

# Effect of Transmembrane Pressure on Performance of Dynamic Membrane for Treatment of Wastewater Containing Humic Acid

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**Abstract:** The effects of transmembrane pressure on the performance of treatment of wastewater containing humic acid using dynamic membrane were studied, *in which* the relative permeate fluxes, pollutant removal efficiency and membrane fouling characteristics were investigated. The results indicated that the removal rates of pollutant UV<sub>254</sub> and UV<sub>436</sub> all gradually reduced with running time, in which the initial removal rates of pollutant UV<sub>254</sub> and UV<sub>436</sub> all exceed 90% and 97%, respectively. In steady states, UV<sub>254</sub> removal rates from high to low were 0.11Mpa, 0.06Mpa, 0.25Mpa and 0.16Mpa. The resistance of fouling dynamic layer took up most of the total membrane resistance. The fouling degree of the dynamic membrane layer was much more serious than that of the support. The resistances of the fouling dynamic layer and total fouling membrane both increased with the rise of transmembrane pressure. The pore blockage rates of support increased from 0.06Mpa to 0.16Mpa, and then reduced as the transmembrane pressure increased to 0.25Mpa.

## Introduction

Nowadays, the pollution problem for nature water body by natural organic matters can not be ignored with the rapid development of industrialization and urbanization. The humic acid is the typical representative of these natural organic compounds which are easy to form DBPs and carcinogens THMs during the disinfection process in waterworks<sup>[1,2]</sup>. Therefore, how to effectively remove the humic acid in natural water is very urgent at present.

Among the many wastewater treatment technologies, membrane technology is one of best promising alternatives which has demonstrated many advantages, such as a small footprint, water quality stability, easy operation, etc.<sup>[3,4]</sup>. Dynamic membrane, as a special kind of membrane technology, has already been well used to treat many kinds of wastewater, such as oily wastewater, dyeing wastewater, domestic wastewater, etc.<sup>[5,6]</sup>. However, the treatment of wastewater containing humic acid using dynamic membrane has rarely been reported. As such, the influence of membrane pressure on the relative membrane flux, contaminants rejection rate and membrane fouling characteristics were discussed in the process of filtrating the wastewater containing humic acid.

## Experimental

### Main apparatus, instruments and materials

The experimental apparatus was described in the literature[7]. The Main instruments: ultraviolet spectrophotometer (UV752, Shanghai Hengping Scientific Instrument Co., Ltd.),

ultrasonic cleaning machine (KQ2200E, Kunshan Ultrasonic Instruments Ltd.Co., P.R.China), Test materials: single channel ceramic membrane ( $\alpha\text{-Al}_2\text{O}_3$ , length 400 mm, outside diameter 13 mm, inside diameter 9 mm, average pore diameter 1.0 microns, Tianya Membrane Separation Ltd.Co., P.R.China); Titanium dioxide (Sharp titanium crystal, average size 25 nm, Shanghai Yue Jiang Titanium Chemical Manufacture Ltd.Co., P.R.China). Humic acid (ash 10%, Tianjin Guangfu Chemical Industry Research Institute). Preparation and preservation methods of humic acid solution were the same with the literature [8].

## Analysis

At different time intervals, the permeate samples were collected by a graduated cylinder to measure the permeate flux  $J_t(\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1})$  which was calculated by Eq. (1). The relative membrane flux  $J_s$  (dimensionless) was calculated by Eq. (2):

$$J_t = \frac{\Delta V}{pDl\Delta t} \quad (1)$$

$$J_s = \frac{J_t}{J_o} \times 100\% \quad (2)$$

where  $D(\text{m})$  and  $l(\text{m})$  were the outside diameter and length of the ceramic membrane.  $\Delta V(\text{L})$  was the permeate volume.  $\Delta t(\text{h})$  was the time of collecting permeate.  $J_o(\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1})$  was the initial membrane flux. After measurement, the permeates were poured back into the liquid storage tank, so as to maintain the liquid in the storage tank basically unchanged. The  $UV_{254}(\text{cm}^{-1})$  and  $UV_{436}(\text{cm}^{-1})$  can be used to stand for the humic acid and chroma pollutants. The rejection rates of the pollutants of  $UV_{254}$  and  $UV_{436}$  were calculated by Eq. (3):

$$h = 1 - \frac{A}{A_0} \quad (3)$$

Where  $A$  represents the permeate absorbance of  $UV_{254}$  or  $UV_{436}$ .  $A_0$  represents the initial absorbance of permeate.  $\eta(\%)$  represents the pollutant removal rate. The hydraulic filtration resistances were calculated using the resistance model by Eq. (4):

