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Tubular Alumina Microfiltration Membrane System with Periodic N_2-back-flushing for Water Treatment

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Abstract: The Gongji stream water of Chuncheon city was filtrated by 2 kinds of tubular alumina ceramic MF membranes with periodic N_2-back-flushing. N_2-back-flushing time (BT) was changed in 0~50 sec at fixed filtration time (FT), or back-flushing period, of 4 min for NCMT-5231 membrane (0.05 μm). Then, FT was changed in 0~32 min at fixed BT of 40 sec for NCMT-7231 (0.1 μm). In the viewpoints of total permeate volume (V_T), dimensionless permeate flux (J/J_0) and resistance of membrane fouling (R_f), the optimal N_2-BT was 50 sec, which was the longest BT, at 4 min FT for NCMT-5231. It means the longest BT was the most effective to minimize the membrane fouling, and we could acquire the most V_T. But the optimal FT for NCMT-7231 was 16 min in the viewpoint of V_T, and was 8 min in the viewpoints of J/J_0 and R_f at fixed BT of 40 sec. The rejection rates were excellent as 80.6~96.6 % for turbidity, 35.2~58.4% for NH_3-N, 16.3~45.2% for T-P and 16.3~45.2% for COD_{Mn}. However, the rejection rate of T-N was very low as 2.7~13.4% and it of TDS below 6.1%.

Keywords: *microfiltration, ceramic membrane, water treatment, N_2-back-flushing, tubular*

1. Introduction

According to pollution deterioration of drinking water source due to various organic and inorganic matters, turbidity, and pathogens, both interests and applications of advanced water treatment have increased in order to remove effectively those pollutants of undesirable drinking water source [1]. Recently researches of water treatment by using membrane separation have achieved actively. Fiksdal and Leiknes [2] could remove viruses in drinking water by MF and UF membrane filtration combined with pre-coagulation/flocculation. And Malek *et al.* [3] investigated photooxidation as a pretreatment to break down the natural organic matter in surface water, and could reduce fouling in microfiltration systems for drinking water treatment.

However, the application of membrane process to drinking water treatment has the problem of membrane fouling and decline of permeate flux, and it shortens membrane lifetime. Membrane fouling was made by inorganic particles (e.g. iron, silica and suspended solids) and organic compounds (e.g. humic substances, polysaccharides, proteins and microorganisms) [4,5]. And it caused concentration polarization and gel layer formation on the surface of membranes, and adsorption and pore blockage in the pores inside membranes.

However, the economic efficiency of membrane separation for water 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.

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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 and time using tubular carbon ceramic MF and UF membranes for lake water [1,13]. Also, we reported effects of periodic N_2-back-flushing in paper waste-water treatment using tubular or multi-channel carbon UF and MF membranes [14-16].

In this study we performed periodic N_2-back-flushing to minimize membrane fouling and to enhance permeate flux in microfiltration system using 2 kinds of tubular ceramic membranes for Gongji stream water treatment. And we tried to find optimal operating conditions by investigating effects of N_2-back-flushing period (FT) and back-flushing time.

2. Theory

The resistance-in-series filtration model in equation (1) was applied to analyze experimental data for calculating filtration resistance and permeate flux (J) in this research. The equation was known well in the application field of membrane separation [1].

$$J = \Delta P / (R_m + R_b + R_f) \tag{1}$$

Where J was the permeate flux through membrane, $\triangle P$ 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).

$$J = \triangle P / R_m \tag{2}$$

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 and Stream Water Source

The optimal conditions were investigated tubular ceramic membranes NCMT-5231 (0.05 μm) and NCMT-7231 (0.1 μm) coated with α-alumina on supporting layer of α-alumina. We purchased the membrane from Nano Pore Materials Co. in Korea, and its surface area was 62.8 cm^2, O.D. 10 mm, I.D. 8 mm, length 250 mm, and its thickness 1 mm. The source water used here was sampled at Gongji stream located in Chuncheon city, and water quality was arranged in Table 1.

