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Impact of recycling alum sludge on coagulation of low-turbidity source waters

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ABSTRACT

This research project evaluated the recycling performance of alum sludge (AS), formed during sedimentation process in drinking water treatment, to enhance coagulation of low-turbidity source water. The results indicated that the recycling of AS effectively enhanced coagulation, when the optimal blended water turbidity was in the range of 13.6–21.7 NTU, with solid content of 0.072–0.124%. Furthermore, recycling AS could reduce up to 40% of fresh coagulant dosage in coagulation–sedimentation process. Bench-scale experiment results showed recycling AS prior to coagulation had insignificant effect on effluent water quality for all measured parameters: color, NH_3-N, COD_{Mn}, UV_{254}, aluminum, and manganese. Scanning electron microscopy evidence revealed that the floc structures with AS were more smooth and more compact than that without AS. It was postulated that the aluminum hydroxide precipitate in AS provide nucleating sites for physical collision and that sweep coagulation might play a key role in the enhancement of coagulation in low-turbidity source water.

Keywords: Recycle; Alum sludge; Blended water; Coagulation; Floc structure

1. Introduction

Large quantities of alum sludge (AS) are generated during coagulation–sedimentation process in drinking water treatment plants (DWTPs) all over the world every day. The proper disposal, recovery, or reuse of AS has thus become a significant environmental issue. In the past, DWTPs residuals, especially AS, have sometimes received insufficient attention in the planning and design of water treatment facilities, leading to < optimal methods of management of these residuals. Many DWTPs have installed residual treatment facilities to comply with increasingly stringent effluent discharge regulations. At present, the methods of management of AS are a treated effluent that is often returned to the source water and a concentrated solid residual stream that is dewatered and disposed of in a landfill [1]. Since the levels of pollutants or hazardous substances in such sludge are relatively low, the recovery and reuse of spent coagulant may be feasible [2]. AS contains a large portion of insoluble aluminum hydroxides that can be utilized as additional coagulant to enhance pollutant removal from wastewater [3,4]. It has been demonstrated that the use of AS was a good way of removing lead or phosphorus from wastewater and reducing the fresh alum dosage [3,5].

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For natural waters of low-turbidity, these essential floc nucleating sites are limited, and need to be provided artificially [6]. Conventional methods include adding more polymer coagulant or coagulant aids to enhancing binding ability [7], or using clay to increase collision sites. All the ways not only increase the cost of water treatment, but also augment the by-product of wastewater. Reservoirs are increasingly constructed to supply raw water instead of river. Reservoir water generally has the property of low turbidity, which brings a great deal of difficulty in water purifying. Based on the existing problems during treatment of low-turbidity water in DWTPs, alternative methods need to be developed. Previous studies demonstrated that recycling water residual stream, especially spent filter backwash water, introduced additional turbidity into the raw water, and might have enhanced coagulation through increased number of collision sites [1,8–10]. Since AS contains much more hydroxide precipitates than filter backwash water, it might be effective on enhancing the coagulation process in drinking water treatment. Existing literature showed the benefits of recycling AS directly to raw water. Bourgeois suggested that recycling 5% of a combined residual stream of filter backwash water and clarifier sludge on a low-turbidity source water could improved settled water quality as quantified by total organic carbon (TOC) and UV_{254} [11]. Zhou et al. found that coagulation efficiency was improved when reusing AS combined with particle activated carbon on drinking water treatment [12]. The study of Qi et al. had demonstrated that sedimentation basin performance was improved concerning turbidity, DOC, and UV_{254} when untreated AS was recycled ahead of the coagulation process [13]. Therefore, recycling AS directly to source water might be an alternative method to improve coagulation efficiency of low-turbidity water and reclaim 2–10% of waste stream simultaneously. This method accomplishes the reuse and the reclamation of water resources, and fulfills a low-operational cost due to low coagulant addition and sludge disposal.

