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제지폐수 처리를 위한 다채널 세라믹 정밀여과 시스템에서 질소 역세척 효과

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Effect of N_2-back-flushing in Multi Channels Ceramic Microfiltration System for Paper Wastewater Treatment

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요 약: 우유 또는 주스 종이용기를 재활용하여 화장지를 생산하고 있는 제지회사에서 배출되는 제지폐수를 대상으로 주기적 질소 역세척이 가능한 세라믹 정밀여과 시스템을 운전하였다. 제지폐수 재활용을 위해 본 연구에서 7개의 채널이 있는 2 종류의 알루미나 분리막이 사용되었다. 질소 역세척 시간을 40초, 막간압력차 1.0 kg_f/cm^2, 역세척 압력 5.0 kg_f/cm^2로 고정하였을 때 0.4 μm의 평균기공 크기를 갖고 있는 HC04 알루미나 분리막의 최적 여과시간 간격은 4분으로 1.0 μm의 평균기공인 HV10 분리막의 16분보다 짧았다. 여과시간 간격과 역세척 시간을 고정한 상태에서 막간압력차(TMP)의 영향을 살펴본 결과, 높은 TMP 조건에서는 쉽게 막표면에 케이크가 형성되고 막 내부 구조에도 막오염이 발생하기 때문에 낮은 TMP 조건이 막오염 제어에 유리한 것을 알 수 있었다. 그러나 TMP는 폐수처리 여과 시스템에서 구동력이기 때문에, 가장 높은 TMP 조건에서 가장 많은 총여과부피를 얻을 수 있었다. 한편, 다채널 세라믹 분리막을 사용한 정밀여과 시스템에서 얻은 투과수는 탁도가 낮기 때문에 제지공정에서 재활용 될 수 있다.

Abstract: The ceramic microfiltration system with periodic N_2-back-flushing was operated for treating paper wastewater discharged from a company making toilet papers by recycling milk or juice cartons. Two kinds of alumina membranes with 7 channels used here for recycling paper wastewater. The optimal filtration time interval for HC04 membrane with 0.4 μm pore size was lower value of 4 min than 16 min for HC10 with 1.0 μm pore size at fixed back-flushing time 40 sec, trans-membrane pressure 1.0 kg_f/cm^2 and back-flushing pressure 5.0 kg_f/cm^2. From the results of TMP effect at fixed filtration time interval and back-flushing time, the lower TMP was better on membrane fouling because high TMP could make easily membrane cake and fouling inside membrane structure. However, we could acquire the highest volume of total permeate at the highest TMP for the reason that TMP was driving force in our filtration system to treat paper wastewater. Then the permeate water of low turbidity was acquired in our microfiltration system using multi channels ceramic membranes, and the treated water could be reused in paper process.

Keywords: *ceramic membrane, multi channel, back-flushing, microfiltration, paper wastewater*

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 waste-

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water 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 microfiltration 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 membranes. 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 back-flushing is a new technology to minimize the membrane fouling, and to maintain a high permeate flux during membrane separation. Many papers related with membrane back-flushing 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 back-flushing 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 back-flushing in the microfiltration of protein cell mixture (BSA, yeast). Heran *et al.*[12] showed that highly frequent back-flushing 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-back-flushing period, TMP, and flow rate using tubular carbon ceramic UF and MF membranes for recycling paper wastewater [13,14]. Also, we recently reported effects of periodic N_2-back-flushing in paper wastewater treatment using carbon UF and MF membranes[15,16].

The high chemical resistance of ceramic membranes used here made it possible to treat the paper wastewater including various chemicals and pollutants. And the ceramic membranes were washable by strong acid or steam with high pressure because of their high mechanical strength, and had a long lifetime. We tried to find the optimal conditions of 7 channels ceramic microfiltration system with periodic N_2-back-flushing for the wastewater discharged from a company making toilet papers, and to reuse in paper process.

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.*[17] investigated the resistance of membrane, cake of bacterial cell, adsorption, and concentration polarization of solution by using equation (1).

