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The effects of coagulation with MF/UF membrane filtration in drinking water treatment

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ABSTRACT

This study proposes coagulation as a pretreatment permitting the optimal operation of drinking water membrane treatment process. Membrane fouling was evaluated for various coagulation control methods and different membrane types [microfiltration (MF)/ultrafiltration (UF)]. A system using MF and UF was operated at a permeate flux of 1.5, 2.0 (m^3/m^2)·day. For the direct membrane filtration process without a pretreatment process (Run A), the days of effective operation (Te) under 1.0 $kg_f cm^{-2}$ of trans-membrane pressure (TMP) reached 33 days. It is thought that a pretreatment process was required to remove micro-particles that cause membrane fouling. In the case of coagulation control, according to raw water turbidity (Run B), the Te was 50 days. On the other hand, it could be seen that the membrane filtration system (Run C) that controls the coagulant dose based on the streaming current value (SCV) measurement operated at higher stability (95 days) than previous tests, even under high turbidity (>200 NTU). Pilot plant operation with a UF membrane was more stable than one with an MF membrane. In addition, the TMP in the coagulation/UF process was maintained at a significantly stable level despite the high permeate flux of 2.0 (m^3/m^2)·day, when coagulation was controlled by SCV. It can be concluded that the membrane filtration process operated with higher stability when the coagulant dose was properly controlled by SCV.

Keywords: Membrane; Ultrafiltration; Microfiltration; Coagulation; Fouling; Streaming current value (SCV)

1. Introduction

Pretreatment, such as coagulation or activated carbon adsorption, is currently being studied as a means to improve permeating efficiency for membrane filtration by fouling mitigation [1–3]. Chemical coagulation can be used as a pretreatment for membrane processes. By implementing the coagulation in conjunction with membrane filtration, enhanced removal of turbidity material, total organic carbon (TOC), natural organic matter (NOM), and trihalomethane (THM) precursors can be expected. The optimum coagulation conditions for a membrane process are expected to prevent membrane fouling [2–4].

Chemical coagulation has been widely used as a method to mitigate membrane fouling in microfiltration (MF)/ultrafiltration (UF) membranes used for drinking water treatment. The optimization of coagulation as a pretreatment for membrane processes has not yet been achieved [5–7]. Also, optimum coagulation conditions obtained for conventional water treatment systems are not necessarily applicable to membrane-based treatment

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systems. Incorrect control of coagulation conditions might cause more severe membrane fouling. The coagulation conditions often determine the degree of membrane fouling because these conditions have significant influence on the fouling mechanisms and characteristics of the flocs [8–10]. Therefore, this study proposes coagulation as a pretreatment method for the optimum operation of membrane drinking water treatment. For effective control of membrane fouling, an appropriate type of membrane was selected after the performance evaluation according to water quality and membrane characteristics.

2. Materials and methods

2.1. Experimental procedure

A pilot plant system was installed in a water treatment plant with river water as its source. The coagulation condition of the coagulation-membrane filtration process was investigated with the raw water condition being the same as that of an actual water treatment process. A pilot scale plant system using membrane filtration (500 m^3/day) was constructed with two trains and was evaluated according to the type of membrane and pretreatment process (Fig. 1). This pilot plant was operated by applying the constant flow control method using the inverter of the raw water feed pump. As a coagulant, polyaluminum chloride (PACl, $[[Al_2(OH)_nCl_{6-n}]m)$ was used, and the injection amount was automatically controlled according to each method to determine the injection amount of coagulant. An external pressure hollow fiber type (PVDF) MF membrane module with a pore size of 0.05 μm and an UF membrane of molecular weight cut off (MWCO) 150,000 Da were used in this system (Table 1). For physical cleaning of the membrane, backwashing and air scrubbing were simultaneously performed using permeate water.

