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Effect of backwash temperature on hollow fiber ultrafiltration membranes fouled by sodium alginate

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ABSTRACT

This study focuses on the effect of backwash temperatures (25, 40, and 55˚C) on hollow fiber ultrafiltration membranes fouled by sodium alginate (SA). The results indicate that a higher backwash temperature could achieve a significant mitigation of total backwash resistance and a cleaning rate promotion. In the following cycle, the initial total fouling resistance was decreased with the temperature increase, while there was an undesirable increase on the fouling rate due to a higher SA concentration in the membrane pool. Although the membrane fouling resistance was more reversible, backwash at 55˚C had a negative impact on the SA removal, because the functional gel layer was washed out. Moreover, all the beneficial effects from 25 to 40˚C were more noticeable than those from 40 to 55˚C. Overall, backwash at 40˚C was relatively a better choice in this study.

Keywords: Ultrafiltration; Sodium alginate; Backwash temperature; Hollow fiber membrane; Total backwash resistance

1. Introduction

Ultrafiltration (UF) has been successfully applied in drinking water treatment for the removal of particles, turbidity, microorganisms, and pathogens from surface water and groundwater during the past decades [1]. However, the generalization of UF was mainly constrained by membrane fouling, which usually brought a serious decline in membrane permeability with foulants intercepted on the surface or into the pores of the membrane over time. Sodium alginate (SA), as a typical dissolved organic matter which was a significant foulant to membrane fouling, previously played a surrogate role for polysaccharide in estimating its fouling mechanism [2–4].

Some "standard" protocols, such as hydraulic backwash and chemical cleaning, have been widely used to mitigate membrane fouling. Periodical hydraulic backwash, as a practical and effective method, makes it possible for the membrane to regain the permeability, and the membrane service life shall be prolonged [5]. Temperature is an important factor in membrane filtration process related to mass transfer [6], dynamic viscosity [7], membrane fouling, and concentration polarization [8]. The effect of feed water temperature on separation performance and fouling mechanism has been investigated previously by researchers [6,9]. By contrast, the effect of backwash

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temperature on mitigating the UF membrane fouling was seldom reported. Although there were several studies concerned, the determination of optimal backwash temperature has not reached a consensus. According to the study of Sohrabi et al. [10], the optimum backwash temperature was 35 ± 1℃ on the reverse osmosis membrane and the nanofiltration membrane which were fouled by licorice aqueous solutions. However, another study indicated that cleaning temperature between 50 and 60℃ could achieve the maximum flux recovery within 5 min in a microfiltration process [11]. Almecija et al. [12] also pointed out that backwash at 50℃ was the optimal temperature to recover flux in ceramic membranes; however, a process of membrane pore erosion found at 60℃. Thus, the temperatures studied in this study ranged from 25 to 55℃. Being compared with other kinds of membranes, the hollow fiber UF membrane is more prevalent on plant scale for the benefits of low costs, easy control, and maintenance [13–15]. Therefore, this kind of membrane was used in our experiments.

The aim of this study was to contribute to a better understanding of backwash performance on hollow fiber membrane fouled by SA under different temperatures (25, 40, and 55℃). Effects of the three backwash temperatures on the fouling potential of SA solution were initially examined via evolution of total filtration resistance (TFR)/total backwash resistance (TBR) as well as fouling/cleaning rates in consecutive filtration/backwash steps. Subsequently, advanced experiments that backwash temperatures changed with fixed intervals were carried out further.

2. Materials and methods

2.1. Feed water

In this study, commercial SA, a polysaccharide model substance, was the only solute that existed in the feed water. The 10 g/L SA stock solution was prepared from deionized water and stored at 4℃. Prior to usage, it was restored to room temperature at 25 ± 1℃. The SA concentration of feed water was 20 mg/L with a pH of 7.5.

