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Recycling ferric water treatment sludge (Fe-WTS) as coagulation aid: A case study in residual aluminum control

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ABSTRACT

To provide an insight into aluminum (Al) control during the treatment of micro-polluted water, recycling ferric water treatment sludge (Fe-WTS) as an alternative approach was innovatively investigated. Particular attention was paid to the recycling ratio of Fe-WTS when coagulated at an optimized dosage of both $Al_2(SO_4)_3$ and polyaluminum chloride (OH/Al = 2.0, denoted as PAC_{20}) respectively. Coagulation performance, i.e., turbidity, organics and Al speciation were also examined. A morphological analysis, characterized via floc size and fractal dimension, was further conducted to explore the fundamental floc operational parameters and to elucidate the mechanism of different Al species removal. The results indicate that an appropriate recycling ratio yields improvements in residual Al control. In comparison, Fe-WTS is more effective in dissolved Al (Al_D) removal when coagulated by $Al_2(SO_4)_3$, while is proper to the reduction of colloidal Al (Al_{CO}) when coagulated by PAC_{20}. Flocs properties are significantly correlated with Al speciation during the treatment. Specifically, floc size is proportional to Al_{CO} removal rate, however, the value of floc fractal dimension is inversely proportional to Al_D removal rate.

Keywords: Ferric water treatment sludge; Recycling; Residual aluminum; Aluminum transformation; Floc property

1. Introduction

Al based coagulants are widely used in the production of potable water [1,2]. Despite its effectiveness during coagulation, it also results in superfluous residual Al in the finished water, especially the source water are of micro-pollution and/or low temperature [3,4]. The national drinking water standard (GB5749-2006) specifies that the Al concentration must be lower than 0.2 mg/L, and even much stricter in the USA (0.05 mg/L) [4,5]. Excessive Al in treated water or in distribution systems can potentially lead to unexpected problems, such as increased turbidity, reduced disinfection efficiency, and a loss in hydraulic capacity [3,6]. Moreover, if the concentration of Al has not been effectively controlled, ingestion may damage the human' nervous system, and even lead to Alzheimer's disease [7]. Because the brain has affinity to Al, sedimentation in the brain caused by ingestion may deteriorate memory and even intelligence. Hence, it is crucial to perform an investigation into residual Al control during the production of potable water.

A large amount of sludge is produced during water treatment for water production, amounting to a total of 10,000 tons per day [8]. As Zhou et al. [9] reported, drinking water treatment sludge must be buried in landfills or dumped into the ocean. The reuse of sludge is a better alternative approach, so that the waste residues from drinking water treatment can be reclaimed. In addition, based on the characterization of elevated concentrations of inorganic and organic substances, as well as hydroxide sediment derived from salt-coagulants, the removal of particulates, phosphorus, arsenate, boron, and heavy metals, i.e., copper (II) and lead (II) during the recycling process has been investigated and verified [9]. However, the literature regarding residual

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Al control of potable water during the recycling process is sparse. Therefore, it is important to discuss the control of Al in finished water when recycling water treatment sludge by coagulation and/or adsorption.

In this study, we adopted ferric water treatment sludge (Fe-WTS), an inexpensive and readily available waste material, as a coagulation aid during the recycling process, and therefore, the goals of this study are as follows: (1) to evaluate the potential feasibility of recycling Fe-WTS in residual Al control during the treatment of synthetic micro-polluted water, and (2) to discuss the removal mechanism of various Al species, including dissolved Al (Al_D) and colloidal Al (Al_{CO}), by analyzing the relationship between flocs properties and the transformation of different Al species.

2. Materials and methods

2.1. Preparation and synthesis of coagulant

In terms of different kinds of Al species, two Al based coagulants were employed in the current study. $Al_2(SO_4)_3$ was purchased from Aladdin Industrial Corporation, and polyaluminum chloride (PAC) was prepared in the laboratory. Specifically, PAC with a B value (OH^-/Al^{3+} ratio) of 2.0, denoted as PAC_{20}, was synthesized using a slow titration method [10]. The prepared stock solution was stored in a refrigerator (4°C) and updated every two weeks for homogeneity and effectiveness.