Experiments were performed using five different operating conditions (Runs A–E). In the case of Run A, membrane filtration was operated independently to evaluate the necessity of the pretreatment process for

Table 1
Specifications of membranes installed in the pilot plant.

Factors	Specifications	
	MF	UF
Pore size/MWCO	0.05 μm	150,000 Da
Membrane type	Hollow fiber	
Active membrane area	72 m^2/module	
Material	Polyvinylidene fluoride (PVDF)	
Size	Ø 216 mm × H 2,160 mm	
Available pH range	1–10	

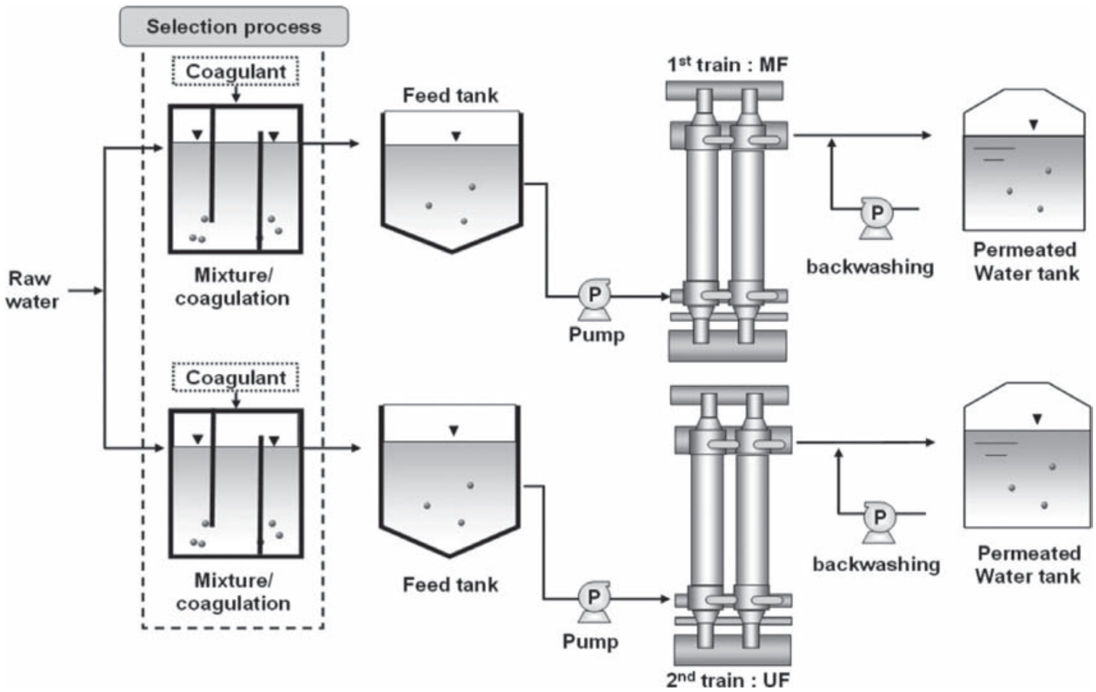


Fig. 1. Schematic diagrams of pilot plant for water treatment system using membrane process.

membrane filtration. In the case Runs B and E, the coagulation process was combined with membrane filtration to evaluate the effect of the coagulation control method on the membrane filtration process. Runs B and D controlled the coagulant injection dosage according to the turbidity of the raw water. Runs C and E controlled the coagulation using a streaming current (SC), which is an electric potential created while ions of the diffusion layer surrounding the particles are moving along with the fluid. Based on the laboratory scale results of the permeate efficiency experiments, an optimum streaming current value (SCV) was selected and reflected on the pilot plant operation. In order to maintain an optimum SCV based on the SC value of the coagulated water, an automatic coagulation control system was operated with a coagulation pump coupled with SCV. In Table 3, turbidity of influent and effluent for the Run A through E was described.

