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Removal of natural organic matter by ultrafiltration with TiO_2-coated membrane under UV irradiation

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ABSTRACT

This study investigates the performance of ultrafiltration (UF) by membranes coated with titanium dioxide (TiO_2) photocatalyst under ultraviolet (UV) illumination in removing natural organic matter (NOM) and possibly in reducing membrane fouling. Experiments were carried out using heat-resistant ceramic disc UF membranes and humic acids as model substances representing naturally occurring organic matter. Membrane sizes of 1, 15, and 50 kDa were used to examine the effects of coating under ultraviolet irradiation. A commercial humic solution was subjected to UF fractionation (batch process); gel filtration chromatography was applied to study the effects of molecular weight distribution of NOM on UF membrane fouling. When compared to naked membranes, UV_{254} (ultraviolet light of $\lambda = 254$ nm) illumination of TiO_2-coated membranes exhibits more flux decline with similar effluent quality. Although the UF membrane is able to remove a significant amount of humic materials, the incorporated photocatalysis results in poor performance in terms of permeate flux. The TiO_2-coated membrane under UV_{254} irradiation alters the molecular weight (MW) distribution of humic materials, reducing them to <1 kDa, which is smaller than the smallest (1-kDa) membrane in this study. Thus, TiO_2-coated membranes under UV_{254} irradiation do not perform any better in removing natural organic matter and reducing membrane fouling.

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1. Introduction

One important issue in water treatment by ultrafiltration (UF) is membrane fouling due to natural organic matter (NOM). NOM, as often represented by dissolved organic matter, will adversely affect productivity, product quality, and process cost [1,2]. NOM can react with chlorine, the most commonly used disinfectant, to form disinfection by-products [3]. Thus it is of interest to consider UF in removing NOM in order to provide safe water to consumers.

Molecular weight distribution of NOM is known to impact membrane performance. Lin and co-workers [4] have shown that the fraction of large humic substances (apparent molecular weight 6.5–22.6 kDa) causes large flux decline but provides high-quality permeate, while small humic materials (160–650 Da) cause little flux decline.

Many approaches have been taken to minimize membrane fouling; these include pretreatment of feed water, hydrodynamic cleaning, optimization of water chemistry such as pH, and membrane surface modification. Studies of membrane surface modification have been undertaken by many researchers; however, most studies have focused on the characteristics of the membrane surface instead of on the application and performance.

Photocatalysis has received considerable attention for several reasons. Porous Teflon sheets coated with TiO_2 can be used to prevent snow from attaching to the surfaces of electric wires and to enable self-cleaning via photocatalytic degradation of organic pollutants [5]. Photocatalysis is shown to remove trichlorophenol and other recalcitrant pollutants and toxic organic substances [6–10]. The application of photocatalysts is based on their ability to promote chemical oxidation. When TiO_2 is illuminated with light of wavelength <400 nm, an electron is promoted from the valence band to the conduction band of the semiconducting oxide to give an electron/hole pair. The valence band potential is positive enough to generate hydroxyl radicals at the surface, and the conduction band potential is negative enough to reduce molecular oxygen. The hydroxyl radical is a powerful oxidizing agent to attack organic pollutants present at the surface of the TiO_2 particle.

Several methods have been developed to modify the membrane characteristics, including ion-assisted deposition [5] and direct filtration of nanoparticles in aqueous solution [10]. In the past few years, TiO_2 has also been successfully coupled with polysulfone and/or $ZnAl_2O_4$ membranes to enhance their water permeability, hydrophilicity, and anti-fouling ability [11–13]. TiO_2/polymer thin

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film composites on reverse osmosis membranes have been developed to mitigate biofouling by photobactericidal effects [14].

The purpose of the present work was to investigate flux performance and rejection on TiO_2-coated ceramic membranes to remove NOM. The specific tasks were mainly focused on (1) comparison of flux performance and fouling mitigation on UV_{254}-illuminated, TiO_2-coated membranes to naked membranes (TiO_2-UV_{254} indicates a membrane that is coated with TiO_2 particles and is under UV_{254} irradiation), (2) NOM removal in terms of dissolved organic carbon (DOC), and (3) the effect of photocatalysis batch processes on the molecular weight distribution of organic particles. Flux and rejection experiments were carried out with ceramic membranes, using humic acid solutions as model substances representing naturally occurring organic matter.

2. Materials and methods

2.1. Feed water

Humic acid (Aldrich) was used to prepare the feed water. The stock solution was prepared by dissolving 1 g of humic acid in 1 L of deionized water (MilliQ) followed by filtration through a 0.45-µm membrane filter (CA, Whatman). The filtrate was stored at 4 °C for subsequent use. Before each experiment, feed water was adjusted to pH 7 with addition of NaOH or H_2SO_4 solution.

2.2. UF membrane system

To observe flux decline within a reasonable time, a single ceramic flat sheet membrane was used. The characteristics of the ceramic membrane obtained from TAMI are shown in Table 1. The setup comprised a ceramic disc UF membrane module, an automatic fraction collector, a nitrogen cylinder, and feed and retentate tanks. Fig. 1 displays the setup of the experimental unit. The feed tanks are three cylinders, with each tank accommodating 3 L of humic acid solutions, totaling 9 L of solution. Air is sent into the feed tank to drive water into the membrane reactor. Transmembrane pressure (TMP) for all experiments was set constant at 10 ± 0.2 psi at room temperature (25 °C). No backwashing was used for these experiments.

