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# Irreversible Fouling in MF/UF Membranes Caused by Natural Organic Matters (NOMs) Isolated from Different Origins

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# Irreversible Fouling in MF/UF Membranes Caused by Natural Organic Matters (NOMs) Isolated from Different Origins

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**Abstract:** For more efficient use of membrane technology in water treatment, it is essential to understand more about the fouling that requires chemical cleaning to be eliminated (i.e., irreversible fouling). In this study, five different MF/UF membranes and four types of organic matter collected from different origins were examined in terms of the degree of irreversible membrane fouling. Experimental results demonstrated that the extent of irreversible fouling differed significantly depending on the properties of both the membrane and organic matter. Among the tested membranes, UF membranes made of polyacrylonitrile (PAN) exhibited the best performance in terms of prevention of irreversible fouling. In contrast, MF membranes, especially one made of polyvinylidenefluoride (PVDF), suffered significant irreversible fouling. Conventional methods for characterization of organic matter such as specific ultraviolet absorption (SUVA), XAD fractionation, and excitation-emission matrix (EEM) were found to be inadequate for prediction of the degree of irreversible fouling. This is because these analytical methods represent an average property of bulk organic matter, while the fouling was actually caused by some specific fractions. It was revealed that hydrophilic fraction of the organic matter was responsible for the irreversible fouling regardless of the type of membranes or organic matter.

**Keywords:** Membrane filtration, irreversible membrane fouling, membrane material, natural organic matter, hydrophilic organic matter

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# INTRODUCTION

Membrane technology is becoming a viable option for water/wastewater treatment. There have been significant increases in installed capacities of microfiltration (MF) and ultrafiltration (UF) for drinking water treatment since the 1990s (1). The main limitation to wider use of membranes in drinking water treatment is the high energy consumption of membranes mainly due to membrane fouling that develops over a long period of operation. Operating costs strongly influence the overall cost-effectiveness of membrane systems (2). Many efforts have been made to control membrane fouling. Several physical membrane cleaning methods such as backwashing, high cross-flow velocity or scouring with air bubbles have been developed and have been used routinely in many existing low-pressure membrane plants to minimize membrane fouling. The resistances in series model is often employed to describe filtration resistance and its build-up in a membrane filtration process:

$$J = \Delta P/\mu R_t = \Delta P/\mu(R_m + R_r + R_{irr}) \tag{1}$$

where $J$ is the permeate water flux, $\Delta P$ is the transmembrane pressure difference, $\mu$ is the water viscosity, $R_t$ is the total filtration resistance, $R_m$ is the intrinsic membrane filtration resistance, $R_r$ is the reversible resistance, and $R_{irr}$ is the irreversible filtration resistance. Over a long period of operation, the membrane filtration resistance gradually increases and the resistance is not completely eliminated by the routine physical cleaning methods mentioned above. Such filtration resistance, denoted in this paper by $R_{irr}$, can only be eliminated by the use of chemical reagents. $R_{irr}$ should be minimized for more efficient use of membranes as it directly increases operation costs. Chemical cleaning of a membrane can reduce or eliminate $R_{irr}$. However, chemical cleaning should be limited to a minimum frequency because repeated chemical cleaning may affect membrane life, and disposal of used chemical reagents is also a problem. Thus, control of $R_{irr}$ is important. For this purpose, information on constituents that cause $R_{irr}$ is indispensable. Natural organic matter (NOM), composed of humic substances as well as non-humic materials, contained in natural water has been demonstrated in a number of studies to be the foulant that causes membrane fouling (3–12). NOM represents a wide range of macro-molecular organic compounds. So far, it is still unclear which fraction of NOM causes irreversible membrane fouling.

In this study, one commercially available humic acid and three types of organic matters isolated from different aquatic environments were filtered with five different MF/UF membranes and the degrees of irreversible fouling were evaluated. The objectives of this study were to

i. determine whether the degree of irreversible membrane fouling differs depending on the properties of both organic matter and membranes,

ii. to explore relationships between the degree of fouling and the properties of organic matter/membranes, and
iii. to investigate which constituents in bulk organic matter cause irreversible fouling.

