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The impact of recycling sludge on water quality in coagulation for treating low-turbidity source water

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ABSTRACT

There is an alternative method for the treatment of low-turbidity source water that reuses coagulation sludge as low-turbidity raw water to enhance conventional coagulation efficiency. This research investigated the impact of the recycling ration by volume of coagulation sludge on enhancing the removal efficiency of specific water quality parameters in a pilot-scale experiment with a capacity of 5 m^3/h. This novel test was first continuously run for 150 h (15 recycling runs), and the recycling sludge was cycle used. The results showed a significantly higher removal efficiency of turbidity from the raw water than that of the traditional process, which was probably attributed to the special long-range Van der Waals attractive force, the enhancement of the collision ratio among particles, and the similar "second dosage of coagulant" strategy after the recycling process. Charge neutralization and absorption have mainly enhanced the removal efficiency of organic matter with recycling sludge. To some extent, this enhancement is due to the open and porous structure of the reused sludge, which could absorb the metal ions and further remove them following the sedimentation process. Recycling sludge is a feasible and successful method to enhance pollutant removal and has a significant effect on the treatment of low-turbidity source water.

Keywords: Recycling sludge; Coagulation efficiency; Low turbidity; Drinking water treatment

1. Introduction

In a traditional low-turbidity water treatment process, the removal ability of particulates would become weaker in coagulation because of the relatively slow hydrolysis of the coagulant, stronger water viscosity, and slower settling velocity of flocs [1,2]. Thus, recycling drinking water treatment (DWT) sludge was proposed as a novel method to optimize the traditional treatment technology as expected for the improvement of the flocculation efficiency for low-turbidity water. Edzwald et al. [3,4] investigated the effect of directly recycling the filter backwash water (FBW) on different treatment units and found that the recycling of FBW was not in proportion to the large increase in the clarifier influent solids. Qi et al. [5] found that upon the reuse of poly aluminium chloride and alum sludge to treat natural surface water prior to ultrafiltration process, the removal efficiency of turbidity could increase to 80.2% than that of the conventional process. Chu et al. [6–8] considered that flocculation and filtration

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units could be strengthened at a lower recycling volume of sludge, whereas this method appreciably reduced the risk of direct recycling. In addition, the process of recycling sludge may potentially risk the drinking water quality. Whether the contaminants (dissolved organic carbon [DOC], UV_{254}, aluminum, and iron) could be further released from the recycling sludge during continuous recycle trials remains unknown.

Based on the discussion above, the overall purpose of this research was to study the feasibility of recycling sludge to enhance the traditional coagulation efficiency for treating low-turbidity water in a pilot-scale experiment; to evaluate some water quality parameters, turbidity, color, DOC, UV_{254}, Fe, Al, and Mn; and to further investigate the possible mechanism of pollutant removal enhancement by sludge.

2. Materials and methods

2.1. Coagulant and characterization of source water and sludge

Polyferric aluminum chloride (PFAC) was industrial grade (with contents of 8.1% Fe_2O_3 and 3.3% Al_2O_3 and a basicity of 6.2%; Zibo, China).

The source water that originated from Nenjiang River was obtained from the intake of the Zhongyin water treatment plant (Daqing, China). The water was characterized as a typical slightly polluted surface water with low turbidity and color. The sludge that was used in this study was taken from the outlet of the sedimentation tank. The sludge samples had serious contamination compared to the raw water samples, and the detailed characteristics of raw water and sludge are shown in Table 1.

2.2. Experimental setup and procedure

The schematic of the pilot treatment processes is shown in Fig. 1. This setup is a typical DWT process with a flow rate of 5 m^3/h. The first unit is a grid flocculation tank with a bottom length L = 1,100 mm, a bottom width D = 400 mm, and a liquid height H = 1,700 mm. The following unit is a plate sedimentation tank with a bottom length L = 2,100 mm, a bottom width D = 800 mm, and a height H = 1,600 mm. In addition, there were 63 plates with a 60° tilt angle and 20-mm interval in this unit. The last unit is a rapid filter tank with a filtration velocity of 8 m/h. The recycle sludge was stored in two homemade cylinder sludge storage tanks with a diameter and height both of 1,500 mm. The sludge was pumped to the head of static pipeline mixer after being completely mixed. The whole hydraulic retention time was 44 min.

