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Application of a Pulsed Electric Field to Cross-flow Ultrafiltration of Protein Solution

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The application of pulsed electric field was investigated in the crossflow ultrafiltration of BSA (bovine serum albumn) to economize the application time of electric current as well as to avoid inherent problems of long-term application of electric field. During the application of various cyclic patterns of pulsed electric current, the averaged filtration flowrate and the degree of concentration were maintained higher than those obtained in the absence of electric current application. The temperature increase, pH change, and BSA loss by electrodeposition were all negligible during the operation. The averaged filtration flowrate increased as the ON/OFF duration ratio of electric current was higher and as the period of ON/OFF cycle was shorter. The re-establishment of concentration polarization was dependent to the duration of current OFF state and, therefore, a longer duration of OFF state was not favorable in maintaining higher filtration flow rate. Although the averaged filtration flowrate was enhanced as the magnitude of electric current increased, the flowrate enhancement became smaller as the magnitude of current increased because there exists a current value above which the degree of electrokinetic depolarization is no further improved.

Key words: crossflow ultrafiltration, concentration polarization, bovine serum albumin, pulsed electric field

INTRODUCTION

Ultrafiltration is being widely used to concentrate proteins and to remove small molecular solutes. The efficiency of ultrafiltration is limited by concentration polarization which is the accumulation of rejected molecules on the membrane surface [1,2]. Concentration polarization gradually decreases filtration flux and alters rejection characteristics of the membrane. The most common solution to this polarization problem is crossflow filtration technique which works by creating high shear on the membrane surface in order to reduce polarization layer. However, vigorous crossflow is not an ultimate solution because some biological materials will be deactivated in the high shear field [3,4].

One of the promising ways of controlling the concentration polarization in crossflow filtration without creating high shear force is using an electric field. An electric field is applied to pull the charged molecules accumulated in the polarization layer in a direction opposite to the pressure-driven convective flow [5,6]. The most critical factor determining filtration efficiency in concentrating protein solution in the presence of an electric field is the operation pH and net charge of the molecule there [7].

Although the application of electric field is very effective in reducing concentration polarization and in maintaining the desired filtration flux, it has several disadvantages including a high energy requirement, temperature change due to the Joule heating, chemistry change of the process solution due to electrode reactions, and electrodeposition of protein molecules on electrode surface [7,8]. Because these problems become severe when the duration of an electric field application is continuous and long, attentions have been recently directed to the use of pulsed electric field to economize the application of electric field and therefore to avoid such problems. Wakeman and Tarleton [9] were the first who pointed out that pulsing the electric field could reduce power consumption and produce filtration flux higher than that attainable with a constant electric field in a microfiltration of inorganic minerals. Bowen and Sabuni [10] utilized the pulsed electric field as a tool for cleaning microfiltration membrane fouled by titanium dioxide colloid. Kim and Lee [11] examined the applicability of the pulsed electric field in a crossflow ultrafiltration for the concentration of bovine serum albumin. The primary objective of this study is to investigate the effects of pulsed electric field on the concentration polarization and filtration rate in a crossflow ultrafiltration to concentrate a protein solution. The effects of different periods and ON/OFF duration ratios of electric pulse are discussed.

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MATERIALS AND METHODS

Apparatus and Materials

A crossflow membrane module was constructed in plexiglass for the study of ultrafiltration with and without an electric field (Fig. 1). Equal volumes of process feed chamber and permeate collecting chamber were divided by a membrane with effective working surface area of 9 cm^2. Platinum wire nets were

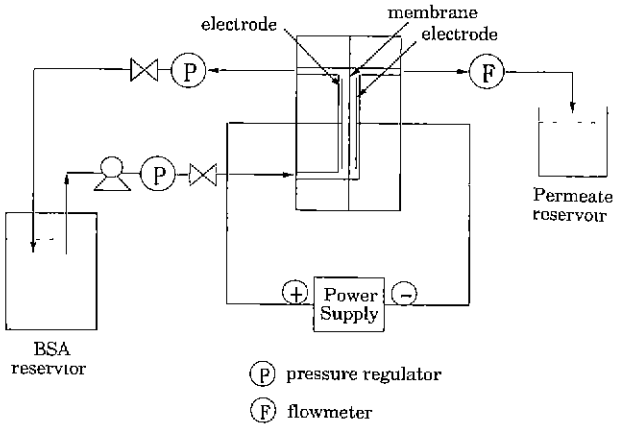

Fig. 1. Schematic experimental system for crossflow ultrafiltration in the presence of electric field.

