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Relationship between feed water quality and membrane performance during the filtration of real secondary effluent

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ABSTRACT

Water quality of feed stream certainly has a close relationship with membrane fouling during the microfiltration of secondary effluent. The current study tested the ceramic membrane performance using secondary effluents from two wastewater treatment plants and a pilot scale A^2/O treatment equipment. Reversible and irreversible fouling were discussed using different water quality parameters. The water quality was characterized using general parameters, as well as particle size distribution (PSD) and fluorescence spectrum of different organic compositions. Higher particle concentration in feed water leads to a higher reversible fouling. Wider PSD results in lower porosity of the cake layer and higher cake layer resistance. Hydraulic pressure above 50 kPa results in the deformation of suspended particles and a more compact cake layer; thus, more dissolved or colloidal materials are retained. Based upon the humic substance content in secondary effluent from different wastewater treatment plants, higher humic substance concentration might contribute greatly to the irreversible fouling in the ceramic microfiltration of secondary effluent.

Keywords: Secondary effluent; Microfiltration; Particle matter; Organic matter; Membrane fouling

1. Introduction

As a highly efficient water treatment technology, membrane filtration can be used to remove microorganisms, suspended particles, and organic matters from water and wastewater. Membrane technology has several advantages, including constant-efficiency, compact size, easy automatic operation, disinfectant saving, and so on [1,2]. Reclamation of secondary effluent using the membrane process is promising because of the scarcity of fresh water in large cities. However, membrane fouling continues to bring down the economic process. As a mixture of dissolved organic matters and suspended particles, secondary effluent has a severe fouling potential toward the low pressure filtration membranes, i.e. microfiltration (MF) and ultrafiltration [3–5].

Many previous publications have suggested that organic matters in secondary effluent (EfOM) plays a significant role during polymeric membrane fouling in the tertiary treatment of wastewater [5–9]. EfOM represents many kinds of organic matters, such as polysaccharide, proteins, aminosugar, nucleic acid, humic and fulvic acids, organic acids, and cell components, among others [6,10], whereas the

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different components of EfOM have different membrane fouling potentials. Some studies reported that the fraction of humic substances from natural organic matters is a major foulant that controls the rate and extent of fouling during ultrafiltration of surface water [11–13], whereas other studies stressed the relevance of soluble microbial products [14], proteins [15], or polysaccharides [16–19] in secondary effluent using membrane fouling. However, the hydrophilic or hydrophobic property of organic matters might be more important to the adhesion ability of the membrane material.

For dead-end low pressure filtration, filtration performance also depends on the content of feed solids, which would cause serious concentration polarization and cake layer fouling [20–22]. Generally, the concentration polarization and cake layer formed using suspended solids are relatively easier to remove physically. Therefore, reversible membrane fouling is mainly contributed by suspended solids. However, taking the particle size into account, different membrane fouling forms might occur. Considering a situation like that, membrane pore size distribution and particle size distribution (PSD) overlap, high transmembrane pressure (TMP) could press parts of small suspended solids into the membrane pores to result in irreversible fouling. However, no previous study has paid attention to such transition from reversible fouling to irreversible fouling.

To date, most related research was conducted using polymeric membrane [20], single organic solution as feed water, or synthetic wastewater [23]. Ceramic membranes have many advantages over polymeric membranes because they are thermally and chemically stable, have longer lifetimes, and are oxidant resistant [24]. Thus, the fouling characteristics of ceramic membrane and polymeric membrane are different from each other. For example, compared with reversible resistance, irreversible resistance plays a major role in the total resistance of ceramic membrane against polymeric membrane [23].

Generally, the feed water quality during the microfiltration process of secondary effluent greatly affects ceramic membrane fouling. Few studies reported on the function of both particulate and organic matters content during the microfiltration of secondary effluent, especially in terms of PSD. Therefore, a study on the effect of water quality on membrane fouling is important.

