Factors Affecting Reversible Pollution of Immersed Ultrafiltration Membrane in Treatment of Surface Water: Pilot Scale Studies

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Abstract: The main factors (i.e., permeate flux, mixture characteristics, and membrane materials) affecting the pollution of immersed ultrafiltration membrane (immersed-UF) during the treatment of surface water were investigated through pilot scale studies. The variation of the running pressure was used as an indicator of membrane fouling. It is indicated that the rate of running pressure increased rapidly when the actual flux exceeded the critical flux, which exhibited to be $20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ as for the membrane used in this study. The higher concentrations of turbidity and organic matter in the mixture contributed to the more significant increase of running pressure and the more serious membrane fouling. The higher temperature could lessen the membrane fouling to a certain extent. It was observed that membrane materials showed obvious effects on membrane fouling. The running pressure of Polyvinylidene Fluoride (PVDF) membrane was lower than that of chemically-modified Polyvinyl Chloride (PVC) at the same permeate flux, indicating the more promising potential of anti-pollution for PVDF membrane.

Keywords: Immersed Ultrafiltration membrane; Membrane fouling; Critical flux; Membrane materials

INTRODUCTION

The immersed ultrafiltration membrane (immersed-UF) device are receiving more and more attention nowadays. The hollow fiber membrane in the device is directly submerged in the reactor. During operation, the water flows from outside to the inside surface under the negative pressure in the membrane cavity, which is produced through the pump suction. The immersed-UF offers advantages such as low energy consumption, high reclamation rate, satisfactory filtration quality, and low land-demand (Thomas *et al.*, 1998). Additionally, the immersed-UF works well when high content of suspended solids is present, which is beneficial to its application in the treatment of surface water with variously contaminative degree. Furthermore, the immersed-UF is not only available in a new waterworks, but also can be installed in the clarification or empty sand filtration for the reconstruction of an existing waterworks with conventional treatment processes (Diana and Mike, 1998; Pierre *et al.* 1998).

In the past decade, many studies have been devoted to the feasibility of employing the immersed-UF to treat the slightly-polluted water, and much effort focused on the removal of pollutants. However, the membrane fouling and its causes are not well investigated for the immersed-UF. The membrane fouling includes reversible pollution and irreversible pollution (Li and Dong, 2008). The reversible pollution, which can be resumed by physical cleaning, may be indicated from the decline of permeate flux or the increase of running pressure (Lidietta and Enrico, 2000; Li and Dong, 2008). If the permeate flux keeps constant during operation, membrane fouling can be indirectly indicated from the running pressure variation. It is of crucial importance to understand main causes and to propose feasible strategies to avoid reversible pollution, which is beneficial to decrease the energy demand and lengthen the lifestyle of the immersed-UF.

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This study aims to investigate the effects of different factors, i.e., permeate flux, mixture characteristics, and membrane materials, on the reversible pollution of the immersed-UF. The immersed-UF was directly used to treat the polluted surface water without any pretreatment, and the variation of running pressure was consistently followed under the same operational conditions.

2. Materials and methods

2.1 Membrane description

Two kinds of immersed Ultrafiltration membrane made of Polyvinylidene Fluoride (PVDF) and Polyvinyl Chloride (PVC) alloy, respectively were employed in the experiments. And each membrane module was fixed with three screens membrane. The material of chemically-modified PVC was studied primarily in the experiment. Characteristics of the membranes were shown in Table 1.

Table 1: Main parameters of the membrane

Parameter	UF membrane	
Type	Hollow fiber	Hollow fiber
Material	chemically-modified PVC	PVDF
Temperature-range (°C)	5-38	5-40
pH-range of operation	2-13	2-12
Molecular weight cut off (Da)	100,000	150,000
Hollow fiber internal diameter(mm)	1.00	0.85
Hollow fiber external diameter(mm)	1.66	1.45
One screen membrane surface area (m ²)	8.03	8.30
Membrane surface area (m ²)	24	24
Packing density (m ² membrane area per m ² footprint)	48	48
Operation pressure recommended (MPa)	0.01-0.06	0.01-0.06
Effective length (mm)	1,360	1,360

2.2. Ultrafiltration test apparatus and procedure

Figure 1 showed the schematic diagram of the immersed UF experimental facility. And the ability of the pilot study was 5.0 m³·h⁻¹. Source water was injected in the raw water pool by submersible pump, and then pumped to the pool for the immersed UF. At the base of the membrane module, perforated pipes were fit for aeration. The permeate water was produced by suction pump. Backwashing was performed by pumping the permeated water from the storage pool.