The experimental procedure consisted of several cycles (5 or 6) of cross-flow filtration (0.05 m/s). The duration of each cycle was roughly 1 h 20 min. The feed solution passed through the ceramic membrane that was mounted on the membrane reactor and the permeate was collected at the bottom side of the membrane reactor by gravity. At constant pressure and velocity, only concentrate (retentate) was recycled to the feed batch, to simulate the actual UF plant operation. After 8 h of operation, each membrane was cleaned for subsequent use with NaOH (1 M) for 8 h, citric acid (1 M) for 1 h, NaOH (1 M) for 1 h, and ultrasonic cleaning for 10 min.

Before the start of the experiment, the membrane was compacted for 5 min while membrane permeability was measured. Pure water was fed to the membrane module with incremental increase and decrease steps of TMP for 5–6 min at 5, 7.5, 10, 12.5, 15, 15, 12.5, 10, 7.5, and 5 psi. Membrane permeability measurements are used to determine the range of UF and to compare the membranes. The measurement of flux began after 3 min when constant TMP of 10 psi was achieved. Membranes used were of 1, 15, and 50 kDa.

2.3. Permeability

All membranes were brand new from the manufacturer (TAMI). Permeability tests were carried out by incrementally increasing TMP as described above. The permeability value was drawn from the slope of each TMP. Fig. 2 is an example of determining the permeability for the membrane of 50 kDa. The permeability of the TiO_2-coated membrane is slightly lower than that of the naked membrane, as shown in Table 2. This result suggests a possibly smaller pore size as a result of TiO_2 coating on the membrane surface. Tighter molecular weight cut-off (MWCO) of membranes after coating outweighs the characteristics of TiO_2

Table 1
Characteristics of ceramic membrane

Support	
Manufacturer	Tami Industries
Material	Alumina, titania, zirconia (ATZ)
Disc diameter	90 mm
Effective surface area	56.3 cm^2
Operating maximum pressure	4 bars
pH operating range	0–14
Solvents	Insensitive
Operating temperature	<350 °C
MWCO	1, 15, and 50 kD
Disc holders	
Material	Stainless steel 316 L
pH operating range	0–14
Operating temperature	<130 °C

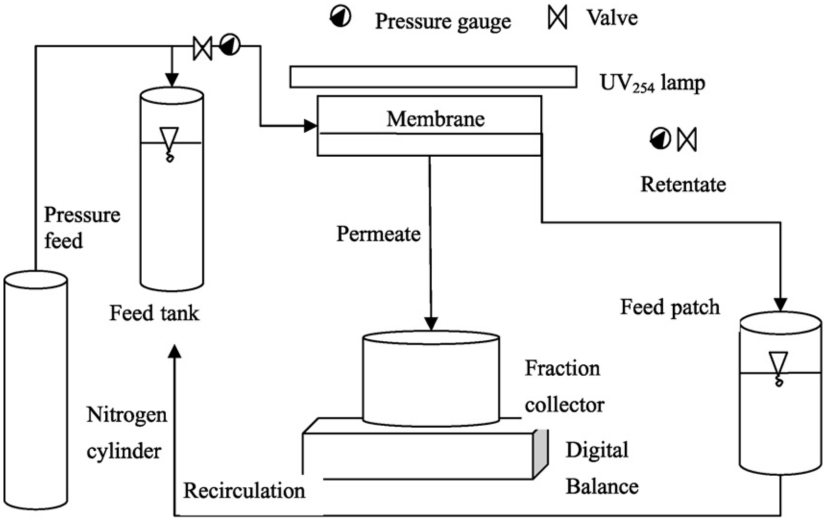


Fig. 1. Apparatus diagram of ceramic disc membrane employed in the filtration experiments.

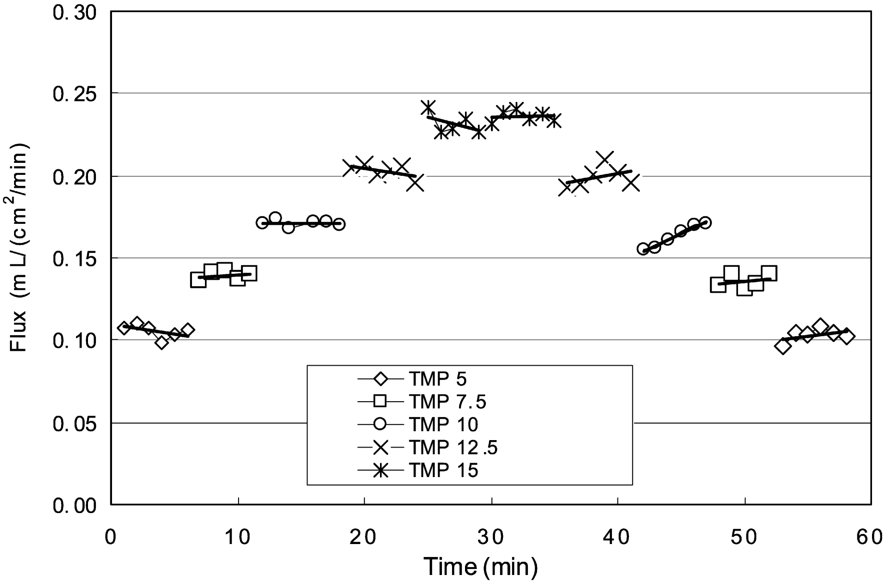


Fig. 2. Determination of permeation of 50-kDa naked membrane (TMP in psi).

