The Role of Humic Acid, PP Beads, and pH with Water Backwashing in a Hybrid Water Treatment of Multichannel Alumina Microfiltration and PP Beads

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Abstract: Photooxidation oxidizes most organic compounds by mineralizing them to small inorganic molecules. In this study, the effects of dissolved organic matter (DOM), pH, and polypropylene (PP) beads concentration on membrane fouling were investigated in a hybrid water treatment process consisting of seven-channel alumina microfiltration (pore size 1.0 μm) and pure PP beads water backwashing with UV irradiation for photooxidation. The synthetic feed was prepared with humic acid and kaolin and flowed inside the microfiltration (MF) membrane. The permeate contacted the PP beads fluidized in the gap of the membrane and module with outside UV irradiation. Membrane fouling resistance (R_f) increased dramatically with an increase in the concentration of humic acid (HA) from 6 mg/L to 8 mg/L. The treatment efficiency of DOM increased dramatically, from 14.3% to 49.7%, with an increase in the concentration of HA. The R_f decreased with an increase of PP beads concentration. However, maximum permeate volume (V_T) was acquired at 5 g/L of PP beads. The maximal treatment efficiency of DOM was 51.3% at 40 g/L of PP beads. The R_f increased with an increase in the pH of feed, and the maximum V_T was acquired at a pH of 5. The maximal treatment efficiency of DOM was 52.5% at pH 9.

Keywords: microfiltration; photooxidation; adsorption; polypropylene bead; hybrid treatment; water treatment; alumina

1. Introduction

The membrane separation process for separation and purification has developed dramatically during the past few decades. It can separate and concentrate all pollutants simultaneously in water by the retention of its microspores without secondary pollution and phase change. Additionally, its equipment is compact, easy to operate, and capable of continuous operation at room temperature with the advantage of low energy consumption [1]. However, membrane fouling due to the adsorption-precipitation of organic and inorganic compounds onto membranes leads to a decrease in the permeate flux, which increases membrane cleaning costs and reduces membrane life. Techniques for controlling membrane fouling remain inadequate, which is the major obstacle in the successful implementation of membrane separation technology, although considerable progress has been made in membrane fouling [2,3]. Natural organic matter (NOM) is a primary component of fouling in low-pressure membrane filtration. Various preventive measures to interfere with NOM fouling have been developed and extensively tested, such as coagulation, oxidation, ion exchange, carbon adsorption, and mineral oxide adsorption [4]. In this study, periodic water backwashing was performed to prevent the membrane fouling.

Recently, the hybrid technology of membrane separation and photooxidation by UV irradiation has effectively solved the above-mentioned membrane fouling problem [5]. Hybrid technology not only

maintains the advantages of each technology, but also produces synergistic effects that overcome the limits of only one technology of membrane separation and photooxidation. In addition, pollutants such as NOM can be oxidized by UV irradiation, and organic species are rejected partially by controlling the residence time in the reacting system. In other words, the membrane is also a selective barrier for the separated molecules. Finally, hybrid technology increases photooxidation efficiency and acquires excellent effluent quality. The effect of UV-irradiation on the nano-hybrid PES-NanoZnO membrane in terms of flux and rejection efficiency has also been discussed in [6]. In addition, a comparison of the efficiency of surface water treatment in hybrid systems by coupling various advanced oxidation processes and ultrafiltration (UF) was presented by [7]. In this study, a hybrid process of ceramic membrane and pure PP beads with UV irradiation was applied for advanced water treatment.

The ceramic membranes, used in this study, have numerous advantages, which are mechanical, thermal, and chemical resistance, as well as a long lifetime. These are economically competitive as compared with polymeric membranes, because of an almost permanent lifetime [8]. Nowadays, ceramic membranes have been widely applied in water or wastewater throughout the world [9,10]. The influence of the soluble algal organic matter (AOM) characteristic on the fouling of a seven-channel tubular ceramic (MF) membrane has been investigated at lab scale by [11]. The influence of the interaction between aquatic humic substances and the algal organic AOM derived from Microcystis aeruginosa on the fouling of a ceramic MF membrane has been studied by [12].

Photooxidation has the characteristics of high efficiency, low energy consumption, and a wide range of application, by mineralizing organic compounds to small inorganic molecules and oxidization of most them, especially nonbiodegradable organic contaminants. In addition, it is an excellent technology for water pollution control. For this reason, the photooxidation technology applied in this study has been applied broadly [13–18]. In addition, degradation of humic acid (HA), which was contained in a synthetic solution used in this study, via the photoelectrocatalysis (PEC) process and the corresponding disinfection byproduct formation potential (DBPFP) were investigated; the PEC process was found to be effective in reducing dissolved organic carbon concentration [19].

In our research group, on the one hand, the results for the effect of operating conditions on a hybrid water treatment process of various ceramic membranes and titanium dioxide (TiO$_2$) photocatalyst-coated polypropylene (PP) were published in "Desalination and Water Treatment" [20] and "Membr. J." [21]. On the other hand, the roles of adsorption and photooxidation in a hybrid water treatment process of tubular carbon fiber UF and pure PP beads with UV irradiation and water backwashing were reported by our group in "Desalination and Water Treatment" [22].

In this study, the roles of humic acid, pure PP beads concentration, and pH were investigated on membrane fouling in a hybrid water treatment process of seven-channel alumina MF membranes and pure PP beads with water backwashing and UV irradiation. This research involved the unique application of pure PP beads to investigate the effect of HA, pure PP beads, and pH on the hybrid process of multichannel MF membrane as compared with our previous results [20–22]. Periodic water backwashing was performed for 10 s per 10 min of filtration to reduce membrane fouling. A hybrid module was composed of the MF membranes and the PP beads, which were fluidized between the gap of carbon fiber membrane and the acryl module case. The results of humic acid, pure PP beads concentration, and pH effects were compared with those of the previous study [22,23] for the hybrid process of the tubular carbon fiber UF (pore size 0.05 μm) membranes and pure PP beads with water backwashing, to investigate the effects on membrane fouling and treatment efficiency depending on membrane module types and materials.