The properties of used coagulants, as shown in Table 1, were determined by the titrimetric method according to the national standard of China (Ferron timed complex-colorimetric method) [11].

2.2. Raw water and sludge

To simulate micro-polluted surface water, the raw water was synthesized by adding a predetermined amount of humic acid (HA), kaolin clay, and filtered domestic wastewater into local tap water (Beijng, China), and then stored at room temperature for 24 h to reach a balance between adsorption and desorption of organic matter. A stock solution of HA and kaolin clay was prepared according to the references [12,13]. The main physicochemical characteristics of the raw water are summarized in Table 2.

Fe-WTS was collected from a drinking water treatment plant in Beijing during the period of the current study. This plant employed a conventional process for potable water production, including stirring coagulation, horizontal sedimentation, sand filtration, and disinfection. The properties of Fe-WTS were as follows: moisture content = 99.31%, total solids = 4.28 g/L, total chemical oxygen demand = 127.07 g/L, soluble chemical oxygen demand in supernatant = 22.98 g/L, pH = 7.51.

2.3. Coagulation trials

To determine the optimum dosage of coagulants and recycling ratio of Fe-WTS, a series of jar tests was conducted using a programmable apparatus (ZR4-6, Zhongrun, China) at a temperature of 22 ± 1°C. The procedure for both started with a rapid mixing at 350 rpm ($G = 449\ s^{-1}$) for 0.5 min; A known dosage of coagulant was added simultaneously with or without Fe-WTS, followed by 1 min of rapid mixing at 120 rpm ($G = 106\ s^{-1}$). Subsequently, the stir speed was declined to 50 rpm ($G = 33\ s^{-1}$) for 15 min followed by a 20-min settlement.

Water samples were collected from the supernatant, 3 cm below the surface, for the measurement of turbidity, UV_{254}, dissolved organic carbon (DOC) and residual Al (seen in the section 2.4).

2.4. Measurement of residual Al

The different Al species were analyzed using the chrome azurol S spectrophotometric method [14]. Nucleopore polycarbonate filters (0.45 μm) were used for the separation of dissolved and colloidal Al. Particularly, the initial filtrate (~30 mL) was discarded to avoid sampling bias caused by potential Al adsorption through the filter material.

2.5. Flocs properties

In this study, two characteristic parameters including its size and fractal dimension were adopted to investigate the removal mechanism of different Al species during the recycling process. Samples of flocs were acquired by a hollow plastic tube at 7 min. A tube open at both ends was inserted into the jar, below the surface, at about 3.0 cm. Flocs were withdrawn by covering one end of the tube and then transferring it on to a flat microscope slide. The flocs images were captured by an optical microscope (Olympus, BX51TF, Japan) equipped with a CCD camera. The camera has a sensor matrix consisting of 1360 (horizontal) ×

Table 1
Species distribution of coagulants

Coagulants	B	Al_a (%)	Al_b (%)	Al_c (%)
$Al_2(SO_4)_3$	0.0	93.8	6.2	0.0
PAC	2.0	25.6	65.5	8.9

Ala: monomer; Alb : medium polymer; Alc: gel or solid

Table 2
Main physicochemical characteristics of raw water

Index	Turbidity	UV_{254}	DOC	SUVA	ZP	Al_T	Al_D	Temperature	pH
Units	NTU	cm^{-1}	mg/L	L/(mg m)	mV	mg/L	mg/L	°C	–
Value	5.3	0.087	2.852	3.05	–15.3	0.334	0.205	22.4	7.67

UV_{254}: ultraviolet light absorption at 254 nm; DOC: dissolved organic carbon; SUVA: UV_{254}/DOC × 100; ZP: zeta potential.

1024 (vertical) pixels. Images were acquired from an interrogation window of about 2952 mm × 1944 mm with a resolution of 1340 pixels/mm. Thus, 1 pixel corresponds to about 0.699 µm. All images were analyzed using Image J software.

The perimeter-based fractal dimension D_{pf} was calculated according to Eq. (1) [15].

$$A \propto^{2/Dpf} \quad (1)$$

where A is defined as the flocs projected area and P is perimeter.