2.2. Analysis method

Permeate flux and trans-membrane pressure (TMP), which were used to monitor the membrane filtration process, was automatically measured in real time. Also the turbidity, water temperature, and pH were automatically measured in real time in order to examine the characteristics of raw water and treated water quality. Monitoring of TMP was performed at 25°C to examine the effect of raw water quality and coagulation on the TMP. In order to investigate the SC value of coagulated water, a streaming current detector (SCD, Chemtrac Systems, Inc.) that can constantly measure the electric charge of the colloid surface of treated water in the mixing/coagulation process was installed. The analysis of inorganic matter (Al, Si, Fe, Mn, Ca) was carried out using an inductively coupled plasma spectrometer (Leeman Co.). The cross-sectional images of the membrane were analyzed by a field emission scanning electron microscope (FESEM) to investigate the characteristics of the membrane surface after the flux test. In addition, the substances absorbed on the membrane surface were analyzed using an energy dispersive X-ray spectrometer (EDAX).

3. Results and discussion

3.1. Operation of MF process without coagulation

In order to evaluate the necessity of the pretreatment process, the raw water was treated using the direct

Table 2
Operation conditions of the pilot plant in Runs A through E.

Operation factors			Run A	Run B	Run C	Run D	Run E
Flux ($m^3/m^2\cdot day$)			1.5	1.5	1.5	1.5	2.0
Membrane			MF	MF	MF	UF	UF
Pretreatment		pH	–	6.8–7.0	6.8–7.0	6.8–7.0	6.8–7.0
	Coagulation	Method	–	Turbidity	SCV	Turbidity	SCV
		Concentration (mg/L)		21–61	16–68	15–60	10–50
Operation period (day)			35	50	96	262	196

Table 3
Turbidity of influent and effluent from the pilot plant in Run A through E.

			Run A	Run B	Run C	Run D	Run E
Temperature (°C)	Max.		13.3	31.8	29.7	32.4	34.5
	Min.		0.4	19.4	9.5	3.2	–0.4
	Avg.		6.0	24.3	20.3	20.7	18.8
Turbidity (NTU)	Influent	Max.	12.3	468.2	145.4	600.0	146.0
		Min.	1.0	4.6	1.3	1.2	1.9
		Avg.	6.5	85.2	16.7	22.1	17.1
	Effluent Avg.	Max.	0.0485	0.0495	0.0495	0.0475	0.0475
		Min.	0.0005	0.0005	0.0005	0.0025	0.02
		Avg.	0.0104	0.0336	0.0342	0.0233	0.0339

membrane filtration process without the pretreatment (Run A). During this period, the turbidity of influent water was measured at a range of 1–12.3 NTU (7.49 NTU on average). As can be seen from Fig. 2, the TMP increased continuously throughout the operating period despite the low turbidity range of the raw water, indicating that the membrane was being fouled.

