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광촉매 및 다채널 세라믹 정밀여과 혼성공정에 의한 고탁도 원수의 고도정수처리: 유기물의 영향

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Advanced Water Treatment by Hybrid Process of Multi-channel Ceramic MF and Photocatalyst: Effect of Organic Materials

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Abstract: For advanced drinking water treatment of high turbidity water, we used the hybrid process that was composed of photocatalyst packing in space of between outside of multi-channel ceramic microfiltration membrane and membrane module inside. Photocatalyst was polypropylene (PP) beads coated TiO_2 powder by CVD (chemical vapor deposition) process. Instead of natural organic matters (NOM) and fine inorganic particles in natural water source, standard NOM solution was prepared with humic acid and kaolin. Water-back-flushing of 10 sec was performed per every period of 10 min to minimize membrane fouling. Resistance of membrane fouling (R_f) increased and J decreased as concentration of humic acid changed from 2 mg/L to 10 mg/L, and finally the highest total permeate volume (V_T) could be obtained at 2 mg/L. Then, treatment efficiency of turbidity and UV_{254} absorbance were above 96.4% and 78.9%, respectively. As results of treatment portions by membrane filtration, photocatalyst adsorption, and photo-oxidation in (MF), (MF + TiO_2), (MF + TiO_2 + UV) processes, turbidity was treated little by photocatalyst adsorption, and photo-oxidation. However, treatment portions of UV_{254} absorbance by adsorption (MF + TiO_2) and photo-oxidation (MF + TiO_2 + UV) at humic acid of 4 mg/L and 6 mg/L were above 9.0, 9.5 and 8.1, 10.9%, respectively.

Keywords: *ceramic membrane, photocatalyst, hybrid process, microfiltration, water-back-flushing*

1. Introduction

Today drinking water sources have been severely polluted by various organic or inorganic matters, turbid materials and pathogens, the interest in and applications for advanced water treatment have increased in order to remove effectively those pollutants of undesirable source. Furthermore, researches of drinking water treatment by membrane separation have been pursued actively [1-5]. Application of membrane separation in the drinking water treatment not only achieves superior quality of treated water, but also requires only a compact facility without limit of installation area compared with conventional water treatment technologies. In addition, the quality of treated water is excellent and independent of fluctuation of water source condition because the membrane separation is physical treatment [6].

Recently, a major pending problem in drinking water treatment plants is to effectively remove NOM that has been known as a precursor of disinfection by-products

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Table 1. Specification of Multi-channel Ceramic Macrofiltration (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. (Korea)

Table 2. Specification of TiO_2 Coated Bead (PP) Employed in This Study

Material of bead	Polypropylene (PP)
TiO_2 coating method	Chemical vapor deposition [20]
Diameter (mm)	4～6
Weight (mg)	21.8～48.3
Average weight (mg)	39.9

(DBPs) such as trihalomethanes (THMs). However, it has been difficult to remove soluble organic materials such as NOM by only microfiltration (MF) [7]. Therefore, a large number of researches for hybrid process of membrane filtration and activated carbon (AC) adsorption have been performed to remove NOM [8-11].

Additionally, NOM is one of the major materials that cause membrane fouling in membrane separation process applied to advanced drinking water treatment. Generally, membrane fouling in drinking water treatment occurs from inorganic particles (e.g., iron, silica and suspended solids) and organic compounds (e.g., humic sub-stances, polysaccharides, proteins and micro-organisms) [12-14]. And the membrane fouling causes concentration polarization [15] and gel layer formation on membrane surface [16], and adsorption and pore blockage inside membrane pores [17].

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. It also has the advantage of preventing damage and pollution, which happen frequently in organic membranes, by microorganisms and bacillus because of inorganic materials. The ceramic membrane will be dramatically applied to the water treatment field, and play an important role in water treatment because it has a high ripple effect on industry [18,19].

The hybrid module for advanced drinking water treatment in this study was composed of a mutli-channel ceramic MF for turbidity removal and Photocatalyst adsorption for NOM removal. We investigated the effects of organic materials on membrane fouling and treatment efficiency of turbidity and UV_{254} absorbance in advanced water treatment process by this hybrid module. And treatment portions of humic acid by membrane filtration, photocatalyst adsorption, and photo-oxidation were investigated in (MF), (MF + TiO_2), (MF + TiO_2 + UV) process.