Table 1. The Quality of Stream Water Source Used in This Study

Experimental conditions	Effect of FT		Effect of BT	
Water quality	NCMT-7231		NCMT-5231	
	Range	Average	Range	Average
TDS (mg/L)	51~73	58	75~79	78
Turbidity (NTU)	2.10~7.02	5.08	2.91~5.03	3.97
COD_{Mn} (mg/L)	1.54~3.18	2.68	2.65~3.97	3.11
NH_3-N (mg/L)	0.098~1.375	0.413	0.098~0.173	0.143
T-N (mg/L)	2.387~3.375	3.007	0.492~2.737	1.970
T-P (mg/L)	0.054~0.332	0.116	0.023~0.063	0.043

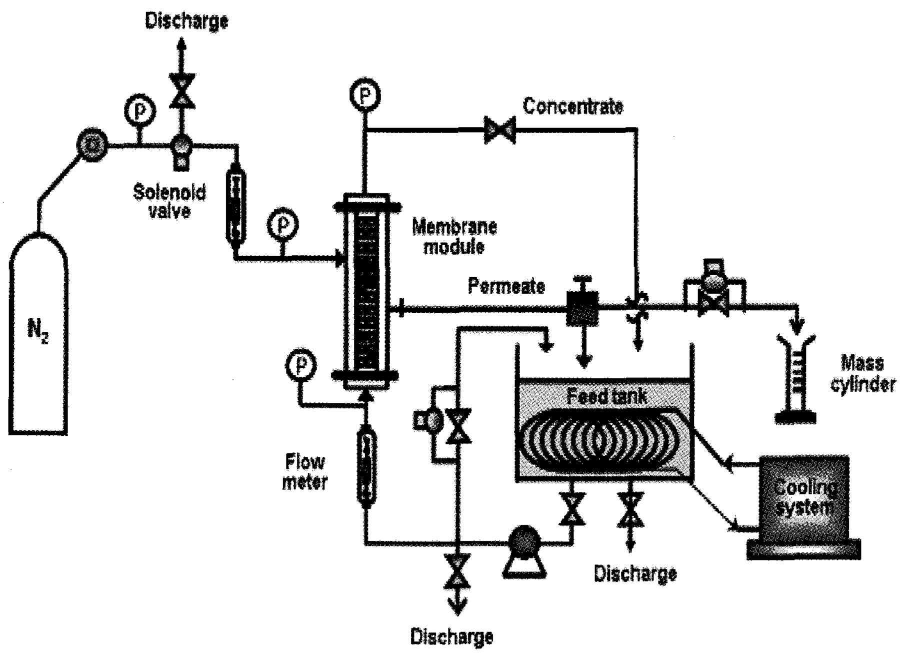

Fig. 1. Apparatus of tubular ceramic microfiltration with periodic N_2-back-flushing system.

3.2. 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 stream this study. The feed tank was filled with 5 L of stream water source, 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 water 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. Also, FT was fixed at 4 min and BT was changed as 10, 20, 30 and 50 sec to inspect effect of BT. At both two experimental conditions, TMP was fixed at 1.80 bar, N_2-back-flushing pressure at 2.0 bar, feed flow rate at 2.0 L/min, and feed water temperature at 20°C.

Then, we measured permeate flux (J) during 3 hrs' operation, and calculated resistance of membrane fouling (R_f) using equation (1) and (2). And we could acquired total permeate volume (V_T) by integrating J

Table 2. Filtration Factors in the Experiments for Effect of N_2-back-flushing Period at BT 40 sec for NCMT-7231 Membrane

Effect of FT	NBF*	4 min	8 min	16 min	32 min
$J_o \times 10^5$ (m/s)	20.81	17.05	18.57	18.38	16.98
$R_m \times 10^{-9}$ (kg/m^2 · s)	0.85	0.87	0.84	0.85	0.87
$R_{f,180} \times 10^{-9}$ (kg/m^2 · s)	3.66	**1.88**	1.94	2.01	2.54
$J_{180} \times 10^5$ (m/s)	3.91	6.05	**6.10**	5.94	4.93
$R_b \times 10^{-9}$ (kg/m^2 · s)	0.00	0.17	0.11	0.11	0.17
V_T (L)	3.44	4.98	5.08	**6.26**	5.06

