



## Preparation and properties of electric-sensitive poly(vinyl alcohol)/poly(acrylic acid) IPN hydrogels as draw agent for forward osmosis

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### ABSTRACT

In this study, electric-sensitive poly(vinyl alcohol)/poly(acrylic acid) (PVA/PAAc) hydrogels were fabricated as draw agent for forward osmosis. By controlling the amounts of cross-linker, chemical cross-linked method integrated with freezing-thawing was adopted, and prepared hydrogels were denoted as PVA3M0.25, PVA3M0.5, PVA3M1.0 and PVA3M1.5. The mass retention ratio examination indicated that PVA/PAAc hydrogels were sensitive to subtle electric stimuli; thus, they could shrink and release water under non-contacted electric field. And swelling ratio experiment suggested that PVA3M1.0 had relatively higher swelling ratio. Swelling exponent  $n$  value was above 0.5, which indicated that the swelling process belonged to the non-Fickian diffusion and water absorbing mechanism was more complex. Besides, swelling-deswelling behavior analysis showed that hydrogels could restore the original state in the next swelling experiment when the hydrogels released water under non-contacted electric field. Thus PVA3M0.5 and PVA3M1.0 were selected as draw agent for forward osmosis; by using deionized water and 2,000 ppm NaCl solution as feed solution, the initial water fluxes obtained were 1.04, 0.72, 0.73 and 0.54 LMH, respectively, with hydrogels against the active layer of membrane. Therefore, using electric-sensitive hydrogels as draw agent can eliminate reverse salt leakage and reduce the complexity of the operation in forward osmosis process.

*Keywords:* Electric-sensitive; PVA/PAAc hydrogels; Forward osmosis; Draw agent

### 1. Introduction

Forward osmosis (FO), as novel membrane separation technology, has attracted more attention in the area of seawater desalination and wastewater reclamation. This process is the transport of water across a selectively permeable membrane from the feed solutions (FS) to the draw solutions (DS). Its driving force comes from the osmotic pressure difference across the membrane between the FS (low osmotic pressure) and the DS (high osmotic pressure). Consequently, compared with pressure-driven membrane processes like reverse osmosis (RO), it has obvious advantages such as minimal energy consumption, less fouling problems and high water recovery

[1,2]. However, the lack of an appropriate draw agent has constrained the development of FO. An ideal DS needs to fulfill the following requirements such as high osmotic pressure, minimal reverse diffusion, easy regeneration and small molecular weight [3].

Inorganic salts such as NaCl, MgCl<sub>2</sub>, KCl, MgSO<sub>4</sub>, KHCO<sub>3</sub>, Ca(NO<sub>3</sub>)<sub>2</sub> and NH<sub>4</sub>HCO<sub>3</sub> were considered to be conventional draw agents and widely used for the various FO processes [4,5]. In general, most inorganic salts produced reasonably high water fluxes and were easily separated by RO or nanofiltration. However, both energy consumption of RO and corresponding reverse salt leakage in FO were found to be high, leading to more operating costs and even contaminating FS. A number of chemicals have been explored as the draw agents for FO, such as sulfur dioxide (SO<sub>2</sub>) [6], glucose [7]

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and polyglycol copolymer [8].  $\text{NH}_4\text{HCO}_3$  was used for the FO desalination due to the high osmotic pressure, and it could be easily separated from the product freshwater through distillation at moderate heat ( $60^\circ\text{C}$ ) and saved energy [9,10], while it had some problems including an unpleasant odor, much higher reverse leakage and underlying health issues. Kravath and Davis [7] first chose glucose as draw agent in the FO process for seawater desalination and made some conclusions that glucose presented in the final product was edible and intended for drinking propose in emergency lifeboats. It avoided the regeneration of draw solute, but its application was limited and more consumption of glucose was unrealistic. Besides, using organic as draw agent could make biofouling much heavier in FO process [11]. Polyelectrolytes composed of a series of poly(acrylic acid) sodium salts (PAA-Na) were explored as the draw solutes by Ge et al. [12] in the FO process, and the literature concluded that they were promising as draw solutes because of the high water flux, insignificant salt leakage and the high salt rejection in recycle via an ultrafiltration process. Bai et al. [13] drew a conclusion that the dextran-coated  $\text{Fe}_3\text{O}_4$  magnetic nanoparticles were ideal draw solutes for FO desalination process because of its high osmotic pressure; then it was easily recovered from the diluted DS by an external magnet. But, the agglomeration of the particles after recovery restricted its applications, because it decreased the osmotic pressure and thus reduced the water flux [2]. Hence, how to select an appropriate DS is crucial for the process performance.

An ideal DS should have good compatibility with membrane, high osmotic pressure, low RO and non-toxic, and could be easily separated from the diluted DS. Recently, it is reported that stimuli-responsive polymer hydrogels has been employed as a new class of draw agent in FO process as the reverse salt diffusion can be avoided completely. Hydrogels can extract large volumes of water attracted by the hydrophilic groups and also have three-dimensional networks of polymer chains, which are cross-linked via either physical or chemical bonds [14]. Stimuli-responsive hydrogels undergo a reversible volume phase change when they are subjected to external stimuli such as temperature, pH, external magnetic field, ionic strength and light [15–19]. Li et al. [20] found that composite hydrogels with light-absorbing carbon particles (e.g., poly(sodium acrylate)-(N-isopropylacrylamide)-Carbon) produced higher water fluxes in the FO process, and the water recovery rates enhanced with the increasing loadings of carbon particles when sunlight radiation was applied to separate pure water from swollen polymer hydrogels. In addition, thermal-responsive semi-interpenetrating network hydrogels [21], bifunctional polymer hydrogel [22] and hydrogel particles [23] were prepared for the FO process to drive water through semi-permeable membrane, and the water could be released from swollen hydrogels by using temperature and solar irradiation as external stimuli. However, compared with other stimuli, electric-responsive hydrogels are easier to control and apply [24]. It was reported in 1965 that poly(vinyl alcohol) (PVA)-poly(acrylic acid) (PAAc) polymer gel had different elongation lengths under the positive and negative 5 V alternately [25]. Electric-sensitive hydrogels are usually made of polyelectrolytes and undergo shrinking or swelling in the presence of an applied electric field, which are able to convert chemical energy to mechanical work, so they have

been commonly applied in controlled drug delivery, muscular contraction, and flagellar and ciliary movement [26].