Table 1
Source water and sludge characteristics

Parameter	Units	Raw water	Sludge
Turbidity	NTU	4–7	60–2,560
Color	CU	28–35	350–420
COD_{Mn}	mg/L	5–9	18–35
UV_{254}	cm^{-1}	0.065–0.076	0.081–0.098
TOC	mg/L	4.4–5.6	4.85–6.2
DOC	mg/L	3.85–4.65	4.70–5.01
Solid content	w/w%	0.001–0.006	0.05–1.8
pH		5.8–6.5	5.7–6.8
Temperature	℃	3–10	4–12

An interesting reusing method was used in our research in which the discharging and recycling processes of sludge were simultaneously performed. The residual sludge was collected in the sedimentation process (label 4 in Fig. 1) every 10 h (10 h equals 1 recycling run), and then this sludge was released into the sludge storage tank (label 7 in Fig. 1) through a PVC pipe that was 100 mm in diameter. The discharged sludge was recycled from the storage tank to the head of the static mixer by a peristaltic pump via a rubber hose of 10 mm in diameter, ensuring that the residual sludge could be completely reused under the optimal treatment combination. Then, the removal efficiencies of turbidity, color, TOC, UV_{254}, Fe, Al, and Mn by the dosage of recycle sludge were studied, and the appropriate recycle ratio was obtained. In addition, the continuous recycle trial was conducted under the optimal sludge recycle ratio (SRR) of sludge to test the stability of this novel process. With this method, this pilot plant test was continuously operated for 150 h (15 recycling run), and the discharged sludge could be repeatedly used many times until the determined parameters exceeded the sanitary standards for drinking water of China. When an odor was produced by sludge in the sludge storage tank, all of the sludge was discharged, and the new sludge was re-collected in the following process cycle.

2.3. Analytical methods

The turbidity was measured with an HACH2100P turbidimeter (Hach Company, Loveland.CO, US). The DOC and total organic carbon (TOC) were analyzed by a TOC-V CHP analyzer (Shimadzu Corporation, Kyoto, Japan). The UV absorbance at 254 nm (UV_{254}) was determined using a DR5000 UV/VIS spectrometer (Hach Company, Loveland.CO, US). Both DOC and UV_{254} were measured after filtration through 0.45-μm membranes. The solid content of the recycled sludge

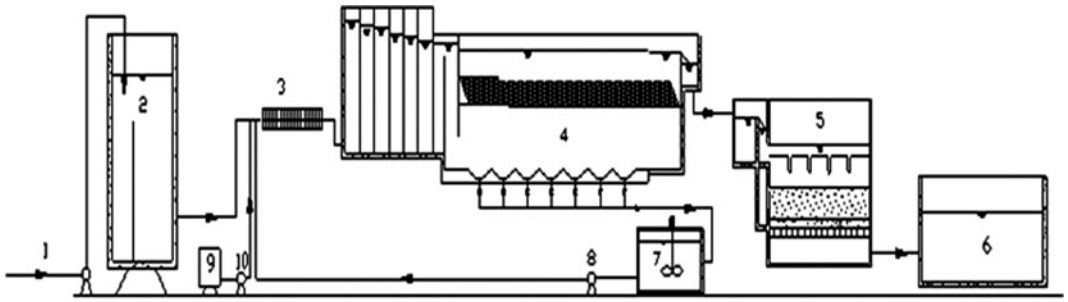

Fig. 1. Diagram of the pilot-scale experimental setup.
Notes: (1) Raw water; (2) Regulating well; (3) Static pipeline mixer; (4) Grid flocculation tank; (5) Conventional rapid filter; (6) Clean water reservoir; (7) Sludge storage tank; (8) Sludge recycle pump; (9) Coagulant dosing pool; and (10) Dosing pump.

was examined following the Standard Methods (APHA, 1995), and the pH was measured using a pH meter (PHS-3C, China). The water samples from the recycling process were simultaneously analyzed for total concentrations of Al, Fe, and Mn by inductively coupled plasma atomic emission spectrometry ICP-AES (Horiba JobinYvon, France).

3. Results

3.1. Effect of the sludge dosage on contaminant removal

3.1.1. Effect of the sludge dosage on the removal efficiency of turbidity and the apparent color

As previously stated, the raw water in this study had low turbidity (i.e. 4–7 NTU). The coagulation performances of different recycling conditions were first investigated in terms of turbidity and apparent color, as demonstrated in Fig. 2. For a 10 mg/L dosage of coagulant, the removal efficiency of turbidity and color exceed 85 and 78%, respectively, with an SRR in the range of 1–3%, whereas these values decreased to 81 and 72%, respectively, at a 5% SRR. At a 7 mg/L dosage of coagulant (E, G), the removal efficiency of turbidity and color was much lower than any other combinations with an SRR of 1 and 5%. For combination F (3% SRR + 7 mg/L PFAC), the removal efficiency was as high as 86% for turbidity and up to 83% for the apparent color. On the premise of guaranteeing the quality of outflow, the main aim was to maximize the reusing amount of sludge and minimize the dosage of coagulant. Therefore, the combination F was competent in this study. The results indicate that the appropriate combination of SRR and dosage of coagulant is crucial for the removal of turbidity and color.