2. Materials and Methods

The seven-channel alumina MF (HC10) membranes, used in the study, were manufactured in Dongseo Industry (Seoul, Korea), with pore sizes of 0.10 μm. The specifications of the HC10 alumina membrane are listed in Table 1. In this study, 4 to 6 mm size polypropylene (PP) beads were applied and the average weight was 39.9 mg. A quantity of HA and kaolin was dissolved in distilled water, instead
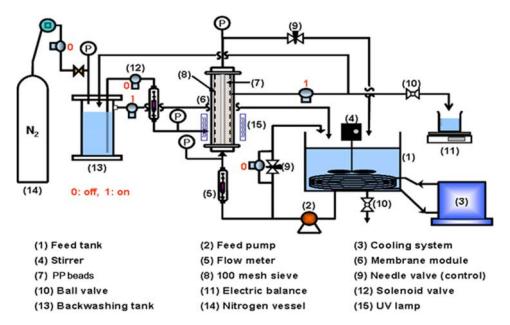
of natural organic matter and fine inorganic particles in a natural water source. In this experiment, it was utilized as synthetic feed water. For photooxidation, two UV lamps (F8T5BLB, Sankyo, Tokyo, Japan) irradiated UV with 352 nm from outside of the acryl module.

Table 1. Specifications of multichannel ceramic microfiltration (MF) membrane used in this study.

Membrane Model	HC10
Pore size (μm)	0.1
No. of channels	7
Outer diameter (mm)	20
Inner diameter (mm)	4
Length (mm)	245
Surface area (cm^2)	215
Material	α-alumina
Company	Dongseo Industry (Korea)

To remove the turbidity and DOM, the hybrid module was packed with pure PP beads in the space between the module inside and outside of the alumina membrane. Additionally, a 100 mesh (0.150 mm) sieve, which was much smaller than 4 to 6 mm particle size of the PP beads used here, was installed at the outlet of the hybrid module to contain the PP beads in the module.

The hybrid water treatment process (6) of multichannel MF membrane and the pure PP beads (7), which were used in the previous study [24], are shown in Figure 1 and Scheme 1. A periodic water backwashing using permeated water was performed for the multichannel MF membrane. The hybrid module (6) was supplied with PP beads fluidizing between the gap of carbon fiber membrane and the acryl module case. Then, 10 L of the synthetic water with HA and kaolin was contained in the feed tank (1). To maintain a constant viscosity of water, the temperature of the feed water was constantly maintained using a temperature circulator (3) (Model 1146, VWR, Atlanta, GA, USA). To maintain the homogeneous condition of the feed water, the synthetic feed water was continuously mixed by a stirrer (4), and it flowed into the inside of the MF membrane by a pump (2) (Procon, Standex Co., Smyna, TN, USA). A flowmeter (5) (NP-127, Tokyo Keiso, Tokyo, Japan) measured the feed flow rate. Controlling valves (9) of both the bypass pipe of the pump (2) and the concentrate pipe maintained constant flow rate and pressure of the feed water that flowed into the hybrid module. An electric balance (11) (Ohaus, Newark, NJ, USA.) measured the permeate flux treated by both the MF membrane and the PP beads. During the permeate, flux had not been measured and the permeate water flowed into the backwashing tank (13). To maintain a constant concentration of the feed water during operation, it was recycled to the feed tank (1) after the treated water was over a certain level in the backwashing tank (13). Physical washing was performed by a brush inside the tubular membrane after each operation, and the permeate flux was measured to calculate the resistances of irreversible and reversible membrane fouling.

(1) Feed tank   (2) Feed pump   (3) Cooling system
(4) Stirrer   (5) Flow meter   (6) Membrane module
(7) PP beads   (8) 100 mesh sieve   (9) Needle valve (control)
(10) Ball valve   (11) Electric balance   (12) Solenoid valve
(13) Backwashing tank   (14) Nitrogen vessel   (15) UV lamp

Figure 1. Apparatus of hybrid water treatment process of tubular carbon fiber ultrafiltration (UF)and microfiltration (MF) membrane and PP beads with periodic water backwashing [24].

(**a**)   (**b**)

Scheme 1. Hybrid water treatment process system of tubular carbon fiber UF/MF membrane and PP beads with periodic water backwashing: (**a**) Total hybrid system and (**b**) hybrid module and UV lamp.

HA was changed from 2 mg/L to 10 mg/L, and kaolin was fixed at 30 mg/L in the experiment of HA effect. To investigate the roles of PP beads concentration and pH, kaolin and HA were fixed at 30 mg/L and 10 mg/L, respectively, in the synthetic feed water. In addition, the water backwashing time (BT) and filtration time (FT) were fixed at 10 s and 10 min, individually. During the total operation time of 180 min, the permeate flux (J) were measured at each condition. Transmembrane pressure (TMP) was maintained constant at 1.8 bar, the water backwashing pressure at 2.5 bar, the feed flow rate at 1.0 L/min, and the feed water temperature at 20 °C, in all experiments. The PP beads concentration

was changed from 0 to 50 g/L in the module, to investigate the effect of PP beads and to investigate the pH effect, the pH was changed from 5 to 9 in the synthetic feed water.