Unlike a Euclidean object, objects with smoothly varying contours (circles) have $D_{pf} = 1$, whereas extremely convoluted plane-filling contours (line) have $D_{pf} = 2$. Thus, the irregularity of a bounding contour is characterized by $1 \le D_{pf} \le 2$ [16]. A smaller D_{pf} indicates a more regular floc structure, demonstrating a higher flocs density that is beneficial to solid-liquid separation [4]. In this study, the fractal dimension was determined for flocs larger than 50 pixels (35 µm) and the 50–80 samples of the aggregates were selected.

3. Results

3.1. Coagulation performance of $Al_2(SO_4)_3$ and PAC under various recycling designs

3.1.1. Turbidity removal and zeta potential

The variation of turbidity removal rate and zeta potential as a function of coagulant dosage are shown in Fig. 1. There existed two similar curves for turbidity removal trend of $Al_2(SO_4)_3$ and PAC_{20}. At a dosage of 17 mg/L (calculated as Al), PAC_{20} achieved an optimum removal efficiency of 79.3%, compared to a lower removal efficiency of 74.2% for $Al_2(SO_4)_3$ at dosage of 11 mg/L. The exact values of residual turbidity corresponding to the highest removal efficiency of $Al_2(SO_4)_3$ and PAC_{20} were 1.38 NTU and 1.09 NTU, respectively. Accordingly, the values of zeta potential of both coagulants continually increased with the increment of its dosage. Isoelectric points of $Al_2(SO_4)_3$ and PAC_{20} were obtained at 20 mg/L and 11 mg/L, respectively. The only difference was the charge of colloids and particles reversed to the positive when PAC_{20} dosage was higher than 11 mg/L.

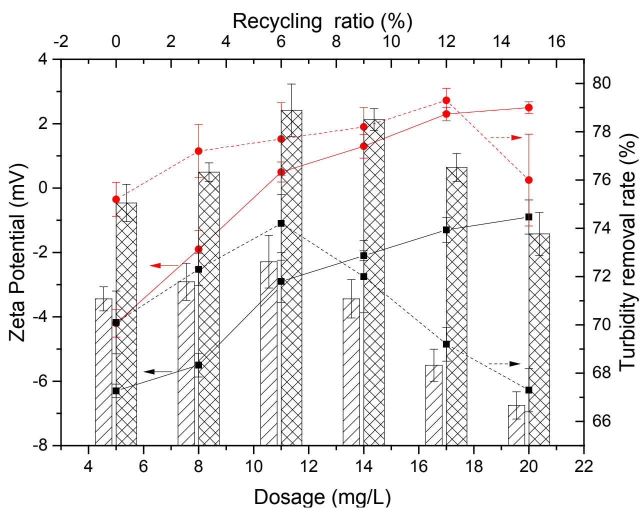

Fig. 1. Effects of dosage and recycling ratio on turbidity removal and zeta potential of both coagulants. Note: (1) Fe-WTS recycling trials were conducted under the optimal dosage of each coagulant, at 11 mg/L of $Al_2(SO_4)_3$ and 17 mg/L of PACl respectively; (2)line: (■) for $Al_2(SO_4)_3$, (●) for PAC_{20}; column: (▨) for $Al_2(SO_4)3$, (▩) for PAC_{20}. (error bars were donated as standard deviation).

It can also be observed from Fig. 1 that the addition of Fe-WTS exerted a different influence on colloid and/or particle removal when coagulated by $Al_2(SO_4)_3$ and PAC_{20}. Compared to the recycling ratio of 0%, as control group, which were determined under the optimal dosage of $Al_2(SO_4)_3$ and PAC_{20} respectively, Fe-WTS obviously improved the performance when PAC_{20} was used. Concretely, the elevated range increased by 6.0%, from 78.2% (0% recycling ratio) to 84.2% (6% recycling ratio). For this reason, with either a decrease or increase in the recycling ratio at 6%, the turbidity efficiency decreased. On the contrary, the superfluous addition (>6% recycling ratio) of Fe-WTS during the $Al_2(SO_4)_3$ process deteriorated the turbidity removal performance in the most cases, and even sharply reduced it to 65.1% at the recycling ratio of 15%, compared to 72.0% of the control group.