As stated above, many studies on the reclamation of secondary effluent with membrane have been carried out, but there have been few studies focusing on the effects of feed water quality, especially PSD, on ceramic low pressure membrane. In order to elucidate the effect of feed water quality on the fouling performance of ceramic membrane filtration process, a pilot scale, ceramic membrane filtration equipment was setup for the treatment tests of secondary effluents from different sources in the current study.

2. Materials and methods

2.1. Source water

The accurate water quality of secondary effluent in real wastewater treatment plants (WWTPs) changes daily; however, a certain secondary effluent still has its own features and fluctuation range. Therefore, different sources of feed water were selected to distinguish the differences. In the current study, secondary effluent from WWTP B employing a conventional active sludge process, G employing A/O processes, and a pilot A^2/O system (P) was used as the feed water. Their average water quality is provided and discussed in Section 3.1.

2.2. Setup and operations

The system, which is initially a combined process, includes a coagulation tank, an ozonation unit, and a microfiltration unit. In the current study, the units of coagulation and ozonation were stopped to examine the direct effects of feed water quality on membrane performance. Fig. 1 shows the flow chart of the process employed. The system consists of a feed pump, a flow meter, a membrane module, a backwashing system, and a data acquisition system. The multi-channel alumina ceramic membrane element was supplied by NGK. The configuration parameters of the ceramic membrane are shown in Table 1, and the permeability given by applied hydraulic pressure and corresponding membrane flux is illustrated in Fig. 2.

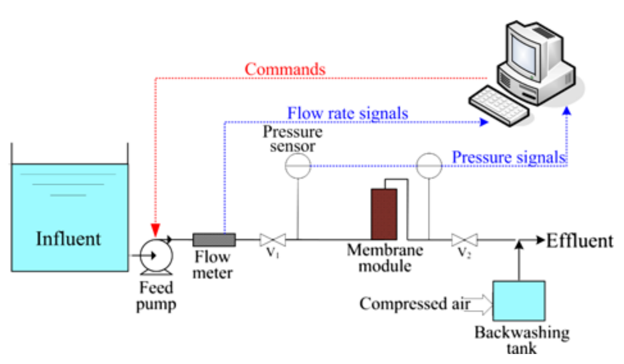

Fig. 1. Schematic diagram of the ceramic membrane microfiltration process.

Table 1
Configuration parameters of the alumina ceramic membrane element

Membrane pore size	0.1 µm
Length	1 m
Channel number	61
Inner diameter of channels	2.5 mm
Membrane filtration area	0.48 m^2

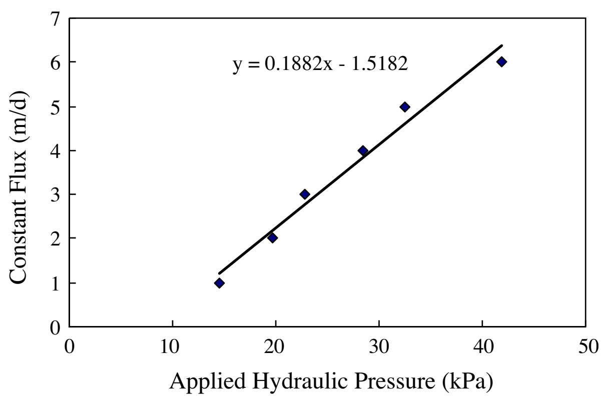

Fig. 2. Applied hydraulic pressure (TMP) and the corresponding flux of the ceramic membrane employed in this study.

The system was operated under constant-flux mode. The feed was first pressured using a centrifugal pump into the channels of the ceramic membrane, which was housed in a stainless steel module. The feed pump was adjusted according to the feedback of the flow rate sensor, and maintained at a constant flux of approximately 1 L/min, corresponding to a filtration flux of 3 m/d. The filtration of the monolithic ceramic membrane was operated under dead-end mode. The pressure values at both upstream and downstream of the ceramic membrane were monitored using sensors, and then recorded on a personal computer. TMP was then calculated. In the current study, the water temperature compensation for TMP was considered based on the following equation (adopted from the ceramic membrane provider NGK Company):

$$\Delta P_{25^\circ C} = \frac{\Delta P_{measure}}{\mu} \times 0.0008935 \quad (1)$$

where μ is the water viscosity (kg/s/m) at real temperature.

