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Pulsed Electrophoretic Membrane Cleaning for Dead-End Ultrafiltration Process

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ABSTRACT

Membrane separation technology has been identified as a new and clean technology and as an alternative to the conventional separation technology since it can offer some advantages over those separation technologies. Nevertheless, membrane fouling appears to be the major bottleneck of this technology. Many approaches have been proposed to reduce membrane fouling. The approach considered in this paper is the application of electric pulse to remove deposits from the membrane surface. An experimental test rig is constructed to study the effect of electric pulse on the membrane cleaning effectiveness. The filter cake is collected after every application of each electric pulse to ensure that the deposited materials which has been removed from the membrane surface after each electric pulse will not be redeposited onto the membrane surface. The dead-end ultrafiltration of silica dioxide (SiO₂) dispersion is conducted with various values of pulse interval, pulse duration and feed concentration. It is found that the application of the electric pulse to reduce membrane fouling and restore high permeation rate is effective in the dead-end filtration process. The application of electric pulses across the membrane gives a higher permeation rate than the conventional ultrafiltration process.

Keywords: membrane separation, membrane cleaning, fouling, ultrafiltration, electric pulse

ABSTRAK

Teknologi pemisahan membran telah dicam sebagai suatu teknologi yang baru dan bersih dan merupakan suatu alternatif kepada teknologi pemisahan lazim kerana teknologi ini menawarkan beberapa kelebihan berbanding teknologi-teknologi pemisahan yang lain. Walau bagaimanapun, membran yang kotor dan tersumbat merupakan masalah utama bagi teknologi ini. Banyak cara telah dicadangkan untuk mengurangkan masalah membran yang tersumbat ini. Cara yang dipertimbangkan di dalam kertas ini ialah penggunaan denyutan elektrik bagi menyingkirkan enapan daripada permukaan membran. Suatu alat uji kaji dibina bagi mengkaji kesan denyutan elektrik terhadap keberkesanan pencucian membran. Kek turasan dikeluarkan selepas setiap denyutan elektrik supaya kek itu tidak terenap semula di atas permukaan membran. Ultraturasan hujung mati bagi penyerakan silika dioksida (SiO2) dijalankan dengan beberapa nilai julat denyutan, masa denyutan dan kepekatan suapan. Kajian ini mendapati bahawa penggunaan denyutan elektrik adalah berkesan bagi mengurangkan kejadian membran tersumbat dan mengekalkan kadar ketelapan yang tinggi bagi proses

ultraturasan hujung mati. Penggunaan denyutan elektrik merentas membran menghasilkan kadar ketelapan yang tinggi berbanding dengan proses ultraturasan lazim.

Katakunci: pemisahan membran, pembersihan membran, pengotoran, ultraturasan, denyutan elektrik

INTRODUCTION

Fouling is one of the major drawbacks in the membrane separation process. Many have attempted to use electric fields continuously (conventional electrofiltration) to control the membrane fouling and filter cake formation and few instances can be seen from the work of Henry et al. (1977), Yukawa et al. (1983), Bowen et al. (1984), Wakeman and Tarleton (1987) and Visvanathan and Ben (1989). It has been shown that by applying the electric fields, the deposit on the membrane surface can be reduced effectively. This approach is successful because it utilizes the inherent surface charge of dispersed materials when brought into contact with a polar (e.g. aqueous) solvent such as water that arises from a combination of mechanisms including ion dissociation, ion adsorption or ion dissolution.

The electrochemical properties of the membrane surface and the dispersed materials or solute can have a significant influence on the nature and magnitude of the interaction between the membrane and the substances being processed and their separation characteristics (Iritani et al. 1992). The utilization of such properties by the application of the external electric fields can potentially give substantial improvement in the performance of separation. In particular, such process makes use of two electrokinetics phenomena. The first phenomena is electrophoretic, the transport of charged surface relative to a stationary liquid by an electric field. An example of this includes the movement of ions or particles between electrodes which occurs when an electric field is imposed on a dispersed system (ion or particles). The schematic representation of this electrophoretic transport of a negatively charged particle is shown in Figure 1.

