot\text{s}$  compared to  $6.60 \times 10^{-7} \text{ m}^2/\text{V}\cdot\text{s}$  for the Red brick.

### **3.5. The electrolyte in upper chambers at the end of experiments**

The solution (tap water) at the upper chambers of the cell used in this study was renewed for each of the specimens. During emptying, a small portion from each side was taken to study the pH change and conductivity.

At the end of each test, it was confirmed that there was no change in pH after treatment from the initial value, demonstrating that the separation of two chambers of solution using glass beads was successful in hindering the change of this parameter caused by the electrolysis of water in the upper chambers as a consequence of electrode reaction in the lower chambers. Furthermore, the polarity

in each test was changed after the water reached 10 cm in the capillary tube. This was repeated 6 times for each specimen. Hence, it was expected that the pH would not change at the end of each test since the time for each polarity change was relatively short (between 63s to 503s). Consequently, it can be considered that no variation of the  $\zeta$ -potential of the brick due to pH changes occurred during a test.

Regarding the conductivity of powdered brick, the variation was low, going from an initial value of 0.8 mS/cm to an average final value of 1.1 mS/cm, so it can be neglected that the thickness of the double layer in the bricks should be affected by a variation of the ionic strength. This conductivity variation could be caused by a slight migration of the ions present in the  $\text{CuCl}_2$  solution, similar results to those obtained when the conductivity of the brick samples were analyzed during the test (section 3.2).

#### **4. Conclusions**

EO has been offered for decades as a process for drying brick masonry exploiting the electrokinetic phenomena, while at the same time, it has been debated whether an EO flow can be obtained in bricks/masonry; literature on the topic is sparse and contradictory. The current work introduces a laboratory setup for the direct measurement of EO coefficients in brick specimens based on the theory of phenomenological coefficients. Two types of Danish bricks were used in the work and EO flow was obtained in both. The electroosmotic permeability coefficient was found to be in the order of  $10^{-11} \text{ m}^2/\text{V}\cdot\text{s}$ , which is about two orders of magnitude lower compared to e.g. bentonites. The key findings of this research are:

- EO was obtained in the two types of bricks used in this study.

- The phenomenological approach based on non-equilibrium thermodynamics was used to measure the electroosmotic permeability coefficient, and it was seen as a suitable tool for this purpose since it considers the physical properties of the bricks including pore structure.
- A relationship between electroosmotic water flow and pore size distribution of the bricks was observed i.e., the Yellow brick with narrower pores exhibited a lower electroosmotic flow rate compared with the Red brick with wider pores and higher capillary coefficient.
- A higher  $\zeta$  potential in the bricks showed an increased electroosmotic permeability coefficient.
- A higher applied current density causes a higher electroosmotic flux.

In summary, EO can be obtained in bricks and could as such be a tool to remove humidity from masonry; however, research is needed to further determine relevant parameters and for the practical use, the influence of mortar must be explored.

#### Data Availability Statement:

Some of the data and models that support the findings of this study, are available from the corresponding author upon reasonable request.

All data, models, and code generated or used during the study appear in the submitted article.

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## FIGURE CAPTIONS

**Figure 1.** EOF in clay matrix caused by the electric current which makes the hydrated ions move, consequently pushing water molecules out (inspired by (Delgado et al., 2007)).

**Figure 2.** Image of how the brick cubes were cut from one brick piece.

**Figure 3.** The schematic drawing of the electro-osmotic setup used.

**Figure 4.** A schematic drawing of that sample was placed between two sample holders and then covered completely so that the only open areas of the bricks were those in contact with the solution.

**Figure 5.** Water content per unit surface area during the capillary absorption test.

**Figure 6.** Pore size distribution of both bricks. Data shows the incremental intrusion volume and the cumulative volume, both in mL/g.

**Figure 7.** Conductivity average values ( $n=4$ ) in  $\mu\text{S}/\text{cm}$  are shown for the two saturated brick specimens.