The pilot system was run in two steps: filtration and backwashing. In the filtration step, feed water was introduced into the vertically set monolithic ceramic membrane module in an upward flow from the bottom side. Hydraulic backwashing with a pressure of 500 kPa was performed after the filtration phase was completed. The filtrate was used for backwashing. Each time, 2.5 L of water was used for backwashing. An air blow process was then performed to flush the membrane foulant loosened from the hydraulic backwashing. Both sides of the membrane were rinsed, and the next filtration phase was initiated. To be specific, the filtration time of each cycle lasted 90 min, and the calculated recovery rates were all 0.968.

Five tests using different effluent sources were conducted. Test #1 used 2nd effluent from B, tests #2 and #3 used 2nd effluent from G, tests #4 and #5 used 2nd effluent from P. To eliminate occasional inaccuracy, two similar tests were conducted using one feed source, except WWTP B because of some uncontrollable reason. For all tests, only the feed source was different; other conditions, such as the flux, were kept constant.

2.3. Analytical methods

Water quality was characterized by its total organic carbon (TOC) concentration, UV absorbance at 254 nm, PSD, and fluorescent excitation-emission spectrum (FEEM) measured using Shimadzu TOC-5000A, Hach DR5000, Ankersmid EyeTech, and Hitachi F2500, respectively. Dissolved organic matters were fractioned using macro-reticular resins (XAD-8, Amberlite, 20–60 meshes), and the fraction method was performed according to the methods described by Wang et al. [25]. Analyses of chemical oxygen demand (COD), ammonia nitrogen, total nitrogen, total phosphorus, and total coliform bacteria were performed according to the standard methods proposed by SEPA of China.

2.4. Theory and assumption

Membrane fouling was discussed based on reversible and irreversible fouling, the definitions of which are illustrated in Fig. 3. Irreversible fouling is more or less affected by backwashing parameters, such as pressure, time interval (filtration phase lasting time), and backwashing duration, which were fixed to 500 kPa, 90 min filtration, and 3 min backwashing, respectively, in the current study. Some specific definitions or calculations used in the current study are summarized as follows (fouling extent is expressed in terms of TMP):

Table 2
Average values of water quality parameters after ozonation in each test

Feed Water Sources	B WWTP	G WWTP		P Pilot	
Test No.	#1	#2	#3	#4	#5
pH	7.54	7.50	7.63	7.80	7.70
Suspended solids (mg/L)	28.0	5.2	5.4	5.5	4.8
Turbidity (NTU)	3.6	1.3	1.4	1.4	1.2
COD_{Cr} (mg/L)	41.88	36.38	33.49	34.50	38.47
TOC (mg/L)	15.76	11.25	7.72	6.22	7.23
UV_{254} (1/cm)	0.1480	0.1341	0.1165	0.1333	0.1405
Ammonia (mg/L)	19.43	0.88	1.18	1.112	2.23
Total nitrogen (mg/L)	33.17	17.21	18.34	19.92	19.64
Total phosphorus (mg/L)	1.55	0.76	0.74	0. 57	0.64

Irreversible fouling gained in the ith cycle = Irreversible fouling at the beginning of the $(i+1)$th cycle − Irreversible fouling at the beginning of the ith cycle;

Reversible fouling gained in the ith cycle = Total fouling at the end of the ith cycle − Irreversible fouling at the beginning of the $(i+1)$th cycle.

Real-time TMP data were acquired and recorded in a computer. One-minute-average TMP data were used for the analysis to eliminate the micro-fluctuations introduced by electronic noise. The first data of the filtration phase were not considered to avoid the cover parts of backwashing, just as other researchers did [26].

