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INHIBITION OF MEMBRANE FOULING FOR TREATMENT OF SURFACE WATER CONTAINING ALGAE: INVESTIGATION BASED ON THE LONG TERM PILOT-SCALE CERAMIC MF FILTRATION EXPERIMENT

HAMOWANIE FOULINGU MEMBRAN STOSOWANYCH PRZY
UZDATNIANIU WÓD POWIERZCHNIOWYCH ZANIECZYSZCZONYCH
GLONAMI: BADANIA OPARTE NA DŁUGOTERMINOWYM
EKSPERYMENCIE W SKALI PILOTOWEJ

Application of membrane filtration processes in large-scale water treatment facilities has expanded in recent years. In particular, they are increasingly used for surface water treatment (river and lake waters) rather than for groundwater treatment. Generally, coagulation is used to remove colloidal and suspended particles in surface water, but the effects of coagulation, which can be used as a pre-treatment process in membrane filtration, on membrane fouling are scarcely reported. In this study, we investigated the effect of coagulation as a pretreatment process of the ceramic membrane filtration system, with the focus being concentrated on the effect of Gt value of coagulation through pilot-scale ceramic MF filtration experiments. Moreover, also through pilot-scale ceramic MF filtration experiments, we investigated the effects of chemically enhanced backwashing (CEB) and adsorption with submicron powdered activated carbon (S-PAC) on its performance by focusing on the removal for dissolved substances (including odors and THM precursor) as well.

1. Introduction

Control of membrane fouling in the course of filtration is of substantial importance in the application of membrane filtration for surface water treatment. Some studies have indicated that coagulation is an effective pretreatment process that can alleviate membrane fouling and at the same time remove a certain percentage of humic substances [1-3]. However, there are also several studies that found that coagulation pretreatment worsened membrane fouling [4-6]. It has been reported that using coagulation conditions appropriate for the conventional chemical precipitation process could not lead to optimum performance for membrane filtration [7]. Based on the results of a series of bench-scale membrane (pore size 0.1 μm) filtration experiments conducted using water containing commercially available humic acid or organic matter isolated from surface water and Polyaluminum chloride (PACl), Kimura *et al.* [8] found that the conditions of coagulation had great impacts on the degree of physically irreversible fouling: acidic coagulation conditions improved the quality of treated water but generally caused greater physically irreversible fouling than coagulation under neutral or alkaline conditions, and the dosage of coagulant affected the extent of membrane fouling, with a higher dosage revealing more severe irreversible fouling. The difference in the physicochemical characteristics of flocs, such as the sizes and the affinity with membrane, formed under different coagulation conditions is a likely reason behind the differences in the extent of membrane fouling. In regard of floc sizes and its affinity with membrane, in addition to the coagulant dose and pH, the mixing strength and the mixing time are also important controlling factors. However, little is known on how these factors affect membrane fouling when coagulation is used as the pretreatment process of membrane filtration and pilot scale investigations are very scarce.

On the other hand, some membrane filtration studies have also reported that membrane fouling can be controlled by pre-chlorination. However, pre-chlorination has the disadvantages relating to its capability to form hazardous chlorinated byproducts, such as the formation of trihalomethanes (THM). For this reason, other alternative oxidation process that can alleviate membrane fouling without sacrificing the filtered water quality is expected, together with some advanced process that can remove trace organic pollutants, such as odors associated with algae.

In this study, two types of ceramic membrane filtration experiments were conducted for objectives of (1) investigation of the effects of coagulation as a pretreatment process by focusing on the effects of Gt value for coagulation and (2) investigation of the effects of chemically enhanced backwashing (CEB) and adsorption with submicron powdered activated carbon (S-PAC) [9] on the performance of ceramic membrane filtration by focusing also on the removal for dissolved organic substances (including odors and THM precursor).

2. Materials and methods

2.1. Raw water

For the first objective of this study, Kiso River water that entered a water purification plant with the conventional rapid sand filtration system was used as the raw water for the experiments. The quality of the raw water during this study is summarized in **Table 1**. The quality of the river water is generally good. During the period of this study, the median values of turbidity, color and DOC were 3.9 NTU, 2.3 degree, 1.0 mg/L, respectively. The maximum values of turbidity, color and DOC, reaching 68.0 (NTU), 11.0 degree and 2.7 mg/L, respectively, were measured only after a heavy rainfall.

Tab. 1. Kiso River water quality during the experiments for the 1st objective of this study

| | Unit | Maximum | Minimum | Average | Median | Number of samples |
|------------|--------|---------|---------|---------|--------|-------------------|
| pH | — | 8.3 | 6.5 | 7.2 | 7.1 | 90 |
| Turbidity | mg/L | 68.0 | 0.5 | 5.8 | 3.9 | 95 |
| Color | degree | 11.0 | 0.6 | 2.6 | 2.3 | 93 |
| Total Fe | mg/L | 1.40 | 0.02 | 0.15 | 0.11 | 90 |
| Soluble Fe | mg/L | 0.18 | <0.01 | 0.03 | 0.02 | 89 |
| Total Mn | mg/L | 0.080 | 0.004 | 0.017 | 0.014 | 90 |
| Soluble Mn | mg/L | 0.014 | 0.001 | 0.003 | 0.002 | 89 |
| Total Al | mg/L | 1.50 | 0.04 | 0.17 | 0.12 | 95 |
| Soluble Al | mg/L | 0.26 | 0.01 | 0.03 | 0.02 | 91 |
| TOC | mg/L | 4.2 | 0.6 | 1.9 | 2.0 | 93 |
| DOC | mg/L | 2.7 | 0.5 | 1.1 | 1.0 | 88 |
| UV260 | 1/5cm | 0.446 | 0.058 | 0.141 | 0.130 | 94 |

For the second objective of this study, Wani River water of Kasumigaura (a lake) that entered another water purification plant in Japan was used as the raw water for the experiments. The quality of the raw water during this study is summarized in **Table 2**. Compared to the water of Kiso River, the water of Wani River is obviously worse. The median values of turbidity, color and DOC of the water of Wani River were 15.7 NTU, 6.5 degree, 3.8 mg/L, respectively, which were approximately three times as high as those of the water of the Kiso River. In addition, the higher content of algae throughout a year is also a feature of the water quality of the lake water.

