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# Behaviours of Natural Organic Matter (NOM) in Ultrafiltration Membrane Filtration for Surface Water Treatment

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**Abstract:** The effect of hydrophobic and hydrophilic fractions on membrane flux is investigated by DAX-8 resin for fractionation. The results show that hydrophobic fraction resulted in rapid decline in flux and hydrophilic fraction resulted in slow decline. It is found that addition of 25 and 100 mg $L^{-1}$ coagulant with 19.2 and 31.6% of TOC removal respectively, obvious enhancement of flux is observed. It can be concluded that the enhancement of flux is depend to a large extent on the removal efficiency of hydrophobicity by pretreatment. The more hydrophobic fraction is removed, the more flux is enhanced. The more hydrophobic fraction is rejected by UF membrane, the more flux is declined. The research also indicates that when Uftrafiltration (UF) membrane filtrates coagulate treated water, UF membrane tends to reject hydrophilicity.

**Key words:** Drinking water treatment, UF membrane, hydrophobicity, hydrophilicity, flux

## INTRODUCTION

Removal of NOM in potable water is a critical concern in both aesthetic aspect and removing precursors to disinfection by products. Recently the application of low-pressure membrane filtration such as Microfiltration (MF) and Ultrafiltration (UF) to produce drinking water has grown in prevalence due to its ability to remove a wide range of contaminants including particulates, colloids and pathogens. In addition, membrane application in surface water treatment offers the extra advantages over conventional treatment such as a small footprint, compact module, consumption, reduced chemical dosing and capability of handling wide fluctuations in feed quality (Jacangelo *et al*., 1997). However the major limitation of membrane filtration is fouling caused by the deposition of materials on or within the structure of the membrane, which results in increase in hydraulic resistances, operational and maintenance costs and deterioration of productivity and product quality (Fane *et al*., 2005).

Many factors effect membrane fouling by NOM, including the nature of the NOM (size, hydrophobicity, charge), the solution (pH, ionic strength, hardness ion concentration), the membrane (pore size, charge, hydrophobicity), module properties, mode of operation and applied flux (Taniguchi *et al*., 2003). In recent years, many researchers have studied a lot on the mechanism of membrane fouling caused by natural organics. The results showed that the characteristics of NOM like relative molecular weight and hydrophilicity greatly affect the membrane fouling. Carroll *et al*. (2000) and someone else separate the raw water into strong and weak hydrophobicities, polar and neutral hydrophilicities with resin and made filer tests respectively. Test results showed that the main organic fraction caused the decline of flux was neutral hydrophilicities. Fan *et al*. (2001) and others conducted the same tests and got the same conclusion with Carroll, T. (Fan *et al*., 2002). However, the results can not explain the fact that coagulation can effectively enhance flux (Chen *et al*., 2007) but with the poor effect of neutral hydrophilicities removal (Sinsabaugh *et al*., 1986). Lee *et al*. (2004) believes that the raw water contained more hydrophilic will cause serious membrane fouling. Some researchers got the opposite results to Carroll, T. and Fan, L.H. through tests. For example, Nilson and DiGiano (1996) tests showed that hydrophobic is the main cause to decline flux and hydrophilic affects flux little. Chen *et al*. (2007) and Dong *et al*. (2007) conclusion was the same as Nilson, J.A.'s, that is hydrophilic only slowly declines flux and the hydrophobic causes the sharp decline. Thus, there are still great differences between the effects of hydrophobic and hydrophilic on membrane fouling, which to be researched further.

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With respect to the behaviours of NOM in UF membrane filtration for surface water treatment is important. Thus, the objectives of this work were to (1) the components of NOM which foul a PES membrane were studied by fractionating the surface water NOM on non-functionalised resins into two specific classes of compounds based on hydrophobicity and hydrophilicity and measuring the rates of fouling by these compounds. (2) the contributions of NOM to the fouling of a Polyethersulfone (PES) UF membrane and the rate controlling fouling mechanisms, were investigated for both untreated and coagulation surface water. The NOM fouling mechanism was inferred from the types of compounds responsible for fouling.