2. Experimental

2.1. Materials

Multi-channel ceramic MF membrane (HC04, Honeycomb 0.4 μm) used in the study was coated with α-alumina on supporting layer of α-alumina, and its pore size was 0.4 μm. We purchased the membrane from Dongseo Inc. in Korea, and specification of multi-channel ceramic membrane is shown in Table 1.

The photocatalyst used was 4～6 mm polypropylene beads coated with TiO_2 powder by CVD (chemical vapor deposition) method in Kim's group [20], as shown in Table 2. Instead of natural organic matters and fine inorganic particles in natural water source, a quantity of humic acid and kaolin was dissolved in distilled water. Then it was used as standard solution in our experiment. UV with 352 nm was radiated from outside of acryl module by 2 UV lamps (F8T5BLB, Sankyo, Japan).

2.2. Hybrid Membrane Module

To remove turbidity and NOM, the hybrid module was composed by packing photocatalyst (TiO_2) in space of between module inside and outside of ceramic membrane. The standard solution was treated by in and

Table 3. Effect of Humic Acid Concentration on Filtration Factors for HC04

Experimental condition		Turbidity (NTU)					
Kaolin (mg/L)	Humic acid (mg/L)	J_0 (L/m^2hr)	$R_m \times 10^{-9}$ (kg/m^2s)	$R_b \times 10^{-9}$ (kg/m^2s)	$R_{f,180} \times 10^{-9}$ (kg/m^2s)	J_{180} (L/m^2hr)	V_T (L)
30	2	413	0.568	0.115	0.513	236	15.46
	4	392	0.670	0.050	0.842	181	12.20
	6	398	0.663	0.046	1.015	164	11.50
	8	391	0.657	0.064	1.247	143	10.48
	10	378	0.683	0.064	1.422	130	9.59

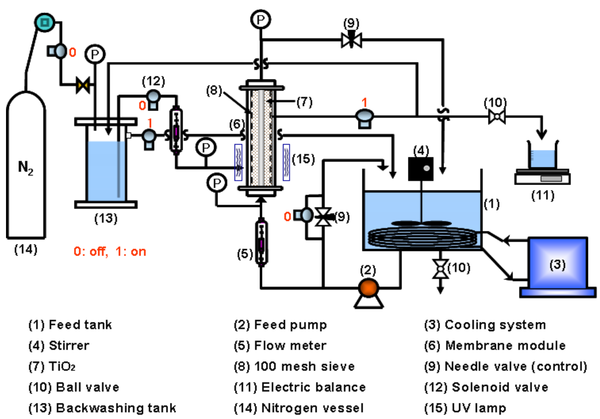

Fig. 1. Apparatus of advanced water treatment system using hybrid module of ceramic microfiltration and TiO_2 bead adsorption with periodic water-back-flushing [21].

out type ceramic membrane, and then by photocatalyst coated PP beads located at membrane outside. In addition, 100 mesh (0.150 mm), which was extremely smaller than 1～2 mm particle size of photocatalyst used here, was installed at the outlet of the hybrid module to prevent photocatalyst (TiO_2) loss to treated water tank.

2.3. Experimental Procedure

The advanced water treatment system using hybrid module (6) of multi-channel ceramic microfiltration and photocatalyst adsorption is shown in Fig. 1. We performed cross-flow filtration for the multi-channel ceramic membrane and periodic water-back-flushing using permeate water. The hybrid module (6) filled with 40 g/L of photocatalyst (TiO_2) was installed in the advanced water treatment system. Then the feed tank (1) was filled with prepared 10 L of synthetic water composed of humic acid and kaolin, and temperature of feed water was constantly maintained by using constant temperature circulator (3) (Model 1146, VWR, U.S.A). Also, the synthetic feed water was continuously mixed by stirrer (4) in order to maintain homogeneous condition of feed water, and it was flowed into the inside of multi-channel ceramic membrane by pump (2) (Procon, Standex Co., U.S.A.). Feed How rate was measured by flowmeter (5) (NP-127, Tokyo Keiso, Japan). Flow rate and pressure of feed water which flows into the hybrid module was constantly maintained by controlling valves (9) of both bypass pipe of pump (2) and concentrate pipe. Permeate flux treated by both multi-channel ceramic membrane and photocatalyst was measured by electric balance (11) (Ohaus, U.S.A.). Permeate water flowed into the back-washing tank (13) when permeate flux was not 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.

