Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.922503

54 (2015) 1457–1469
April

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Effect of water back-flushing and PP beads in hybrid water treatment of multi-channel alumina MF and photocatalyst-coated PP beads

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Received 15 January 2014; Accepted 29 April 2014

ABSTRACT

The effects of water back-flushing period (filtration time, FT) and time (BT) and polypropylene (PP) beads concentration were investigated in hybrid process of seven-channel ceramic microfiltration and PP beads coated with titanium dioxide (TiO_2) photocatalyst for advanced drinking water treatment. Instead of natural organic matters (NOM) and fine inorganic particles in natural water source, modified solution was prepared with humic acid and kaolin. As results of the effect of water back-flushing condition, FT 2 min and BT 10 s were the most effective to reduce membrane fouling and to maintain high permeate flux, and the highest total permeate volume (V_T) could be acquired at FT 2 min and BT 10 s in our experimental range. Thus, the optimal condition for the hybrid water treatment process would be FT 2 min and BT 10 s. The final permeate flux (J) and total permeate volumes (V_T) were the maximum at 40 g/L after 180 min's operation. Also, the treatment efficiencies of turbidity and humic acid were the maximum at 40 g/L of PP beads in our experimental range of 5−50 g/L. It means that the UV light could not transport to the PP beads located at the end position by blocking of another PP beads when PP beads concentration increase above 40 g/L. Finally, the optimum concentration of photocatalyst-coated PP beads would be 40 g/L in our experimental range.

Keywords: Microfiltration; Photocatalyst; Hybrid process; Ceramic membrane; Multi-channel; Water back-flushing

1. Introduction

Photocatalytic oxidation, a new type of water pollution control technology, with the characteristics of high efficiency, low energy consumption, and a wide range of application, can oxidize most organic compounds, especially non-biodegradable organic contaminants, by mineralizing them to small inorganic molecules. For this reason, photocatalytic oxidation technology has broad prospects for application. Among various semiconductor photocatalysts, there is a general consensus among researchers that TiO_2 is more superior because of its high activity, large stability to light illumination, and low price [1–4]. In

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Presented at the 6th International Conference on the "Challenges in Environmental Science and Engineering" (CESE-2013), 29 October–2 November 2013, Daegu, Korea

photocatalytic degradation, two modes of TiO_2 application are adopted: (1) TiO_2 immobilized on support materials and (2) TiO_2 suspended in aqueous medium [5,6]. Application of TiO_2 in suspension instead of immobilizing the TiO_2 on solid carriers has shown an improvement in organic degradation efficiencies due to the uniform distribution and large specific surface area. However, classical solid–liquid separation processes such as sedimentation, centrifugation and coagulation used for separation of the fine TiO_2 particles (typically less than 1 μm), are not effective [7]. In addition to the low reutilization rate, there is also a chance of secondary pollution caused by fine TiO_2 particles in the effluent. Therefore, the recovery of the photocatalysts is one of the main concerns that affect its engineering application on a large scale. A lot of investigations have been conducted aiming at solving this problem [8–12]. The TiO_2 particles were retained in the system by means of a membrane filtration unit that was configured externally to the membrane but operated in an air lift, low pressure manner equivalent to that of a submerged membrane system [11].

A major pending problem in drinking water treatment plants is to effectively remove natural organic matters (NOM). However, it has been difficult to remove soluble organic materials such as NOM by only microfiltration (MF) [13]. Additionally, NOM is one of the major materials that cause membrane fouling in membrane separation process applied to advanced drinking water treatment. Generally, the membrane fouling in drinking water treatment occurs from inorganic particles (e.g. iron, silica, and suspended solids) and organic compounds (e.g. humic substances, polysaccharides, proteins, and microorganisms) [14–16]. And the membrane fouling causes concentration polarization [17] and gel layer formation on membrane surface [18], and adsorption and pore blockage inside membrane pores [19]. The application of membrane process has the problem of membrane fouling and decline of permeate flux. So the membrane back-flushing is a general technology to minimize the membrane fouling, and to maintain a high permeate flux. Ceramic membrane used in this study has excellent chemical resistance, high mechanical strength, stable characteristics at high pressure and temperature, wide available range of pH and long lifetime compared with organic membranes [17]. The ceramic membrane has been applied to the water treatment field, especially in Japan, and recently operated at the Yeoncho water treatment plant located at Geoje Island in Korea (2012).

In this study, the effects of water back-flushing period (FT) and time (BT) and polypropylene (PP) beads concentration were investigated in hybrid process of seven-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for advanced drinking water treatment. The optimal operating conditions of water back-flushing and photocatalyst-coated PP beads concentration were induced experimentally. And the results were compared with our previous study [20] for the hybrid process of tubular ceramic ultrafiltration and PP beads coated with TiO_2 photocatalyst, which were the same PP beads utilized in this study to investigate the effect of membrane type, and our other result [21] of polyethersulfone (PES) beads loaded with TiO_2 photocatalyst and seven-channel ceramic MF membrane, which was the same membrane used here to observe the effect of photocatalyst bead classification.

2. Experiments

2.1. Materials

The multi-channel ceramic MF membrane (HC04) used in the study was coated with α-alumina on a supporting layer of α-alumina, and its pore size was 0.4 μm. We purchased the membrane from Dongseo Inc. in Korea, and the specifications of the multi-channel ceramic membrane are arranged in Table 1. The photocatalyst used was 4−6 mm PP beads coated with TiO_2 powder manufactured by Prof. Kim's group [22] using chemical vapor deposition method, presented in Table 2.

