



Coagulation performance and flocs characteristics of variable sludge recycling designs for the synthetic low-turbidity water treatment

Zhiwei Zhou^a, Yanling Yang^{a,*}, Xing Li^a, Wei Gao^b

^aKey Laboratory of Beijing for Water Quality Science and Water Environment Recovery Engineering, Beijing University of Technology, No.100 Xi Da Wang Road, Chao Yang District, Beijing 100124, P.R. China
Email: yangyanling@bjut.edu.cn

^bState Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, P.R. China

Received 31 August 2012; Accepted 7 May 2013

ABSTRACT

Drinking-water-precipitated sludge, characterized as accumulated suspended solids and organic or inorganic components is produced in large quantities during the coagulation process. The proper recycling of drinking water residual sludge is an effective method to enhance low-turbidity water treatment. The current research primarily focused on the settled water quality in terms of bulk parameters, i.e. fine particle distribution, turbidity, COD_{Mn}, total organic carbon, dissolved organic carbon and UV₂₅₄ removal. Residual aluminum under variable sludge recycle designs was also studied. In addition, flocs characteristics was analyzed using optical microscopy and Image J software. The results indicated that recycled sludge combined with activated silica (AS) exhibited excellent potential for turbidity and particulates removal, which might be attributed to jointed effects of sweep floc, adsorption, and bridging. Moreover, powdered activated carbon (PAC) addition was significantly further improving organic matters elimination. Residual total aluminum or dissolved aluminum exceeded the requirement of drinking water treatment standard (0.2 mg/L) in China, indicating that pretreatments or subsequent treatments should be implemented or optimized to avert secondary pollution. Image analysis of regrowth flocs confirmed that recycled sludge combined with AS or/and PAC could be significant for the coagulation treatment of low-turbidity water.

Keywords: Sludge recycling; Coagulation; Low-turbidity; Flocs characteristics

1. Introduction

Waste residual streams, such as filter backwash water, clarifier sludge, or the mixture of both refers to as combined filter backwash water are largely

generated from drinking water treatment plants. Commonly, those residual streams are characterized by elevated concentrations of organics and inorganics, as well as the chemical precipitates derived from inorganic alum- or ferric-based coagulant. Various studies [1–4] have also reported that elevated concentrations of pathogenic microorganism, e.g. *Cryptosporidium* and

*Corresponding author.

Giardia, and enriched poisonous and deleterious substances, as well as other disinfection-resistant pathogens exist in residual streams. Since large amounts of waste residual streams are daily generated all over the world, the proper disposal, regeneration, or recycling of waste residual streams has thus become a significant environmental issue.

Conventional coagulation with recycling residual streams is an alternative way to improve low-turbidity water treatment. It is proposed that increased particle concentration and adsorption sites, as well as changes in coagulation chemistry can give rise to the enhanced removal of particulates and organic matters. Also, this process can accomplish the reuse and reclamation of water resources, and fulfill a low-operational cost due to lower coagulant addition and lower cost for residuals disposal. Currently, there are three dominant issues regarding recycling residual streams in drinking water treatment: (1) safety evaluation with or without pretreatments before recycling [1,4–6]; (2) operating conditions, i.e. recycling ratio, storage or mixing terms [1,7]; (3) mechanism of enhanced removal efficiency for pollutants [8]. Previous investigation [4] demonstrated higher removal efficiencies of *Cryptosporidium* oocyst and other pollutants were achieved when residual streams were recycled in particular terms. However, Cornwell and Lee [9] assumed that the chemical risk of the recycling designs could be reduced only if some effective measures were made.

Generally, charge neutralization and sweep-flocculation are the two main mechanisms for pollutants removal during coagulation. Charge neutralization occurs at quite low coagulant concentrations. Under conditions of rapid and extensive hydroxide precipitation, impurity particles are enmeshed in a growing hydroxide precipitate and are effectively removed from suspension. This process has become known as “sweep flocculation.” Meanwhile, hydroxide precipitates tend to have a rather open structure, so that even a small mass can give a large effective volume concentration and, hence, a high probability of capturing other particles. It is also possible that binding (“bridging”) of particles by precipitated hydroxide may give stronger aggregates. Additionally, the adsorption and bridging of coagulation aids could assist inorganic coagulant to exert charge neutralization or sweep-flocculation.