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

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

Table 1. Specifications of the Tubular Ceramic Membranes used in this Study

Material (Model #)	Alumina (HC10)	Alumina (HC04)
Average pore size (μm)	1.0	0.4
Company	Dongseo Inc. (KOREA)	
No. of channel	7	
Outer diameter (mm)	20	
Inner diameter of a channel (mm)	4	
Length (mm)	245	
Surface area (cm^2)	216	

Table 2. The Quality of Paper Wastewater used in this Study

Module	Experimental	Conditions	Turbidity (NTU)	COD (mg/L)	TDS (mg/L)
HC10	Effect of Filtration time interval	Range	18.1~31.5	293.3~489.5	115~120
		Average	26.7	405.3	117
	Effect of TMP	Range	25.4~35.9	208.3~489.5	108~117
		Average	29.4	329.9	113
HC04	Effect of Filtration time interval	Range	3.38~5.12	162.5~195.5	174~177
		Average	4.06	181.1	175
	Effect of TMP	Range	7.94~9.62	296.8~491.8	156~172
		Average	8.78	349.3	165

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

The optimal conditions were investigated using 7 channels ceramic membrane HC10 (average pore size: 1.0 μm) and HC04 (0.4 μm) made in Dongseo Inc., KOREA. They were prepared by alumina coating on alumina supporting membrane. Table 1 showed the characteristics and specifications of tubular alumina membrane used in this research. The tubular membrane was used here for making a crossflow to reduce the membrane fouling.

3.2. Wastewater Source

We used here 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 experiments to see one of effects on the membrane fouling was done using the paper wastewater sampled at the same time.

3.3. Experimental Procedures

The ceramic membrane module and microfiltration system with periodic N_2-back-flushing as shown in Fig. 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 con-

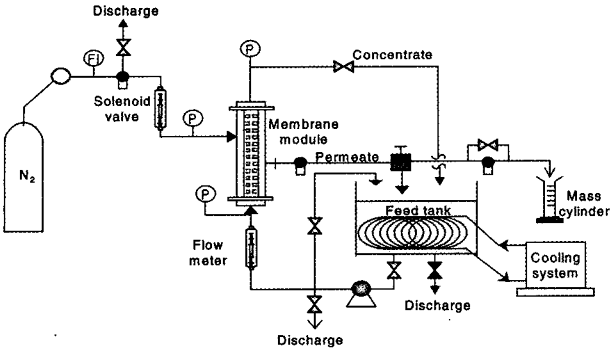

Fig. 1. Apparatus of filtration with periodic N_2-back-flushing system.

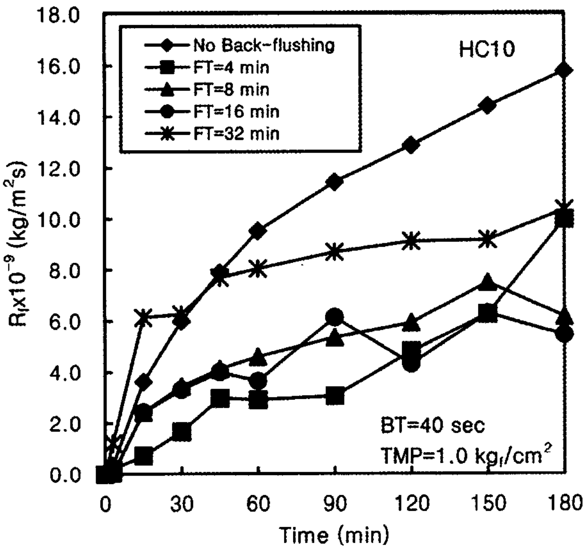

Fig. 2. Effect of filtration time interval on resistance of membrane fouling for HC10 membrane at BT=40 sec and TMP=1.0 kg_f/cm^2.

centrate flow were recycled to the feed tank to maintain the concentration of the feeding wastewater almost constant during operation. The back-flushing nitrogen gas flowed periodically to the outside of the tubular membrane.

To see the effect of N_2-back-flushing, back-flushing time (BT) was fixed at 40 sec and filtration time interval (FT) was varied as 4, 8, 16 and 32 min. Then the N_2-back-flushing pressure was fixed at 5.0 kg_f/cm^2, TMP at 1.0 kg_f/cm^2 and the feed flow rate at 2.0 L/min. To see effect of TMP we fixed FT at 16 min and BT at 40 sec. And then the permeate volume was measured by mass cylinder during 3 hours' operation. Then for

