



## Factors influencing critical flux of UF membrane in drinking water treatment

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Received 31 March 2014; Accepted 17 September 2014

### ABSTRACT

Very few fouling or even no fouling would happen when ultrafiltration (UF) was operated below the critical flux. Factors influencing the critical flux of UF for drinking water treatment were systematically investigated in both short-term and long-term tests. Results showed that critical flux could be increased by 5 L/m<sup>2</sup> h by continuous aeration with the air flow rate of 10 m<sup>3</sup>/m<sup>2</sup> h, and the same effect could be obtained by intermittent aeration with the air flow rate of 40 m<sup>3</sup>/m<sup>2</sup> h which could not be further improved by increasing air flow rate. Intermittent filtration had no positive effect on improving critical flux, while intermittent filtration combined with aeration could effectively improve critical flux by 5 L/m<sup>2</sup> h and reduce fouling a lot in long-term filtration. Critical flux could be increased by 10 L/m<sup>2</sup> h when coagulation and sedimentation pretreatment was applied, and could be increased by 15 L/m<sup>2</sup> h for combined process of coagulation combined with continuous aeration or coagulation combined with intermittent filtration and intermittent aeration. Coagulation coupled with intermittent filtration and aeration would produce a synergy effect on critical flux improvement. Critical flux determined by flux-step tests could help to instruct the UF operation in application.

*Keywords:* Critical flux; UF; Aeration; Filtration modes; Coagulation; Drinking water treatment

### 1. Introduction

Critical flux was initially defined in two ways: one definition is the flux through membrane which does not increase the trans-membrane pressure (TMP) of the membrane with time [1], and the other definition is the flux below which there is no deposition of colloids on the surface of membrane [2]. Above the critical flux, irreversible fouling of suspended solids forms a stagnant, consolidated, and aggregated layer on the

membrane surface, which can make the flux to decline rapidly. On the other hand, below the critical flux, called subcritical flux, it has been reported that the fouling is not observed [3].

Aeration has a positive effect on improving critical flux. The addition of gas leads to increases in the Reynolds and wall shear stress numbers which subsequently lead to increased critical fluxes [4]. Gas-liquid two-phase flow enhanced the microfiltration for treating wastewater; the flux being more than tripled [5]. Increasing the air flow rate from 1.2 to 3.6 m<sup>3</sup>/m<sup>2</sup> h, it was possible to decrease the total resistance and

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increase the filtrate flux by a ratio of three in a pilot submerged bioreactor [6]. A study adopted two coarse bubble aeration (sparging) modes in a submerged tubular membrane bioreactor and reported that the flux was found to increase by 43% when aeration was introduced to the airlift module [7].

A few investigations showed that coagulation could improve flux indeed [8–10]. The pretreatment of flocculation/pretreatment removed approximately 50% of organics, while producing uniform microflocs of 13–16  $\mu\text{m}$  size. In addition, the critical flux was enhanced by 70%, which resulted in 30–70% of the remaining organic removal in the cross-flow microfiltration [11]. Larger flocs typically yielded higher critical fluxes as in monodisperse suspensions, and the effect of particle size was less pronounced for the more porous flocs [12,13]. Few studies researched the effects of coagulation and sedimentation on critical flux especially in drinking water treatment.

Critical flux is very important for ultrafiltration (UF) operation because fouling would be very little or even not happen, and chemical cleaning would not be usually adopted when operating below it, which could simplify the UF operation of drinking water treatment plant. There have been already so many works studying the critical flux in municipal wastewater treatment especially for membrane biological reactor [13–17], while few studies reported the critical flux and its influencing factors of UF in drinking water treatment. Therefore, the main objective of this study is to systematically investigate the factors influencing the critical flux of UF membrane in both short-term and long-term tests for drinking water treatment including aeration, filtration modes and coagulation pretreatment. This article also aims to evaluate the effects of critical flux determined by flux-step method on the actual operation in application.