Activated silica (AS) has been detected to be an efficient coagulant aid and has been noticed to enhance the bridging ability of poly-aluminum chloride (PACl) [10]. However, the recycling sludge combined with AS may be not effectively eliminating low or medium molecule weight and biodegradable

organic substances. Meanwhile, the trapped organic matters may release from the sludge into water during recycling, which ultimately may lead to a compromised performance for organic matter (OM) removal. The removal efficiency of the recycle process can be improved in case the recycled sludge contains powdered activated carbon (PAC), which is an excellent adsorbent commonly used for the removal of organics including OM and synthetic organic compounds [11–13]. Additionally, the retention time of PAC can be prolonged, as that in conventional process is limited to only 10–20 min.

Therefore, the overall purpose of the research was to investigate the impacts of precipitated sludge combined with AS and/or PAC on settled water qualities in terms of bulk parameters, e.g. fine particle distribution, turbidity, COD_{Mn} , total organic carbon (TOC), dissolved organic carbon (DOC), and UV_{254} removal, and the residual aluminum speciation with respect to total alum Al_{T} , dissolved alum Al_{D} and particle alum Al_{P} . Additionally, surface morphology analysis of flocs formed at the end of flocculation was conducted. It is intended to investigate the feasibility of combined effects of AS, PAC, and recycling sludge for low-turbidity water treatment.

2. Materials and methods

2.1. Materials and simulated test water

All reagents used were analytical grade. PAC made of wood was 180 mesh (Actview carbon technology Inc., China); carbon size 98% > 180 mesh. PACl used was reagent grade (with 28% Al_2O_3 content, basicity was 72.3%, GongYi, China). AS was prepared by adding 57.442 g $\text{Na}_2\text{SiO}_3 \cdot 9\text{H}_2\text{O}$ into Milli-Q water and diluted to 0.3 M (calculated as SiO_2). Afterwards, 33.8 mL H_2SO_4 (2M) as activated medicament was added to react 1 h under rapidly mixing, and final pH was adjusted to ~ 2 to yield a 0.15 M AS. The natural colloid stock solution (about 1,000 NTU) prepared according to the reference [14] was used as turbidity-causing matter.

The synthetic raw water was prepared through adding natural colloids and filtered domestic wastewater into local tap water (Harbin, China) at a volumetric ratio of 4: 5: 142, then stabilized at room temperature for 1 day. Main water characteristics of the simulated water were as follows: temperature = 24 ± 1 °C; pH 7.19 ± 0.12 ; turbidity = 17.1 ± 1.14 NTU; COD_{Mn} = 3.61 ± 0.17 mg L⁻¹; TOC = 3.051 ± 0.075 mg L⁻¹; DOC = 2.820 ± 0.112 mg/L; UV_{254} = 0.039 ± 0.001 cm⁻¹; Al_{T} = 0.369 ± 0.044 mg L⁻¹; Al_{D} = 0.236 ± 0.086 mg L⁻¹ and Al_{P} = 0.177 ± 0.063 mg L⁻¹.

2.2. Experimental set-up and sampling

Before on-line recycling trials, jar tests were performed on a programmable jar test apparatus (ZR4-6, Water Industry Technology Development Co. Ltd., China) to determine the optimal PACl and AS dosage. The operation procedures as follows: 1.0 L of raw water was transferred into a 1.4-L square beaker with sampling port 3 cm below water surface; the jar tester was initially started with a rapid mixing at 300 rpm; after 1 min, a certain dose of PACl or/and AS was added, with simultaneously 1 min of rapid mixing at 120 rpm again; the sample was subsequently exposed to a slowing mixing at 50 rpm for 15 min; and then after 20 min of quiescent settling, samples were taken for water quality measurement. The optimal dosage of PACl and AS was 9 and 1.485 mg/L (calculated as SiO₂), respectively. Considering the changes in concentration and characteristics of OM released from sludge, the PAC dosages were investigated at 4, 6, 8 and 10 mg/L in this study. Variable recycle designs were listed in Table 1.

Fig. 1 presents the flow diagram of the experimental system. As indicated, the experimental system included a mixer, 3-stage flocculating reactor, sedimentation unit and sludge recycling system. In detail, the retention time and stirring speed of the mixer was 2 min, 600 rpm accordingly. The 3-stage flocculating reactor of each with retention time was 6,

6, and 3 min, respectively, and the stirring speed was 350, 250, and 150 rpm in sequence. The upstream flow rate of sedimentation unit was 1.0–2.0 mm/s and retention time of 6–8 min.