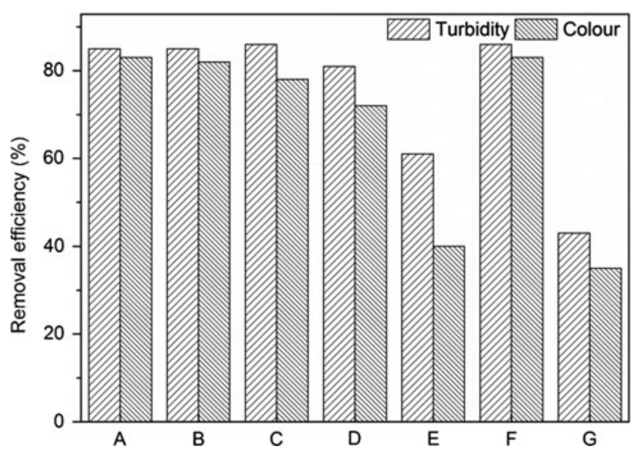

Fig. 2. Effect of sludge recycling ratio and different dosage of coagulants on turbidity and color removal.
Notes: (A-raw water + 10-mg/L PFAC, B-1% SRR + 10-mg/L PFAC, C-3% SRR + 10-mg/L PFAC, D-5% SRR + 10-mg/L PFAC, E-1% SRR + 7-mg/L PFAC, F-3% SRR + 7-mg/L PFAC, and G-5% SRR + 7-mg/L PFAC).

3.1.2. Effect of the sludge dosage on the removal efficiency of DOC and UV_{254}

Both DOC and UV_{254} are important surrogate parameters that can represent the content of dissolved organic matter (DOM) in DWT. DOM poses potential threats to human health and is difficult to be removed by the conventional treatment process. In this study, the removal of DOC and UV_{254} during this recycling process was also investigated to evaluate the security of drinking water, and the results are illustrated in Fig. 3. The UV_{254} removal rates for the four types of recycling conditions were as follows: C > B > A > D. The optimal removal efficiency was at 3% SRR and

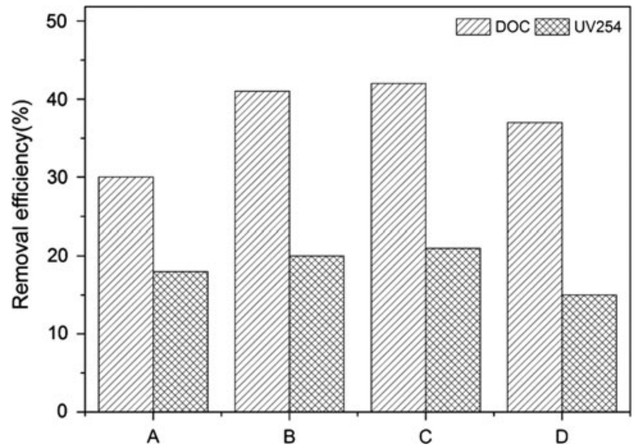

Fig. 3. Effect of sludge recycling ratio and different dosage of coagulants on DOC and UV_{254} removal.
Notes: (A-raw water + 10-mg/L PFAC, B-3% SRR + 10-mg/L PFAC, C-3% SRR + 7-mg/L PFAC, and D-5% SRR + 7-mg/L PFAC).

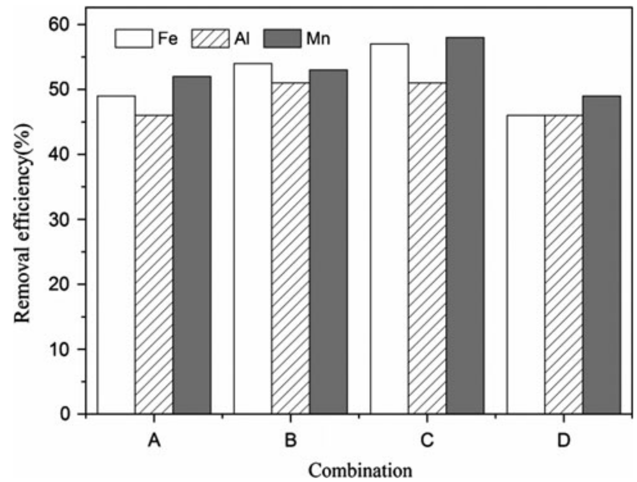

Fig. 4. Effect of sludge recycling ratio and different dosage of coagulants on Fe, 、Al, and Mn removal.
Notes: (A-raw water + 10-mg/L PFAC, B-3% SRR + 10-mg/L PFAC, C-3% SRR + 7-mg/L PFAC, and D-5% SRR + 7-mg/L PFAC).