Instead of NOMs and fine inorganic particles in natural water source, a quantity of humic acid sodium salt (Aldrich, MW 2,000−500,000 Dalton) and kaolin (Sigma–Aldrich, MW 258.16 Dalton, $Al_2O_3{\cdot}2SiO_2{\cdot}2H_2O$) was dissolved in distilled water. It was then utilized as synthetic water in our experiment. The kaolin concentrations were fixed at 30 mg/L and the humic acid was 10 mg/L in the synthetic feed water to investigate the effect of water back-flushing period (FT) and time (BT), but the humic acid was 6 mg/L for that of PP beads concentration. Particle size distribution of the synthetic feed was measured by Laser particle size analyzer (Mastersizer 2000, MALVERN, UK) as shown in Fig. 1. The average particle sizes of (humic acid 6 mg/L + kaolin 30 mg/L) and (humic acid 10 mg/L + kaolin 30 mg/L) solutions were 4.418 and 4.692 μm, respectively, which were much higher than the pore size of the multi-channel ceramic MF membrane (HC04). UV light with 352 nm was radiated from

Table 1
Specifications of the multi-channel ceramic MF (HC04) used in this study

Membrane	HC04
Pore size (μm)	0.4
No. of channels	7
Outer diameter (mm)	20
Inner diameter (mm)	4
Length (mm)	235
Surface area (cm^2)	206.7
Material	α-alumina coating on α-alumina support
Company	Dongseo Inc. in Republic of Korea

Table 2
Specification of the TiO_2 coated PP beads used in this study

Material of the beads	Polypropylene (PP)
TiO_2 coating method	Chemical vapor deposition
Diameter (mm)	4–6
Weight (mg)	21.8–48.3
Average weight (mg)	39.9

outside of the acryl module by 2 UV lamps (F8T5BLB, Sankyo, Japan) having 8 W power.

2.2. Hybrid membrane module

For eliminating the turbidity and dissolved organic matters (DOM), the hybrid module was constructed by packing PP beads coated with TiO_2 powder between the module inside and outside of a ceramic membrane. In addition, 100 meshes (0.150 mm), which was extremely smaller than 4–6 mm particle size of the PP beads utilized here, was installed at the outlet of the hybrid module to prevent PP beads loss into the treated water tank.

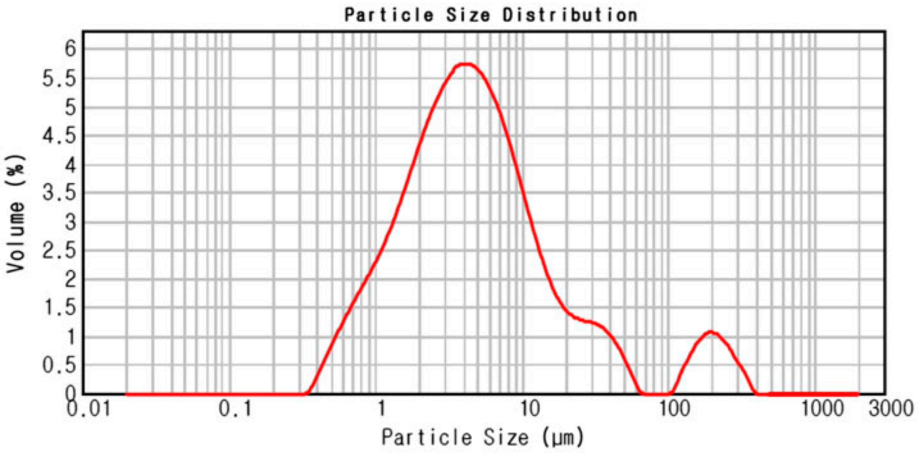
2.3. Experimental procedure

The advanced water treatment system utilizing a hybrid module (6) of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst was shown in Fig. 2, which was utilized at our previous study [23]. We performed cross-flow filtration for the multi-channel ceramic membrane and periodic water back-flushing utilizing permeated water. The hybrid module (6) filled up with 40 g/L of PP beads coated with TiO_2 photocatalyst in the gap of ceramic membrane and the acryl module case, and those were fluidized in the advanced hybrid water treatment system. Then the feed tank (1) was filled with 10 L of prepared synthetic water composed of humic acid and kaolin, and the temperature of the feed water was constantly maintained by using a constant temperature circulator (3) (Model 1146, VWR, USA). Also, the synthetic feed water was continuously mixed by a stirrer (4) in order to maintain the homogeneous condition of the feed water, and it was caused to flow into the inside of the multi-channel ceramic membrane by a pump (2) (Procon, Standex Co., USA). The feed flow rate was measured by a flow meter (5) (NP-127, Tokyo Keiso, Japan). The flow rate and pressure of the feed water that flowed into the hybrid module was constantly maintained by controlling valves (9) of both the bypass pipe of the pump (2) and the concentrate pipe. The permeate flux treated by both the multi-channel ceramic membrane and photocatalyst was measured by an electric balance (11) (Ohaus, USA). The permeate water flowed into the back-washing tank (13) when the permeate flux had not been measured. After the treated water was over a certain level in the back-washing tank (13), it was recycled to the feed tank (1) to maintain a constant concentration of the feed water during operation.