Tab. 2. Wani River water quality during the experiments for the 2nd objective of this study

| | Unit | Feb 2013 - Feb 2014 | | | | Number of samples |
|-------------------------------------|-----------|---------------------|---------|---------|--------|-------------------|
| | | Maximum | Minimum | Average | Median | |
| pH | — | 9.0 | 7.8 | 8.2 | 8.2 | 12 |
| Turbidity | NTU | 26.7 | 8.4 | 15.7 | 15.5 | 12 |
| Color | degree | 9 | 3 | 6.5 | 6.5 | 12 |
| Total Fe | mg/L | 0.63 | 0.12 | 0.29 | 0.26 | 12 |
| Soluble Fe | mg/L | 0.050 | 0.010 | 0.010 | 0.020 | 12 |
| Total Mn | mg/L | 0.160 | 0.043 | 0.089 | 0.029 | 12 |
| Soluble Mn | mg/L | 0.018 | 0.006 | 0.010 | 0.009 | 12 |
| Total Al | mg/L | 0.53 | 0.09 | 0.24 | 0.22 | 12 |
| Soluble Al | mg/L | 0.050 | 0.020 | 0.028 | 0.030 | 12 |
| TOC | mg/L | 4.4 | 3.1 | 3.8 | 3.8 | 12 |
| DOC | mg/L | 3.8 | 2.7 | 3.4 | 3.4 | 12 |
| UV260 | 1/ 5cm | 0.489 | 0.316 | 0.406 | 0.387 | 12 |
| Trihalomethanes formation potential | mg/L | 0.154 | 0.068 | 0.101 | 0.094 | 40 |
| Algae count ¹ | Counts/mL | 58810 | 5770 | 32310 | 30930 | 40 |

1: blue green algae + diatom + green algae

2.2. Membrane

Ceramic membrane having a nominal pore size of 0.1 μm was used. The detailed information on the specifications of this membrane is shown in **Table 3**.

Tab. 3. Membrane specifications

| Parameter | Ceramic MF |
|-----------------------|-----------------------------|
| Nominal pore size | 0.1 μm |
| Length / Diameter | 1m / 0.03m |
| Channels per membrane | 55 |
| Channel diameter | 2.5 mm |
| Membrane area | 0.43 m ² |
| Pure water flux | 45 m/d (at 25°C, 100kPa) |

2.3. Experiment units and conditions

2.3.1. Experiments for investigation of the effects of pre-coagulation

The configuration of one of the three identical experiment units operated in parallel for investigation of the effect of pre-coagulation (the 1st objective of this study) is shown in **Figure 1**. Each unit comprised of five mixing chambers and a ceramic module inserted with the membrane described in **Table 3**. On the left side wall of each chamber, five openings were designed at different vertical levels. By selecting the opening for use, the water level in each chamber can be changed, which allowed more flexible adjustment of the mixing time for the whole coagulation system, in addition to the adjustment based on the number of chambers used among the total five installed. Raw water of Kiso River after coagulation was pumped to the membrane module under a constant flow rate.

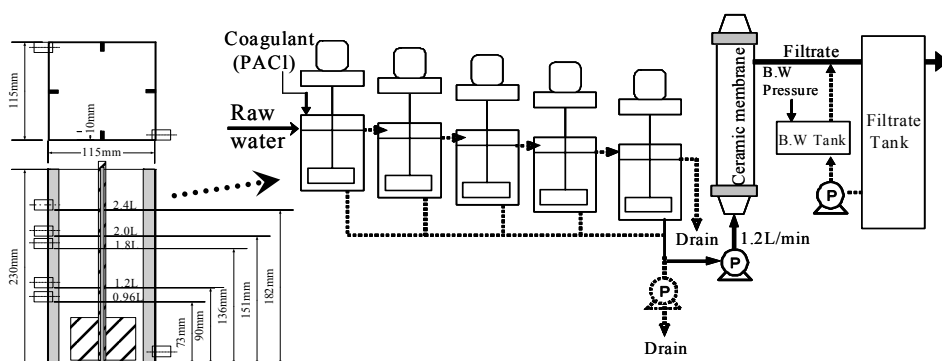


Fig. 1. Experiment unit for investigation of the effects of pre-coagulation

Polyaluminum chloride (PACl; basicity 54%, Al_2O_3 10.0%) was used as the coagulant and was added to the first chamber with a constant dosage of 20 mg/L for all filtration runs. The mixing strength, described by G value (s^{-1}), was changed by adjusting the rotation speed of the mixing blade installed in each mixing chamber. The mixing time t (sec) was varied by changing the number of mixing chambers and the water level of each chamber used. Membrane filtration was terminated before the transmembrane pressure (TMP) reached about 100 kPa. The increasing rate of TMP (kPa/d) was estimated based on the initial TMP after water backwashing and the TMP increasing profile with the least-square analysis method. Water pH was controlled by adding diluted sulfuric acid to raw water to maintain pH after addition of PACl at 6.8 [10]. Chemical backwashing was not used throughout this study.

The general experiment conditions are summarized in **Table 4**, and the specific mixing conditions of coagulation for all experiment runs are listed in **Table 5**. Totally 18 experiment runs (Run 1 - Run 18) were conducted in order over the whole study period (2006/8/25 - 2008/4/28). The average operation time for each run was about 30 days. In most cases, each run was further consisted of three series runs (Series A - Series C) operated in parallel for comparison of the effect of the mixing strength and the mixing time t within the ranges of $G = 30\text{-}1375 \text{ s}^{-1}$ and $t = 1.5\text{-}600 \text{ sec}$. Different combinations of

G and t within the ranges were selected, which enabled generation of membrane filtration results related to the Gt values in the broader range of 2060-180,000. Experiment runs with very short mixing time but very strong mixing strength, for instance the run at $t=2.5$ sec and $G=1375$ s⁻¹ (Run 4 / Series B) could probably show the membrane performance when coagulation was insufficient to have substances causing irreversible membrane fouling, for instance, humic molecules, be captured onto/into flocs that can be rejected by membrane filtration. On the other hand, experiment runs with longer mixing time but weaker mixing strength, for instance, the run at $t=600$ sec and $G=30$ s⁻¹ (Run 16 / Series B) may generate information for the condition when irreversible fouling causing substances are captured onto/into probably fragile and less dense flocs containing more aluminum originated from the added coagulant.

