

Electroosmotic flow through particle beds packed with conditioned sludges

Ching-Jung Chuang, Pan-Wei Wang, Che-Chia Hu and Kuo-Lun Tung

ABSTRACT

Electroosmotic flow through packed beds filled with kaolin slurries conditioned with various chemicals (KCl, alum and polymeric flocculant etc.) was experimentally studied. A theoretical expression relating electroosmotic flux to physicochemical properties of the bed was derived and agreed with experimental data at low KCl concentration. When slurries were coagulated with high alum dosages without pH adjustment, a threshold electric field strength commenced electroosmosis flow, and a hysteresis phenomenon in the electric field strength dependent on electroosmotic flow were observed. However, when slurries were conditioned by adding polymeric flocculant, both the electroosmotic flux and its flow direction were qualitatively consistent with the prediction based on the measured values of zeta potential of particles.

Key words | coagulation, electroosmotic flow, flocculation, packed bed, sludge bed

Ching-Jung Chuang (corresponding author)
Pan-Wei Wang
Che-Chia Hu
Kuo-Lun Tung
R&D Center for Membrane Technology and
Department of Chemical Engineering,
Chung Yuan Christian University,
Taoyuan 320, Taiwan
Fax: +886 32654199
E-mail: cjchuang@cycu.edu.tw

NOMENCLATURE

- d diameter of the capillary [m]
 d_e equivalent diameter of flow channel in packed bed [m]
 d_p diameter of particle [m]
 D dielectric constant of bulk fluid [$F m^{-1}$]
 E electric field strength [$V m^{-1}$]
 q_e electroosmotic flux through bed [$m^3 m^{-2}.s$]
 \bar{v} average velocity of flow in a channel [$m s^{-1}$]
 ζ zeta potential [V]
 μ fluid viscosity [$kg.m.s^{-1}$]
 ε porosity of the bed [–]
 κ Debye constant [$1/m$]
 Φ_s sphericity of particles [–]

INTRODUCTION

In solid–liquid separation processes, sludges hard to dewater such as colloidal or gelatinous materials, are usually chemically conditioned, generally using coagulants and/or flocculants, to improve their settling and/or subsequent mechanical dewatering. But the dewatering

problem experienced during filtration or expression processes often remains due to very low hydraulic permeability, and may even be magnified because flocculation generally reduces the solid content to a very low extent for mechanical dewatering techniques. Further, considerably expensive thermal drying is required to obtain the desired final moisture content. This implies that any method for dewatering these sludges to a higher efficiency is of interest.

One method is applying a direct current (DC) electric field to induce electroosmosis and enhance sludge dewatering (Lockhart 1983a, b; Buijs *et al.* 1994; Smollen & Kafaar 1995; Dussour *et al.* 2000; Yoshida *et al.* 2001). According to the Helmholtz-Smoluchowski model for large capillaries with respect to double layer thickness, electroosmotic flow induced in sludge beds is generally considered independent of flow channel size. Therefore, this technique is particularly attractive in achieving a substantial increase in dewatering rate and greater solid content for fine-particles and gelatinous sludges difficult to dewater by conventional methods. The classical Helmholtz-Smoluchowski model indicates that electroosmotic flow in porous media is dependent on physicochemical properties such as the zeta

potential and ionic strength. Since these sludge properties are significantly affected by added chemical compounds, chemical pretreatments adopted in most solid–liquid separation processes might play a vital role in filtration and dewatering stages if assisted by applying electric fields.

Experimental results from Lockhart (1983a, b) in electroosmotic dewatering of sand and coal washing tailings, has shown that both salt concentration and types of exchangeable cation in the sludges are of significant influence in dewatering. However, non-ionic flocculants do not impair electroosmosis, but usually have an adverse effect on energy requirements. With respect to flocculation pretreatment, Smollen & Kafaar (1995) pointed out experimentally that using polyelectrolyte is not necessary to increase the final solid content for electroosmotic dewatering; however, it may decrease energy requirement in some cases. On the contrary, a study presented by Buijs *et al.* (1994) indicated that flocculation can accelerate electroosmotic dewatering, making it more economically attractive when flocculants used can increase both size and absolute value of aggregate zeta potential. Orsat *et al.* (1999) conducted the dewatering of a vegetable sludge by a combined electroosmosis and pressure action, and found that the addition of NaCl, MgCl₂ or CaCl₂ electrolytes does not increase the total water removed compared with that of a non-treated sludge. In a study on the influence of chemical pretreatment of kaolin suspensions on both filtration and electroosmotic dewatering stages carried out successively, Dussour *et al.* (2000) revealed that: (1) the addition of electrolytes leads to a liquid volume increase obtained during the electroosmotic dewatering stage, but the duration of dewatering is somewhat longer when compared with the process without additives; and (2) the addition of flocculant can markedly decrease the filtration time and increase the volume of filtrate in the filtration stage; however, it is less efficient in decreasing the energy consumption on the electroosmotic dewatering stage. In contrast, surfactants can increase the electroosmotic rate and decrease energy consumption during the electroosmotic dewatering stage.