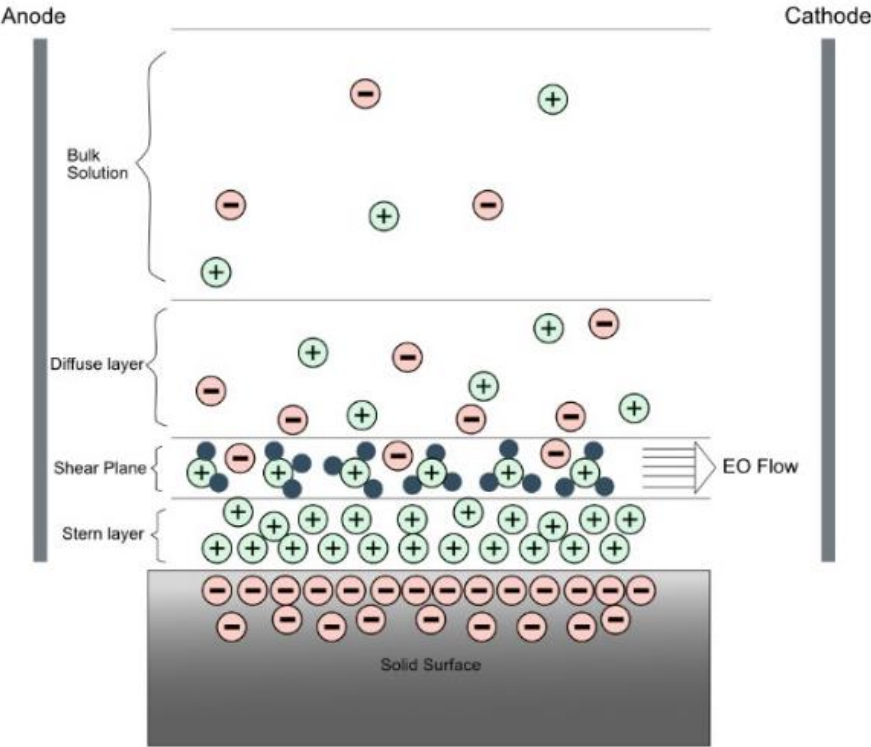
**Figure 8.** Average EO rate (in  $\text{cm}/\text{s}$ ) of the Red and Yellow bricks under two different electric currents, 2.0 mA and 1.0 mA, calculated based on 10 cm in the capillary tube.

**Figure 9.** Average EO flow rate (in  $\text{m}/\text{s}$ ) of the Red and Yellow bricks under two different electric currents, 2.0 mA and 1.0 mA, calculated based on 10 cm in the capillary tube.

**Figure 10.** Electroosmotic permeability coefficient at different currents, 1 and 2 mA, for the bricks used in this study. The error bars show the dispersion between the results obtained in each of the six iterations performed, i.e., in each polarity change.

**Figure 11.** Average values of electroosmotic permeability coefficient in two bricks based on 10cm of water movement in the capillary tube.