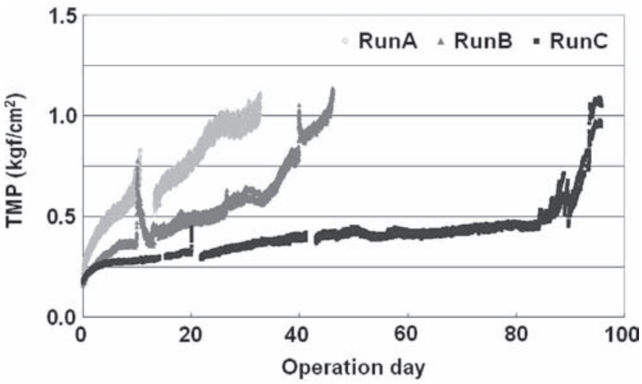
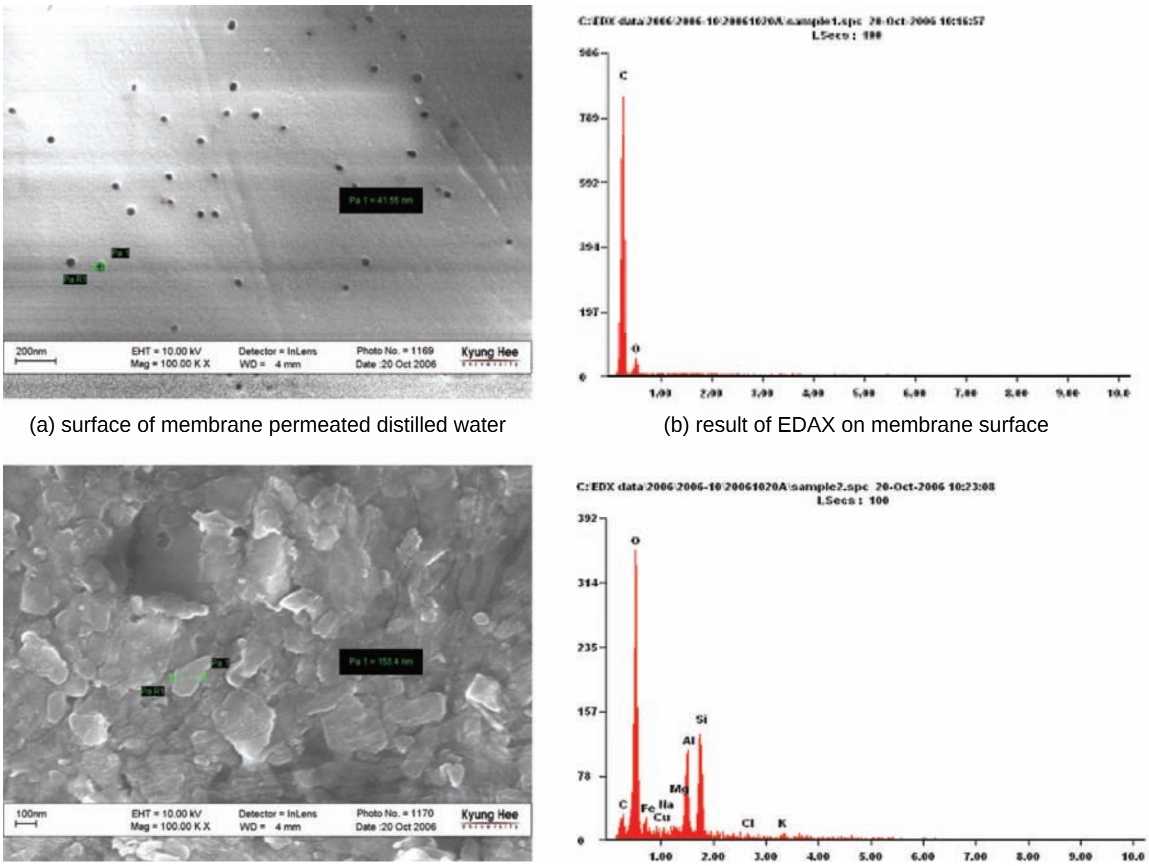


Fig. 2. Variation of TMP of MF in Run A through C.

The effective days of operation (Te), days in which the system operated under 1.0 kgfcm^{-2} of TMP, was about 33 days. The operation of membrane filtration without coagulation for a long period of time (Run A) proved to be quite difficult.

Analysis was performed to determine the cause of membrane fouling based on the results of Run A. The surface of the membranes permeated by distilled water and raw water was compared by an FESEM to evaluate the membrane's fouling characteristics. In addition, the surfaces of the membrane were analyzed using an EDAX to examine the substances attached to them. As can be seen from Fig. 3(a), the surface of the membrane through which only distilled water had permeated was so clear that even the pores could be seen. From the FESEM image, the pore size of the membrane was estimated to be 41 nm. As shown in Fig. 3(c), the size of unspecified particles was measured as 153.4 nm from the result of the analysis on the membrane surface through which raw water was permeated. It could be predicted that membrane pores were clogged by particles as large as the membrane pore size, 41 nm (Fig. 3(a)). EDAX