3. Results and discussion

3.1. The feed water characteristics

The average values of water quality in different tests are given in Table 2. As can be seen in the table, the secondary effluents from G and P were close to each other, and were at a good level. The effluent from B contained more pollutants than G and P, both on particulate concentration (in terms of suspended solids and turbidity) and organic matter concentration (in terms of COD, TOC, UV, and so on) because WWTP B was long-term overloaded. WWTP B was left behind when #1 was finished because its capacity has expanded since then. The feed from G was similar to that of the P pilot equipment on most parameters of water quality, except for the lower ammonia nitrogen concentration.

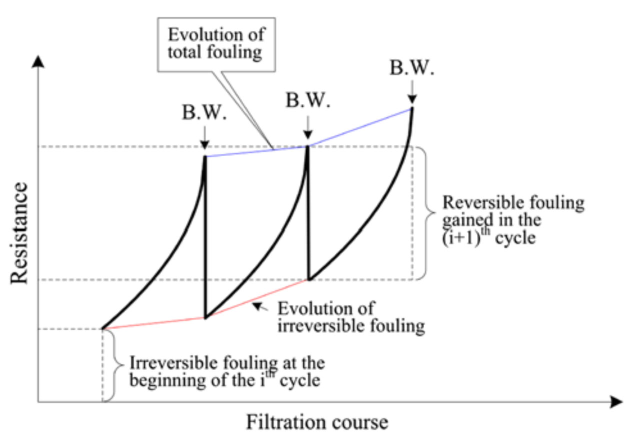

Fig. 3. Definitions of reversible and irreversible fouling (B. W. – backwashing).

3.2. Effect of particle concentration on reversible fouling

Fig. 3 shows how the TMP developed from the beginning of tests #1 to #5. The scales of the abscissas and ordinates in Fig. 4 are the same for clear comparison. Based on the definition given in Fig. 3, the TMP profiles from irreversible fouling in Fig. 4 were easy to draw, i.e. the bottom borderlines of dense-dotted areas. Moreover, the longitudinal length of the dense-dotted area at any time (section) represents the reversible fouling at that moment. Most of the time, the irreversible fouling profiles in all five tests gradually increased at different rates, which is consistent with the definition of irreversible fouling. The increasing rates relate with colloidal and dissolved materials, and will be discussed in Section 3.5. Unlike irreversible fouling, reversible fouling profiles have extinct fluctuation, especially in Fig. 4(A)–(C). The fluctuation in the suspended solids concentration in feed water was one of the reasons for the fluctuation. Presuming the suspended solids amount was the sole factor affecting reversible fouling extent, the fluctuation in suspended solids concentration would obviously result in the fluctuation of reversible fouling. Based on the data in Table 2, feed water quality from B was different with that from the other two sources.