3.1.2. Organics removal

As shown in Fig. 2, PAC_{20} demonstrated a greater capability of eliminating organic matter than $Al_2(SO_4)_3$. Generally, UV_{254} and DOC removal rates continually ascended when the dosages of $Al_2(SO_4)_3$ and PAC_{20} increased. As to UV_{254} removal, peak values were achieved at 20 mg/L and 14 mg/L for 39.5% and 42.1% of $Al_2(SO_4)_3$ and PAC_{20}, respectively. Correspondingly, peak values of DOC removal rate were 45.7% and 52.2% at the dosage of 17 mg/L sequentially.

If there was recycling Fe-WTS during coagulation, the organic removal displayed by UV_{254} and DOC was signifi-

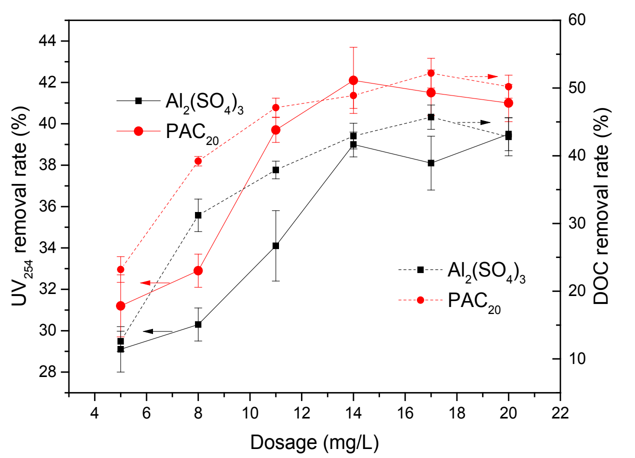

Fig. 2. Effect of dosage on organics removal of both coagulants (error bars were donated as standard deviation).

cantly different. As shown in Fig.3, the addition of Fe-WTS exerted a nearly negative effect on UV_{254} removal at the most recycling ratio. Especially when coagulated by $Al_2(SO_4)_3$, the rate of UV_{254} removal descended from 39.0% (0% recycling ratio) to 34.1% (15% recycling ratio). By comparison, Fe-WTS showed an improved UV_{254} removal performance when coagulated by PAC_{20} at 6% recycling ratio (UV_{254} removal rate 43.7%), compared with 0% recycling condition (UV_{254} removal rate 42.1%). On the contrary, the results of DOC indicated an obvious positive improvement on organics removal at a certain recycling ratio. The peaks of the two curves (shown in Fig. 3) occurred at the recycling ratio of 6%, and the corresponding removal rates were 49.7% and 54.6% for $Al_2(SO_4)_3$ and PAC_{20}, respectively.

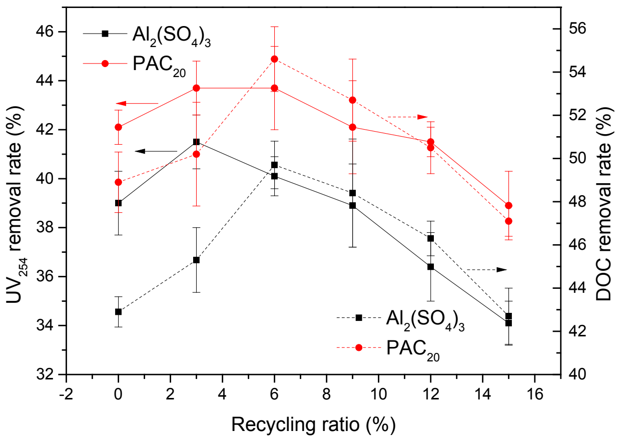

Fig. 3. Effect of recycling ratio on organics removal of both coagulants. Note: (1) Fe-WTS recycling trials were conducted under the optimal dosage of each coagulant, at 11 mg/L of $Al_2(SO_4)_3$ and 17 mg/L of PACl respectively. (error bars were donated as standard deviation).