* NBF : No back-flushing

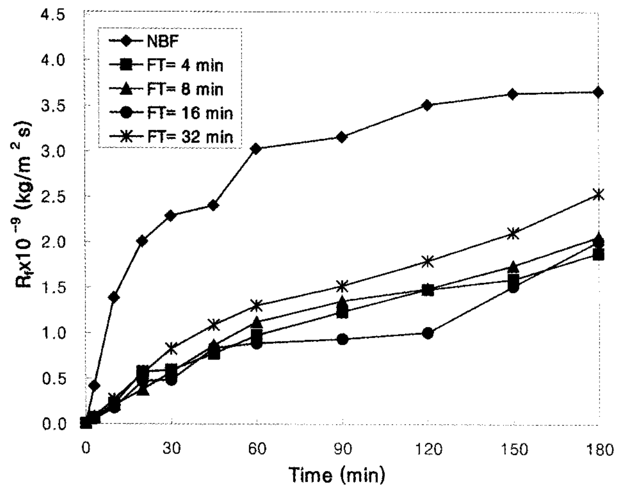

Fig. 2. Effect of N_2-back-flushing period (FT) on resistance of membrane fouling for NCMT-7231 membrane.

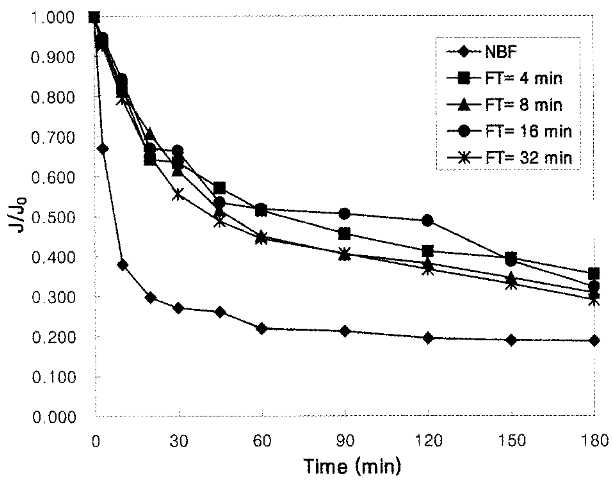

Fig. 3. Effect of N_2-back-flushing period (FT) on dimensionless permeate flux for NCMT-7231 membrane.

from starting time to 3 h [14]. And we analyzed TDS (total dissolved solid), turbidity, COD_{Mn} (chemical oxygen demand), NH_3-N, T-N and T-P of supplied and permeate water by standard method of water analysis.

4. Results and Discussions

4.1. Effect of N_2-back-flushing Period (FT) for NCMT-7231 (0.1 μm)

The membrane fouling was investigated according to N_2-back-flushing period and time in Gongji-lake water treatment using tubular ceramic MF membrane. The result of FT effect was given in Fig. 2 at fixed 40 sec back-flushing time. As shown in Fig. 2, R_f was the lowest at FT 16 min during 60~120 min, but the lowest final value of R_f was represented at FT 4 min and BT 40 sec, in which membrane fouling could decrease to 51.4% of R_f in No back-flushing (NBF) condition. And FT 4 min was the shortest back-flushing period, thus it means that the shorter back-flushing period was more effective to reduce membrane fouling. It was the same result of water-back-flushing for high turbid water treatment at discharged position of Soyang Dam in our previous study [1].

