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Hydraulically irreversible membrane fouling during coagulation–microfiltration and its control by using high-basidity polyaluminum chloride

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ABSTRACT

The extent of hydraulically irreversible membrane fouling in a coagulation–filtration system depends on several factors, including properties of the coagulant. Effects of polyaluminum chloride (PACl) coagulant properties, specifically basidity and sulfation, were investigated by conducting long-term direct filtration experiments. Elemental analysis determined Al and Si to be the major foulants, though the Si/Al ratios of the foulants differed from those of coagulated floc particles. While floc particle size depended on the concentrations of sulfate ions and polymeric species in the PACls, floc-size changes did not affect transmembrane pressure (TMP) buildup and thus did not affect irreversible fouling. Differences in PACl basidity, which affected the distribution of aluminum species, resulted in changes to the degree of irreversible fouling.

Pretreatment with high-basidity (71%) PACl was superior to pretreatment with normal-basidity (51%) PACl in reducing irreversible fouling and attenuating TMP buildup during filtration. Higher basicities resulted in less Al breakthrough and a decrease in the Si/Al ratio of the foulants. However, TMP buildup was the same for PACls with basicities of 71% and 90%; therefore, TMP buildup is not simply related to Al breakthrough and deposition. Increasing the basidity of PACls would be an effective way to reduce the amount of foulant deposited on the membrane by decreasing the amount of aluminum that passes through the membrane.

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1. Introduction

Coagulation, adsorption, and oxidation are widely used as pre-treatment processes for microfiltration (MF) in water purification to alleviate membrane fouling and enhance the removals of micropollutants and disinfection byproduct precursors [1]. In MF with ceramic membranes, coagulation–flocculation with polyaluminum chloride (PACl) is a successful pretreatment for removing soluble substances and reducing the decrease in membrane permeability during long-time operation [2]. This process is commonly used in full-scale water treatment. Coagulation pretreatment destabilizes and agglomerates the colloidal and particulate foulants, increasing their size and thereby mitigating pore constriction and blockage and the formation of a porous cake layer. Additionally, increased particle size reduces the specific cake resistance, according to the Carman–Kozeny relationship, and thus increases permeability. However, membrane fouling is not

completely avoided since aquatic colloids are not removed, which cause fouling by narrowing or blocking membrane pores, and substances retained on the membrane that form a gel or cake layer still contribute resistance.

The permeability of the cake layer formed from floc particles during coagulation has been extensively studied for dependence on floc size, strength, and fractal structure [3–5]. Coagulated flocs with a high fractal dimension have low compressibility, leading to low membrane permeability [6]. Other studies, however, found high compressibility in flocs related to a higher specific resistance of the cake layer [7,8]. Coagulated flocs with a high fractal dimension formed by PACl have a more compact structure than flocs formed by alum [9]. Therefore, the MF membrane permeability deteriorates more severely during PACl coagulation than during alum coagulation due to the higher specific resistance of the cake layer. Liu et al. [10], in contrast, report that floc particles of a high fractal dimension as well as a large size formed by two-stage coagulant dosing mitigated TMP development more than those formed by a single dose. The strength (resistance toward shear stress) of floc particles formed by coagulation also plays an important role in the permeability of the cake layer

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[11–13]. The increase in transmembrane pressure (TMP) in an ultrafiltration (UF) system is lower with floc breakage, which lowers the fractal dimension of flocs, than without breakage [14]. Xu and Gao [3], however, reported that an increased shear for floc breakage considerably decreased the floc size and increased the floc compactness, thus increasing resistance and lowering the permeability of the cake layer. Therefore, an increase in floc strength could enhance the permeability of the cake layer [4]. Overall, findings on the relationship of floc characteristics to membrane performance are not consistent, though it is clear the structure of the cake layer plays an important role in membrane permeability.

In a ceramic membrane MF system, the permeability of the cake layer may not be a crucial issue, because an integrated, intensive, hydraulic backwash process would eject most of the cake layer. After coagulation, the affinity of the membrane for destabilized contaminants and their aggregates is lower than without coagulation, which leads to a more effective backwash. Hence, with an integrated, hydraulic backwash, the degree of fouling from cake layer formation would be minimized. Hydraulically irreversible fouling is the main concern in full-scale membrane filtration facilities because it determines energy consumption for long-term membrane filtration and affects the sustainable operation of the facility. Irreversible fouling is caused by contaminants that do not react with or adsorb to hydrolytic species formed by the coagulant and thus are not destabilized [15]. Many studies have been conducted to better understand the behavior of membrane foulants and elucidate fouling mechanisms, but these studies have seldom identified practical solutions to the membrane-fouling problem.