7-mg/L PFAC. In terms of DOC, 3% SRR performed better than did 5% SRR under the same dosage. Additionally, 7-mg/L PFAC exhibited superior DOC removal to that of 10 mg/L, and the corresponding DOC removing efficiency was as high as 42%. Without recycling sludge, the removal efficiencies were barely 30 and 18% with these monitoring parameters. Generally, the removal capacity for DOM has been appreciably improved via the recycling sludge process in contrast to that of the traditional coagulation process. The removal efficiency and reagent cost are considered the two important indexes for feasible treatment. Therefore, combination C (3% SRR + 7 mg/L) was chosen as the optimal condition, corresponding to the removal efficiency of turbidity and apparent color.

3.1.3. Effect of the sludge dosage on the removal efficiency of metal ions

Hydrolyzing metal ions are very widely used as coagulants to remove many impurities from polluted water. In contrast, the enrichment of these ions in drinking water will potentially threaten human health. In our research, the amorphous aluminum and iron and manganese that were contained in PFAC were usually detected in raw water. Therefore, the typical ions of aluminum, iron, and manganese were chosen as indicators in our sludge-recycling process. Fig. 4 shows the effect of various recycling ratios on the removal efficiencies of iron, aluminum, and manganese. In terms of iron, combination C (3% SRR + 7-mg/L PFAC) attained the best removal efficiency, whereas the corresponding efficiency decreased to 46% with combination D (5% SRR + 7-mg/L PFAC). Comparing the two procedures, the removal of iron was dependent on the sludge recycling ratio under the same coagulant dosage. As for the removal of aluminum, a similar removal efficiency was found in combinations A (raw water + 10-mg/L PFAC) and D (5% SRR + 7-mg/L PFAC), at 46 and 45%, respectively. In terms of the slightly higher removal efficiency in combinations B (3% SRR + 10-mg/L PFAC) and C (3% SRR + 7-mg/L PFAC), the removal percentages were as high as 52%, independent of the dosage of the coagulant. Based on the variation in manganese against coagulation procedures, the order of the removal efficiency changed to C > B > A > D.

It was interesting to find that the optimal removal efficiency of turbidity, DOM, and metal ions simultaneously increased under the same conditions that reused 3% residual sludge and dosed 7-mg/L PFAC. Based on the comprehension consideration of the coagulant costs and the removal performance of metal ions, 3% SRR and 7-mg/L PFAC were selected to conduct the subsequent continuous trials.

3.2. Investigation of the continuous sludge-recycling trials

3.2.1. Investigation on the effluent quality during the continuous trials

To test the stability of the sludge recycling process on the effluent of drinking water quality, the continuous recycling trials were conducted for 150 h

(15 recycling runs) under the optimal conditions discussed above. The removal efficiency of traditional physical parameters, DOM, and inorganic metal ions is displayed in Fig. 5. The turbidity of the effluent slightly fluctuated vs the continuous running times, whereas the apparent color was sustainably steady. The corresponding values of these two monitoring parameters were in the range of 0.4–0.7 NTU and 4.5–8 ACU, respectively. This result was probably due to the increased number of particles that were introduced with the recycling sludge stream, and details will be discussed later [9]. The similar changing trends of DOC and UV_{254} are shown in sub-image (b) in Fig. 5. The removal efficiency of both DOC and UV_{254} increased slightly in the effluent of the filter. This increase may indicate that the DOM could release again after the sludge was recycled in raw water. However, the concentration of these two parameters wavered within ranges of 4.75–4.97 mg/L and 0.063–0.077 cm^{-1}, respectively. In addition, the corresponding DOC values in the filter effluent dramatically decreased after three and six recycling runs but appreciably increased after nine recycling runs. Interestingly, there was a similar changing trend for the values of UV_{254} and DOC in the discharged sludge as shown in Fig. 6. This result suggests that the initial load of soluble organic substances would impact the effluent of the water quality. The relevant analysis between DOC and UV_{254} is shown in the sub-image. The R value was as high as 0.945, meaning that there was an excellent correlation between DOC and UV_{254}; hence, the value of UV_{254} could be replaced by the