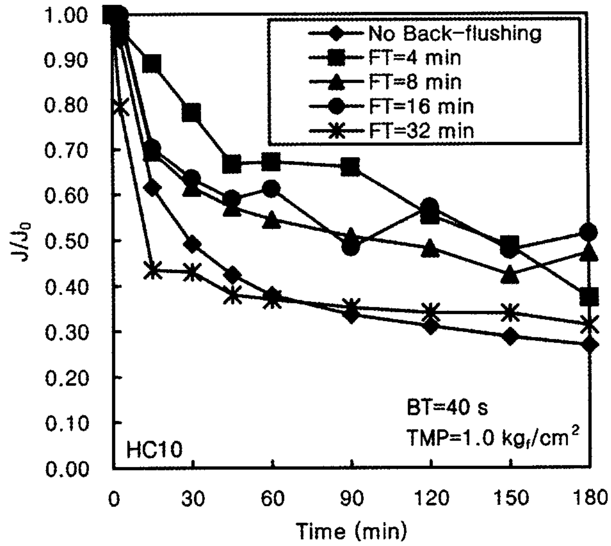

Fig. 3. Effect of filtration time interval on dimensionless permeate flux for HC10 membrane at BT=40 sec and TMP=1.0 kg_f/cm^2.

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

4.1. Effect of Filtration Time Interval

The effect of filtration time interval on membrane fouling resistance R_f for HC10 (pore size 1.0 μm) was shown in Fig. 2, in which the lowest values of R_f could be maintained at FT=16 min and BT=40 sec during 3 hours operation. Also, the highest value of the permeate flux on time (J) vs. the initial permeate flux (J_0) could be found at FT=16 min and BT=40 sec as shown in Figure 3. It means FT=16 min was the most effective filtration time interval at BT=40 sec to reduce membrane fouling and to maintain high permeate flux during filtration in our system. Then the highest total permeate volume (V_T) of 2.22 L could be acquired at FT=16 min and BT=40 sec as shown in Table 3, and it should be the optimal N_2-back-flushing

Table 3. Effect of Filtration Time Interval (FT) on Total Permeate Volume (V_T) at BT=40 sec During 3 Hours Filtration

FT (min)		No back-flushing	4	8	16	32
$V_T \times 10^3$ (m^3)	HC10 (1.0 μm)	1.56	2.10	2.09	2.22	1.84
	HC04 (0.4 μm)	2.53	4.14	3.48	3.03	2.94

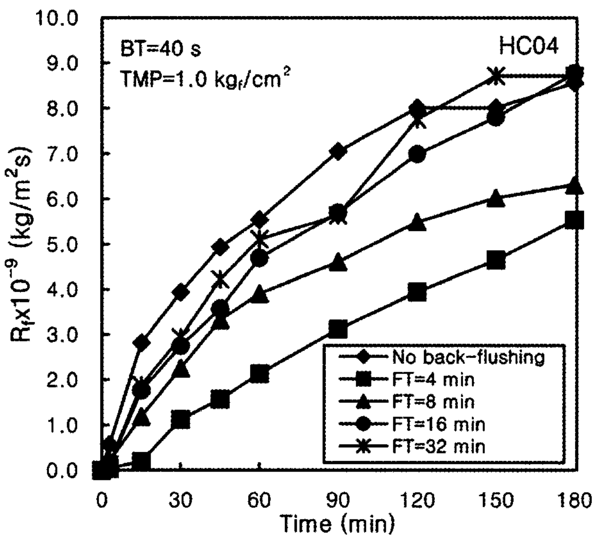

Fig. 4. Effect of N_2 filtration time interval on resistance of membrane fouling for HC04 membrane at BT=40 sec and TMP=1.0 kg_f/cm^2.

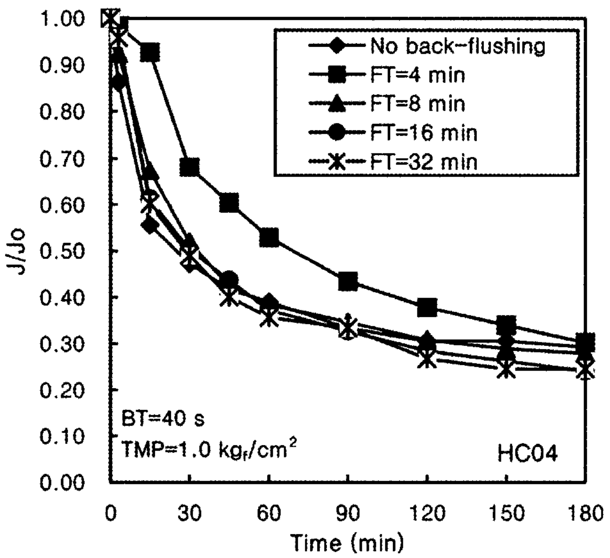

Fig. 5. Effect of N_2 filtration time interval on dimensionless permeate flux for HC04 membrane at BT=40 sec and TMP=1.0 kg_f/cm^2.