In the present study, we attempt to prepare intelligent hydrogels that is sensitive to electric stimuli. Using electric-sensitive polymer hydrogels as draw agent are less used compared with the other published literatures. Chemical cross-linked method integrated with freezing-thawing is adopted to prepare the interpenetrating polymer networks (IPN) hydrogels using PVA and PAAc, which are easy and cheap to obtain. The swelling ratio (SR), mass retention rate, repeated swelling-deswelling behaviors and the water flux are investigated to advance the FO performances.

## 2. Materials and methods

### 2.1. Materials

Acrylic acid (AAc, analytical reagents (AR),  $\geq 99.0\%$ ) monomer was purchased from the Tianjin Guangfu Fine Chemical Research Institute (Tianjin). PVA (molecular weight:  $1,750 \pm 50$ ,  $\geq 99.0\%$ ) monomer was provided by the Sinopharm Chemical Reagent Co., Ltd. (Shanghai). *N,N'*-methylenebisacrylamide (MBAAm, AR,  $\geq 99.0\%$ ), as a cross-linker, was purchased from the Tianjin Kermel Chemical Reagent Co., Ltd. (Tianjin). Ammonium peroxydisulfate (APS) and sodium bisulfate ( $\text{NaHSO}_3$ ), as the redox initiator, were obtained from the Tianjin Guangfu Technology Development Co., Ltd. (Tianjin) and Tianjin DaMao Chemical Reagent Factory (Tianjin), respectively. All other chemicals, which were of reagent grade, were employed without any further purification. The FO membrane used in this work was purchased from the Hydration Technologies Inc. (Albany, OR) and was denoted as CTA-FO. It was an asymmetric structure membrane, which was composed of cellulose acetate supported by an embedded polyester mesh.

### 2.2. Preparation of PVA/PAAc hydrogel

PVA/PAAc hydrogels were prepared by the chemical cross-linked method integrated with the freezing-thawing. Kim et al. [24] explained that an initial network of PAAc was prepared in a PVA solution by using UV irradiation, and PVA networks as a secondary network were formed by a repetitive freezing-thawing process. Unlike the reference, the redox initiator was adopted to spur an initial network of PAAc in a PVA solution. The synthesis routes of PVA/PAAc hydrogels are shown in Fig. 1.

In this study, PAAc was copolymerized in a PVA solution and generated the segment chain polymer containing MBAAm structure by using APS and  $\text{NaHSO}_3$  as the redox initiator and MBAAm as the cross-linker. It led to form the primary networks through intermolecular hydrogen bond. Then, via the hydrogen bond, the primary network and PVA formed the second network using freezing-thawing method. Repeated freezing-thawing process would strengthen the hydrogen bonding, and the two forces caused stable three-dimensional networks of hydrogels.

First, the desired amount of PVA solution was heated and stirred in  $80^\circ\text{C}$  water bath for 5 h to make it colorless and transparent. Second, the AAc monomer and MBAAm were added in the PVA solution with desired proportions

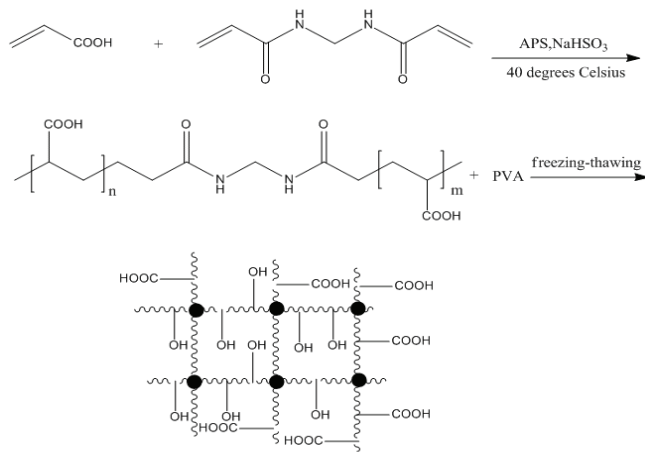


Fig. 1. Synthesis routes of PVA/PAAC hydrogels.

(AAc: PVA = 3:5 wt%) and mixed thoroughly when it was cooled to 40°C. The initiator content was 1 wt% of the monomers, and the content of cross-linker was 0.25, 0.5, 1.0 and 1.5 molar ratio of the monomers, respectively. The cross-linker had an effect on the number of cross-linked dots in the polymer network. Then the redox initiator APS and NaHSO<sub>3</sub> were added in the solution and stirred magnetically at the room temperature for 1.5 h. Finally, it was put statically in the air to remove air bubbles after the mixed solution was stirred; then it was poured into the mold and frozen in the refrigerator of -20°C for 15 h. Subsequently, the sample was removed and thawed at room temperature for 9 h. This process was repeated for five times to obtain the composite hydrogels, which had certain intensity and elasticity. Prior to use, the sample was soaked into the deionized water for 7 d, and the deionized water was changed every day.

### 2.3. Swelling properties of IPN hydrogel

To measure the SR, swollen hydrogels cut into the small pieces (3 cm × 2.5 cm × 0.4 cm) were dried in vacuum oven at 35°C for 8 h and weighed accurately. The sample was immersed in deionized water of 200 mL and taken out every 30 min at room temperature. Then it was weighed after the surface water of the hydrogel was removed by using the filter paper gently. The above steps were repeated until the weight was no longer increased (i.e., the swelling equilibrium). The SR was determined according to the following equation:

$$SR(g/g) = \frac{M_t - M_0}{M_0} \quad (1)$$

where  $M_t$  was defined as the mass of the hydrogel at a certain time  $t$ , and  $M_0$  was the mass of the semi-dry hydrogel.