Although numerous studies have been conducted concerning chemical pretreatment effects on sludges and subsequent electroosmotic dewatering, very few have explicitly been concerned with the role of zeta potential of particles in the processes and, thus, a current understanding

of the principles of such processes is very limited. A detailed examination of electroosmotic flux through kaolin particle beds pretreated by adding various chemicals (KCl, alum and polymeric flocculant) was performed in this study. Emphasis was placed on understanding chemical dosage effects, particle zeta potential and electric field strength on electroosmotic flux through the slurry beds.

THEORETICAL EQUATIONS

Electroosmotic flow induced by applying an electric field across a capillary has been formulated in a variety of ways. By considering electrokinetic phenomena dependence in fine capillaries on the electrical double-layer overlap, the average electroosmotic velocity expression in a cylindrical flow channel of diameter, d , has been obtained by Kobayashi *et al.* (1979) and Rice & Whitehead (1965):

$$\bar{v}_e = -\frac{D\zeta E}{\mu} \left(1 - \frac{I_1(\kappa d/2)}{\kappa d I_0(\kappa d/2)} \right) = -\frac{D\zeta E}{\mu} F \quad (1)$$

where, D is the dielectric constant of the medium, ζ the zeta potential of the flow channel, E the electric field strength, κ the Debye Huckel parameter, and I the modified Bessel function of the first kind. The bracket term is generally considered as a correction factor, F , for the classical formulation based on the Helmholtz-Smoluchowski model, as $\kappa d > 50$ the values of F approach 1.0.

The fluid flow channels in a packed bed are generally regarded as a bundle of capillary tubes. To effectively replace the actual channels in a packed bed, an equivalent diameter d_e of a set of identical parallel tubes is usually determined using the definition of the hydraulic radius, r_H :

$$d_e = 4r_H = \frac{2}{3} \left(\frac{\varepsilon}{1 - \varepsilon} \right) \Phi_s d_p \quad (2)$$

where, ε denotes the porosity of the particle bed and d_p and Φ_s are the size and sphericity of the particles, respectively. Substituting the equivalent diameter into the Hagen-Poiseuille equation derived for the laminar flow through a capillary,

$$\bar{v} = \frac{d^2}{32\mu} \left(-\frac{dP}{dx} \right) \quad (3)$$

the flux for flow through packed bed at very low Reynolds number is obtained (McCabe *et al.* 2001),

$$q = \frac{\Phi_s^2 d_p^2}{72 k_p \mu (1 - \varepsilon)^2} \left(- \frac{dP}{dx} \right) = \frac{1}{k_p} (\varepsilon \bar{v}) \quad (4)$$

where k_p is a correction factor added to account for the fact that flow channels in beds are actually tortuous and not straight and parallel. The constant k_p is generally designated as the tortuous factor in packed beds and is dependent on porosity, particle shape and other factors. An empirical value of 150/72 for the constant has been used in the well-known Kozeny-Carman equation. Happel & Brenner (1965) have solved the Navier–Stokes equation for a creeping flow through packed beds and derived an expression for the constant k_p ,

$$k_p = \frac{\varepsilon^3}{(1 - \varepsilon) \left[\ln \left(\frac{1}{1 - \varepsilon} \right) - \frac{1 - (1 - \varepsilon)^2}{1 + (1 - \varepsilon)^2} \right]} \quad (5)$$

For a given packed bed, it is a reasonable assumption that both the electroosmotic flow and the hydraulic flow have the same tortuous factor. Therefore, similar to the expression in Equation (4), the electroosmotic flux through a packed bed is then given by:

$$q_e = \frac{1}{k_p} (\varepsilon \bar{v}_e) \quad (6)$$

where, \bar{v}_e is evaluated from Equation (1) using the equivalent diameter defined in Equation (2).