During the experiments, PACl or/and AS blended with raw water were pumped into the mixer. When necessary, PAC was pumped into 1-stage flocculation by metering pump. In the individual sludge recycle, the optimal sludge recycling ratio by volume was determined to be 8%, from sludge control tank to the beginning of 1-stage flocculation. Before each recycle trial, the sludge in control tank was completely updated. The solid content of recycling sludge in every recycle tests was found to be 0.085–0.10 w/w%.

All recycling trials were carried out in continuous flow, and the settled water samples and flocs samples were intermittently collected twice per day. The location of water samples and flocs samples were labeled in Fig. 1.

2.3. Analytical methods

TOC was analyzed by a TOC Analyzer (Shimadzu, TOC-V_{CPN}, Japan). DOC was monitored by TOC analyzer. UV absorbance at 254 nm (UV₂₅₄) was determined using a spectrometer (UV754, Cany, China). Both DOC and UV₂₅₄ were measured after filtration through 0.45 μm membrane. COD_{Mn} was analyzed by acid potassium permanganate oxidation method. Turbidity was measured by Hach 2100 P Turbidimeter. The total number of particulates and the particle diameter distribution was determined with Versa Count TM particle counter (IBR Inc., USA). The size was selected in the ranges 2.00–28.95 μm. Alum concentration was determined with Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES, PerkinElmer Optima 2000, USA). Al_T and Al_D concentrations were measured without and with sample filtration through 0.45 μm membrane, respectively. Prior to analysis, the samples were digested by nitric acid. Al_P concentration was the difference between the Al_T and Al_D. Each trial was conducted in duplicated. pH was determined by pHS-3C (Shanghai, China) pH meter, which was calibrated daily using pH buffer solution. Solid content was measured using standard methods.

Table 1
Variable sludge recycling processes

	PACl (mg/L)	Sludge recycling ratio (%)	AS (mg/L)	PAC (mg/L)
A	9	0	0	0
B	9	8	0	0
C	9	8	1.485	0
D	9	8	1.485	10
E	9	8	1.485	8
F	9	8	1.485	6
G	9	8	1.485	4

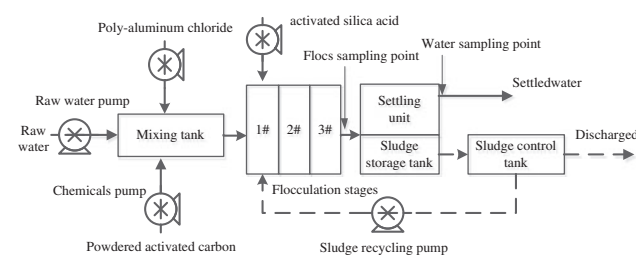


Fig. 1. Diagram of on-line sludge recycling trial.

2.4. Image analysis of the flocs

In this study, samples of flocs formed at the end of flocculation were acquired by a hollow plastic tube. The tube was open at both ends; one end of the tube was inserted gently about 25 mm into the flocculated solution. The flocs in the tube were withdrawn carefully by covering the other end of the tube with a

finger. After transferring the sample to a flat microscope slide, the images of the flocs were captured by an optical microscope (Olympus, BX51TF, Japan) equipped with a video camera (Olympus, C-5,050 200M, Japan). The camera had a sensor matrix consisting of 1,360 (horizontal) \times 1,024 (vertical) pixels. The floc dimensions were obtained by processing the floc image using Image J software to obtain values for the average size, perimeter P and the projected area A . The perimeter-based fractal dimension D_{pf} was obtained from Eq. (1) [15,16].

$$A \propto P^{2/D_{pf}} \quad (1)$$

Thus, the $2/D_{pf}$ may be found as the slope of a plot of $(\lg A)$ vs. $(\lg P)$. Unlike that of a Euclidean object, the value of D_{pf} of a fractal object is not an integer but a decimal, ranging from 1 (a circle) to 2 (a line). 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 $1 \leq D_{pf} \leq 2$. As the object area becomes larger, the perimeter increases more rapidly than for Euclidean objects, indicating that the boundary of the objects is becoming more convoluted and the shape of the objects is more irregular. Generally, a smaller D_{pf} represents a more regular floc structure, meaning a higher flocs density that has a major effect on solid/liquid separation.