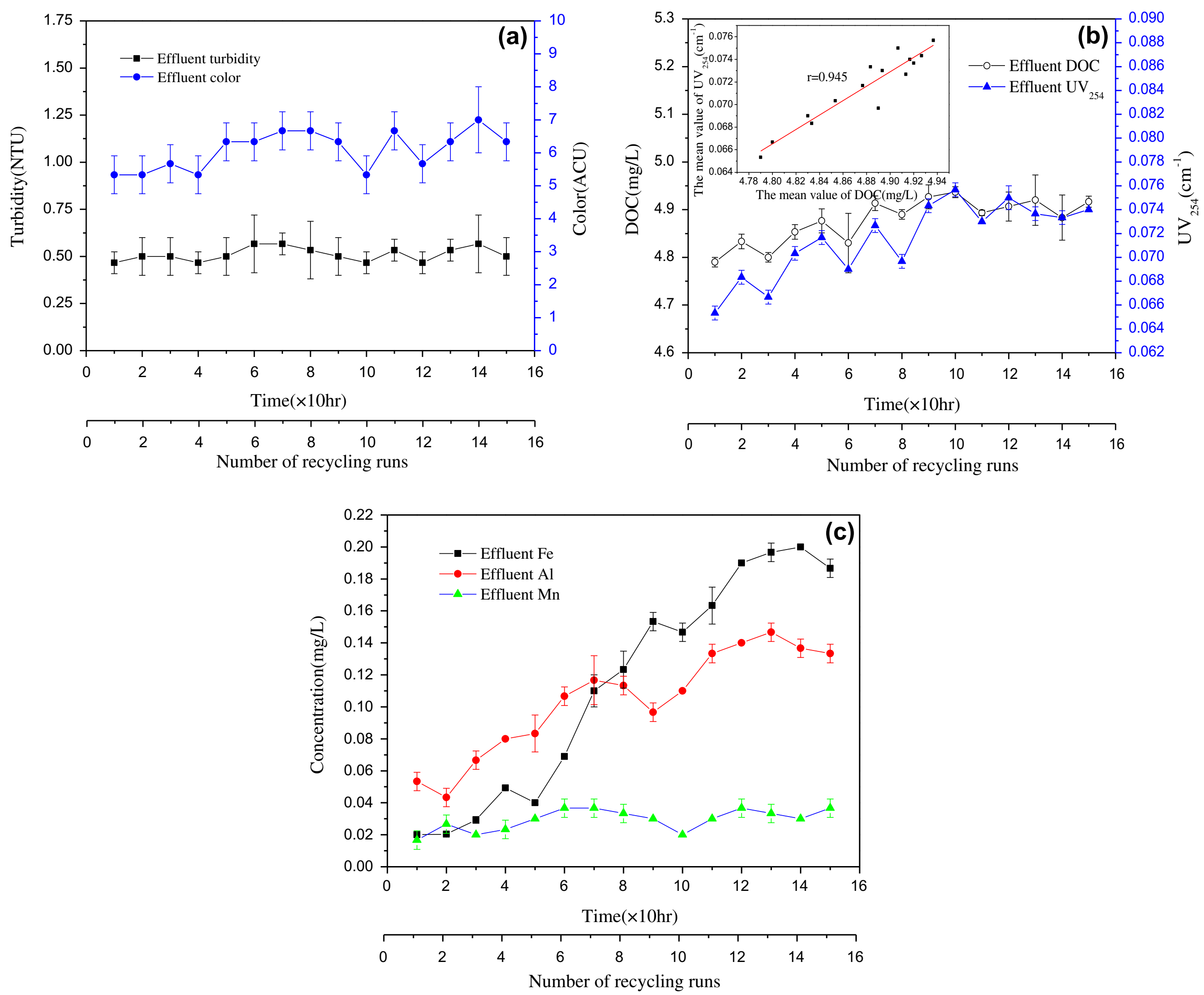

Fig. 5. Coagulation performance of continuous running for 150 h under 3% SRR and 7-mg/L PFAC: (a) turbidity and apparent color removal efficiency, (b) DOC and UV_{254} removal efficiency, and (c) metal ions removal efficiency.