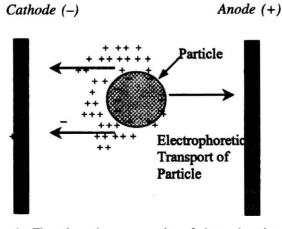


FIGURE 1. The schematic representation of electrophoretic transport of a negatively charged particle

The second electrokinetics phenomena is electroosmosis, the transport of the liquid relative to an immobile charged surface by an electric fields. An example of this category is the movement of an electrolytes solution relative to a membrane pore or filter cake under the influence of potential gradient (Hunter 1991). The schematic representation of electroosmotic transport through a single pore of negatively charged membrane is shown in Figure 2.

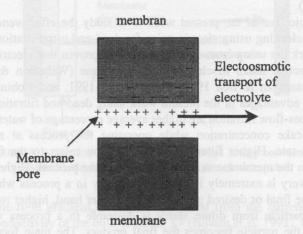


FIGURE 2. The schematic representation of electroosmotic transport through a single pore of negatively charged membrane.

Both electrokinetic and electroosmosis are similar phenomena. The only distinction between them is on the frame of reference either solid or liquid. The magnitude of both phenomena depends on the applied electric field gradient and the zeta potential (electric potential at the particle's shear plane between the mobile and immobile parts of the double layer).

The conventional electrofiltration, which applies electric fields continuously, unfortunately has some drawbacks which renders the method uneconomical and difficult to handle for certain processes. Among the disadvantages of this process are:

- 1. The limitation of the process stream or relatively low conductivity since it requires electrolyte for current to flow between two electrodes
- 2. A high-energy requirement
- 3. Substantial production of heat
- 4. Changes in the process feed (conductivity) due to reaction at the electrode

For this reasons, attention has been directed to use of pulsed electric fields as suggested by Bowen et al. (1991/2), Ahmad (1997), Bowen and Ahmad (1997), and Ahmad et al. (1999/2000). This process has the same mechanism at work in preventing fouling as conventional electrofiltration. The distinction is that the application of electric field is at certain intervals, which can be adjusted to suit the process. The electrically enhanced membrane process has been applied to electrically conducting membranes, such as stainless steel microfilter which pushed the foulant materials out into the

feed stream termed in-situ electrolytic membrane cleaning, IEMC (Bowen et al. 1989). This process does not depend on the surface charge of the dispersed materials for its primary action. It is applicable to process stream of a wide range of conductivity. The process can also improve the permeation rate by using nonconducting polymeric or organic membrane (Wakeman & Tarleton 1987, and Bowen et al. 1987). An important advantage of such process is that they eliminate the need of high cross-flow velocity (Bowen et al. 1991).

The objective of the present work is to study the effectiveness of the membrane cleaning using electric pulse for dead-end ultrafiltration process. Similar work but using cross-flow filtration has proven that electric pulse is a very effective mean in cleaning the membrane (Wakeman & Tarleton 1987; Visvanathan & Ben 1989; Bowen et al. 1991; and Robinson et al. 1993). The advantages of the present work using dead-end filtration process over the cross-flow filtration are higher average percentage of water recovery and filter cake concentration while operating the process at acceptable permeation rate. Higher filter cake concentration refers to the filter cake collected on the membrane surface at the end of the process. Higher average water recovery is extremely important especially in a process where water becomes the final or desired product. On the other hand, higher recovery of dispersed particle from dilute slurry is desirable in a process where the amount of the particle becomes the final product. The main focus of this work is to study the effect of operating conditions such as pulse interval, pulse duration and feed concentration on the effectiveness of this cleaning technique.

FORCE BALANCE ANALYSIS

A model for an electric-field pulse in a dead-end filtration system is developed based on balance of forces that act on a particle at the surface of the filter cake with slight modification from the established model for cross-flow filtration by Bowen and Sabuni (1992). The modification of the model for the dead-end filtration from the work of Bowen and Ahmad (1997) and Ahmad et al. (1999) is adapted in this study. The analysis is also extended to provide an analytical expression that predicts the rate or filtration expected immediately after the application of an electrical pulse. Figure 3 shows a schematic diagram of all the forces acting between the electrodes during the application of the electric pulse.

The criterion for the removal of a particle from the surface of the filter cake during the application of an electric field pulse is given as:

$$F_{y} + F_{e} < 0, \tag{1}$$

where, F_y is the viscous drag on the particle due to the flow of permeate and Fe is the electrophoretic force. Positive sign refers to a force acting toward the membrane.