EXPERIMENTAL

The experimental system is shown schematically in Figure 1. The electrokinetic cell made of an acrylic cylinder of 4.0 cm diameter consists of three parts, two electrode chambers (a) and (b), and a section (c) for positioning the particle bed. Both electrodes were made of stainless steel plates and the solutions filled in both electrode chambers were the same as that within the sludge beds. Two voltage measurement ports, 0.5 cm apart, were installed on the wall in the bed section (c) and connected to a voltmeter. At each side of the particle bed, a filter medium (Whatman #2) was set up in contact with the bed surface and, on the outside surface of the filter medium, a perforated plate was placed to support the bed.

Kaolin particles (reagent-grade; from Nacalai Tesque Co., Japan) with a median size of 6.37 μm and sphericity of 0.7 were used in the electroosmotic experiments. The zeta potential of particles was determined by electrophoresis using Malvern Zetasizer (Malvern Instrument, Malvern, UK). In the slurry preparation, 5 wt% slurries were first washed twice in distilled water, and jar tests were used to pretreat the slurries by adding KCl or alum or flocculant

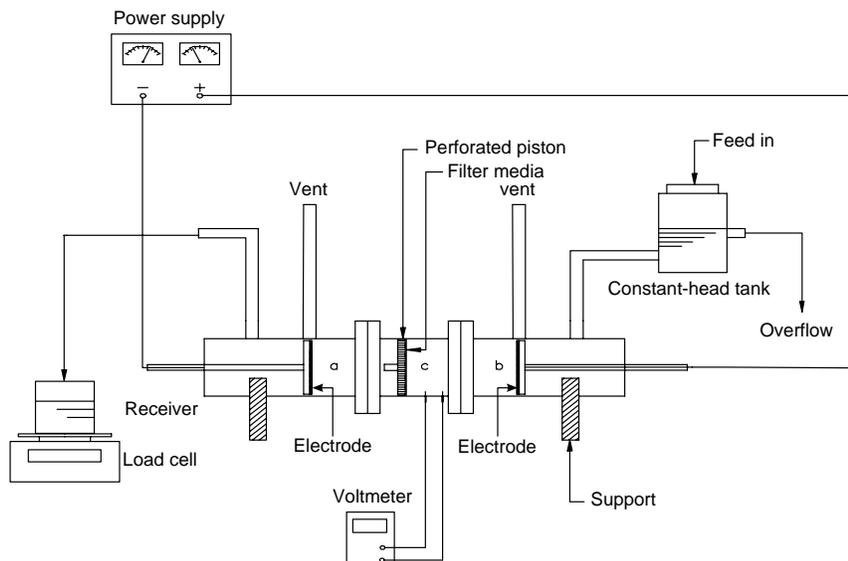


Figure 1 | Experimental set-up to measure the electroosmotic flow through the sludge bed.

(cationic polyacrylamide, $MW = 1.0 \times 10^7$) to form flocs. After settling of the flocs and then draining the sediment by gravity, the concentrated slurries were taken out to fill the section (c) of the measuring cell and compressed by a perforated piston to a given thickness. Based on the slurry amount and the bed thickness, the average packed bed porosity was calculated.

A constant-head tank filled with solution was installed to keep a hydraulic pressure equilibrium between its overflow and the permeate discharge from the bed. In the experiments, a constant electric field strength across the particle bed was controlled by regulating the voltage applied, via a DC power supply. The electroosmotic rate was determined by using a load cell to measure the volume of liquid discharged from chamber (a) within a 5 min lapse of running time after the start of electric field application.

RESULTS AND DISCUSSION

Effect of KCl concentration

By knowing the zeta potential values, the Debye constant and bed porosity, the electroosmotic flux through packed beds was predicted from Equation (6). Figure 2 compares the predicted values with the experimental data of the electroosmotic flux for kaolin particle beds pretreated with 10^{-4} M KCl. The electroosmotic flux increases with the applied electric field as well as the variance in the experimental data for the reagent-grade kaolin. Also included in the figure is a comparison of the experimental data from Chen (1999) who used industrial-grade kaolin along with the calculated values from Equation (6). It can be noted that, except under higher field strengths, both results agree fairly well.