Table 2
Permeability value of membrane

Description	Membrane MWCO (kDa)	Permeability (mL/(cm^2 min psi))
Without photocatalyst	50	0.0129
	15	0.0131
	1	0.0055
Combined with photocatalyst	50	0.01
	15	0.0104
	1	0.0052

coating that can increase the membrane surface affinity to water [13].

2.4. TiO_2 coating

The TiO_2 powder was P25 Degussa. An aqueous suspension containing 1.25% Degussa P25 (99.5% purity and about 80% anatase), 3.75% acetylacetone, and 5 drops of Triton-X was stirred for 1 h. While the coating solution was stirred, the membrane was prepared in such a position that the membrane disk was maintained horizontal to ensure equal distribution of solution within the membrane surface. After 1 h, the solution was then poured carefully on top of the membrane.

Temperature was initially set to 100 °C for 1 h and was gradually increased to 450 °C. The setting was done to ensure equal evaporation of acetylacetone to disperse the build up of TiO_2 on the membrane uniformly.

2.5. Molecular weight distribution

To determine the molecular weight distribution, UF fractionation and gel filtration chromatography (GFC) were employed. Prior to UF fractionation, photocatalytic processing was performed to obtain a humic solution. An aqueous suspension of TiO_2 was stirred by a magnetic stirrer in a water-jacked reactor vessel (20 °C) in which a UV lamp (Philips 254 nm, TUV 8W/G8 T5) in an inner periphery of the quartz glass tube was placed. A humic acid solution of 9 mg TOC/L was filtered by a 0.45-µm membrane filter (Whatman, cellulose acetate) and subjected to photocatalytic treatment. TiO_2 at 0.1 g (0.125%) was used as a photocatalyst and suspended by stirring it in 3 L of humic acid solution. The solution was aerated with a constant air flow rate. The pH was measured and adjusted to 7 by adding NaOH or H_2SO_4 solution. The UV_{254} illumination started after the addition of TiO_2. A sample was collected after 1 h, filtered with a 0.45-µm membrane to separate TiO_2 particles, and then measured for dissolved organic carbon by TOC analyzer. The processed humic acid solution was then fed to the membrane module for UF fractionation.

UF fractionation and GFC were conducted to determine the apparent molecular weight distribution (AMW) of the dissolved organic compounds. UF fractionation was carried out after TiO_2-UV_{254} batch processing. UF membranes used were of 1, 15, and 50 kDa. The method used was the same with the membrane system mentioned above. TMP used was 10 psi and was visually monitored. Humic acid concentration for each sample was determined by a TOC analyzer and UV absorbance measurements at wavelength 254 nm with a UV/vis spectrophotometer (Cintra).

Molecular weight distributions were also analyzed by GFC. The GFC system consisted of a C26/70 column (Pharmacia). The carrier solvent was made up of a 20 mM phosphate buffer (sodium phosphate, MW 141.96) and was adjusted to an ionic strength of 0.1 M with sodium chloride (MW 58.44) at pH 7. A flow rate of 2.0 mL/min was used. The gel bead matrix was Sephadex G-75, comprising 75% slurry under phosphate buffer solutions. The system was calibrated with PEG standards of molecular weights 1500, 6000, 15,000, 35,000, and 72,000 Da (Merck), prepared at 1 g/L concentration. The PEG standards and NOM were detected by UV absorbance at 254 nm and measured as DOC as well. A linear equation of the form $\log(\mathrm{MW}) = a - b(V_e)$ was obtained ($R = 0.98$), where MW is the molecular weight and V_e is the eluted volume.

2.6. Analysis

The DOC was analyzed for filtered samples (0.45 µm) in an organic carbon analyzer (TOC analyzer O. I. Analytical 1010). UV absorbance was measured with a UV/vis spectrophotometer (Cintra) at a single wavelength, 254 nm, with a 1-cm quartz cell; pH was determined with a Sension pH meter.

