



Impacts of organic coagulant aid on purification performance and membrane fouling of coagulation/ultrafiltration hybrid process with different Al-based coagulants



Weiying Xu^{a,b,*}, Qinyan Yue^{c,**}, Baoyu Gao^c, Bin Du^a

^a Key Laboratory of Chemical Sensing & Analysis in Universities of Shandong, School of Resources and Environmental Sciences, University of Jinan, Ji'nan 250022, Shandong, People's Republic of China

^b State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

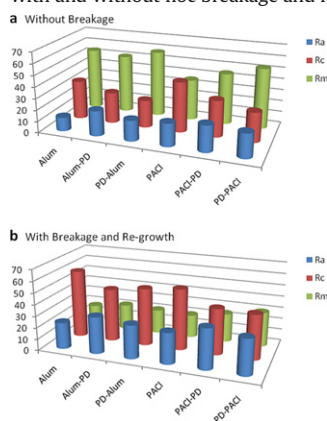
^c Shandong Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Shandong University, Ji'nan 250100, Shandong, People's Republic of China

HIGHLIGHTS

- PD aid in alum and PACI coagulations could improve HA removal by C–UF process.
- Floc size had more important effect on UF than compact degree.
- PD–alum/PD–PACI led to the highest fluxes but was most susceptible to high shear.
- R_c and R_a were chiefly affected by floc size and D_f , respectively.

GRAPHICAL ABSTRACT

Resistance analyses of the investigated systems: coagulations by different alum-based and PACI-based coagulants with and without floc breakage and re-growth phases.



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ABSTRACT

Effects of polydimethylallylammonium chloride (PD) on coagulation behaviors of different Al-based coagulants were investigated in coagulation–ultrafiltration (C–UF) hybrid process, regarding the impurity removal efficiency, floc properties and membrane foulings. Floc characteristics, including floc size, compact degree, strength and re-growth ability were studied using a laser diffraction particle sizing device. Resistance analyses were implemented to explore the membrane fouling mechanisms. The results indicated that PD aid could increase the purification efficiency of C–UF, especially at low coagulant doses. PD–alum/PD–PACI contributed to large flocs, while alum–PD/PACI–PD gave rise to flocs with high R_f and D_f values. The results of ultrafiltration experiments showed that conventional coagulant, i.e., alum led to a flux reduction of 52%; while the reductions for alum–PD and PD–alum were 53% and 34%, respectively. The flux reductions for PACI, PACI–PD and PD–PACI were 60%, 57% and 39%. Flux declines became more severe when the coagulated suspensions were exposed to increased shears and the suspensions coagulated by aluminum plus PD resulted in the least reductions in fluxes.

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* Correspondence to: W. Xu, Key Laboratory of Chemical Sensing & Analysis in Universities of Shandong, School of Resources and Environmental Sciences, University of Jinan, Ji'nan 250022, Shandong, People's Republic of China.

** Corresponding author.

E-mail addresses: stu_xuwy@ujn.edu.cn (W. Xu), qyyue@sdu.edu.cn (Q. Yue).

1. Introduction

Low pressure driven membrane filtration, ultrafiltration (UF), has been receiving a lot of attention and is now increasingly used as a technology for surface water purification. UF can effectively remove particles, turbidity, microorganisms (bacteria, protozoa, algae), and some viruses. Nevertheless, the main drawback of this technology is membrane fouling, which is a major obstacle to the widespread application in water and wastewater treatment of UF throughout the world. Generally, the accumulation of the retained matter on the membrane surface leads to an increase in operating costs, due to increased energy consumption and the necessity of periodic cleaning. Up to now, numerous researchers have studied membrane foulants for many types of membranes with various feed waters and they concluded that most of the natural organic matter (NOM) components are major membrane foulants [1–3]. Humic acid (HA), as the main component of NOM, is considered to be closely associated with membrane fouling [4,5]. Using pre-treatment to lower the NOM concentration of feedstock has been a useful approach to prevent membrane fouling.

Chemical coagulation could significantly enhance the performance of UF in water treatment. Indeed, it is now a common practice to add coagulants into feed water to improve the NOM removal and filtration flux of ultrafiltration unit [6,7]. Our previous study [8] employed various Al-based coagulants in C–UF process and the results revealed that the membrane permeate fluxes varied for different coagulants. That was essentially induced by the distinct properties of aggregates formed by different coagulants in the coagulation/flocculation processes. Actually, there is a great amount of literatures supporting the importance of floc properties in C–UF and/or C–MF (coagulation–microfiltration) hybrid processes. Hwang and Liu [9] reported that effective conventional coagulation conditions generally produced larger particles and this reduced membrane fouling by reducing adsorption in membrane pores, increasing cake porosity and transport of foulants away from the membrane surface. Cho et al. [10] suggested that coagulated flocs with lower fractal dimension could improve membrane permeability due to their higher porosity and relative loose aggregation. Barbot et al. [11] found that the effect of coagulation system on the permeate flux depended on the ability of floc resistance to shear stress. Large flocs, which were highly resistant to shear, led to a 20% increase in permeate flux because they were not easily broken; while large flocs but with weaker resistance to shear resulted in a decrease of 50% in permeate flux. The results of our previous research [12] proved that flocs formed by pre-hydrolyzed Al coagulants, such as PACl, were stronger than those by conventional alum. However, they commonly had more compact structures and hence, pre-hydrolyzed Al species could not essentially improve the permeate flux despite of its superior coagulation behaviors.