## 2. Materials and methods

### 2.1. Experimental setup

Bench-scale immersed UF was employed in this study. A schematic illustration of the experimental setup is shown in Fig. 1. The UF membrane modules were made of polyvinylidene fluoride (PVDF), with a nominal pore size of 0.01  $\mu\text{m}$  and a total membrane area of 0.01  $\text{m}^2$ . The characteristics of UF membrane are shown in Table 1. The effluent was drawn directly from the membrane module by using a peristaltic pump. A pressure sensor was set between the membrane module and the suction pump to monitor and TMP was recorded automatically by software. Aeration was provided at the bottom of the membrane

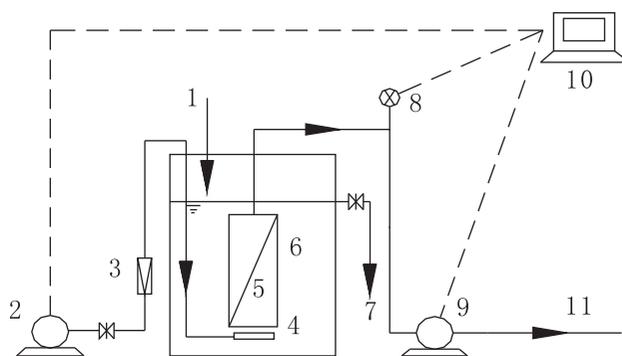


Fig. 1. Schematic diagram of the experimental setup. (1) Feed water; (2) air pump; (3) air flowmeter; (4) air diffuser; (5) UF membrane module; (6) UF tank; (7) overflow; (8) pressure sensor; (9) peristaltic pump; (10) computer and software; and (11) effluent.

Table 1  
Physical characteristics of the UF membranes

| Parameters                   | Characteristics   |
|------------------------------|-------------------|
| Material                     | PVDF              |
| Type                         | Hollow fiber      |
| Filtration mode              | External pressure |
| Weight cutoff (kDa)          | 100               |
| Contact angle ( $^{\circ}$ ) | 56.5              |
| Inner diameter (mm)          | 0.85              |
| Outside diameter (mm)        | 1.45              |

tank to reduce fouling. UF was operated in a constant flux, and change of TMP was taken to express the extent of fouling.

### 2.2. Raw water supply

The raw water was taken from the Songhua River in northeast of China, which represented a kind of typical surface water in China. The main water quality characteristics of raw water are summarized in Table 2.

Table 2  
Characteristics of raw water

| Parameters                                | Results     |
|---|-------------|
| Turbidity (NTU)                           | 19.5–21.2   |
| pH  | 7.62–7.72   |
| DOC (mg/L)                                | 7.718–7.832 |
| UV <sub>254</sub> ( $\text{cm}^{-1}$ )    | 0.088–0.096 |
| Total hardness ( $\text{CaCO}_3$ ) (mg/L) | 75.5–81.0   |
| Total alkalinity (mg/L)                   | 62.3–72.0   |
| Water temperature ( $^{\circ}\text{C}$ )  | 22.0–24.0   |

### 2.3. Experimental methods

A short-time flux-step method [18–20] was used to evaluate the effect of different operation modes on critical flux, which was determined by averaging the flux in which the TMP starts to increase and its former step flux. Different aeration air flow rates and frequency were studied and the filtration modes included continuous filtration without aeration, continuous filtration combined with continuous aeration, continuous filtration combined with intermittent aeration, and intermittent filtration combined with intermittent aeration. Aeration was operated for 1 min on and 4 min off in the operation of continuous filtration combined with intermittent aeration. For the mode of intermittent filtration combined with intermittent aeration condition, aeration was operated when the filtration stopped and the operation was 4 min off and 1 min on. Coagulation experiments were performed with jar tests, and polymer aluminum chloride (Act-view Carbon Technology Inc., China) was used as coagulant with an addition of 10 mg/L and sedimentation of 30 min being after that. Before every test was done, clean membrane modules were dipped in pure water for 5 d and the pure water was replaced every day.