To evaluate the treatment efficiencies of turbid materials and DOM, the quality of feed and treated water was analyzed for each experiment. To measure turbid materials and DOM, turbidity was measured using a turbidimeter (2100N, Hach, Ames, IA, USA) and UV_{254} absorbance was analyzed using a UV spectrophotometer (Genesys 10 UV, Thermo, Pittsburgh, PA, USA). The detection limits of turbidimeter and UV spectrophotometer were 0 to 4000 NTU (±0.001 NTU) and −0.1 to 3.0 cm^{-1} (±0.001 cm^{-1}) correspondingly. Before measuring UV_{254} absorbance, each sample was filtered using a 0.2 μm syringe filter to remove turbid materials.
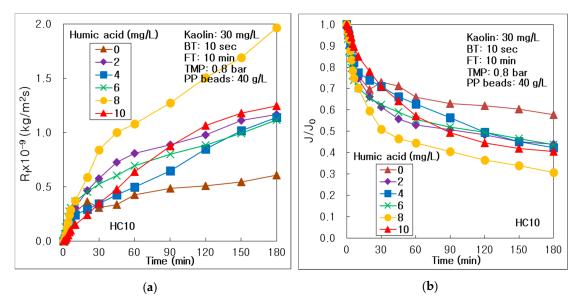
To clean the ceramic membrane and apparatus after each experiment was finished, all of the synthetic solution was discharged from the hybrid water treatment system, and distilled water was circulated through the line of the system for 15 min. To combust fouling materials inside the membrane, the PP beads and the membrane were separated from the module, and the membrane was ignited at 550 °C in a furnace for 30 min. To wash out organic or inorganic pollutants after cooling the membrane, the membrane was immersed in a nitric acid (HNO_3) of 15% for 24 h, and in a sodium hydroxide (NaOH) solution of 0.25 N for 3 h, and kept in distilled water for 24 h for rinsing. Before beginning a new experiment, the water permeated flux (J_w) was measured to check the membrane recovery when a normal operation was performed with distilled water, after the recovered membrane was installed inside the module. The recovered membrane was used in all of the experiments to reduce the influence of membrane condition on the treatment efficiency.

3. Results and Discussions

The effects of HA, pure PP beads concentration, and pH were investigated in the hybrid water treatment process of seven-channel alumina MF (HC10) membrane and pure PP beads with periodic water backwashing and UV irradiation. Membrane resistance, boundary layer, and membrane fouling (R_m, R_b, and R_f) were calculated from permeate flux (J) data using the resistance-in-series filtration equation ($J = \Delta P/(R_m + R_b + R_f)$) which was the same method as the previous study [24], where ΔP is the transmembrane pressure. For a new membrane, the equation was simplified to $J = \Delta P/R_m$ because there were no boundary layer resistances, and membrane fouling and R_m could be determined using J data for a new membrane. For the synthetic water of HA and kaolin, the equation was modified to $J = \Delta P/(R_m + R_b)$ at initial time, and R_b could be determined using J_0 and R_m data. Irreversible and reversible membrane fouling resistances (R_{if}, R_{rf}) could be found from J data after physical washing using a brush inside the membrane.

3.1. Effect of HA Concentration on Membrane Fouling and Treatment Efficiency

The membrane fouling resistance (R_f) was significantly influenced by humic acid (HA), which was one of the major natural organic matters (NOM) in lakes or rivers, and R_f was the minimum at HA 0 g/L, and almost constant at 2 to 6 mg/L of HA; however, it increased dramatically with an increase in the concentration of HA from 6 mg/L to 8 mg/L, and then, decreased suddenly in a concentration of HA from 8 mg/L to 10 mg/L , as shown in Figure 2a. This means that DOM, similar to HA, could drive membrane fouling more severely on the surface and inside the alumina membrane, with an increase of HA concentration from 6 mg/L to 8 mg/L in water. However, the membrane fouling decreased from 8 mg/L to 10 mg/L of HA, because the thick fouling cake on the membrane could be removed by water backwashing at 10 mg/L of HA. As summarized in Table 2, the membrane resistance (R_m) was controlled at a constant value by combustion in a furnace and washing with an acid and alkali solution. The final R_f ($R_{f,180}$) value after operating for 180 min at HA 8 mg/L was 1.963×10^9 kg/m^2s, which was 3.24 times higher than 0.606×10^9 kg/m^2s of the $R_{f,180}$ value at HA 0 mg/L.

Figure 2. Effect of humic acid concentration in the hybrid process of multichannel alumina MF and PP beads with UV irradiation and periodic water backwashing: (**a**) Resistance of membrane fouling and (**b**) dimensionless permeate flux.

In the previous study [22] for the hybrid water treatment process of the same tubular carbon fiber UF and the same pure PP beads, R_f increased dramatically with an increase in the concentration of HA from 2 mg/L to 10 mg/L. The increasing rate of R_f was very high, when HA increased from 6 mg/L to 10 mg/L, in particular. This means that DOM, similar to HA, could drive membrane fouling more severely on the surface and inside the carbon fiber membrane, with an increase of HA concentration in water, and specifically at a high HA concentration. There was a little different trend as compared with the study of HC10, depending on the membrane type and material. The $R_{f,180}$ value after operating for 180 min at HA 10 mg/L was 6.998×10^9 kg/m^{2}s, which was 3.94 times higher than 1.775×10^9 kg/m^{2}s of the $R_{f,180}$ value at HA 2 mg/L. The $R_{f,180}$ value was much higher than those of this result of HC10 in this study, meaning that membrane fouling could be much less developed in the seven-channel HC10 membrane as compared with the tubular C005 membrane.

As shown in Figure 2b, the dimensionless permeate flux (J/J_0), where J_0 was the initial permeate flux predicted using the initial two data by an extrapolation method, was plotted according to HA concentration. The J/J_0 overlapped in the range of HA 2 to 6 mg/L, and it showed the highest values at HA 0 mg/L and the lowest at HA 8 mg/L during 180 min; however, it was much higher at HA 10 mg/L. This means that the permeate flux could be maintained low at HA 8 mg/L and increased at HA 10 mg/L, because the membrane fouling was developed severely by DOM, and the cake layer could be removed at HA 10 mg/L by water backwashing.

As arranged in Table 2, the final J/J_0 after operating for 180 min (J_{180}/J_0) was 0.578 at HA 0 mg/L, which was 1.88 times higher than 0.307 at HA 8 mg/L. In the previous work [22] of the C005, J/J_0 tended to decrease with an increase in the HA concentration from 2 to 10 mg/L, specifically from 6 to 8 mg/L, because of the membrane fouling development by the more DOM. The J_{180}/J_0 was 0.241 at HA 2 mg/L, which was 2.84 times higher than 0.085 at HA 10 mg/L. This means that the permeate flux decline rate was affected more severely by HA concentration in the hybrid process of C005 than HC10, because the membrane fouling increased dramatically with an increase in HA concentration.