$$J = \frac{\Delta P}{hR} \quad (4)$$

where  $\Delta p(\text{Pa})$  represents transmembrane pressure,  $\eta(\text{Pa}\cdot\text{s})$  represents the permeate viscosity, and  $R(\text{m}^{-1})$  represents the filtration resistance. The total membrane resistance can be composed of several parts described as follows:

$$R_t = R_o + R_{dr} + R_{sr} + R_d = \frac{\Delta P}{hJ_t} \quad (5)$$

$$R_o = \frac{\Delta P}{hJ_o} \quad (6)$$

$$R_o + R_d = \frac{\Delta P}{hJ_d} \quad (7)$$

$$R_o + R_{sr} = \frac{\Delta P}{hJ_c} \quad (8)$$

where  $R_o$ ,  $R_{dr}$ ,  $R_{sr}$ ,  $R_d$  and  $R_t$  represent the resistances of new support, fouling dynamic layer, fouling support, dynamic layer, and the total fouling membrane, respectively. Before the test, at the end of forming dynamic membrane, at the end of filtration and after flushing off the fouling dynamic layer using tap water at a transmembrane pressure of 0.2 MPa, the pure water fluxes  $J_o$ ,  $J_d$ ,

$J_c$  and  $J_t$  were all measured at a transmembrane pressure of 0.15 MPa, respectively. The pore blockage rates  $f_j$  (%) of the support was calculated by Eq. (9):

$$f_j = \frac{J_0 - J_c}{J_0} \times 100\% \quad (9)$$

## Experimental procedure

Titanium dioxide was used as coating agent. The preparation methods of dynamic membrane was described in the literature[9]. After preparation, the valve was switched to separate the humic acid solution. During the operation, the permeate was realigned to the feed tank for recycling and the membrane fluxes and humic acid retention ratios were determined. The crossflow velocity, humic acid concentration and operating temperature maintained  $1.60 \text{ m}\cdot\text{s}^{-1}$ ,  $75\text{mg}\cdot\text{l}^{-1}$  and  $293\text{K}$ , respectively. The effects of transmembrane pressures 0.06, 0.11, 0.16 and 0.25Mpa were compared and investigated. Before the experiments, the support was thoroughly cleaned to make sure the pure water fluxes completely recovered.

## Results and discussion

### Relative membrane flux

During the process of treatment of wastewater containing humic acid using the dynamic membrane, the variations of the relative permeate fluxes at different transmembrane pressures were measured. The results are showed in Fig.1.

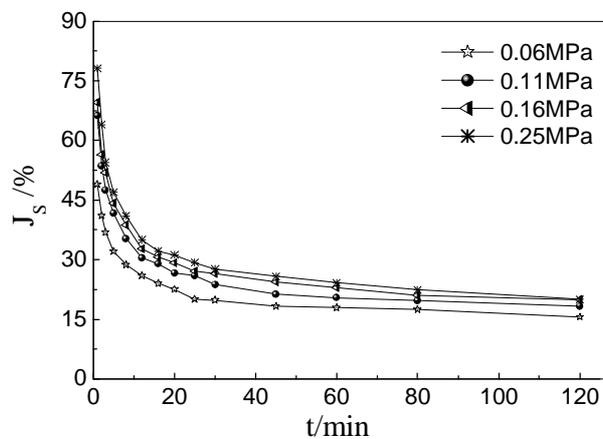


Fig. 1. Variations of relative permeate fluxes with running time

Fig.1 shows that all the relative permeate fluxes rapidly reduced and reached their steady states in about 30 min and then the membrane flux began to gradually reduce. The higher the transmembrane pressure, the higher the relative permeate flux. In the high range of transmembrane pressure, the permeate flux reduced more obvious than that of low range of transmembrane pressure. When the transmembrane pressure increased from 0.16Mpa to 0.25Mpa, the steady permeate flux increased little. The convective flow of solution towards the membrane could be enhanced with an increase in transmembrane pressure, leading to improvement of the permeate flux. On the other hand, the pollution layer deposited on the membrane by the pollutants is more dense, which could form higher filtration resistances. Thus, the permeate fluxes were not increased linearly with the transmembrane pressure in high range.

## Pollutants removal rates

During the process of treatment of wastewater containing humic acid using the dynamic membrane, the variations of the pollutant removal rates at different transmembrane pressures were measured. The removal rates of  $UV_{254}$  and  $UV_{436}$  are showed in Fig.2(a) and Fig.2(b), respectively.