mounted at both chambers as electrodes with 0.4 cm apart. Electric field of constant current was created by using a power supply EC570 from E-C Apparatus Corporation (New York, NY, U.S.A.). Amicon ultrafiltration membrane YM10 with nominal cutoff of 10,000 molecular weight was used. YM10 is a regenerated cellulose membrane which is hydrophilic and is known that nonspecific binding of proteins are negligible according to the manufacturer. Separation barrier was not used between electrode and process solution in order to prevent an excessive Joule heating and conductivity loss due to the barrier itself.

The molecular weight and pI of BSA (Sigma) are known about 67,000 and 5.4, respectively. BSA solutions were prepared by dissolving in Trizma buffer (Sigma) with minimal agitation. BSA concentration was determined with a Shimadzu 2401 UV spectrophotometer (Japan). Temperature was maintained at 20℃ throughout all experiments.

Experiments and Membrane Cleaning

Crossflow ultrafiltration was carried out to concentrate BSA in aqueous solution. Process feed chamber was initially filled with BSA solution at pH 8 and permeate chamber with blank buffer solution at pH 8. A centrifugal pump was used to drive filtration. Filtration flowrate was measured with a liquid flowmeter. Since the pI of BSA was 5.4, net charge of BSA at pH 8 would be negative and thus its direction of electrophoretic migration was towards anode when the direction of electric field is as in Fig. 1. The pore surface of YM10 membrane is known carrying a weak negative zeta potential in aqueous solution. Therefore the direction of electroosmotic flow was towards cathode and its flowrate was estimated less than 0.1 mL/min below 20 mA [12]. The filtration flowrates during the experiments were designed to be maintained high enough so that the contribution of electroosmosis could be negligible.

Once a run of ultrafiltration experiment was finished, membrane cleaning procedures were conducted to minimize the membrane fouling and to reuse the membrane. After discharging the residual solutions in both chambers and refilling with fresh buffer solution, reversed pressure was applied for 4 hr at 50 mL/ min and then reversed electric field was additionally applied for 30 min. This cycle was repeated until the UV absorbance at 280 nm fell to the level of pure buffer solution. Generally two or three cycles were enough to complete membrane cleaning in this study.

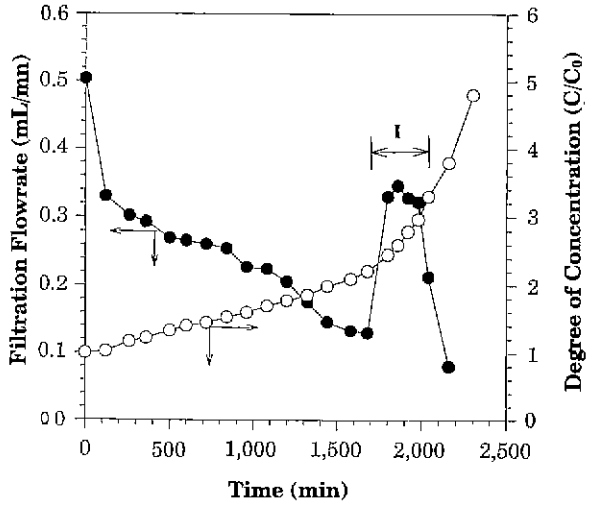

Fig. 2. Effect of electric current on filtration rate and the degree of concentration. Initial BSA concentration C_o= 0.5 g/L; pH=8; ΔP=1.2 bar; electric current I=10 mA; crossflow rate, 50 mL/min. 'I' in the plot represents the period of electric current application.