In addition, the permeate volume (V_T) of 14.18 L at HA 0 mg/L was 1.57 times higher than 9.05 L of V_T at HA 8 mg/L, as shown in Table 2, because high flux was maintained at HA 4 mg/L during 180 min as compared in Figure 2b. In [22], the result of the tubular carbon fiber C005, V_T at HA 2 mg/L was 4.98 L, which was 2.59 times higher than 1.92 L of V_T at HA 10 mg/L. The difference rate of V_T was

higher in the hybrid process of C005 than HC10. This means that DOM affected membrane fouling more at tubular UF C005 membrane than multichannel MF HC10.

As listed in Table 3, the treatment efficiency of turbidity showed an increasing trend, with an increase of HA concentration. Furthermore, in the result by [22] with C005, the treatment efficiency of turbidity was almost constant. This means that the DOM could affect the treatment of suspended particles, such as kaolin, in the hybrid process of the multichannel alumina MF and the pure PP beads, because the thick membrane fouling layer rejected kaolin particles at a high HA concentration. However, DOM could not affect the process of tubular carbon fiber UF membrane, because of the tubular membrane type.

Table 2. Effect of humic acid (HA) on filtration factors in the hybrid process of seven-channel alumina MF (HC10) membrane and PP beads with periodic water backwashing (BT 10 s, FT 10 min) as compared with the results of tubular carbon fiber UF (C005) membrane.

Membrane	Humic Acid (mg/L)	0	2	4	6	8	10
7 channels alumina MF (HC10)	$R_m \times 10^{-9}$ (kg/m²s)	0.823	0.840	0.803	0.864	0.831	0.798
	$R_b \times 10^{-9}$ (kg/m²s)	0.006	0.072	0.030	0.001	0.038	0.053
	$R_{f,180} \times 10^{-9}$ (kg/m²s)	0.606	1.165	1.137	1.113	1.963	1.246
	$R_{if} \times 10^{-9}$ (kg/m²s)	0.432	0.044	0.228	0.408	0.453	1.037
	$R_{rf} \times 10^{-9}$ (kg/m²s)	0.173	1.121	0.909	0.705	1.510	0.209
	J_0 (L/m²h)	341	310	339	326	325	332
	J_{180} (L/m²h)	197	136	143	143	100	135
	J_{180}/J_0	0.578	0.439	0.423	0.437	0.307	0.406
	V_T (L)	14.18	10.59	12.48	11.30	9.05	9.39
Tubular carbon fiber UF (C005) [22]	$R_m \times 10^{-9}$ (kg/m²s)	-	0.429	0.397	0.418	0.418	0.415
	$R_b \times 10^{-9}$ (kg/m²s)	-	0.133	0.185	0.174	0.174	0.237
	$R_{f,180} \times 10^{-9}$ (kg/m²s)	-	1.775	2.135	2.676	5.311	6.998
	$R_{if} \times 10^{-9}$ (kg/m²s)	-	0.389	0.016	0.215	0.194	0.768
	$R_{rf} \times 10^{-9}$ (kg/m²s)	-	1.386	2.119	2.461	5.118	6.230
	J_0 (L/m²h)	-	1129	1092	1073	1072	974
	J_{180} (L/m²h)	-	272	234	194	108	83
	J_{180}/J_0	-	0.241	0.214	0.181	0.100	0.085
	V_T (L)	-	4.98	4.89	3.83	2.67	1.92

Table 3. Water quality and treatment efficiency of turbidity in the hybrid process of 7-channel alumina MF (HC10) and PP beads for the effect of humic acid concentration as compared with results of tubular carbon fiber UF (C005) membrane.

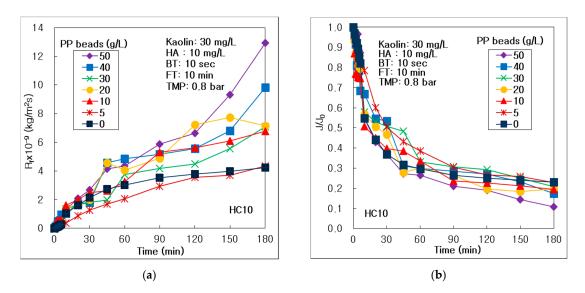| Kaolin (mg/L)	Humic Acid (mg/L)	Turbidity (NTU)				Average Treatment Efficiency (%)	
		Feed Water		Treated Water		Adsorption Media	
		Range	Average	Range	Average	7 Channels Alumina MF	Tubular Carbon Fiber UF [22]
	0	12.1–13.5	12.6	0.342–0.396	0.371	97.0	-
	2	12.1–16.3	13.6	0.370–0.446	0.396	97.1	99.1
	4	18.2–24.5	20.9	0.312–0.549	0.397	98.1	98.0
30	6	13.2–20.2	15.6	0.187–0.469	0.260	98.3	98.9
	8	23.9–37.2	31.1	0.324–0.534	0.415	98.7	98.0
	10	24.5–32.4	29.4	0.391–0.557	0.457	98.4	99.0

As shown in Table 4, the treatment efficiency of UV_{254} absorbance, which means the concentration of DOM, increased with an increase in HA concentration, and finally showed the maximum 49.7% at HA 10 mg/L. This means that DOM could be treated more effectively at a high DOM condition in the hybrid water treatment process of seven-channel alumina MF and pure PP beads. The maximum treatment efficiency of DOM was 69.3% at HA 6 mg/L, in the previous work [22] for the hybrid process of the tubular carbon fiber MF membrane and the same PP beads. From 2 mg/L to 6 mg/L of HA, most of DOM could be adsorbed on fouling materials inside the membrane or retained by cake layer

on the membrane, and the remained things passed through the membrane adsorbed or oxidized by the PP beads and UV. It happened that the treated water quality of HA increased less slowly than the feed water quality. However, above 6 mg/L of HA, most of DOM passed through the membrane and could not be treated by adsorption or photooxidation by the PP beads and UV. And the treated water quality of HA increased more rapidly than the feed water quality.