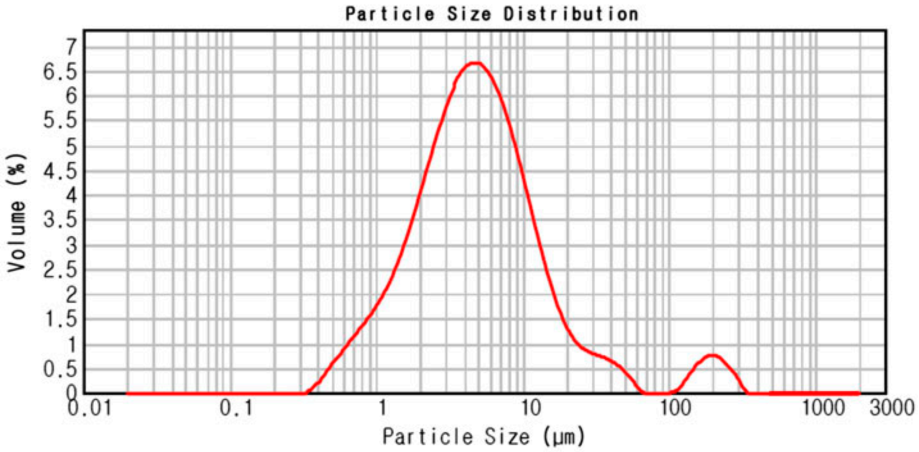
The kaolin and humic acid concentration was fixed at 30 and 10 mg/L, respectively, in the synthetic feed water to observe the effect of water back-flushing condition. The back-flushing time (BT) was fixed at 10 s and filtration time (FT) was changed as 2, 4, 6, 8, and 10 min to investigate the effect of back-flushing period. Also, FT was fixed at 10 min and BT was changed as 2, 4, 6, 8, and 10 s to research the effect of BT. The kaolin and humic acid concentrations were fixed at 30 and 6 mg/L, respectively, in the synthetic feed water to investigate the effect of photocatalyst-coated PP beads concentration. The TiO_2 photocatalyst-coated PP beads concentration was changed as 5, 10, 20, 30, 40, 50 g/L to observe the effect of PP beads concentration.

The resistance of membrane fouling (R_f), dimensionless permeate flux (J/J_0), and permeate flux (J) were calculated and compared during 180 min of total filtration time under each condition, where J_0 was the initial permeate flux which was estimated by extrapolation using initial 2 data at 1 and 2 min. Under all experimental conditions, trans-membrane pressure (TMP) was maintained constant at 0.8 bar, the water back-flushing pressure at 1.0 bar, the feed flow rate at 1.0 L/min, and the feed water temperature at 20℃.

The quality of feed water and treated water was analyzed in order to evaluate the treatment efficiencies of turbid materials and DOM in the hybrid process of multi-channel ceramic MF and PP beads



(a) Humic acid 6 mg/L and kaolin 30 mg/L (Average particle size: 4.418 μm)



(b) Humic acid 10 mg/L and kaolin 30 mg/L (Average particle size: 4.692 μm)

Fig. 1. Particle size distribution of the synthetic feed solution including humic acid and kaolin. (a) Humic acid 6 mg/L and kaolin 30 mg/L (average particle size: 4.418 μm). (b) Humic acid 10 mg/L and kaolin 30 mg/L (average particle size: 4.692 μm).

coated with TiO_2 photocatalyst. Turbidity was measured by a turbidimeter (2100 N, HACH, USA) and UV_{254} absorbance was analyzed by a UV spectrophotometer (GENESYS 10 UV, Thermo, USA). The detection limits of turbidimeter and UV spectrophotometer were 0−4,000 NTU (±0.001 NTU) and $-0.1-3.0\,cm^{-1}$ ($\pm 0.001\,cm^{-1}$). Before the measurement of UV_{254} absorbance, the sample was filtered by 0.2 μm syringe filter to remove turbid materials.

3. Results and discussion

The effects of water back-flushing period (FT), time (BT) and PP beads concentration were investigated in the hybrid process of seven-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for advanced water treatment. Resistances of membrane, boundary layer, and membrane fouling (R_m, R_b, R_f) were calculated from permeate flux (J) data using the resistance-in-series filtration equation as the same method as our previous study [23]. The effects of water back-flushing condition and PP beads concentration could be compared clearly during 180 min's operation.

3.1. *Effect of water back-flushing period (FT)*

To investigate the optimal condition of FT, FT was changed as 2, 4, 6, 8, and 10 min at fixed BT 10 s in the hybrid water treatment process. The resistances of membrane fouling (R_f) were calculated from the experimental J data of FT effect, and compared at fixed BT of 10 s in Fig. 3. As shown in Fig. 3, R_f was

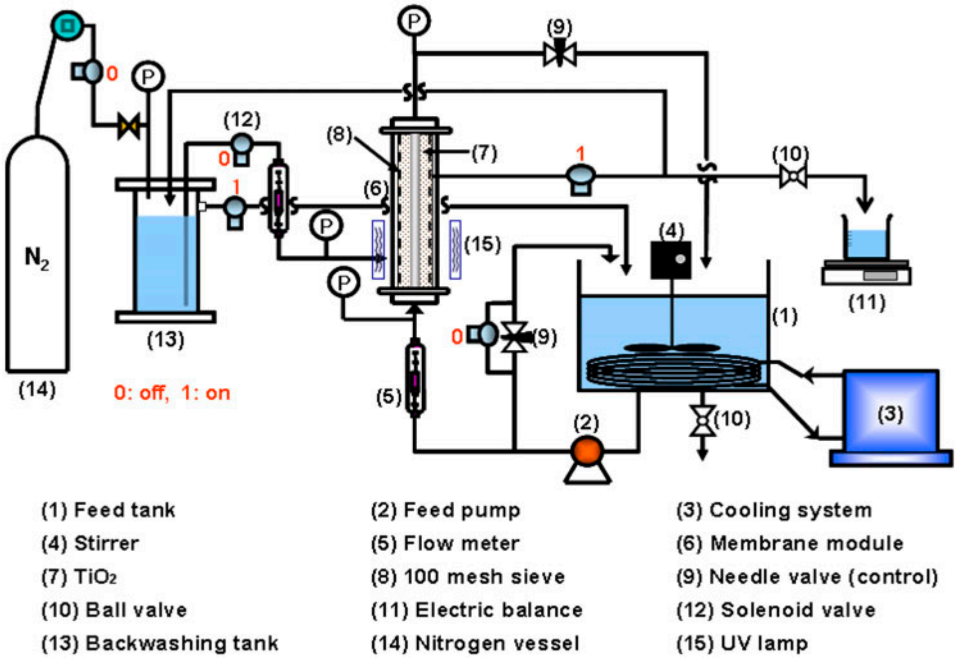


Fig. 2. Apparatus of advanced water treatment system using hybrid module of ceramic MF and PP beads coated with TiO_2 photocatalyst with periodic water back-flushing [20].

the minimum at FT 2 min, and increased dramatically with rising FT from 2 to 10 min, and finally had the maximum value at no back-flushing (NBF) condition. It means that the shorter water back-flushing period was the more effective to reduce a cake layer on the membrane surface and a fouling inside the membrane, and the resistance of membrane fouling presented the maximum value at the shortest FT. These results were the same trends with our previous work [20] for the hybrid process of tubular ceramic ultrafiltration and TiO_2 photocatalyst-coated PP beads, which were the same PP beads utilized in this study, and our other result [21] of photocatalyst-loaded PES beads and seven-channel ceramic MF membrane, which was the same membrane used here.