RESULTS AND DISCUSSION

Effect of Application of Electric Field

Fig. 2 shows how the application of electric field affected filtration flowrate when concentration polarization had been already proceeded. Ordinary ultrafiltration of BSA feed solution was carried out at the beginning. BSA solution with initial concentration of 0.5 g/L at pH 8 was circulated between reservoir and electrode chamber with a crossflow rate of 50 mL/min. The measured pressure difference was 1.2 bar. The filtration rate was 0.5 mL/min initially and was gradually reduced to 0.14 mL/min within 1,500 minutes due to the establishment of concentration polarization. Then an electric current of 10 mA was applied for 5 hr, from 1,700 min to 2,000 min. During the application of electric field, filtration rate was increased fast and recovered to 0.35 mL/min within 1 hr. This is because the direction of electrophoretic migration of BSA was opposite to that of convective fluid flow in the system shown in Fig. 1. BSA was negatively charged at pH 8 and the polarity of electric field shown in Fig. 1 made BSA migrate towards anode from the layer of concentration polarization on the membrane surface. After the electric current was turned off at 2,000 min, filtration rate decreased again rapidly.

The degree of concentration in Fig. 2 was defined as the ratio of the BSA concentration in reservoir at time t (C) to the initial BSA concentration (C_o), 0.4 g/L. In the absence of electric field, the degree of concentration was increased up to 2 for 1,500 min. Meanwhile, the degree of concentration was higher than 3 after 300 min of electric field application, which implies that the change of the degree of concentration in BSA reservoir was accelerated during the electric field application. This is because BSA molecules depolarized away from the membrane surface were circulated to the reservoir and thus filtration flux of solvent

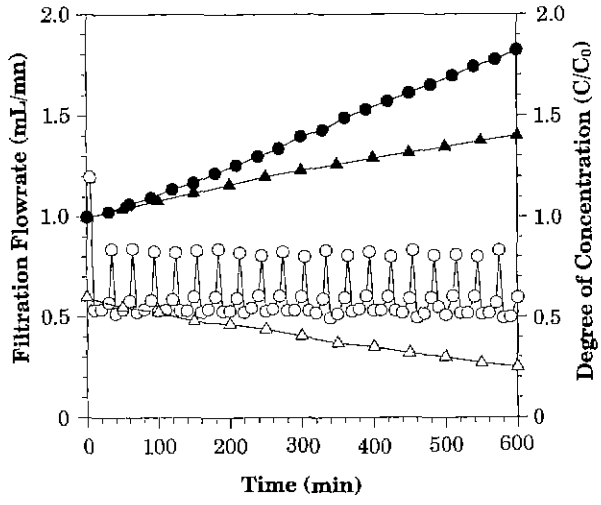

Fig. 3. Effect of pulsed electric current application. Initial BSA concentration C_o=0.5 g/L; pH=8; ΔP=1.2 bar; electric current I=10 mA; cycle of pulsed electric current, 5 min ON /25 min OFF; crossflow rate, 50 mL/min. ●, degree of concentration with pulsed electric current; ○, filtration flowrate with pulsed electric current; ▲, degree of concentration without electric current; △, filtration flowrate without electric current.

through the membrane increased.

During the 300 min of electric current application, some problems were observed. Firstly, the reservoir temperature started rising after 1 hr approximately at a rate of 0.6℃/min due to the Joule heating. Secondly, electrodeposition of BSA molecules at anode surface was began observed after 4 hours. The total 4.8% loss of BSA was estimated by material balance after the completion of experiment. Thirdly, the pH of system changed due to the electrode reactions. Lastly, filtration flowrate was slowly lowered to a level below 0.3 mL/min at the end of electric current application. The reason for the decrease of filtration rate in continuous application of electric field was not clear, but it was assumed that first three phenomena affected unfavorable in a coupled manner. These observations implied that a long-term continuous application of electric field was not desirable and thus optimization of application time would be required to minimize heating and electrodeposition in this system. In this study, the usage of a pulsed electric field was investigated to minimize such problems in the electrokinetic control of the concentration polarization.

Effect of Pulsed Application of Electric Field

From the beginning of ultrafiltration with 0.5 g/L of BSA solution at pH 8 and with 50 mL/min of crossflow rate, a pulsed electric field was applied for 5 min every 30 min (5 min ON/25 min OFF) with constant electric current of 10 mA. In Fig. 3, the results of flowrate and the degree of concentration were compared to those obtained without the application of electric field. During the 600 min of operation with the pulsed electric field, filtration rate was oscillated between 0.5 mL/min and 0.85 mL/min, resulting in about 0.6 mL/min on average, and the degree of concentration was increased to 1.8. The temperature change, pH change, and BSA loss by electrodeposition were all negligible for 600 min. Meanwhile in the absence of