## 3. Results and discussion

### 3.1. Effect of aeration on critical flux

#### 3.1.1. Effects of aeration modes and air flow rates on critical flux

Cake layers which were formed on the membrane surface during the filtration could be brushed off by friction of bubbles and the libration of membrane fiber in aeration process, which could be used as an effective method to control membrane fouling. Effects of different air flow rates on critical flux for continuous aeration are shown in Fig. 2. Critical flux of UF was 17.5 L/m<sup>2</sup> h when raw water was filtered directly without aeration. For continuous aeration, critical fluxes were 17.5, 22.5, and 22.5 L/m<sup>2</sup> h in the air flow rate of 2.5, 10, and 40 m<sup>3</sup>/m<sup>2</sup> h, respectively. Results showed that critical flux could be increased by 5 L/m<sup>2</sup> h in the air flow rate of 10 m<sup>3</sup>/m<sup>2</sup> h. It can be inferred from the results that critical flux could be improved by continuous aeration, as the air flow rate reached up to 10 m<sup>3</sup>/m<sup>2</sup> h which could not be further improved by air flow rate. Results also indicated that reasonable air flow rate of aeration should be chosen in application, and oversized aeration could neither alleviate membrane fouling nor save energy.

Effects of different air flow rates on critical flux by intermittent aeration are shown in Fig. 3. Critical fluxes were 17.5, 17.5, 22.5, and 22.5 L/m<sup>2</sup> h in the air flow rate of 10, 20, 40, and 80 m<sup>3</sup>/m<sup>2</sup> h, respectively. It can be seen that critical flux could be improved by intermittent aeration, when the air flow rate reached up to 40 m<sup>3</sup>/m<sup>2</sup> h which also could not be further improved by air flow rate. Results indicated that critical flux could not be greatly improved by aeration regardless of the air flow rate for continuous filtration.

#### 3.1.2. Alleviation of fouling by aeration in long-term run

Effects of aeration on TMP development in long-term filtration are shown in Fig. 4. Three kinds of flux were chosen for the principle that one flux (25 L/m<sup>2</sup> h) was beyond that of filtrating raw water directly without aeration (17.5 L/m<sup>2</sup> h), and two fluxes (17 and 12 L/m<sup>2</sup> h) below 17.5 L/m<sup>2</sup> h. It can be seen that the duration reaching up to the same TMP could be extended by both continuous aeration and intermittent aeration, and UF membrane fouling was alleviated. For the flux of 25 L/m<sup>2</sup> h, TMP always increased quickly when filtrating raw water without aeration, which had a low-speed increase in a period of 18 d, and then a high-speed increase for continuous aeration and intermittent aeration. The duration was about 7 d for TMP reaching up to 69.5 kPa for filtration without aeration, while it was about 23 d reaching up to the same TMP for aeration. For the flux of 17 L/m<sup>2</sup> h, TMP increased quickly in filtration without aeration, which reached up to 67 kPa after 17 d duration. Development of TMP experienced a low-speed increase and then a high-speed increase for both continuous aeration and intermittent aeration. Period of the low-speed increase to the TMP of 48.5 kPa lasted for 30 d for continuous aeration, and about 27 d for intermittent aeration. For the flux of 12 L/m<sup>2</sup> h, development of TMP experienced a low-speed increase for about 24 d and then a high-speed increase when filtrating raw water without aeration, while the TMP nearly did not increase for continuous aeration and intermittent aeration. It could be inferred from the results that continuous aeration had a greater potential to reduce fouling than intermittent aeration, especially at the flux below the critical flux determined by the flux-step method. Membrane fouling could be effectively alleviated by aeration which is usually a necessary mode of continuous UF operation [4,5,7,21]. Results also indicated that the critical flux determined by the flux-step method was higher than the actual