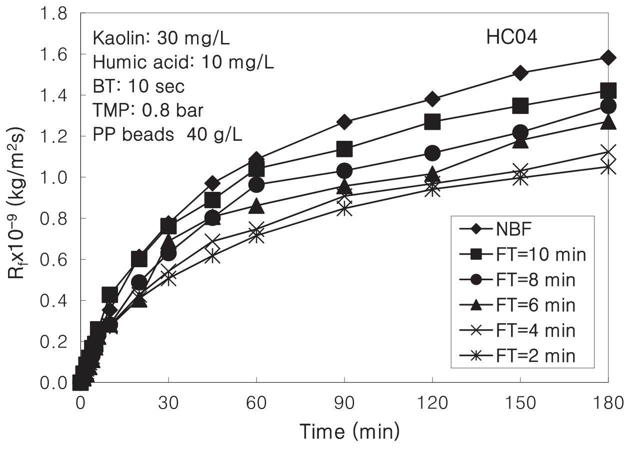


Fig. 3. Effect of water back-flushing period on resistance of membrane fouling in hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst.

The lowest value of the final dimensionless permeate flux (J_{180}/J_0), where J_{180} was the final permeate flux after 180 min's operation, was 0.322 at NBF as shown in Fig. 4 and Table 3. And the curves of J/J_0 increased as decreasing FT from 10 to 2 min, and finally the maximum value of J_{180}/J_0 was 0.408 at FT 2 min. In Table 3, R_m was fluctuated below ±5% because the alumina membrane was used again after chemical cleaning by alkaline and acid solution during 2 and 12 h, respectively. As results, FT 2 min should be the most effective water back-flushing period to reduce membrane fouling and to maintain high permeate flux during three hours' operation at our experimental range in this hybrid water treatment process. Then, the highest total permeate volume (V_T) was 11.50 L at the lowest FT 2 min because the lowest final R_f ($R_{f,180}$) was $1.051 \times 10^9\ kg/m^2s$ at FT 2 min, as arranged in Table 3. It was the same trends with our previous results [20,21].

As shown in Table 4, the highest treatment efficiency of turbidity was 98.1% at NBF condition, because the severe fouling inside the membrane and

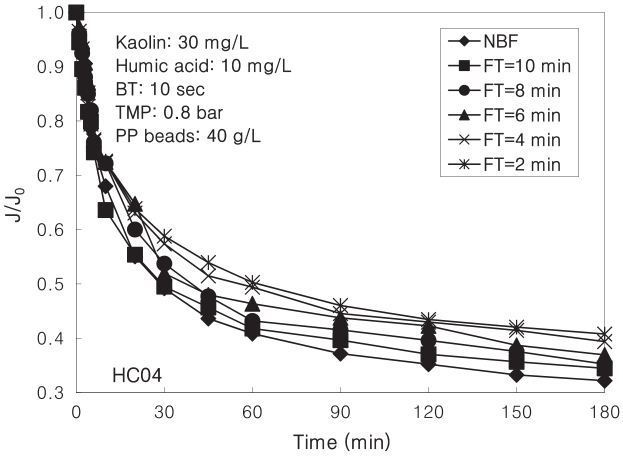


Fig. 4. Effect of water back-flushing period on dimensionless permeate flux in hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst.

the thick cake layer on the membrane surface have generated without any water back-flushing and could remove kaolin effectively. The average treatment efficiencies in Table 4 was averaged the treatment efficiency values measured at every 30 min. And the treatment efficiency of turbidity decreased as dropping FT from 10 to 2 min. It means that the more frequent water back-flushing could make the cleaner surface of the photocatalyst-coated PP beads, and adsorption by PP beads could remove kaolin the more effectively. Then, the highest treatment efficiency of humic acid (UV_{254} absorbance) was 85.3% at FT 2 min, because the more frequent water back-flushing should make the cleaner photocatalyst-coated PP beads and those could activate the more effectively the photooxidation and adsorption for humic acid, as arranged in Table 5. Absolutely, the highest treatment efficiency of humic acid was 85.2% at NBF, because the thickest cake layer on the membrane surface and the most severe membrane fouling inside the membrane could remove humic acid the most excellently.

The results of Tables 4 and 5 could be compared with our previous study [20] for the hybrid process of tubular ceramic ultrafiltration and photocatalyst-coated PP beads, and our other result [21] of photocatalyst-loaded PES beads and seven-channel ceramic MF membrane. The average treatment efficiencies of turbidity and humic acid increased as decreasing FT except NBF in the previous study [20,21], which was the exactly same trend of this study. But the treatment efficiencies of turbidity and humic acid were the lower than those of the previous study [20], because the membrane in this study had the larger pore size of 0.4 μm than 0.05 μm in the previous study [20], and the smaller pore size could accumulate the denser cake layer on the membrane surface and the cake layer removed the kaolin or humic acid the more effectively. Then, the turbidity treatment efficiency in this study was almost same with that of the previous study [21], but the treatment efficiency of humic acid was the lower than that of the previous study [21]. It means that PP beads coated with photocatalyst by the chemical vapor deposition was the less effective to remove humic acid than PES beads loaded with photocatalyst by the phase inversion in this hybrid water treatment process.