The electrophoretic force may be written

$$F_e = -fu_p E = -fu_p I / \lambda \circ \tag{2}$$

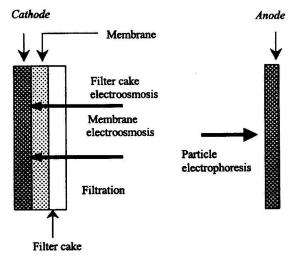


FIGURE 3. Schematic diagram of all the forces acting between the electrodes during the application of the electric pulse

The electric-field gradient, E can be calculated from the current density, I and the conductivity of the bulk solution, λ + (Bowen et al. 1992). Calculation of the viscous force is more complex, as the effects of electroosmosis in the membrane and the filter cake must be considered (Henry et al. 1977). The force is calculated from

$$Fy = f\Delta P/\mu (R_m + R_c) \tag{3}$$

 R_m and R_c are estimated as follows (Bowen et al., 1992),

$$R_m = R_{om}/(1 + v_{eom} I/v_{om}) \tag{4}$$

and,

$$R_{c} = R_{oc} / [1 + (u_{p} I / \lambda_{o} V_{oc})]$$
 (5)

When an electric field is applied, particles are removed from the filter cake and the filtration rate increases. This removal process continues until the hydrodynamic and electrical forces become balanced. Thus, the criterion for no further removal of particles is,

$$F_{y} + F_{e} = 0 \tag{6}$$

MATERIALS AND METHOD

Filtration experiment was carried out in a flat sheet dead-end filtration module. The overall dimensions of the process feed chamber were 11.2 cm x 3.4 cm x 0.2 cm with an effective membrane area 23 cm². The feed chamber refers to the column inside the filtration module where the feed is accumulated. The filtration module on the other hand refers to the overall membrane filtration unit. The membrane was supported on a stainless steel

sheet of 200 μ m pore size, which was in turn supported on a stainless steel mesh. Both the sheet and the mesh formed the cathode, while the anode was a stainless steel sheet mounted flush with the other side of the process feed chamber. The schematic diagram of the experimental set up is shown in Figure 4. The system reservoir held up to 5 L of feed and a centrifugal pump would drive the filtration process at a constant pressure of 100 kN/m².

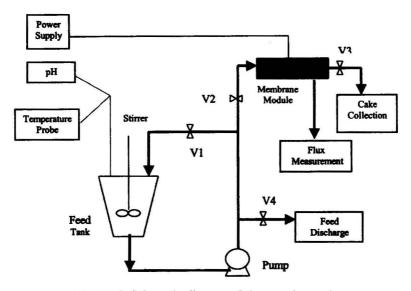


FIGURE 4. Schematic diagram of the experiment rig

The power supply used to provide electric pulses to the membrane module was obtained from Farnell Instrument Limited. The unit could supply voltage up to 60V. It was linked directly to the membrane surface via a negative electrode. This power supply uses switching technique that results in a fast rise time (0.6s) and fall time (1.0s). The flux was calculated based on time taken to collect a volume of permeate. In the operation of the filtration membrane module, a valve on the concentrate side (V3) was opened manually after the application of each electric pulse to allow collection of the filter cake. For every experiment, the dispersion was recycled for about 10 minute by opening valve V1 and closing valve V2 to make sure that the particles are very well dispersed before starting the filtration process.

The membrane used was a Millipore membrane with a molecular-weight cut-off (MWCO) of 30, 000 (Millipore Cat No HATF 202 00). The electrolyte used in this experiment was 0.01M Potassium Nitrate, KNO₃. The feed system used was Silica Dioxide (SiO₂) dispersed in 0.01M KNO₃ as the electrolyte. SiO₂ was chosen due to its natural rigidity in that its physical and chemical properties do not change with electric pulse. Dilute NaOH and H₂SO₄ was used to maintain the pH. Since the electrophoretic mobility of particle might change with pH, changing pH might change the electrophoretic force resulting change in the particle size of the feed system(Bowen and Ahmad, 1997). It is therefore imperative to maintain pH at a common value (pH 8 in this work). A pH probe and thermometer were

inserted into the feed tank to monitor the pH and the temperature throughout the experiment. Voltage strength of 60V was applied for all the experiments. All the experiments were run for 180 minutes.