Table 4. Water quality and treatment efficiency of dissolved organic matter (DOM) (UV_{254} absorbance) in the hybrid process of 7-channel alumina MF (HC10) and PP beads for the effect of humic acid concentration as compared with results of tubular carbon fiber UF (C005) membrane.

Kaolin (mg/L)	Humic Acid (mg/L)	UV_{254} Absorbance (cm^{-1})				Average Treatment Efficiency (%)	
		Feed Water		Treated Water		Adsorption Media	
		Range	Average	Range	Average	7 Channels Alumina MF	Tubular Carbon Fiber UF [22]
30	0	0.003–0.005	0.004	0.002–0.004	0.003	14.3	-
	2	0.147–0.176	0.161	0.110–0.140	0.131	18.6	67.2
	4	0.132–0.241	0.164	0.121–0.147	0.132	19.5	65.9
	6	0.164–0.211	0.186	0.131–0.151	0.145	22.0	69.3
	8	0.184–0.370	0.244	0.135–0.152	0.146	40.3	59.3
	10	0.203–0.302	0.247	0.110–0.142	0.124	49.7	60.9

3.2. Effect of Pure PP Beads on Membrane Fouling and Treatment Efficiency

In this study, the pure PP beads were induced for adsorption of DOM and turbid matters. The effect of pure PP beads concentration was investigated at the most severe HA 10 mg/L condition, at kaolin 30 mg/L, and pH 7. The membrane fouling resistances (R_f) showed the highest at 50 g/L and the lowest at 5 g/L during the 180 min operation, as shown in Figure 3a. This means that the optimal PP beads concentration could be 5 g/L to control the membrane fouling and high permeate flux in this hybrid process of 7-channel alumina MF HC10 and PP beads.

(a)　　　　　　　　　　　　(b)

Figure 3. Effect of PP beads concentration in the hybrid process of multichannel alumina MF and PP beads with UV irradiation and periodic water backwashing: (**a**) Resistance of membrane fouling and (**b**) dimensionless permeate flux.

In the previous work by [23] for the hybrid process of the tubular carbon fiber UF C005 and the same PP beads, the R_f showed the highest at 50 g/L during the 180 min operation, and the lowest at 5 g/L until 120 min, and at 0 g/L of PP beads after 120 min. This result almost agreed with the trend of PP beads effect in this study and showed that the effect of PP beads concentration on membrane

fouling could not depend on the membrane type and materials in this hybrid water treatment process of ceramic membrane and pure PP beads.

In the hybrid process of ceramic membrane and PP beads, the boundary layer resistance (R_b), which was produced by concentration polarization on the membrane surface, was the lowest at 50 g/L of PP beads independent of membrane type and materials, as shown in Table 5. This means that the frequent colliding of more PP beads on the membrane surface could reduce the concentration polarization for both HC10 and C005. The $R_{f,180}$ after 180 min was the highest, i.e., 12.94×10^9 kg/m^{2}s, at 50 g/L, which was 3.06 times higher than 4.23×10^9 kg/m^{2}s for the $R_{f,180}$ at 0 g/L of PP beads. The R_{rf} showed an increasing trend, with an increasing PP beads concentration from 0 g/L to 50 g/L; however, the minimum R_{if} was at 40 g/L and the maximum was at 0 g/L of PP beads. This means that the reversible membrane fouling could be inhibited at 50 g/L of PP beads, because the optimal amount of PP beads captured the turbid or organic materials by adsorption.

In the previous result by [23] for the hybrid process of the C005 and the pure PP beads, the $R_{f,180}$ after 180 min was the highest of 4.892×10^9 kg/m^{2}s at 50 g/L, which was 1.48 times higher than 3.306×10^9 kg/m^{2}s of the $R_{f,180}$ at 0 g/L of PP beads. These $R_{f,180}$ were much higher than those of HC10 in this study; however, the trends depending on PP beads concentration was exactly the same in this hybrid process. Additionally, the R_{rf} increased with an increasing PP beads concentration from 0 g/L to 50 g/L of PP beads; however, the R_{if} was the minimum at 50 g/L and the maximum at 10 g/L of PP beads. This result was also almost in agreement with that of this study using HC10.

The J/J_0 was compared to investigate the effect of PP beads on the relative decline of permeate flux, as shown in Figure 3b. The J/J_0 maintained higher until 90 min at PP beads 5 g/L than those at other PP beads concentration, and showed the lowest at PP beads 50 g/L after 60 min. As arranged in Table 5, the J_0 and J_{180} decreased to 40 g/L and to 50 g/L, respectively, with an increase in PP beads concentration, because the R_b and R_f increased to 40 g/L and to 50 g/L of PP beads, correspondingly. Finally, the J_{180}/J_0 after 180 min of operation at 0 g/L of the PP beads was the maximum 0.230, which was 2.13 times higher than 0.108 at 50 g/L. However, the V_T was the highest, 11.75 L at 5 g/L of PP beads, because J maintained higher all through the operation than those of other PP beads conditions.

In the previous work by [23] for the hybrid process of the C005 and the pure PP beads, the J/J_0 showed higher during the 5 to 90 min at 5 g/L than those at other PP beads concentration. The J_0 decreased and J_{180} increased as increasing PP beads concentration, because the R_b was the minimum at 50 g/L of PP beads. In addition, the J_{180}/J_0 after 180 min of operation at 0 g/L of the PP beads was the maximum 0.126, which was 1.62 times higher than 0.078 at 50 g/L. The increasing rate, 1.62 of J_{180}/J_0 for C005, was much lower than 2.13 for HC10, because PP beads controlled more effectively the membrane fouling in the hybrid process of HC10 than C005. Also, the V_T was the highest 4.99 L at 5 g/L of PP beads for the C005 process, which was the exact same trend of HC10 in this study.