3.2. Effect of water back-flushing time (BT)

To induce the optimal BT condition, BT was changed as 2, 4, 6, 8, and 10 s at fixed FT 10 min in the hybrid water treatment process. The resistance of membrane fouling (R_f) increased dramatically as dropping BT from 10 to 2 s, as shown in Fig. 5. Finally, $R_{f,180}$ value after 180 min's operation at BT 2 s should be much smaller than the $R_{f,180}$ at BT 10 s, and the lowest $R_{f,180}$ was 1.421 kg/m^{2}s at BT 8 s, which was

Table 3
Effect of water back-flushing period on filtration factors for hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst (BT 10 s)

FT (min)	NBF*	10	8	6	4	2
$R_m \times 10^{-9}$ (kg/m^2 s)	0.664	0.683	0.668	0.671	0.683	0.663
$R_b \times 10^{-9}$ (kg/m^2 s)	0.087	0.064	0.065	0.073	0.047	0.060
$R_{f,180} \times 10^{-9}$ (kg/m^2 s)	1.583	1.422	1.347	1.272	1.123	1.051
J_w (L/m^2 h)	420	428	423	420	413	426
J_0 (L/m^2 h)	376	378	385	379	387	390
J_{180} (L/m^2 h)	121	130	136	140	152	159
J_{180}/J_0	0.322	0.344	0.352	0.369	0.394	0.408
V_T (L)	9.24	9.59	10.33	10.55	11.18	11.50

*No back-flushing.

Table 4
Water quality and treatment efficiency of turbidity in the hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for effect of water back-flushing period (BT 10 s)

Experimental condition			Turbidity (NTU)				
			Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	FT (min)	Range	Average	Range	Average	Average treatment efficiency (%)
30	10	NBF*	18.0–29.8	24.2	0.380–0.601	0.471	98.1
		10	25.1–32.1	28.5	0.905–1.200	1.011	96.4
		8	28.5–32.9	31.9	0.770–1.181	0.956	97.0
		6	29.7–33.5	31.7	0.734–0.896	0.811	97.4
		4	25.4–32.8	30.0	0.698–0.988	0.759	97.5
		2	30.1–33.1	31.9	0.586–0.815	0.673	97.9

*No back-flushing.

Table 5
Water quality and treatment efficiency of humic acid (UV_{254} absorbance) in the hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for effect of water back-flushing period (BT 10 s)

Experimental condition			UV_{254} absorbance (cm^{-1})				
			Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	FT (min)	Range	Average	Range	Average	Average treatment efficiency (%)
30	10	NBF*	0.104–0.181	0.142	0.014–0.030	0.021	85.2
		10	0.115–0.190	0.152	0.023–0.039	0.032	78.9
		8	0.117–0.205	0.152	0.022–0.038	0.030	80.4
		6	0.109–0.191	0.151	0.015–0.035	0.026	82.6
		4	0.100–0.193	0.138	0.015–0.030	0.022	84.0
		2	0.108–0.195	0.152	0.015–0.029	0.022	85.3

*No back-flushing.

almost same with that of BT 10 s, as shown in Table 6. However, the highest $R_{f,180}$ was 1.583 kg/m^2s at NBF. It means that the longer water back-flushing time was the more effective to reduce membrane fouling. These results were the same trends with our previous work [20] for the hybrid process of tubular ceramic ultrafiltration and photocatalyst-coated PP beads, but there was not the effect of BT in our other result [21] about the hybrid process of photocatalyst-loaded PES beads and seven-channel ceramic MF membrane.

As shown in Fig. 6, the dimensionless permeate flux (J/J_0) decreased a little as decreasing BT from 10 to 2 s. However, the lowest J_{180}/J_0 was 0.322 at NBF after 180 min's operation, as arranged in Table 6. Also, the highest total permeate volume (V_T) was 9.59 L at the longest BT 10 s. It means that the longer water back-flushing time could reduce the membrane fouling the more effectively, and improve the permeate flux during operation, and finally, acquire the highest V_T. It was the same trends with our previous results [20].

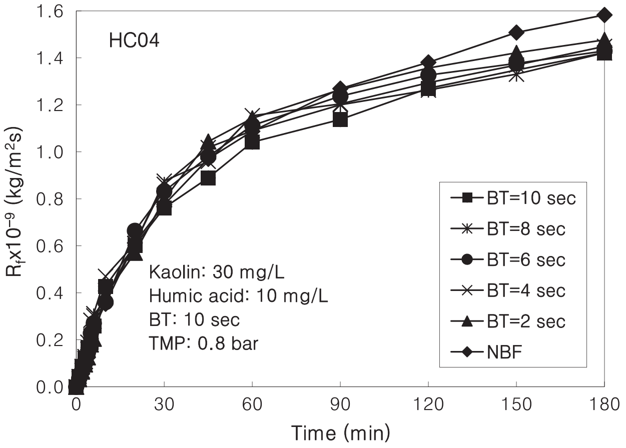


Fig. 5. Effect of water back-flushing time on resistance of membrane fouling in hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst.

Table 6

Effect of water back-flushing time on filtration factors for hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst (FT 10 min)

BT (sec)	10	8	6	4	2	NBF*
$R_m \times 10^{-9}$ (kg/m^{2}s)	0.683	0.690	0.687	0.689	0.697	0.664
$R_b \times 10^{-9}$ (kg/m^{2}s)	0.064	0.061	0.063	0.059	0.057	0.087
$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)	1.422	1.421	1.429	1.449	1.477	1.583
J_w (L/m^{2}h)	413	409	411	410	405	425
J_0 (L/m^{2}h)	378	376	381	378	374	376
J_{180} (L/m^{2}h)	130	130	130	128	127	121
J_{180}/J_0	0.344	0.346	0.340	0.340	0.338	0.322
V_T (L)	9.59	9.41	9.32	9.30	9.25	9.24

*No back-flushing.