The following exponential relation equation was introduced to describe the diffusion of water [27]:

$$SR(g/g) = \frac{M_t - M_0}{M_0} = Kt^n \quad (2)$$

And the form of Eq. (3) was obtained by calculating the logarithm of Eq. (2), where  $k$  is the swelling constant, and  $n$  is the swelling exponent calculated from the slopes of the  $\ln SR$  lines vs.  $\ln t$  plots. The  $t$  was the determination time of SR.

$$\ln SR = \ln K + n \ln t \quad (3)$$

### 2.4. The mass retention rate of IPN hydrogels

Swollen hydrogels were cut into the small pieces (as shown above), and its surface water was wiped and weighted. Then, it was laid in the measuring apparatus, which had a non-contacted electric field. Copper electrode was used, and its distance was 6 mm. It was removed from the device every 10 min and weighed after the surface water of the hydrogels was removed by using the filter paper softly. A generalized expression of the mass retention rate ( $R_w$ ) equation could be written as:

$$R_w(\%) = \frac{m_t}{m_0} \times 100\% \quad (4)$$

where  $m_t$  represented the mass of hydrogels at a certain time after non-contacted field was applied, and  $m_0$  indicated the mass of hydrogels before it was laid in the device.

### 2.5. Characterizations

Swollen polymer hydrogels were cut and dried by vacuum freeze-dryer. A scanning electron microscope (SEM; QUANTA 450, USA) was used to observe the surface morphology of PVA/PAAC hydrogels.

### 2.6. The water flux assessment

The prepared hydrogels were semi-solid substance and were kept in deionized water. Before FO test, the PVA/PAAC hydrogels were cut into the bulk material and subjected to a non-contacted electric field, which made the hydrogels shrink and release water. The mass of hydrogels was around 10 g. Then the hydrogels were put in the channel with the volume of 3 cm × 8 cm × 0.4 cm (width, length and height, respectively). And the hydrogel and membrane were firmly attached.

The water flux of electric-sensitive polymer hydrogels was examined by using a cross-flow FO membrane module with an effective membrane area of 24 cm<sup>2</sup>. The FS was pumped by a peristaltic pump at a volumetric flow rate of 200 mL/min (a cross flow velocity of 11.11 cm/s). The dehydrated hydrogel DS facing the different layers of CTA membrane (support layer [SL] and active layer [AL]) was investigated. Before each test in the homemade FO apparatus (Fig. 2), the membrane was immersed in deionized water in 1.5 h to maintain hydration. Water flux,  $F$ , in L/m<sup>2</sup>·h (LMH), was calculated from Eq. (5):

$$F = \frac{\Delta V}{A \times \Delta t} \quad (5)$$

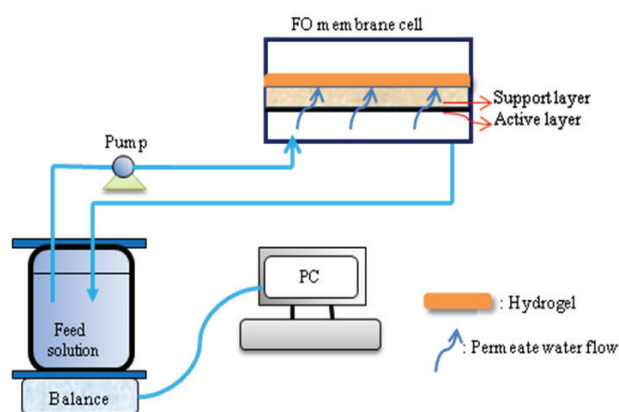


Fig. 2. Experimental setup diagram of the FO by using polymer hydrogels as draw agent.

where  $\Delta V$  indicated the volume of water permeated through the FO membrane (L), and it was based on the mass decrease of the FS at a certain time,  $\Delta t$  (h);  $A$  was the effective area of membrane. The water flux was measured for three times under the same testing conditions, and the mean value and the standard deviation were obtained.

### 3. Results and discussion

#### 3.1. Swelling behavior

To test the swelling property of the four kinds of hydrogels, the SR was investigated. As shown in Fig. 3, polymer hydrogels had faster SR in the first 300 min, with the time increasing, and the SR of four hydrogels grew slowly until the equilibrium was reached at the values of 0.67, 0.90, 1.12 and 0.88, respectively, for the increasing cross-linker amount. In conclusion, PVA3M1.0 hydrogels had better SR than others.

In addition, the literature [28] discussed swelling behavior of anionic copolymer gels and came to conclusion that for a planar geometry, the value of swelling exponent  $n = 0.5$  signified a Fickian water diffusion mechanism, while  $n = 1.0$  indicated a Case II diffusional mechanism. For  $0.5 < n < 1.0$ , the diffusional mechanism was non-Fickian, where both diffusion and polymer relaxation controlled the overall rate of water uptake. It could be seen in Fig. 4 that the values of  $n$  were 0.77, 0.79, 0.83 and 0.89, respectively, when the dose of cross-linker increased, so they belonged to the non-Fickian diffusion. The swelling exponent  $n$  value enhanced with the increasing cross-linker dose. Also, it indicated that the water absorbing mechanism was more complex because the swelling process was controlled by the interaction of water molecular diffusion and relaxation of the chain segments.