2.2. Experimental apparatus

This study was performed in a bench-scale apparatus developed in the laboratory, which is schematically shown in Fig. 1. The hollow fiber UF membrane module (Litree, China) made of polyvinyl chloride was used in this study. The total surface area of a membrane was 0.02 m^2, and its nominal pore size was 0.01 μm. The new membrane was immersed in DI water for about 24 h to remove any residual storage agent.

As shown in Fig. 1, in order to keep the same water head, the feed water was pumped into a high-level water tank and then flowed down to a constant-level tank. The membrane module was submerged in the membrane pool. The permeated water was collected directly from the membrane module using a suction peristaltic pump. A pressure sensor was installed between the membrane module and the peristaltic pump to monitor the TMP continuously. The UF process was conducted utilizing a dead-end filtration mode at a constant flux of 30 L/(m^2 h), corresponding to a hydraulic retention time of 2 h. Permeated samples were collected periodically for subsequent concentration analysis. The temperature of the permeated solution, collected in the backwash tank for backwashing, was controlled at 25, 40, and 55℃ (within ±2℃), respectively, by a temperature regulator. The backwash flux amounted to 0.6 L/h. The permeated sample within the initial 5-min filtration cycle was excluded from the collection in order to eliminate the influence of the unsteady SA removal that occurred at the beginning of filtration.

2.3. Fouling and backwash experiments

Experiments were divided into two parts: primary experiments and advanced experiments.

In the primary experiments, three groups of UF membrane modules with identical characteristics and operating conditions were used to compare the effects of three different backwash temperatures (25, 40, and 55 ± 2℃, respectively) on the SA viscosity, TFR/TBR, and fouling/cleaning rate in a relatively short-term operation period through three successive filtration/backwash cycles. Each cycle contained a 58-min filtration step and a 2-min backwash step.

In order to investigate the effect of backwash temperature in a relatively long-term operation period, advanced experiments were carried out. Three groups of UF membrane modules went through eight successive filtration/backwash cycles at room temperature (25 ± 2℃), except for the sixth backwash step, which used varying temperatures (25, 40, and 55 ± 2℃). Each cycle identically contained a 58-min filtration step and a 2-min backwash step. Intermittent backwash tests at specific temperatures were further evaluated by the evolution of TFR/TBR, fouling/cleaning rates, constitution of membrane fouling resistance, SA removal, and its mass transfer.

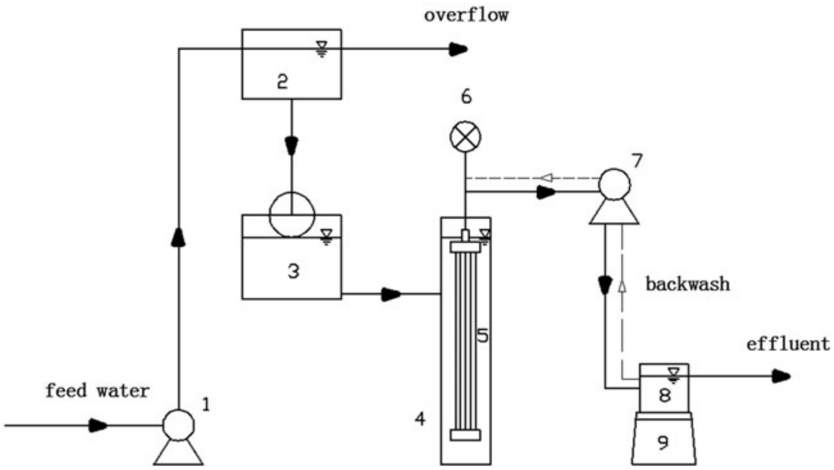

Fig. 1. Schematic diagram of the single hollow fiber apparatus employed in the filtration/backwash experiments. Note: 1. feed peristaltic pump; 2. high-level water tank; 3. constant-level water tank; 4. UF system; 5. UF membrane module; 6. pressure sensor; 7. suction peristaltic pump; 8. collected backwash tank; and 9. temperature regulator.