3.1.3 Residual Al speciation

The variation of different residual Al species including Al_T, Al_D and Al_{CO} under conventional coagulation is shown in Fig. 4. Fig. 4a displays the Al_T removal tendency as a function of coagulant dosage. The residual Al_T concentration descended when the coagulant dosage was kept at a relatively low range (5–11 mg/L). The result indicated an appropriate dosage, 11 mg/L and 14 mg/L, for Al_T removal of $Al_2(SO_4)_3$ and PAC_{20} separately. In comparison, there was a significant difference for the Al_D removal compared to the Al_T removal, as shown in Fig. 4b, in that the increased dosage of coagulant continually caused a further decrement of Al_D content, and the lowest concentrations of Al_D were 0.199 mg/L and 0.151 mg/L of $Al_2(SO_4)_3$ and PAC_{20} respectively. However, the corresponding Al_D concentration of the optimized dosage of both coagulants (11 mg/L and 14 mg/L), as noted above, were 0.289 mg/L and 0.163 mg/L. As to Al_{CO} reduction, shown in Fig. 4c, the variation presented similar trends regardless of coagulant type.

Based on the analysis of the transformation of various Al species in conventional coagulation, the comparative investigation of recycling Fe-WTS was carried out under the predetermined optimal dosage of 11 mg/L and 14 mg/L of $Al_2(SO_4)_3$ and PAC_{20} respectively. The results show (Fig. 5) that the recycling ratio exerted a distinct influence on the

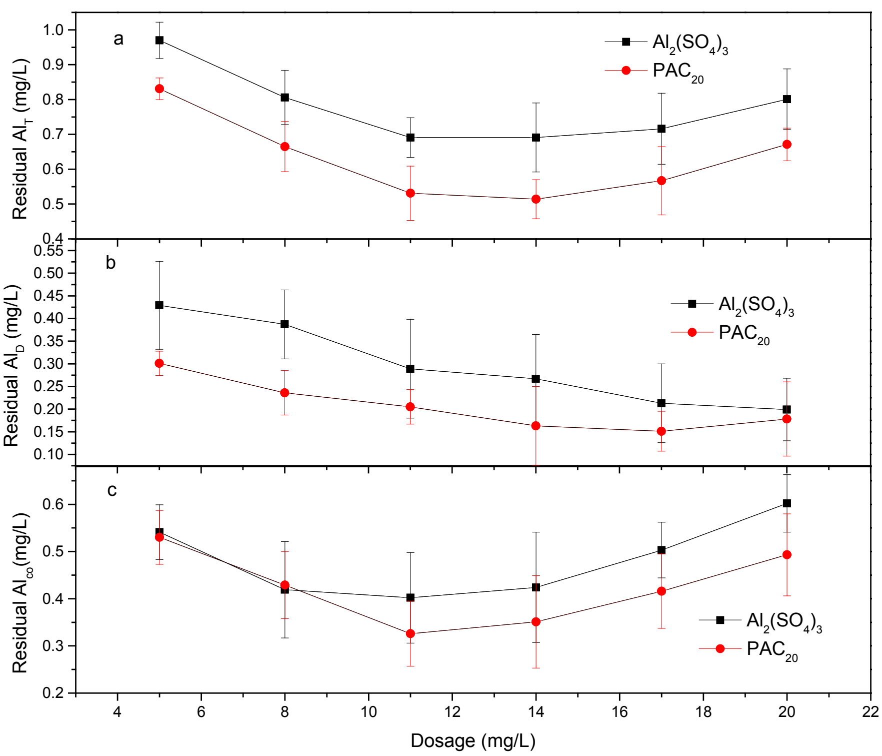

Fig. 4. Effect of dosage on Al (Al_T, Al_D and Al_{CO}) removal of both coagulants(error bars were donated as standard deviation).