The experiment apparatus had flow and pressure indicators, and had the capabilities of automatic suction, aeration and backwashing. The running mode of the immersed membrane was permeate flux invariable. The running parameters in the experiment were as follow as the interval time of aeration 30 min, the aeration time 3min, the intensity of aeration 45 m³·m⁻²·h⁻¹, the period

of backwashing and sewage discharging 24 hr, the backwashing time 5 min, and thereinto, the intensity of backwashing $60 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, the intensity of aeration $45 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$.

1, raw water tank ; 2, feed pump ; 3, 11, 13, liquid flow meter ; 4, immersed membrane module ; 5, membrane pool ; 6, gas flow meter ; 7, gas pressure gauge ; 8, blower ; 9, vacuum pressure gauge ; 10, suction pump ; 12, back-washing pressure gauge ; 14, back-washing pump ; 15, storage tank

Figure 1: Flow diagram of the experiment

2.3. Raw water characteristics

One river water was used as the raw water. The average characteristics were as follows as pH 8.03~8.29, Turbidity 9.50~17.4 NTU, Temperature 5.0~20.5 °C and COD_{Mn} 4.65~5.30 $\text{mg} \cdot \text{L}^{-1}$.

2.4. Analysis apparatus and methods

The water samples were analyzed for chemical oxygen demand (COD_{Mn}), Turbidity, Temperature and pH. COD_{Mn} was determined according to the method described in Sakol and Konieczny (USEPA, 1983). The turbidity was measured by a Hach turbidity meter (model 2100N). The pH was measured by a pH meter (model PHs-3c).

3. Results and discussion

3.1. Effect of permeate flux on reversible pollution of immersed-UF

It was reported that there existed a critical permeate flux for a specified membrane module, and the membrane fouling would be severe if the operation flux was above the value. However, the membrane fouling would be well mitigated and the cycles of chemical cleaning would be satisfactorily prolonged if the running flux was controlled to be below this value (Field *et al.*, 1995; Cho and Fane, 2002). Figure 2 investigated the effect of membrane running pressures at each specified permeate flux when the temperature was 10.5°C.

Figure 2: The effect of permeate flux on membrane running pressure

As being indicated in Fig.2a, the initial running pressures increased steadily at higher flux values. When the flux increased from 15 L·m⁻²·h⁻¹ to 35 L·m⁻²·h⁻¹, the initial running pressures correspondingly increased from 0.0085MPa to 0.021MPa. The variation of running pressures was observed to show different trends at different fluxes. When the permeate flux was 15 L·m⁻²·h⁻¹, the running pressure kept constant in 24 hours. The higher flux of 20 L·m⁻²·h⁻¹ contributed to slight increase of running pressures, with the increasing rate of 13.8% in one period. The further increase of permeate flux to 30 L·m⁻²·h⁻¹ and 35 L·m⁻²·h⁻¹ led to more significant increase of running pressure, which was as high as 123% and 152%, respectively. The running pressure was observed to be positively correlated to the permeate flux ($R^2=0.985$), as being indicated from Figure 2b.

Basing on these results, the critical flux of the chemically-modified PVC membrane used in this study was determined to be $20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, which was used as the operation parameter to investigate the effects of mixture qualities, temperature, and materials on membrane fouling in the following experiments.

3.2. Effects of mixture characteristics on membrane fouling

In engineering to reconstruct a waterworks with conventional processes, the immersed-UF may be installed in the pools of sedimentation, clarification, and filtration to substitute these processes. The influent of immersed-UF shows great difference in the quality under these conditions, which is quite different from that of inner pressure UF. Furthermore, in one period of backwashing and sewage discharging, the concentrations of different pollutants in the membrane pool, i.e., turbidity, and organic matter, would increase with longer time due to the enriching effects, which leading to the variation of mixture characteristics and the possible side effects on membrane fouling.

3.2.1. Effect of turbidity on membrane fouling

Figure 3 indicated that the running pressure increased with higher turbidity in the membrane pool, which showed a binomial relationship between them ($R^2=0.985$). The rate of running pressure increase was relatively low when the turbidity is below 60 NTU. However, with time prolonging and the increase of turbidity in membrane pool, the running pressure increased significantly and indicated the serious membrane fouling under this condition.

(Temperature = 9.0 °C, Permeate flux = $20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, chemically-modified PVC)

Figure 3: The effect of the mixture water turbidity on membrane running pressure

The formation of pollutants layer on the membrane surface was important for the increase of running pressure. During membrane aeration, pollutants layer would break off into the membrane pool. However, the enrichment of suspended solids contributed to the formation of compact and dense layer on membrane surfaces, and to the severe phenomenon of membrane aperture adsorption and jam. On the other hand, the polarization phenomena would be distinct with the accumulating of

pollutants in the pool (Zia *et al.*, 2008). Consequently, the resistance of membrane filtration became remarkable with the increased turbidity in the membrane pool, which required higher running pressure to keep constant permeate flux.