It is worth mentioning that during the advanced experiments, we found that the concentrated solution in the membrane pool had a greater influence on restraining membrane fouling. Thus, in check experiments, the SA solutions, after the sixth backwash cycle with different temperatures, were drained. Then, the pool was refilled with the feed water to investigate the impact of the SA concentration.

2.4. The determination of TFR/TBR of membranes

Darcy's law (Eq. (1)) describes the degree of membrane fouling during the filtration operation:

$$\mathrm{TFR} = \frac{\mathrm{TMP}}{\eta J_\mathrm{w}} \tag{1}$$

where TFR is the total filtration resistance (m^{-1}); TMP is trans-membrane pressure (Pa); η is the dynamic viscosity (Pa s); J_w is membrane flux ($\mathrm{m}^3/\mathrm{m}^2\,\mathrm{s}$). For constant-flux filtration, the resistance-in-series model is expressed in Eq. (2):

$$\mathrm{TFR} = R_\mathrm{m} + R_\mathrm{f} = R_\mathrm{m} + R_\mathrm{r} + R_\mathrm{ir} \tag{2}$$

R_m is the intrinsic membrane resistance (m^{-1}) and R_f is the total fouling resistance, including the physically reversible fouling resistance (R_r, m^{-1}) and physically irreversible fouling resistance (R_ir, m^{-1}).

$$\mathrm{TBR} = \frac{\mathrm{BTMP}}{\eta J_\mathrm{w}} \tag{3}$$

TBR is the total backwash resistance (m^{-1}) as a new concept mentioned in this study, being used to estimate the effect of the backwash on the degree of membrane fouling (calculated by Eq. (3)); BTMP is the backwash trans-membrane pressure (kPa).

The rate of foulant accumulation on the membrane over time was measured using fouling rate values (V_f, kPa/h) on a per cycle basis (Eq. (4)).

$$V_\mathrm{f} = \frac{\mathrm{TMP}_\mathrm{end} - \mathrm{TMP}_\mathrm{ini}}{t_\mathrm{end} - t_\mathrm{ini}} \tag{4}$$

The rate of foulant cleaning on the membrane over time was measured using cleaning rate values (V_c, kPa/h) on a per cycle basis (Eq. (5)).

$$V_\mathrm{c} = \frac{\mathrm{BTMP}_\mathrm{end} - \mathrm{BTMP}_\mathrm{ini}}{t_\mathrm{end} - t_\mathrm{ini}} \tag{5}$$

The initial TMP and final TMP in each cycle were expressed by $\mathrm{TMP}_\mathrm{ini}$ and $\mathrm{TMP}_\mathrm{end}$. The initial BTMP and final BTMP in each cycle were expressed by $\mathrm{BTMP}_\mathrm{ini}$ and $\mathrm{BTMP}_\mathrm{end}$. The initial time and final time in each cycle were expressed by t_ini and $t_\mathrm{end.}$

2.5. Viscosity measurement

Under normal pressure, the dynamic viscosity of SA solution was expressed in Eq. (6):

$$\eta = \eta_0 \eta_\mathrm{r} = \eta_0 \frac{t_\mathrm{u}}{t_\mathrm{v}} \tag{6}$$

The relative viscosity (η_r) was measured using an Ubbelohde-type capillary viscometer 0.40 mm in diameter and 10.0 cm in length [7]. The measured temperature of backwash water at 25, 40, and 55 ± 2℃ was controlled by a circulating water bath. Deionized water was regarded as a solvent, and its flow time (t_v, s) was measured by the viscometer. Similarly, the flow time of SA solution (t_u, s) was measured as well. The ratio of flow time of solution to that of solvents, t_u/t_v, was regarded as the relative viscosity. The absolute viscosity (η_0, Pa s) of deionized water could be found in the viscosity database [16].