Tab. 4. Experiment conditions for investigation of the effects of pre-coagulation

| | |
|---------------------------------|---|
| Coagulant dose | 20 mg/L (as PAC I) |
| Coagulation tank pH | 6.8 (controlled) |
| G value | 30, 100, 110, 300, 400, 500, 600, 1375 [s ⁻¹] |
| Rotation speed | 40 - 500 [rpm] |
| Mixing time, t | 1.5, 2.5, 30, 40, 45, 48, 60, 100, 300, 600 [s] |
| Gt value [-] | 2060, 3440, 9000, 15000, 18000, 28800, 30000, 40000, 50000, 55000, 61875, 66000, 82500, 90000, 123750, 180000 |
| Membrane filtration flux | 4.0 [m ³ /m ² /d] |
| Water backwashing time interval | 180 [min] |
| Water backwashing pressure | Backwashing: 500 [kPa], and Air blow: 200 [kPa] |

For all runs, Series A that was operated under the same mixing condition ($G=110$ s⁻¹, $t=600$ sec) was included. This series was used as the reference for comparing the effect of the mixing conditions since the quality of raw water from Kiso River fluctuated with time throughout the study period. The mixing condition for Series A was determined based on the result of a bench scale membrane filtration experiment for controlling of membrane filtration resistance by coagulation. The G value was calculated using the following equation [11]:

$$G = \sqrt{\frac{\rho \cdot C \sum i (a_i v_i^3)}{2\mu V}} \quad (1)$$

ρ : water density (=1.0×10³ kg/m³, 20°C)

C : coefficient of resistance of mixing blade (=1.5)

a_i : area of the mixing blade i , which is perpendicular to rotating direction (m²)

v_i : average speed of the mixing blade i (m/s)

μ : water viscosity (=1.0×10⁻³ kg/m·s, 20°C)

V : mixed water volume (m³)

2.3.2. Experiments for investigation of the effects of chemical enhanced backwashing (CEB) and adsorption with submicron powdered activated carbon (S-PAC)

The experiments were conducted by installing three experimental systems (A, B and C) shown in **Figure 2** in a water purification plant that uses the Wan River water as its source water. The experiment conditions are shown in **Table 6**.

The coagulant dose was controlled at 60 mg/L as PACI and the water backwashing

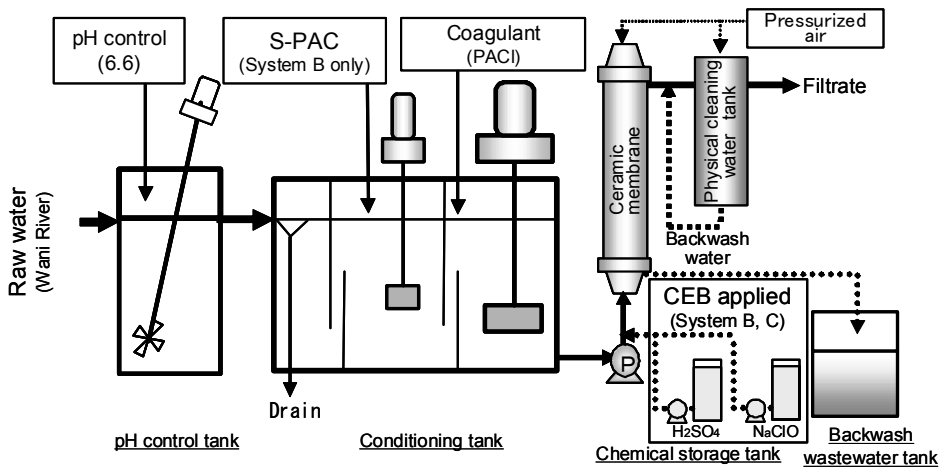


Fig. 2. Experiment system for investigation of the effects of CEB and S-PAC adsorption