## MATERIALS AND METHODS

### Organic Matter

One commercially available humic acid and three organic isolates obtained from different aquatic environments were used in this study. The commercially available humic acid was obtained from Aldrich (Milwaukee, USA) and is designated hereafter as AHA. Two drinking water sources and one tertiary wastewater treatment effluent were chosen for isolation of organic matter. Chitose River (DOC = 2.1 mg/L) and Toyohira River (DOC = 1.3 mg/L) are currently being used as drinking water sources in the cities of Chitose and Sapporo, Japan, respectively. The tertiary treatment effluent was collected at the Soseigawa Wastewater Treatment Plant where conventional activated sludge process and sand filtration are carried out. DOC of the effluent was 4.0 mg/L. The organic matters isolated from Chitose River, Toyohira River and the wastewater treatment plant effluent are denoted hereafter as CTR, TYR, and WWTP, respectively. Eight hundred L of the water collected from each source was processed with reverse osmosis (RO) for isolation of organic matter by the method of Sun et al. (13) immediately after sample collection. Pretreatment with microfilters (10 μm, 2 μm and 0.45 μm) was carried out to remove suspended matter, and a cation exchange softener was used to remove calcium and magnesium. Then the organic matter was concentrated using reverse osmosis (Nanomax 95, Millipore). The recoveries of organic matter, defined here as (mass of DOC after concentration by RO)/(mass of DOC before concentration), were 0.95, 0.81, and 0.91 for CTR, WWTP, and TYR, respectively. Following concentration by RO, electric dialysis was applied for desalination and then the samples were lyophilized and preserved until use.

### Membranes

Four different hollow-fiber membranes were used in this study. Two of them were UF membranes and were made of polyacrylonitrile (PAN). They were made of the same type of polymer but had different nominal molecular weight cutoffs (MWCOs): 100,000 and 80,000. The other two were MF membranes that had the same nominal pore size, 0.1 μm. These two MF membranes were made of different organic polymers. One was made of polyethylene (PE) and the other was made of polyvinylidenefluoride (PVDF). Tiny membrane modules of 40 cm$^2$ in membrane area were assembled with the four types of membrane strings and were used in filtration experiments. A flat-sheet type membrane made of polyether sulphone (PES), which has an MWCO of 100,000, was also examined. Table 1 summarizes the zeta potential of the tested membranes.

## Filtration Protocol

Filtration was carried out by water level difference in the experiments using the hollow-fiber membranes. The four membranes were submerged in one filtration basin and fed organic matter at the same time. Membrane filtration was carried out by a water level difference of 3 m. Variation of water level in the filtration basin was maintained within 2 cm by pumping the same amount of feed water as the amount filtered. Water temperature of the filtration basin was maintained at 20°C during the experiment. Feed water was prepared daily by dissolving organic matter to a concentration of 3 mg-C/L in Milli-Q water. $Na_2CO_3$ was also added to the solution at a concentration of 1 mM, resulting in approximately neutral pH of the solution. Filtration was continued for 23 hours and the change in permeate flux was monitored by electric balances. Following the 23 hours of filtration, the tiny modules were taken out from the basin and were physically cleaned. The physical cleaning performed in this study was backwashing (50 kPa, 3 minutes) using Milli-Q water followed by gentle wiping with a sponge. Following the physical cleaning, the tiny modules were placed back in the filtration basin and the filtration was re-started. Depending on the available amount of each organic matter, the 23-hour filtration was repeated 4–10 times. At the time of each physical cleaning, the feed water remaining in the filtration basin was discarded in order to avoid excessive accumulation of rejected organic matter in the system. At the end of the filtration experiment, the tiny modules were disassembled and the foulants were desorbed from the membranes by sodium hydroxide. Consequently, new membranes were used in every experimental run. The flat-sheet type membrane made of PES was examined in the manner of cross-flow filtration. A membrane cell (C70-F, Nitto-Denko, Osaka, Japan) was used for the cross-flow filtration. Effective membrane area in the cell was 32 cm$^2$. The cross-flow

***Table 1.*** Zeta potential of the tested membranes[a]

| PAN (MWCO: 100,000) | PAN (MWCO: 80,000) | PE | PVDF | PES |
|---|---|---|---|---|
| −7 mV | −4 mV | −7 mV | −2 mV | −10 mV |

[a] Measurements were carried out by using a zeta potential meter (ELS-8000, Otsuka Electronics, Osaka, Japan) at pH 7.0 and 5 mM KCl.

filtration using the PES membrane was carried out in a manner basically the same as that in the case of the hollow-fiber membranes. That is, the applied pressure was fixed at 30 kPa, corresponding to the water level difference of 3 m, which was used in the experiments with hollow-fiber membranes. Also, physical cleaning (backwashing and wiping with a sponge) was carried out after each 23-hour filtration.