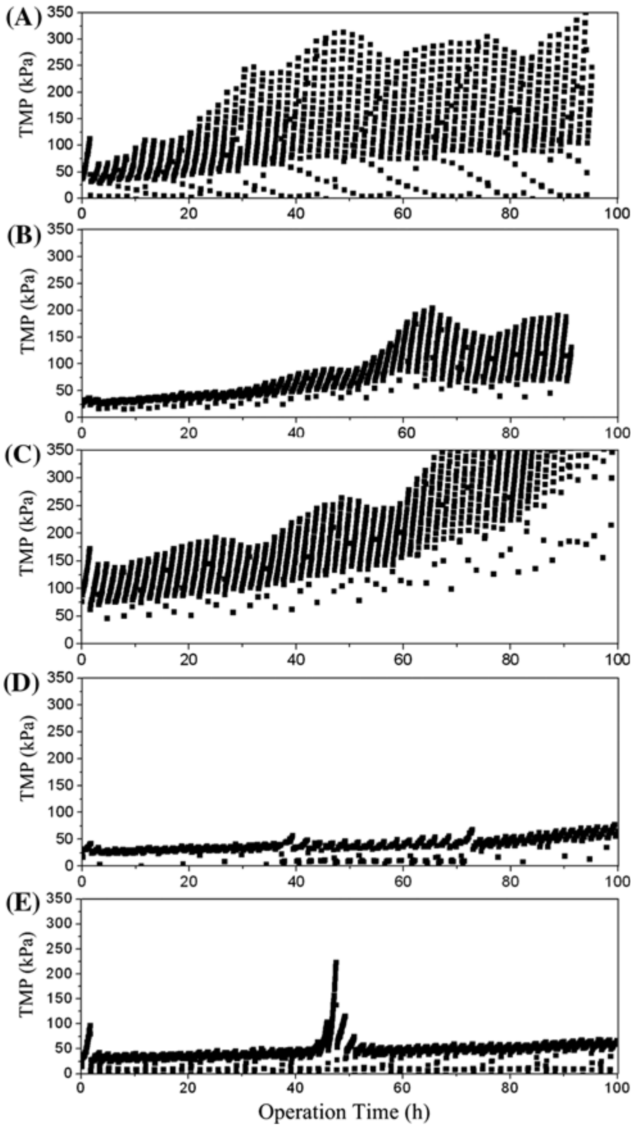

Fig. 4. TMP profile in all five tests: (A) #1 – secondary effluent from B, (B) #2 – secondary effluent from G, (C) #3 – secondary effluent from G, (D) #4 – secondary effluent from P, and (E) #5 – secondary effluent from P.

The feed from B contained more suspended particles and organic substances compared with that from the others mainly because of the aforementioned phenomenon. By contrast, the feed from P had a lower concentration of suspended particles, and thus the reversible fouling of each filtration cycle was lower (Fig. 4(D) and (E)).

However, the suspended solids concentration does not seem to be the sole factor affecting reversible fouling, at least under some conditions. As shown in Fig. 4(C)–(E), the reversible fouling values of each cycle throughout the filtration course were close to one another, which is reasonable because the suspended solids concentration values during the five tests were all close to one another (most data were between ±20% of the average level). On the other hand, the reversible fouling profiles shared an increasing trend from the beginning (Fig. 4(A) and (B)). The authors believe that this phenomenon is related to the critical filtration pressure (Section 3.4). However, this could not undermine the conclusion that suspended solids concentration is a main factor in determining reversible membrane fouling.

3.3. Effect of PSD on reversible fouling

A comparison between Fig. 4(B) from G and Fig. 4 (D) and (E) from P reveals that the feed water from G and P did not produce similar reversible fouling profiles although they had close suspended solids concentrations. Thus, aside from particle concentration, other factors can also affect reversible membrane fouling during dead-end filtration. Particle matter is mainly known to form a cake layer on the surface of membranes. This cake layer increased the filtration resistance. The permeability of the cake layer determines its filtration resistance, whereas the permeability is