(a) surface of membrane permeated distilled water

(b) result of EDAX on membrane surface

(c) membrane of surface permeated raw water

(d) result of EDAX on membrane surface

Fig. 3. (a, c) FESEM image for surface analysis and (b, d) ingredients analysis attached on membrane surface (EDAX).

analysis for the substances attached on the membrane surface showed that only carbon (C, weight standard 89.6%) and oxygen (O, 10.4%), which comprise the membrane material, were detected when the distilled water was permeated (Fig. 3(b)). On the other hand, oxygen (O, 41.6%), ferric (Fe, 13.6%), aluminum (Al, 10.99%), and silicon (Si, 15.5%) were detected from the membrane surface when raw water was permeated. It was thought that oxygen originated from organic material and ferric, aluminum and silica from fine soil particles. From the analysis of the membrane surface operated in Run A, it was found that membrane fouling was caused by both organic and inorganic materials. Thus, the pretreatment process must remove micro-particles that cause membrane fouling.

3.2. Operation of coagulation/MF process

Based on the results obtained from Run A, further investigation was made on the characteristics of the TMP change according to the injection method or the coagulant dosage with coagulation pretreatment (Runs B and C). In the case of Run B, a coagulant injection was performed by applying an empirical equation for the relationship between the raw water turbidity and the TMP change, which was obtained from the results of previous experiments.

$$Y=1.8057\sqrt{X} + 20579 \quad (1)$$

where X is turbidity of raw water and Y is injection amount of coagulant. The effective days of operation (Te) in Run B were estimated to be over 50 days (Fig. 2), indicating that the effect of membrane fouling was still significant. Coagulant dosage (PACl) was varied over a range of 21–61 mg/L as turbidity of influent raw water changed rapidly from 0.1 to 496 NTU. Since TMP was continuously increased as time passed, it was considered that the applied coagulant dose and conditions were inappropriate.

In the case of Run C, based on the permeate efficiency (flux) experiment of the laboratory scale, an optimum SC value was selected and reflected on the pilot plant operation. In order to maintain an optimum SC value for the treated water, an automatic coagulation control system was operated with a coagulation pump coupled with an SCV. The influent raw water turbidity changed from 2.35 to 144.6 NTU (16.6 NTU in average), and the concentration of the injected coagulant through the SCV control was within the range of 16–68 mg PACl/L during Run C. As a result, the Te under Run C conditions was over 95 days. The method of coagulant injection using the SCV of coagulated water was more effective than the coagulant injection determined by turbidity of the raw water. Therefore, it is thought that a proper coagulation based on the SCV is required for the successful operation of a membrane filtration process to treat river water. Moon et al. also reported that appropriate control of the coagulation process resulted in stable TMP in the membrane filtration process [11]. Therefore, it is concluded that the membrane filtration process can be effectively applied for the river water treatment, as the SCV-based coagulation is used as a pretreatment method by reducing the membrane fouling.

3.3. Observation of the surface of membrane after operations

After experiments (Runs A through C) were completed, chemical cleaning (1N HCl + 1% oxalic acid) of the used membrane module was performed to measure the substances attached and accumulated on the memb rane surface and pores. Then, an analysis was made on the substances causing membrane fouling. The analysis on the supernatant concentration was divided by the days of operation after chemical cleaning and showed that it contained a high amount of aluminum in Runs B and C (Table 4). Under each condition, the aluminum concentration was 5.378 mg/L/day (Run B) and 1.884 mg/L/day (Run C), respectively. For Run A, in which coagulant was not added, the detected aluminum concentration was low. In the case of Runs B and C, the non-reacted coagulant and coagulated flocs were attached on the membrane surfaces, resulting in a high concentration of aluminum. In addition, it is thought that a high concentration of silica accumulated on membrane surfaces during runs with a raw water influent of high turbidity (Runs B and C). On the other hand, there was no tendency for accumulation of Fe, Mn, or Ca, in particular. Therefore, the major substances causing membrane fouling were non-reacted aluminum and coagulation flocs due to the coagulant injection and silica contained in the raw water influent with high turbidity.

