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Effect of N_2-backflushing Time in Carbon Ceramic UF & MF System for Paper Wastewater Treatment

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Abstract: The wastewater discharged from a paper plant was filtrated by 3 kinds of tubular carbon ceramic UF and MF membranes with N_2-backflushing. The filtration time (FT) was fixed at 8 min or 16 min, and N_2-backflushing time (BT) was changed in 0~60 sec. The optimal condition was discussed in the viewpoints of total permeate volume (V_T), dimensionless permeate flux (J/J_0) and resistance of membrane fouling (R_f). In the viewpoints of V_T, J/J_o and R_f, the optimal N_2-BT was 40 sec at both FT for M9 (MWCO: 300,000 Daltons) and C005 (0.05 μm) membranes. However, for C010 (0.1 μm) it was 10 sec at FT=8 min, and 20 sec at FT=16 min in the viewpoints of J/J_o and R_f, and 5 sec at both FT in the viewpoints of V_T. It means that the short N_2-BT could reduce the membrane fouling and recover the permeate flux sufficiently for MF membrane having a large pore size as C010. Average rejection rates of pollutants were higher than 99.0% for turbidity and 22.8~59.6% for COD_{Cr}, but rejection rates of total dissolved solid (TDS) were lower than 8.9%. Therefore, the low turbidity water purified in our system could be reused for paper process.

Keywords: *ceramic membrane, nitrogen-backflushing, microfiltration, paper wastewater, ultrafiltration*

1. Introduction

Nowadays the recycling rates of industrial wastewater should be increased, and the dual water system extended to solve the shortage of water source and water pollution by dramatic economic development of developing countries. Various technologies for advanced wastewater treatment have been developed to satisfy such a demand, and one of them was membrane separation. Recently the membrane separation has been applied to wastewater treatment for reuse.

Many researchers have published the results of wastewater treatment by membrane separation. Tchobanoglous *et al.*[1] treated highly the domestic wastewater by ultrafiltration (UF), and Cheryan *et al.*[2] studied the economic efficiency of oil-water emulsion treatment by combining method of the traditional chemical treatment and membrane separation. Roorda *et al.*[3] investigated the characteristics of ultrafiltration for effluents of two wastewater treatment plants. As an example of applications of ceramic membranes used in this study Li *et al.*[4] used ceramic microfiltration (MF) membranes to separate cells from *E. coli*-containing fermentation broth. Then, Nazzal *et al.*[5] reported the effect of pH and ionic strength in water treatment by ceramic micofiltration membranes.

However, the economic efficiency of membrane separation for wastewater treatment should depend on the power cost of operation, the permeate flux, and the membrane lifetime. The lifetime of membranes has a deep relation with membrane fouling during the operation. It was well known that the membrane fouling was made by concentration polarization and gel layer formation on the surface of membranes, and adsorption and pore blockage in the pores inside mem-

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branes. Therefore, a lot of researches have been accomplished for solving the membrane fouling in the world. For an example, Taylor vortex was applied to microfiltration to reduce the membrane fouling by Park *et al.*[6] and Choi *et al.*[7]. Then, the membrane backflushing is a new technology to minimize the membrane fouling, and to maintain a high permeate flux during membrane separation. Many papers related with membrane backflushing have been published nowadays. Davis *et al.*[8] built up a modeling of concentration and depolarization with high frequency backpulsing. Srijaroonrat *et al.*[9] applied the backflushing to ultrafiltration of oil/water emulsion. And Sondhi *et al.* [10] researched that the membrane fouling could be minimized by backpulsing in the crossflow filtration of chromium hydroxide suspension using ceramic membranes. And Kuberkar *et al.*[11] could reduce the fouling resistance of pollutant layer on the membrane by backflushing in the microfiltration of protein cell mixture (BSA, yeast). Heran *et al.*[12] showed that highly frequent backflushing could be effective on the microfiltration of 3 kinds of suspended solids through inorganic tubular membranes. Then, we published membrane fouling control effects of periodic water-backflushing period, TMP, and flow rate using tubular carbon ceramic UF and MF membranes for recycling paper wastewater[13]. Also, we recently reported effects of periodic N_2-backflushing in paper wastewater treatment using carbon UF and MF membranes[14].