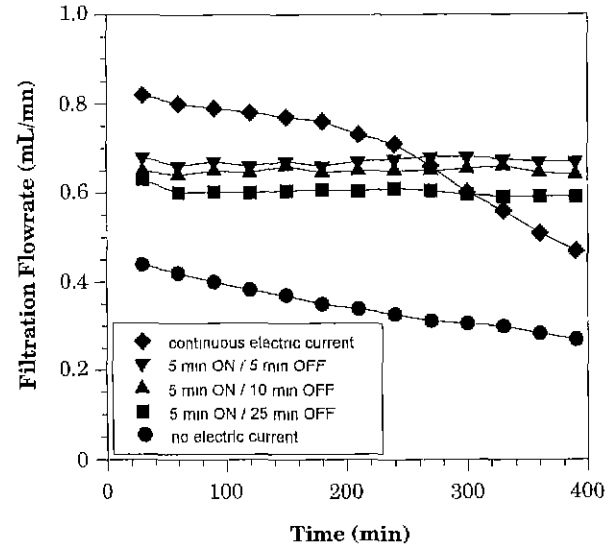

Fig. 4. Averaged filtration flowrates under pulsed electric current with different ON/OFF duration ratios. Initial BSA concentration C_o=0.5 g/L; pH=8; ΔP=1.2 bar; electric current I=10 mA; crossflow rate, 50 mL/min.

electric field, filtration rate decreased gradually from 0.6 mL/min to 0.25 mL/min and the degree of concentration was only 1.4 after 600 min. Results in Fig. 3 confirmed that the application of pulsed electric field could be effective in controlling concentration polarization and thus in maintaining stable filtration rate without the problems of heating and electrodeposition.

Effect of Different Pulse Cycles

To economize the use of electric power consumption, effects of different periods of current pulse on filtration rate were examined. In Fig. 4, the duration of current application was kept constant for 5 min, and three different pulsing cycles were used. Electric current of 10 mA was applied for 5 min every 30 min (5 min ON/25 min OFF), 15 min (5 min ON/10 min OFF), and 10 min (5 min ON/5 min OFF). Other experimental conditions were the same as in Fig. 3. The curves in the Fig. 4 represent the filtration flowrate averaged on each cycle of periods. Fig. 4 shows that the averaged filtration rate increased when the ON/OFF duration ratio was high, which meant that longer duration of current OFF state was disadvantageous in controlling polarization and in maintaining higher filtration rate. Fig. 5 shows the effect on filtration rate of different pulsing periods with an identical ON/OFF duration ratio. Experimental conditions were the same as in Fig. 4. Application of an electric pulse with shorter period of ON/OFF cycle showed increased filtration flowrate even though ON/OFF duration ratios were all identical. Another set of three different pulsing periods with a duration of current OFF for 10 min was carried out (data not shown), where 10 mA was applied for 5 min every 15 min (5 min ON/10 min OFF), for 10 min every 20 min (10 min ON/10 min OFF), and for 15 min every 25 min (15 min ON/10 min OFF). These three cases showed little difference for 600 min of operation. We could conclude that the duration of current OFF state was more critical in controlling the concentration polarization, and it was maybe because re-esta-

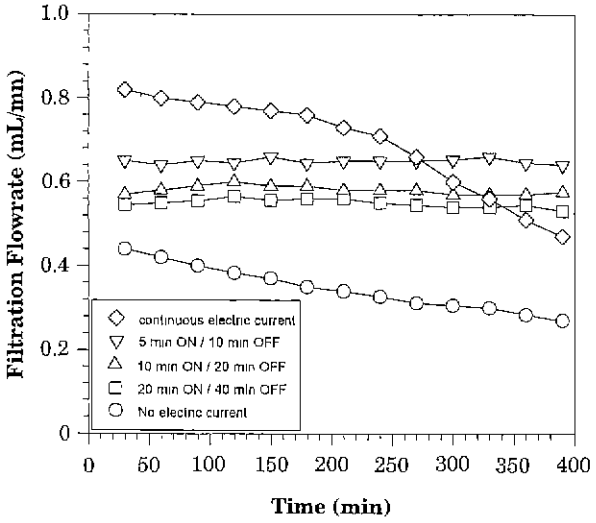

Fig. 5. Averaged filtration flowrate under different pulsing periods with identical ON/OFF duration ratio. Other conditions are the same as in Fig. 4.