Kaolin was fixed at 30 mg/L and Humic acid was changed as 2, 4, 6, 8 and 10 mg/L in the synthetic feed water to study the effect of humic acid (HA). Also, humic acid was fixed at 4 mg/L and 6 mg/L respictively, was investigated portion of treatment efficiency of multi-channel ceramic MF, adsorption, and photo-oxidation in the hybrid process of multi-channel ceramic MF and photocatalyst. We observed the resistance of membrane fouling (R_f) and permeate flux (J) during total filtration time of 180min at each condition. At all experimental conditions, TMP was maintained at

Table 4. Water Quality and Treatment Efficiency of Turbidity in the Hybrid Process of Multi-channel Ceramic MF and Photocatalyst for Effect of Humic Acid Concentration

Experimental condition		Turbidity (NTU)				Average treatment efficiency (%)
		Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	Range	Average	Range	Average	
30	2	25.4～29.2	26.9	0.290～0.410	0.330	98.8
	4	23.2～30.6	26.3	0.318～0.575	0.444	98.3
	6	24.3～31.9	27.8	0.452～0.614	0.541	98.1
	8	25.1～32.0	28.0	0.494～0.658	0.590	97.9
	10	25.1～32.1	28.5	0.905～1.200	1.011	96.4

Table 5. Water Quality and Treatment Efficiency of UV_{254} Absorbance in the Hybrid Process of Multi-channel Ceramic MF and Photocatalyst for Effect of Humic Acid Concentration

Experimental condition		UV_{254} absorbance (cm^{-1})				Average treatment efficiency (%)
		Feed water		Treated water		
Kaolin (mg/L)	Humic acid (mg/L)	Range	Average	Range	Average	
30	2	0.023～0.070	0.039	0.006～0.008	0.007	81.4
	4	0.041～0.091	0.062	0.008～0.018	0.011	81.8
	6	0.060～0.138	0.089	0.014～0.024	0.018	79.8
	8	0.086～0.166	0.115	0.018～0.028	0.023	80.0
	10	0.115～0.190	0.152	0.023～0.039	0.032	78.9

0.81 bar, water-back-flushing pressure at 1.0 bar, feed flow rate at 1.0 L/min, and feed water temperature at 20˚C. Periodic water-back-flushing using permeate water was performed during 10 sec per filtration of 10 min.

Quality of feed water and treated water was analyzed in order to evaluate treatment efficiency of turbid materials and dissolved organic matters. Turbidity was measured by turbidimeter (2100N, HACH, U.S.A.) and UV254 absorbance was analyzed by UV spectrophotometer (GENESYS 10 UV, Thermo, U.S.A).

3. Results and Discussions

We investigated the effect of organic matters and role of microfiltration (MF), adsorption, and photo-oxidation in hybrid process of multi-channel ceramic microfiltration and photocatalyst oxidation for advanced drinking water treatment.

3.1. Effect of Organic Matters

Kaolin was fixed at 30 mg/L and humic acid was changed from 2 to 10 mg/L in synthetic feed water in the experiment in order to investigate the effect of organic matters as like humic acid. As a result, both resistance of membrane fouling (R_f), which was calculated by resistance-in-series filtration equation as same method as our previous result [21], and permeate flux (J) were highly influenced by humic acid, and R_f was dramatically dropped with decreasing concentration of humic acid from 10 mg/L to 2 mg/L as shown in Fig. 2. In particular, R_f clearly decreased when the concentration of humic acid was changed 2 mg/L. As summarized in Table 3, $R_{f,180}$ value at humic acid 2 mg/L was 0.513 × 10^9 kg/m^{2}s. This $R_{f,180}$ value should be much smaller than the $R_{f,180}$ value of 1.422 × 10^9 kg/m^{2}s at humic acid 10 mg/L.