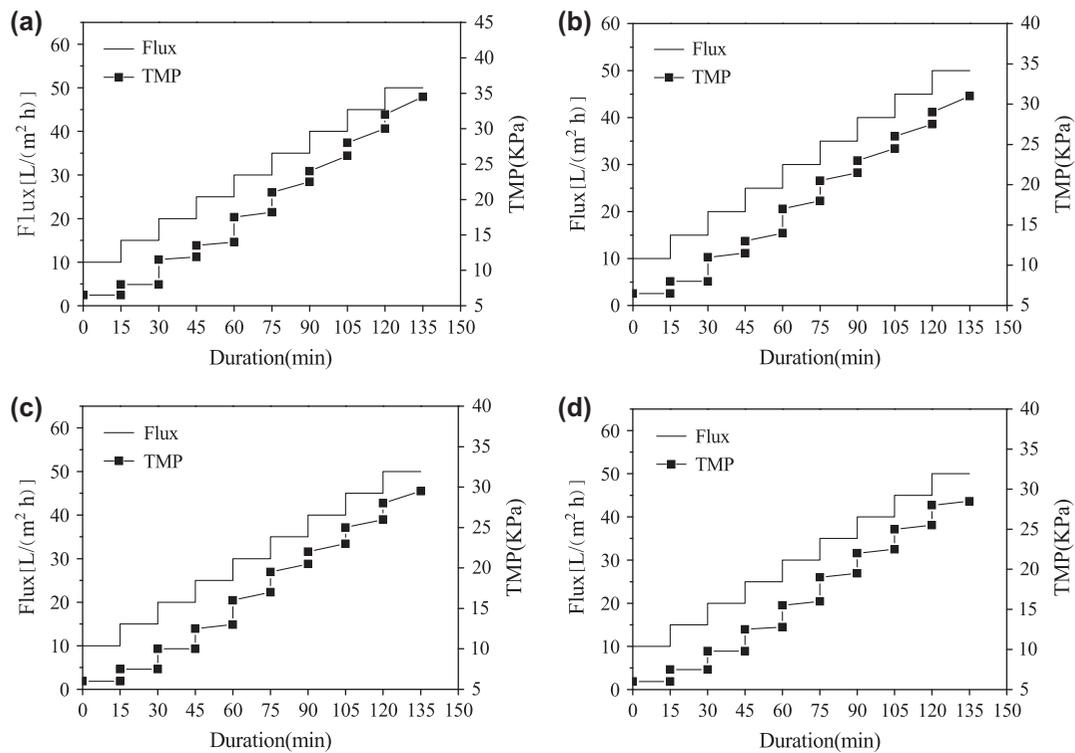


Fig. 2. Effect of aeration air flow rate on critical flux of UF membrane under continuous aeration condition. (a)  $0 \text{ m}^3/\text{m}^2 \text{ h}$ ; (b)  $2.5 \text{ m}^3/\text{m}^2 \text{ h}$ ; (c)  $10 \text{ m}^3/\text{m}^2 \text{ h}$ ; and (d)  $40 \text{ m}^3/\text{m}^2 \text{ h}$ .