In this study we treated the wastewater discharged from a company making toilet papers by using 3 kinds of tubular carbon ceramic UF and MF membranes for reuse as industrial water. And we tried to find out the optimal BT of periodic N_2-backflushing system to minimize membrane fouling and to maintain a high permeate flux for the maximum total volume of purified water. The high chemical resistance of ceramic membranes used here made it possible to treat easily the paper wastewater including various chemicals and pollutants.

2. Theory

The resistance-in-series filtration equation shown in equation (1) was applied to analyze the experimental data of this research. The equation was known well in the application field of membrane separation. Carrene *et al.*[15] investigated the resistance of membrane, cake of bacterial cell, adsorption, and concentration polarization of solution by using equation (1).

$$J = \Delta P / (R_m + R_b + R_f) \qquad (1)$$

Where J was the permeate flux through membrane, ΔP was TMP (trans-membrane pressure), R_m the resistance of membrane, R_b the resistance of boundary layer, and R_f the resistance of membrane fouling.

For filtration of pure water, R_b and R_f did not exist because of no boundary layer by concentration polarization and no membrane fouling by pollutants. The equation (1) could be simplified to equation (2).

$$J = \Delta P / R_m \qquad (2)$$

Now R_m could be calculated from the experimental data of permeate flux for pure water using equation (2). Then, the plot of $R_b + R_f$ vs. t (operation time) could be obtained from the permeate flux data using wastewater. The intercepting value of y-axis (t=0) in this plot using only initial 2 or 3 data was R_b because of no R_f at the initial time of filtration, and finally R_f could be calculated using equation (1).

3. Experiments

3.1. Ceramic Membranes

In this study the optimal BT was investigated using 3 kinds of tubular carbon ceramic membranes as UF membrane M9 (MWCO: 300,000) made by Tech-Sep Inc. in France, and MF membranes C005 (average pore size: 0.05 μm) and C010 (average pore size: 0.1 μm) made by Koch Inc. in USA. Tech-Sep Inc.'s membrane

Table 1. Specification of Tubular Carbon Ceramic Membranes Used in this Study

Membrane	M9	C005	C010
MWCO* or Pore size	300,000 Daltons	0.05 μm	0.1 μm
Filtration type	Ultrafiltration	Microfiltration	Microfiltration
Outer diameter (mm)	10	8	8
Inner diameter (mm)	6	6	6
Length (mm)	250	244	244
Surface area (cm^2)	47.1	46.0	46.0
Company	Tech-Sep (France)	Koch (USA)	
Material	Active layer: Zirconium dioxide Support layer: Carbon	Carbon fiber	

*MWCO: Molecular weight cutoff

Table 2. The Quality of Wastewater Used in this Study

Membrane	M9	C005		C010	
FT (min)	8	8	16	8	16
Turbidity (NTU)	5.63	9.44	10.15	8.65	6.59
COD (mg/L)	110	150	156	157	147
TDS (mg/L)	520	1338	1358	5655	5029

was prepared by zirconium dioxide coating on carbon supported membrane, but Koch Inc.'s membranes made of only carbon fibers themselves. Table 1 showed the characteristics and specifications of tubular carbon ceramic membranes used in this experiments. The tubular membranes were used here for making a cross-flow inside the tube to reduce the membrane fouling.

3.2. Wastewater Source

The wastewater source in this research was the wastewater discharged from a company making toilet papers by recycling milk or juice cartons. The water quality of the paper wastewater was shown in Table 2. The quality was varied a lot depending on the condition of wastewater treatment plant in the paper company. However, a set of experiment at a fixed FT for a membrane was done using the paper wastewater sampled at the same time.

