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## Q1 Influence of coagulation mechanisms and floc formation 2 on filterability

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### A B S T R A C T

Minimizing particles in water is a key goal for improving drinking water quality and safety. 20  
The media filtration process, as the last step of the solid–liquid separation process, is largely 21  
influenced by the characteristics of flocs, which are formed and controlled within the 22  
coagulation process. In a laboratory-based study, the impacts of the physical characteristics 23  
of flocs formed using aluminum sulfate on the filtration treatment of two comparative 24  
water samples were investigated using a photometric dispersion analyzer and a filterability 25  
apparatus. In general, the optimum dosage for maximizing filterability was higher than that 26  
for minimizing turbidity under neutral pH conditions. For a monomeric aluminum-based 27  
coagulant, the charge neutralization mechanism produced better floc characteristics, 28  
including floc growth speed and size, than the sweep flocculation mechanism. In addition, 29  
the charge neutralization mechanism showed better performance compared to sweep 30  
flocculation in terms of DOC removal and floc filterability improvement for both waters, 31  
and showed superiority in turbidity removal only when the raw water had high turbidity. 32  
For the different mechanisms, the ways that floc characteristics impacted on floc 33  
filterability also differed. The low variation in floc size distribution obtained under the 34  
charge neutralization mechanism resulted in the flocs being amenable to removal by 35  
filtration processes. For the sweep flocculation mechanism, increasing the floc size 36  
improved the settling ability of flocs, resulting in higher filter efficiency. 37

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## 48 Introduction

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52 Minimizing the particles in treated drinking water is funda-  
53 mental to the successful operation of a water treatment  
54 facility. In water treatment processes, media filtration is the

55 final solid–liquid separation step, (Boller and Blaser, 1998) and  
56 its performance is mainly evaluated by filtered water qualities  
57 (turbidity and dissolved organic carbon (DOC)), head loss  
58 development (rate and time to backwash) and water produc-  
59 tion (unit filter run volume). Parameters that determine the

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filtration performance include particle size, particle concentration and the surface chemical properties of particles, and these are usually influenced by pre-treatment methods. Coagulation, applied for the removal of turbidity and natural organic matter (NOM), is the most common treatment process prior to the filtration process (Xiao et al., 2009; Jiao et al., 2014).

During coagulation treatment, most natural particles can be effectively aggregated in floc formation and then removed by sedimentation. However, a certain amount of flocs will not settle readily due to small size, low density and/or weak floc strength that leads to floc breakage under hydraulic shear forces (Jarvis et al., 2005). These flocs can then break up through the filtration process. Typically, small fine particles with the size of 1–10  $\mu\text{m}$  have been considered the most challenging elements to be removed in media filters. This is because particles of this size range can pass straight through the filter bed, detach from the filter media, or overload the filter (Fabrizi et al., 2010; Kim and Tobiasson, 2004). Therefore, the development of floc characteristics that result in efficient filtration and high quality drinking water is of high importance.

To date, in studies on floc characteristics, researchers have focused on the evaluation of the physical characteristics of flocs under different coagulation conditions. For example, studies found that the formation of flocs depends on the type and dosage of coagulant, applied mixing shear, and the concentration and characteristics of the floc particles, e.g., size (Yu et al., 2010a; Ehrl et al., 2008). Besides hydraulic conditions, coagulation mechanisms are largely influenced by the coagulant type, the demand for coagulant and the characteristics of particles. From a control point of view, coagulation mechanisms determine the characteristics of flocs. For the commonly used hydrolyzing coagulants, the removal of particles mainly depends on two mechanisms (charge neutralization and sweep flocculation) that have been widely studied (Duan and Gregory, 2003; Yu et al., 2010b). Some studies have described the influence of coagulation mechanisms on floc characteristics (Kim et al., 2001; Li et al., 2006). For example, Yu et al. (2011) compared the floc strength and reforming of broken flocs from aluminum sulfate formed at pH 5 and pH 7 using a photometric dispersion analyzer (PDA). They found that the flocs formed by charge neutralization were much stronger (resistant to breakage) than those by sweep flocculation (Yu et al., 2011). In the study of Xu et al. (2011), the floc characteristics of two different coagulants, pre-hydrolyzed  $\text{Al}_{13}$  and  $\text{AlCl}_3$ , in humic acid (HA) coagulation were compared using a laser diffraction particle sizer. It was found that flocs formed by preformed  $\text{Al}_{13}$ , where formation mainly depends on the neutralization mechanism, contributed to flocs with larger fractal dimension than those formed using  $\text{AlCl}_3$  (Xu et al., 2011).