Table 4. Effect of Trans-membrane Pressure (TMP) on V_T at FT=16 min and BT=40 sec During 3 Hours Filtration.

TMP (kg_f/cm^2)		0.5	0.8	1.0	1.2	1.5	2.0
$V_T \times 10^3$ (m^3)	HC10 (1.0 μm)	1.21	1.73	2.22	2.53	-	-
	HC04 (0.4 μm)	1.68	2.36	2.69	-	3.02	4.52

condition for HC10 in our N_2-back-flushing system.

Then HC04 (pore size 0.4 μm) membrane showed the lowest value of R_f, and the highest value of J/J_0 at FT=4 min and BT=40 sec as shown Fig. 4 and 5. Also we acquired the highest V_T of 4.14 L at FT=4 min and BT=40 sec as shown in Table 3, and it could be the optimal N_2-back-flushing period for HC04 in our system. The optimal FT of 4 min for HC04 should be much lower than the optimal FT of 16 min for HC10 membrane. It means that HC04 with smaller pore size than HC10 should need frequent N_2-back- flushing to reduce membrane fouling and to maintain high flux.

4.2. Effect of Trans-membrane Pressure

At the fixed FT=16 min and BT=40 sec, we tried to find an optimal trans-membrane pressure (TMP) for HC10 membrane. The lowest value of membrane fouling resistance could be found at TMP=0.5 kg_f/cm^2 as shown in Fig. 6. Then we got the highest J/J_0 values at TMP=1.0 kg_f/cm^2 as shown in Fig. 7. Therefore the low TMP conditions could reduce the membrane fouling, but could not maintain high flux during filtration because TMP was a driving force in our membrane system. However, the highest V_T=2.53 L value could be obtained at TMP=1.2 kg_f/cm^2 during 3 hours as shown in Table 4, and it should be the best TMP in our experimental conditions.

The effect of TMP on membrane fouling resistance for HC04 membrane was shown in Figure 8. We could keep the lowest value of R_f at TMP=0.5 kg_f/cm^2 which was the lowest condition in our experiments. Also the highest value of J/J_0 could be taken at the same TMP= 0.5 kg_f/cm^2 as shown in Fig. 9. It means the lower TMP should reduce the membrane fouling during

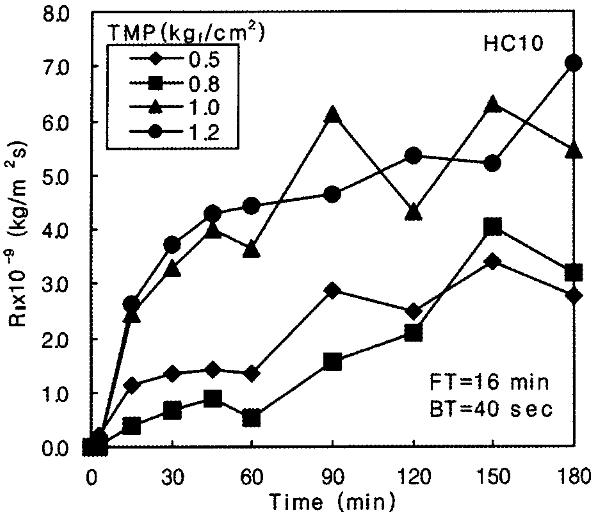

Fig. 6. Effect of trans-membrane pressure on resistance of membrane fouling for HC10 membrane at FT=16 min and BT=40 sec.

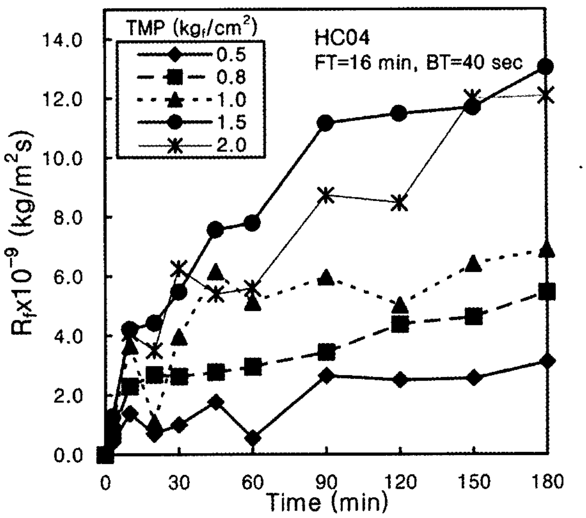

Fig. 8. Effect of trans-membrane pressure on resistance of membrane fouling for HC04 membrane at FT=16 min and BT=40 sec.