Figure 3 shows electric field strength dependence on the electroosmotic flux through kaolin beds under KCl

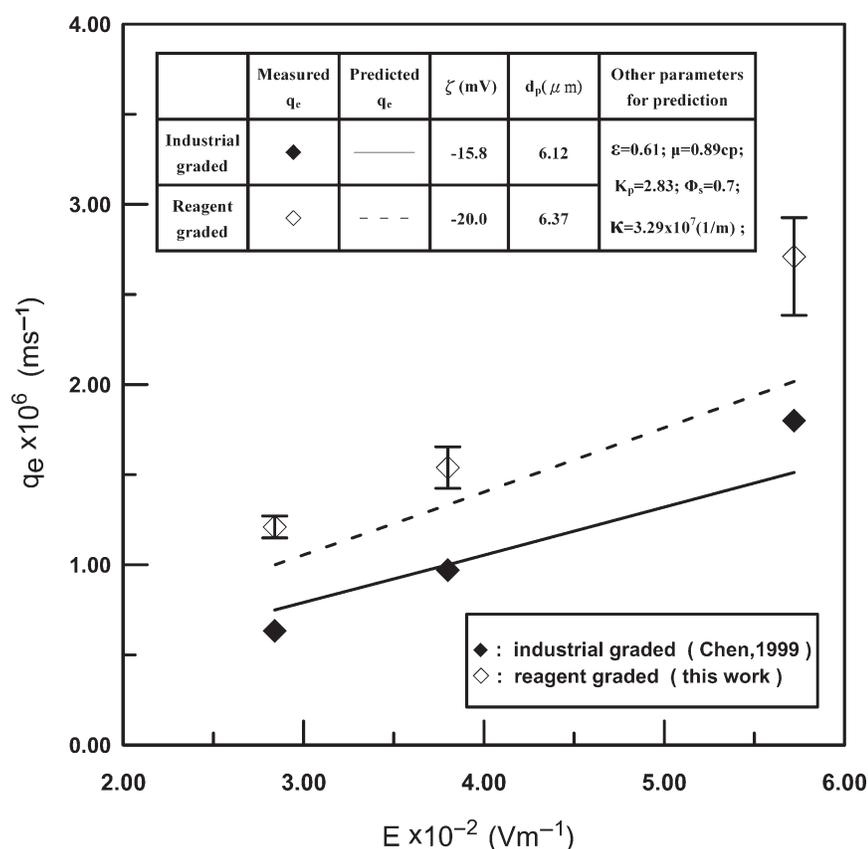


Figure 2 | Comparison between measured values and model predictions for the electroosmotic flux of kaolin beds at 10^{-4} M KCl. (Error bars depict the maximum and minimum flux from duplicated tests at each electric field strength.)

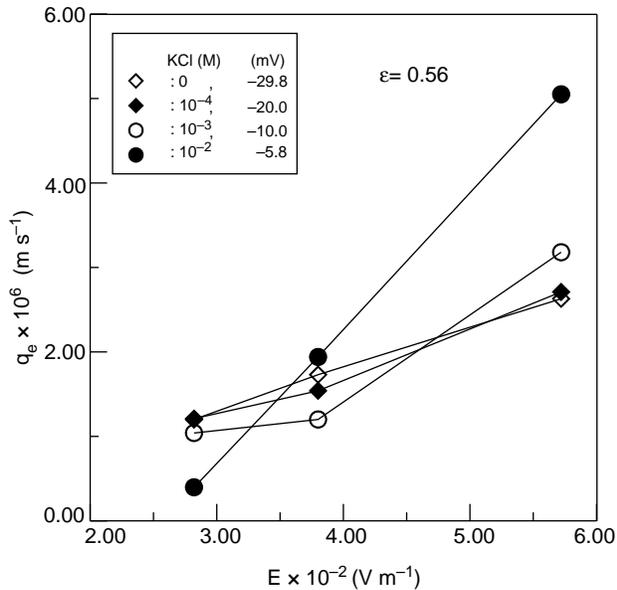


Figure 3 | Effect of KCl concentration on the electroosmotic flux of a kaolin bed.

concentrations ranging from 0 to 10^{-2} M. The corresponding zeta potentials (ζ) ranging from -29.8 to -5.8 mV are also listed in the figure. Based on the theoretical model of Equation (6), it is expected that electroosmotic flux will decrease with the increase of KCl concentration. However, this was only valid for low KCl concentrations ($<10^{-3}$ M) and low electric field strength ($E < 400 \text{ V m}^{-1}$). A reason for the unexpected result of a rapid increase in electroosmotic flux at high KCl concentrations, where zeta particle potential is very small, is that our experimental set-up is not suitable for cases with concentrated electrolytes in the measuring cell, which will result in a considerable amount of gas generated by electrolysis in the electrode chamber. The vent tube will be filled with foams and thus part of the liquid initially at the chamber (a) (see Figure 1) will be forced to flow into the permeate receiver. This will lead to an over measured flux, measured based on the liquid amount received, compared with the actual permeation rate through the bed.