This research project focused on the activities of untreated AS and examined the impact of recycling AS to enhance coagulation of low turbidity water. Bench-scale experiments were conducted using a standard jar-test apparatus to evaluate removal of turbidity and some water quality parameters, color, NH3-N, COD_{Mn}, UV_{254}, aluminum, and manganese. Scanning electron microscopy (SEM) images were also taken to compare the changes in the floc structures with or without AS recycling for investigating the possible mechanism involved in pollutant removal enhancement by AS.

2. Materials and methods

2.1. Source water and AS

The raw water and the AS used in the bench-scale experiments were obtained from one drinking water treatment plant in Harbin, China (Mopanshan DWTP). It has a conventional flocculation–sedimentation–filtration process. The Mopanshan DWTP has two sets of $45 \times 10^4\ m^3/d$ flow rate that mainly serves the area of Harbin City, China. The plant uses poly-aluminum chloride (PACl) as the primary coagulant at an average dosage of 80 mg/L. Lime is added to adjust pH. For one-set, the sedimentation sludge is reported to be wasted at a rate of approximately $1.5 \times 10^4\ m^3/d$. The settled water then passes onto dual-media filters that are backwashed on average every 72 h. The average daily backwash flow was calculated to be approximately $0.62 \times 10^4\ m^3/d$. Thus, the total plant residual volume is approximately 5% of the total plant outflow.

The raw water is from Reservoir Mopanshan. A summary of the raw water parameters is presented in Table 1. Temperature is 0–15℃, turbidity is 0.5–5.0 NTU, color is 18–45 CU, and COD_{Mn} is 2.2–7.0 mg/L. The raw water is a typical case of low temperature, low turbidity, and high color water. The characteristics of AS also is stated in Table 1. Turbidity is 200–600 NTU, color is 500–1,500 CU, and COD_{Mn} is 15–20 mg/L. The substance which cause color mainly is humic acid.

2.2. Bench-scale equipment

Bench-scale experiments were performed using a standard jar test instrument (Zhongrun Model ZR4-6, China) consisting of six 1.5-L square jars marked with sample ports at 10 cm depth filled to the 1-L blended water, prepared by compositing variation of AS concentration to raw water at a volumetric ratio of

Table 1
Water quality characteristics

	Units	Raw water	Alum sludge
Temperature	℃	0–15	0–15
pH	Units	6.6–7.0	7.0–7.2
Turbidity	NTU	0.5–5.0	200–600
Color	CU	18–45	500–1,500
NH_3-H	mg/L	0.04–0.30	–
COD_{Mn}	mg/L	2.2–7.0	15–20
UV_{254}	cm^{-1}	0.100–0.300	–
Aluminum	mg/L	0.110–0.212	0.211–0.412
Manganese	mg/L	0.011–0.023	0.014–0.028
Zeta potential	mV	−11.4 to −13.8	2.99–6.40

5:100. Fresh alum coagulant was injected using graduated syringes and rapidly mixed at 300 rpm for 1 min. Following the rapid mix, three-stage flocculation was employed for 5 min each at 120, 60, and 40 rpm, respectively. In addition, control tests were performed without recycling AS. Sedimentation effluent was sampled at 20 min settling for turbidity, color, NH_3-N, COD_{Mn}, and UV_{254}. Manganese and aluminum were also sampled.

2.3. Analytical methods

Water quality parameters were measured using standard testing procedures as recommended by the USEPA and/or as outlined by standard methods. In particular, temperature and pH were measured using an Orion pH meter. Turbidity was measured using a HACH 2100P turbidity meter. Color, aluminum, and manganese were all measured using a HACH DR/2010 spectrophotometer. UV_{254} was measured with a UV/VIS spectrophotometer (Phillips PIE INICAM). COD_{Mn} and NH_3-N measurement followed standard procedures of the Chinese Environmental Protection Bureau. Zeta potentials (ZP) of the samples were measured with a Zeta potential analyzer (Nano-Z, England). SEM (JSM6335) was used to analyze the samples of coagulation flocs with and without recycling AS. After freeze–drying in the lyophilizer, the samples were coated with a thin layer of gold to make the specimen conductive and emit secondary electrons. Samples were observed in SEM, using scan voltage 10 kV, with 5,000 times magnification.