3.3. Experimental Procedures

The apparatus of filtration with periodic N_2-backflushing as shown in Figure 1 was designed and made by us in our laboratory for this study. The feed tank was filled with 5 L of paper wastewater, and it flowed to the inside of the tubular ceramic membrane. The permeate flow and the concentrate flow were recycled to the feed tank to maintain the concentration of the feeding wastewater almost constant during operation. The backflushing nitrogen gas flowed periodically to the outside of the tubular membrane.

To see the effect of N_2-BT, FT was fixed at 8 min or 16 min and BT was varied as 0, 5, 10, 20, 40 and 60 sec. However, the N_2-backflushing pressure was fixed at 2.0 kg$_f$/cm^2, TMP at 1.5 kg$_f$/cm^2, and the feed flow rate at 2.0 L/min. And then the permeate volume was measured by mass cylinder during 3 hours' operation. Then, for water quality of the feeding wastewater and the permeated water, we measured TDS (Total dissolved solid) by Conductivity meter (ATI Orion, Model 162), Turbidity by Turbidimeter (HF Scientific Inc., DRT-15CE), and COD (Chemical oxygen demand) by Chromium method.

4. Results and Discussion

In this study the experimental results were compared with each other in the viewpoints of dimensionless permeate flux (J/J_o) (J: permeate flux at a given time,

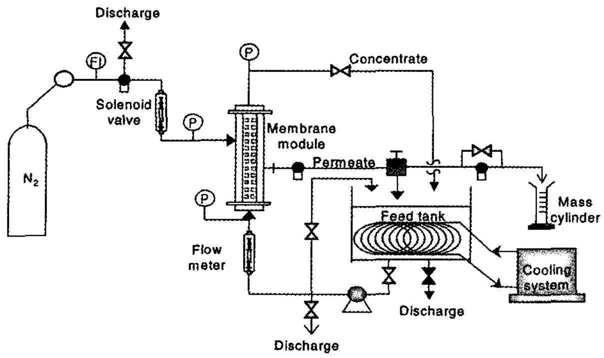

Fig. 1. Apparatus of filtration with periodic N_2-backflushing system.

J_0: initial permeate flux at t=0), total permeate volume (V_T), and resistance of membrane fouling (R_f) during 3 hours' operation. The resistances R_b, R_m and R_f were calculated from experimental values of permeate flux using the resistance-in-series filtration model of equation (1).

4.1. Effect of N_2-backflushing Time (BT)

The effect of N_2-BT on R_f for M9 (MWCO: 300,000 Daltons) UF membrane was shown in Figure 2, in which the lowest values of R_f could be maintained at BT=40 sec and FT=8 min during 3 hours' operation. The values of R_f at BT=60 sec were higher than those at BT=40 sec. Then, we can see the effect of N_2-BT on J/J_0 for M9 membrane in Figure 3. Also, the highest values of J/J_0 could be maintained at BT=40 sec, and the values of J/J_0 at BT=60 sec were lower than those at BT=40 sec. It means the optimal condition was BT=40 sec at FT=8 min for M9 to treat paper wastewater used here in the viewpoints of R_f and J/J_0. In other words BT=40 sec was the most effective to minimize the membrane fouling for M9, and BT=60 sec was too long to minimize the membrane fouling and to maintain the high permeate flux at FT=8 min for M9 UF membrane.

Table 3 and Figure 4 showed the effect of N_2-BT on final R_f ($R_{f,180}$) after 3 hours' operation for all UF and MF membranes used here. As shown in Table 3 and Figure 4, $R_{f,180}$ for C005 MF membrane

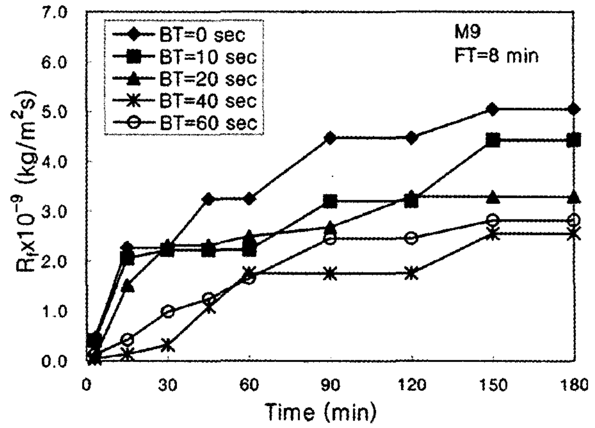

Fig. 2. Effect of N_2-backflushing time on resistance of membrane fouling for M9 membrane at FT=8 min.