The effectiveness of this membrane cleaning method was tested by changing 3 experimental conditions; the pulse interval, the pulse duration and the feed concentration at pH 8 and with ionic strength (electrolyte) of 0.01M KNO₃. In the first experiment, the pulse interval was varied for 20, 30, 45, and 60 minutes at constant pulse duration and feed concentration of 10 seconds and 1 g/L respectively. In the second experiment, the pulse duration was varied for 5, 10, 20 and 40 seconds at constant pulse interval and feed concentration of 20 minutes and 1 g/L respectively. In the subsequent experiment, the feed concentration was varied in the order of 0.1, 0.5, 0.8 and 1.0 g/L at constant pulse interval and pulse duration of 20 minutes and 10 seconds respectively.

RESULTS AND DISCUSSION

EFFECT OF PULSE INTERVAL

Typical results on the effect of pulse interval at 1 g/L dispersion, 10 second pulse duration and 60V applied voltage on the average flux is shown in Table 1. When the pulse interval was increased from 20 to 60 minutes, the average flux decreases from 100.29 to 56.27 L/m²h respectively. The longer the pulse interval, the more SiO₂ particle would be deposited on the membrane surface before any membrane cleaning take place. In other words, longer pulse interval means the membrane being cleaned at longer frequency, which resulted in lower flux.

Voltage	Concentration (gL ⁻¹)	Pulse Interval (min)	Pulse Duration (s)	Average Filtration Flux (Lm ⁻² h ⁻¹)
60	1.0	20	10	100.29
60	1.0	30	10	74.27
60	1.0	45	10	64.22
60	1.0	60	10	56.37

TABLE 1. Variation of pulse interval

Figure 5 shows the normal filtration of 0.5 g/L SiO₂ dispersed in 0.01M KNO₃ without any cleaning of the membrane surface. The graph shows how the flux varies with time. It is clearly shown that the flux keep on decreasing continuously with time throughout the experiment. This is due to the deposit of SiO₂ particles on the membrane surface, which slowly block and foul the pores of the membrane resulting in the reduction of the flux with time.

Figure 6 and 7 show the plot of flux with time at different pulse interval. Figure 6 shows that the lowest flux for 20 minutes interval is slightly higher compared to 30 minutes interval. Most of highest flux after each cleaning

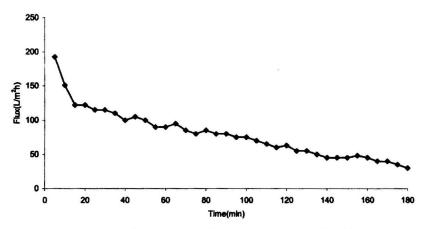


FIGURE 5: Flux vs. time for normal filtration(0.5 g/L)

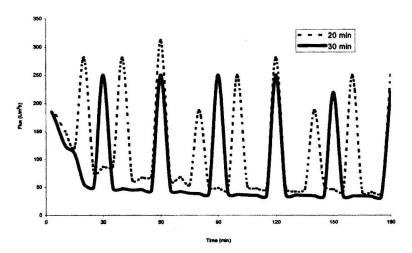


FIGURE 6. Flux vs. time at 20 and 30 min pulse interval (60V, 10s pulse duration, 1g/L, SiO_2)

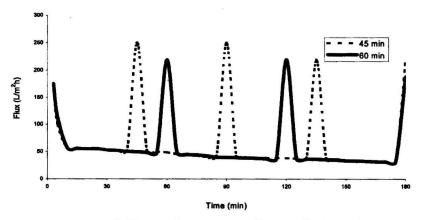


FIGURE 7: Flux vs. time at 45 and 60 min pulse interval (60V, 10s pulse duration, 1g/L, SiO_2)

interval is higher for 20 minutes compared to 30 minutes pulse interval. Whereas, Figure 7 shows that the lowest flux and the highest flux for both 45 minutes and 60 minutes are quite comparable.

It is well known that membrane fouling is one of the main phenomena responsible for the decline in flux with time. The other major factors include adsorption and chemical interactions. The occurrence of fouling affects the performance of the ultrafiltration either by deposition of a layer onto the membrane surface or by blockage or partial blockage of the pores. This changes the effective pore size distribution. After long periods of time, internal pore blocking occurs within the pores. Internal pore blocking is a particularly serious fouling mechanism, even a small amount of adsorption can lead to a considerable change in the membrane's rejection characteristics (Scott & Hughes 1996). When applying electric pulse, after a certain period of filtration process, it can remove particles that build up on the external layer of the membrane but difficult to reduce the internal pore blocking by the particles. Thus the average filtration fluxes decrease when the pulse interval is increased.