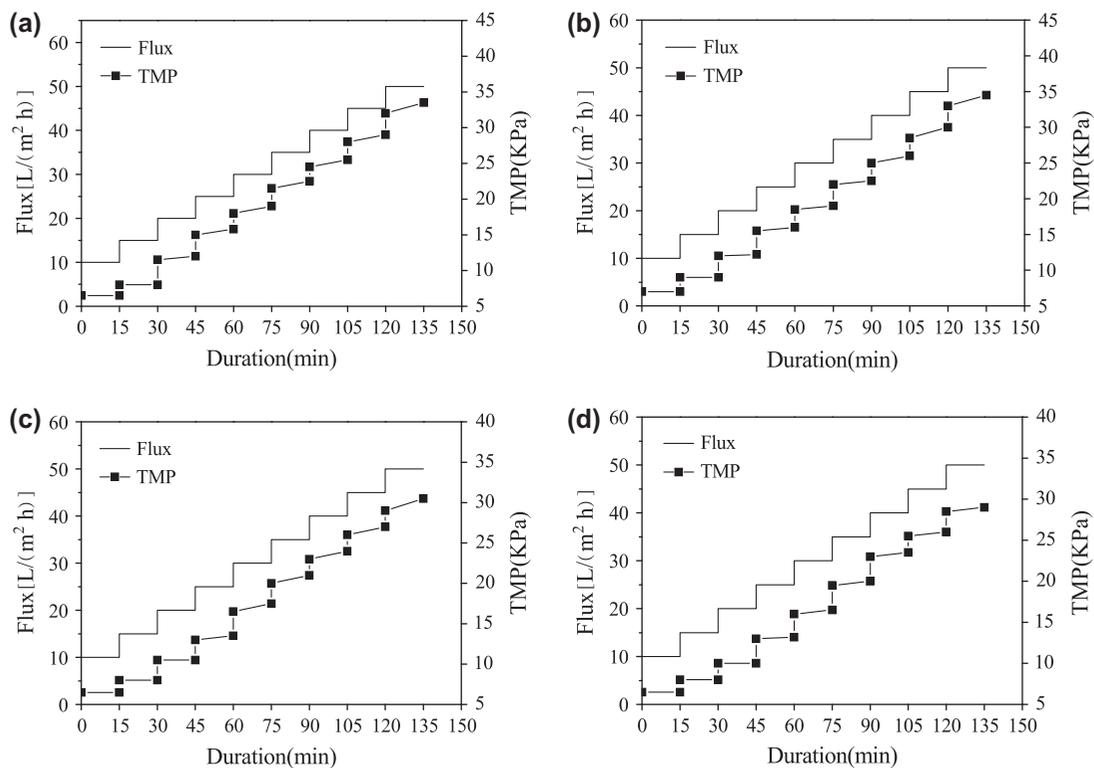


Fig. 3. Effect of aeration air flow rate on critical flux of UF membrane under intermittent aeration condition. (a)  $10 \text{ m}^3/\text{m}^2 \text{ h}$ ; (b)  $20 \text{ m}^3/\text{m}^2 \text{ h}$ ; (c)  $40 \text{ m}^3/\text{m}^2 \text{ h}$ ; and (d)  $80 \text{ m}^3/\text{m}^2 \text{ h}$ .

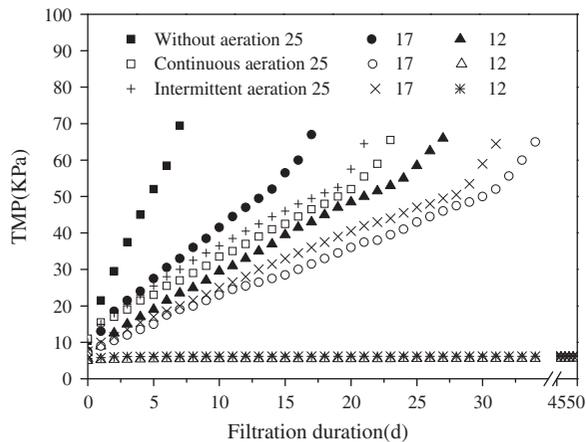


Fig. 4. Effect of aeration on TMP with different air flow rates in long-term run.

critical flux in application, but it could have a positive effect on the operation in the application.

### 3.2. Effects of filtration modes on critical flux

#### 3.2.1. Effects of intermittent filtration combined with and without aeration on critical flux

Effects of two different filtration modes on critical flux are shown in Fig. 5. Critical fluxes were 17.5 and 22.5 L/m<sup>2</sup> h for intermittent filtration without aeration and intermittent filtration combined with aeration, respectively. Results showed that critical flux could not be improved by intermittent filtration without aeration, but could be improved by 5 L/m<sup>2</sup> h for intermittent filtration combined with aeration. Filtration duration could be reduced by intermittent filtration mode, and the fouling on the membrane surface was less serious than that for

continuous filtration, but would further happen as filtration went on. As the intermittent filtration combined with aeration, the cake layers could be partly removed by liberation of membrane fiber and friction of bubbles in aeration, and a dense layer could not be formed.