#### 3.2. Analysis of the mass retention rate

In order to evaluate the sensitivity of four hydrogels toward electric stimuli, the mass retention rate ( $R_w$ ) was tested. The measurement procedure was described in the experimental section. The  $R_w$  of PVA/PAAc hydrogels was displayed in Fig. 5. It indicated that there was some phenomenon of shrinkage for the PVA/PAAc hydrogels under non-contacted electric field, and it made the volume become

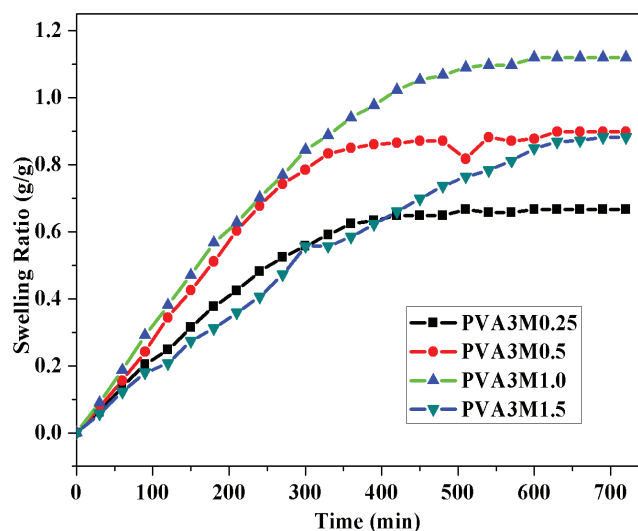


Fig. 3. Swelling ratio charts of PVA/PAAc hydrogels.

smaller, so the hydrogels released water. Kim et al. [29] proposed that electric-responsive hydrogels would shrink and release water when an extra electric field was added, because the electric field caused the migration of the positively charged counter-ion in the polymer network resulting in varying of osmotic pressure and ionic state, and then the volume and shape of the hydrogels changed. The  $R_w$  was examined in 5, 10 and 15 V, respectively, and we found that it decreased slowly with the increasing voltages. Overall, the longer time for electric field applied was, the lower  $R_w$  was. The lower  $R_w$  meant that it released more water. And the PVA3M1.0 and PVA3M1.5 released less water than the PVA3M0.25 and PVA3M0.5.

The  $R_w$  of PVA3M0.25, PVA3M0.5, PVA3M1.0 and PVA3M1.5 reached 79.22%, 77.37%, 81.69% and 81.19%, respectively, under 5 V after 50 min. The  $R_w$  of PVA3M0.25, PVA3M0.5, PVA3M1.0 and PVA3M1.5 reached 77.45%, 78.60%, 78.77% and 81.12%, respectively, under 10 V after 50 min. The  $R_w$  of PVA3M0.25, PVA3M0.5, PVA3M1.0 and PVA3M1.5 reached 77.69%, 78.52%, 82.23% and 82.64%, respectively, under 15 V after 50 min. However, it almost did not decrease entirely with the increasing voltage. So it signified that higher voltages did not trigger more water to release. PVA3M0.5 hydrogel had relatively lower  $R_w$  at lower voltage. It could be concluded that subtle electric field had an effect on the PVA/PAAc hydrogels, and 5 V was selected to evaluate the dehydration test of hydrogels. The swollen hydrogels could release water when it was subjected to electric stimuli, and they became dehydration state under the non-contacted electric field before the FO process.

#### 3.3. The morphology of PVA/PAAc hydrogels

The morphology of PVA/PAAc hydrogels was also investigated using SEM. The SEM micrograph of PVA/PAAc hydrogels' upper surface is shown in Fig. 6. It could be seen that there were some inhomogeneous pores in the PVA3M0.25 hydrogels, and the surface became dense with the increasing cross-linker content. The network structure was relatively loose, and it led to pore when the content of



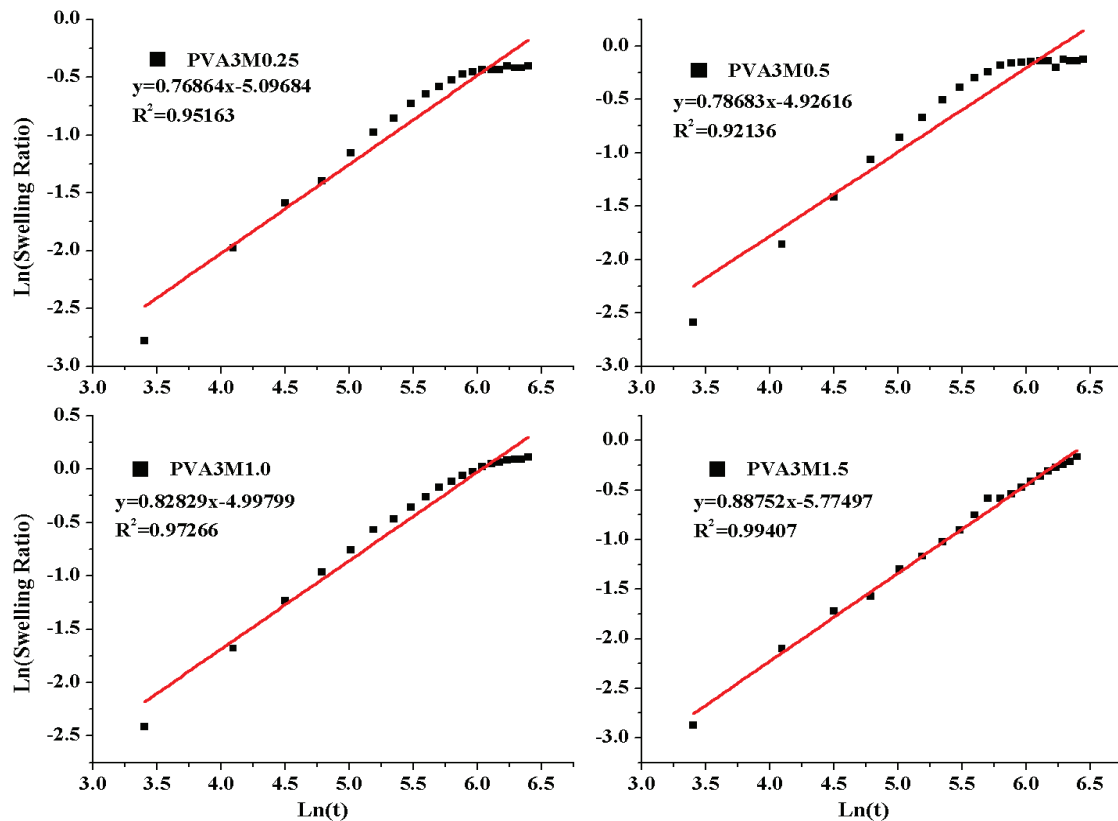


Fig. 4. Swelling kinetic relation charts of the hydrogels.