3. Results and discussion

3.1. Coagulation performance of AS

Generally, coagulant dosage is much higher in this DWTP since the raw water has the characteristics of low turbidity. In order to examine whether the solids in AS has the equivalent active performance for coagulation, the first set of trials with jar test apparatus were performed without adding the coagulant. These results were shown in Fig. 1. The turbidity difference meant the residual turbidity difference between the control trials and that of blended water after 20 min settling. The blended water was composted by raw water with recycling AS. The blended water turbidity (BWT) increased with the increasing of recycling ration of AS, and there was a good linear correlation between BWT and blended water solid content (SC), as shown in Fig. 2. Color difference, COD_{Mm} difference, and UV_{254} difference in Fig. 1 had the same meaning.

Results in Fig. 1(a) and (b) shows that the removals of turbidity and color are increased with the addition of AS. The turbidity difference is negative at low BWT and the reversal occurs around BWT at 33 NTU, with SC of 0.218%. At higher BWT, the turbidity difference increases slightly. More color removal was achieved for the BWT increasing. The color difference changed from negative to positive when the BWT was 15 NTU, with SC of 0.074%. Similar tendencies of the COD_{Mn} difference and UV_{254} difference are presented in Fig. 1(c) and (d). With the BWT increasing, both of the differences increased. The COD_{Mn} difference changed from negative to positive with the BWT at 19 NTU, while UV_{254} difference changed at 12 NTU, with SC of 0.105 and 0.061%, respectively. These findings indicate that the AS does contain active coagulant ingredients, which can play a positive role as a recycling agent.

3.2. Effect of AS to enhance coagulation

In order to investigate the effect of recycling AS to enhance coagulation, the second trials with jar test apparatus were performed by adding fresh alum coagulant (Fig. 3). Obviously, with the same dosage of

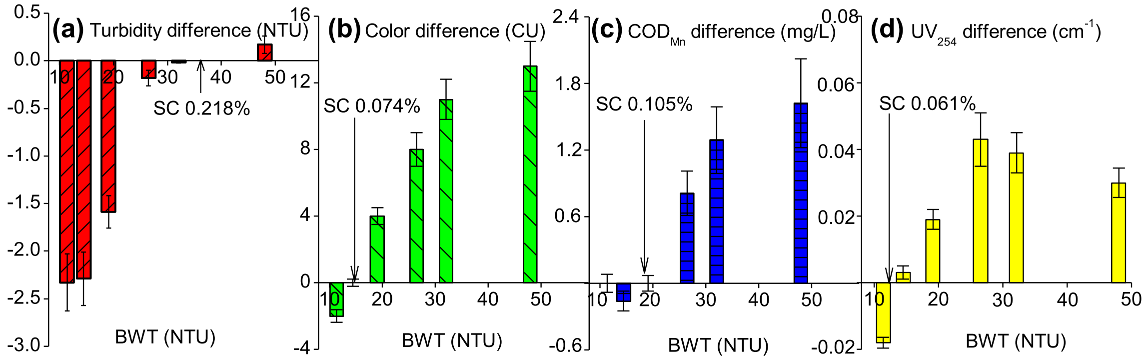

Fig. 1. Effects of recycling AS on coagulation.

coagulant, adding AS in to raw water gave the better turbidity removal efficiency in comparison to the control test. At a low coagulant dosage of 40 mg/L, the residual turbidity was 1.0 NTU in raw water, while it was lower than 1.0 NTU in all blended water. With increasing BWT, the residual turbidity gradually decreased to 0.6 NTU. Increasing BWT to 21.7 NTU, SC 0.124%, further reduced residual turbidity to 0.4 NTU. Subsequently, the turbidity removal was limited when the BWT was followed by 32.5 NTU. These findings indicated that particles existing in AS increased the concentration of particles in raw water and became the cores of flocculation itself to enhance coagulation, consistent with the findings of Zhou et al. and Qi et al. Both studies claimed that reusing AS was an alternative method to enhanced traditional coagulation.