Effect of alum dosage

The relation between electroosmotic flux and the strength of the electric field for alum conditioned kaolin slurries without pH adjustment are shown in Figure 4. As a low alum dosage such as $50 \sim 100 \text{ mg l}^{-1}$ is applied, the

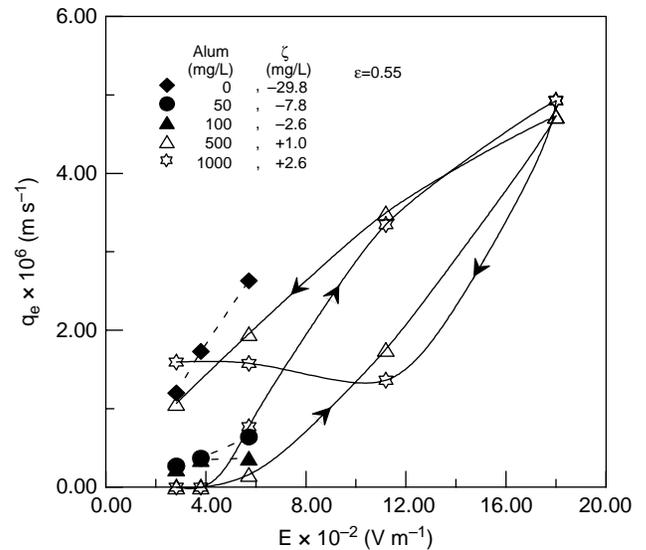


Figure 4 | Electroosmotic flux through bed packed with alum-coagulated slurry.

electroosmotic flux is much smaller than that without pretreatment. This is mainly due to the significant decrease of zeta potential of particles after pretreatment. As alum dosages are increased to $500 \sim 1,000 \text{ mg l}^{-1}$ ($\text{pH} = 3 \sim 4$), the ζ of the treated particles is very small and there is almost no electroosmotic flow at low electric field strength ($E < 400 \text{ V m}^{-1}$); that means a finite electric field strength (called threshold electric field strength) is needed to initiate the electroosmotic flow. As E applied is greater than the threshold strength, the flow rate increases steeply. Although zeta potential polarities are positive under high alum-dosage pretreatment, it is observed that electroosmotic flow is still directed to the cathode, the same as that without pretreatment in which ζ is negative. A hysteresis behaviour of electroosmotic flux versus electric field strength can be observed in Figure 4. It is noticed that, as the applied voltage is reduced from a specific value higher than the threshold strength, a considerable electroosmotic flux is still maintained even if E is decreased below the threshold strength. This phenomenon can be interpreted by the variation of streaming potential across the bed, since the streaming potential across porous media is proportional to the zeta potential of the flow channels (Benaventea & Jonsson 1998). In order to reveal why the electroosmotic flux can be maintained as the applied electric field reduced below the threshold value, the apparatus shown in Figure 1

has been modified to measure the streaming potential across the sludge bed which has gone through the electroosmotic run with high electric field strength. Results of measurement depict that the streaming potential of the sludge bed after exerting the electric field is quite different from that without imposing the external electric field. That means the electrokinetic properties of the bed are altered after a higher electric field strength has been exerted. Thus, the hysteresis phenomenon of electric field strength dependence on electroosmotic flux may result from the fact that some aluminium complexes adsorbed on particle surfaces are detached by the exerted electric field.

The electroosmotic flux of the alum conditioned sludge bed under similar conditions as depicted in Figure 4 but with pH adjustment was also conducted. Since the pH value of the alum conditioned sludge will decrease with the increase of alum dosage, a dosage of $1,000 \text{ mg l}^{-1}$ will result in a pH value of 3 which is too acidic for practical application in alum coagulation. It is more practical to carry out the coagulation at pH in the range of 6.0 to 7.0. (Pernitsky & Edzwald 2006). In order to examine the effect of pH adjustment on the electroosmotic flux of the alum conditioned sludge bed, the coagulation was adjusted to a pH value of 6.8 for analysis. Experimental results showed that, under this condition, no obvious threshold strength was observed and the electroosmotic flux relation against electric field strength was rather reversible. The higher the zeta potential, the larger electroosmotic flux was obtained. These results are quite different from that illustrated in Figure 4 for the slurry coagulated without pH adjustment.