cross-linker was less. The cross-linker could promote the cross-linking polymerization of polymer molecules to form a network structure. With increasing amount of the cross-linker, the polymer chain would be further enhanced, so that the network structure of gel was dense. Therefore, it was seen that PVA3M1.5 hydrogel became much denser and less porous. The literature [30] reported that morphology of PVA/PAAc hydrogels was related to the PVA content, because PVA had excellent chemical resistance and physical properties. SEM images of PVA/PAAc hydrogels' fracture surface are shown in Fig. 7. Polymer network structure was observed in the PVA/PAAc hydrogels, and it became apparent with the increasing amount of cross-linker.

### 3.4. Swelling–deswelling behavior

In order to further investigate the swelling–deswelling behavior of the PVA/PAAc hydrogels, five-cycle continuous swelling–deswelling experiments were conducted. In brief, SR was tested while the hydrogels was immersed in deionized water for 720 min. Then the hydrogels were taken out from water, and the SR was tested while the hydrogels were subjected to the electric stimuli in 5 V until SR did not change with the time. This was one swelling–deswelling cycle. Continuous swelling–deswelling process was repeated for another four cycles. And the following curves are shown in Fig. 8. It can be observed that the PVA/PAAc hydrogels showed reversible behavior according to the applied electric field. In general, in the first 10 min the SR decreased rapidly after a 5-V voltage was employed, and it indicated that they

dehydrated quickly at the beginning. Then the SR dropped slowly with the time increasing. In addition, the SR decreased slowly as the amount of cross-linker increased because of too much cross-linked point. The order of equilibrium SR was PVA3M1.0 > PVA3M0.5 > PVA3M1.5 > PVA3M0.25, while the order of dehydration rate after 50 min under the 5 V was PVA3M0.25 > PVA3M0.5 > PVA3M1.0 > PVA3M1.5. With the increasing amount of cross-linker, the dehydration of the hydrogels slowed down when the electric field was applied. That result was in agreement with the result of the mass retention rate. However, the profile of swelling–deswelling behavior was similar, and it suggested that polymer hydrogels had a good reversibility and could be reused. Above results indicated that PVA/PAAc hydrogels had the potential in FO as draw agent, which could be easily recovered through electric stimuli. Furthermore, according to the above discussions, PVA3M0.5 and PVA3M1.0 were chosen as the draw agent for the FO during the following experiments.

During the dehydration of the hydrogels, only the direct current (DC) consumed energy. Electrical energy consumption, abbreviated  $W$  (J), was obtained from Eq. (6):

$$W = UIt \quad (6)$$

where  $W$  (J) is the electrical consumption caused by the DC power supply;  $U$  and  $I$  are the voltage and current of the DC power supply, respectively;  $t$  is the time that electric field is applied. Meanwhile, the voltage and current of DC power supply are 5 V and 3 A, respectively. So the energy