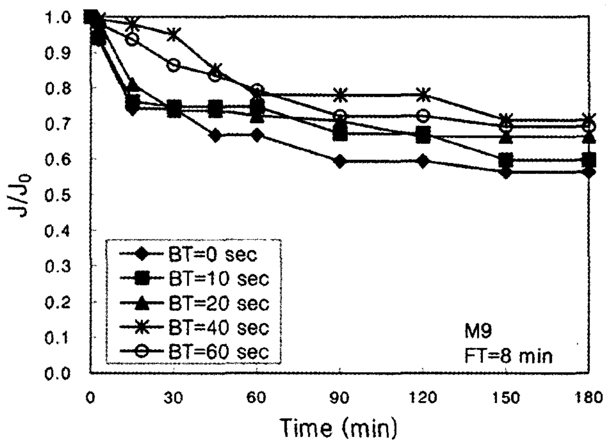

Fig. 3. Effect of N_2-backflushing time on dimensionless permeate flux for M9 membrane at FT=8 min.

got the lowest values at BT=40 sec in both cases of FT=8 and 16 min. Then, we could see the effect of N_2-BT on final J/J_0 (J_{180}/J_0) after 3 hours' operation for all membranes in Table 4 and Figure 5. Also, J_{180}/J_0 for C005 got the highest values at BT=40 sec in both cases of FT=8 and 16 min. It means that the best BT of C005 was 40 sec in both viewpoints of R_f and J/J_0 in our experimental range.

However, $R_{f,180}$ for C010 (0.1 μm) MF membrane, which had the larger pore size than C005 (0.05 μm), got the lowest value at BT=10 sec when FT=8 min and at BT=20 sec when FT=16 min as shown in Table 3 and Figure 4. And $R_{f,180}$ at BT=20 sec and

Table 3. Effect of N_2-BT on Final Resistance of Membrane Fouling ($R_{f,180}$) after 3 hours' Filtration

BT (sec)	$R_{f,180} \times 10^{-9}$ (kg/m^{2}s)				
	M9	C005		C010	
	FT=8 min	FT=8 min	FT=16 min	FT=8 min	FT=16 min
0	5.05	20.5	22.2	6.21	6.68
5	-	5.31	6.24	6.18	6.51
10	4.43	7.77	12.7	4.69	4.87
20	3.30	2.95	3.29	5.19	3.98
40	2.56	2.39	2.46	6.57	5.21
60	2.83	-	-	-	-

Table 4. Effect of N_2-BT on Final Dimensionless Permeate Flux (J_{180}/J_0) after 3 hours' Filtration

BT (sec)	J_{180} / J_0				
	M9	C005		C010	
	FT=8 min	FT=8 min	FT=16 min	FT=8 min	FT=16 min
0	0.564	0.189	0.175	0.422	0.378
5	-	0.494	0.449	0.181	0.243
10	0.597	0.382	0.281	0.444	0.434
20	0.663	0.623	0.587	0.421	0.537
40	0.709	0.695	0.686	0.298	0.431
60	0.692	-	-	-	-

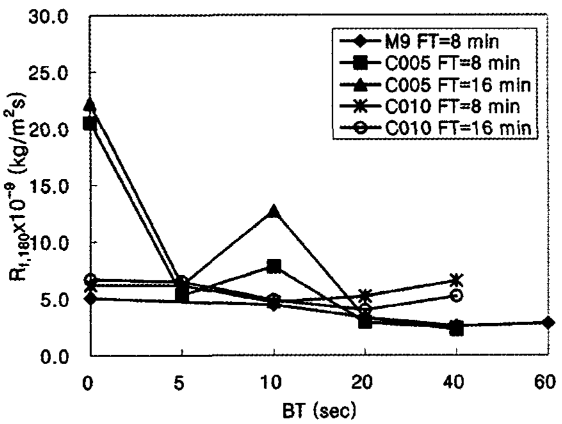

Fig. 4. Effect of N_2-backflushing time on final resistance of membrane fouling after 3 hours' operation.