Figure 1



629 **Figure 2.**

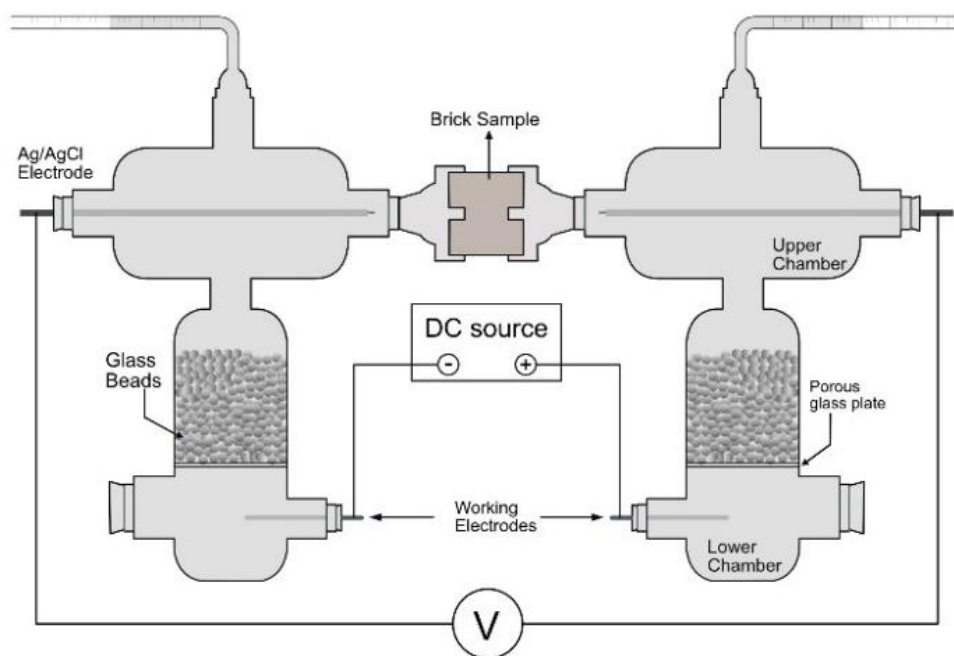
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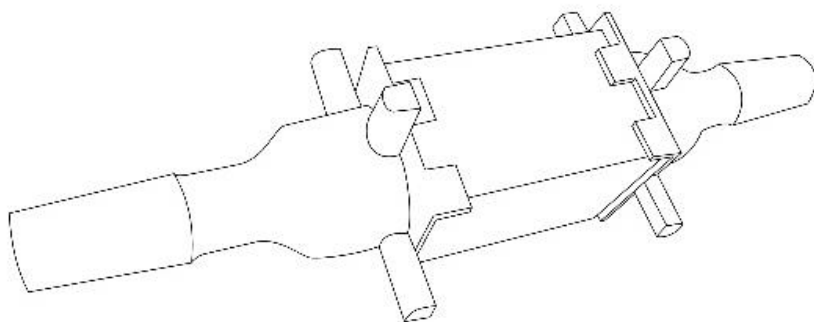
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**Figure 3.**



637 **Figure 4.**

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Figure 5.