As arranged in Table 6, the treatment efficiencies of turbidity were almost constant in the range of 97.5% and 98.9% in the hybrid process of HC10 MF, independent of the pure PP beads concentration. This means that the tubid matters could be treated effectively, independent of PP beads concentration in this hybrid process. In the previous result by [23] for the hybrid process of the C005 and the pure PP beads, the treatment efficiencies of turbidity were the highest, i.e., 99.3% at 30 g/L. This means that the optimal PP beads concentration could be 30 g/L to treat the turbid matter in the hybrid process of C005 UF membrane.

Table 5. Effect of PP beads concentration on filtration factors in the hybrid process of 7-channel alumina MF (HC10) membrane and PP beads with periodic water backwashing (BT 10 s, FT 10 min) as compared with the results of tubular carbon fiber UF (C005) membrane.

Membrane	PP Beads (g/L)	0	5	10	20	30	40	50
7-channel alumina MF (HC10)	$R_m \times 10^{-9}$ (kg/m^{2}s)	1.24	1.27	1.53	1.75	1.82	1.93	1.49
	$R_b \times 10^{-9}$ (kg/m^{2}s)	0.020	0.024	0.106	0.009	0.041	0.142	0.076
	$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)	4.23	4.35	6.79	7.15	7.09	9.83	12.94
	$R_{if} \times 10^{-9}$ (kg/m^{2}s)	1.75	1.24	1.20	3.24	0.66	0.43	1.44
	$R_{rf} \times 10^{-9}$ (kg/m^{2}s)	2.486	3.110	5.587	3.910	6.423	9.403	11.500
	J_0 (L/m^{2}h)	503	491	388	361	342	307	405
	J_{180} (L/m^{2}h)	116	113	75	71	71	53	44
	J_{180}/J_0	0.230	0.229	0.194	0.198	0.208	0.174	0.108
	V_T (L)	10.11	11.75	7.56	6.94	7.91	6.55	6.85
Tubular carbon fiber UF (C005) [23]	$R_m \times 10^{-9}$ (kg/m^{2}s)	0.413	0.409	0.409	0.411	0.403	0.405	0.403
	$R_b \times 10^{-9}$ (kg/m^{2}s)	0.062	0.043	0.119	0.031	0.042	0.048	0.009
	$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)	3.306	3.596	3.683	4.767	4.967	4.918	4.892
	$R_{if} \times 10^{-9}$ (kg/m^{2}s)	0.008	0.004	0.021	0.006	0.015	0.020	0.002
	$R_{rf} \times 10^{-9}$ (kg/m^{2}s)	3.298	3.596	3.662	4.761	4.953	4.898	4.890
	J_0 (L/m^{2}h)	1336	1407	1221	1435	1426	1401	1543
	J_{180} (L/m^{2}h)	168	157	151	122	117	118	120
	J_{180}/J_0	0.126	0.112	0.124	0.085	0.082	0.084	0.078
	V_T (L)	4.13	4.99	3.54	3.35	2.73	3.16	3.29

Table 6. Water quality and treatment efficiency of turbidity in the hybrid process of 7-channel alumina MF (HC10) and PP beads for the effect of PP beads concentration as compared with results of tubular carbon fiber UF (C005) membrane.

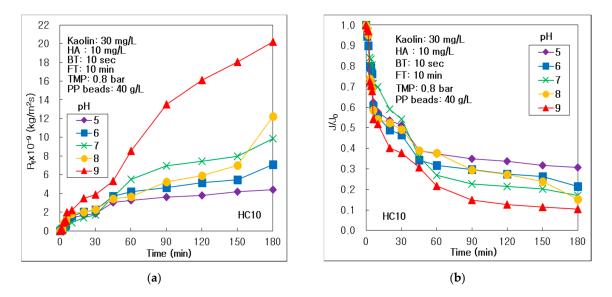| PP Beads (g/L)	Turbidity (NTU)				Average Treatment Efficiency (%)	
	Feed Water		Treated Water		7 Channels Alumina MF	Tubular Carbon Fiber UF [23]
	Range	Average	Range	Average		
0	20.6–25.9	23.1	0.207–0.326	0.264	98.9	98.6
5	22.8–27.3	25.5	0.338–0.587	0.438	98.3	98.5
10	22.8–25.7	24.4	0.297–0.434	0.349	98.6	99.1
20	20.7–28.8	24.0	0.298–0.408	0.341	98.6	97.9
30	15.3–20.4	17.8	0.353–0.542	0.447	97.5	99.3
40	22.8–25.8	24.1	0.254–0.333	0.297	98.8	98.7
50	27.3–34.3	29.7	0.308–0.354	0.339	98.9	98.4

As shown in Table 7, the treatment efficiency of DOM (UV$_{254}$ absorbance) did not show a trend; however, that was the maximal of 51.3% at 5 g/L of PP beads. This proves that the optimal PP beads concentration was 5 g/L to remove DOM in this hybrid process of HC10 MF and pure PP beads. In the previous work by [23] for the hybrid process of the C005 and the pure PP beads, the treatment efficiency of DOM showed a trend to increase dramatically from 75.9% to 84.1%, with an increase in the PP beads concentration. It was much higher than those for the hybrid HC10 MF process in this study, because the secondary gel layer could be formed denser on the UF membrane surface of smaller pore size than MF. Finally, the more PP beads could adsorb the more efficiently DOM on the surface of PP beads in the hybrid C005 UF process.

Table 7. Water quality and treatment efficiency of DOM (UV_{254} absorbance) in the hybrid process of 7-channel alumina MF (HC10) and PP beads for the effect of PP beads concentration as compared with results of tubular carbon fiber UF (C005) membrane.