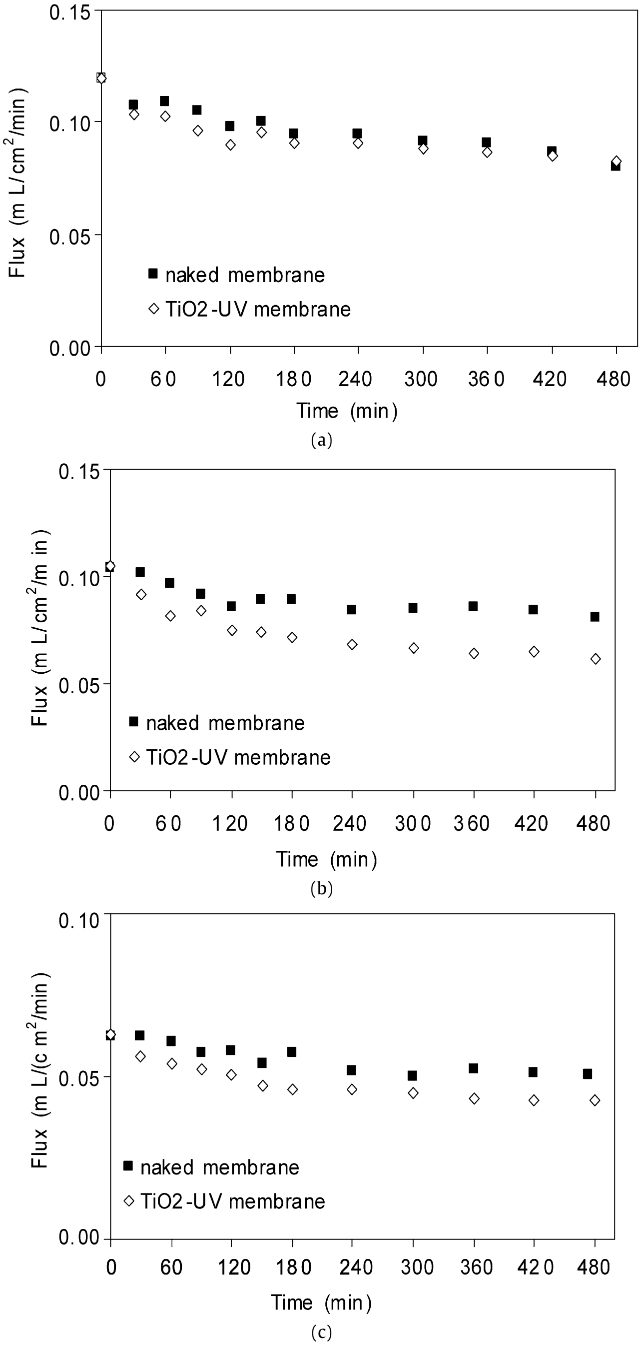


Fig. 3. Flux of (a) 50-kDa, (b) 15-kDa, and (c) 1-kDa membrane, naked membrane, and TiO_2-UV_{254} membrane.

3. Results and discussion

3.1. Flux performance

Similar initial fluxes were observed for TiO_2-coated membranes of 1, 15, and 50 kDa when compared with naked membrane, as shown in Fig. 3. TiO_2-coated membranes under illumination seem to show a more severe decline of flux. The coated membrane of 50 kDa under illumination declined in flux within the first 2 h until it reached a steady-state flux at 0.09 mL/(cm^2 min). On the other hand, the unmodified membrane of 50 kDa showed a steadily decreasing flux. The results were similar with other membranes.

The flux decline through an illuminated, coated membrane of 15 kDa took place within 6 h, while for 1 kDa it occurred within 3 h. These results suggested that more fouling occurred with TiO_2-coated membranes under UV_{254} irradiation than with naked membranes. This was estimated by measuring the permeability value for each membrane. Permeability values of TiO_2-coated membranes were lower than those of naked membranes. The presence of TiO_2 either produced tighter MWCO of the membrane or changed the physical and/or chemical properties of NOM. Thus more humic materials were retained on the membrane surface, resulting in less water passing through and the flux declining. A lower steady-state flux and a higher loss of flux for coated membranes were also evident in Fig. 4 and Table 3.

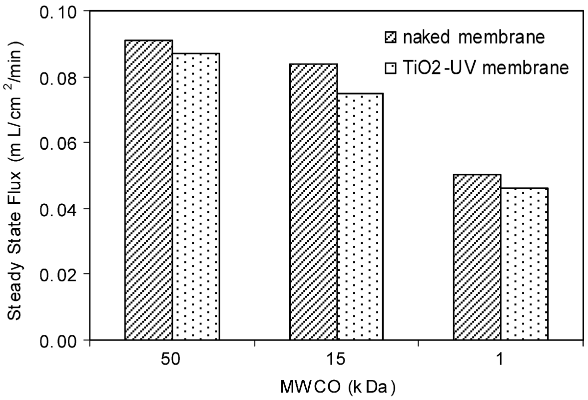


Fig. 4. Steady state flux for naked and TiO_2-UV_{254} membrane.

Table 3
Initial and steady-state flux of naked membrane and TiO_2-UV_{254} membrane

Description	Membrane MWCO (kDa)	Initial flux (mL/(cm^2 min))	Steady state flux (mL/(cm^2 min))	Percentage flux loss (%)
Naked membrane	50	0.01198	0.0909	24.13
	15	0.01044	0.0838	19.76
	1	0.0623	0.0503	19.32
TiO_2-UV_{254} membrane	50	0.01199	0.0868	27.60
	15	0.0943	0.0748	20.68
	1	0.0631	0.0462	26.73

Higher flux decline during operation of illuminated, coated membrane was probably due to clogging of pore openings on the membrane surface. While large molecules are rejected by a size exclusion mechanism, smaller molecules are attached to the TiO_2 particles and adsorbed within the membrane pores. The results of this study are not in agreement with those of a previous study using a different feed solution [15]. Different membrane materials may have been a factor. The materials can affect the overall hydrodynamic condition, especially in the chemical interaction between foulants and the membrane.

3.2. Removal characteristics

Dissolved organic carbon removal by both modified and unmodified membranes is shown in Fig. 5. DOC removal by an unmodified membrane of 50 kDa ranged from 51 to 71%, while removal by illuminated, coated membrane ranged from 57 to 73%. Removal by an illuminated, coated membrane was slightly higher than that by a naked membrane. However, this result was not reproduced for the illuminated, coated membrane of 15 kDa, which achieved 70–79% removal of DOC compared to naked membrane at 79–86% removal.