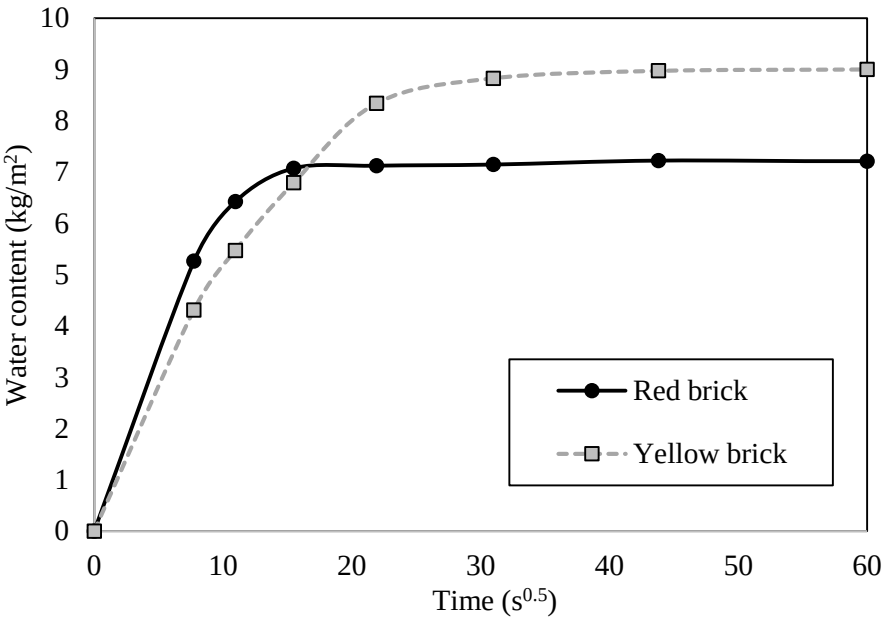


**Figure 6.**

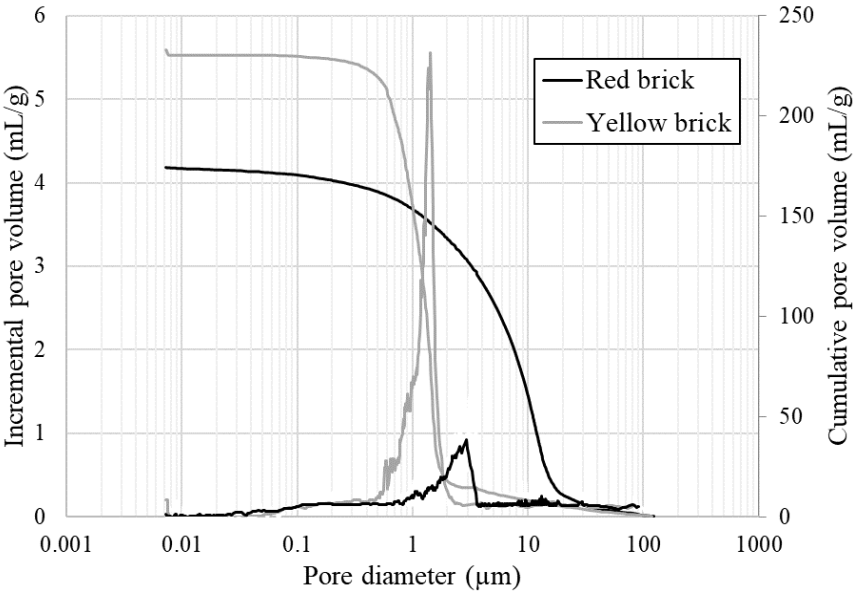


**Figure 7.**

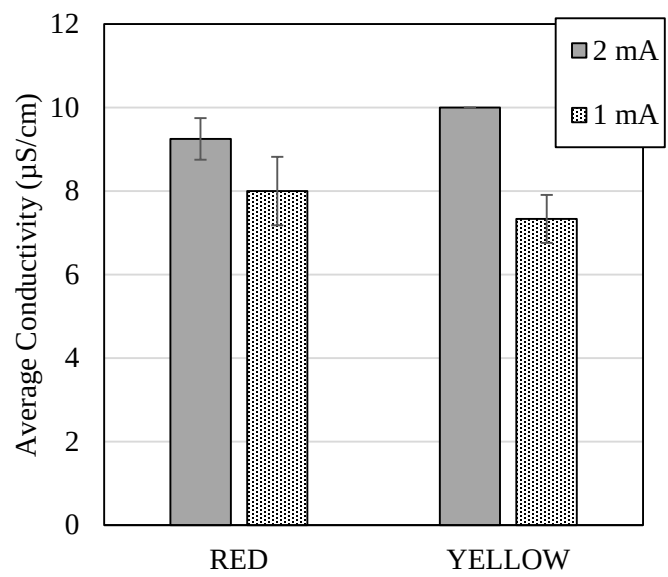


