



Maximizing biopolymer removal by coagulation for mitigation of fouling in the following membrane process



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ABSTRACT

In recent studies dealing with membrane fouling in microfiltration (MF)/ultrafiltration (UF) processes for drinking water production, biopolymers, which are hydrophilic macromolecules, have been identified as the key foulants. However, removal of biopolymers from water has been rarely focused on in previous studies. In this study, removal of biopolymers by coagulation, which is the most common pretreatment for MF/UF in existing full-scale facilities, was investigated. Surface water samples from three rivers used as raw water for drinking water production were examined in this study. Sampling was repeated in different seasons to examine a wide range of water quality. It was demonstrated that dosages commonly used in existing facilities were insufficient to maximize biopolymer removal by coagulation conducted at neutral pH. It was shown that an extremely high dosage of the coagulant was necessary to maximize biopolymer removal. A new dosage index, aluminum (Al, mg/L)/biopolymer (Bp, $\mu\text{g/L}$), was used in this study. Al/Bp of about 20 was found to be necessary at pH 7 to almost maximize biopolymer removal by coagulation regardless of the water tested. Maximized biopolymer removal brought about least evolution of membrane fouling, which was separately verified in bench-scale filtration tests. Efficiency of coagulation for biopolymer removal varied depending on the season: samples collected in the cold season exhibited poor removal of biopolymers. Detailed examinations of biopolymers in each sample suggested that properties of the biopolymers changed seasonally.

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1. Introduction

Application of low-pressure membranes (microfiltration (MF) and ultrafiltration (UF)) to potable water production has drawn much attention due to their good performance. The safety of drinking water is improved by using membrane processes because pathogens such as *Cryptosporidium* can be completely removed by such processes. However, membrane fouling, which causes deterioration in membrane permeability, increases both the initial and operational costs of membrane processes and hinders the benefits gained from the technology. Mitigation of membrane fouling is necessary for widespread use of membrane processes in drinking water production. To do so, identification of agents that cause membrane fouling, which should lead to a detailed understanding of the phenomena of membrane fouling, is important.

Many previous studies showed that natural organic matter (NOM), a mixture of various types of high-molecular organic compounds, played important roles in the development of membrane fouling in application of MF/UF to drinking water production. In

early studies dealing with membrane fouling in MF/UF for drinking water treatment [1–3], hydrophobic humic substances, which generally account for a large fraction of NOM in surface water [4], were identified as major foulants. Recently, the importance of hydrophilic organic compounds in evolution of membrane fouling in MF/UF for drinking water production has been increasingly reported [5–10]. Analysis of the molecular weight distribution of dissolved organic matter revealed the presence of very large hydrophilic organic molecules, larger than hydrophobic humic substances, in natural water. These large hydrophilic organic molecules are mainly composed of polysaccharides and proteins and are called “biopolymers” [11]. Dimensions of biopolymers would be close to the sizes of micro-pores of MF/UF membranes, leading to plugging of the pores. It has also been suggested that hydroxyl functional groups that are abundant in polysaccharides formed hydrogen bonds with polymer materials used for membrane production, leading to membrane fouling [12].

In most full-scale membrane facilities, membranes are routinely cleaned by backwashing or air scouring. Despite the implementation of this routine physical cleaning, some constituents gradually accumulate on and in the membrane and eventually cause fouling during long-term operation. This type of fouling that develops even with routine physical cleaning is defined as irreversible fouling and

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should be focused on [5]. The authors demonstrated that the abovementioned biopolymers were the major players in evolution of irreversible fouling in MF of various surface water samples and that a conventional water quality index such as dissolved organic carbon (DOC) or ultraviolet (UV) absorption did not show a good correlation with membrane fouling [13,14].

In many full-scale MF/UF facilities, membrane processes are coupled with pretreatment processes. Coagulation is a major option as pretreatment combined with membrane processes [15]. Pre-coagulation removes a certain range of NOM [16] and also improves the quality of the permeate [17]. Conditions for pre-coagulation such as pH and dosage have huge impacts on fouling in the following membrane processes as well as the quality of treated water. Coagulation conditions that are regarded as “optimum” from a conventional view based on turbidity/color removal may not be identical to the conditions for prevention of membrane fouling. Many studies have shown that inappropriate coagulation conditions caused more severe membrane fouling [18–22].

Considering the abovementioned importance of biopolymers in evolution of fouling in MF/UF, optimum conditions for the pre-coagulation process should be determined on the basis of maximizing removal of biopolymers by coagulation. However, removal of biopolymers by coagulation has been overlooked, partly because it is believed that coagulation is not good for removal of hydrophilic organic matter [23,24]. Thus, in this study, we focused on removal of biopolymers by coagulation and examined the relationship between conditions in pre-coagulation and degrees of membrane fouling. Multiple surface water samples collected in different seasons were examined in this study.