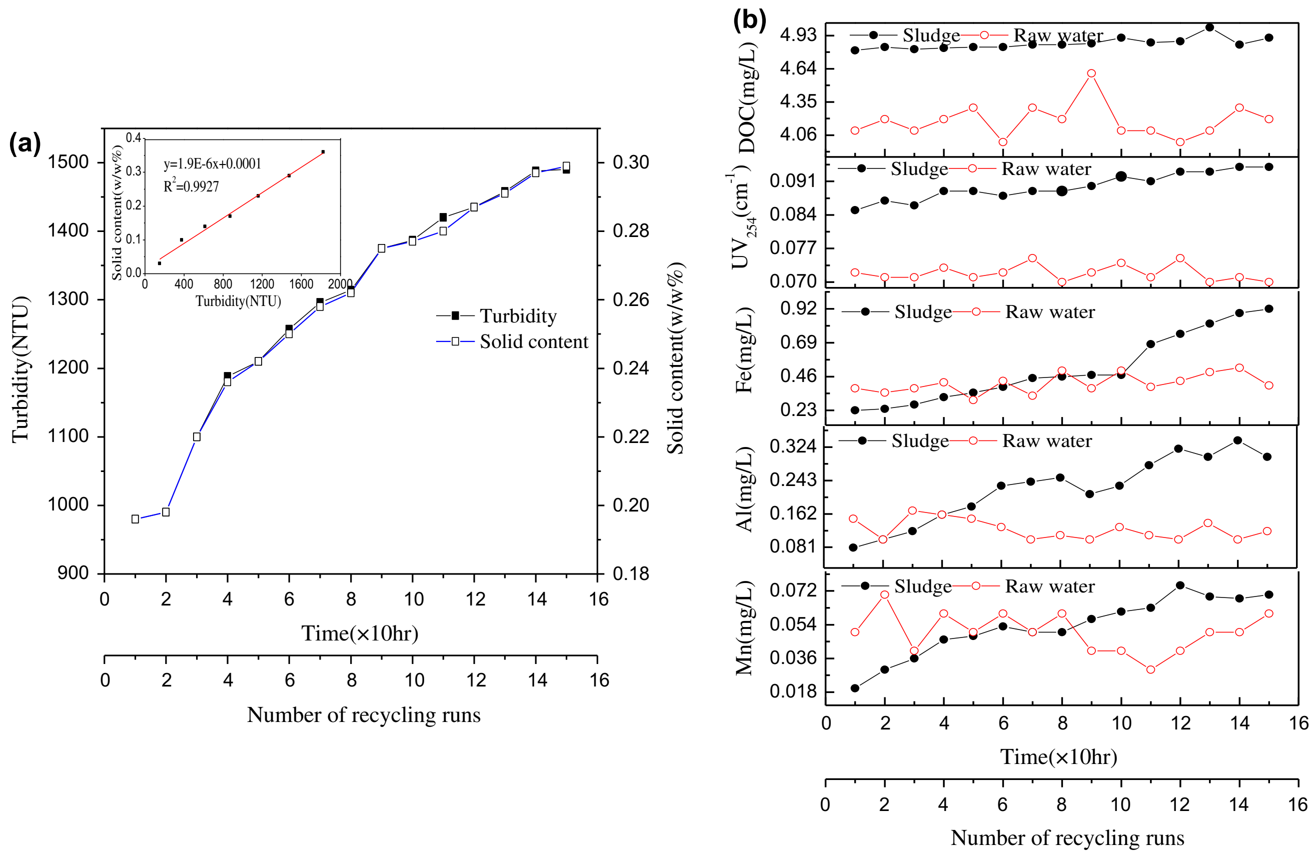

Fig. 6. (a) Effect of recycle process (3% recycling ratio + 7 mg/L) on turbidity and solid content and (b) concentration of DOC, UV_{254}, Fe, Al, and Mn of the discharged sludge in the continuous recycle trial.

DOC value to evaluate the DOM in our research. In terms of the changing trend for Al and Fe, the contents of the two ions in the effluent increased with the running time. Specifically, the concentration of aluminum was as high as 0.142 at the 130th h, and the density of iron peaked at 0.21 mg/L at the 140th h. Compared with the experimental results in Fig. 6, the density of aluminum and iron in the discharged sludge both increased with increase in cycling times. It is likely that the enrichment of the inorganic contaminant in the residual sludge could release again during this cycling process. Additionally, the aluminum content obviously decreased by the ninth d, which is consistent with the results in Fig. 6, in that the density of aluminum decreased within 9 h. The concentration of manganese in the outflow was always steady and independent of the running times. The results of this continuous experiment suggest that instead of increasing coagulant dosages, there should be a higher removal efficiency of traditional physical parameters, organic, and inorganic matters for low turbidity.

3.2.2. *Effect of the recycling process on the material accumulation*

The repeated use of the discharged sludge 15 times in this pilot plant test was performed. Fig. 6(a) demonstrates that there was a relatively preferable correlation between the solid content and water turbidity. Both of these parameters increased with the recycling time; however, the sludge content did not accumulate in large quantities when continuously running for 150 h. Specifically, the turbidity of the sludge increased by 50–100 NTU, whereas that of the solid content increased to only 0.005–0.3% for every recycling time. This result indicates that the low-turbidity testing water contained relatively few suspended particles and produced a small volume of residual sludge. We also inferred that this recycling sludge process was not suitable for high-turbidity water, probably due to the fairly larger sludge emissions.

Using a comparison of all of the concentration of measurements in raw water and sludge, Fig. 6(b) shows that these measurements sustainably increased in the discharged sludge and increased with the recycling time. It is most likely that these pollutant matters continuously accumulated in the reused sludge. However, based on the results in section 3.1, the removal efficiency of all of the measurements was maintained in higher ranges in the effluent during the continuous running process. This result suggests that these pollutant concentrations were not released again in the supernatant.

The enrichment of pollutants in the sludge has demonstrated that the preferable performance of reused sludge could effectively promote the coagulation treatment. The continuous use of sludge in this test has an important meaning for the re-design of the traditional treatment.