3.4. Operation of coagulation/UF process

In Runs D and E, the characteristics of the membrane drinking water treatment system were evaluated when

Table 4
Analysis of materials attached on membrane module through chemical cleaning.

	Run A	Run B	Run C
Al (mg/L/day)	0.079	5.378	1.884
Si (mg/L/day)	0.091	2.676	0.343
Fe (mg/L/day)	0.306	0.724	0.074
Mn (mg/L/day)	0.055	0.101	0.035
Ca (mg/L/day)	1.194	0.372	0.400

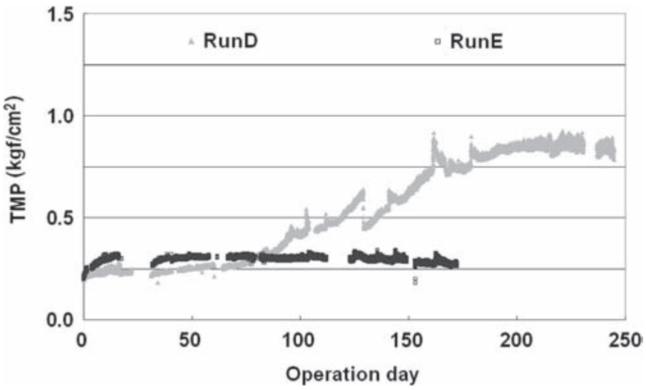


Fig. 4. Variation of TMP (values converted at 25°C) in Runs D and F.

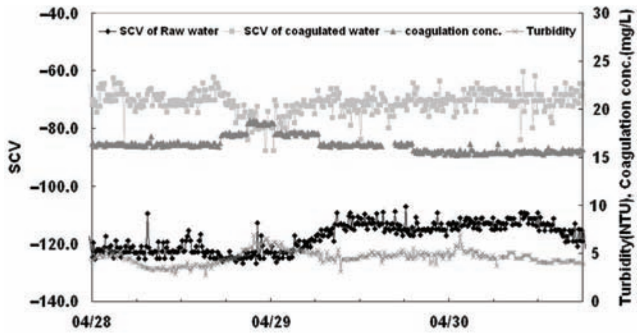


Fig. 5. Variation of raw water SCV and coagulation control according to raw water turbidity.

UF membranes with a MWCO 150,000 Da were used, instead of an MF membrane with a pore size of 0.05 μm (Runs A–C). In the case of Run D with a permeate flux of 1.5 m^3/m^2·day, the coagulant injection was performed by applying the empirical formula for the relationship between the raw water turbidity and the TMP change in the same manner as was done for Run B. The turbidity of influent raw water changed from 0.1 to 600 NTU, and the coagulant dosage was injected within a range of 15–60 mg/L. A low and stable TMP was maintained as the turbidity of raw water influent was low at the initial operation stage. Even when the turbidity was relatively high by comparison (above 100 NTU), the TMP temporally increased and then became stable after a decrease in raw water turbidity. With suspended matter increased according to the influent of high turbidity, the TMP increased temporarily. However, TMP was restored and became stable as backwashing was applied. This indicated that fouling caused by added coagulants, natural organics, and oxides was reversible.

For Run E, the operation characteristics of the UF membrane system was estimated by increasing the permeate flux up to 2.0 m^3/m^2·day. Run E was conducted using a coagulant injection within a range of 10–50 mg/L with an SCV coagulant control in the same manner as was done for Run C. The turbidity of influent raw water was changed from 1.9 to 380 NTU, and the Te under Run E conditions was measured at 196 days despite the high permeate flux. Overall, Run E showed the most stable operation among the five different types (i.e., A, B, C, D, and E) of conditions.