2.6. SA removal

The purifying effect of UF process on SA solution was measured by SA removal, which was calculated by the ratio of C_{feed} and C_{permeate} (Eq. (7)).

$$\text{SA removal} = \frac{C_{\text{feed}} - C_{\text{permeate}}}{C_{\text{feed}}} \times 100\% \quad (7)$$

C_{feed} and C_{permeate} stand for the SA concentration of feed water and permeate water, respectively, being measured by total organic carbon detector (Elementar, Germany) with the NPOC method.

3. Results and discussion

3.1. Primary experiments

Filtration/backwash experiments were performed with SA solutions in a short period to investigate the effect of backwash temperature on the mitigation of membrane fouling. TFR/TBR behaviors in the different backwash temperatures are shown in Fig. 2. Significant TFR increases occurring in each filtration step indicated a severe membrane fouling. A quick TFR buildup occurred at the initial stage of each filtration cycle, because the dominant fouling mechanism in the beginning of filtration was pore blocking, which was more severe than the fouling of the gel layer forming subsequently [17–19].

The TBR (in Fig. 2) was quite different when the backwash temperature varied. A higher backwash temperature resulted in a lower TBR and higher cleaning rate (Fig. 3), which indicated a greater reduction of foulant. This reduction was caused by the improved mobility of the backwash solution related to its dynamic viscosity (Table 1), which attenuated with the increase of temperature.

Meanwhile, the dissolving capacity increased when the backwash temperature was enhanced due to a higher diffusive coefficient [6]. As a result, a lower initial TFR was subsequently displayed in the next filtration cycle. Although backwashing at a higher temperature and in such a short period of time had shown its potential of mitigating membrane fouling, the advantage was not obvious in a slight fouling condition.

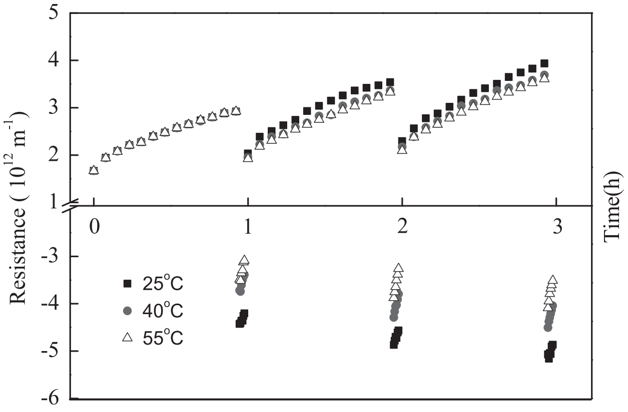

Fig. 2. Effects of backwash temperatures on TFR/TBR in primary experiment.

3.2. Advanced experiments

In advanced experiments, effects of the varied backwash temperatures, using intervals instead of cycles, were further investigated to cope with more severe fouling accumulation.

3.2.1. Effect of temperature on TFR/TBR and fouling/cleaning rate

In this advanced experiments, the influence of the concentrated solution was taken into account for a longer period accumulation.

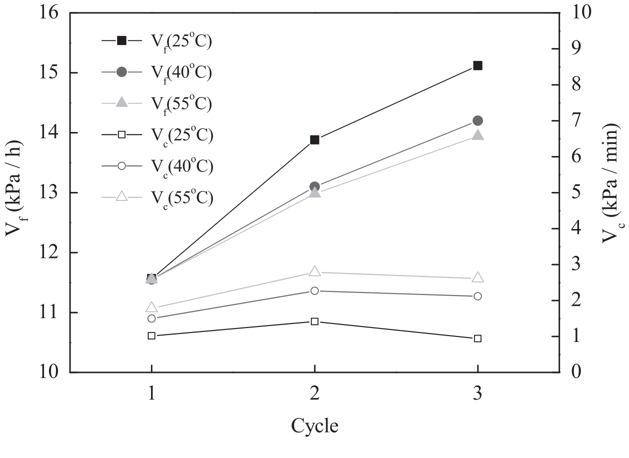

Fig. 3. Effects of backwash temperatures on fouling/cleaning rate in primary experiment.