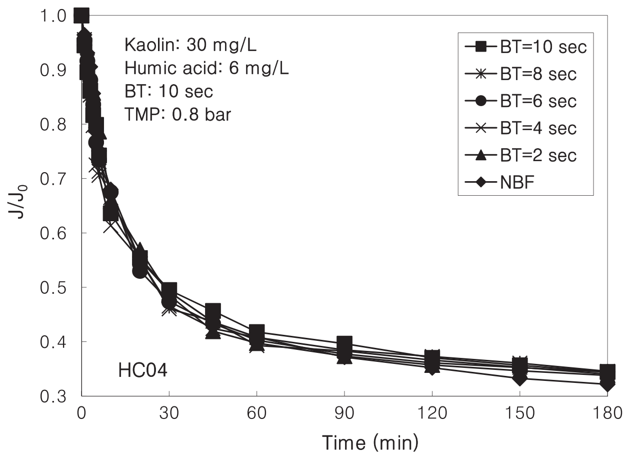


Fig. 6. Effect of water back-flushing time on dimensionless permeate flux in hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst.

The treatment efficiency of turbidity was almost constant, but that of humic acid (UV_{254} absorbance) increased as dropping BT from 10 to 2 s, as shown in Tables 7 and 8, because the membrane fouling increased dramatically as decreasing BT and the cake layer on the membrane surface and fouling inside the membrane could remove the humic acid the more effectively. Finally, the highest treatment efficiencies of turbidity and humic acid was 98.1 and 85.2% at NBF, respectively, because the pore size could be reduced by pore blocking and cake layer on the membrane surface, and the turbid materials and humic acid removed the most excellently at NBF.

Finally, the optimal water back-flushing condition of the hybrid water treatment process of multi-channel ceramic MF membrane and PP beads coated with TiO_2 photocatalyst should be FT 2 min and BT 10 s in our experimental range because of the lowest resistance of membrane fouling and the highest total permeate volume as compared in Tables 3 and 6.

The results of Tables 7 and 8 could be compared with our previous study [20] for the hybrid process of tubular ceramic ultrafiltration and photocatalyst-coated PP beads. The average treatment efficiency of turbidity was almost constant independent of BT in this study, but those of turbidity in the previous study [20] increased as rising BT except NBF. Also, the treatment efficiency of humic acid in the previous study [20] increased as rising BT except NBF, which was a reverse trend of this study, because the longer water back-flushing time could make the cleaner surface of PP beads, and kaolin adsorbed and humic acid photo-oxidized on the PP beads' surface the more effectively in the result [20]. However, the treatment efficiencies of turbidity and humic acid were smaller than those of the previous study [20], because the membrane utilized in this study had the larger pore size of 0.4 μm than 0.05 μm in the result [20], and the smaller pore size could make the denser cake layer on the membrane surface and the cake layer remove the kaolin and humic acid the more effectively.

3.3. Effect of photocatalyst-coated PP beads concentration

To observe the effect of photocatalyst-coated PP beads concentration and to investigate the optimal PP beads amount, the photocatalyst-coated PP beads were fluidized from 5 to 50 g/L concentration between the gap of ceramic membrane and the acryl module case in the hybrid process of seven-channel ceramic MF and the PP beads. As shown in Fig. 7, the resistance of membrane foiling (R_f) dropped as increasing the PP beads concentration from 5 to 40 g/L, but increased dramatically at 50 g/L after 180 min's operation. The lowest $R_{f,180}$ was 1.015×10^9 kg/m^{2}s at 40 g/L of PP beads, and the highest $R_{f,180}$ was 1.333×10^9 kg/m^{2}s at

Table 7
Water quality and treatment efficiency of turbidity in the hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for effect of water back-flushing time (FT 10 min)

Experimental condition			Turbidity (NTU)				
			Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	BT (sec)	Range	Average	Range	Average	Average treatment efficiency (%)
30	10	10	25.1–32.1	28.5	0.905–1.200	1.011	96.4
		8	25.8–32.9	28.5	0.825–1.210	1.018	96.4
		6	23.8–31.2	27.8	0.875–1.040	0.948	96.3
		4	21.9–31.1	27.1	0.521–1.100	0.851	96.9
		2	23.0–32.5	28.8	0.709–1.200	0.954	96.7
		NBF*	18.0–29.8	24.2	0.380–0.601	0.471	98.1

*No back-flushing.

Table 8
Water quality and treatment efficiency of humic acid (UV_{254} absorbance) in the hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for effect of water back-flushing time (FT 10 min)

Experimental condition			UV_{254} absorbance (cm^{-1})				
			Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	BT (sec)	Range	Average	Range	Average	Average treatment efficiency (%)
30	10	10	0.115–0.190	0.152	0.023–0.039	0.032	78.9
		8	0.108–0.196	0.149	0.018–0.034	0.026	82.9
		6	0.108–0.188	0.147	0.018–0.032	0.024	83.4
		4	0.105–0.186	0.146	0.016–0.029	0.024	83.9
		2	0.110–0.192	0.146	0.018–0.030	0.023	84.1
		NBF*	0.104–0.181	0.142	0.014–0.030	0.021	85.2

*No back-flushing.

50 g/L, as shown in Table 9. Therefore, the optimal concentration of photocatalyst-coated PP beads could be 40 g/L in this hybrid process of multi-channel ceramic MF and photocatalyst-coated PP beads in the view point of membrane fouling. These results were the same trends with our previous work [20] for the hybrid process of tubular ceramic ultrafiltration and the photocatalyst-coated PP beads, and our other result [21] of photocatalyst-loaded PES beads and seven-channel ceramic MF membrane.