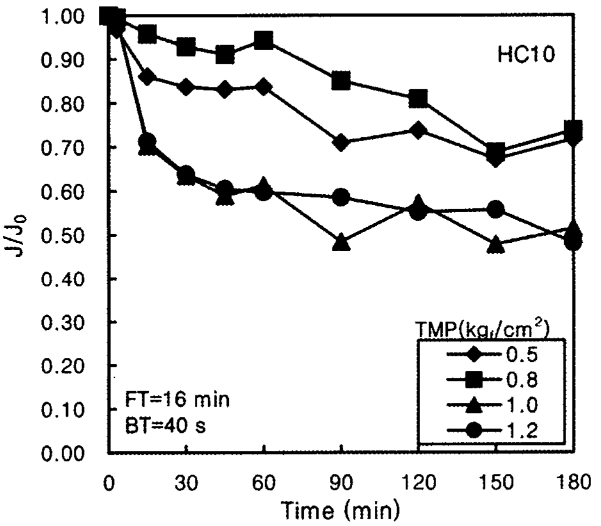

Fig. 7. Effect of trans-membrane pressure on dimensionless permeate flux for HC10 membrane at FT=16 min and BT=40 sec.

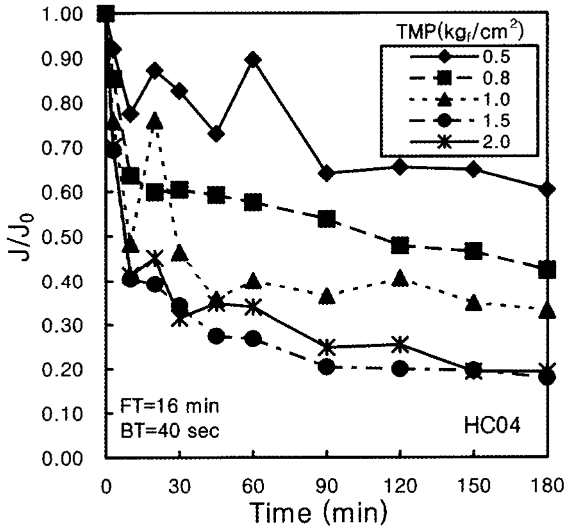

Fig. 9. Effect of trans-membrane pressure on dimensionless permeate flux for HC04 membrane at FT=16 min and BT=40 sec.

filtration of paper wastewater, and could maintain high permeate flux compared to initial flux. However, TMP was driving force in our filtration system, and the largest value V_T of 4.52 L could be obtained at the highest TMP condition of 2.0 kg_f/cm^2 as shown in Table 4.

4.3. Rejection Rates of Pollutants

The average rejection rates were above 90% for turbidity, and above 25% for COD_{Cr}, and below 1% for TDS because microfiltration membranes were used here. And the permeate water treated in our system could be reused in paper plant because its turbidity was low under 5 NTU.

5. Conclusions

For HC10 membrane with 7 channels, FT=16 min at fixed BT=40 sec and TMP=1.0 kg_f/cm^2 could be the optimal N_2-back-flushing period in our microfiltraion system for recycling wastewater discharged from a plant making toilet papers. However, FT=4 min at fixed BT=40 sec and TMP=1.0 kg_f/cm^2 should be the best condition for HC04 membrane with 7 channels. Therefore, membrane fouling in HC04 membrane should progress easily than HC10 because HC04 had smaller pore size than HC10. And HC04 needed more frequent N_2-back-flushing than HC10 to maintain high flux and largest total permeate volume. From the results of TMP effect on resistance of membrane fouling (R_f) and permeate flux vs. initial flux (J/J_0), the lower TMP was better on membrane fouling because high TMP accumulated easily membrane cake on membrane surface and made fouling inside membrane structure. However, we could acquire the highest volume of total permeate at highest TMP for the reason that TMP was driving force in our filtration system to treat paper wastewater. Then the permeate water with low turbidity below 5 NTU was acquired in our microfiltration system using two kinds of alumina ceramic membranes with 7 channels. And this wastewater treated in our system could be reused in paper process.

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