### Analytical Methods

Concentrations of total organic carbon (TOC) and dissolved organic carbon (DOC) were determined by a TOC analyzer (TOC-5000, Shimadzu, Kyoto, Japan). UV absorbance was measured by a spectrophotometer (U-2000, Hitachi, Tokyo, Japan). Fractionation of organic matter contained in the isolates was achieved by using the hydrophobic/hydrophilic fractionation method described by Aiken et al. (14) to obtain three different fractions (i.e., hydrophobic (HPO), hydrophilic (HPI), and transphilic (TPI) fractions). Amberlite resins, DAX-8 (Supelco, Bellefonte, USA) and XAD-4 (Supelco, Bellefonte, USA), were used for the fractionation. All resins were purified by Soxhlet extraction prior to being used in the process. To obtain information on organic matter from different aspects, fluorescence excitation-emission matrices (EEMs) of the samples were generated by using a fluorescence spectrophotometer equipped with a 150 W ozone-free xenon lamp (RF-5300PC, Shimadzu, Kyoto, Japan). In the measurement of fluorescence, wavelength of both emission and excitation was varied stepwise by 5 nm. Prior to the measurements of UV and fluorescence, pH of the samples was adjusted to ~7.

## RESULTS AND DISCUSSION

### Characteristics of the Organic Matter

NOM is composed of a variety of organic molecules with high molecular weights. Its characteristics reflect its origin and geographic environment and consequently will differ greatly depending on where the NOM is obtained (15). The degree of membrane fouling would differ depending on the characteristics of organic matter that is filtered. Thus, characteristics of the four organic matters used in this study were investigated based on specific ultraviolet absorbance (SUVA), hydrophobic/hydrophilic fractionation, and fluorescent excitation-emission matrix (EEM).

Table 2 summarizes SUVA values determined for the organic matters used. SUVA is considered as a surrogate measurement for DOC aromaticity (16). It can be seen from Table 2 that AHA showed a much higher SUVA value than the values of the other three organic matters. This demonstrates

***Table 2.*** $SUVA_{254}$ values of the tested organic matter

| | AHA | CTR | WWTP | TYR |
|---|---|---|---|---|
| $SUVA_{254}$ ($L\,mg^{-1}\,m^{-1}$) | 8.6 | 2.9 | 3.7 | 2.9 |

that the commercially available humic acid does not necessarily represent the features of real organic matter found in aquatic environments. Organic matters isolated from the two surface waters (i.e., CTR and TYR) showed comparable SUVA values which were slightly lower than the value for WWTP. Table 3 presents results of hydrophobic/hydrophilic fractionation of the organic matter used. The fractionation method used definitively divides organic matter into three fractions: hydrophobic (HPO), transphilic (TPI), and hydrophilic (HPI). Based on the definition given by Thurman and Malcolm (17), the HPO fraction is referred to humic substances. The HPI fraction was the most abundant in WWTP, while HPO content was highest in TYR. As stated above, TYR and CTR were found to be similar in terms of results of SUVA analysis (Table 2). Nonetheless, a considerable difference was found in results of XAD analysis, reflecting the fact that SUVA only shows an "average" property.

EEMs generated for the organic matter are presented in Fig. 1. Locations of peaks in an EEM reflect characteristics of organic matter. Large peaks can be seen near 430 nm/320 nm (emission/excitation) in EEMs for AHA and WWTP. On the other hand, four different peaks can be seen in the EEMs for CTR and TYR. The shapes of the peaks in the EEMs generated for CTR and TYR were similar. The EEM analysis revealed that features of fluorescence-sensitive organic matters contained in the surface waters (TYR and CTR) and the wastewater effluent (WWTP) were considerably different. Based on the EEM analysis, features of the commercial humic acid were found to be similar to those of organic matter in wastewater effluent rather than those of organic matters in the surface waters used. As described above, features of the organic matters used in this study were considerably different, and the degrees of membrane fouling caused by them were therefore expected to differ even when the loading the same.