Tab. 5. Transmembrane pressure (TMP) development for all filtration runs

| Run No. | Series No. | G value [s ⁻¹] | Mixing time, t [sec] | Gt value [-] | TMP increase rate [kPa/d] | Relative TMP increase rate [-] [*] | Run period |
|---------|------------|----------------------------|----------------------|--------------|---------------------------|---|------------------|
| 1 | A | 110 | 600 | 66000 | 1.01 | | 2006/8/25-9/25 |
| 1 | C | 300 | 600 | 180000 | 1.32 | 1.31 | 2006/8/25-9/25 |
| 2 | A | 110 | 600 | 66000 | 2.53 | | 2006/10/6-10/26 |
| 2 | B | 300 | 300 | 90000 | 2.75 | 1.10 | 2006/10/6-10/26 |
| 2 | C | 1375 | 90 | 123750 | 3.17 | 1.25 | 2006/10/6-10/26 |
| 3 | A | 110 | 600 | 66000 | 2.53 | | 2006/10/26-11/30 |
| 3 | C | 1375 | 40 | 55000 | 3.33 | 1.32 | 2006/10/26-11/30 |
| 4 | A | 110 | 600 | 66000 | 1.43 | | 2006/11/30-12/21 |
| 4 | B | 1375 | 2.5 | 3438 | 1.98 | 1.38 | 2006/11/30-12/21 |
| 4 | C | 1375 | 60 | 82500 | 1.14 | 0.80 | 2006/11/30-12/21 |
| 5 | A | 110 | 600 | 66000 | 1.43 | | 2007/1/12-2/7 |
| 5 | B | 1375 | 60 | 82500 | 1.14 | 0.80 | 2007/1/12-2/7 |
| 5 | C | 1375 | 48 | 66000 | 1.43 | 1.00 | 2007/1/12-2/7 |
| 6 | A | 110 | 600 | 66000 | 1.74 | | 2007/2/7-3/5 |
| 6 | B | 1375 | 45 | 61875 | 2.55 | 1.47 | 2007/2/7-3/5 |
| 6 | C | 1375 | 48 | 66000 | 1.78 | 1.02 | 2007/2/7-3/5 |
| 7 | A | 110 | 600 | 66000 | 1.16 | | 2007/3/9-4/12 |
| 7 | B | 1375 | 48 | 66000 | 1.10 | 0.95 | 2007/3/9-4/12 |
| 8 | A | 110 | 600 | 66000 | 3.70 | | 2007/4/12-5/7 |
| 8 | B | 300 | 30 | 9000 | 4.80 | 1.30 | 2007/4/12-5/7 |
| 8 | C | 600 | 48 | 28800 | 2.90 | 0.78 | 2007/4/12-5/7 |
| 9 | A | 110 | 600 | 66000 | 1.06 | | 2007/5/16-6/5 |
| 9 | B | 1375 | 1.5 | 2060 | 2.24 | 2.11 | 2007/5/16-6/5 |
| 9 | C | 1375 | 60 | 82500 | 1.10 | 1.04 | 2007/5/16-6/5 |
| 10 | A | 110 | 600 | 66000 | 3.60 | | 2007/6/22-7/25 |
| 10 | B | 600 | 30 | 18000 | 2.43 | 0.68 | 2007/6/22-7/25 |
| 11 | A | 110 | 600 | 66000 | 2.90 | | 2007/7/27-8/23 |
| 11 | C | 500 | 30 | 15000 | 3.50 | 1.21 | 2007/7/27-8/23 |
| 12 | A | 110 | 600 | 66000 | 3.40 | | 2007/8/23-9/14 |
| 12 | B | 500 | 30 | 15000 | 3.80 | 1.14 | 2007/8/23-9/14 |
| 12 | C | 100 | 500 | 50000 | 3.05 | 0.90 | 2007/8/23-9/14 |
| 13 | A | 110 | 600 | 66000 | 1.64 | | 2007/9/14-10/17 |
| 13 | B | 300 | 100 | 30000 | 1.23 | 0.75 | 2007/9/14-10/17 |
| 13 | C | 500 | 60 | 30000 | 1.30 | 0.79 | 2007/9/14-10/17 |
| 14 | A | 110 | 600 | 66000 | 3.07 | | 2007/10/17-11/13 |
| 14 | B | 100 | 500 | 50000 | 2.76 | 0.90 | 2007/10/17-11/13 |
| 14 | C | 500 | 100 | 50000 | 1.68 | 0.55 | 2007/10/17-11/13 |
| 15 | A | 110 | 600 | 66000 | 1.51 | | 2007/11/26-12/26 |
| 15 | B | 300 | 600 | 180000 | 1.37 | 0.91 | 2007/11/26-12/26 |
| 15 | C | 400 | 100 | 40000 | 1.08 | 0.72 | 2007/11/26-12/26 |
| 16 | A | 110 | 600 | 66000 | 0.89 | | 2008/1/8-2/8 |
| 16 | B | 30 | 600 | 18000 | 0.77 | 0.87 | 2008/1/8-2/8 |
| 16 | C | 400 | 100 | 40000 | 0.86 | 0.97 | 2008/1/8-2/8 |
| 17 | A | 110 | 600 | 66000 | 2.01 | | 2008/2/12-3/3 |
| 17 | B | 30 | 600 | 18000 | 1.65 | 0.82 | 2008/2/12-3/3 |
| 17 | C | 400 | 100 | 40000 | 1.67 | 0.83 | 2008/2/12-3/3 |
| 18 | A | 110 | 600 | 66000 | 2.92 | | 2008/4/1-4/28 |
| 18 | C | 400 | 100 | 40000 | 2.80 | 0.96 | 2008/4/1-4/28 |

* Relative TMP increase rate is the rate difference when the TMP increase rate of the reference series A in each filtration run is assumed as "1", which equals to the ratio of the TMP increase rate of series B and/or C to that of the reference series A in each filtration run.

Tab. 6. Experiment conditions for investigation of the effects of CEB and S-PAC adsorption

| Membrane filtration conditions | System A (control 1) | System B (experimental) | System C (control2) |
|---|--|-------------------------------------|------------------------|
| G value | 339 [s ⁻¹] | | |
| Mixing time, <i>t</i> | 120 [s] | | |
| Coagulant dose | 60 [mg/L (as PACl)] | | |
| Membrane filtration flux | 4 [m ³ /m ² /d] | | |
| Water backwashing time interval | 60 [min] | | |
| Water backwashing pressure | Backwash:500 [kPa], and Air blow:200 [kPa] | | |
| Pre-chlorination dose [mg/L] | 3 - 5 | 0 | |
| Submicron powdered activated carbon (S-PAC) dose [mg-dry-C/L] | 0 | 10, 20, 25 | 0 |
| Chemical enhanced backwashing (CEB) | Not applicable | 2 - 14 times / week, or, Every time | |

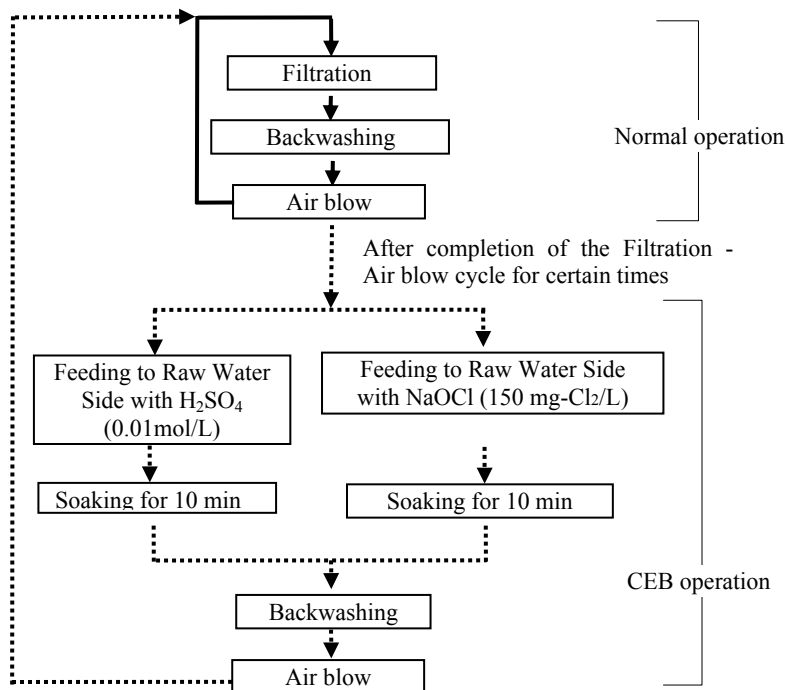


Fig. 3. The process for chemical enhanced backwashing (CEB)