Also, the highest final value of the permeate flux on time (J) vs. the initial permeate flux (J_o) could be found at FT 4 min, as shown in Fig. 3. J/J_o was 0.335 at this operating condition, but 0.188 at NBF condition. It means FT 4 min was the most effective N_2-back-flushing period to reduce membrane fouling and to maintain high permeate flux during 3 hrs' operation in our MF system. However, the highest total permeate

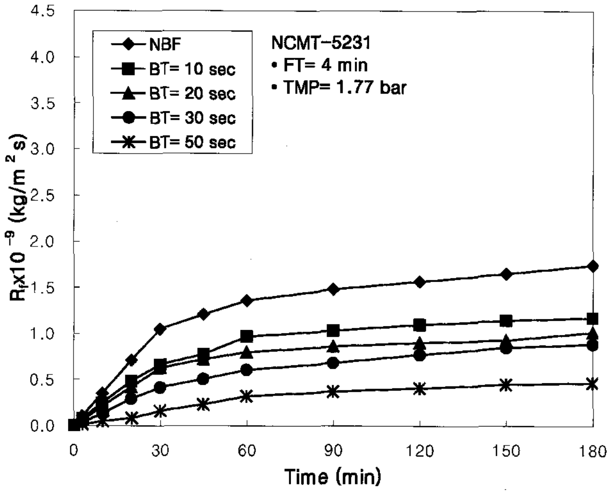

Fig. 4. Effect of N_2-back-flushing time (BT) on resistance of membrane fouling for NCMT-5231 membrane.

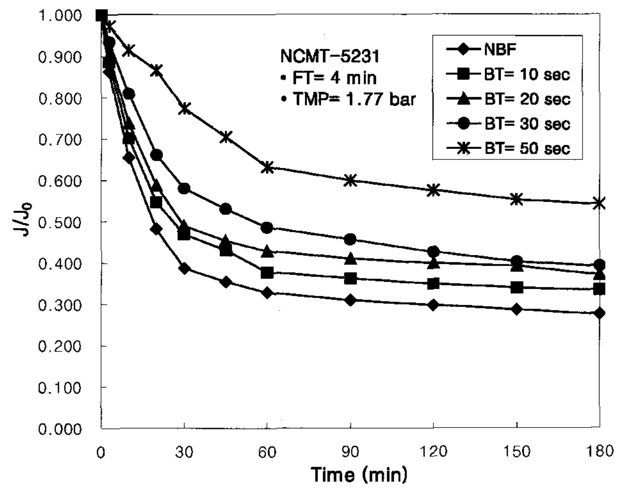

Fig. 5. Effect of N_2-back-flushing time (BT) on dimensionless permeate flux for NCMT-5231 membrane.

volume (V_T) of 6.26 L could be acquired at FT 16 min, as arranged in Table 2, because J/J_o at FT 16 min was the higher than that of FT 4 min during 60~120 min.

Then, final J after 3 hrs' operation was the highest value of 6.10×10^{-5} m/s at FT 8 min, and final R_f was the lowest value of 1.88×10^9 $kg/m^2 \cdot s$ at FT 4 min. Therefore, the optimal condition for Gongji stream water treatment by NCMT-7231 (0.1 μm) ceramic MF was FT 16 min and BT 40 sec in the experimental range of this study, because of the highest V_T.

4.2. Effect of N_2-back-flushing Time (BT) for NCMT-5231 (0.05 μm)

BT were adopted as 10, 20, 30 and 50 sec to see effect of BT at the fixed FT 4 min, which was the shortest N_2-back-flushing period in our FT effect experiment. The lowest value of membrane fouling resistance could be found at BT 50 sec and FT 4 min, in which membrane fouling could decrease to 27.0% of R_f in NBF condition, as plotted in Fig. 4. This optimal condition was the longest BT in our BT effect experiment, and it was same as the result [1] for high turbid water source at discharged position of Soyang Dam.

Also, we have got the highest dimensionless permeate flux (J/J_o) value of 0.541 at BT 50 sec, as shown in Fig. 5. It means that the longest BT should reduce the membrane fouling effectively during operation. And as arranged in Table 3, the most V_T of 9.37 L, which was much more than 6.26 L of optimal condition in FT effect experiment using NCMT-7231 (0.1 μm), could be obtained at BT 50 sec and FT 4 min. And final J after 3 hrs' operation was the highest value of 17.3×10^{-5} m/s and final R_f was the lowest value of 0.47×10^9 $kg/m^2 \cdot s$ at FT 4 min.