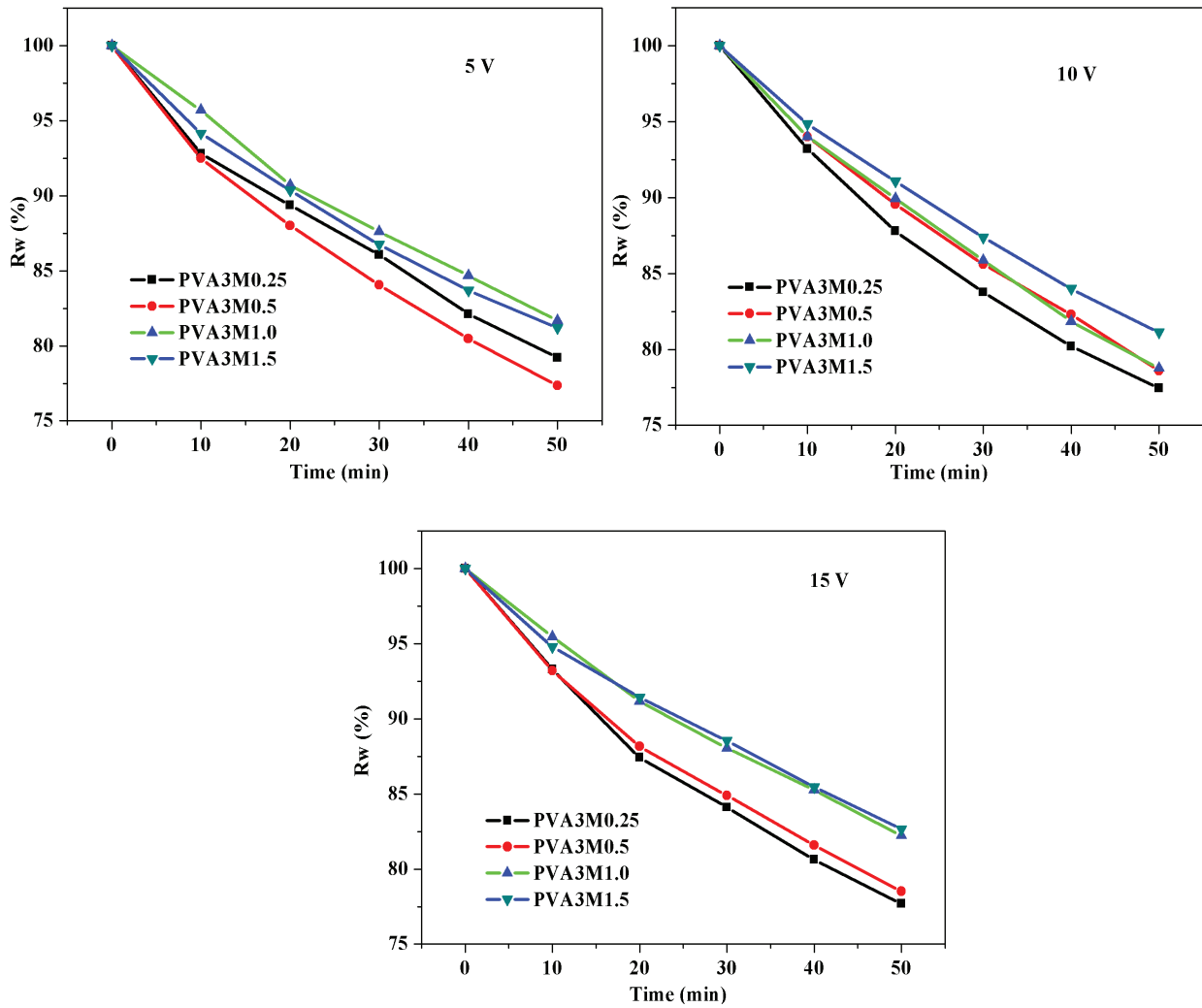


Fig. 5. Charts of the mass retention rate about PVA/PAAc hydrogels under different voltages.

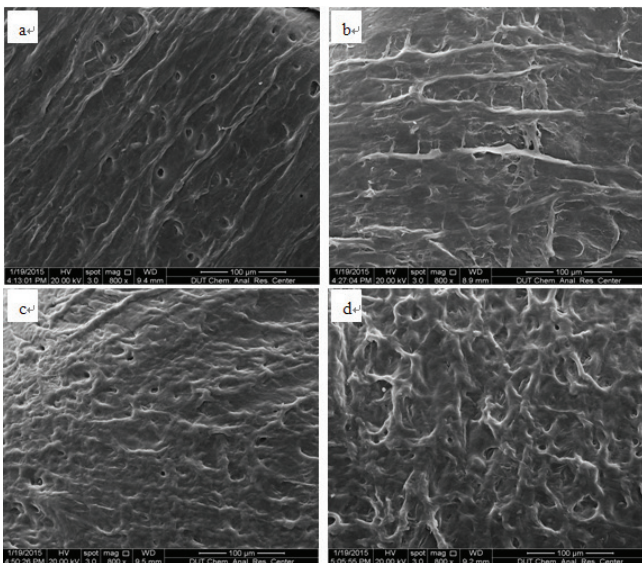


Fig. 6. SEM graphs of PVA/PAAc hydrogels' upper surface: (a) PVA3M0.25; (b) PVA3M0.5; (c) PVA3M1.0 and (d) PVA3M1.5.

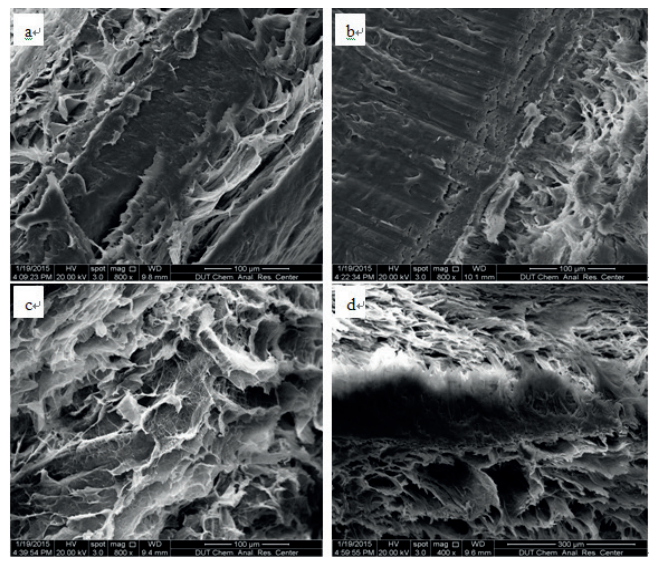


Fig. 7. SEM images of PVA/PAAc hydrogels' fracture surface: (a) PVA3M0.25; (b) PVA3M0.5; (c) PVA3M1.0 and (d) PVA3M1.5.