On average, the DOC removal performance (data not shown) for naked membranes was slightly better than that for illuminated coated membranes. It is possible that illuminated coated membranes exhibit less DOC removal due to the alteration of the

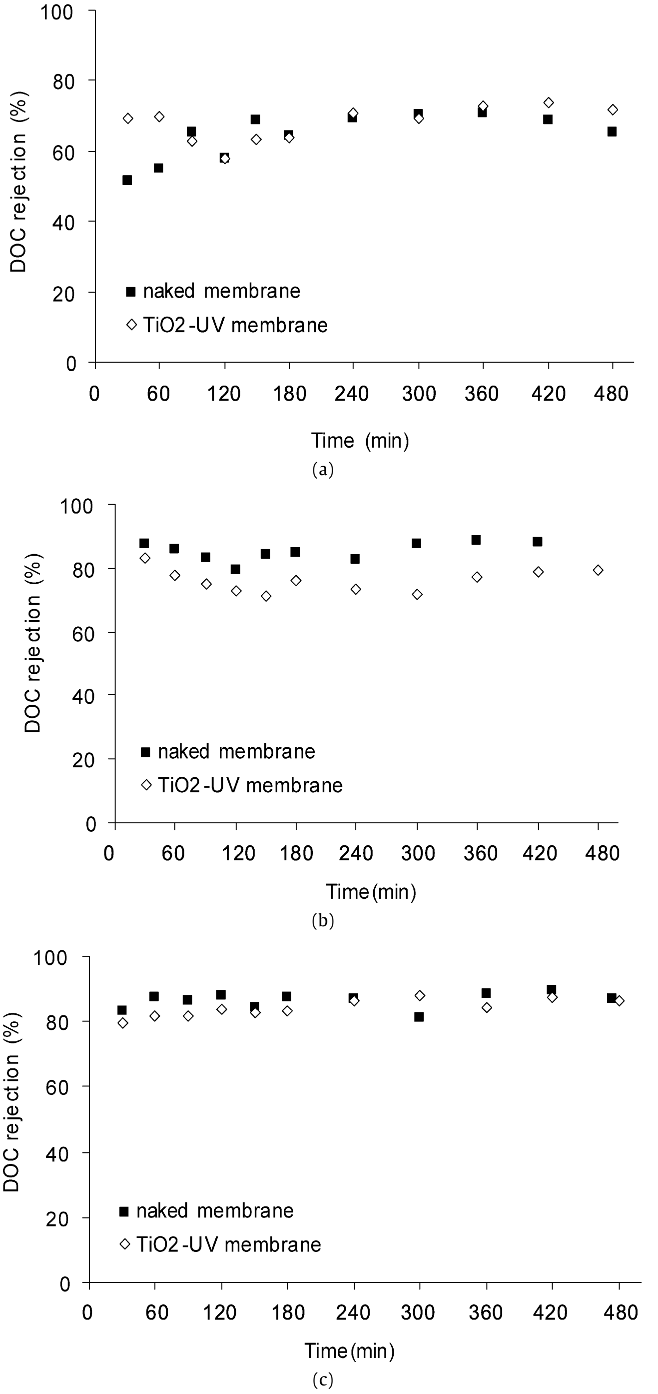


Fig. 5. DOC removal of (a) 50-kDa, (b) 15-kDa, and (c) 1-kDa naked and TiO_2-UV_{254} membranes.

molecular weight (MW) distribution of humic acid. The organic fraction in 1–50 kDa was reduced to below 15 kDa through bond breaking by OH radicals. The OH radicals were produced by photocatalyst TiO_2 under UV_{254} irradiation. However, Figs. 5b and 5c provide another perspective. After 6 h of operation, DOC rejection appeared to increase. This was because of the smaller membrane MWCO, which was an effect of organic materials on both TiO_2 particles and membrane pores. DOC removal by a naked membrane of 1 kDa was 81–87% and that by an illuminated coated membrane was 79–87%, which shows a result similar to that with a 50-kDa membrane.

From results of flux and DOC removal, it appears that the alteration of MW distribution in the feed humic acid solution occurs

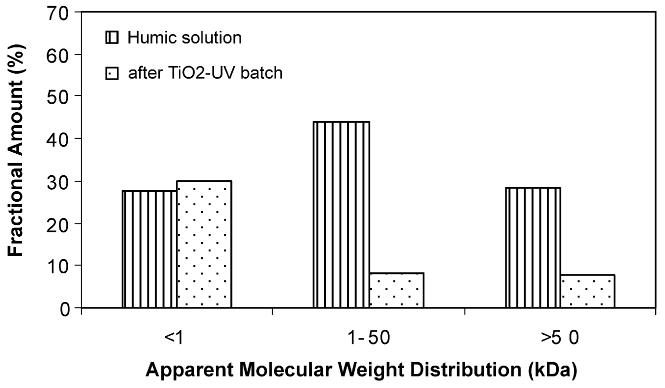


Fig. 6. Apparent molecular weight distribution after 1 h TiO_2-UV batch relative to initial concentration 9 mg/L based on DOC.

for organics in the range 1–50 kDa. The transformation of molecular size occurred toward below 1-kDa and above 50 kDa ranges. Our results are similar with the work of Bae et al. [15], where rejections by membrane with TiO_2 entrapped on the surface were similar to original membrane.