Typical results for the variation of pulse duration for 1 g/L dispersion with 20 minutes pulse interval and 60 V applied voltage is shown in Table 2. When the pulse duration is increased from 5 to 40 seconds, the average flux is decreased from 107.93 to 91.3 L/m²h. The particles need to be removed at certain distance from the membrane surface in order to be able to be swept out from the membrane module at the end of each application of the pulse (every 20 minutes). The results show that 5s pulse duration is good and long enough to move the particle to the minimum distance away from the membrane surface since increasing the pulse duration resulted in decrease in flux. Obviously, longer pulse duration result in increase of resistance for the permeate to flow. At this stage, the particles are also experiencing forward force by viscous drag due to the pressure driven flow and the forces arising from electroosmosis in the cake and in the membrane as shown in Figure 3. During the application of electric pulse to the membrane surface, the filter cake will be removed from the membrane surface, which is placed on the negative electrode (cathode) and moves towards the positive electrode (anode). This surface charge or electric charge gives the particle of the cake, electrophoretic mobility to move away from the surface of the membrane. When this happens, the surface of the membrane was cleaned resulted in the increase of permeation rate. When the

TABLE 2. Variation of pulse duration

The second secon							
Voltage	Concentration (gL ⁻¹)	Pulse Interval (min)	Pulse Duration (s)	Average Filtration Flux (Lm ⁻² h ⁻¹)			
60	1.0	20	5	107.93			
60	1.0	20	10	100.29			
60	1.0	20	20	93.75			
60	1.0	20	40	91.30			

application of electricity to the surface is terminated, the cake particles will lose its electrophoretic mobility. The filter cake starts to build up on the membrane surface and cause membrane fouling.

EFFECT OF PULSE DURATION

With longer pulse duration it seems that the properties of the filter cake might have changed, on the application of longer pulses, in such away as to produce an increase in the resistance to fluid flow. This may be due to the changes in the morphology of firmly adhering layers produced by the action of electric pulses (Bowen et al. 1992). Local changes of pH and conductivity in the membrane module due to the application of longer pulses could produce changes in surface chemistry of SiO₂ particles that might lead to such effects.

Figure 8 and 9 show the relationship of the flux with time at different pulse duration. The permeate rate decreases continuously during the course of experiment as the SiO₂ filter builds up on the membrane surface. When pulses were applied, and the filter cake was released for collection, the

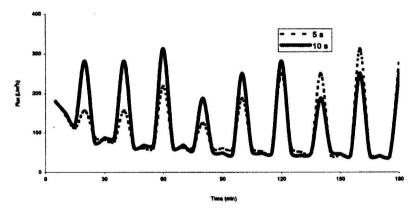


FIGURE 8. Flux vs. time at 5 and 10s pulse duration (60V, 20min pulse interval, 1g/L SiO₂)

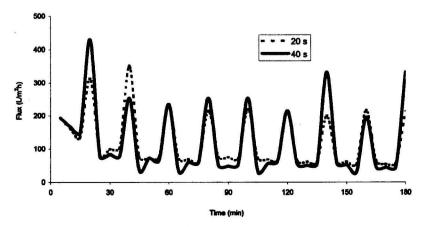


FIGURE 9. Flux vs. time at 20 and 40s pulse duration (60V, 20min pulse interval, 1g/L SiO₂)

permeation rate increased as obviously shown in those Figures. The pulses were applied at every pulse interval chosen and as shown at every low point of the saw teeth pattern of those Figures.