Fig.2(a) shows that the removal rates of pollutant  $UV_{254}$  gradually reduced with running time at different transmembrane pressures. The initial removal rates all exceed 90%. In steady states,  $UV_{254}$  removal rate from high to low were 0.11Mpa, 0.06Mpa, 0.25Mpa and 0.16Mpa. Fig.2(b) shows that the removal rates of pollutant  $UV_{436}$  also gradually reduced with running time at different transmembrane pressures. The initial removal rates all exceed 97%. In steady states, the highest and lowest  $UV_{436}$  removal rate were at 0.11Mpa and 0.16Mpa. The pollutants could more easily flow towards the membrane and incline to penetrate the membrane, which would reduce the removal rates of the pollutants. The more dense fouling layer formed at higher transmembrane pressure could exhibit better retention performances. Then the pollutants removal rate at 0.25Mpa was higher than that at 0.16Mpa instead.

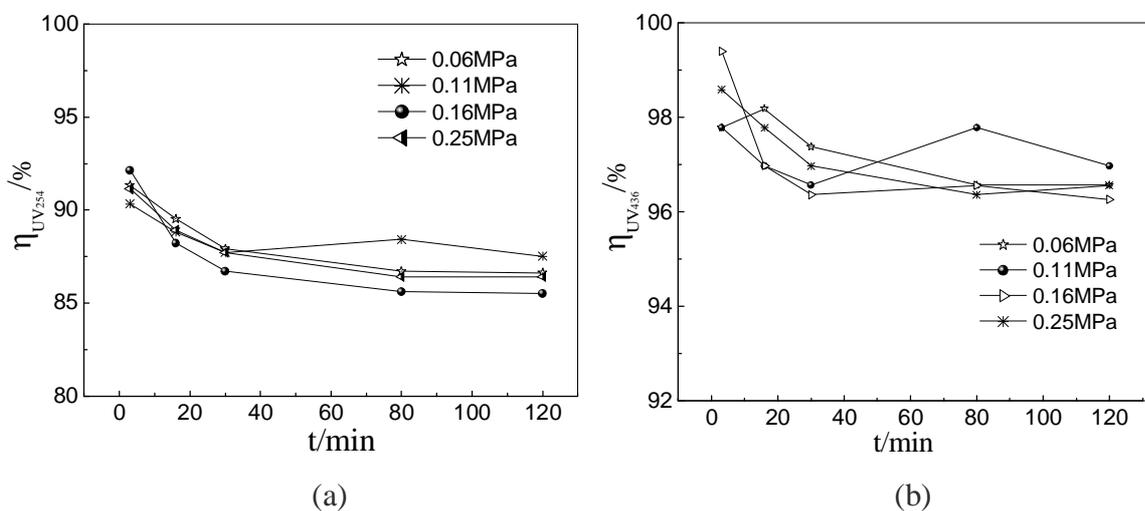


Fig. 2. Variations of the pollutant removal rates: (a)  $UV_{254}$ , (b)  $UV_{436}$

## Membrane fouling characteristic

After treatment of wastewater containing humic acid, the membrane resistance distribution and pore blockage rates of the support at different transmembrane pressure were calculated, respectively. The results are showed in Fig.3 and Fig.4.