#### 3.2.2. Effects of filtration modes on fouling in long-term run

Effects of filtration modes on TMP development in long-term filtration are shown in Fig. 6. Three kinds of flux were chosen for the principle that one flux (25 L/m<sup>2</sup> h) was beyond that of filtrating raw water directly without aeration (17.5 L/m<sup>2</sup> h), and two fluxes (17 and 12 L/m<sup>2</sup> h) below 17.5 L/m<sup>2</sup> h. It can be seen that the increase speed of TMP development was slightly lower for intermittent filtration than that of continuous filtration due to less filtration duration, but fouling was not greatly reduced. Development of TMP was greatly slower for intermittent filtration combined with aeration than that of continuous filtration. At the flux of 25 L/m<sup>2</sup> h, TMP experienced a low-speed increase for 18 d and then a high-speed increase that reached up to 64.5 kPa after 21 d operation. The duration was about 14 d for continuous filtration to reach up to the same TMP. At the flux of 17 L/m<sup>2</sup> h, the low-speed TMP increase lasted for 26 d and end TMP was 64 kPa after 30 d operation, which was longer than continuous filtration by 15 d. TMP nearly did not increase for intermittent filtration combined with aeration at the flux of 12 L/m<sup>2</sup> h. Results indicated that fouling could be effectively reduced by intermittent filtration combined with aeration for the possible reason that the liberation of membrane fiber and friction of bubbles had a better effect on cake layer shedding when the membrane stopped to filtration.

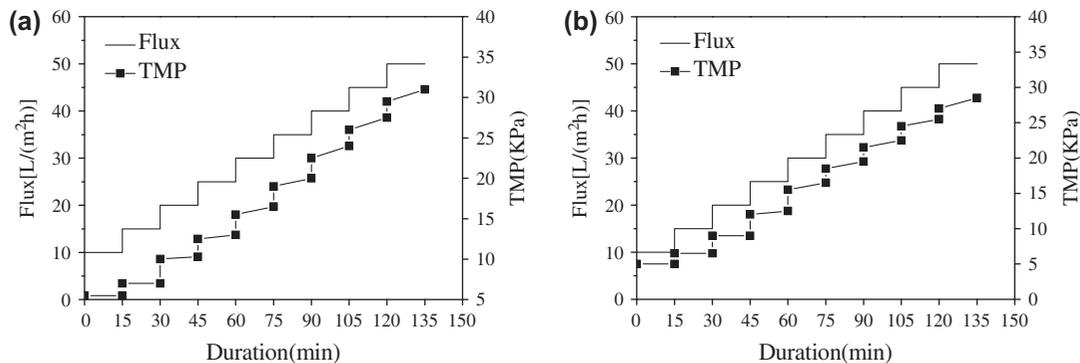


Fig. 5. Effect of filtration modes on critical flux. (a) Intermittent filtration without aeration and (b) intermittent filtration combined with aeration.

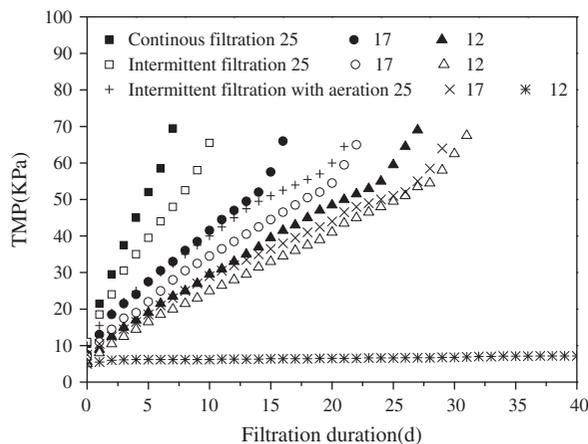


Fig. 6. Effect of operating modes on TMP of UF membrane in long-term run.