Effect of polymeric flocculant dosage

Figure 5 shows the electroosmotic flux through beds packed with kaolin particles, flocculated by adding cationic polyacrylamide over a wide dosage range. The negative value of flux means that the electroosmotic flow is towards the anode. Several phenomena can be revealed from the results depicted in the figure: (1) electroosmotic flux is very small as the flocculant dosages close to the slurry zero-charged point; (2) electroosmotic flux increases with absolute zeta potential value; (3) the direction of electroosmotic flow depends on the zeta potential polarity of particles; and (4) no obvious threshold electric field

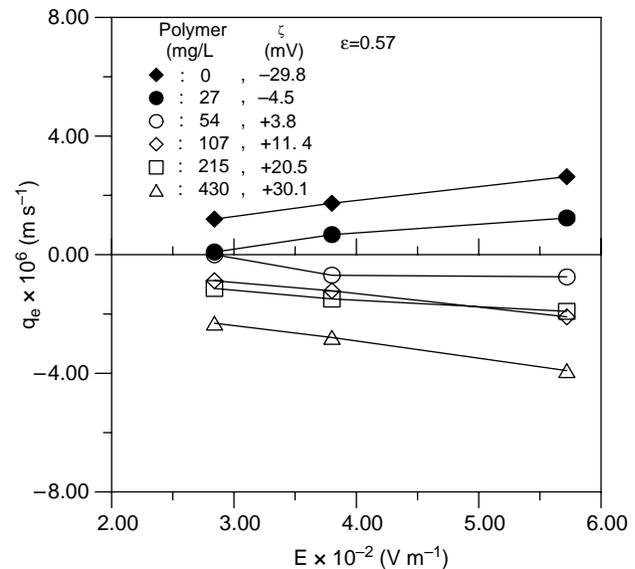


Figure 5 | Electroosmotic flux through bed packed with polymer flocculated slurry.

strength was observed. These results imply that both the magnitude and direction of electroosmotic flow in such beds are qualitatively consistent with the prediction based on the measured values of zeta potential of particles.

In practice, coagulation and flocculation processes are generally carried out successively in most chemical pretreatment operations to increase both size and strength of the flocs. The electroosmotic flux of sludge beds with such coagulation-flocculation pretreatments is illustrated in

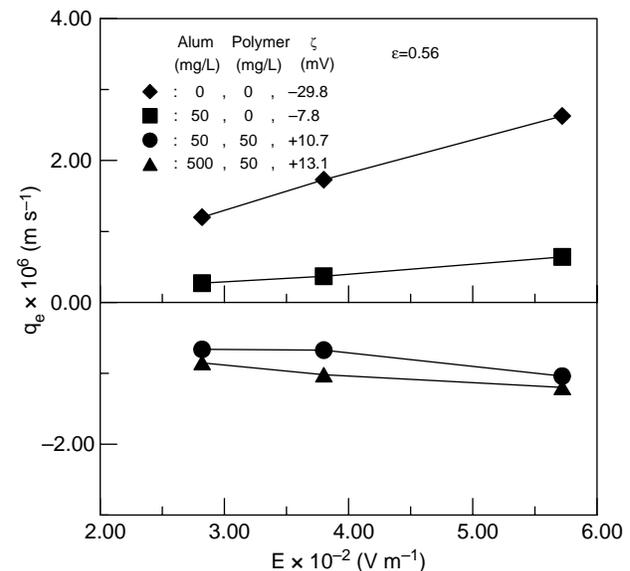


Figure 6 | Electroosmotic flux through bed packed with cationic flocculant conditioned alum-coagulated slurry.

Figure 6. It appears that, for the slurries dosed with both alum and polymeric flocculant, the electroosmotic flow direction is dependent on the zeta potential polarity of particles. Furthermore, it can be noted that no obvious threshold electric field strength was observed as in the case of pretreatment with polymeric flocculant only. Therefore, it is concluded that the electroosmotic behaviour of sludge beds with coagulation-flocculation pretreatment is similar to that with flocculation pretreatment.