Organic polymeric coagulants, like polydimethyldiallylammonium chloride (PDMDAAC or PD for short), have been used in water purification for several decades. Many researchers have considered using PDMDAAC in drinking water treatment due to its wider working pH range and lower dosage demand than metal coagulants for equivalent treatment efficiency [13,14]. However, the high cost of PDMDAAC limits its comprehensive application in water treatment and thus it was generally used in combination with metal coagulants [15]. In this circumstance, PD was used as primary coagulant aid of two different aluminum coagulants, a commercial product (alum) and lab-prepared material (polyaluminum chloride, i.e., PACl), expecting to obtain superior aggregates propitious to the ultrafiltration performance. This has rarely been reported and could have great practical significance.

Besides, various mixing modes were implemented in the coagulation process to explore the effect of increased shear on floc properties and membrane foulings. In natural as well as technical aquatic systems particles and flocs in suspension undergo highly different and varying flow conditions, which are always accompanied by various shear forces. In C–UF system, the circulation of suspension also commonly induces variation of shear stress. Depending on the binding strength of the

particles or aggregates, the action of shear forces may lead to deformation and eventual floc break-up. Over the last three decades, numerous studies have been carried out to obtain information on the particle characteristics under certain hydraulic shear and to bring considerable insight into the mechanisms involved in floc formation and break-up. However, most of the previous studies focused on the relationship between shear conditions and floc properties, while the effect of different shear conditions on the subsequent membrane performances remains unclear, which is necessary and of significant meaning.

The present paper was thus focused on (1) the influences of PDADMAC aid on floc properties and ultrafiltration performances for different Al coagulants in C–UF processes; (2) floc properties and membrane behaviors for the coagulants with PD aids when the pre-formed flocs were exposed to enhanced shear rates; and (3) comparison of membrane fouling mechanisms by coagulation effluents with and without PD aids.

2. Materials and methods

2.1. Humic acid solution and coagulant preparations

The biochemical reagent HA was purchased from the Jufeng Chemical Technology Co. Ltd., Shanghai, China. HA sample water with the concentration of 10.0 mg/L was prepared as previously described [8].

Two Al-coagulants, alum and PACl with a basicity value (B, OH/Al molar ratio) of 2.2 were used in this study and the preparation details can be found in other paper [16,17]. Cationic polymer PDMDAAC (40% w/w aqueous solution, 100% charge density) was purchased from Bin Zhou Chemical Co., Shandong, China and its intrinsic viscosity was 1.02 dL/g. PD solution with a target concentration of 1.0 g/L was prepared by dissolving pre-determined PD into DI water under continuous stirring.

2.2. Pre-coagulation–ultrafiltration (C–UF) experiments

Jar tests were carried out using a programmable jar test apparatus (ZR4-6, Zhongrun Water Industry Technology Development Co. Ltd., China) composed of six 1.0 L cylindrical plexiglass beakers. HA water samples were first stirred at 200 revolutions per minute (rpm) for 30 s with the purpose of uniform mixing, and then the prepared coagulants were added. PD aid was added in two ways. For the first one, Al coagulant was dosed firstly at the start of rapid mixing period, followed by PD addition after 30 s, and then the rapid mixing continued for another 1 min. This dual-coagulant was referred as alum–PD/PACl–PD. For the second one, referred as PD–alum/PD–PACl, the addition sequence of Al and PD coagulants was reversed. After the rapid mixing stage, 15 min of slow stirring period at 40 rpm was introduced to allow floc growth. Zeta potentials (ZP) were measured a few seconds after coagulant addition using a Zetasizer 3000HSa (Malvern Instruments, UK). A dead-end batch UF unit was set up to filter the coagulated water as shown in Fig. 1. Commercial polyethersulfone (PES) flat membrane (Mosu Co. Ltd., China) was used throughout the experiments and the characteristic parameters of the membrane were listed in Table 1. Nitrogen gas was used to maintain precise supply of constant pressure at 220 kPa. For more detailed procedures of the C–UF process, please refer to the literature [8].

2.3. Floc characterizations

2.3.1. On-line measurement of floc size

A laser diffraction instrument (Mastersizer 2000, Malvern, UK) was used to continuously measure the dynamic floc size as the coagulation proceeded and the details have been reported in other paper [18].

2.3.2. Floc strength, recovery ability and structure analyses

A series of jar tests with 5 min of breakage periods at 200 rpm, performed after the floc had reached their steady stage, were conducted to

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