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## Investigation of pre-coagulation and powder activate carbon adsorption on ultrafiltration membrane fouling

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## ABSTRACT

Pre-coagulation by alum before ultrafiltration without (CUF) and with (CAUF) powdered activated carbon (PAC) addition was investigated to explore the influence of PAC on membrane fouling. It was found that the continuous addition of a low dosage of PAC reduced membrane fouling for operational run periods < 20 days, both for reversible and for irreversible components of fouling. The presence of PAC increased the removal of dissolved organic matter (DOM), particularly proteinaceous substances, and some fractions of humic-type substances. For a longer operation time (> 20 days) an increase in mainly reversible fouling was evident and was attributed to microorganism growth in the cake layer, as indicated by the enhanced concentrations of extracellular polymeric substances, such as polysaccharides and proteins. A process of crystallization of the coagulant floc in the cake layer with increasing operation time was believed to cause desorption of DOM from primary coagulation particles and a decrease in their size, and consequently a higher density of cake layer. PAC adsorption of desorbed DOM within the cake layer, and residual DOM after coagulation, reduced DOM reaching the membrane surface and the extent of subsequent internal membrane fouling.

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## 1. Introduction

Ultrafiltration (UF) membrane systems are an attractive alternative for the treatment of ground waters and surface waters for potable water supply since they can economically achieve a consistently high quality of drinking water. However, natural organic matter (NOM) is often a major foulant during the ultrafiltration of surface water [1,2], since it contains a complex mixture of humic and fulvic acids, proteins, carbohydrates of various molecular sizes and functional groups [2–5], which can either bind to the membrane surface or support the development of microbiological fouling.

Pretreatment before UF, especially by “in-line” chemical coagulation or coagulation–hydraulic flocculation, has been shown to be an effective way to control membrane fouling [6–8], and improve general water quality [1,9,10]. However, not all contaminants can be fully removed by the coagulation process, particularly biopolymers, such as polysaccharides and proteins [3–5]. In view of this, the combination of powder activate carbon

(PAC) adsorption and UF has been reported to be more effective in the removal of organic contaminants, either as represented by total organic carbon or any other special pollutants [11–16]. However, the addition of PAC may influence membrane fouling.

Currently there is no clear consensus concerning the effect of PAC addition on the flux of membrane filtration. Some researchers have reported that PAC was ineffective at reducing membrane fouling despite it enhancing the removal of micromolecular NOM [13,17–19], and regardless of the NOM characteristics (hydrophobicity and protein content) and inorganic content of the water [20]. Alternatively, some researchers have found that PAC can improve membrane flux [13,21–23]. The reason for the improved membrane performance was because the PAC acts as a scouring agent which limits foulant deposition or controls irreversible membrane fouling, and the effect of the combined adsorption and biodegradation by the biologically active PAC on the foulant components [20,24].

In contrast, other studies have indicated that the addition of PAC caused higher membrane fouling [25–27]; the reasons for this have been stated to be because PAC adsorbed non-fouling molecules preferentially over foulants, although PAC removed a larger fraction of the organic matter [28], and that the attachment of NOM to PAC particles led to an increase in cake layer resistance [29]. Overdosing of PAC contributes to fouling by blocking the membrane pores [30]

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or by forming a cake layer which increases the reversible resistance [31]. Therefore, it can be concluded that PAC, as a sole pretreatment, can improve organic matter removal, but it may have an adverse impact on membrane fouling.

Previous studies involving coagulation have compared combined coagulation/microfiltration (MF) and PAC/MF, and found that there was a similar performance for organic matter removal, but coagulation/MF resulted in a significant decrease in flux [32]. Alternatively, some authors have recognized the potential benefits of a combined coagulation and PAC adsorption process before membrane filtration [33]; however, in these cases the membrane operation process lasted for only 30 min [12], or less than 5 h [17]. Matsui et al. [34] found that both physically reversible and irreversible membrane fouling were reduced by a combination of PAC and polyaluminum chloride (PACl) pretreatment, because of a high NOM removal and the formation of larger and more porous flocs. Dialynas and Diamadopoulos [35] found that after the addition of PAC to a coagulation–UF system, the trans-membrane pressure increased rapidly due to the formation of the PAC cake on the membrane surface, but there was a high degree of organic matter removal.

In light of the previous studies summarized above, there is no consensus as to whether the addition of PAC can mitigate or aggravate membrane fouling. In this paper, the impact of adding a low dosage of PAC with alum as the pretreatment before UF membrane filtration has been examined in terms of the nature and extent of membrane fouling. This paper provides additional information, including that relating to bacteria and extracellular polymeric substances (EPS), and the adsorption of organic matter released from the cake layer by PAC in short and long ages, to the limited work reported so far concerning the combined use of PAC and alum coagulant before UF membrane filtration.