CONCLUSIONS

The electroosmotic flow through packed beds filled with kaolin slurries under different chemical pretreatments has been investigated experimentally. A theoretical analysis has been proposed for relating the electroosmotic flux and the physical properties of the sludge bed. The measured values of electroosmotic flux agree with the theoretical predictions at low electrolytic concentrations. When the suspensions are coagulated with a high dosage of alum without pH adjustment, a threshold electric field strength is needed to commence electroosmotic flow. As the electric field imposed is greater than threshold strength, the flow increases rapidly. There is also a hysteresis phenomenon in the electric field strength dependence on electroosmotic flow, which is always directed towards the cathode, independent of the zeta potential polarity of the alum coagulated flocs. However, when the slurries are pretreated with cationic flocculant, both the rate and direction of electroosmotic flow are qualitatively consistent with the prediction based on the measured values of zeta potential of flocs. According to these results, it is concluded that chemical pretreatment is not only an important stage in the traditional mechanical separation process, but also plays an important role in subsequent filtration and/or dewatering processes if they are assisted by employing an electric field.

ACKNOWLEDGEMENT

The authors wish to express their sincere gratitude to the Ministry of Economic Affairs (MOEA), ROC, for the grant

from the Technology Development Program for Academia (TDPA) project and the National Science Council (NSC) and the Center-of-Excellence (COE) Program on Membrane Technology from the Ministry of Education (MOE), ROC, for their financial support.

REFERENCES

- Benaventea, J. & Jonsson, G. 1998 Electrokinetic characterization of a microporous non-commercial polysulfone membrane: Phenomenological coefficients and transport parameters. *Colloids and Surfaces, A: Physicochem. Eng. Aspect* **140**, 339–346.
- Buijs, P. J., Van Diemen, A. J. G. & Stein, H. N. 1994 Efficient dewatering of waterworks sludge by electroosmosis. *Colloids and Surfaces A: Physicochem. Eng. Aspects* **85**, 29–36.
- Chen, S. F. 1999 *Study on the Electroosmosis of Conditioned Sludges and its Dewatering by the Combined Action of Electric Field and Mechanical Expression*. MS thesis, Chung Yuan Christian University, Chungli, Taiwan.
- Dussour, C., Favoriti, P. & Vorobiev, E. 2000 Influence of chemical additives upon both filtration and electroosmotic dehydration of a kaolin suspension. *Sep. Sci. Technol.* **35**, 1179–1193.
- Happel, J. & Brenner, H. 1965 *Low Reynolds Number Hydrodynamics*. Prentice Hall, Englewood Cliffs, New Jersey, pp. 235–430.
- Kobayashi, K., Iwata, M., Hosoda, Y. & Yukawa, H. 1979 Fundamental study of electroosmotic flow through perforated membrane. *J. Chem. Eng. Japan* **12**, 466–471.
- Lockhart, N. C. 1983a Electroosmotic dewatering of clays II. Influence of salt, acid, and flocculants. *Colloids and Surfaces* **6**, 239–251.
- Lockhart, N. C. 1983b Electroosmotic dewatering of clays III. Influence of clay type, exchangeable cations, and electrode materials. *Colloids and Surfaces* **6**, 253–269.
- McCabe, W. L., Smith, J. C. & Harriot, P. 2001 *Unit Operations of Chemical Engineering*, 6th edition. McGraw-Hill, New York.
- Orsat, V., Raghavan, G. S. V., Sotocinal, S., Lightfoot, D. G. & Gopalakrishana, S. 1999 Roller press for electro-osmotic dewatering of bio-materials. *Drying Technol.* **17**, 523–538.
- Pernitsky, D. J. & Edzwald, J. K. 2006 Selection of alum and polyaluminum coagulants: Principles and applications. *J. Wat. Suppl.: Res. & Technol.-AQUA* **55**(2), 121–141.
- Rice, C. L. & Whitehead, R. 1965 Electrokinetic flow in a narrow cylindrical capillary. *J. Phys. Chem.* **69**, 4017–4024.
- Smollen, M. & Kafaar, A. 1995 *Development of Electroosmotic Sludge Dewatering Technology*, Report to the Water Research Commission by the Division of Water Technology, CSIR, South Africa (No. 412/1/95).
- Yoshida, H., Tanaka, K. & Komatsu, M. 2001 Influence of on and off times of power application on electro-osmotic dewatering under intermittent electric field. *Filtration* **2**(1), 27–32.

First received 15 March 2006; accepted in revised form 20 May 2006