### 3.3. Effects of coagulation pretreatment on critical flux

#### 3.3.1. Effects of coagulation and coagulation combined with aeration on critical flux

Effects of coagulation combined with different filtration modes and aeration on critical flux are shown in Fig. 7. Critical fluxes were 27.5, 32.5, and 32.5 L/m<sup>2</sup> h for continuous filtration without aeration, continuous filtration combined with continuous aeration, and intermittent filtration combined with intermittent aeration, respectively in filtrating the water treated by coagulation and sedimentation. Results indicated that critical flux could be improved by 10 L/m<sup>2</sup> h by coagulation and sedimentation. Suspended particles and macromolecular organic substances, which are the main pollutants resulted in fouling, could be effectively removed by coagulation [22,23]. Accumulation of pollutants on membrane surface and within the membrane pores is less serious for coagulation pretreatment than that without pretreatment [9,10]. Continuous aeration could alleviate membrane fouling by libration of membrane fiber and friction of bubbles in the process of filtration. Coagulation coupled with continuous aeration could produce a synergy effect on critical flux improvement. Intermittent filtration could make the membrane have relaxation time in the filtration progress, and aeration could have a better effect on fouling control during the relaxation period. Integration of coagulation and continuous aeration or coagulation and intermittent filtration combined with intermittent aeration could significantly improve critical flux, which could be better operation modes for UF in drinking water treatment.

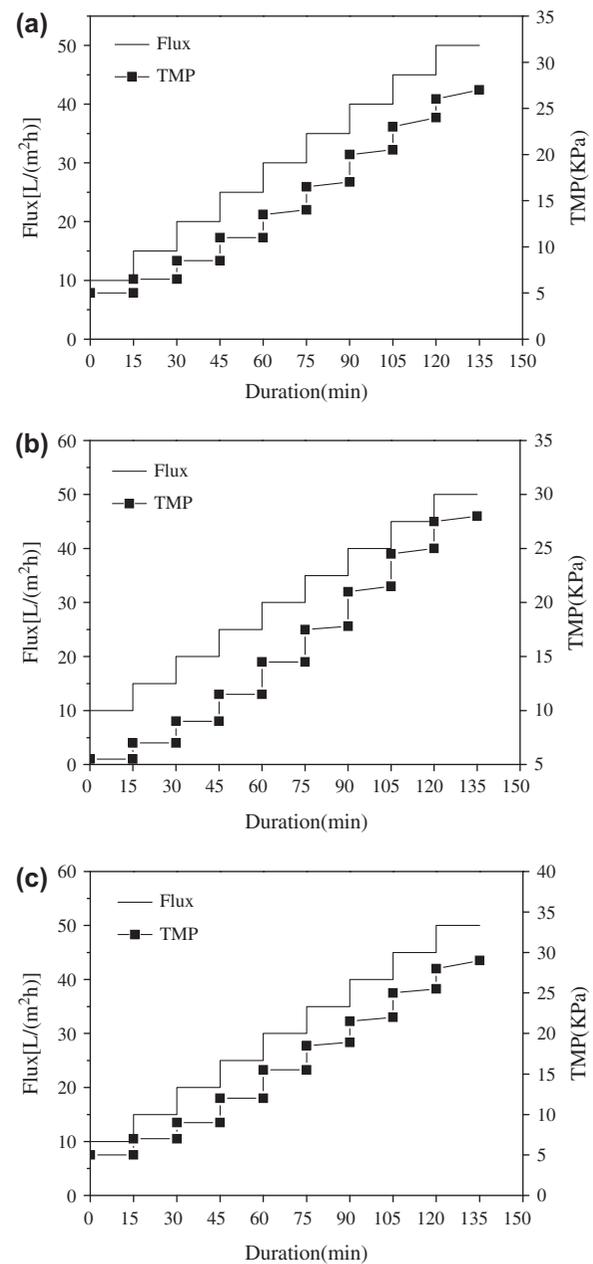


Fig. 7. Effect of coagulation on critical flux. (a) Continuous filtration; (b) continuous filtration with continuous aeration; and (c) intermittent filtration with intermittent aeration.