***Table 3.*** XAD fractionation of the tested organic matter

| | Hydrophobic (%) | Transphilic (%) | Hydrophilic (%) |
|---|---|---|---|
| AHA | 45 | 21 | 34 |
| CTR | 40 | 26 | 34 |
| WWTP | 32 | 23 | 45 |
| TYR | 50 | 25 | 25 |

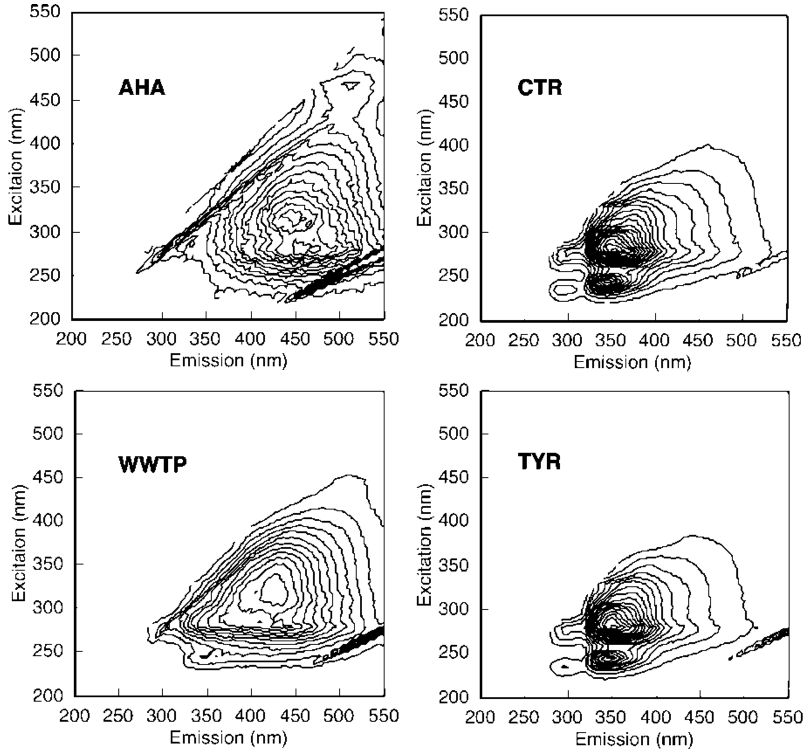

*Figure 1.* Fluorescence EEM of the examined organic matters.

## Membrane Filtration Driven by a Constant Pressure Difference

When membrane filtration is carried out under a constant pressure difference, permeate flux gradually decreases over time. By applying physical cleaning (e.g., backwash), permeate flux can be increased but membrane permeability can never be restored to the initial state due to deposition of foulant on/in the membrane that cannot be removed by physical cleaning. Figure 2 shows representative data obtained in filtration of CTR by the PVDF membrane under constant pressure difference. In Figure 2, permeate flux was normalized to the flux observed at the initiation of the experiment. All data regarding permeate flux shown in this paper were treated in this manner. As was expected, permeate flux decreased with elapse of time and was partially restored by physical cleaning. Membrane fouling can be divided into two categories: reversible fouling, which can be eliminated by physical cleaning, and irreversible fouling, which requires a chemical reagent(s) to be eliminated. In Fig. 2, evolution of irreversible fouling can be evaluated by connecting points recorded just after the physical cleaning (dashed line). This study focused on irreversible membrane fouling as defined above.

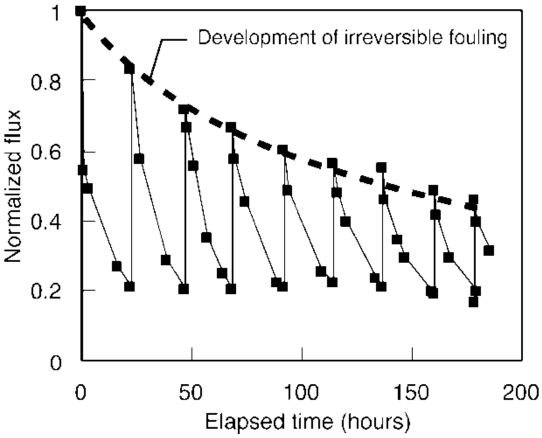

*Figure 2.* Change in normalized flux in the filtration of CTR with a PVDF membrane.