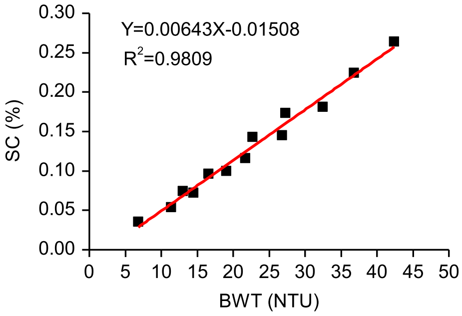

Fig. 2. Linear correlation between BWT and SC.

Moreover, recycling AS can reduce the dosage of fresh alum coagulant. In many scientific papers, optimal coagulation conditions are judged on the optimal turbidity removal after a fixed period of settling. In this work, optimal coagulant dosage was judged on 0.6 NTU in settled water (Mopanshan DWTP’s internal guideline). As presented in Table 2, most of the optimum coagulant dosage (OCD) of blended water were lower than that of raw water with OCD at 56.7 mg/L. The minimum value appeared with the OCD in the BWT range of 13.6–21.7 NTU, with SC range of 0.072–0.124%. When the BWT was within this range, AS recycling had a positive effect on reduction of coagulant dosage. The maximum reduction could reach up to 40%.

SEM examination of floc structure was conducted. Without recycling AS, the surface morphology of the flocs was porous, as shown in Fig. 4(a). There were apparent crevices and branches on the flocs surface without AS. On the contrary, with AS, the surface morphology of the flocs (Fig. 4(b)) is more smooth and more compact than that in Fig. 4(a). And there are particles absorbed on the flocs, which were conducive to the enmeshment of impurity particles in raw water through adsorption bridging.

3.3. Impact of recycling AS on water quality

Previous studies suggest that recycling untreated AS introduced additional turbidity into the raw water

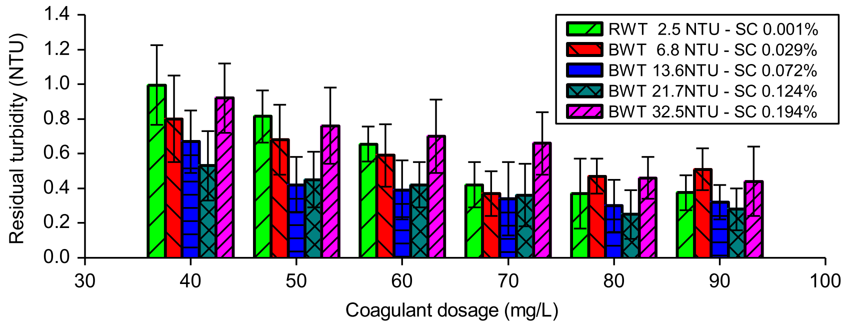

Fig. 3. Residual turbidity in settled water under different coagulant dosages at variation of BWT.

Table 2
OCD of the raw water and different BWT

	Raw water		Blended water					
Turbidity (NTU)	2.5	6.8	13.6	18.2	21.7	26.8	32.5	44.2
OCD (mg/L)	56.7	51.2	42.8	38.6	32.1	47.8	73.0	62.2
SC (%)	0.001	0.029	0.072	0.102	0.124	0.157	0.194	0.269