4. Discussion and mechanism analysis

4.1. The mechanism analysis of turbidity removal

The results indicate that the physical adsorption and sweep flocculation by hydroxide precipitates play significant roles in enhancing the removal efficiency of organic and inorganic substances. The improvement in removing turbidity was also obvious, resulting from the collision frequencies and efficiencies among particles appreciably increasing [10,11]. Some studies have reported that residual sludge was composed of un-reacting coagulant and relatively large particles [10,12]. In addition, the reused PFAC and fresh preload sludge not only increased the concentration of large particles, but also provided many aggregated cores of flocs in the flocculation process.

Based on the classical Derjaguin–Landau and Verwey–Overbeck theory, the stability of colloids is determined by the relative difference between the attractive potential energy (U_a) and the repulsion potential energy (U_r). The suspended small particles that were close to the new cores could stick to only the border of the aggregated cores because of the repulsion dominant; hence, rather loose and fragile flocs will be produced during this process. In this regard, the enhancement of the turbidity removal efficiency depends on the U_r. An attractive force also existed between the particles. The attractive force between colloids is actually the Van der Waals force, which is a type of a short-range force. However, some studies [13,14] have reported that if a particle size is much larger than others, there is a type of a long-range Van der Waals force to attract the small scattered particles to move toward the large cluster. Although dependence on the Van der Waals attractive force cannot form a stable aggregation, there is no doubt that it will significantly influence the moving direction of the particles [15]. In our study, we postulated that the reused large-size particles such as "attractive cores" will urge the distant particles to move towards the attractive cores. In addition, the guiding role of the attractive force also encouraged particles to easily enter the inner portion of the cores, which will greatly improve the characterization of aggregation.

In addition, the higher concentration of large particles in the recycling sludge made significant contributions to the collision ration, which was also beneficial for the flocculation process, especially for low-turbidity water. In this recycling process, the residual sludge significantly increased the density of the particles. As a result, the collision frequencies and efficiencies between the particles dramatically increased [16]. However, the particle concentration of blended water mixed with the recycling sludge and raw water needed to remain within a suitable range. If particle active collision sites were much more or less than the adhesion sites, it would worsen the quality of the effluent, which may be a reasonable explanation for combinations G and E in Fig. 2.

The reusing sludge also could be considered as a type of rather a low-active coagulant. The recycling process seems to be a second dosage strategy, which could obviously improve the structure of flocs and effluent quality. Yu et al. [15] reported a significant progress on the floc structure after the second little dosage of fresh coagulant. In our research, the removal efficiency was promoted via the recycling method, as seen in Fig. 7.

4.2. The mechanism analysis of DOC and UV_{254} removal

The removal efficiency of DOC and UV_{254} was 43.5 and 21%, both of which were higher than the conventional process. Zhao and Xu et al. [16,17] stated that the removal of organic substance by a hydrolyzed metal coagulant is likely due to the neutralizing effect. The anionic sites on the surface of organic materials could be for binding metal species, such as Al^{3+} and Fe^{3+}, in the PFAC; thus, particles that are formed by these means can be removed during sedimentation or filtration. We speculated that the remaining metal composition that was present in the discharge sludge produced the precipitation of the metal–organic complex, again with the charge neutralization mechanism during the recycling process. In addition, the adsorption of organic substances in amorphous metal hydroxide precipitation played a significant role in the removal of DOC and UV_{254} [18]. Sludge with a porous and large specific surface area can strongly adsorb the soluble organic materials. These results indicate that