## Irreversible Membrane Fouling caused by Isolated Organic Matter

Figure 3 shows the changes in permeate fluxes observed during the filtration experiments with the four hollow-fiber membranes. In Fig. 3, only data recorded immediately after the physical cleaning were plotted in order to focus on the evolution of irreversible fouling. The experiments using CTR and TYR were carried out for 9 days and 4 days, respectively, while experiments using AHA and WWTP were carried out for 10 days. The durations of the experiments using CTR and TYR were shorter because available amounts of isolated organic matters were limited. It is clearly demonstrated in Fig. 3 that the degree of the irreversible fouling was different depending on the combination of membrane and organic matter examined.

As mentioned before, AHA showed a much higher SUVA value than the values of the other three isolates. However, with all of the tested hollow-fiber membranes, AHA caused the least irreversible fouling. In experiments using the UF (PAN) membranes, evolution of the irreversible fouling was not apparent with any of the tested organic isolates. In contrast, in experiments using the MF membranes (PVDF and PE), evolution of irreversible fouling was significant with all of the tested organic isolates. The degree of irreversible fouling followed the order: WWTP > TYR > CTR > AHA. Although the two MF membranes used in this study had the same nominal pore size (0.1 $\mu$m), the rate of the fouling was faster in the experiments using the PVDF membrane. This demonstrated that the material of the membrane had an influence on the degree of irreversible fouling. The most significant irreversible fouling was found in the case of a combination of a PVDF membrane and WWTP.

As mentioned above, irreversible fouling was not significant in the experiments using UF membranes. In general, UF membranes produce smaller

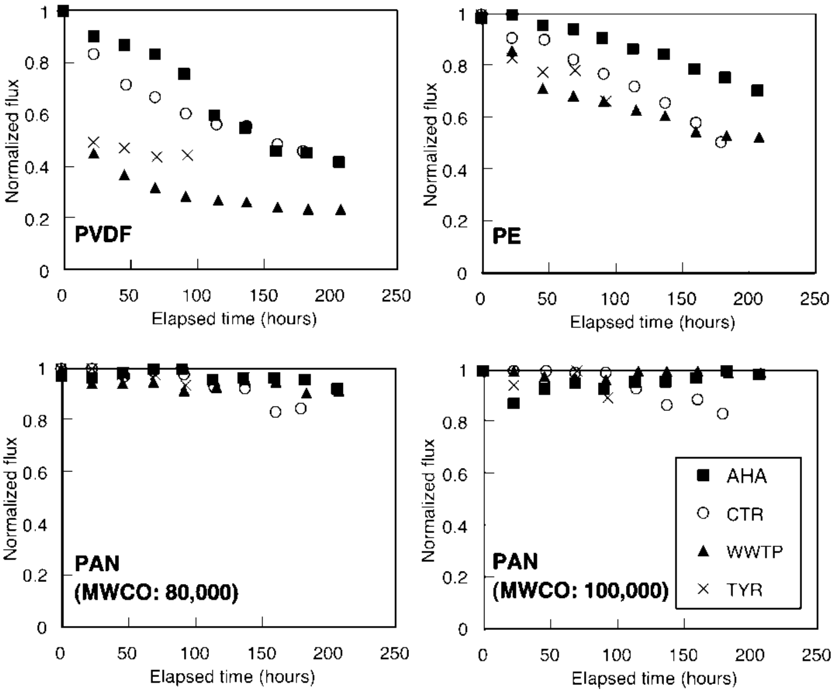

*Figure 3.* Evolution of irreversible fouling during filtration of 3 mg/L of each of the organic matters.