Previous studies have mostly focused on floc characteristics during the coagulation stage. However, the impacts of the physical characteristics of flocs on filtration treatment have often been overlooked. Therefore, the aim of the study reported here was to investigate the influence of different raw water turbidities on coagulation mechanisms and floc properties, plus the relationships between floc characteristics and filterability under different mechanisms. In many laboratory-based studies, paper filters have been used for filtration of coagulant-treated waters (in jar tests) in the evaluation of floc filterability. However, filter papers used for filtration have smaller pore sizes than

those present in sand filtration in full-scale operations. Further, it is not feasible to evaluate full-scale rapid sand filter head losses by the use of paper filtration under laboratory conditions. In this study, sand filtration was used under laboratory conditions to better simulate the practical water treatment process.

## 1. Materials and methods

### 1.1. Raw water

Two waters, of low and high turbidity (spiked with clay) were used in the experiments. The first (Water I) was collected from the Happy Valley Reservoir at the inlet to the water treatment plant (WTP), located 15 km south of Adelaide, South Australia. The Happy Valley Reservoir water is sourced from the River Murray and from the Mt. Bold catchment. During the period of study, the turbidity of the reservoir water was  $4.6 \pm 0.5$  nephelometric turbidity units (NTUs), and DOC was  $9.7 \pm 0.3$  mg/L. For the second water (Water II), clay was added into Water I to significantly elevate the turbidity. The clay was obtained from a zero order catchment (grassland) of the Myponga Reservoir (a drinking water reservoir located 50 km south of Adelaide). The clay sample was sieved to less than 420  $\mu\text{m}$ . The elemental composition of the clay was determined by a field emission scanning electron microscope (FE-SEM) and is shown in Fig. 1 of the supplementary information (Appendix A Fig. S1). The turbidity of Water II was  $46 \pm 2$  NTU, with raw water DOC of  $9.5 \pm 0.5$  mg/L.

### 1.2. Jar testing

Jar testing was performed using a PB-900 programmable six-paddle gang stirrer (Phipps and Bird, USA). Flash mixing of the alum coagulant was performed at 200 rpm for 1 min with the addition of coagulant at  $t = 30$  sec. This was followed by 14 min slow mixing at 40 r/min and then 15 min of sedimentation. Samples of settled water were then analyzed for turbidity and DOC.

Aluminum sulfate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ ) obtained from a full-scale water treatment plant (WTP) was used as the coagulant. Stock solutions were prepared at a concentration of 20,000 mg/L. All coagulant doses are expressed as mg/L  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$  in this study.

Sodium hydroxide (NaOH) and sulfuric acid ( $\text{H}_2\text{SO}_4$ ) of 0.5 mol/L were used to adjust the pH level.

### 1.3. Filterability index

A filterability index was used to assess flocs. The structure of the apparatus is shown in Appendix A Fig. S2. In the experiments, dual media comprising anthracite and sand was chosen to model a full-scale filter process (Appendix A Fig. S3). The filterability index can be calculated as follows:

$$\text{Filterability Index} = \frac{H \cdot C}{C_0 \cdot T \cdot V} \quad (1)$$

where,  $H$  (cm) is the head loss, which can be calculated by comparing the difference between the initial and final values

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