#### 3.3.2. Effects of coagulation on fouling in long-term run

Effects of coagulation on TMP development in long-term filtration are shown in Fig. 8. Three kinds of flux were tested as 25, 20, and 15 L/m<sup>2</sup> h. At the flux of 25 L/m<sup>2</sup> h, TMP increased in a low speed for about 20 d and then in a high speed until 26th day for

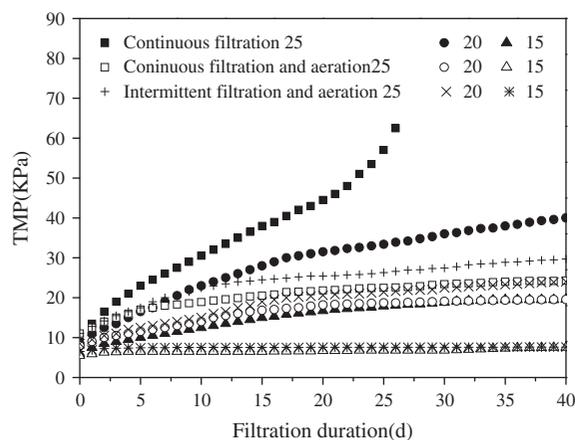


Fig. 8. Effect of coagulation on TMP of UF membrane in long-term run.

continuous filtration. Different with continuous filtration, TMP grew rapidly in the first 4 d and then increased in a very low speed for 40 d with a TMP increase from 11 to 24.3 kPa for continuous filtration combined with continuous aeration, which was similar as intermittent filtration combined with intermittent aeration. At the flux of 20 L/m<sup>2</sup> h, TMP for all the three kinds of operation mode increased very slowly, which grew from 8.5 to 40 kPa, 8.0 to 19.5 kPa, and 8.5 to 24.1 kPa for continuous filtration without aeration, continuous filtration combined with continuous aeration, and intermittent filtration combined with intermittent aeration, respectively. At the flux of 15 L/m<sup>2</sup> h, TMP increased in a very low speed in 40 d and reached up to an end value of 19.6 kPa for continuous filtration. Compared with continuous filtration, TMP nearly had no increase in the whole period of run for other two modes, which might be the actual critical flux in application.

Results indicated that coagulation and coagulation combined with intermittent filtration or aeration could significantly improve critical flux in long-term filtration. This improvement would be better for the operation below the flux determined by flux-step tests. It can also be inferred that critical flux determined by flux-step tests could help to instruct the operation of UF in the application.

#### 4. Conclusion

Factors influencing the critical flux of UF for drinking water treatment were systematically investigated in both short-term and long-term tests. Critical flux could be improved when the air flow rate reached up to 10 m<sup>3</sup>/m<sup>2</sup> h for continuous aeration and 40 m<sup>3</sup>/m<sup>2</sup> h

for intermittent aeration, which could not be further improved by increasing air flow rate. Continuous aeration had a better effect on fouling alleviation than intermittent aeration in long-term filtration. Intermittent filtration combined with aeration could improve critical flux and reduce fouling a lot in long-term filtration. Coagulation and sedimentation could greatly improve critical flux, and integration of coagulation and continuous aeration or coagulation and intermittent filtration combined with intermittent aeration had a better effect on critical flux improvement. Critical flux determined by flux-step tests could help to instruct the UF operation in application, which could be determined by short-term flux-step tests at first for a given water quality in actual application. Appropriate operation modes including intermittent filtration and aeration should be chosen within the reasonable flux range for applications in drinking water treatment.

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