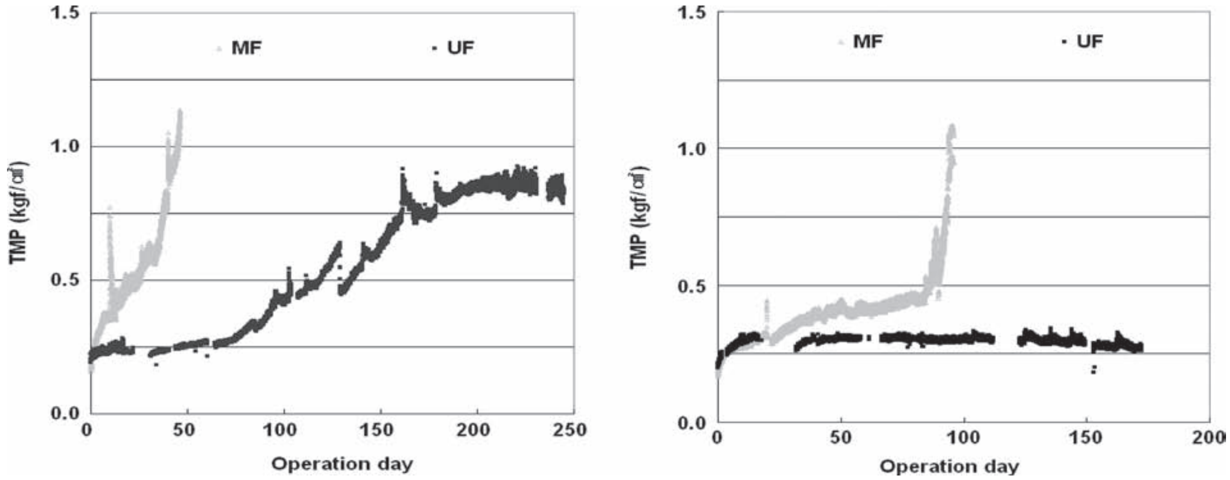
The TMP maintained its stability below 0.3 $kg_f cm^{-2}$ during not only the low turbidity period, but also the high turbidity period over 380 NTU. Therefore, it was thought that the coagulant injection based on the SCV of coagulated water can be more adequately operated than those based on the turbidity of raw water. Fig. 5 shows the SCV of raw water and coagulated water, turbidity, and coagulation dosage with time. On April 29th, the SCV of raw water decreased by approximately 8 when the raw water turbidity increased by 5 NTU, resulting in a decrease of the SCV of the coagulated water. Thus, coagulant dosage was increased to keep an optimum SCV for the coagulated water. The average value of the measured SCV of the raw water and coagulated water were –116 and –71.2 respectively, showing a difference of approximately 45. The SCV of the coagulated water controlled by the coagulation pump was maintained within the range of the optimum SCV ± 8.

In previous study, a close relationship between SC and zeta potential of suspended solids was observed as the water quality parameters were varied [12]. Additionally, it was reported that coagulant dosage could be reduced by controlling the coagulant dosage using SCV [13].

3.5. Comparisons of permeate efficiency in coagulation/MF/UF process

Fig. 6(a) compares the operation results of MF and UF using the turbidity coagulation control method, with a permeate flux of 1.5 m^3/m^2·day (Runs B, D). As a result, the Te of UF (Run D) was five times longer than the Te of MF (Run B). The UF membrane has a smoother increase in TMP than the MF membrane. Fig. 6(b) compares the operation results of MF and UF using the SCV coagulation control method. Once again, the TMP of UF with a permeate flux of 2.0 m^3/m^2·day showed very low and stable values, as compared with the TMP of MF with a permeate flux of 1.5 m^3/m^2·day.