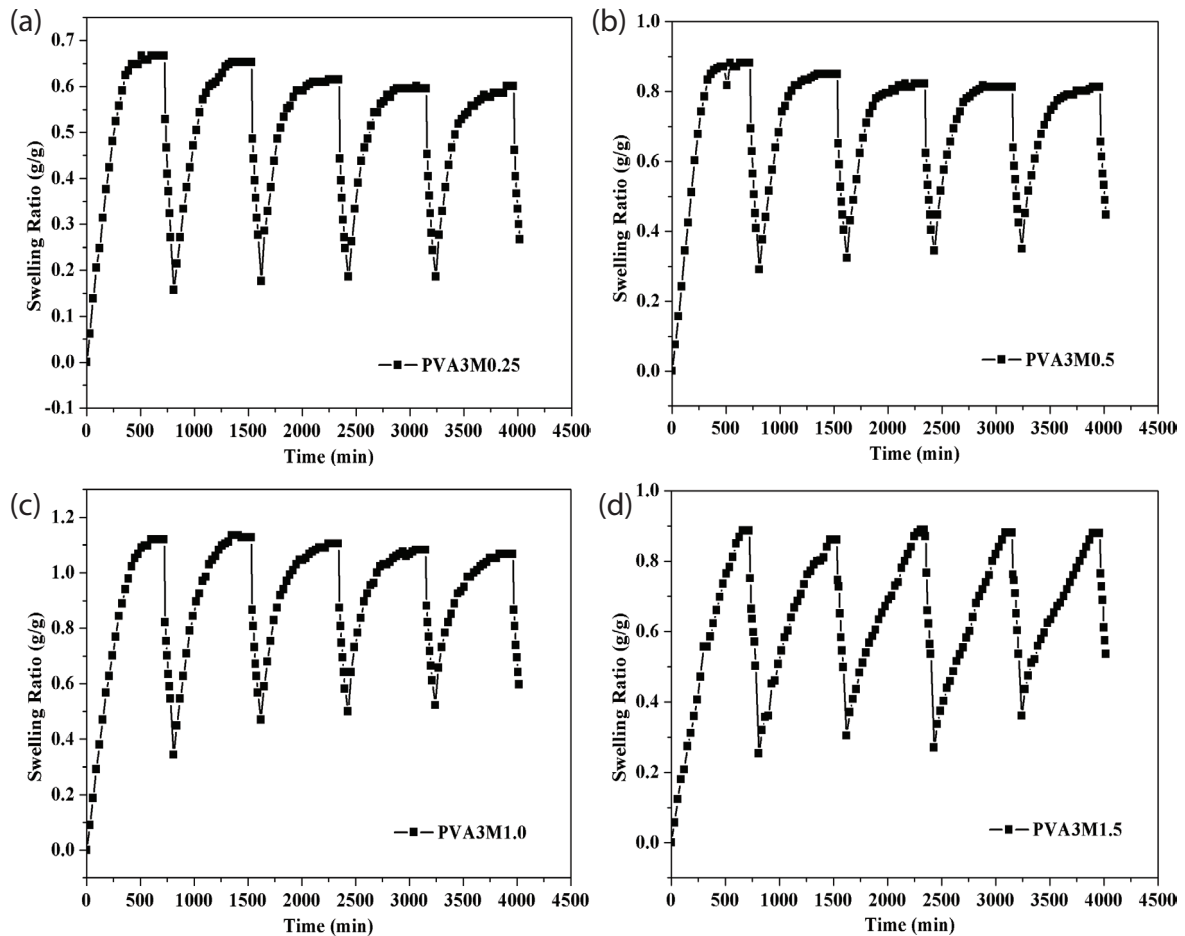


Fig. 8. Swelling–deswelling charts of PVA/PAAc hydrogels: (a) PVA3M0.25; (b) PVA3M0.5; (c) PVA3M1.0 and (d) PVA3M1.5.

consumption of DC power supply in 50 min would be  $4.5 \times 10^4$  J after one cycle of dehydration. Five cycles of dehydration consumed  $2.25 \times 10^5$  J.

### 3.5. Water fluxes in FO process

To evaluate the performance of electric-sensitive polymer hydrogels as draw agents in FO, PVA/PAAc hydrogels were tested in homemade FO systems at room temperature. As shown in Fig. 9, the water fluxes gradually decreased as the FO process proceeded by using PVA3M0.5 and PVA3M1.0 hydrogels. In the first 9 h, the water fluxes decreased quickly, and then water fluxes became relatively stable. It could be explained that osmotic difference gradient between the draw agents and the FS was higher when the FO process started, and high osmotic difference led to the higher initial water flux, while the hydrogels absorbed water in FO process, subsequently the water flux reduced slowly as the osmotic difference gradient decreased. Before the FO process, the hydrogels was dehydrated under the electric field of 5 V. By using PVA3M0.5 and PVA3M1.0 polymer hydrogels as draw agents and deionized water as the FS, the initial water fluxes were 0.51 and 0.31 LMH, respectively, with the DS against the SL. And with the PVA/PAAc hydrogels facing the AL, the initial water fluxes were 1.04 and 0.72 LMH, respectively. The water flux of hydrogels facing AL was better

than that of hydrogels facing SL. It could be attributed to the more severe dilutive concentration polarization in FO mode.

And water fluxes were examined by using 2,000 ppm NaCl solution as FS and PVA3M0.5 and PVA3M1.0 hydrogels as draw agent, as shown in Fig. 10. Water fluxes declined with time because the driving force became low when the hydrogels drew water. The initial water fluxes of PVA3M0.5 and PVA3M1.0 were 0.73 and 0.54 LMH, respectively, in AL–DS mode when 2,000 ppm NaCl solution as the feed. And the initial water fluxes of PVA3M0.5 and PVA3M1.0 were 0.38 and 0.23 LMH, respectively, in AL–FS mode. The water flux of hydrogels was generally low according to the currently published in the literature. For example, Li et al. [20] demonstrated that the water flux for composite polymer hydrogels (poly(sodium acrylate)-carbon (PSA-C)) was 1.32 LMH in the first 0.5 h when 2,000 ppm NaCl solution was applied as the FS. A series of hydrogels based on poly(*N*-isopropylacrylamide) (PNIPAm)-PSA and PNIPAm-PVA prepared by Cai et al. [21] had low energy consumption and quasi-continuous characteristics that made semi-IPN hydrogels attractive as draw agents for FO desalination, but the initial water flux was 0.24 LMH. Relatively speaking, as-prepared hydrogels in the study were in medium level. Water fluxes decreased rapidly in the first 9 h; subsequently, it tended to be stable. Compared with the deionized water as

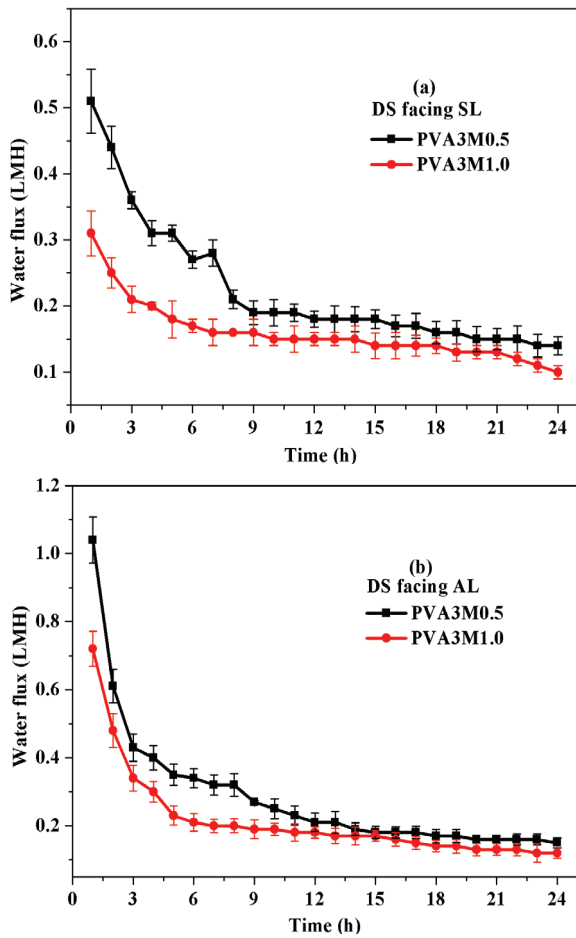


Fig. 9. Water fluxes vs. time in FO process using the deionized water as the feed and PVA/PAAc hydrogels as draw agent: (a) hydrogels DS facing SL of CTA membrane and (b) hydrogels DS facing AL of CTA membrane.

the feed, the initial water fluxes were slightly lower because of its lower driving force.