interval was 60 minutes. The filtration flux was set at $4 \text{ m}^3 / \text{m}^2 / \text{d}$. The coagulation Gt value during this long-term filtration test was controlled at 40,500 ($G=339 \text{ s}^{-1}$ and $t=120 \text{ sec}$) based on the results of pre-coagulation. For comparison, pre-chlorination experiments were conducted using the experimental system A, for which, the pre-chlorine dose was adjusted at around 4 mg/L to ensure the residual chlorine concentration of 0.5 mg/L could be maintained in the filtrate. The process of CEB is depicted in **Figure 3**. In brief, after membrane filtration for a designated time length, sulfuric acid or the aqueous solution of sodium hypochlorite was introduced to the raw water side of membrane filtration module. After the module was immersed in the solution chemical solution for 10 minutes, backwashing and air blow were conducted to prevent mixing of the chemicals into the filtrate side. The application frequency for CEB was 2 - 14 times a week.

3. Results and discussion

3.1. Effect of mixing conditions of coagulation on TMP development

The values of the increasing rate of TMP for all filtration runs under different combinations of the mixing strength G and the mixing time t are summarized in **Table 4**. For the reference runs of Series A, the TMP increase rate differed obviously, with the smallest and the largest rate values being 0.89 and 3.7 kPa/day for Run 16 / Series A and Run 8 / Series A, respectively. This difference was considered to be mainly caused by fluctuations in the raw water quality as could be seen from **Table 1**.

To evaluate the impact of mixing strength and mixing time on the development of

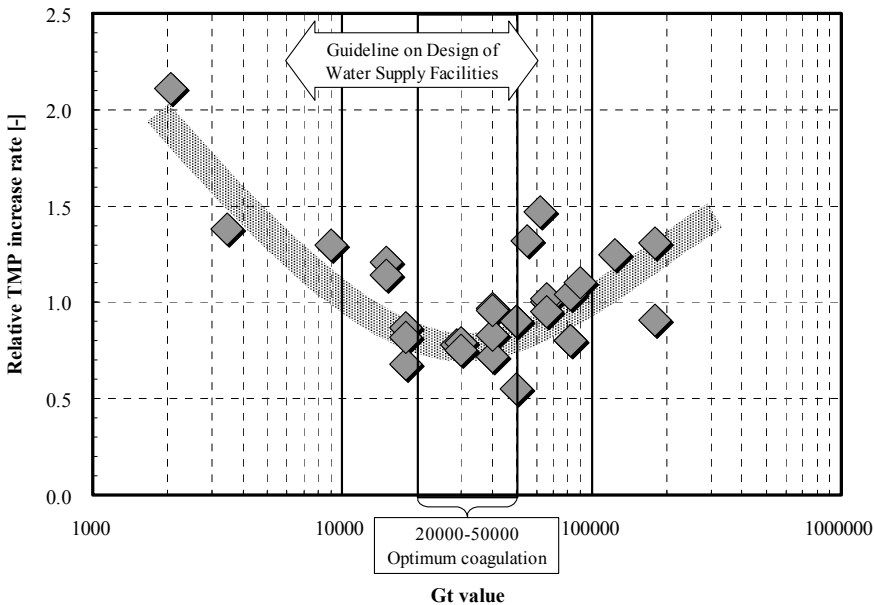


Fig. 4. Optimum coagulation range for membrane filtration

TMP, the relative TMP increase rate, a parameter reflecting the relative increase of TMP was adopted. This parameter was computed by dividing the TMP increase rate of Series B and/or C by that of the reference run Series A. The computed values for all runs are also summarized in **Table 5**, and are plotted in **Figure 4** to show the relationship between TMP development and Gt value. In the Guideline on Design of Water Supply Facilities [11], a rapid mixing G value range of 100-200 s^{-1} and a mixing time t range of 60-300 sec are suggested [11]. This corresponds to a Gt range in 6,000-60,000, which is also revealed in **Figure 4**. As is evident from this figure, an optimum coagulation range that could suppress the TMP increase rate seemed to be existent in the Gt range of about 20,000-50,000. Small Gt values (below 10,000) were found ineffective in suppressing the irreversible TMP development. This could be easily seen from the results with the runs of Run 4 / Series B ($G=1375 s^{-1}$, $t=2.5$ sec) and the Run 9 /Series B ($G=1375 s^{-1}$, $t=1.5$ sec). As shown in Table 5, for these two runs, even if the largest mixing strength ($G=1375 s^{-1}$) was adopted, since the mixing time was too short (2.5 and 1.5 sec), the resulting relative TMP increase rate was comparatively larger: 1.38 and 2.11, respectively. Insufficient coagulation that probably failed to capture irreversible fouling-causing substances, such as larger humic molecules and fine turbid particles that could form flocs with other coexisting impurities under optimum coagulation conditions, was a likely reason behind. Li *et al.* studied the characteristics of micro-flocs of peaty water NOM coagulated by PACl and found that at the G value of 660 s^{-1} , the distribution of micro flocs (0.1-5.0 μm) could reach steady stage after rapid mixing for 30 sec and increasing the mixing time till 300 sec increased the captured NOM content (i.e., the ratios of UV260/Al and DOC/Al) in flocs with sizes above 5 μm , the major floc fraction in the final solution after coagulation at pH 5.5 [13].

It is also interesting to see that for the run (Run 16 / Series B), even if the G value was very small ($G=30 s^{-1}$), the smallest investigated in this study, a mixing time of 600 sec led to a lower relative TMP increase rate (the rate value of 0.87). The Gt value under this condition was 18,000, close to the optimum Gt range mentioned above. Increasing the mixing time probably enhanced the capture of irreversible fouling-causing humic molecules (by sorption, for instance) onto /into fragile and less dense aluminum flocs formed under weaker mixing strength.

Membrane fouling involves complicated mechanisms. This is particularly true when membrane filtration is applied to treat surface water that contain a polydisperse mixture of impurities, many of which are not yet clarified. The optimum range obtained in this study may serve as an important reference for determining the coagulation conditions for practical application of membrane filtration to surface water; however, the mechanisms behind have to be clarified in coming studies through combined investigation of the floc characteristics and the physicochemical features of non-coagulated constituents remaining in water under different coagulation conditions. For this, bench scale membrane filtration experiments will be conducted along with pilot scale studies.