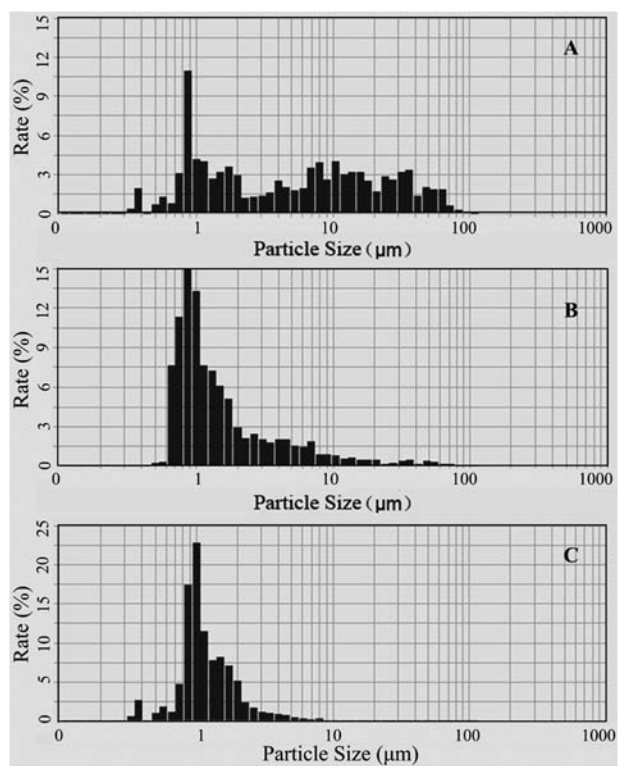

Fig. 5. Particle size distributions of secondary effluent from (A) WWTP B, (B) WWTP G, and (C) pilot equipment P (system model: Eye Tech S/N60294; laser lens: A100; meas. range: 0.1–100.0).

determined by thickness of the cake layer and the size distribution of the particles involved.

Based on the PSD of the three secondary effluent solutions (Fig. 5), the particles in the effluent from B, with sizes ranging from 0.2 μm to 70 μm, had a wider range compared with that from the other two effluent streams from G and P, respectively. Over 50% of the particles in the effluent from P had diameters between 0.9 and 1.1 μm. The PSD of the effluent from P is the most concentrated among the three. That is, majority of the particles in the effluent from P were similar in size. Assuming that the particles are spheres, then the porosity of a randomly packed bed of uniform spheres is 0.3–0.44 [27], whereas a mixture of widely sized distributed spheres can give a porosity as low as 0.0384 [28]. Therefore, if the compressibility of particles is ignored, the porosity of the cake layer accumulated by particles in the secondary effluent would have a strong relationship with its PSD, i.e. wide PSD leads to low porosity and high filtration resistance, and vice versa. The comparison between Fig. 5(B) and (C) reveals that the effluent from G (Fig. 5(B)) had a wider PSD, and had more small particles with diameters of approximately 0.7–0.8 μm. Thus, the effluent from G would result in a higher reversible fouling than the effluent from P. This result might explain the different reversible fouling profiles obtained in Fig. 4 (B), (D), and (E) even though their suspended solids concentration values were close to one another.

The comparison above was based on similar suspended solids concentration values between the effluents from G and P. If both suspended solids concentration and PSD largely differs, then the former factor is believed to be dominant.

3.4. Interaction of irreversible membrane fouling and suspended solids

Aside from the formation of a cake layer, suspended solids could also affect irreversible fouling through pore blocking. Huang et al. [29] reported that a critical range of particle sizes relative to membrane pore diameter could lead to efficient pore blocking, which probably means irreversible fouling. However, no direct evidence is available to support this viewpoint. As aforementioned, the particles with diameters less than 0.2 μm in all three secondary effluents were negligible. Therefore, whether the suspended solids cake layer could also affect irreversible fouling under some specific conditions should also be investigated.

Fig. 4(C) and (D) illustrates the TMP variations in tests #3 and #4, respectively. A comparison between these two figures demonstrates that both reversible and irreversible fouling in each cycle of test #3 were greater than those of test #4, although the feed suspended solids and organic concentration values of the former were slightly lower, which can be accounted to the high starting TMP (irreversible fouling) of test #3. Unlike in the other tests, the membrane used in test #3 was not sufficiently washed with chemicals before filtration. Therefore, its initial TMP was approximately 60 kPa, which was higher than that in the other tests (20–30 kPa). As shown in Fig. 4(A) and (B), reversible fouling significantly increased when TMP reached approximately 50 kPa (on abscissas, 30 h in Fig. 4(A) and 60 h in Fig. 4(B)). Thus, irreversible fouling or TMP may affect reversible fouling.