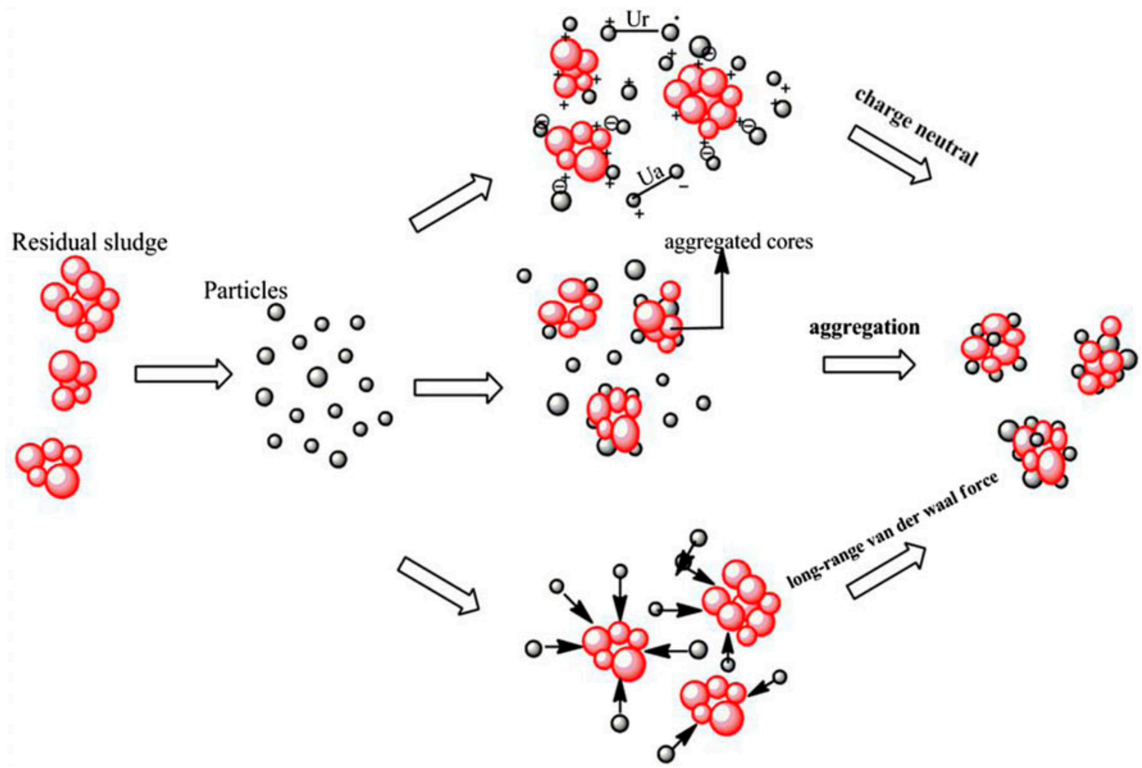

Fig. 7. Diagram of mechanism analysis of turbidity removal.

the reusing sludge was a good adsorbent for organic substances. However, in many practical cases, it is difficult to evaluate the difference between the charge neutralization and absorption mechanisms. There was an interesting finding that the removal rates of these two determined indexes were slightly lower at the 5% SRR (as shown in combination D in Fig. 3) than the optimal combination C. This result indicates that the charge potential of the water may exceed the optimal point (such as the zero zeta point) when too many particles with positive charges exist during the coagulation process. Under these conditions, absorption plays an important role. Due to the relatively worse removal efficiency of the organic matter under a slightly larger value of SRR, there was a hypothesis that the effect of charge neutralization on the removal of organic substances was better than that of the absorption mechanisms. As a result, the greater recycling ratio of sludge could deteriorate the removal efficiency of organic materials.

4.3. The mechanism analysis of Fe, Al, and Mn removal

This result indicates that rather lower volume of reusing sludge was beneficial for removing the aluminum and that the dosage of PFAC has little impact on the treatment effect. Considering the removal efficiency of manganese, there was no evident change in any combination, suggesting that there was almost no influence of the reusing sludge on the removing efficiency of manganese. The experimental results showed that the removal efficiency of inorganic metal materials was significantly enhanced by the improved recycling sludge treatment. Chiang et al. and Gibbons et al. [19,20], using water treatment residuals (WTRs) as a sorbent to absorb multi-heavy metals, suggested that the WTRs were highly amorphous and contained significantly more micropores. In addition to the removal efficiency, cost is also an important factor in feasible production. Therefore, combination C (as shown in Fig. 3) was chosen as the optimal method for the sequential continuous tests.

5. Conclusions

The results of this study clearly indicate that recycling sludge could greatly improve the efficiency of the coagulation mechanism in this pilot-scale test.

(1) The removal efficiency of turbidity and apparent color were obtained by recycling 3% SRR and 7-mg/L PFAC. This recycling is attributed

to a type of a special long-range Van deer Walls force that existed in the larger clusters and scattered particles. This force has a significant guiding effect on the moving direction of particles, urging free particles to move toward the larger clusters and further improving the characterization of flocs. In addition, the collision frequencies between particles improved after recycling the sludge, which was beneficial for the formation of large flocs. We considered the discharge sludge to be a lower active coagulant, as the removal efficiency of turbidity was obviously enhanced after this recycling procedure.

(2) As for the removal of organic materials, the concentration of DOC and UV_{254} was reduced by 43.5 and 21%, respectively, under the 3% SRR and 7 mg/L dosage of PFAC. Both the charge neutralization and absorption simultaneously affected the removal of organic substance after the recycling process. Due to the worse removal efficiency at the slightly higher SRR value, we inferred that the charge neutralization was even more effective than was the absorption for the removal of organic matter.

(3) The removal efficiency of iron, aluminum, and manganese was 56, 50, and 57%, respectively. The reused sludge has an open structure that could intensively absorb metal ions in raw water; thus, the removal efficiency of these parameters in the recycling process was higher than that of the traditional treatment.

(4) Under the optimal operating conditions, this pilot test was continuously run for 150 h to investigate the stability of the recycling process. There was no previous similar report. All of the measurements were still within the reasonable ranges after the recycling process, indicating sufficient stability of the continuous running process and providing a profound and lasting practical meaning.

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