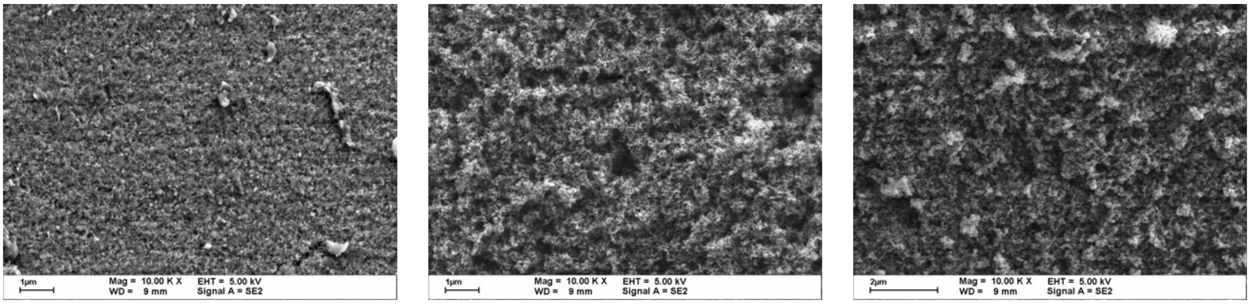
These results might have resulted from the intrinsic nature of TiO_2 particles, which acted to increase the interdiffusion velocity of the water solvent and because of the particles' hydrophilic nature made the membrane more porous. Even though the presence of TiO_2 on the membrane surface made the surface more hydrophilic and increased the affinity between water and the membrane, fouling still occurred rapidly. This was due to the presence of TiO_2 particles attached to the membrane surface and possibly on the pore surface inside the membrane, thus resulting in lower permeability.

Lowered permeability is expected to yield increased rejection; however, this work shows the opposite. It seems that the MW distribution change occurs during operation. Although photodegradation occurred on illuminated, coated membranes, it did not help overall performance. Photodegradation of humic materials is evident by the difference in fractional amount (in term of DOC) between the raw feed water (humic solution) and the UF-filtered water after the illuminated, coated membrane batch process (Fig. 6). The organics within the humic solution were photodegraded by as much as 45%.

On one hand, the pore size that becomes smaller will produce more rejection; on the other, possible transformations of organics from higher MW to lower will allow increased passage through the TiO_2-coated membrane. The MW transformation, which adversely impacts performance, outweighs the benefit of the illuminated, coated membrane, which includes photodegradation and adsorption for organic removal.

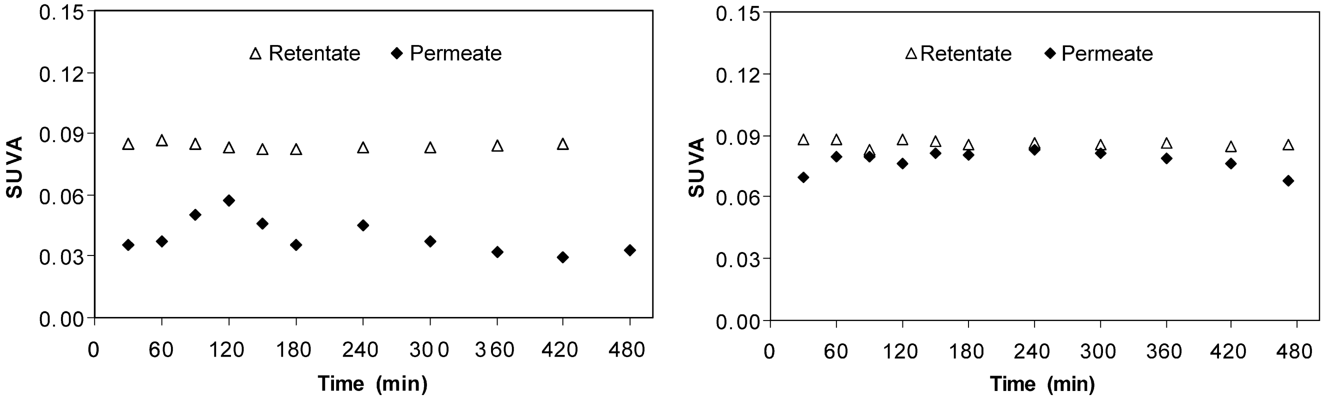
Under the scanning electron microscope (SEM), a neat flat surface was observed for the naked membrane with MWCO of 15 kDa (Fig. 7a). Coating of TiO_2 created coarse and deep valleys on the membrane surface (Fig. 7b). A smoother surface was also observed for a fouled coated membrane of 15 kDa after 8 h of operation. This shows that the removal of organics on the TiO_2 surface was by adsorption, while photocatalytic degradation contributed some removal.