Table 1
Effect of backwash temperature on viscosity

		Backwash temperature (℃)		
	Unit	25	40	55
η	Pa s	0.992×10^{-3}	0.733×10^{-3}	0.568×10^{-3}

3.2.1.1. Concentrated solution retained. As shown in Fig. 4(a) a steady buildup of TFR/TBR was depicted in each cycle, and a significant recovery of TFR was clearly observed after each backwash step. During the sixth backwash cycle, a significant mitigation of TBR and a cleaning rate promotion were found in a higher temperature. These results were more prominent than those of primary experiments. And the difference values of TBR and cleaning rate, respectively, between 40 and 25℃ were about two times the figure for those between 55 and 40℃. This indicated that the effect of the backwash temperature on both the TBR and the cleaning rate was not linear under a long-term fouling.

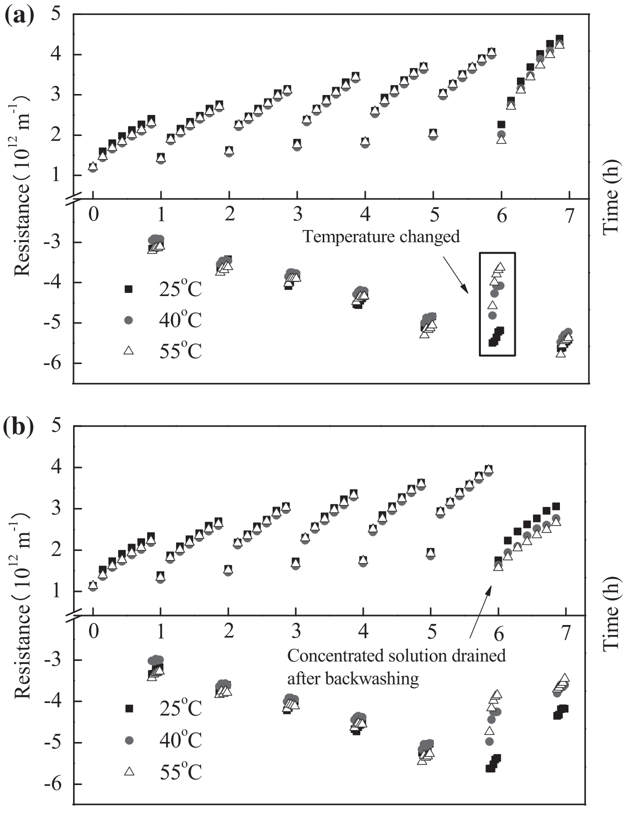

Fig. 4. Effects of backwash temperature and concentrated solution in membrane pool on TFR/TBR in advanced experiment (a) retaining concentrated solution and (b) discharging concentrated solution.

Backwashing at a higher temperature led to a lower initial TFR in the next cycle as expected, but an undesirable increase of the fouling rate was observed, accidentally. This can be explained by a more augment of the SA concentration measured in the membrane pool after the sixth backwash step (Table 2); in other words, it was the retentate removed from the membrane that was concentrated in the membrane pool and in next cycle reformed the gel layer. A previous study via direct observation found that the formation of the loose fouling gel layer after periodic backwashing rapidly recompressed upon resuming filtration [19]. This phenomenon indicated that the backwash temperature played a significant role in controlling the initial TFR but had an adverse effect on the restraint of the fouling rate in the following filtration cycle.

3.2.1.2. Concentrated solution drained. In order to eliminate the negative impact of the concentrated SA solution on reforming gel layer, check experiments were implemented to illuminate the individual effects of the backwash temperature and concentrated solution on alleviating membrane fouling. After the sixth backwash cycle, the concentrated SA solution in the membrane pool was drained and refilled with the feed water.