3.2. Verification of the optimum Gt range of coagulation

Verification of the optimum Gt range of coagulation will be given and discussed from the following aspects.

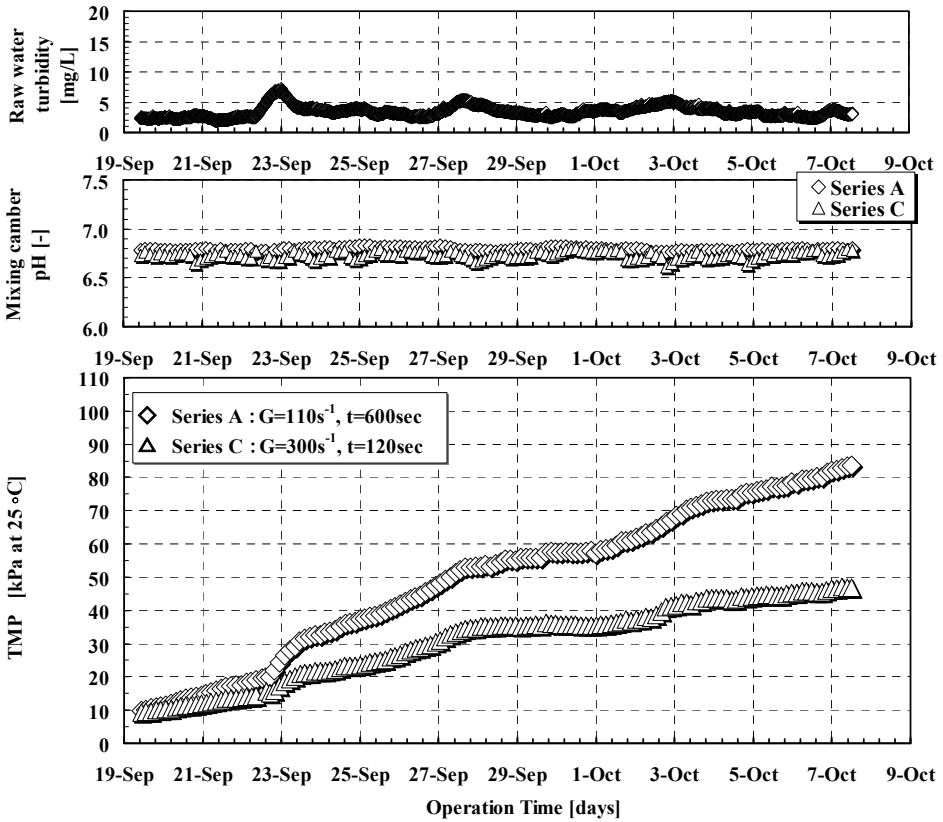


Fig. 5. Membrane filtration performance in the optimum coagulation range

3.2.1 Membrane performance and filtrate quality

Figure 5 shows the profiles of raw water turbidity, mixing chamber pH and TMP with running time for the run of Series A with the Gt value of 66,000 ($G=110\text{ s}^{-1}$, $t=600$ sec) and the run of Series C with the Gt value of 36,000 ($G=300\text{ s}^{-1}$, $t=120$ sec). For both runs, the filtration flux was $4\text{ m}^3/\text{m}^2/\text{day}$, the physical water cleaning interval was 3 hours and the coagulant dosage was 20 mg/L as PACl.

As is evident from this figure, TMP of the Series C with the Gt value of 36,000 ($G=300\text{ s}^{-1}$, $t=120$ sec) was always lower than the Series A with the Gt value of 66,000 ($G=110\text{ s}^{-1}$, $t=600$ sec). The increasing extent of TMP during the time of raw water quality deteriorations due to rainfall (for instance, around Sep. 23), as could be seen from the time profile of the raw water turbidity, was also smaller. After running for three weeks, the difference in TMP was nearly two-folds.

The analytical results of water quality in the course of filtration are shown in Table 7. For TOC, the values were confirmed to be of the same level in filtrate of both series runs. However, for the index of UV260 and the index of total residual Al, the filtrate of the Series C with the Gt value of 36,000 ($G=300\text{ s}^{-1}$, $t=120$ sec) revealed lower values than the Series A with the Gt value of 66,000 ($G=110\text{ s}^{-1}$, $t=600$ sec). Given the mixing time

of coagulation is sufficient, a mixing strength that could lead to a Gt value falling in the optimum Gt range of about 20,000-50,000 seems to be effective to alleviate the development of TMP. Enhanced capture of humic molecules detected by UV260 and aluminum onto and/or into coagulated flocs, and thus reduced levels of these fouling causing substances in the water after coagulation, as could be estimated from the filtrate water quality, might be the reason behind the better performance with the run of Series C. For verification, as mentioned earlier that further investigation of the physicochemical characteristics of flocs and the remaining constituents not captured by flocs under different mixing conditions of coagulation is necessary.

Thomas *et al.* [13] reported that the residual aluminum concentration decreased with increases of the mixing time. Even if our result did not support their result, the difference in the mixing strength G might be a reason and it is evident that the coagulation condition affects the performance of membrane filtration.

Tab. 7. Water quality before and after filtration (Samples of 2008/09/21)

| | unit | Raw water | Series A filtrate | Series C filtrate |
|-----------------|-----------------|-----------|-------------------|-------------------|
| pH | - | 7.2 | 7.1 | 7.0 |
| Temperature | °C | 15.0 | 14.0 | 14.0 |
| Turbidity | mg/L | 9.0 | <0.1 | <0.1 |
| Color | degree | 2.6 | 0.9 | 0.7 |
| Total Fe | mg/L | 0.12 | <0.01 | <0.01 |
| Soluble Fe | mg/L | 0.03 | - | - |
| Total Mn | mg/L | 0.012 | 0.002 | 0.002 |
| Soluble Mn | mg/L | 0.001 | - | - |
| Total Al | mg/L | 0.10 | 0.03 | 0.01 |
| Soluble Al | mg/L | 0.01 | - | - |
| TOC | mg/L | 2.4 | 0.7 | 0.7 |
| DOC | mg/L | 1.1 | - | - |
| UV260 | 1/5cm | 0.151 | 0.063 | 0.060 |
| G value | s ⁻¹ | - | 110 | 300 |
| Mixing time t | sec | - | 600 | 120 |

3.2.2. Responses of membrane filtration to variation of water pH

The variations of pH and turbidity in the water to be treated and the TMP development with time are shown in **Figure 6**. The coagulant dose was controlled identical at 20 mg/L (as PACl). For most time of the experiment, pH of the raw water was controlled at 7.1. A running period without pH adjustment was allowed for about one week. During this week, pH of the raw water was slightly higher, with its values being about 7.5. The water backwashing interval and the flux were maintained not changed: 3 hours and 4 m³/m²/day, respectively.