When humic materials were in contact with TiO_2-UV_{254} membrane, the fraction of unsaturated C–C bonds of NOM was altered. The specific ultraviolet absorbance (SUVA) represented by the ratio between UV_{254} and DOC can be indicative of complexity resulting from the presence of aromaticity and other unsaturated chemical bonds. Interestingly, more conversion of complex aromatics and unsaturated chemical bonds was observed with unmodified membranes (Figs. 8 and 9) of 15 and 1 kDa. The SUVA in the retentate ranged from 0.08 to 0.09 L/(mg cm) (15 and 1 kDa), and the per-



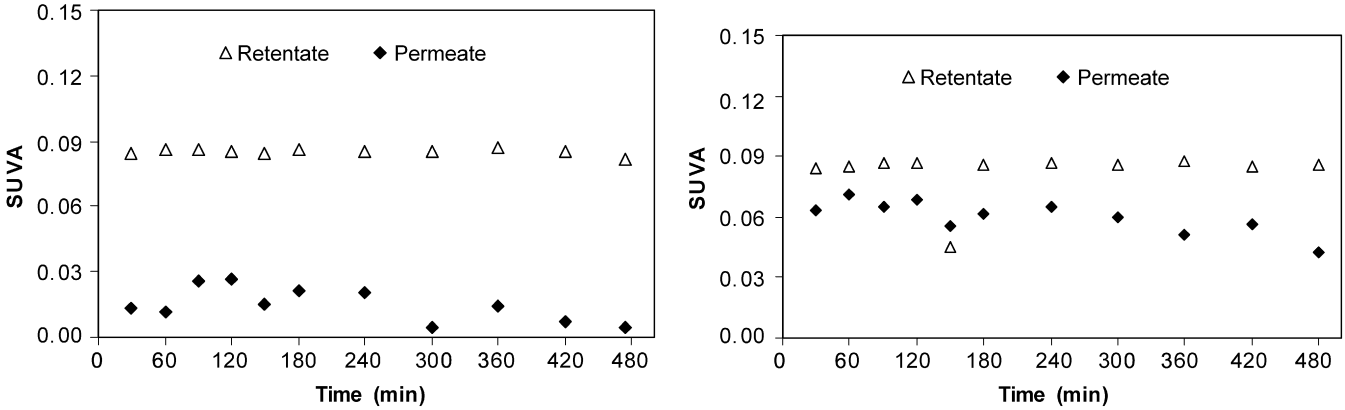
(a) (b) (c)

Fig. 7. Scanning electron microscopy of 15-kDa (a) naked membrane, (b) TiO_2-coated membrane, and (c) fouled TiO_2-UV membrane.



(a) (b)

Fig. 8. SUVA for 15-kDa membranes: (a) naked membrane and (b) TiO_2-UV_{254} membrane.



(a) (b)

Fig. 9. SUVA for 1-kDa membranes: (a) naked membrane and (b) TiO_2-UV_{254} membrane.

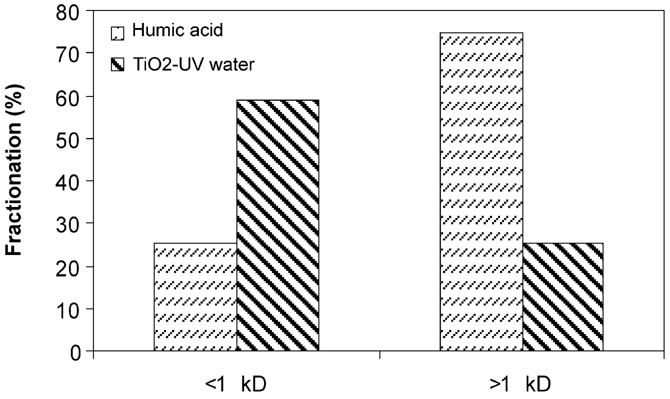
meate SUVA observed was found to be in the range from 0.005 to 0.06 L/(mg cm). In contrast, lower conversion was observed after treatment by illuminated, coated membranes of 15 and 1 kDa (Figs. 8 and 9). The retentate SUVA was similar to those from naked membrane, but with a slight difference in permeate ranging from 0.04 to 0.07 L/(mg cm). It appeared that oxidation occurred during photocatalysis, which inhibited further conversion to simpler forms of humic groups.

3.3. Molecular weight distribution

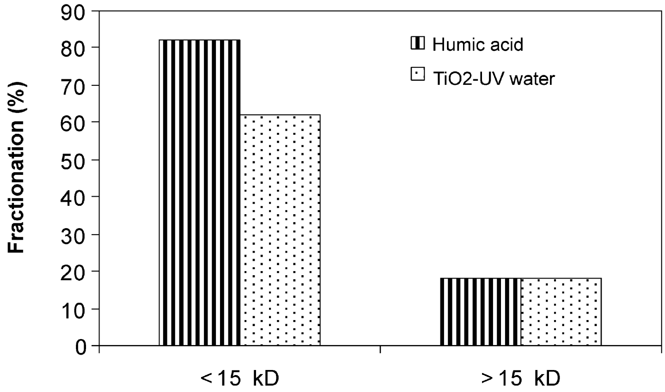
Fig. 10 shows molecular weight distribution. It should be noted that each graph was under slightly different initial DOC concentrations. Therefore these graphs only depict the distribution of MW after reaction with TiO_2 under irradiation with UV_{254}. Initial DOC concentrations of UF fractionation for 1, 15, and 50 kDa membranes were 8.7, 9.0, and 9.4 mg/L, respectively. The operation time was 1 h to easily identify MW changes as subjected to the photocatalytic process.

The MW of humic materials below 1 kDa in raw water was around 25.2% and above 1 kDa was 74.8%. After 1 h of the illuminated, coated membrane batch process, removal of DOC was as much as 15.8%. The concentration of the sample was 7.4 mg/L. This evidence showed that removal was due to photodegradation of humic materials. The MW distribution after photocatalysis was 58.9% below 1 kDa and 25.3% above 1 kDa, as shown in Table 4.