As shown in Fig. 4(b), after backwashing at 25, 40, and 55℃, each initial TFR in the seventh cycle was lower than that in Fig. 4(a), because the dissolution of gel layer elevated with the decrease of the SA concentration. Hence, the resistance of concentration polarization based on the gel layer formation was whittled down. During the ensuing filtration cycle, the fouling rate (Fig. 5(b)) achieved a significant decrease compared with the result of Fig. 5(a). This proved that the SA concentration in membrane pool played a dominant role in fouling rate. Moreover, the difference of fouling rates at 25℃ between Fig. 5(a) and (b), in the seventh filtration cycles, could reflect the individual effect of concentrated solution. Correspondingly, the difference of the fouling rates between different temperatures

Table 2
The SA Concentration in membrane pool before/after the sixth backwash step

		Backwash temperature (℃)		
	Unit	25	40	55
C_{before}	mg/L	32.6	32.1	30.1
C_{after}	mg/L	40.0	45.6	45.8
ΔC	mg/L	7.4	13.5	15.7

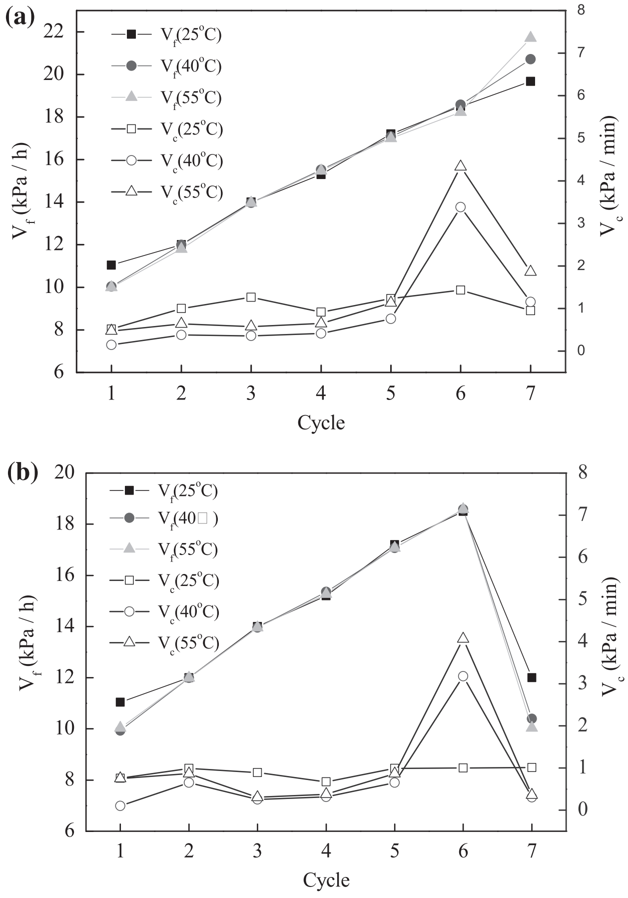

Fig. 5. Effects of backwash temperature and concentrated solution in membrane pool on fouling/cleaning rate in advanced experiment (a) retaining concentrated solution and (b) discharging concentrated solution.

(Fig. 5(b)) indicated the individual effect of the backwash temperatures as well. According to the results above, we found the fouling rate was mainly determined by the actual feeding of the SA concentration in the membrane pool. A previous result was also found that a higher SA concentration decreased the permeability of UF membranes [4]. Additionally, the difference of cleaning rates at 25℃ (Fig. 5(a) and (b)) was due to the difference of final their TFRs. Thus, a combination of rising backwash temperature and then draining the concentrated solution in time improved the performance on the mitigation of the membrane fouling.

3.2.2. Effect of temperature on SA removal and distribution

The evolution of the SA removal at different backwash temperatures over eight filtration cycles is illustrated in Fig. 6. In spite of some system errors among these three groups, the trends of the SA removal were clear and accordant.