As shown in Fig. 8, the dimensionless permeate flux (J/J_0) increased as rising PP beads concentration from 5 to 40 g/L, but dropped dramatically at 50 g/L. Finally the J_{180}/J_0 value at 40 g/L of PP beads was the maximum as shown in Table 9. The highest permeate flux (J) after 180 min's operation was 164 L/m^{2}h at 40 g/L of PP beads. Also, the total permeate volumes (V_T) at 40 and 50 g/L of PP beads were 11.50 and 9.80 L, respectively, which were the maximum and the minimum values in our experimental range. It means that too many PP beads could generate the more membrane fouling

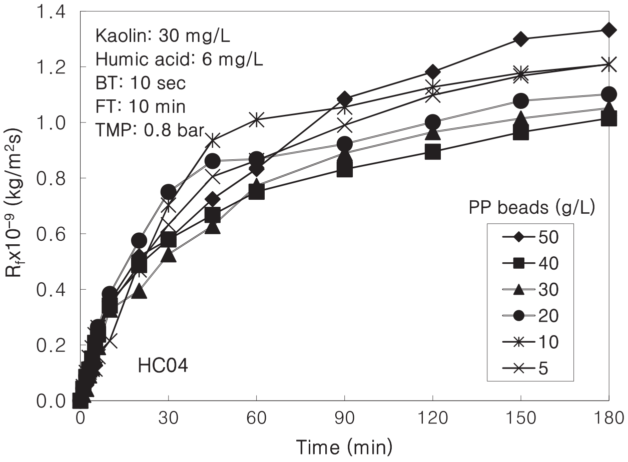


Fig. 7. Effect of PP beads concentration on resistance of membrane fouling in hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst.

Table 9
Effect of PP beads concentration on filtration factors for hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst

PP beads (g/L)	50	40	30	20	10	5
$R_m \times 10^{-9}$ (kg/m^2 s)	0.777	0.663	0.660	0.643	0.668	0.760
$R_b \times 10^{-9}$ (kg/m^2 s)	0.030	0.046	0.048	0.047	0.021	0.069
$R_{f,180} \times 10^{-9}$ (kg/m^2 s)	1.333	1.015	1.052	1.102	1.209	1.210
J_w (L/m^{2}h)	363	426	428	439	423	371
J_0 (L/m^{2}h)	349	398	399	409	410	341
J_{180} (L/m^{2}h)	132	164	160	158	149	138
J_{180}/J_0	0.377	0.411	0.402	0.385	0.363	0.407
V_T (L)	9.80	11.50	11.47	10.88	10.46	9.96

and reduce the permeate flux because the UV light could not transport to the PP beads located at the end position by blocking of another PP beads. It was the same results with our previous studies [20,21].

As arranged in Tables 10 and 11, the treatment efficiencies of turbidity and humic acid (UV_{254} absorbance) increased from 95.9 to 98.1% and from 73.4 to 79.8%, respectively, when PP beads concentration changed from 5 to 40 g/L. But the treatment efficiencies of turbidity and humic acid at 50 g/L of PP beads dropped to the lower 97.3 and 76.7% than those at 40 g/L. Finally, the highest treatment efficiencies of turbidity and humic acid were at 40 g/L of PP beads, and the lowest treatment efficiencies of those were at 5 g/L. Therefore, the optimal concentration of photocatalyst-coated PP beads should be at 40 g/L in the view point of treatment efficiencies in this hybrid water treatment process of multi-channel ceramic MF and photocatalyst-coated PP beads, too. This result was the same with our previous work [20]. It means that the optimal concentration of photocatalyst-coated PP beads could be independent of membrane classification in this hybrid water treatment process. However, the treatment efficiency of humic acid was the maximum at 50 g/L of PES beads in our previous work [21]. It means that the optimal beads concentration could be different depending on the classification of photocatalyst beads.

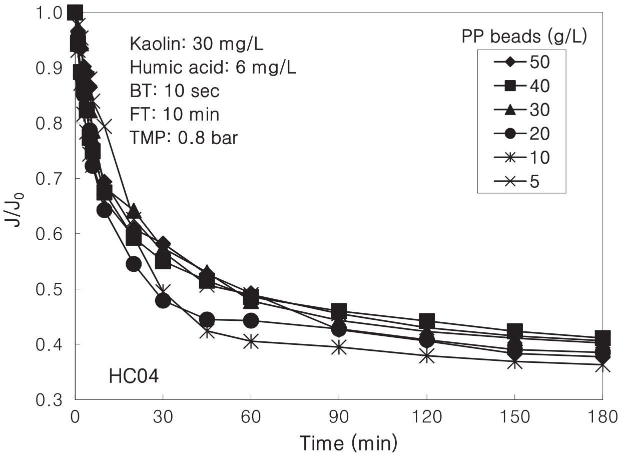


Fig. 8. Effect of PP beads concentration on dimensionless permeate flux in hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst.

All of photocatalysts on the PP beads surface could be active under UV light when PP beads concentration was 40 g/L and less. But when PP beads concentration increase above 40 g/L, the UV light could not transport to the PP beads located at the end position by blocking of another PP beads. Therefore, some photocatalyst on PP beads should not be active, and treatment efficiencies of turbidity and humic acid decreased. This result showed a similar trend with the other's research [24] about photocatalytic degradation of organic matter by pure TiO_2 powder.