Al removal performance. Specifically, for Al_T, the residual contents achieved the lowest values of 0.609 mg/L and 0.403 mg/L at the same recycling ratio of 6% of $Al_2(SO_4)_3$ and PAC_{20} separately. Additionally, with either a larger or smaller recycling ratio, Fe-WTS showed a negative effect on Al removal. Taking Al_D into consideration, the recycling ratio displayed an obvious discrepancy between the two coagulants. It was found that an appropriate recycling rate of 3% enhanced the Al_D removal performance since its concentration sharply dropped to 0.179 mg/L from 0.289 mg/L when coagulated by $Al_2(SO_4)_3$. By contrast, Fe-WTS resulted in an obvious reduction of Al_{CO} content while the recycling ratio increased to 6%, as shown in Fig. 5c, and the corresponding lowest residual Al_{CO} concentration declined to 0.243 mg/L.

3.2. Relationship between residual Al and flocs properties

With regard to technical and economic considerations, four optimized operating conditions were determined as follows (shown in Table 3): A for 11 mg/L $Al_2(SO_4)_3$; B for 11 mg/L $Al_2(SO_4)_3$ and 3% recycling ratio of Fe-WTS; C for 14 mg/L PAC_{20}; and D for 14 mg/L PAC_{20} with 6% recycling ratio of Fe-WTS.

As indicated in Table 4, the relative flocs properties under the above mentioned conditions (A, B, C and D) were summarized. It was found that Fe-WTS enhanced the average floc size dramatically, compared B and D with A and C condition as control groups. In this regard, when Fe-WTS was added, the size of floc increased from 83.5 μm (A condition) to 93.2 μm (B condition), and also went up by 16.1 μm from C condition (91.4 μm) to D condition (107.5 μm). As to D_{pf}, the influence exerted by Fe-WTS varied obviously with coagulants used. Fe-WTS notably decreased the D_{pf} value from 1.533 to 1.457 of $Al_2(SO_4)_3$ formed flocs; however, did not exerted a significant effect on PAC_{20} formed flocs.

The analysis of Al speciation during the conventional coagulation (A and C) and the recycling Fe-WTS process (B and D) is shown in Table 4. It was apparent that the recycling process improved the Al_T removal efficiency, reflected by the corresponding Al concentration in the B and D conditions of 0.516 and 0.388 mg/L, compared with the A and C conditions of 0.691 and 0.514 mg/L respectively. The variation of different Al species also demonstrated this tendency. Specifically, the reduction of Al_D when coagulated by $Al_2(SO_4)_3$ and the Al_{CO} transformation when coagulated by PAC_{20} were both derived from the addition of Fe-WTS (interpretation follows in section 4).

Table 3
Variable operating conditions

Conditions	Dosage (mg/L)		Recycling ratio (%)
	$Al_2(SO_4)_3$	PAC_{20}	Fe-WTS
A	11	–	0
B	11	–	3
C	–	14	0
D	–	14	6

Table 4
Analysis of influence of floc properties on residual Al speciation

Parameters	A	B	C	D
Size (μm)	83.5	93.2	91.4	107.5
D_{pf}	1.533	1.457	1.412	1.405
Qty	89	76	74	81
Al_T(mg/L)	0.691	0.516	0.514	0.388
Al_D (mg/L)	0.289	0.179	0.163	0.160
Al_{CO} (mg/L)	0.402	0.337	0.351	0.228

Qty: sample quantity; Al_T/Al_D/Al_{CO}: residual Al content.

4. Discussion

Superfluous residual Al (> 0.2mg/L) in drinking water has drawn greater attention due to its potential for nervous system damage [17]. Zhou et al. pointed out that the residual Al in the sludge recycling process during low turbidity water treatment exceeded the requirements of the drinking water treatment standard of China. In their study, Zhou et al. indicated that pre-treatments or subsequent treatments should be implemented or optimized to avert secondary pollution [4]. Based on the above statement, we carried out a series of trials of residual Al control by recycling Fe-WTS. From previous research, it is known that the use of Al salt coagulants may either increase or decrease the Al concentration in treated water. The corresponding results depend on Al species in the source water, the Al species in coagulant, and its transformation during water treatment [18–20].