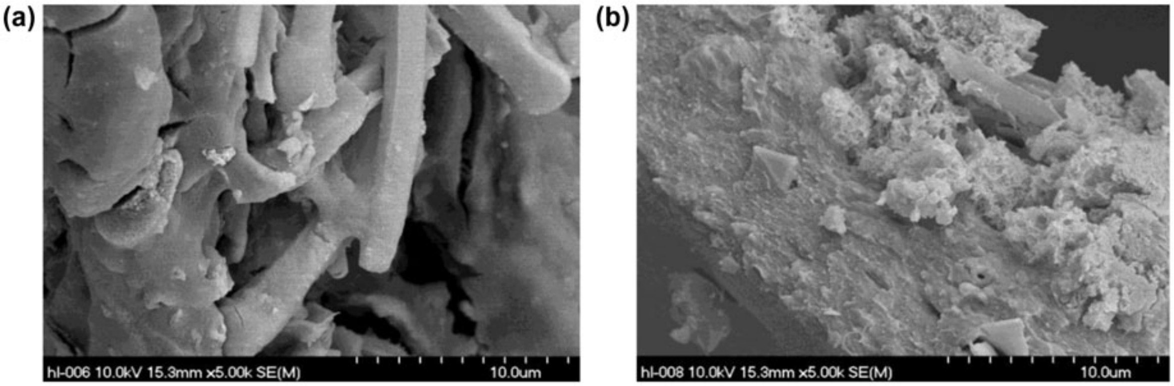

Fig. 4. SEM images of flocs surface (5000× magnification) of (a) raw water with OCD at 60 mg/L and (b) blended water with OCD at 35 mg/L.

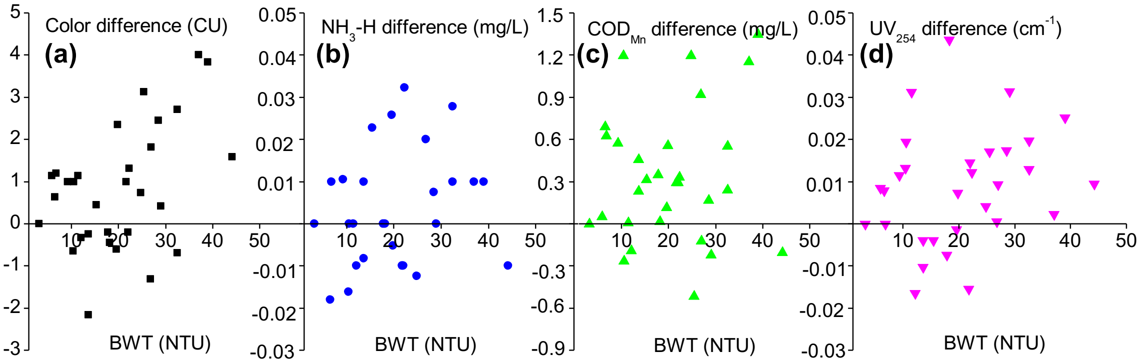

Fig. 5. Effect of BWT on water quality parameters (a) color, (b) NH_3-N, (c) COD_{Mn}, and (d) UV_{254}.

that may have improved settled water quality as quantified by TOC and UV_{254} measurements [13–15]. In order to determine the effect of recycling as on water quality comparison with that of raw water, some water quality parameters were measured, including residual color, NH_3-N, COD_{Mn}, and UV_{254}. Fig. 5 showed the difference of all measured parameters. This trial of tests was operated with the optimal coagulant dosage that attained base on residual turbidity settled at 0.6 NTU (internal guideline).

From Fig. 5(a) and (b), we can see that most data of color difference and NH_3-N difference are greater than zero, accounting for 66.7 and 70.0%, respectively. The findings indicated that the turbidity-causing matter was advantageous for removing color through coagulation [16]. Since in this kind of raw water, the color-causing matter mainly is humic acid, color was removed with the removal of nature organic matter.

It is becoming more important to utilize additional water quality parameters to evaluate AS recycling performance and optimize operations. COD_{Mn} and UV_{254} are important parameters to evaluate natural organic matter (NOM) removal performance in DWTPs. Fig. 5(c) and (d), shows that most data of COD_{Mn} difference and UV_{254} difference are greater than zero, accounting for 80.0 and 83.3%, which are higher than that of color difference and NH_3-N difference. The result indicated NOM could be attributed to the increased number of attachment sites with the introduction of AS for precipitating dissolved organic matter in the raw water. At natural water treatment conditions (pH 6–8), aluminum hydroxide fraction dominates among the hydrolysis reaction products in water and the NOM removal efficiency is dependent on the adsorption of the humic substances on the aluminum hydroxide crystals, and sweeping flocculation plays a key role [17]. In this research, coagulation achieved a moderate removal for COD_{Mn} and UV_{254}, and recycling AS had an enhancement effect to coagulation, which may be due to the adsorption and sweeping flocculation by a large number of aluminum hydroxide complexes and precipitates in AS. Similarly, the collective results of improved reduction in TOC, UV_{254}, and color found by Bourgeois were attributed to the increased number of attachment sites with the introduction of AS for precipitating dissolved organic matter in the raw water [11]. Besides, in the optimal BWT range of 13.6–21.2 NTU, most of COD_{Mn} difference and UV_{254} difference were greater than zero, indicating that recycling untreated AS in this concentration range under a low coagulant dosage enhanced the removal of NOM instead of causing the accumulation of organic matter. This phenomenon was interpreted as that the majority of the organic matters present in untreated AS were not in the dissolved form and the majority of absorbable organic matters would be enmeshed in alum flocs through coagulation in the main process train [18].