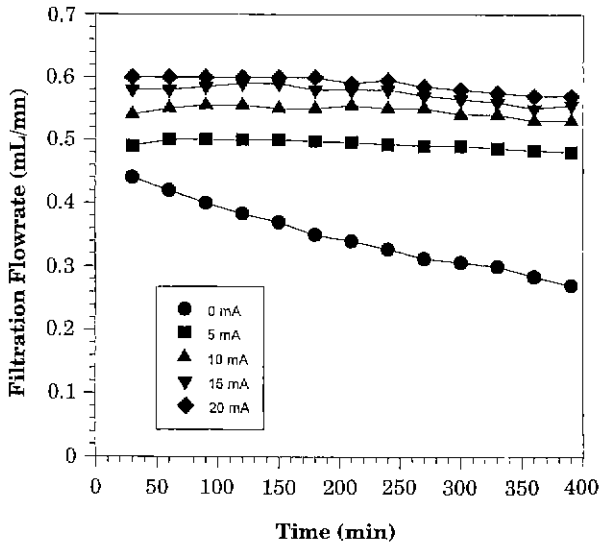

Fig. 6. Effect of the magnitude of electric current on averaged filtration flowrate. The cycle of pulsed electric current, 10 min ON/20 min OFF. Other conditions are the same as in Fig. 4.

blishment of concentration polarization was dependent on the duration of the OFF state and thus re-depolarization by current ON became less effective as the duration of OFF increased.

In both Figs. 4 and 5, the initial filtration flowrates for the first 5 min from $t=0$ (the first ON state of electric current) of 10 mA application were all identical as 0.85 mL/min, except the case of no electric current where the flowrate was 0.48 mL/min. The filtration rates with 10 mA began showing differences one another right after ON state of the first ON/OFF cycle ended because the ON/OFF duration ratios were all different.

In Figs. 4 and 5, continuous application of electric field showed the highest filtration rate initially, but filtration rate was slowly decreased as time passed. As mentioned earlier, the decrease of filtration rate after a long application of continuous electric field would be the combined results of heating, pH change, and electrodeposition. It is possible that an excessive heating and pH change influenced the physicochemical properties of membrane, such as swelling and electroosmosis. Also, severe electrodeposition could reduce working area of electrode surface and thus made electrokinetic control of concentration polarization less effective. All these factors could act unfavorably against normal filtration.

Effect of Magnitude of Electric Current

Different magnitudes of constant electric current were applied for 10 min every 30 min (10 min ON/20 min OFF). Fig. 6 shows that the averaged filtration flowrate was enhanced as the magnitude of electric current increased. However, they were not linearly proportional. The flowrate enhancement was largest when current was raised from 0 mA to 5 mA. Subsequent increases by 5 mA showed that the flowrate enhancement became smaller as the magnitude of current was higher. This result implies that there might exist a current value above which the degree of electrokinetic depolarization is no further improved for a specific pulsed electrofiltration condition.

CONCLUSION

Although the concentration polarization phenomena in ultrafiltration processes can be controlled by using an electric field, a long term application of electric current evokes undesirable problems such as temperature increase, pH change, electrodeposition of solutes, and the resultant decrease of filtration flux. The application of pulsed electric field was investigated in the crossflow ultrafiltration of BSA to economize the application time of electric current as well as to avoid such problems. Effects of various ON/OFF cycles of pulsed current were compared. During the application of pulsed electric current, the averaged filtration flowrate and the degree of concentration were maintained higher than those obtained in the absence of electric current application. The temperature increase, pH change, and BSA loss by electrodeposition were all negligible during the operation. The averaged filtration flowrate increased as the ON/OFF duration ratio of electric current was higher and as the period of ON/OFF cycle was shorter. The re-establishment of concentration polarization was dependent on the duration of current OFF state and, therefore, a longer duration of OFF state was not favorable to maintain higher filtration flowrate. The averaged filtration flowrate was enhanced as the magnitude of electric current increased. However, the flowrate enhancement became smaller for the higher magnitude of current because there might exist a certain current value above which the degree of electrokinetic depolarization is no further improved.

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