The toxicity of Al is highly dependent on its speciation and mobility (dissolved, colloidal, and precipitated) [21]. As shown in Fig. 4b, $Al_2(SO_4)_3$ and PAC_{20} presented a similar tendency for Al_D removal and the concentrations of the A and C conditions in treated water were 0.289 and 0.163 mg/L respectively. Hence, only the C condition could achieve the requirement, which was considered as 0.2 mg/L due to the elimination of Al_{CO} during the following filtration unit. Compared with the conventional coagulation, Fe-WTS resulted in a significant difference between Al_D removal of $Al_2(SO_4)_3$ and PAC_{20} (Fig. 5b). This phenomenon can be deduced by different Al species in both coagulants, listed in Table 1. With the increased dosage of $Al_2(SO_4)_3$, a large amount of Al_a, introduced and partially hydrolyzed to Al_b and Al_c, was redundant since the negatively charged pollutants were nearly kept at charge-neutralization saturation state, and there was no more spared adsorption sites for Al_a. Therefore, a proper ratio (3%) of Fe-WTS provides more sites to attract Al_a, and the relative two reaction processes were simulated in Fig. 6a comparatively. In addition, it can be concluded (Table 4) that a lower concentration of residual Al_D combined with the decreased value of D_{pf} (comparison between A and B), which is in accordance with the findings of Jiao et al [22]. This result can be explained by the higher charging of individual Al_a, which caused the flocs to be more compact.

As to Al_{CO} removal, $Al_2(SO_4)_3$ and PAC_{20} show a similar trend, as indicated in Fig. 4c. With an increased dosage of $Al_2(SO_4)_3$, more Al_b and Al_c formed by transformation of Al_a and the quantity was not enough to precipitate. How-

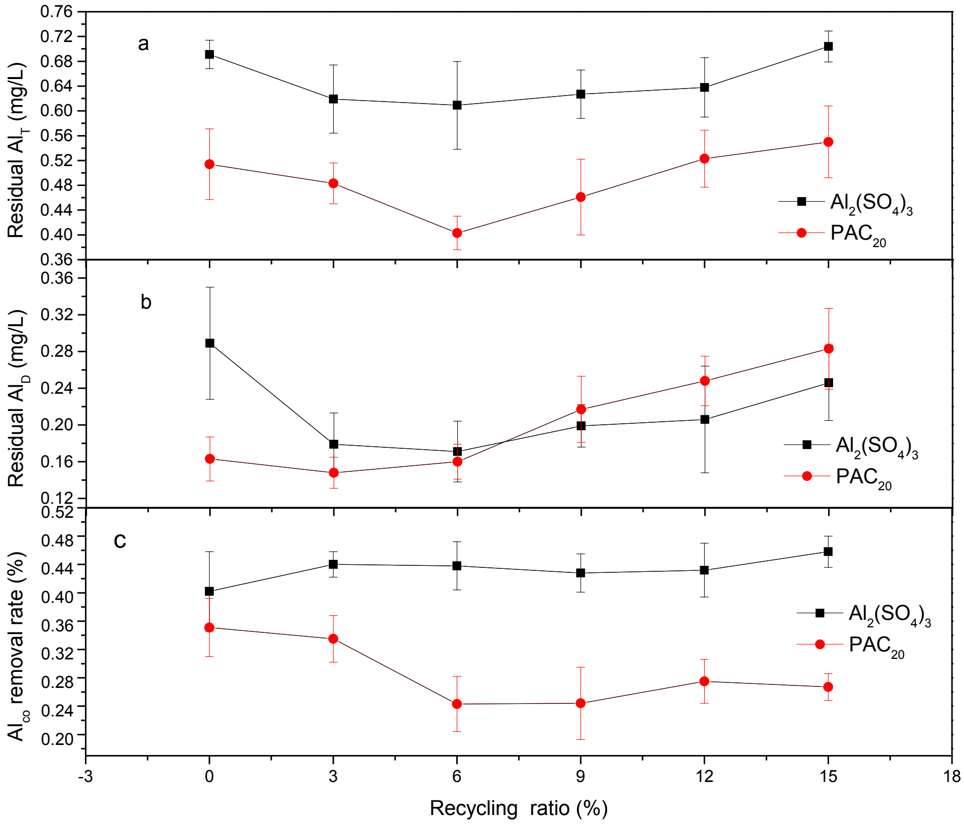

Fig. 5. Effect of recycling ratio on Al (Al_T, Al_D and Al_{CO}) removal of both coagulants. Note: (1) Fe-WTS recycling trials were conducted under the optimal dosage of each coagulant, at 11 mg/L of $Al_2(SO_4)_3$ and 17 mg/L of PACl respectively. (error bars were donated as standard deviation).