amounts of permeate than MF membranes when operated under the same pressure difference. If this were the case in this study, a smaller amount of organic matter would have been delivered to the surface of the UF membrane compared with that in the case of the MF membrane and this might have been a reason why the UF membrane did not show significant irreversible fouling. However, this explanation was discounted as there was not a considerable difference between the accumulative permeate volumes obtained during the experiments using UF and MF membranes. For instance, in the case of filtration of WWTP, the accumulative permeate volumes obtained by using PVDF (MF) and PAN (UF) membranes were approximately 60 L and 50 L, respectively. This reflects the fact that a high degree of membrane permeability of the UF membrane was maintained even in the late stage of the experiments. Consequently, it was more likely that PAN was a material that did not exhibit significant irreversible membrane fouling with the tested organic matter or that the UF membranes did not allow permeation of particles that could pass through the large pores of the MF membranes and cause irreversible fouling. In order to examine these hypotheses, a UF membrane with a comparable MWCO (100,000) made of a different material (i.e., PES) was tested. As a hollow-fiber type of membrane was not available in our laboratory at that time, a flat-sheet type of membrane was examined in the cross-flow mode of operation. Physical cleaning was carried out under the same conditions and at the same frequency as those in the experiments using the hollow-fibers. That is, after each 23-hour filtration, backwashing was conducted at 50 kPa for 3 minutes and the surface of the membrane was gently wiped with a sponge. Fig. 4 shows the change in permeate flux observed in the experiments using the flat- sheet PES membrane. TYR and CTR were not examined because sufficient amounts of the isolates were not left. As shown in Figure 4, irreversible fouling was significant in the experiment using the UF membrane made of PES. Consequently, the fact that the PAN membranes did not exhibit significant irreversible fouling was attributed not to the sieving effect but to the features of the material (PAN). At present, however, it is not known which features of PAN are relevant for the prevention of irreversible fouling. It would be interesting to identify the features of PAN that are responsible for the prevention of fouling and to determine the range in which PAN can be used without suffering severe irreversible fouling, as observed in this study. This would lead to a development of a new membrane material that would reduce the extent of irreversible fouling.

## Relationship between Irreversible Fouling and Properties of the Organic Matter

There was no significant difference between CTR and TYR in terms of results of SUVA or EEM analyses (Table 2 and Fig. 1). However, the rates of evolution of irreversible fouling in the experiments using these organic isolates were different, especially when MF membranes were used. The degrees of

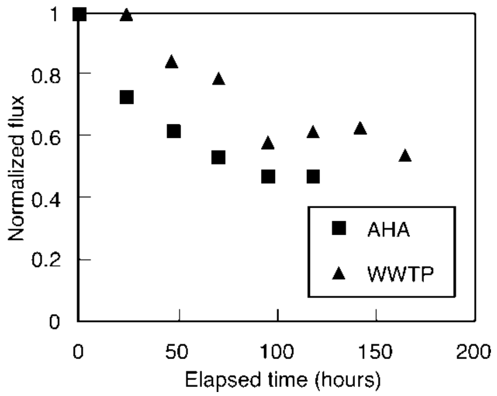

*Figure 4.* Changes in normalized fluxes in the filtration of AHA and WWTP with a PES membrane (MWCO = 100,000 Da).

irreversible fouling in the MF membranes caused by WWTP and TYR were comparable though no similarity was found between the two isolates on the basis of results of SUVA and EEM analyses (Table 2 and Fig. 1). These results clearly demonstrated that the degree of irreversible fouling could not be predicted on the basis of results of SUVA or EEM analysis of bulk organic matter. Fractionation using XAD resins is also not likely to be adequate for predicting the degree of fouling. This can be seen from a comparison of WWTP and TYR in terms of XAD fractionation (Table 3). Although the two isolates showed similar degree of irreversible fouling in the experiments using the MF membranes, the compositions of them based on XAD fractionation were quite different: the HPI fraction was abundant in WWTP, while hydrophobic fraction was greatest in TYR. These findings give the overall impression that analyses of feed organic matter based on SUVA, EEM, and XAD fractionation are not adequate for predicting the trend in evolution of irreversible membrane fouling. This reflects the fact that analyses based on SUVA, EEM, and XAD fractionation represent a heterogeneous mixture of molecular structures in averaged manners. In those analytical methods representing the integrated features of organic matter, the presence of specific fractions is buried in bulk of the organic matter. Irreversible fouling is likely to be caused by specific fractions of organic matter.

## Constituents Responsible for Irreversible Membrane Fouling

It has been stated that the degree of irreversible membrane fouling differs depending on characteristics of both the membrane and organic matter. Constituents responsible for irreversible membrane fouling will be discussed in this section. To identify the constituents responsible for membrane fouling, organic matter that had caused irreversible fouling during the filtration experiments was desorbed and analyzed. Analysis was carried out for only the two MF membranes that suffered from significant irreversible fouling.