3.2.2. Effect of organics on membrane fouling

The main course for membrane fouling was reported to be the adsorption and jam of organics species (Kerry and Mark, 2002). Figure 4 showed the relationship between running pressure and the organics content (COD_{Mn}) in the membrane pool in one period.

(Temperature = 11.5 °C, Permeate flux = 20 L·m⁻²·h⁻¹, chemically-modified PVC)

Figure 4: The effect of the mixture water COD_{Mn} on membrane running pressure

As being indicated, the running pressure was positively correlated with higher COD_{Mn} concentrations in the membrane pool, showing an exponential relationship between these two parameters. Similar to that of turbidity, running pressure showed higher rate of increasing at higher COD_{Mn} concentration in the pool. However, there did not show a critical value of COD_{Mn} concentration, above which the running pressure began to obviously increase. The higher organics content facilitated the adsorption of organics on the membrane surface or pore (Yiantsios and Karabelas, 2001; Wang *et al.*; 2002), which led to the formation of compact gel layer on the membrane surface. Additionally, the hypo-grade film gel layer, which has been demonstrated to be dense (Schlesinge *et al.*, 2005), would form on the membrane surface with higher organics content.

3.2.3 Effect of temperature on membrane fouling

Temperature was reported to show effect on membrane fouling. The high temperature could not only reduce the viscosity of mixture to facilitate membrane filtration, but also change the thickness of filtration layer on surface and aperture of membranes to alleviate membrane fouling (Nilsson *et al.*, 2006). Figure 5 showed the effect of temperature on the running pressure of immersed-UF.

(Permeate flux = $20 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, chemically-modified PVC)

Figure 5: The effect of temperature on membrane running pressure

The running pressure showed a linear relation with temperature, and the higher temperature decreased the running pressure and alleviated membrane fouling (Figure 5a). When the permeate flux was controlled to be $20 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, the running pressure declined with higher temperature.

Figure 5b showed the variation of running pressure at different temperatures of 7.5°C, 15.0°C, and 20.5°C. As being indicated, the increasing rate of running pressure at 7.5°C was faster than those at 15.0°C and 20.5°C, indicating the positive effect of temperature on membrane fouling alleviation. The elevated temperature enhanced the activity, reduced viscosity of water molecular, and increased mass transfer coefficient, therefore diminishing the filtration resistance and reducing the running pressure. At low temperature, the water molecular showed high viscosity and low activity, which resulted in the increase of filtration resistance and the running pressure.

3.3. Effect of membrane materials on membrane fouling

PVDF and chemically-modified PVC are the two kinds of membrane that were widely used in engineering. Figure 6 compared the variation of running pressure at different permeate flux between these two membrane materials. The running pressure of PVDF was obviously lower than that of PVC at the same permeate flux, indicating the better permeability of PVDF than that of PVC.

Figure 6: The difference of running pressure on permeate flux with different membrane materials

Figure 7: The effect of membrane materials on membrane running pressure

Figure 7 compared the trends of running pressure variation in four periods between PVDF and chemically-modified PVC at the same experimental conditions, confirming that PVDF required lower energy than PVC. Additionally, PVDF showed the lower rate of running pressure increase than chemically-modified PVC did in this experiments. The average rate of running pressure

increase in one period of backwashing with PVDF and chemically-modified PVC was respectively 33.3% and 41.5%. It was indicated that PVDF showed stronger ability of anti-pollution than that of chemically-modified PVC.

The different membrane materials differed greatly in the size of membrane aperture, the thickness and degree of membrane dense layer, which resulted in the difference in the anti-pollution ability between PVDF and PVC. Additionally, it was reported that the extent of membrane fouling, which resulted from the formation of gel layer and the jamming of membrane aperture, varied between different materials (Choo, 2000). The membrane fouling trend of a specified material was consistent with the dispersion component trend of membrane surface tension, and the greater of dispersion component trend of membrane surface tension was, the membrane was more easily polluted.

CONCLUSIONS

The membrane fouling of immersed-UF was greatly affected by permeate flux, mixture characteristics, and membrane materials. There existed a critical value ($20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) of permeate flux for the chemically-modified PVC membrane used in this study. The increase of turbidity and organics content in the membrane pool led to the more significant membrane fouling. Temperature significantly affected the running pressure and the higher temperature is beneficial to alleviate membrane fouling. Comparatively, the PVDF showed higher ability of anti-pollution than that of chemically-modified PVC.

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