The values of permeate flux (J) tended to be increased with decreasing the humic acid concentration as shown in Fig. 3, because reducing membrane foul-

Table 6. Roles of Microfiltration, Adsorption, and Photo-oxidation on Filtration Factors for HC04

Experimental condition		Turbidity (NTU)					
Humic acid (mg/L)	Process	J_0 (L/m^{2}hr)	$R_m \times 10^{-9}$ (kg/m^{2}s)	$R_b \times 10^{-9}$ (kg/m^{2}s)	$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)	J_{180} (L/m^{2}hr)	V_T (L)
4	MF + TiO_2 + UV	392	0.670	0.050	0.842	181	12.20
	MF + TiO_2	385	0.714	0.019	0.845	179	12.11
	MF	392	0.653	0.067	0.837	181	12.27
6	MF + TiO_2 + UV	398	0.663	0.046	1.015	164	11.50
	MF + TiO_2	391	0.664	0.058	1.042	160	10.95
	MF	374	0.663	0.092	1.052	156	10.88

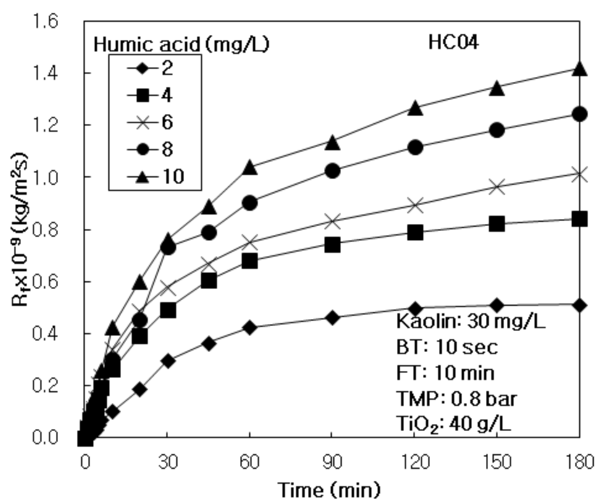

Fig. 2. Effect of humic acid concentration on resistance of membrane fouling in hybrid process of multi-channel ceramic MF and photocatalyst.

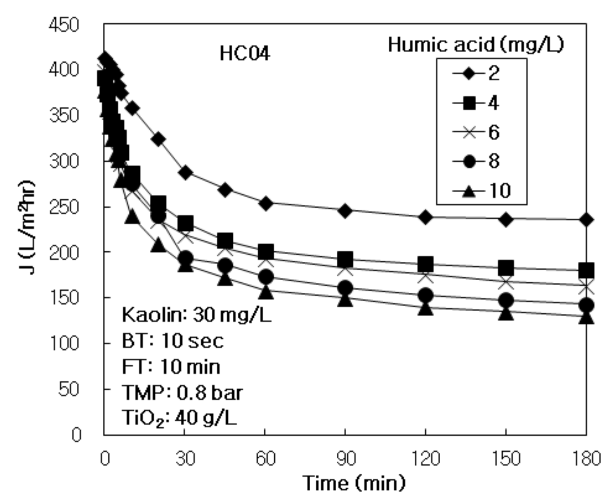

Fig. 3. Effect of humic acid concentration on permeate flux in hybrid process of multi-channel ceramic MF and photocatalyst.

ing by organic macromolecules was a rather complex combination process involving adsorption of macromolecules to the membrane surface and within membrane pores, and formation of a gel-like cake layer. Thus, the J_{180} value of 236 L/m^{2}hr at humic acid 2 mg/L was the higher than J_{180} value of 130 L/m^{2}hr at humic acid 10 mg/L.

Finally, the experimental values of $R_{f,180}$ were significantly decreased according to reduction of the humic acid concentration. It proved that NOM like humic acid could be a more important factor on membrane fouling in drinking water treatment than fine inorganic particles. Then, treatment efficiency of turbidity and UV_{254} absorbance was very outstanding at 96.4～98.8% and 78.9～81.8%, respectively, as shown in Table 4 and Table 5. The highest influence of turbidity and UV_{254} absorbance was 98.8% at humic acid 2 mg/L and 81.8% at 4 mg/L as near as 81.4% at humic acid 2 mg/L. But the lowest efficiency of that treatment was 96.4%, 78.9% at humic acid 10 mg/L.