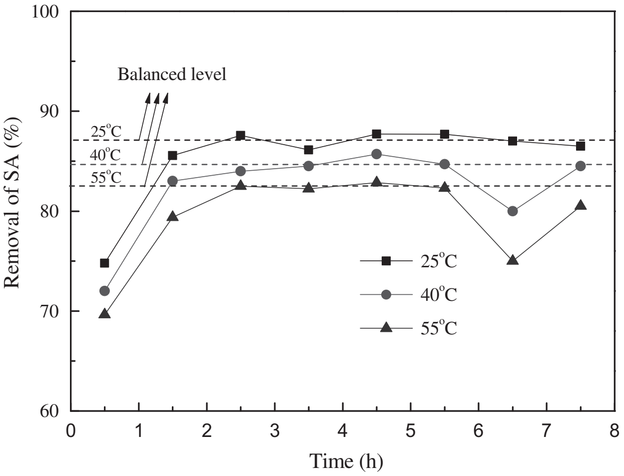

Fig. 6. SA removals at different backwash temperatures in advanced experiment.

Removals increased rapidly in the initial three cycles. With the deposition of SA molecules on the membrane surface, the gel layer was formed gradually as an additional barrier to the solute transport [20]. In the first cycle, the gel layer was initially formed, but this layer was not quite compacted. Its loose part was washed out in the first backwash step. In the second cycle, the gel layer became more compacted and its rejection performance was improved. As a result, the SA removal was further enhanced. From the third to the sixth cycle, the removals were stabilized because the formation of gel layers developed into a dynamic equilibrium under periodic filtration/backwash alternations. However, changing the backwash temperature, at the end of the sixth cycle, broke this equilibrium. With the temperature increased, the SA removal suddenly was reduced, because part of the functional gel layer was removed. On one hand, the SA solubility increased with the backwash temperature [7]; hence, more SA molecules were dissolved into backwash solution from the gel layer. Besides, the reduced dynamic viscosity made the Reynolds number increased [11] and hence raised the turbulence intensity surrounding the membrane surface. A similar theory was also discussed concerning the feed water temperature of the reverse osmosis membrane [21]. In the following seventh filtration cycle, the SA layers were recreated varying from the backwash temperatures. After an excessive backwash at 55℃, the SA layer recreated an incomplete gel layer as a result of the decrease of the SA removal. However, the removal was still higher than the first cycle. This indicated that the residual stubborn gel layer still kept some functions of rejection and adsorption. It is worth mentioning that the SA removal at 55˚C did not bounce back to the balanced level within the two cycles. By contrast, backwashing at 40˚C as a gentle cleaning to the SA layer not only had a right control on the increase of the TFR but also recreated a relatively complete gel layer in the seventh filtration cycle. Therefore, its SA removal had an acceptable decrease and then rapidly bounced back to the balanced level in the eighth cycle.

The SA distribution in the UF process was an important parameter for illustrating the process of mass transfer. The SA mass distribution accumulated within the first six constant filtration cycles was depicted in Fig. 7. Relying on the restored positions, the SA molecules were classified into four parts: concentrated solution, reversible layer, irreversible layer, and permeated solution. The SA concentration in the concentrated and permeated solution was quite similar (around 50 and 20% collectively). Yet, the main differences appeared on the reversible and irreversible layer after suffering different temperature backwash. With the backwash temperature increased, the SA layer was more reversible, and the irreversible part of the residual layer on the membrane surface decreased. There were 22, 10, and 7% of the irreversible layer left on the membrane, while 11, 21, and 24% of the reversible layer was flushed out in the backwash process at 25, 40, and 55˚C, respectively. A greater decrease of the irreversible layer appeared at 40˚C than 25˚C, whereas at 55˚C, it had a much less improvement in removing irreversible layer, and this was consistent with the TBR behaviors. This phenomenon indicated that the decrease of the irreversible SA layer determined the decline of the TBR, as well as the reduction of the initial TFR in the next filtration cycle.