**Figure 8.**

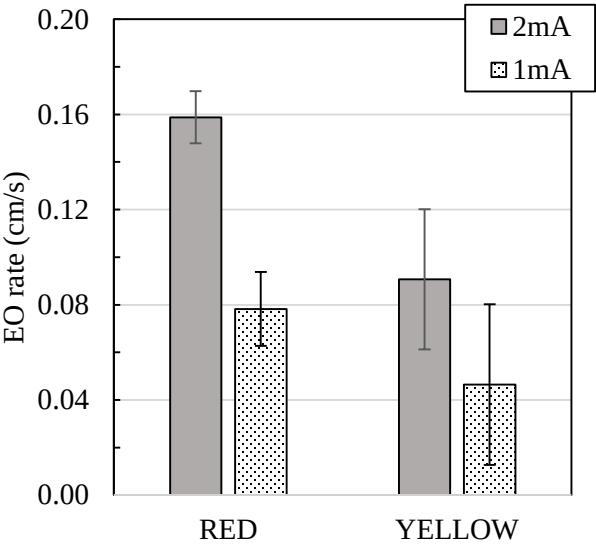
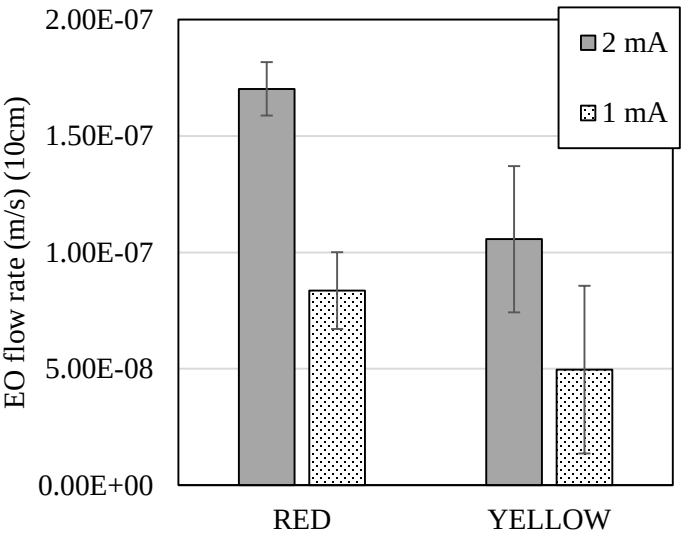
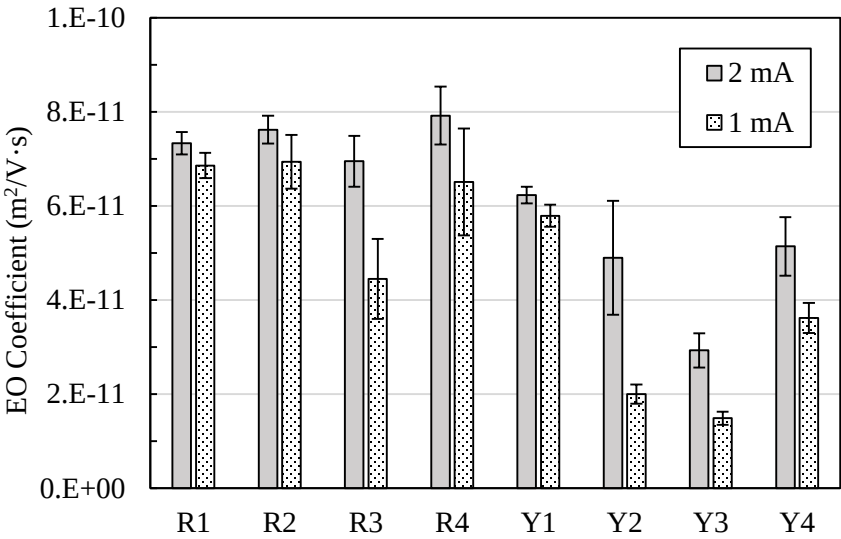