The cake layer is known to form with suspended solids from activated sludge. These suspended particles are compressible, rather than rigid. If operated in a constant pressure mode, the particles could be viewed as rigid, just like the gel layer formed by organic molecules. Foulant–foulant electrostatic repulsion was suggested to play a significant role in membrane fouling via the effluent organic matter under constant pressure condition [30]. Compared with the constant pressure mode, the filtration pressure increased with rising resistance in the constant flux mode. Normally, the cake layer would have a higher porosity under a relatively low hydraulic pressure, allowing dissolved and colloidal materials to permeate. However, higher filtration pressures result in a more compact fouling layer that is more difficult to hydraulically backwash [31]. Moreover, a compact fouling layer might retain more materials and result in the rapid increase in the rate of reversible fouling. Geng et al. reported a similar phenomenon [32]. They found that large flocs from mixed liquor were able to exert dual effects on membrane filtration. The flocs could either act as membrane foulants that cause fouling via particle deposition and cake formation, or serve as filters that entrap soluble and colloidal substances and thus alleviate the fouling. In the current study, 50 kPa might be a critical value for suspended solids in the effluent used. Hydraulic pressure above this critical value would result in the deformation of suspended particles and cake layer.

3.5. Effects of organic substances on irreversible membrane fouling

Dissolved and colloidal organic matters in irreversible fouling are believed to play a dominant role because their sizes are small enough to enter into the membrane pores. In test #1, the feed water evidently contained more organic substances than G and P