Desorption of organic matter from the membrane was carried out by soaking the membrane in alkaline solution (sodium hydroxide). At first, desorption was attempted at pH of 10 so as to prevent undesirable membrane decomposition. This condition, however, turned out to be so insufficient for desorption of the foulant that the subsequent analysis did not give us informative results (data not shown). Therefore, membrane specimens soaked in the solution of pH 10 were subsequently transferred to a stronger alkaline solution (pH 12) and left in the alkaline solution for 24 hours. Prior to alkaline extraction, physical cleaning of the membrane was carried out in the same manner as that done in the filtration experiment (i.e., backwashing and wiping with a sponge).

Table 4 summarizes the results of desorption tests. Data for AHA are not shown in Table 4 because a desorption test was not carried out with it.

***Table 4.*** Properties of the organic matter desorbed from the fouled membranes

| | DOC (mg/L) | | $UV_{254}$ absorbance (1/cm) | | SUVA (1/m/mg/L) | |
|---|---|---|---|---|---|---|
| Samples | PVDF | PE | PVDF | PE | PVDF | PE |
| CTR | 2.9 | 0.9 | 0.021 | 0.005 | 1.19 | 0.83 |
| WWTP | 8.1 | 1.1 | 0.028 | 0.007 | 0.37 | 0.58 |
| TYR | 2.3 | 1.1 | 0.017 | 0.005 | 1.60 | 0.20 |
| Unused membrane | 1.4 | 0.6 | 0.003 | 0.004 | | |

In the calculation of SUVA for the desorbed components, DOC and UV absorbance measured for an unused membrane (bottom row in Table 4) were subtracted from those measured for the used membranes so as to eliminate the influence of the organic matter originating from the membranes themselves. Comparison of SUVA of the membrane foulants and that of the feed organic matter (Table 2) showed that SUVA of the former was clearly lower than that of the latter. This demonstrates that a relatively hydrophilic fraction of each organic matter, which is likely to exhibit a lower SUVA, was responsible for evolution of the irreversible fouling. Fractions that have features of protein and/or polysaccharides are contained in NOM and exhibit hydrophilic characteristics (e.g., low SUVA). The authors previously conducted a long-term, pilot-scale ultrafiltration at an existing water purification plant and reported that polysaccharides-like organic matter was responsible for the irreversible membrane fouling (18), which is in accordance with the results of this study in which different membranes and feed organics were examined.

It should be noted that the SUVA values determined for the foulants from the PVDF and PE membranes were considerably different despite the fact that the same organic matter was filtered for the same duration. This implies that composition of the desorbed organic matter (i.e., irreversible membrane foulant) was different depending on the membrane material even when the same organic matter was fed. The difference in SUVA values of the desorbed organic matter was most pronounced when TYR was examined.

## SUMMARY

In this study, the influence of properties of membranes and organic matters on the evolution of irreversible membrane fouling was investigated using five different MF/UF membranes and four different types of organic matter. Experimental results indicated that the degree of irreversible fouling was dependent on the properties of both the membrane and organic matter. Among the tested membranes, the UF membranes made of PAN exhibited the best performance in terms of prevention of irreversible fouling. This was attributed to unidentified features of the membrane material, PAN. Determination of the reason why PAN membranes do not suffer irreversible fouling should lead to the development of a new anti-fouling membrane material. The two MF membranes suffered significant irreversible fouling. With the organic matters used in this study, fouling of the PVDF membrane was greater than that of the PE membrane. Conventional methods for characterization of organic matter such as SUVA, XAD fractionation, and fluorescence excitation-emission matrix (EEM) were found to be inadequate for prediction of the degree of irreversible fouling. This is because these analytical methods represent an average property of bulk organic matter, while the fouling was actually caused by some specific fractions. It was revealed that relatively hydrophilic fractions of the organic matter were responsible for the irreversible fouling regardless of the type of organic matter or membrane. Composition of organic matter causing irreversible fouling is likely to differ depending on the membrane material. Further analysis of desorbed organic matter should lead to a more fundamental understanding of irreversible fouling.

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