## MATERIALS AND METHODS

**Raw water fractionation:** The surface water source was the Sanhaowu River. This water had a turbidity of 28.4 NTU, a Dissolved Organic Carbon (DOC) concentration of 10.96 mg $L^{-1}$, pH of 7.63, TDS of 561 mg $L^{-1}$ and UV254 of 0.212 cm-1. The most common techniques for isolation of NOM fractions are gel filtration and adsorption using non-ionic macroporous ion-exchange resins DAX-8 (Gray *et al*., 2004). The raw water concentrate was filtered though a 0.45-μm, adjusted to pH 2 and fed onto a Supelite DAX-8 nonfunctionalised resin which retains hydrophobicity matter attributed to humic and fulvic acids. The unabsorbed concentrate from the DAX-8 resin, were consists of hydrophilic (non-humic) organic matter attributed to proteins, amino acids.

**Membrane fouling:** Membrane Technique R and D Center Shanghai Institute of Nuclear Research Chinese Academy of Sciences provided UF membrane and cell for this study. We used a fresh sheet 10 kDa Polyethersulphone (PES) and effect filtration area was 3.32×10-3m2 for each experiment. Before filtration, the new membrane was soaked in the ultra-pure water (Milli-Q) for over 24 h. Dead end filtration was used in this test. Milli-Q water was first introduced into cell (350 mL), pressured at 100 kPa for 1-2 h until stable flux was reached and the 300 mL of NOM sample was introduced into cell for filtration. When permeated volume was reached 210mL, filtration was stopped and retentive and permeate sample was collected for analysis. Prior to each experiment run, the pure water of flux was measured. The ratio of the flux measured (J) to pure water flux (J0) was designated as J/J0 for comparison of the effect for each experiment run.

UF experiments were carried out on the two NOM fractions and on the unfractionated raw water, subjected to following pretreatments; coagulation with alum (Al2(SO4)3·18H2O). All UF experiments were done at pH 7.

**Analytical methods:** A UV spectrophotometer (Shimadzu UV-2201) and TOC analyzer (Shimadzu TOC-VCPH) were used to measure UV254 and DOC, respectively. All samples were filtered through a 0.45-μm filter. The turbidity was determined using a turbidimeter (Hach 2100N).

## RESULTS AND DISCUSSIONS

**Membrane fouling of the reconstituted raw NOM fractions:** The DOC and UV254 concentrations and relative percentages in the two raw NOM fractions are shown in Table 1. Hydrophilic fraction was 61.9% counted on TOC and 57.7% in terms of UV254.

The raw water is condensed by rotation evaporation and fed onto DAX-8 resin. The unabsorbed concentrate from the DAX-8 resin, which consists of hydrophilic was diluted by pore water with the same DOC with the raw water (DOC 11.45 mg $L^{-1}$).

The declines in permeate flowrates with permeate throughput during UF of the hydrophilic fraction and raw water are shown in Fig. 1.

At the beginning of filtration, the raw water flux was 0.83 (J/J0) significantly lower than the hydrophilic fraction 0.94 (J/J0) but in the late, hydrophilic fraction flux was lower than the raw water. This showed that the direct

Table 1: DOC and $UV_{254}$ concentrations of raw NOM fractions[a]

| Fraction | TOC (mg $L^{-1}$) | UV254($cm^{-1}$) |
|---|---|---|
| Hydrophilic | 6.933 | 0.155 |
| Hydrophobic | 4.157 | 0.044 |
| Unfractionated | 11.190 | 0.199 |