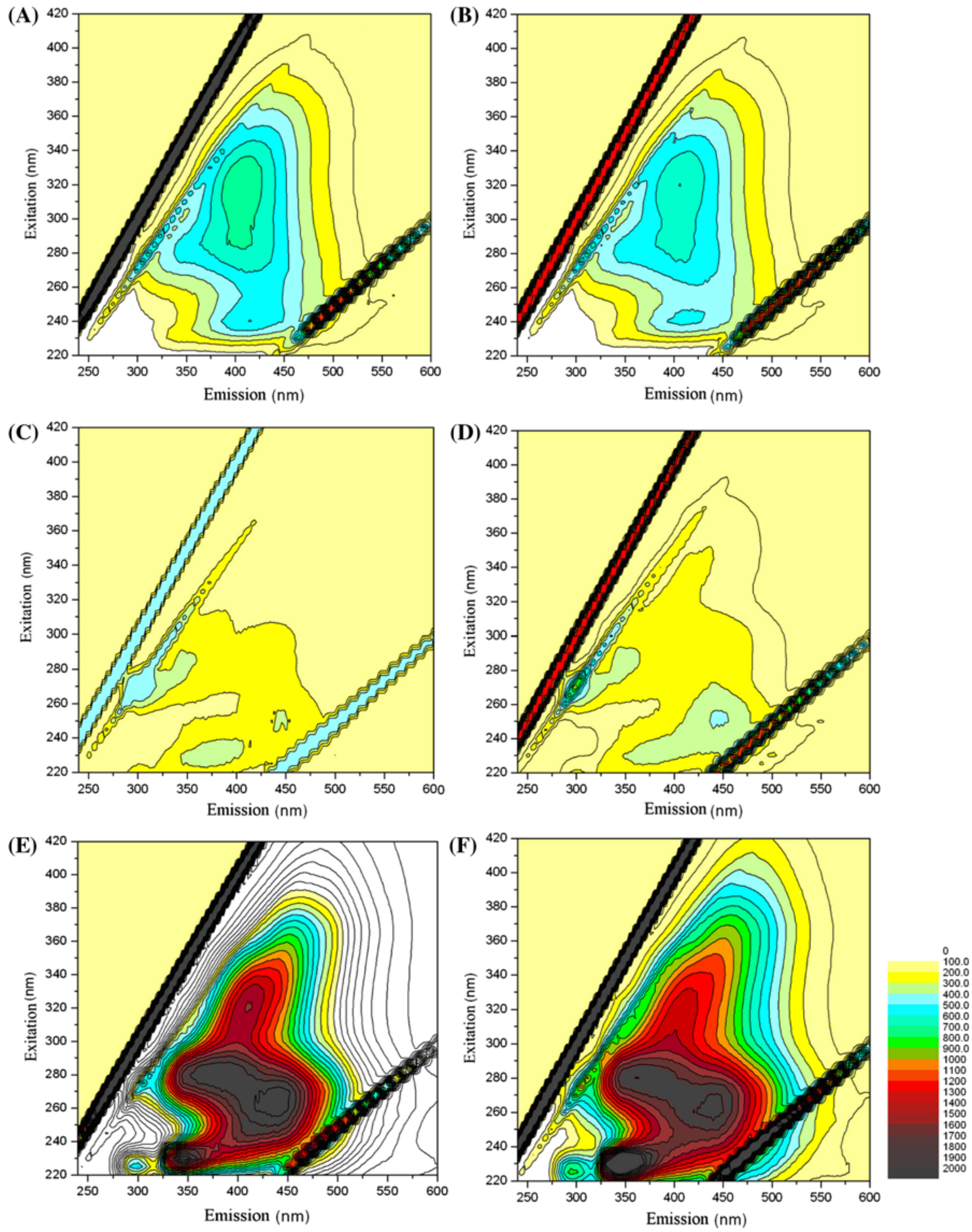

Fig. 6. Fluorescence spectrum of different organic compositions: (A) hydrophilic organics of P, (B) hydrophilic organics of G, (C) hydrophobic base of P, (D) hydrophobic base of G, (E) hydrophobic acid of P, and (F) hydrophobic acid of G (XAD-8 resin, Amberlite, 20–60 meshes).

(Table 2). Moreover, the effluent water from G and P had close organic substance concentration values. The organic substances in tests #2 and #4 were fractioned into hydrophilic substance, hydrophobic base, and hydrophobic acid with XAD-8 resin to explore further their differences. Fig. 6 shows the characterization of the different fractions using FEEM. FEEMs were reported to be very useful in distinguishing different types of organic matters; in addition, FEEM revealed that protein-like substances, rather than polysaccharides, are correlated with hydraulically irreversible fouling [33]. As shown in Fig. 6(A) and (B), the effluent from P had a similar but stronger fluorescent intensity than the effluent from G. Therefore, hydrophilic organics might not be the main reason for the fouling differences between tests #2 and #4. A peak area at excitation wavelengths (230–260 nm) and emission wavelengths (400–470 nm) appeared on the spectrum of the effluent from G, whereas no peak area appeared on the spectrum of the effluent from P (Fig. 6(C)–(D)). The substances relating to this peak may play key roles in the pore blockage and inner pore adsorption fouling of ceramic microfiltration membrane. These materials were reported to be humic acid-like or fulvic acid-like substances [34,35]. For the hydrophobic acid (Fig. 6(E) and (F)), there was no distinct difference. Therefore, materials, including humic acid, fulvic acid, humic acid-like, and fulvic acid-like substances might significantly contribute to the irreversible fouling of membrane during the microfiltration of secondary effluents. Moreover, these results coincide with the results of Jermann et al. [36], rather than those of Haberkamp et al. [33].

4. Conclusions

Three of the secondary effluents from the two WWTPs and one pilot scale, wastewater treatment equipment were used for the ceramic membrane filtration tests. The effects of feed water quality on the membrane fouling were concluded as follows:

(1) Higher particle concentration in feed water leads to a higher reversible fouling.
(2) Particle size distribution greatly affects reversible fouling during dead-end filtration. Wider PSD results in lower porosity of cake layer and higher cake layer resistance.
(3) A critical value for suspended solids in the effluent may have been used in the current study. A hydraulic pressure of above 50 kPa results in the deformation of suspended particles and a more compact cake layer.
(4) Compared with the effluent from P, the effluent from G had more humic substances, including humic acid, fulvic acid, and humic/fulvic acid-like substances. Moreover, based upon the analysis of FEEM results (Section 3.5), this humic substance might contribute greatly to the irreversible fouling during the ceramic microfiltration of secondary effluent.

Acknowledgments

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