**Figure 9.**



663 **Figure 10.**

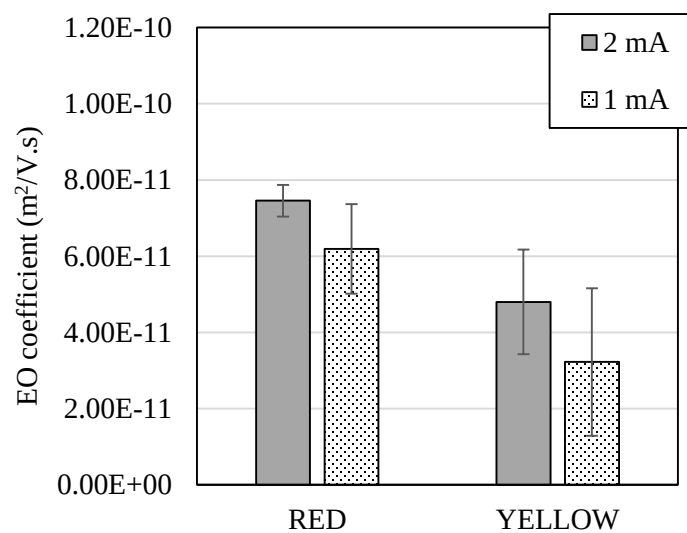


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666 **Figure 11.**

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Tables

**Table 1:** Calculation of conductivity ( $L_{22}$ ) and EO coefficients ( $L_{12}$ ) from the measured data

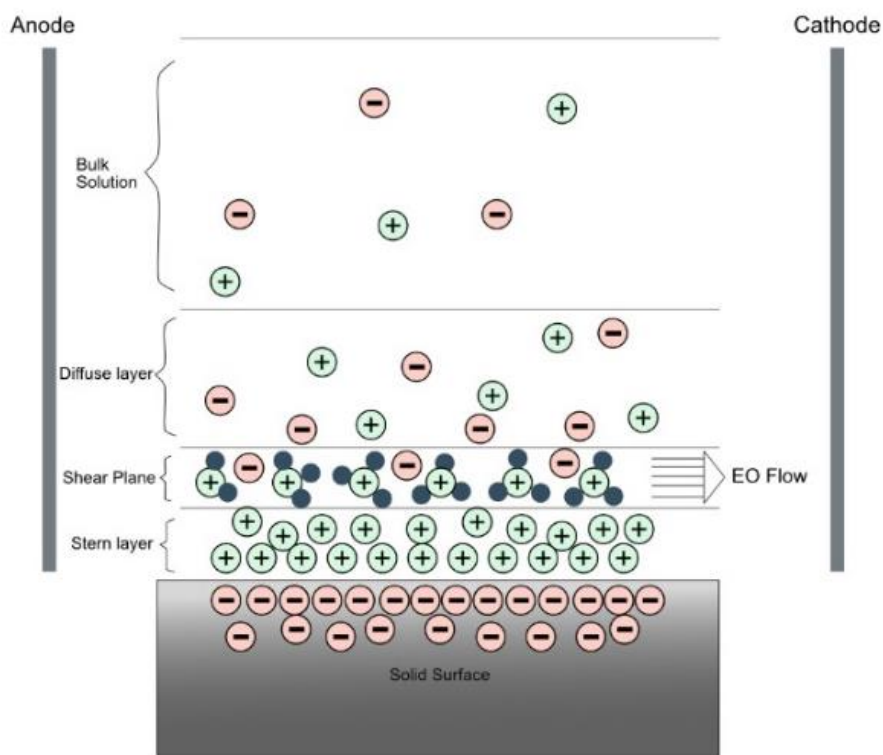
|              |                                 |                                                                                             |
|--------------|---------------------------------|---------------------------------------------------------------------------------------------|
| Constants    | Cross section of the brick cube | $A = 0.0009 \text{ m}^2$                                                                    |
|              | Length of brick cube            | $L = 0.03 \text{ m}$                                                                        |
|              | Volume in 5 cm capillary tube   | $4.81 \times 10^{-9} \text{ m}^3$                                                           |
|              | Volume in 10 cm capillary tube  | $9.6 \cdot 10^{-9} \text{ m}^3$                                                             |
| Measurements | Voltage                         | $U \text{ [V]}$                                                                             |
|              | Current                         | $I \text{ [A]}$                                                                             |
|              | Time for 5 cm                   | $t_5 \text{ [s]}$                                                                           |
|              | Time for 10 cm                  | $t_{10} \text{ [s]}$                                                                        |
| Calculations | Potential gradient              | $\nabla (-\psi) = U/L \text{ [V/m]}$                                                        |
|              | Current density                 | $J_q = I/A \text{ [A/m}^2\text{]}$                                                          |
|              | Electrical conductivity         | $L_{22} = J_q/ \nabla (-\psi) \text{ [S/m]}$                                                |
|              | Flow of water                   | $J_{v,5} = V_5/(A \cdot t_5) \text{ and } J_{v,10} = V_{10}/(A \cdot t_{10}) \text{ [m/s]}$ |
|              | coefficient for EO              | $L_{12} = J_w/ \nabla (-\psi) \text{ [(m}^2\text{)/(V}\cdot\text{s)]}$                      |