[a]The raw of fractions and unfraction adjusted to pH 7

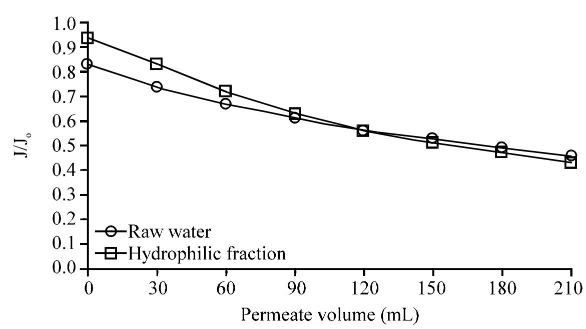


Fig. 1: Changes of flux in prefiltered raw water and hydrophilic faction

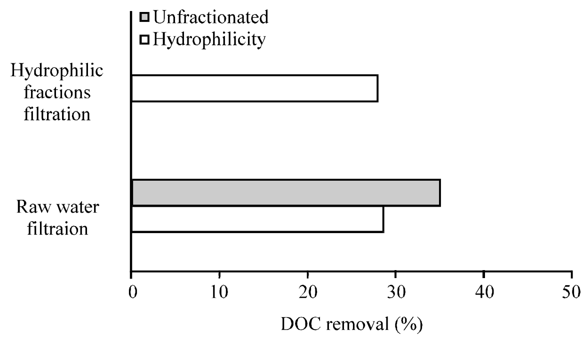


Fig. 2: Unfractioned and fractionated DOC removal by membrane filtration

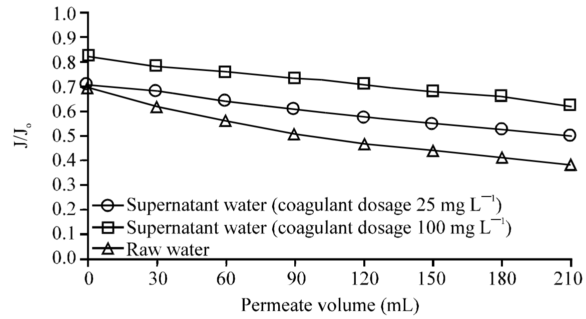


Fig. 3: Effect of coagulation pretreatment on flux

filtration of raw water would have even more serious membrane fouling and the effects of hydrophilic fractions on membrane fouling would show in the longer filtration. Fig. 2 shows the organics removal of raw water and hydrophilic fractions filtration. Thus, the membrane removed 35% of the DOC when the raw water is directly filtrated. It is noted that the hydrophilic fraction was only about 6%, only 17% of organic rejection, which meaned that the hydrophilic was the major factor of the decline of flux. Filtering hydrophilic fraction, the removal of TOC by the membrane is 28%, which showed that despite organics in water was the same, different compositions of it caused the different effects of rejection. Test shows that hydrophobic is easier to be rejected by membrane.

Aluminum sulfate is used as the coagulant. Respectively add 25 mg and 100 mg coagulants into 1L water samples, rapidly mix (100 r $min^{-1}$) 1 min, slowly churn up (30 r $min^{-1}$) 30 min and then after 30 min rest, the supernatant is filtered by 0.45 μm membrane. Filtrate is used for ultrafiltration membrane test and the changes of filter flux are shown in Fig. 3.

**Membrane fouling of raw water with coagulation pretreatment:** From Fig. 3, It can be seen the flux has been remarkably enhanced after adding coagulants. 25 mg $L^{-1}$ added, the initial flux filter is 0.85 (J/J0), at the end is 0.54 (J/J0); when dosage is 100 mg $L^{-1}$, the flux has been further enhanced, with the initial filter flux of 0.99 (J/J0) and the end of 0.69 (J/J0). This result shows that adding coagulants can effectively enhance membrane flux.

The effect of coagulant on removal of total organic carbon in raw water and hydrophilic fractions is shown in Fig. 4. Adding 25 mg $L^{-1}$, the organic removal rate was 19.2% and hydrophilic fractions removal is only 3.9%. Hydrophilic fraction is about 20% of the organics removed, which shows that coagulation mainly removed hydrophobic. When the dosage was added to 100 mg $L^{-1}$, organics removal also increased to 31.6% and hydrophilic fractions for the removal rate is 10.3%. Despite the hydrophilic fractions of the organic removal by the ratio has raised to about 30%, hydrophobic fraction was still the majority. Hydrophilic and hydrophobic play the important roles in it. The more hydrophobic fraction the pretreatment removes, the more obvious the flux enhances and vice versa.