ever, Al_b in PAC_{20} was easier to transform to sediment due to the dominating percentage in the coagulant that was used. Hence, there existed a larger amount of Al_{CO} in settled water of $Al_2(SO_4)_3$ at a higher dosage range (14–20 mg/L), compared with PAC_{20}. Additionally, Fe-WTS had a more positive effect on Al_{CO} removal when coagulated by PAC_{20} as shown in Fig. 5c. This phenomenon can be explained by a mass of Al_b with larger charge intensity that attracted and combined with Fe-WTS, which enhanced the function of sweep and entrapment of Al_{CO}, as displayed in Fig. 6b. It can also be seen in Table 4 that Al_{CO} in the D condition (0.228 mg/L) was lower than in the C condition (0.351 mg/L), and the floc size of the D condition (107.5 µm) was larger than that of the C condition (91.4 µm). Consequently, we propose that Al_{CO} removal efficiency is proportional to floc size.

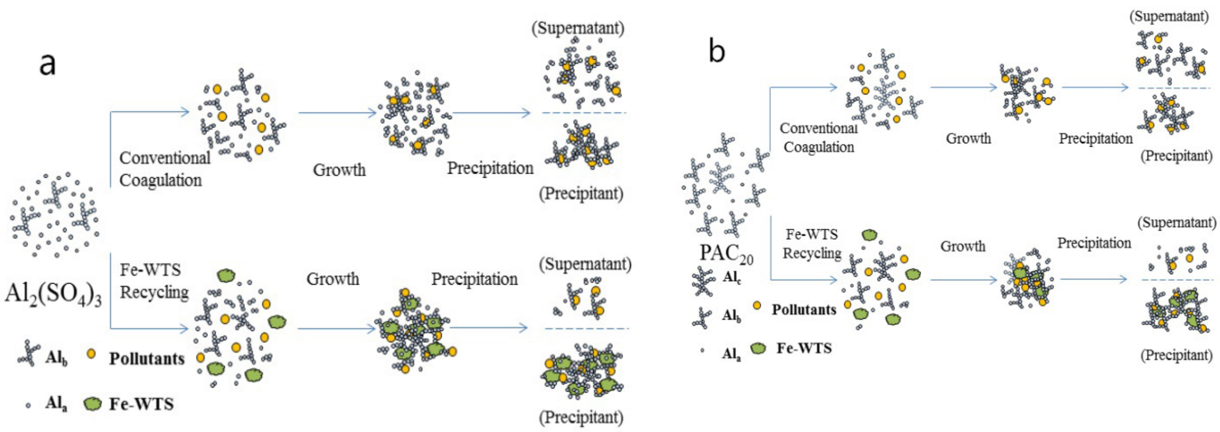

Fig. 6. A schematic diagram of Al transformation when coagulated by $Al_2(SO_4)_3$ and PAC_{20} under conventional coagulation and Fe-WTS recycling.

5. Conclusion

This research was conducted to study the effects of recycling Fe-WTS as a coagulation aid on coagulation performance and the mechanism of various Al species transformation during coagulation. The main conclusions can be drawn as follows:

(1) A proper recycling ratio of Fe-WTS enhances Al removal during coagulation. An improvement of Al_D removal when coagulated by $Al_2(SO_4)_3$ and a better reduction of Al_{CO} when coagulated by PAC_{20} were demonstrated.

(2) The flocs properties are significantly correlated with Al speciation in the treatment. Specifically, floc size is proportional to Al_{CO} removal rate, however, the value of floc fractal dimension is inversely proportional to Al_D removal rate.

(3) There is a potentially significant implication based on this work. With regard to Al removal, the Al based coagulant can be divided into two species: $Al_2(SO_4)_3$ (nearly all monomeric Al_a) and PAC_{20} (nearly all Al_b). The improvement of different Al species elimination should be based on this property.

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