In order to investigate why the UF operation was stable compared to MF, the permeate efficiency according to membrane pore size was estimated by using a mini-module (active membrane area 0.003 m^2/module) experiment. In this system, a hollow-fiber type MF membrane (PVDF) module with a pore size of 0.05 μm and an UF membrane of MWCO 150,000 Da were used, the same



(a) Coagulation control by raw water turbidity (Run B, D) (b) Coagulation control by SCV (Run C, E)

Fig. 6. Comparisons of MF/UF according to coagulation control method in Runs B through D.

Table 5
Properties of MF and UF membrane filtration.

Properties	Membrane	
	MF	UF
Pore size or MWCO	0.05 μm	150,000 Da
Pure water flux (J, m^3/m^2·day at 100 kPa)	31	26
Increasing rate of filtration resistance (R_r, $10^{12}/m^2$)	17	11
Backwashing recovery percent (R_b, %)	79.3	85.9
Increasing rate of irreversible resistance (R_{ir}, $10^{12}/m^2$)	0.41	0.22

membrane material as in the pilot plant. An increase in the filtration resistance and irreversible filtration resistance were expressed as the recovery rate and the average increased rate per filtration cycle. In addition, cake resistance was not considered because physical cleaning was performed completely in this study so that the membrane surface had almost no cake resistance.

Although pure water flux of UF (26 m^3/m^2·day at 100 kPa) was lower than that of the MF membrane (31 m^3/m^2·day at 100 kPa), the backwashing recovery percent of UF (85.9%) was superior to that of the MF membrane (79.3%), Also, the increasing rate of filtration resistance of UF ($11 \times 10^{12}/m^2$) was lower than that of the MF membrane ($17 \times 10^{12}/m^2$), and the increasing rate of the irreversible filtration resistance of UF ($0.22 \times 10^{12}/m^2$) was lower than that of the MF membrane ($0.41 \times 10^{12}/m^2$),

Results from the pilot plant tests showed that operation with UF was quite stable, compared to that with MF. It appeared that UF was less influenced by raw water turbidity and membrane fouling materials. The backwashing recovery percent of UF was superior to that of the MF membrane, and the membrane fouling of UF was less than that of MF. The average pore size of the MF membrane is relatively larger than that of UF membrane. This resulted in a more severe accumulation of micro-particles with the MF membrane. Continuous accumulation of micro-particles reduces the permeation efficiency of the membrane, leading to unstable operation of the membrane filtration process [2]. Also, this pore blocking for MF is difficult to recover through backwashing.

4. Conclusion

This study proposed coagulation as a pretreatment for the optimum operation of membrane drinking water treatment. Operation characteristics as a function of coagulation control method and the type of membrane (MF vs. UF) were evaluated. The summary of the results obtained from the study is described below;

1. In the case of membrane filtration without coagulation (Run A), the TMP showed a tendency of

continuous increase. Analysis of the membrane surface showed that membrane fouling was caused by organic and inorganic materials. It is thought that a pretreatment process is required to remove micro-particles which cause the membrane fouling.

2. With temporal variation of raw water quality parameters, coagulant control methods based on the SCV are more effective than the turbidity coagulation control method.
3. Analysis of supernatant obtained from the chemical cleaning of membrane used for coagulation/MF process (Runs A–C) showed that membrane fouling could be aggravated by un-reacted coagulant and coagulated flocs. It is observed that optimum coagulation control could reduce the membrane fouling.
4. The operation of a pilot plant with an UF membrane was more stable than that with MF. In addition, stable TMP is maintained despite the high permeate flux of 2.0 $m^3/m^2 \cdot day$ when coagulation dose is controlled by SCV.
5. Micro-flocculation or colloidal particles caused pore blocking and membrane fouling of the MF membrane, which were not easily removed during backwashing. Considering the specifications of the membrane applied to the water treatment process for river water, it is thought that the permeation efficiency of UF membranes is superior to that of MF membranes when coagulation is used as a pretreatment process.

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