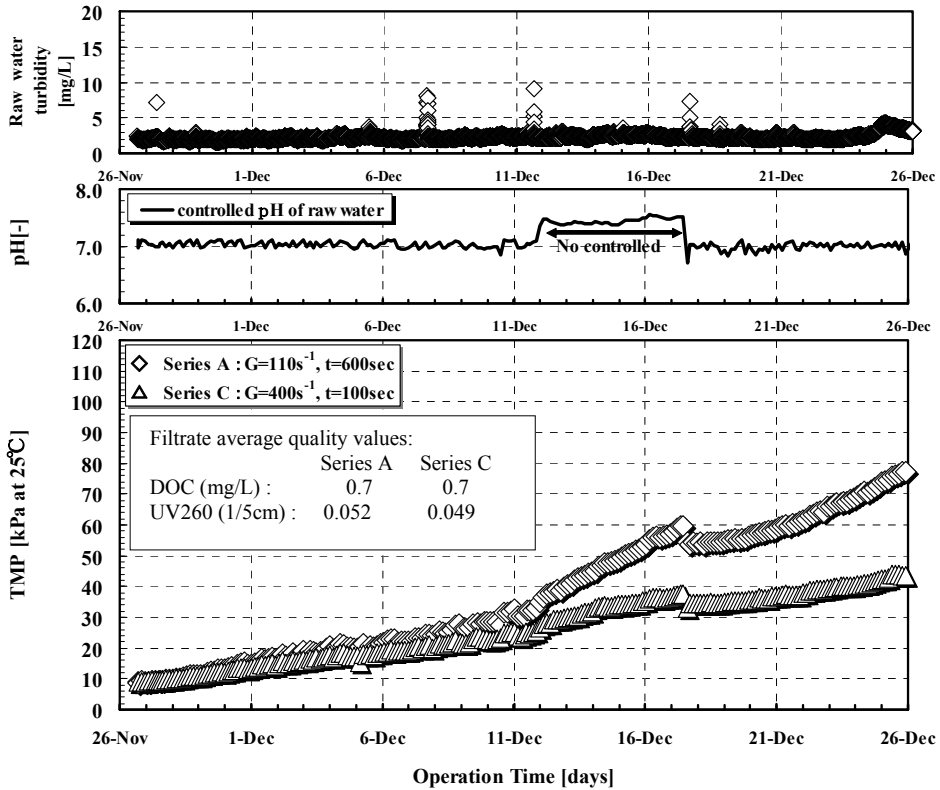


Fig. 6. Membrane filtration performance under varied pH

It can be seen from this figure that TMP in Series A with the Gt value of 66,000 ($G=110\text{ s}^{-1}$, $t=600\text{ sec}$) suddenly increased for the period from December 11 to December 17 when pH was not controlled. However, for Series C with the Gt value of 40,000 ($G=400\text{ s}^{-1}$, $t=100\text{ sec}$) that fell in the optimum coagulation Gt range shown in **Figure 4** suffered less changes in the increasing pace of TMP. Lowering water pH within a certain range was found to be able to enhance the content of humic molecules captured into micro flocs coagulated by either Al or Fe coagulant [13]. Taking into consideration this finding, it is reasonable to consider that the observed increases in the trend of TMP during the period of higher pH were caused partly, if not totally, by those humic molecules that were not coagulated under higher pH; and coagulation under the condition with a suitable Gt probably reduced the remaining level of such fouling causing molecules (as could be estimated from the lower average UV260 value in the filtrate shown in Fig. 6). The likely differences in the physicochemical characteristics of flocs and residual Al species in the water after coagulation under these two different conditions may also contribute to the observed difference, for which investigation is desired and will be do in coming studies. The results shown in this figure further suggest the adversary influence of raw water quality variation on the development of TMP can be minimized by optimizing the coagulation conditions during membrane filtration.