EFFECT OF FEED CONCENTRATION

Table 3 shows the effect of variation of feed concentration at constant pulse interval of 20 min., 10s pulse duration and magnitude of voltage of 60 V. As feed concentration is increased from 0.1 to 1.0 g/L, the average filtration flux decreases from 231.19 to 100.29 (L/m,h). This is because when the feed concentration is increased, number of particles that contains in the solution is also increased. Higher concentration will result in the increase of particles in the solution whereas the electrophoretic mobility is still at the same rate. This means, the force which removes the particles away from the membrane surface, is getting smaller relative to the amount of particles. Therefore, not all the particles can be removed from the membrane surface (Ahmad et al. 1999). Since higher concentration feed was used, probability of concentration polarization to occur will become greater and thus the permeation rate (flux) will become lower. This can be described based on the rejection of higher molar mass solutes by the membrane; their concentration in the fluid boundary layer adjacent to the membrane surface can be much greater than that in the bulk solution. This accumulation of high molar mass solutes on the membrane surface is called 'concentration polarization'. Its occurrence will determine, to a large extent, the efficiency of the membrane processes. Concentration polarization may reduce the net permeate flux due to osmotic pressure effects and will increase the resistance due to fouling, which further impede the flow of permeate through the membrane.

TABLE 3. Variation of concentration

Voltage	Concentration (gL ⁻¹)	Pulse Interval (min)	Pulse Duration (s)	Average Filtration Flux (Lm ⁻² h ⁻¹)
60	0.1	20	10	231.19
60	0.5	20	10	220.12
60	0.8	20	10	120.51
60	1.0	20	10	100.29

Figure 10 and 11 show the relationship of the flux with time at different feed concentration. Figure 10 shows the comparison of flux for 0.1 and 0.5 g/L respectively. It shows that the initial flux and the lowest flux are much less for 0.5 g/L than 0.1 g/L. It also shows that for the first 2 hours, its highest flux after cleaning process is higher for 0.1 g/L than 0.5 g/L feed concentration. After 2 hours, the highest flux for 0.1 g/L than 0.5 g/L feed concentration is quite comparable. Figure 11 shows the comparison of flux for 0.8 and 1.0 g/L respectively. For the lowest flux, Figure 11 shows result

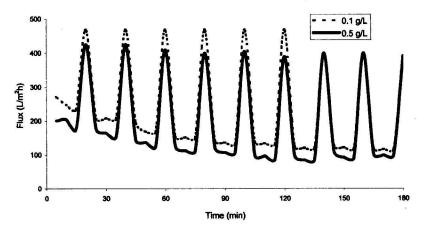


FIGURE 10. Flux vs. time at 0.1 and 0.5 g/L concentration (60V, 20min pulse interval, 10s pulse duration)

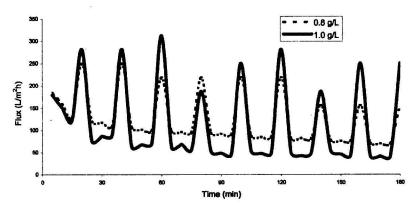


FIGURE 11: Flux vs. time at 0.8 and 1 g/L concentration (60V, 20min pulse interval, 10s pulse duration)

with similar trend as in Figure 10. The highest flux is less for 0.8 than 1 g/L except at about 80 minutes where the highest flux is less for 1 than 0.8 g/L.

CONCLUSION

It has been discovered that the application of the electric pulse to clean the membrane surface is effective in minimizing the presence of membrane fouling, thus, enabling high permeation rate to be restored. The application of electric pulses across the membrane has resulted in an increase of permeation rate immediately after each pulse was activated. In a long run, this technique can potentially improve recovery of the desired product. The results of this work can be utilized to identify the best operating condition for the most effective membrane cleaning in the dead-end module using pulse. In summary, high average filtration flux from dead-end cleaning employing electric pulse was attained when the pulse interval decreased, pulse duration decreased and feed concentration decreased.

ACKNOWLEDGEMENT

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NOTATIONS

f = friction factor, N s m⁻¹

= ionic strength, M, in the tables.

 R_c = Hydraulic resistance of the filter cake, m⁻¹ R = hydraulic resistance of the membrane, m⁻¹

 R_{\parallel} = hydraulic resistance of the filter cake in the absence of an electric field, m⁻¹

 v_{ac} = flux through the cake in the absence of an electric field, m⁻¹

 v_{∞} = flux through the membrane in the absence of an electric field,

m s⁻¹

 $v = \text{membrane-specific electrophoretic flow rate, m s}^{-1}$

 $\lambda_{\rm o}^{\rm o}$ = bulk electrolyte conductivity, S m⁻¹

 μ = electrolyte viscosity, N s m⁻¹

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