**Table 2:** Properties of the Red and Yellow bricks.

| Parameters                                              | Red   | Yellow |
|---------------------------------------------------------|-------|--------|
| Water-accessible porosity (% v/v)                       | 23.4  | 30.6   |
| Mercury accessible porosity (% v/v)                     | 30.9  | 37.9   |
| Capillary porosity (% v/v)                              | 6.5   | 8.3    |
| Capillary Coefficient ( $\text{kg/m}^2\text{s}^{0.5}$ ) | 0.60  | 0.51   |
| Water absorption coefficient (under vacuum) (% w/w)     | 16.1  | 19.9   |
| Water absorption coefficient (by capillarity) (% w/w)   | 10.4  | 15.7   |
| Total anisotropy, dM (%)                                | 3.68  | 7.78   |
| Zeta potential (mV)                                     | -36.0 | -32.4  |
| Dry density ( $\text{g/cm}^3$ )                         | 2.31  | 1.95   |
| Saturated density ( $\text{g/cm}^3$ )                   | 2.68  | 2.34   |

**Table 3:** Intruded mercury volumes for different pore size ranges (both in ml/g and %).

|              |                              | <b>Pore size ranges (µm)</b> |       |        |        |      |
|--------------|------------------------------|------------------------------|-------|--------|--------|------|
| <b>Brick</b> | Total intruded volume (ml/g) | <0.1                         | 0.1-1 | 1-10   | 10-100 | >100 |
| Red          | 174.42                       | 3.80                         | 17.25 | 93.19  | 59.70  | 0.48 |
| Yellow       | 232.82                       | 3.21                         | 74.51 | 146.98 | 7.35   | 0.77 |
| <b>Brick</b> | Total intruded volume (%)    | <0.1                         | 0.1-1 | 1-10   | 10-100 | >100 |
| Red          | 100.00                       | 2.18                         | 9.89  | 53.43  | 34.23  | 0.28 |
| Yellow       | 100.00                       | 1.38                         | 32.00 | 63.13  | 3.16   | 0.33 |





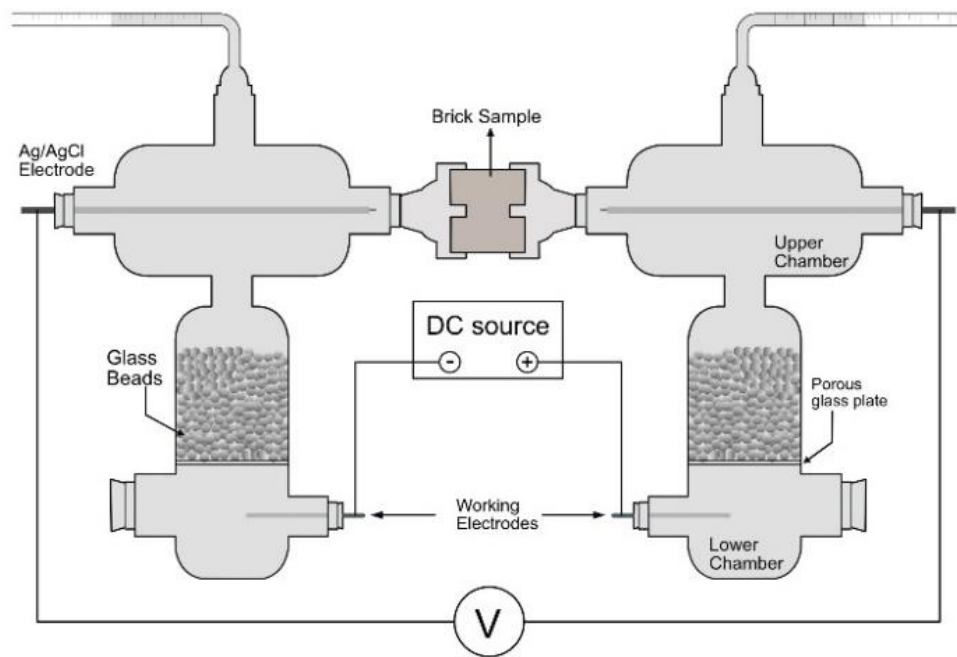


**Figure 3.** The schematic drawing of the electro-osmotic setup used.