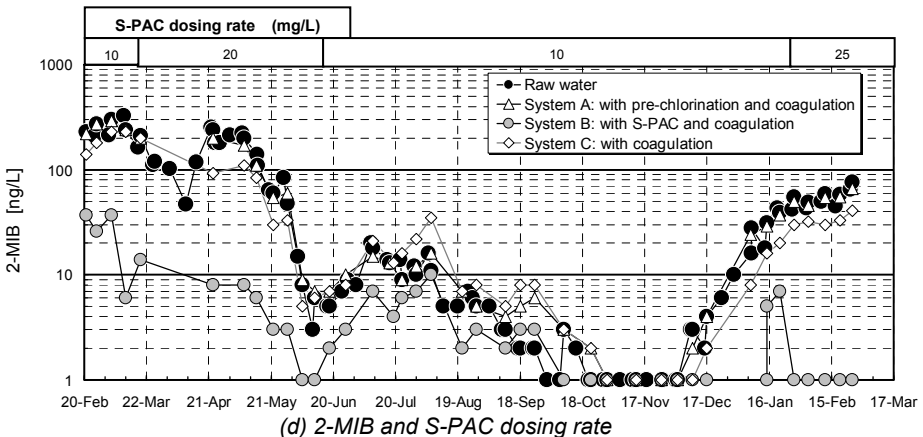
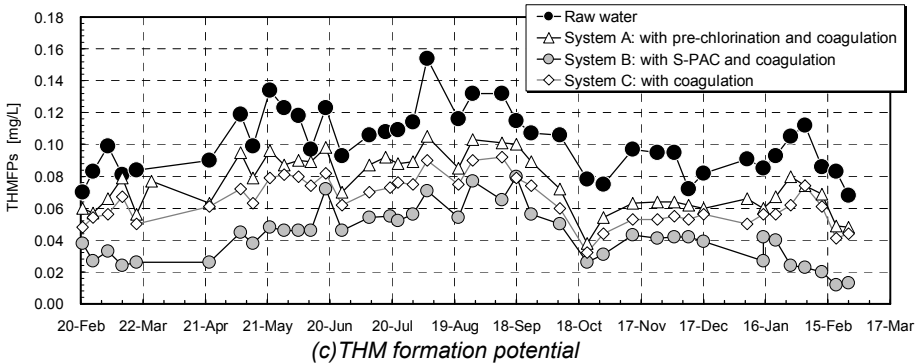
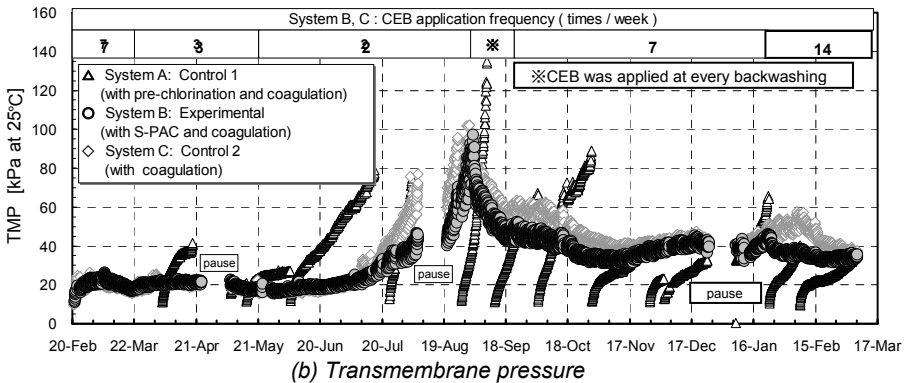
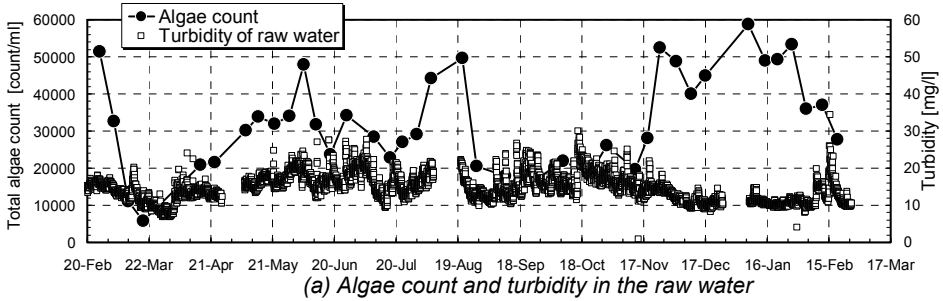


Fig. 7. Changes of raw water quality (a), and the effects of CEB and adsorption with S-PAC on transmembrane pressure (b), and THMFPs (c) and 2-MIB in filtrate (d).

3.3. Effects of CEB and adsorption with S-PAC on membrane filtration performance and filtrate water quality

Figure 7(a) shows the algae count of raw water during the period from February, 2013 to March, 2014 while **Figure 7(b)** shows the turbidity and the transmembrane pressure (TMP) after water backwashing. The TMP of System A (as shown with \triangle in the figure) increased substantially partly due to the membrane filtration flux of $4 \text{ m}^3/\text{m}^2/\text{d}$, so that the filtration period before applying chemical cleaning under the TMP condition of 80 - 100 kPa was 14 to 50 days. On the other hand, the TMP (as shown with \circ in the figure) of System B, in which CEB was applied, was stable immediately after start of operation, and the CEB frequency was gradually decreased since February 20. After that, the TMP reached 100 kPa on September 2 after consistent operation for 6 months. After this time point, the CEB was implemented at each time of backwashing (with an interval of 60 min) for about 14 days. This practice enabled the recovery of the TMP. As a result, the TMP decreased, indicating that membrane performance can be recovered while continuing its filtration operation.

Figure 7(c) and **Figure 7(d)** show the concentrations of THMFPS and the odor-causing compound (2-MIB) in the raw water and the filtrate during filtration. 2-MIB in raw water was extremely high during the January - May period and low during June - December. On the other hand, THMFPS showed a trend different from 2-MIB. It was low during January - May and increased in the period of June - December. 2-MIB in the filtrate was removed by the adsorption with S-PAC, with the removal efficiency being dependent on the dosage of the adsorbent. In the experiment the dosage of S-PAC was reduced from 20 mg/L to 10 mg/L during the period of June - November, following the decreases of 2-MIB in the raw water. This resulted in concentration increases of 2-MIB and THMFPSs in the filtrate. It is therefore considered necessary to keep the dosage of the adsorbent at about 20 mg/L if the odor-causing compound and THM precursors are to be removed in the stable manner.

4. Conclusions

Based on the results of long term pilot scale membrane filtration experiments with pre-coagulation to treat the Kiso River water, following conclusions can be drawn.

Coagulation under suitable mixing strength and time was important to control the development of irreversible membrane fouling when coagulation was used as a pretreatment process to treat surface water used as the water source of conventional water purification plants. Over the Gt ranges of 2060-180,000 investigated in this study from selected combinations of G in the range of $30\text{-}1375 \text{ s}^{-1}$ and the mixing time t of 1.5-2.5 sec, an optimum coagulation range that could suppress the increasing rate of TMP seemed to be existent in the range about 20,000-50,000. This range is narrower than the range of 6,000-60,000 suggested in the Guideline on Design of Water Supply Facilities [11] and can be used as a reference for facilitated determination of suitable pre-coagulation conditions. Gt values below 10,000 associated with larger values of the relative increase rate of TMP.

Pre-coagulation with optimum Gt values alleviated the influence of variations in raw water quality on the development of irreversible membrane fouling during filtration and

could reduce the coagulant dosage. For water sources with algae obstacles, application of chemical enhanced backwashing and powdered activated carbon adsorption in ceramic membrane filtration could enhance its performance, with the removals for dissolved organic compounds (odors, THM precursors, etc.) being also increased.

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