The results of Tables 10 and 11 were compared with our previous study [20,21]. The average treatment efficiency of turbidity was almost constant independent of photocatalyst-coated PP beads concentration in the previous study [20]. But those of turbidity in this study decreased as dropping the PP beads concentration because the membrane in this study had the larger pore size of 0.4 μm than 0.05 μm in the previous study [20]. Also, the average treatment efficiency of humic acid decreased as dropping the PP beads concentration in the previous study [20]. Then, treatment efficiency of humic acid in this study was much lower than those of the previous study [20], because the smaller pore size could accumulate the denser cake layer on the membrane surface and the cake layer remove humic acid the more effectively. Then the turbidity treatment efficiency in this study was almost same with that of the previous study [21], but the humic acid treatment efficiency was the lower

Table 10
Water quality and treatment efficiency of turbidity in the hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for effect of PP beads concentration

Experimental condition			Turbidity (NTU)				
			Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	PP beads (g/L)	Range	Average	Range	Average	Average treatment efficiency (%)
30	6	50	23.4−29.9	26.5	0.587−0.855	0.721	97.3
		40	24.3−31.9	27.8	0.452−0.614	0.541	98.1
		30	25.1−31.5	28.4	0.513−0.801	0.638	97.8
		20	25.3−31.8	28.3	0.928−1.310	1.128	96.0
		10	23.5−31.4	26.1	0.894−1.217	1.037	96.0
		5	22.4−29.5	25.7	0.800−1.310	1.042	95.9

Table 11
Water quality and treatment efficiency of humic acid (UV_{254} absorbance) in the hybrid process of multi-channel ceramic MF and PP beads coated with TiO_2 photocatalyst for effect of PP beads concentration

Experimental condition			UV_{254} absorbance (cm^{-1})				
			Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	TiO_2 (g/L)	Range	Average	Range	Average	Average treatment efficiency (%)
30	6	50	0.078−0.126	0.100	0.020−0.026	0.023	76.7
		40	0.060−0.138	0.089	0.014−0.024	0.018	79.8
		30	0.061−0.128	0.086	0.016−0.028	0.020	76.6
		20	0.057−0.122	0.083	0.014−0.023	0.020	76.4
		10	0.060−0.131	0.086	0.015−0.027	0.020	76.3
		5	0.070−0.120	0.090	0.022−0.028	0.024	73.4

than that of the previous study [21]. It proved again that PP beads coated with photocatalyst by the chemical vapor deposition was the less effective to remove humic acid than PES beads loaded with photocatalyst by phase inversion in this hybrid water treatment process.

4. Conclusions

In this study, the effects of water back-flushing period (FT), time (BT), and photocatalyst-coated PP beads concentration were observed and the optimal conditions were investigated in a hybrid water treatment process of seven-channel alumina MF and the PP beads for advanced water treatment. The seven-channel ceramic MF membrane was coated with α-alumina on a supporting layer of α-alumina, and its pore size was 0.4 μm. The photocatalyst was PP beads coated with TiO_2 powder manufactured by the chemical vapor deposition method. Instead of NOMs and fine inorganic particles in natural water source, a quantity of humic acid sodium salt and kaolin was dissolved in distilled water, and utilized as synthetic water.

(1) To investigate the optimal condition of FT and BT, FT was changed as 2, 4, 6, 8, and 10 min at fixed BT 10 s, and BT was changed as 2, 4, 6, 8, and 10 s at fixed FT 10 min in the hybrid water treatment process. As results of the effect of FT and BT, FT 2 min and BT 10 s were the most effective to reduce membrane fouling and to maintain high permeate flux, and the highest total permeate volume (V_T) of 11.50 L could be acquired at FT 2 min and BT 10 s in our experimental range of the hybrid water treatment process. Thus, the optimal water back-flushing condition in the hybrid water treatment process should be FT 2 min and BT 10 s. However, the humic acid could be removed the most effectively at NBF because of the thickest cake layer on the

membrane surface and the most severe membrane fouling inside the membrane.

(2) The results of FT effect could be compared with our previous study [20] for the hybrid process of tubular ceramic ultrafiltration and photocatalyst-coated PP beads, and other result [21] of photocatalyst-loaded PES beads and seven-channel ceramic MF membrane. The treatment efficiencies of turbidity and humic acid were the lower than those of the study [20], because the membrane in this study had the larger pore size of 0.4 μm than 0.05 μm in the study [20], and the smaller pore size could make the denser cake layer on the membrane surface and the cake layer remove the kaolin or humic acid the more effectively. Then, the humic acid treatment efficiency was the lower than that of the study [21]. It means that PP beads coated with photocatalyst by the chemical vapor deposition was the less effective to remove humic acid than PES beads loaded with photocatalyst by phase inversion in this hybrid water treatment process.

(3) To investigate the optimal amount of photocatalyst-coated PP beads in the hybrid water treatment process, the PP beads were fluidized from 5 to 50 g/L. The permeate flux (J) after 180 min's operation and the total permeate volumes (V_T) were the maximum at 40 g/L, but the minimum at 50 g/L of PP beads. Therefore, the optimal concentration of photocatalyst-coated PP beads could be 40 g/L in this hybrid process in the view point of membrane fouling. It means that too many PP beads could generate the more membrane fouling and reduce the permeate flux because the UV light could not transport to the PP beads located at the end position by blocking of another PP beads. It was the same results with our previous studies [20,21].

(4) The treatment efficiencies of turbidity and humic acid increased when PP beads concentration changed from 5 to 40 g/L. But the treatment efficiencies of turbidity and humic acid at 50 g/L of PP beads dropped than those at 40 g/L. Therefore, the optimal concentration of photocatalyst-coated PP beads should be at 40 g/L in the view point of treatment efficiencies in this hybrid water treatment process, too. This result was the same with our previous work [20]. It means that the optimal concentration of photocatalyst-coated PP beads could be independent of membrane classification in this hybrid water treatment process. However, the treatment efficiency of humic acid was the maximum at 50 g/L of PES beads in our previous work [21]. It means that the optimal beads concentration could be different depending on the classification of photocatalyst beads.

Acknowledgment

This research was supported by Korea Ministry of Environment as "The Eco-Innovation project (Global Top project)" (GT-SWS-11-01-004-0) and by Hallym University Research Fund, 2014 (HRF-201404-008).

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