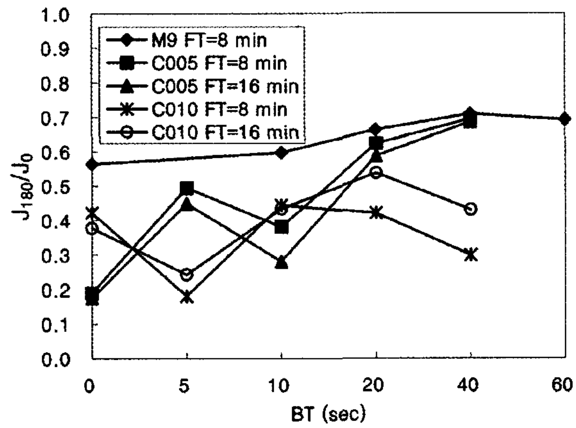

Fig. 5. Effect of N_2-backflushing time on final dimensionless permeate flux after 3 hours' operation.

FT=16 min was lower than that of BT=10 sec and FT=8 min. Also, J_{180}/J_0 for C010 got the highest values at BT=10 sec when FT=8 min and at BT=20 sec when FT=16 min as shown in Table 4 and Figure 5. And J_{180}/J_0 at BT=20 sec and FT=16 min was higher than that of BT=10 sec and FT=8 min. It means that BT=20 sec and FT=16 min was the optimal condition for C010 MF membrane. Therefore, we could find out there was an optimal N_2-BT, and longer BT than optimal BT was not helpful to minimize the membrane fouling and to maintain high permeate flux for C010 MF membrane. And the optimal N_2-BT for C010 depended on FT, but it for C005 did not as shown in Figure 4 and Figure 5.

Then, the effect of N_2-BT on total permeate volume V_T was shown in Table 5 and Figure 6 for all membranes used here. In this study V_T was the most important factor in paper wastewater treatment using UF and MF membranes, because the purpose of this membrane separation was to acquire the largest volume of purified water for recycling to the paper process. As shown in Table 5 and Figure 6, V_T for M9 and C005 got the highest values at BT=40 sec in both cases of FT=8 min and 16 min. But V_T for C010, which had the largest pore size in membranes used here, got the highest values at BT=5 sec in both FT=8 min and 16 min. In other words the optimal N_2-BT for M9 and C005 was BT=40 sec, and it was BT=5 sec for C010. It means that the short N_2-BT could reduce the membrane fouling and recover the permeate flux sufficiently for MF membrane having a large pore size as C010. However, longer N_2-BT was necessary to acquire the maximum volume of purified water for UF and MF membranes having a small

Table 5. Effect of N_2-BT on Total Permeate Volume (V_T) after 3 hours' Filtration

BT (sec)	J_{180} / J_0				
	M9	C005		C010	
	FT=8 min	FT=8 min	FT=16 min	FT=8 min	FT=16 min
0	8.00	6.39	7.52	9.46	9.68
5	-	9.42	9.49	13.29	13.19
10	8.50	8.24	7.93	12.54	12.65
20	8.79	10.69	10.65	11.40	12.04
40	10.45	11.49	11.61	11.09	11.55
60	9.28	-	-	-	-

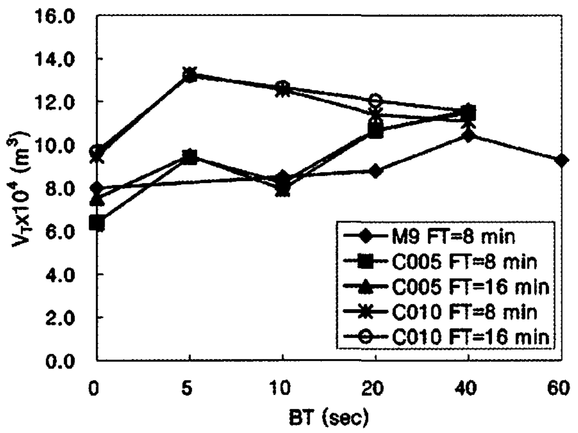

Fig. 6. Effect of N_2-BT on the total permeate volume during 3 hours' operation.

pore size as M9 or C005.