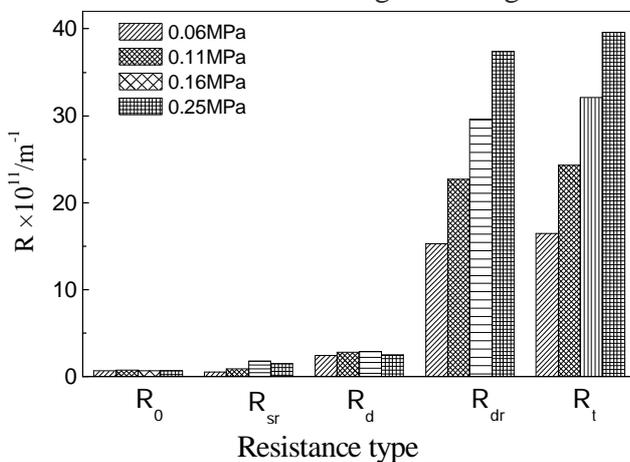


Fig. 3. Membrane resistance distribution at different transmembrane pressures

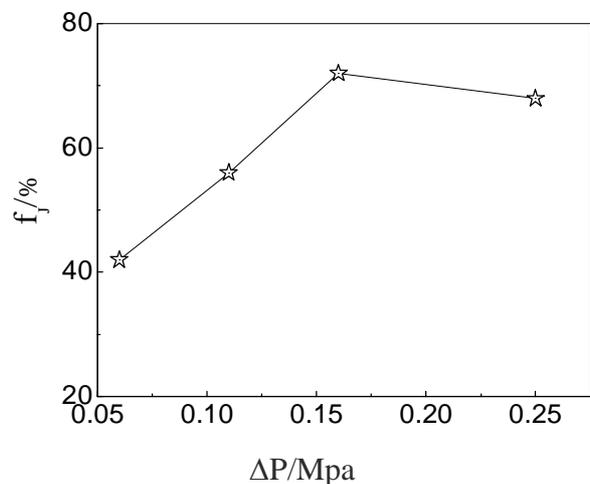


Fig. 4. Pore blockage rates of support at different transmembrane pressures

Fig.3 shows that the resistance of the fouling dynamic layer took up most of the total membrane resistance at different transmembrane pressure, which indicated that the fouling degree of the dynamic membrane layer was much more serious than that of the support. The resistances of the fouling dynamic layer and total fouling membrane both increased with the rise of transmembrane pressure. The resistance of the support pollution reached highest level at 0.16Mpa. Fig.4 shows that pore blockage rates of support increased from 0.06Mpa to 0.16Mpa, and then reduced as the transmembrane pressure increased to 0.25Mpa. In the process of filtration, the pollutants directly contact with the dynamic membrane layer, leading to its most serious pollution. The higher the transmembrane pressure, the more easily the pollutants depositing and blocking the membrane, and then the fouling degree of the dynamic membrane layer and support increased. On the other hand, the more dense fouling layer formed at higher transmembrane pressure could prevent the pollutants from enter into the pore of the support, leading to lower rate of the pore blockage at 0.25Mpa than that 0.16Mpa instead.

## Conclusions

- (1) The relative permeate fluxes increased with the rise of transmembrane pressure. The permeate flux reduced more obvious in high range of transmembrane pressure than that in low range of transmembrane pressure. The steady permeate flux increased little as the transmembrane pressure varied from 0.16Mpa to 0.25Mpa.
- (2) The removal rates of pollutant  $UV_{254}$  and  $UV_{436}$  all gradually reduced with running time, in which the initial removal rates of pollutant  $UV_{254}$  and  $UV_{436}$  all exceed 90% and 97%, respectively. In steady states,  $UV_{254}$  removal rates from high to low were 0.11Mpa, 0.06Mpa, 0.25Mpa and 0.16Mpa, and the highest and lowest  $UV_{436}$  removal rate were 0.11Mpa and 0.16Mpa.
- (3) The resistance of fouling dynamic layer took up most of the total membrane resistance. The fouling degree of the dynamic membrane layer was much more serious than that of the support. The resistances of the fouling dynamic layer and total fouling membrane both increased with the rise of transmembrane pressure. The resistance of the support pollution reached highest level at 0.16Mpa. The pore blockage rates of support increased from 0.06Mpa to 0.16Mpa, and then reduced as the transmembrane pressure increased to 0.25Mpa.

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## References

- [1]Krasner S.W. , Weinberg H. S. , Richardson S. D. , et al. Occurrence of a of disinfection new generation byproducts. *Environmental Science and Technology*, 2006,40(23): 7175-8185
- [2] Katsoufidou K , Yiantsios S G , Karabelas A J. A study of ultrafiltration membrane fouling by humic acids and flux recovery by backwashing: Experiments and modeling[J] *J .Membr. Sci.* ,2005, 266(1-2): 40-50.

- [3] Juang R. S., Lin K. H. Flux recovery in the ultrafiltration of suspended solutions with ultrasound. *J. Membr. Sci.*, 2004, 243(1-2): 115-124.
- [4] Jermann D, Pronk W, Meylan S. Interplay of different NOM fouling mechanisms during ultrafiltration for drinking water production[J]. *Water Res.*, 2007, 41(8): 1713-1722.
- [5] Pessoa de Amorim M T, Ramos I R A. Control of irreversible fouling by application of dynamic membranes. *Desalination*, 2006, 192(1-3): 63-67
- [6] Noor M J M M, Ahmadun F R, Mohamed T A, et al. Performance of flexible membrane using kaolin dynamic membrane in treating domestic wastewater [J]. *Desalination*, 2002, 147(1-3): 263~268.
- [7] Yang T, Ma Z F, Yang Q Y. Formation and performance of Kaolin/MnO<sub>2</sub> bi-layer composite dynamic membrane for oily wastewater treatment: effect of solution conditions[J]. *Desalination*. 2011, 270(1-3): 50-60.
- [8] Yan X J, Yu S L, Li L Z, et al. Effect of Inorganic Ions on Photocatalytic Degradation of Humic Acid[J]. *China water & wastewater (In Chinese)*, 2012, 8(13): 80-83.