Flory [31] pointed out that the swollen hydrogel was similar to solution. Wang et al. [32] suggested that the swelling pressure derived from polymer-water mixing, elastic reaction force of the polymer network structure and osmotic pressure of ionizable groups. Thus, these literatures showed that the swelling pressure of polymer hydrogels was related to concentration of free mobile ions in the network structure. Wack and Ulbricht [33] demonstrated that no effect on the swelling pressure could be found by variation of monomer concentration and cross-linker content; conversely, the content of sodium acrylate had an effect on the swelling pressure. The prepared hydrogels were mostly composed of  $-\text{COOH}$ , which was free to move and able to dissociate less, so it was inferred that the swelling pressure became lower. The osmotic pressure difference was so small that it led to the lower water flux. Although the water fluxes were relatively lower compared with those using saline solutions as draw agents, hydrogels showed its advantages such as easy recovery, no reverse salt leakage, etc. Its potential used as draw agents in FO is worthy of further investigation.

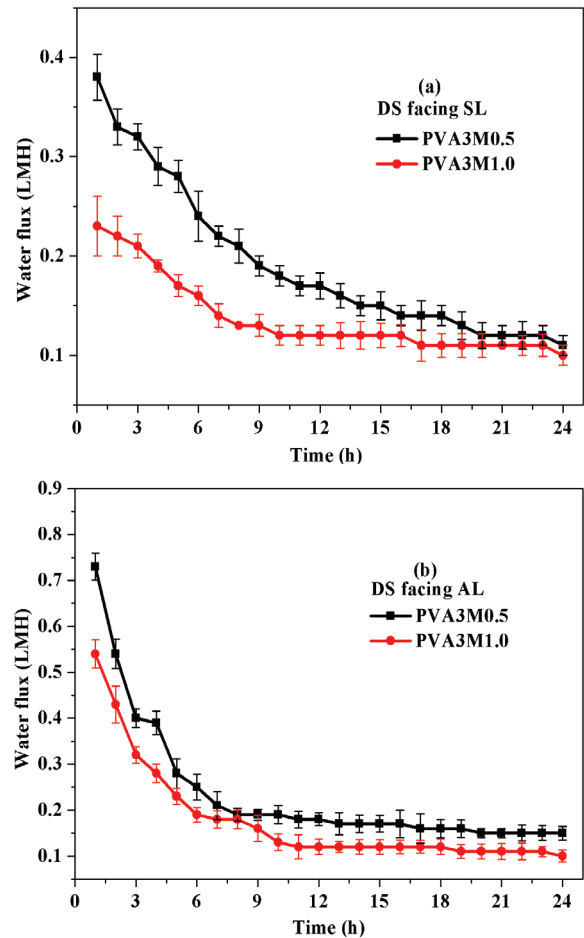


Fig. 10. Water fluxes vs. time in FO process using 2,000 ppm NaCl solution as the feed and PVA/PAAc hydrogels as draw agent: (a) hydrogels DS facing SL of CTA membrane and (b) hydrogels DS facing AL of CTA membrane.

#### 4. Conclusions

In this paper, chemical cross-linked method integrated with the freezing-thawing was used to fabricate electric-sensitive hydrogels as draw agent for FO. The monomers of PVA and AAc were adopted to prepare the PVA/PAAc hydrogels with different amounts of cross-linker, and they were designed as PVA3M0.25, PVA3M0.5, PVA3M1.0 and PVA3M1.5. The experiment of mass retention rate demonstrated that the PVA/PAAc hydrogels were sensitive to the subtle electric field; thus, they could shrink and release water under non-contacted electric field. Swelling–deswelling behavior analysis showed that the hydrogels could absorb water again in the next swelling experiment when the hydrogels released water under non-contacted electric field, and PVA3M0.5 released water quickly when the 5 V was applied. These results proved that the hydrogels could be reused and also had enough mechanical strength. Therefore, by using PVA3M0.5 and PVA3M1.0 as draw agent for FO, with the hydrogels facing the AL, the initial water fluxes were 1.04, 0.72, 0.73 and 0.54 LMH, respectively, when the deionized water and 2,000 ppm NaCl were used as the FS. Besides, reverse salt leakage of draw agent could be avoided by using polymer hydrogels as draw agent



in FO process. So there is great potential for application in the field of food and drug concentration that is not allowed to be contaminated for the FS.

### Acknowledgment

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### Symbols

$M_t$	—	The mass of the hydrogel at a certain time $t$ , g
$M_0$	—	The mass of the semi-dry hydrogel, g
SR	—	Swelling ratio, g/g
$k$	—	Swelling constant
$n$	—	Swelling exponent calculated from the slopes of the $\ln SR$ lines vs. $\ln t$ plots
$m_t$	—	The mass of hydrogels at a certain time after non-contacted field was applied, g
$m_0$	—	The mass of hydrogels before it was laid in the device, g
$F$	—	Water flux, LMH
$\Delta V$	—	Volume of water permeated through the FO membrane, L
$A$	—	Effective area of membrane, $m^2$
$W$	—	Electrical energy consumption, J
$U$	—	Voltage, V
$I$	—	Current, A
$t$	—	Time that electric field is applied, s

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