4.2. Rejection Rate of Pollutants

In this study the wastewater discharged from paper plant was treated by 3 kinds of UF and MF tubular carbon ceramic membranes. As a result, the rejection rates of Turbidity, TDS, and COD were obtained as shown in Table 6. The rejection rates were varied as 99.0~99.8% for Turbidity, 22.8~59.6% for COD_{Cr}, and 3.21~8.92% for TDS. The rejection rates of Turbidity had almost same values above 99.0% for all kinds of membranes used here, and it means that MF membranes having as large pore size as C010 could remove sufficiently most of Turbidity in paper wastewater. Therefore, the purified water acquired in our UF and MF system could be reused for paper process because of low Turbidity. However, the rejection rates

Table 6. Average Rejection Rates of Various Tubular Ceramic Membranes Used in this Study

Membrane	M9	C005		C010	
FT (min)	8	8	16	8	16
Turbidity (%)	99.0	99.7	99.8	99.6	99.7
COD (%)	59.6	37.5	35.4	25.5	22.8
TDS (%)	3.21	5.18	4.82	8.92	6.95

of COD were varied highly depending on the pore size of membranes, and M9 UF membrane (MWCO: 300,000 Daltons) got higher rejection rate of COD than those of C005 and C010. And the UF and MF membranes used here could not remove TDS sufficiently as shown in Table 6, and NF (nanofiltration) or RO (reverse osmosis) membranes were required to remove TDS in paper wastewater.

5. Conclusions

The optimal conditions of N_2-BT could be found in the carbon ceramic UF and MF system for effluent of paper plant to make toilet papers by recycling milk or juice cartons. In the viewpoints of V_T, J/J_o and R_f, the optimal N_2-BT was BT=40 sec at both FT=8 min and 16 min for M9 (MWCO: 300,000) UF and C005 (0.05 μm) MF membranes. However, it was BT=20 sec at FT=16 min for C010 (0.1 μm) MF membrane in the viewpoints of J/J_o and R_f. And in the viewpoints of V_T it was BT=5 sec at both FT=8 min and 16 min for C010, which had the largest pore size in membranes used here. It means that the short N_2-BT could reduce the membrane fouling and recover the permeate flux sufficiently for MF membrane having a large pore size as C010. However, longer N_2-BT was necessary to acquire the maximum volume of purified water for UF and MF membranes having a small pore size as M9 or C005. Then, The rejection rates were varied as 99.0~99.8% for Turbidity, 22.8~59.6% for COD_{Cr}, and 3.21~8.92% for TDS. The rejection rates of Turbidity had almost same values above 99.0% for all kinds of membranes used here, and it means that C010 MF membrane could remove sufficiently most of Turbidity in paper wastewater. Therefore, the purified water acquired in our UF and MF system with periodic N_2-backflushing could be used for paper process. However, the rejection rates of COD were varied highly depending on the pore size of membranes.

Nomenclature

BT	:	Backflushing time	[s]
FT	:	Filtration time	[s]
J	:	Permeate flux at a given time	[m/s]
J_o	:	Initial permeate flux at t=0	[m/s]
J/J_o	:	Dimensionless permeate flux	[dimensionless]
P	:	TMP (Trans-membrane pressure)	[kg_f/cm^2]
R_b	:	Resistance of boundary layer	[kg/m^2s]
R_f	:	Resistance of membrane fouling	[kg/m^2s]
R_m	:	Resistance of membrane	[kg/m^2s]
t	:	Operation time	[min]
V_T	:	Total permeate volume	[m^3]

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