The results of Table 4 and Table 5 could be compared with previous studies [22,23] of our laboratory. The average treatment efficiency of turbidity was almost constant in the previous study [22], but those of turbidity in this study were decreased as humic acid increased because the membrane of this study had larger pore size than that of the study [22]. But the average treatment efficiency of UV_{254} absorbance was increased corresponding to humic acid increasing in the result [22], which was opposite to those of this study.

Table 7. Water Quality and Treatment Efficiency of UV_{254} Absorbance in the Hybrid Process of Multi-channel Ceramic MF and Photocatalyst for Roles of Microfiltration, Adsorption, and Photo-oxidation at HA 4 mg/L and 6 mg/L

Experimental condition			Average treatment efficiency (%)	
Kaolin (mg/L)	Humic acid (mg/L)	Process	Turbidity	UV_{254} absorbance
30	4	MF + TiO_2 + UV	98.3	81.8
		MF + TiO_2	96.1	72.3
		MF	94.5	63.3
	6	MF + TiO_2 + UV	98.1	79.8
		MF + TiO_2	96.7	68.9
		MF	93.7	60.8

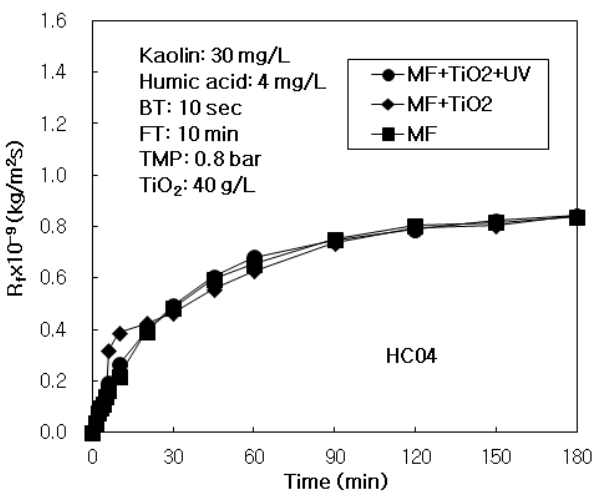

Fig. 4. Role of adsorption and photo-oxidation on resistance of membrane fouling in hybrid process of multi-channel ceramic MF and photocatalyst at humic acid 4 mg/L.

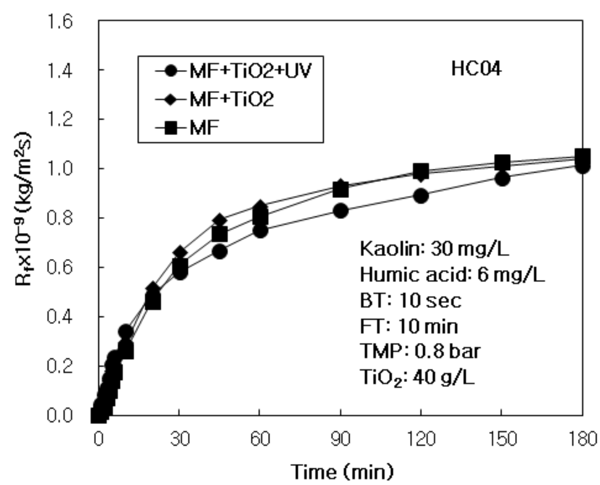

Fig. 5. Role of adsorption and photo-oxidation on resistance of membrane fouling in hybrid process of multi-channel ceramic MF and photocatalyst at humic acid 6 mg/L.

It was thought that fouling cake layer could be created the denser by humic acid in the membrane surface because it had 0.1 μm of the smaller pore size than that of this study. Therefore, the turbidity and UV_{254} absorbance of our treated water were higher than those of the study [22].

In the another study [23] of our laboratory, the average treatment efficiency of turbidity and UV_{254} absorbance was decreased with increasing of humic acid, which was a same trend of this study. Also, the treatment efficiency of UV_{254} absorbance was higher than that of this study, because the membrane in this study was honey-comb type with the larger pore size of 0.4 μm than that of the result [23].