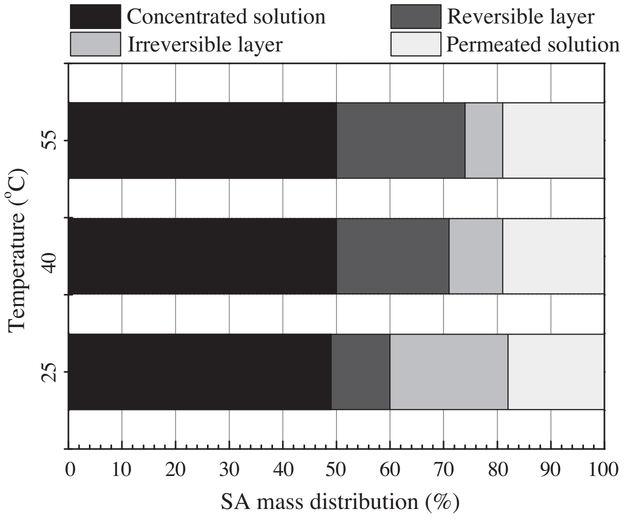

Fig. 7. SA mass distribution at different backwash temperatures in advanced experiment.

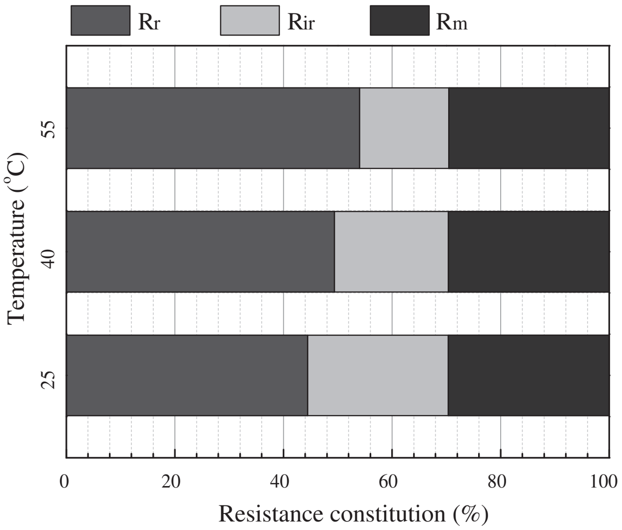

Fig. 8. Constitutions of membrane fouling resistance at different backwash temperatures in advanced experiment.

3.2.3. *Effect of temperature on membrane fouling resistance constitution*

The different resistance constitutions of the three backwash temperatures are shown in Fig. 8. It was found that the three investigated backwash temperatures had unobvious influence on $R_{m,}$ which was determined by the property of the membrane used. However, R_r took up a higher percentage of total resistance with increasing temperature and a lower percentage was observed in R_{ir} correspondingly. This phenomenon was in accordance with the consequence of the SA distribution in Fig. 7, revealing that the TFR at a 55˚C backwash was more reversible.

4. Conclusion

In this study, bench-scale experiments were developed to investigate the effect of the backwash temperature on the UF membrane fouling. The chosen foulant was SA, a typical model polysaccharide for dissolved organic matter. The core parameter considered was the backwash temperature.

(1) Backwash at a higher temperature could achieve a significant mitigation of TBR and a cleaning rate promotion. As a result, the initial TFR was decreased in the following cycle.

(2) A higher temperature removed more gel layer, which would increase the SA concentration in the membrane pool, thus essentially leading to a higher fouling rate in the next cycle.
(3) After a long-term operation, backwash at a higher temperature had a negative effect on the SA removal, although the membrane fouling resistance was more reversible.
(4) In this study, all the beneficial effects from 25 to 40℃ are more noticeable than those from 40 to 55℃. Moreover, considering the reduction of the fouling rate and SA removal at 55℃, backwash at 40℃ was proven to be overall a better choice.

Acknowledgements

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