PP Beads (g/L)	UV_{254} Absorbance (cm^{-1})				Average Treatment Efficiency (%)	
	Feed Water		Treated Water		7 Channels Alumina MF	Tubular Carbon Fiber UF [23]
	Range	Average	Range	Average		
0	0.339–0.411	0.389	0.145–0.265	0.205	47.4	75.9
5	0.498–0.569	0.526	0.214–0.298	0.256	51.3	77.0
10	0.399–0.487	0.431	0.185–0.251	0.220	48.9	77.8
20	0.305–0.363	0.342	0.155–0.228	0.182	46.8	83.2
30	0.403–0.598	0.527	0.192–0.423	0.273	48.1	82.3
40	0.318–0.359	0.335	0.154–0.177	0.168	49.8	82.4
50	0.310–0.410	0.374	0.159–0.215	0.200	46.5	84.1

3.3. Effect of pH on Membrane Fouling and Treatment Efficiency

To investigate the pH effect on membrane fouling and treatment efficiency, the pH of synthetic feed water was controlled by nitric acid (HNO_3) and sodium hydroxide (NaOH). As shown in Figure 4a, the R_f maintained the highest values at pH 9, and the lowest at pH 5 during 180 min. There was a dramatic trend to increase as the pH increased from 5 to 9 in the hybrid process of HC10 MF. As arranged in Table 8, the $R_{f,180}$ and R_{rf} were the minimum at pH 5; however, the R_b was the lowest at pH 6. Conclusively, the $R_{f,180}$ increased dramatically with an increase in pH, and was the highest, 20.20×10^9 kg/m^2s, at pH 9, which was 4.58 times higher than 4.41×10^9 kg/m^2s at pH 6. The R_{rf} and R_{if} were the maximum at pH 9 and pH 8, respectively. This means that the reversible and irreversible membrane fouling, and concentration polarization could be inhibited at acid condition, because both the membrane and humic materials had a negative surface charge at acid conditions below a pH of 7, as reported that the surface charge of ZrO_2 membrane was changed depending on the pH [25]. The surface charge of seven-channel alumina HC10 membrane, used in this study, could be changed depending on the pH, because those were the similar ceramic membranes as ZrO_2 membrane.

(**a**) (**b**)

Figure 4. Effect of pH on the hybrid process of multichannel alumina MF and PP beads with UV irradiation and periodic water backwashing: (**a**) Resistance of membrane fouling and (**b**) dimensionless permeate flux.

In the previous result by [23] for the hybrid process of the C005 and the pure PP beads, the R_f showed the highest at a pH of 9 after 90 min and the lowest at a pH of 5 after 150 min, and finally have a

trend to increase, with an increase in pH from 5 to 9. This result agreed exactly with that in the hybrid process of HC10 in this study. The R_b and R_{if} were the minimum at a pH of 5; however, the $R_{f,180}$ and R_{rf} were the lowest at a pH of 6. However, this trend did not match the result for HC10 in this study. Conclusively, the $R_{f,180}$ was the highest, 5.51×10^9 kg/m^{2}s, at a pH of 9, which was 1.32 times higher than 4.16×10^9 kg/m^{2}s at a pH of 6. The R_{rf} and R_{if} were the maximum at pH 9. The increasing rate, i.e., 1.32 of $R_{f,180}$ for C005 was much lower than 4.58 for HC10, because pH controlled more effectively the membrane fouling in the hybrid process of HC10 than C005.

As compared in Figure 4b to investigate the pH effect on relative permeate flux, the J/J_0 showed a trend to decrease dramatically, with an increase in the pH from 5 to 9. The J_{180}/J_0 after 180 min of operation at pH 5 was 0.306, which was 2.94 times higher than 0.104 at pH 9, as shown in Table 8. And the J_{180} was the maximum 327 L/m^{2}h at pH 6, and the minimum 270 L/m^{2}h at pH 9. This means that the high permeate flux could be acquired at pH 5, because the membrane fouling was inhibited by repulsion force between the carbon fiber membrane and humic materials, which had the same negative surface charge, as reported that the surface charge of ZrO$_2$ membrane was changed depending on the pH [25]. Finally, the V_T was the highest of 8.12 L at pH 5, because the J could maintain highly during a 180 min operation.

In the previous work by [23] for the hybrid process of the C005 and the pure PP beads, the J_{180}/J_0 after 180 min of operation at a pH of 6 was 0.099, which was 1.46 times higher than 0.068 at a pH of 9. The increasing rate, 1.42 of J_{180}/J_0 for C005, was much lower than 2.94 for HC10, because the pH maintained higher the permeate flux in the hybrid process of HC10 than C005. Additionally, J_{180} was the maximum 138 L/m^{2}h at a pH of 6, and the minimum, 107 L/m^{2}h, at a pH of 9. Finally, the V_T was the highest of 3.85 L at pH 8, because the permeate flux could maintain highly during operation. These trends did not match exactly with the result for HC10 in this study, because of the different membrane type and material.

Table 8. Effect of pH on filtration factors in the hybrid process of 7-channel alumina MF (HC10) membrane and PP beads with periodic water backwashing (BT 10 s, FT 10 min) as compared with results of tubular carbon fiber UF (C005) membrane.