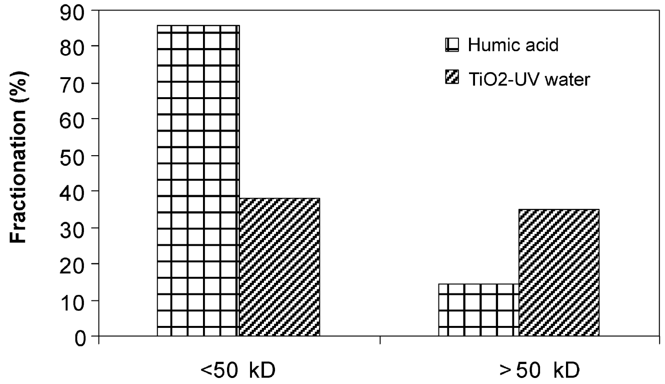
Initial DOC of UF fractionation with 15-kDa membrane was 9 mg/L, and after removal by 1 h of the photocatalytic process it was 19.8 mg/L. Before photocatalysis, approximately 82% of organic matter was below 15 kDa, leaving 18% of organic matter



(a) Apparent Molecular Weight Distribution



(b) Apparent Molecular Weight Distribution



(c) Apparent Molecular Weight Distribution

Fig. 10. Apparent molecular weight distribution obtained by UF fractionation after 1 h photocatalysis batch: (a) 1-kDa, (b) 15-kDa, and (c) 50-kDa membranes.

above 15 kDa. The photocatalysis process alters the MW distribution. The organic matter with MW below 15 kDa was 62.1%, and above 15 kDa was 18.1%, as noted in Table 4.

The initial DOC for UF fractionation of 50 kDa was 9.4 mg/L, composed of 85.6% below 50 kDa and 14.4% above 50 kDa. The removal by the photocatalytic process was 26.7%. However, there was a change in MW distribution after the photocatalysis process. MW became 38.1% below 50 kDa and became 35.2% above 50 kDa. Fig. 10 also shows that most organic MW above 15 kDa, either 15–50 kDa or above 50 kDa, will be converted during photocatalysis to below 15 kDa, and a portion to above 50 kDa. The result is supported by Fig. 11, which shows that the MW distribution moves to below 1 kDa. However, since the initial concentration of each experiment was different, no exact comparison could be made other than an overview.

Table 4
Molecular weight (MW) distribution of humic solution (raw water) and TiO_2-UV photocatalysis batch

MW distribution/water (kDa)	Raw water (%)	TiO_2-UV water (%)
<1	25.17	58.9
>1	74.83	25.29
<15	82.09	62.10
>15	17.91	18.13
<50	85.64	38.08
>50	14.36	35.19

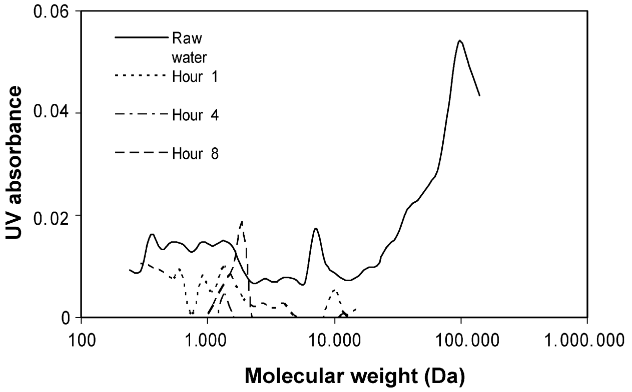


Fig. 11. Molecular weight distribution by GFC.

Molecular weight distribution of UF fractionation is confirmed by GFC. Fig. 11 confirms that the molecular weight of the permeate decreases, but a significant peak around 2 kDa was observed, with the highest peak at 8 h of membrane operation. These results show that photocatalysis alters the MW distribution of organic matter in the water, especially for larger molecules.

More membrane fouling resulted from the illuminated, coated membrane of 15 kDa. Since different membranes affect the fractionation process, it is difficult to accurately isolate the MW distribution. Therefore, it is important to use a better method to determine MW distribution during illuminated, coated membrane operation. GFC has provided excellent additional information for molecular weight distribution of the humic materials, as shown by Fig. 11.

4. Summary

The results of the present study show that the use of an UV_{254}-illuminated TiO_2-coated membrane system in UF did not enhance flux and the removal of humic substances in comparison to unmodified membrane.

On average, UF of TiO_2-UV_{254} systems of 1, 15, and 50 kDa did not improve flux or alleviate fouling, as indicated by the decline of flux being more rapid compared to that on naked membranes. Humic substances removal was found to be better with naked membranes than with TiO_2-UV_{254} UF membrane systems. However, increased humic removal was observed on TiO_2-UV_{254} membranes suggesting increased rejection due to MWCO of membranes becoming smaller with the presence of TiO_2, which increased adsorption of humic substances. Humic removal by photodegradation was also observed.

Illuminated TiO_2 on membranes alters the molecular weight distribution of humic materials. After operation, a fraction of humic substances was converted to below 1 kDa and a small portion to above 50 kDa, as evident from results of UF fractionation and GFC.

Acknowledgments

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