Further make analysis of rejection of organics by membrane, as shown in Fig. 4. It can be seen, adding 25 mg $L^{-1}$ coagulant, the rejection of organics by membrane is almost all the hydrophilic fraction; adding 100 mg $L^{-1}$ coagulant, the rejection of organics by membrane was 27%, in which the hydrophilic fraction was 20 and 74% of rejection of organics by membrane, which showed that in coagulation as pretreatment, the rejection of organics by membrane was more hydrophilic. Thus, coagulation removes the most hydrophobic and most organic into the membrane is hydrophilic. As hydrophilic fraction affects less on flux, flux enhances significantly.

The results indicate that the rejection of hydrophilicity by membrane affects less on flux; opposite, the rejection of hydrophbicity by membrane determines membrane fouling. However, these results conflicts with the result of the hydrophilic fraction filtration in this study and get the different conclusions with Fan, L.H. and Carroll, T.. It should be noted the contributions of the NOM fractions with and without fractionated to fouling are different possibly, even are the opposite. Cause of this result may be that some parameters of the raw water changed after the fractionation. For example, TDS in hydrophilic fractions after resin separation (TDS: 1118mg $L^{-1}$) was double than the raw water, even though their TOC were same in this study.

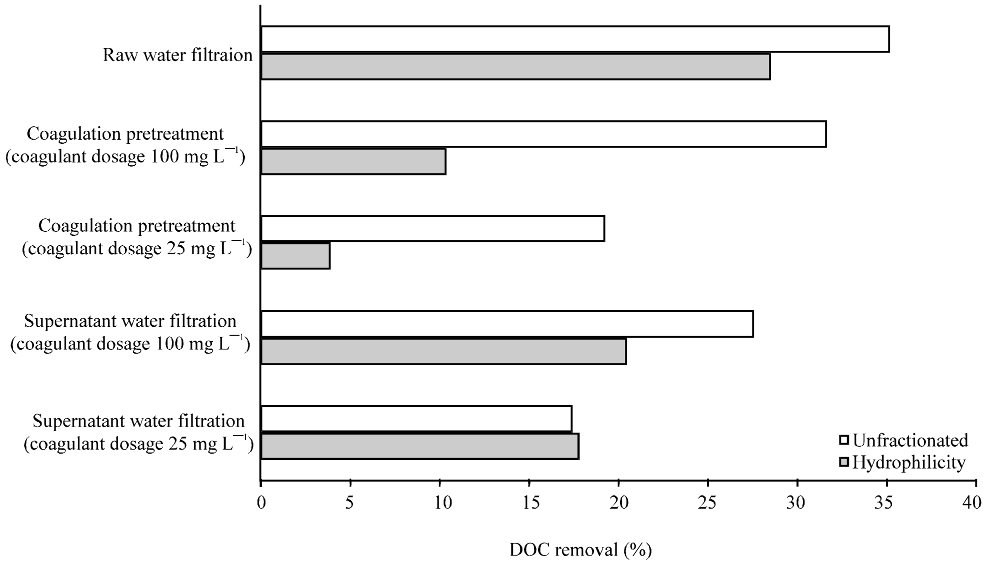


Fig. 4: Unfractionated and hydrophilicity DOC removal by coagulation and membrane filtration

## CONCLUSION

In this study, natural organic matter can play a rate-determining role in the fouling of dringking-water ultrafiltration membranes. The contribution of NOM to fouling depends on the raw water quality, characteristics of the NOM and type and level of any pretreatment. The following conclusions were obtained:

- For UF filtration without any pretreatment rejected less NOM, severe fouling also occurred. This may be contributed to the hydrophobicity deposited in the pores or on the surface of the membrane in UF of raw water. Hydrophobicity causes the rapid decline of ultrafiltration membrane flux
- When coagulation was used as a pretreatment for UF, coagulation pretreament mainly removes hydrophobicity and avoids membrane's contact with a large number of hydrophobicity thereby effectively enhances flux

## ACKNOWLEDGMENT

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