PACl was added in this study and concentration of soluble residual aluminum in settled water was evaluated in this study, which poses a risk towards Alzheimer's disease as recycling back to the main trial. Fig. 6(a) showed the concentration difference of residual aluminum between raw water with blended water matrix. All data of alum difference were larger than zero. The concentration of residual aluminum of blended water was lower than that of raw water. The raw water of this DWTP was low-turbidity water, 1.56 NTU. Duan and Gregory had found that at low particle concentration, low coagulant dosages should be required if charge neutralization was the predominant destabilization mechanism [19]. However, under dosage of coagulant gave charge neutralization and formed micro-floc, which had not enough density to settle from water. The residual turbidity at this range does not meet the internal guideline. Higher dosage needed to clarify the low-turbidity water, resulting in high level of residual aluminum, which was not incorporated into micro-floc for lacking of binding sites. In the presence of AS, i.e. particles increased, the OCD was lower than control trial, which not only enhanced the coagulation but also reduced the concentration of residual aluminum. This finding noted that recycling AS prior to coagulation had an advantage on aluminum removal.

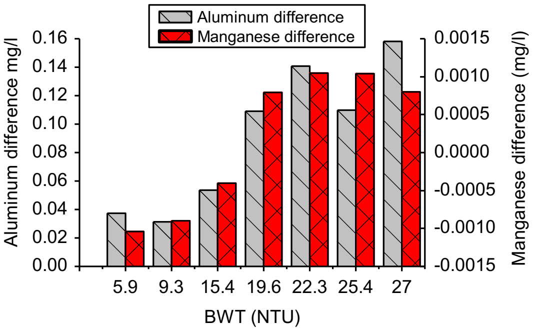

Fig. 6. Effect of BWT on removal of alum and manganese (a) alum, and (b) manganese.

The potential for high manganese concentrations in recycled AS was also evaluated at this study. Fig. 6(b) showed the concentration difference of soluble residual manganese between the control trials with blended water matrix. The manganese difference increased from −0.0010 to −0.0004 mg/L with the BWT increasing from 5.9 to 15.4 NTU. With the BWT continuing rising up to 27 NTU, the difference continued increasing through a maximum, with BWT at 22.3 NTU, then began to deteriorate. Within the range of 19.6–27 NTU, the differences of residual manganese in the settled water were larger than zero, which was meant for recycling suitable concentration of AS enhanced manganese removal efficiency. Some of similar effects had previously been described. Cornwell suggested the proper management of waster steams could render manganese suitable for recycling filter backwash water through investigating 24 water treatment plants [17]. He found that the manganese concentration in the sludge would increase with storage time as more manganese was released from the solids. Anaerobic conditions should theoretically promote the release of manganese from the solids into the liquid state. Bourgeois et al. noted that manganese levels were much lower in the sedimentation effluent opposed to the dissolved air flotation and gravity thickener. In gravity thickener, effluent quality with respect to manganese began to deteriorate because of re-solubilizaton [14]. This study noted recycling untreated AS had less than potential of manganese releasing from sludge than recycling effluent of gravity thickener, lagoon, and dissolved air flotation.

In short, the water quality from recycling trials met or exceeded the control trials quality of the source water for all measured parameters, color, NH_3-N, COD_{Mn}, UV_{254}, aluminum, and manganese.