2. Materials and methods

2.1. Water samples

Water samples were collected at drinking water treatment plants using surface water from different rivers. Toyohira River water was collected at Moiwa Water Purification Plant (Sapporo, Japan). Chitose River water was collected at Kamiebeta Water Purification Plant (Ebetsu, Japan). Yoichi River Water was collected at Yoichigawa Water Purification Plant (Yoichi, Japan). For Toyohira River water and Chitose River water, raw water without any treatment was transported to the university laboratory and examined. Full-scale MF membranes are used at Yoichigawa Water Purification Plant. Their source water (Yoichi River water) contains a certain level of manganese, which has been identified as a problematic foulant. Therefore, as a pretreatment, manganese is oxidized with chlorine and the oxidized manganese is removed by sand filtration. Effluent from the sand filter is used as feed water for the full-scale MF. In this study, for Yoichi River water, the effluent from the sand filter was transported to the university laboratory and examined. To quench residual chlorine, an equivalent amount of reducing agent (sodium bisulfite) was added upon arrival. To suppress microbial activity, sodium azide (2 mg/L) was added to all samples collected from the three sites upon arrival. Samples were kept in a refrigerator until the experiments. Experiments were conducted within 10 days after the samples had been collected. Samples were repeatedly collected from the three sites in different seasons. Table 1 shows the quality of the water examined in this study. Chitose River water contained larger amounts of organic matter than those in water from the other two rivers. Turbidity in Yoichi River water was low, reflecting the implementation of sand filtration as described above.

2.2. Jar tests

One day before each experiment, samples were taken out of the refrigerator and coagulation was performed under an

Table 1
General water quality of the tested water.

	TOC (mg/L)	Turbidity (TU)	Biopolymers ($\mu\text{g/L}$)	pH
Toyohira River (average)	2.7	1.7	46	7.2
July	0.8	0.6	70	7.4
September	1.3	1.9	39	7.1
October	5.2	3.5	37	6.7
January	3.4	0.6	37	7.4
Chitose River (average)	6.5	7.0	234	7.0
July	– ^a	1.3	370	6.8
August	6.2	26.8	473	6.7
September	2.2	4.2	141	7.3
November	8.0	3.6	127	6.7
December	10.4	2.6	198	7.1
January	5.6	3.2	97	7.2
Yoichi River (average)	1.7	0.3	70	6.9
August	1.5	0.7	130	6.9
September	1.0	0.1	85	6.9
November	– ^a	0.3	40	6.7
December	2.7	0.2	23	6.8

^a Not available due to instrumental malfunction.

air-conditioned room temperature (about 23 °C). Poly-aluminum chloride (PACl) was used as the coagulant in this study. Removal of biopolymers was examined in a series of jar tests with variable dosages and fixed pH of 7. Control of pH was carried out by using sodium hydroxide (Wako Pure Chemical, Tokyo, Japan) and chloric acid (Wako Pure Chemical, Tokyo, Japan). In existing conventional drinking water treatment facilities, coagulant dosage is often controlled on the basis of aluminum (Al, mg/L)/turbidity (T, Turbidity unit) ratio. In full-scale treatment facilities in Japan using the conventional treatment method (i.e., coagulation/sedimentation followed by sand filters), this Al/T ratio is usually controlled in the range between 0.05 and 0.2. As described later, this dosage of the coagulant commonly used in existing treatment facilities was insufficient to maximize the removal of biopolymers. A new dosage index, Al (mg/L)/biopolymer Bp (mg-C/L) ratio, was used in this study to investigate maximum removal of biopolymers by coagulation, and the effects of high dosages were examined. Jar tests were conducted with a jar tester (Miyamoto Riken Industry, Osaka, Japan) holding six 1-L beakers. Samples were mixed with coagulants with rapid mixing (G -value of 100 s^{-1}) for 2 min and were filtered by a polytetrafluoroethylene (PTFE) filter with 0.50- μm openings. The filtered samples were subject to analyses.

2.3. Bench-scale MF experiments

Tiny-scale membrane elements (surface area: 0.0128 m²) were assembled in the laboratory and used in the filtration tests. A commercially available hollow-fiber MF membrane made from PVDF polymer (Asahi Kasei, Tokyo, Japan) was used in this study. Nominal pore size of the membrane was 0.1 μm . A single new membrane element was immersed in the filtration tank in each experiment, and permeate flow rate was fixed at 62.5 LMH by using a peristaltic pump (EYELA, Tokyo, Japan). Membrane filtration was carried out in outside-in mode, and periodic backwashing (every 30 min for 30 s each time, at 94 LMH) was performed using a separate backwashing pump from the inside to the outside of the membrane. To prevent excessive accumulation of rejected particles in the filtration tank, the concentrated suspension was drained from the tank every 4 h. Trans-membrane pressure (TMP) was monitored with a digital pressure meter (Nagano Keiki, Tokyo, Japan). Each operation was continued for >20 h unless TMP reached 40 kPa.

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