3.2. Role of Microfiltration (MF), Adsorption, and Photo-oxidation

To investigate role of microfiltration (MF), adsorption, and photo-oxidation, experiment without UV irradiation (MF + TiO_2) and only microfiltration (MF) was performed at humic acid 4 mg/L. Then, R_f values of experiment without UV irradiation (MF + TiO_2) and only MF at humic acid 4 mg/L were arranged in Fig. 4, and compared with the previous experiment (section 3.1) with UV irradiation (MF + TiO_2 + UV). As shown in Fig. 4, curves for process of MF + TiO_2 + UV, MF + TiO_2 and for only MF were shown almost same.

After humic acid concentration was changed to 6 mg/L, the R_f values were calculated from the ex-

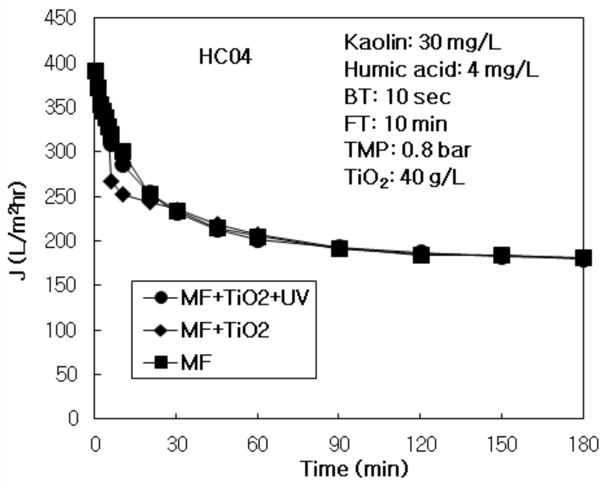

Fig. 6. Role of adsorption and photo-oxidation on permeate flux in hybrid process of multi-channel ceramic MF and photocatalyst at humic acid 4 mg/L.

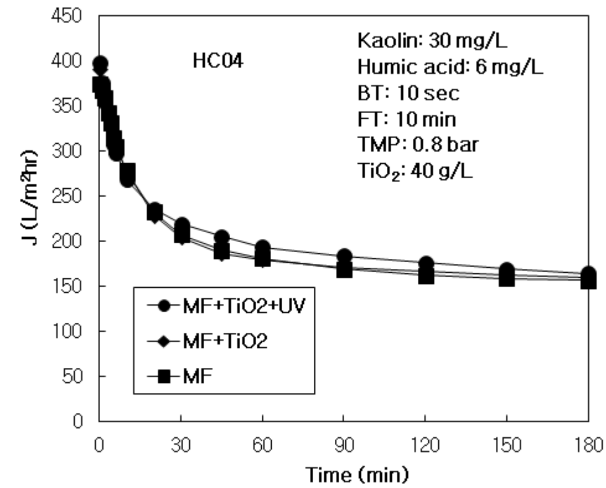

Fig. 7. Role of adsorption and photo-oxidation on permeate flux in hybrid process of multi-channel ceramic MF and photocatalyst at humic acid 6 mg/L.

perimental result of the previous MF + TiO_2 + UV process (section 3.1), MF + TiO_2 process without UV lamp and only MF process, and then compared in Fig. 5. Then, it dropped a little in MF + TiO_2 process packing TiO_2 coated beads in the module, and the hybrid process of MF and photocatalyst (MF + TiO_2 + UV) showed the lowest membrane fouling during 180 minutes's operation. This result proved that photocatalyst adsorption and photocatalyst-oxidation by UV radiation could control membrane fouling in hybrid system. However, the membrane fouling at 6 mg/L of humic acid was much clear dependent on each process compared with it of 4 mg/L.