3.4. Mechanisms

AS contains 60–90% of the total flocculated solids in DWTPs. One of the mechanisms on recycling AS to enhance coagulation performance attributed to the increase in the number of collision sites through increasing concentration of particles. Natural turbidity provides a ready source of nucleating sites for floc development. For low-turbidity source water, the absence of nucleating sites resulted in poor macro-floc formation. One of the greatest practical problems faced in treating low-turbidity source waters is the inability to produce an acceptable floc. Even if the coagulant dosage conditions for micro-floc formation have been optimized, macro-floc development and floc settling may be limited [6]. In this work, recycling AS prior to coagulation increased particles into low-turbidity source water, resulting in providing sufficient nucleating sites for floc development.

For further understanding of the AS mechanism in water treatment process, ZP of AS and raw water were measured (Table 1). Specifically, charge analysis showed that the particles present in the AS stream were predominately destabilized (i.e. ZP = 2.99–6.40 mV) in comparison to the raw water that contained predominantly stabilized particles (i.e. ZP = −11.4 to −13.8 mV). The introduction of the AS with alum-destabilized particles would result in a change on surface charge of the particles in the blended water matrix. These results demonstrated that the addition of AS into raw water, forming a coating on the impurity particles, resulted in a shift from negative to positive in the bulk surface charge of water particle. These ZP measurements were in agreement with bench-scale experiment results. According to the DLVO theory, ZP is a measure of the excess number of electrons found on the surface of all particle matter. The magnitude of the charges determines whether colloidal-size particles in suspension will repel one another and remain in suspension or agglomerate eventually settled. The more negative the ZP, the stronger the repelling force. The optimal ZP is close to zero. Since the ZP of AS was greater than that of the raw water, recycling AS can increase the ZP of raw water to get closer to zero. So the AS recycle neutralizes or reduces ZP of the colloidal-size particles allowing the force of attraction to pull particles together, as the role of coagulant, indicating that recycling AS has the effect of strengthening coagulation and saving coagulant.

In conclusion, in the case of conventional coagulation process, sufficient coagulant dosage gives charge neutralization leading to micro-flocs forming rapidly. Subsequently, as the alum dosage is increased, extensive hydroxide precipitate formed *in-suit* gives sweep flocculation, entrapping particles out of water. However, if the source water had the characteristics of low-turbidity, floc nucleating sites need to be provided artificially. Recycling AS took the effect of providing particles for enhancing coagulation. In the case of recycling AS at specific recycle rates ahead of coagulation, bulk hydroxide precipitates contained in AS initially form very small colloidal particles (a few nm in size), which are positively charged at around neutral pH. It is likely that some of these particles form a coating on the impurity particles in the low-turbidity source water, reversing their charge. Simultaneously, the AS provided sufficient nucleating sites for floc development. Subsequently, with addition of fresh coagulant, new precipitate coats the flocs so that conventional aggregation of the colloidal hydroxide particles occurs, either on the particle surfaces or in bulk solution, corresponding to macro-floc forming.

4. Conclusions

This work draws three main conclusions: (1) AS had the function of enhancing coagulation effect in low-turbidity source water, and reducing coagulant dosage. Save coagulant rate was up to 50% in the optimal BWT range, 13.6–21.7 NTU, with the recycling SC of 0.072–0.124%. (2) Water quality from the optimal trials met or exceeded the control trial quality of the source water for all measured parameters, color, NH_3-N, COD_{Mn}, UV_{254}, aluminum, and manganese. (3) AS recycling could strengthen coagulation performance dominantly by the sweep coagulation of amorphous hydroxide precipitate in AS, which enmesh impurity particles into floc and provide nucleating sites for floc forming, and slightly positive particles in AS, which could reverse surface charge of floc.

Recycling AS to enhance coagulation performance is not only effective, but also a potentially economical approach to reducing wastewater volume. Therefore, this study strengthens the body of literatures of reusing such sludge directly back to the head of coagulation in low-turbidity source water treatment engineering.

Acknowledgments

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