Membrane	pH	5	6	7	8	9
7-channel alumina MF (HC10)	$R_m \times 10^{-9}$ (kg/m^{2}s)	1.87	1.89	1.97	2.07	2.27
	$R_b \times 10^{-9}$ (kg/m^{2}s)	0.071	0.060	0.065	0.136	0.080
	$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)	4.41	7.11	9.86	12.25	20.20
	$R_{if} \times 10^{-9}$ (kg/m^{2}s)	0.60	0.63	0.88	3.47	0.20
	$R_{rf} \times 10^{-9}$ (kg/m^{2}s)	3.802	6.472	8.975	8.780	19.997
	J_0 (L/m^{2}h)	100	70	53	44	28
	J_{180} (L/m^{2}h)	327	326	313	288	270
	J_{180}/J_0	0.306	0.215	0.171	0.152	0.104
	V_T (L)	8.12	7.00	6.30	6.26	3.87
Tubular carbon fiber UF (C005) [23]	$R_m \times 10^{-9}$ (kg/m^{2}s)	0.412	0.413	0.405	0.405	0.401
	$R_b \times 10^{-9}$ (kg/m^{2}s)	0.001	0.047	0.048	0.020	0.003
	$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)	4.87	4.16	4.92	4.62	5.51
	$R_{if} \times 10^{-9}$ (kg/m^{2}s)	0.003	0.010	0.020	0.006	0.020
	$R_{rf} \times 10^{-9}$ (kg/m^{2}s)	4.865	4.149	4.898	4.616	5.487
	J_0 (L/m^{2}h)	1537	1382	1401	1494	1573
	J_{180} (L/m^{2}h)	120	138	118	126	107
	J_{180}/J_0	0.078	0.099	0.084	0.084	0.068
	V_T (L)	3.30	3.68	3.17	3.85	3.21

As arranged in Table 9, the treatment efficiency of turbidity decreased slightly from 98.6% to 98.0%, with an increase in pH from 5 to 9. This means that the turbid matter, such as kaolin, could be removed more effectively at acidic pH conditions in the hybrid process of the alumina HC10 MF membrane with periodic water backwashing. Y. Zhao et al. [26] reported that zeta-potential of alumina

membrane decreased with an increase in pH. This phenomenon could be explained that the treatment efficiency of turbidity decreased slightly, because the decrease of zeta potential of HC10 membrane resulted in a decrease of the electroviscous effect. However, in the previous result [23] for the hybrid process of the C005 and the pure PP beads, the treatment efficiency of turbidity was almost constantly above 98.7%, independent of the pH. This means that the pH could not affect treating the turbid matter for carbon fiber UF process with water backwashing.

Table 9. Water quality and treatment efficiency of turbidity in the hybrid process of 7-channel alumina MF (HC10) and PP beads for the effect of pH as compared with results of tubular carbon fiber UF (C005) membrane.

pH	Turbidity (NTU)				Average Treatment Efficiency (%)	
	Feed Water		Treated Water		7 Channels Alumina MF	Tubular Carbon Fiber UF [23]
	Range	Average	Range	Average		
5	21.9–25.1	23.4	0.297–0.379	0.331	98.6	98.8
6	22.4–24.5	23.5	0.229–0.347	0.321	98.6	98.8
7	23.9–26.0	25.0	0.387–0.439	0.411	98.4	98.7
8	23.4–25.7	24.5	0.411–0.511	0.471	98.1	99.0
9	20.6–21.4	20.9	0.399–0.487	0.423	98.0	98.7

As shown in Table 10, the treatment efficiency of DOM showed the highest value at a pH of 9. This means that DOM could be removed effectively at alkalic condition, because of the secondary layer on the membrane surface accumulated by the most severe membrane fouling. However, in the previous result [23] for the hybrid process of the C005 and the pure PP beads, the treatment efficiency of DOM was the highest at a pH of 5. This means that DOM could be removed effectively at acid condition, because of repulsion force between the carbon fiber membrane and humic materials.

Table 10. Water quality and treatment efficiency of DOM (UV_{254} absorbance) in the hybrid process of 7-channel alumina MF (HC10) and PP beads for the effect of pH as compared with results of tubular carbon fiber UF (C005) membrane.

pH	UV_{254} Absorbance (cm^{-1})				Average Treatment Efficiency (%)	
	Feed Water		Treated Water		7 Channels Alumina MF	Tubular Carbon Fiber UF [23]
	Range	Average	Range	Average		
5	0.421–0.511	0.493	0.211–0.264	0.243	50.7	85.1
6	0.478–0.513	0.499	0.219–0.289	0.252	49.4	81.6
7	0.489–0.509	0.497	0.239–0.269	0.256	48.6	82.4
8	0.458–0.527	0.493	0.222–0.279	0.253	48.7	82.3
9	0.511–0.521	0.516	0.213–0.271	0.245	52.5	81.7

4. Conclusions

In this study, the roles of humic acid, PP bead concentration, and pH were investigated on membrane fouling and treatment efficiency of turbid matter or DOM in the hybrid process of the multichannel alumina MF membrane and pure PP beads with periodic water backwashing. The results of pH effect were compared with those of the previous study [22,23] in the hybrid process of the tubular carbon fiber UF and the same PP beads with water backwashing. In conclusion, the following results could be extracted from these investigations.

(1)　DOM, such as HA, could drive membrane fouling more severely on the surface and inside the alumina membrane, with an increasing HA concentration in water; however, the thick fouling cake on the membrane could be removed by water backwashing at 10 mg/L of HA. DOM could affect

the treatment of suspended particles, such as kaolin, in the hybrid process of the multichannel alumina MF and the pure PP beads; however, it could not affect the process of tubular carbon fiber UF membrane. DOM could be treated more effectively at high DOM condition in the hybrid water treatment process of seven-channel alumina MF and pure PP beads.

(2) The optimal PP beads concentration could be 5 g/L to control the membrane fouling in this hybrid process of seven-channel alumina MF HC10 and PP beads. The tubid matters could be treated effectively, independent of PP beads concentration in this hybrid process. The optimal PP beads concentration was 5 g/L to remove DOM in this hybrid process. The optimal PP beads concentration to reduce the turbid matter could be 30 g/L in the hybrid process of C005 UF membrane.

(3) The reversible and irreversible membrane fouling, and concentration polarization could be inhibited at acid condition, because both the membrane and humic materials had a negative surface charge at acid conditions below pH 7. The turbid matter could be removed more effectively at an acidic pH condition in the hybrid process of the alumina HC10 MF membrane; however, pH could not affect treating the turbid matter for carbon fiber UF process. DOM could be removed effectively in alkalic condition, because of the secondary layer on the membrane surface accumulated by the most severe membrane fouling.

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