As summarized in Table 6, $R_{f,180}$ at humic acid 4 mg/L was almost same in processes of MF + TiO_2 + UV, MF + TiO_2 and only MF. But the $R_{f,180}$ at humic acid 6 mg/L dropped a little as adding photocatalyst adsorption and photocatalyst-oxidation, which the lowest value was 1.015 kg/m^{2}s for hybird process of MF and photocatalyst (MF + TiO_2 + UV). It means that photocatalyst adsorption and photo-oxidation could reduce membrane fouling at the higher humic acid concentration the more efficiently. Furthermore, the permeate flux (J_{180}) and V_T showed the largest value at MF + TiO_2 + UV process. Finally, this result means that the highest permeate flux could be maintained during operation because the hybrid process of MF and photocatalyst controled membrane fouling effectively, and finally the largest V_T was obtained in our hybrid process.

Also, permeate flux (J) in process of MF, MF + TiO_2, and MF + TiO_2 + UV at humic acid 4 mg/L and 6 mg/L were arranged in Fig. 6 and Fig. 7. The J at humic acid 4 mg/L was almost same in each process, but J at 6 mg/L increased as adding photocatalyst adsorption and photocatalystoxidation. After 180 min's operation the highest J_{180} at humic acid 6 mg/L was 164 L/m^{2}hr in process of MF + TiO_2 + UV, as shown in Table 6. This result proved that photocatalyst adsorption and photocatalyst-oxidation by UV radiation could control memrbane fouling in our hybrid process.

Turbidity and UV_{254} absorbance of feed and treated water at 6 mg/L of humic acid were arranged for each process in Table 7. Turbidity treatment efficiency was 93.7% for only MF, 96.7% for MF + TiO_2 process added photocatalyst adsorption, and 98.1% for MF + TiO_2 + UV process added photo-oxidation. However, treatment efficiency of UV_{254} absorbance, which meaned organic matters, was 60.8% for only MF, 68.9% for MF + TiO_2, and 79.8% for MF + TiO_2 + UV process.

Table 8. Portion of Treatment Efficiency of Multi-channel Ceramic MF, Adsorption, and Photo-oxidation in the Hybrid Process of Multi-channel Ceramic MF and Photocatalyst

Humic acid concentration (mg/L)	4		6	
Portion of treatment Efficiency	Turbidity	UV_{254} absorbance	Turbidity	UV_{254} absorbance
Membrane filtration (%)	94.5	63.3	93.7	60.8
Adsorption (%)	1.6	9.0	3.0	8.1
Photo-oxidation (%)	2.2	9.5	1.4	10.9
Total treatment efficiency (%)	98.3	81.8	98.1	79.8

The treatment efficiency of UV_{254} absorbance increased rapidly compared with that of turbidity. It proved that photocatalyst adsorption and photocatalyst-oxidation could be more excellent for removal of organic matters than suspended particles as turbidity.

The treatment portion of MF, adsorption, and photo-oxidation in our process was calculated from the treatment efficiency of turbidity and organic matters as shown in Table 7, and then arranged in Table 8. In turbidity treatment efficiency the portions of MF were very high as 94.5 and 93.7% at humic acid 4 mg/L and 6 mg/L, respectively. And adsorption was very small portion of turbidity, and those of photo-oxidation were very low under 2.2%. However, in treatment efficiency of humic acid the portions of adsorption was high as 9.0 and 8.1% at humic acid 4 mg/L and 6 mg/L, respectively. Those of photo-oxidation in himic acid treatment were very important as 9.5 and 10.9%, compared with turbidity. Therefore, it proved that adsorption and photocatalyst-oxidation of TiO_2 photocatalyst could be more important in the removal of organic matters than inorganic matters in our hybrid process of MF and photocatalyst.

4. Conclusions

We investigated the effect of organic materials role of microfiltration (MF), adsorption, and photo-oxidation in advanced water treatment by hybrid process of multi-channel ceramic MF and photocatalyst. The important conclusions are summarized as follows:

1) Both R_f and J were highly dependent on the concentration of humic acid in the synthetic water. Therefore, Organic matters like humic acid could be one of the main factors on membrane fouling in drinking water treatment.

2) As results of the efficiency of turbidity and UV_{254} in the process of MF only, MF + TiO_2, and MF with photocatalyst and UV radiation (MF + TiO_2 + UV), the highest values of treatment efficiency was observed in process of MF + TiO_2 + UV. It means that photocatalyst-oxidation is more important in the removal of organic matters than inorganic matters in our hybrid process.

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