## 2. Materials and methods

### 2.1. Synthetic raw water and coagulant

Synthetic raw water was chosen for the tests in order to simplify the study to provide sample consistency and reproducibility. Domestic sewage was added to the local (Beijing, China) tap water with a volumetric ratio of 1:50 to simulate a surface water supply slightly polluted by sewage discharges. In addition, 5 mg/L of humic acid (HA, sodium salt, Aldrich, Cat: H1, 675-2) was added to the raw water. Prior to mixing with domestic sewage and humic acid solution, the tap water was left overnight to ensure the complete decay of residual chlorine, which may inactivate/kill the bacteria. The characteristics of the synthetic raw water are listed in Table 1. During the course of the experimental program the temperature of the water was  $20 \pm 2$  °C.

**Table 1**  
Water quality of raw water and UF influents<sup>a</sup>/filtrates.<sup>b</sup>

Parameter <sup>c</sup>	Raw water	CUF influent	CAUF influent	CUF filtrate	CAUF filtrate
UV <sub>254</sub> (cm <sup>-1</sup> )	0.115 ± 0.006	0.029 ± 0.002	0.023 ± 0.002	0.027 ± 0.003	0.020 ± 0.002
DOC (mg/L)	4.956 ± 0.415	2.843 ± 0.185	2.532 ± 0.302	2.572 ± 0.234	2.188 ± 0.287
Turbidity (NTU)	3.69 ± 0.16	2.03 ± 0.18	1.75 ± 0.21	0.05 ± 0.02	0.06 ± 0.03
Al (mg/L)	0.050 ± 0.002	0.079 ± 0.001	0.069 ± 0.003	0.067 ± 0.001	0.065 ± 0.003
P (mg/L)	0.259 ± 0.061	0.062 ± 0.014	0.073 ± 0.010	0.061 ± 0.013	0.060 ± 0.010
NH <sub>4</sub> <sup>+</sup> -N (mg/L) <sup>d</sup>	0.934 ± 0.145	0.252 ± 0.069	0.482 ± 0.104	0.125 ± 0.035	0.305 ± 0.045
pH	7.93 ± 0.06	7.02 ± 0.04	7.01 ± 0.04	7.04 ± 0.05	7.03 ± 0.03

<sup>a</sup> Influent – within membrane tank, immediately after flocculation units.

<sup>b</sup> The values in Table 1 are averages for all the measurements during the 3 stages.

<sup>c</sup> Number of measurements (n): For turbidity, UV<sub>254</sub>, and DOC, n=9; for residual Al, and P, n=5.

<sup>d</sup> NH<sub>4</sub><sup>+</sup>-N (mg/L) in CAUF was measured in the early period (less than 20 days) of the three stages, and it was nearly the same as that in the CUF at the end of the first stage.

Aluminum sulfate hydrate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·18H<sub>2</sub>O; Bodi, China, > 99%), ‘alum’, was used as a coagulant in this study. Stock alum solution was prepared at a concentration of 0.1 M in de-ionized (DI) water. A stock powdered activated carbon suspension (PAC, < 20 μm, Shanghai) was prepared at a concentration of 5 g/L.

### 2.2. Pretreatment before UF processes

Conventional coagulation and flocculation by alum without and with PAC addition (at a concentration of 10 mg/L) before ultrafiltration (CUF/CAUF, respectively) were operated in parallel. A schematic illustration of the CUF and CAUF arrangements is shown in Fig. 1. Synthetic raw water was supplied to a constant-level tank to maintain the water head for the membrane tanks. An optimal alum dose of 0.15 mM (determined by pre-experiments) was continuously added to the rapid mixing tanks, which had a stirring speed of 200 rpm ( $G=184$  s<sup>-1</sup>; determined in our previous work [36]) and a hydraulic retention time (HRT) of 1 min. After mixing, the flow passed to three flocculation tanks, with each having a stirring speed of 50 rpm ( $G=23$  s<sup>-1</sup>) and an HRT of 5 min. For tests with PAC, 10 mg/L PAC was added to the first flocculation tank to ensure sufficient contact time for adsorption prior to UF separation. After the flocculation tanks, the flow passed directly to the membrane tanks. Each tank contained a submerged polyvinylidene fluoride (PVDF) hollow-fiber UF membrane module (Tianjin Motimo Membrane Technology Co., Ltd., China) where the membrane had a nominal pore size of 0.03 μm and a surface area of 0.025 m<sup>2</sup>. The permeate from the submerged membrane module was continuously withdrawn by a peristaltic pump at a constant flux of 20 L m<sup>-2</sup> h<sup>-1</sup>, operated in a cycle of 30 min filtration and 1 min backwash (40 L m<sup>-2</sup> h<sup>-1</sup>). For each backwash, air was supplied to each reactor at 100 L/h (air:water=200:1). The trans-membrane pressure (TMP) was continuously monitored by pressure gauges. The HRT of the membrane tanks was maintained at 0.5 h and accumulated sludge was released every 3 days. The cake layer on the surface of the membrane was washed away by a high-pressure tap water and sponge application twice during the whole operation process (63 days). It is noted that direct ultrafiltration without pretreatment caused significantly higher membrane fouling than with pretreatment, and this was not compared here [37].

### 2.3. Samples of cake layer and sludge

At the end of operation, the fouled membrane modules were taken out from the membrane tanks. The cake layer on the membrane surface was carefully scraped off with a plastic sheet (Deli, China), and analyzed by the following methods to characterize the external membrane fouling. The extraction of internal fouling is described later.

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