A limited amount of research has concerned the extent to which different coagulant types might be exploited to most effectively reduce the extent of irreversible fouling. Tran et al. [16] reported that polysilicato-iron coagulants were better at mitigating irreversible fouling than aluminum-based coagulants at a higher dose while aluminum-based coagulants worked better at a lower dose. Their study suggests that the effect on membrane fouling is a complex phenomenon where many factors including the residual DOC and the property of small-size flocs influence the fouling to various extents. Membrane fouling may also be caused by hydrolytic species of coagulants, though it has not been fully studied [1]. The key consideration is that coagulant characteristics required for membrane

pretreatment are not necessarily the same as those for coagulation and settling. Conventional coagulation is designed to form large-size floc particles that settle out, whereas, for membrane pretreatment, coagulation should allow for direct filtration of floc that results in improved filtrate water quality and alleviates membrane fouling.

In this study, we investigated five PACI coagulants suitable for direct MF. The effect of PACI properties (basicity and sulfated/non-sulfated) on hydraulically irreversible membrane fouling (hereafter called irreversible fouling), which results in a long-term TMP rise, was studied, in particular, by focusing on the residual aluminum concentration in filtrates and aluminum deposits on membranes.

2. Materials and methods

2.1. Coagulants

Four PACIs were obtained from the Taki Chemical Co. (Kakogawa, Japan): conventional normal-basicity (51%) sulfated PACI (designated as PACI-51s), high-basicity (71%) sulfated PACI (PACI-71s), high-basicity (71%) nonsulfated PACI (PACI-71), and very-high-basicity (90%) nonsulfated PACI (PACI-90). A second very-high-basicity (90%) nonsulfated PACI (PACI-90b) was prepared in the authors' laboratory by the base titration method using NaOH (0.3 M) and AlCl₃ (0.5 M) [17]. The distributions of aluminum species in the coagulants were determined by the ferron method [17]. These species were assumed to be monomeric, polymeric, and colloidal aluminum species on the basis of their reaction rates with ferron reagent (8-hydroxy-7-iodo-5-quinolinesulfonic acid; Wako Pure Chemical Industries, Osaka, Japan), denoted Ala, Alb, and Alc, respectively [18]. Ala denotes aluminum species that reacted with ferron instantaneously (within 30 s); Alb denotes species that reacted with ferron within 120 min; and Alc denotes species that did not react. Properties of the PACIs are listed in Table 1S (Supplementary data).

2.2. Pilot-scale MF system

Experiments were conducted with the coagulation-direct MF pilot plant at the Water Quality Center of the Sapporo Waterworks Bureau, Japan. The plant has two parallel lines (Lines A and B) with the same

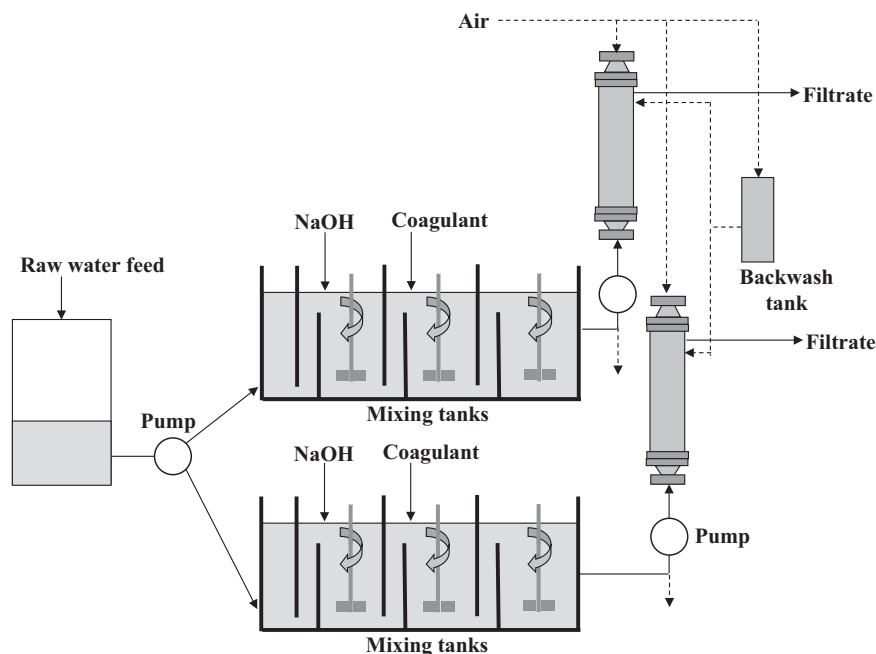


Fig. 1. Pilot-scale MF systems.

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