Finally, the optimal condition for Gongji stream water treatment by NCMT-5231 (0.05 μm) ceramic MF was BT 50 sec and FT 4 min in the experimental range of this study, because of the highest V_T, and J/J_o, and also the lowest R_f.

4.3. Water Quality and Rejection Rate of Pollutants

As results of water quality analysis for feed and treated water, average rejection rate of TDS, turbidity, COD_{Mn}, NH_3-N, T-N and T-P were arranged in Table 4. The rejection rate of turbidity was excellent as 80.6~96.6%, but it of TDS was much lower than 6.1%. Then, the rejection rates were 13.2~59.6% for COD_{Mn}, 35.2~58.4% for NH_3-N, 2.7~13.4% for T-N and 16.3~45.2% for T-P, because we used MF membranes here and there seemed to be a limit to remove soluble organic materials.

Table 3. Filtration Factors in the Experiments for Effect of N_2-back-flushing Time at FT 4 min for NCMT-5231 Membrane

Effect of BT	NBF*	10 sec	20 sec	30 sec	50 sec
$J_o \times 10^5$ (m/s)	26.68	30.12	29.54	30.91	31.98
$R_m \times 10^{-9}$ (kg/m^2 · s)	0.41	0.44	0.41	0.41	0.40
$R_{f,180} \times 10^{-9}$ (kg/m^2 · s)	1.74	1.35	1.02	0.89	**0.47**
$J_{180} \times 10^5$ (m/s)	7.33	10.33	10.94	12.10	**17.30**
$R_b \times 10^{-9}$ (kg/m^2 · s)	0.248	0.18	0.19	0.17	0.15
V_T (L)	6.46	8.43	8.41	9.28	**9.37**

* NBF : No back-flushing

Table 4. Average Rejection Rate of Tubular Ceramic Microfiltration with Periodic N_2-back-flushing System

Pollutants	Effect of FT	Effect of BT
TDS (%)	0.11	4.20
Turbidity (%)	93.7	92.8
CODMn (%)	33.1	20.3
NH_3-N (%)	46.5	44.7
T-N (%)	5.25	11.6
T-P (%)	25.9	36.9

5. Conclusions

The Gongji stream water of Chuncheon city in Korea was filtrated by 2 kinds of tubular alumina ceramic MF membranes made in Korea with periodic N_2-back-flushing. N_2-back-flushing time (BT) was changed in 0~50 sec at fixed filtration time (FT), i.e. back-flushing period, of 4 min for NCMT-5231 membrane (0.05 μm). Then, FT was changed in 0~32 min at fixed BT of 40 sec for NCMT-7231 (0.1 μm). However, the N_2-back-flushing pressure was fixed at 2.0 bar, TMP was at 1.80 bar, and the feed flow rate was at 2.0 L/min in all experiments. In the viewpoints of total permeate volume (V_T), dimensionless permeate flux (J/J_0) and resistance of membrane fouling (R_f), the optimal N_2-BT was 50 sec, which was the longest BT, at 4 min FT for NCMT-5231. It means the longest BT was the most effective to minimize the membrane fouling, and we could acquire the most V_T. But the optimal FT for NCMT-7231 was 16 min in the viewpoint of V_T, and was 8 min in the viewpoints of J/J_0 and R_f at fixed BT of 40 sec. Therefore there was a suitable N_2-back-flushing period of 8 min for reducing membrane fouling of NCMT-7231, but we could accumulate the most V_T at longer FT conditions of 16 min, in which the stream water was filtrated during longer operation time than 8 min.

Then, the rejection rate of Turbidity was excellent as 80.6~96.6%, but it of TDS was much lower than 6.1 %. And the rejection rates were 13.2~59.6% for COD_{Mn}, 35.2~58.4% for NH_3-N, 2.7~13.4% for T-N and 16.3~45.2% for T-P, because we used MF membranes here and there seemed to be a limit to remove soluble organic materials.

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