3. Results and discussion

3.1. Turbidity and particulate removal

As shown in Fig. 2, a substantial reduction in residual turbidity was initially observed, afterwards, it became steady in all processes at about 180 min. As

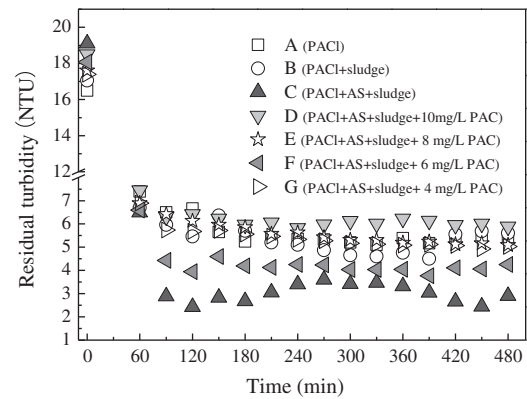


Fig. 2. Comparisons of residual turbidity in the settled water vs. time.

compared to A process, the lowest residual turbidity was 3.22 ± 0.23 NTU in process C, and the residual turbidity varied according to the following order: $D > E > G > B > F > C$. It can be inferred that AS combined with recycling sludge played the most important role in facilitating coagulation.

Former studies [14] have reported that single recycling sludge showed less potential of favoring coagulation, but the increase in seeding and nuclei for flocs was more significant. PAC in recycling sludge and continuous fresh preloaded was significant to increase the number of particles or nuclei in flocculated water; moreover, the PAC adsorption capacity on organic was available to compress the space between the droplets or minimize the exclusion effect of the double layer. However, the results of PAC and AS combined with recycling sludge in D/E/F/G processes were not as expected that residual turbidity were lower.

The higher particles concentration and larger particles diameter contributed to higher probability of collision, which favored the flocculation process, especially for low turbidity water according to Eq. (2) [14].

Table 2
Particle size distribution in settled water

Particle size (μm)	Raw water	A	B	C	D	E	F	G	
Total particles	23,675	8,897	11,762	8,596	9,876	9,767	9,456	9,633	
2	Percentage (%)	40.4	50.2	60.9	28.8	50.4	44	53.8	48.2
2.79		28.4	32.8	4	25.6	30.7	27.6	28.9	33.3
3.90		18.2	13.3	23.4	22.7	13.4	17.7	9.8	15.4
5.45		7.7	1.6	8.1	12.3	3.7	5.5	6.1	2
7.61		2.9	0.9	2.2	5.4	1.1	3	0.6	0.8
10.63		2.3	0.7	0.8	3	0.4	1.1	0.4	0.2
14.84		0.7	0.3	0.3	1.7	0.2	0.6	0.3	0
20.73		0.3	0.1	0.1	0.5	0.1	0.3	0.2	0
28.95		0.1	0	0	0.1	0	0.1	0	0

$$N = 12\pi\beta n^2 d^3 \sqrt{\xi/\mu} \quad (2)$$

where N is frequency of collision ($1/(\text{cm}^3\text{s})$); β is a constant; n is concentration of particles ($\text{counts}/\text{cm}^3$); d is diameter of particles (cm); ξ is dissipation rate of water energy in per time per volume; μ is dynamical viscosity degree of water ($\text{Pa}\cdot\text{s}$). As indicated in Eq. (2), the collision probability between particles was positively relative to “ d^3 ” and “ n^2 .” In recycling process, the “ d ” and “ n ” improved since the addition of recycling flocs particles. As a result, the collision frequencies and efficiencies were promoted. In process C, the adsorption and bridging of AS was benefit to produce larger and denser flocs. The incorporation of PAC into larger flocs may reduce the collision probability, and the turbidity removal efficiency could be somewhat decreased.

Although the lowest residual turbidity was obtained in C process, the particles size distribution of each process was investigated, as summarized in Table 2.

There were about 23,675 particles ($>2\mu\text{m}$)/mL in raw water, and 8,897, 11,762, 8,596, 9,876, 9,767, 9,456, and 9,633 particles ($>2\mu\text{m}$)/mL, respectively, after being treated by A, B, C, D, E, F, and G processes. As observed in Table 2, the particle number in the range $>3.9\mu\text{m}$ were greatly decreased by process A, which could be attributed to the adsorption of these Al(OH)₃ complexes and precipitates [17]. Conversely, the number of particles ranging from $3.9\text{--}5.45\mu\text{m}$ and $<2\mu\text{m}$ increased from 25.9 to 31.5%, 40.4 to 60.9% respectively in process B. In process C, the enhanced removal of turbidity may be attributed to the removal of particles with size $<2\mu\text{m}$. The varied dosages of PAC (D/E/F/G processes) on particle removal showed the similar trends that fine particles with the size $<2\mu\text{m}$ increased while that of $>3.9\mu\text{m}$ decreased. Some investigations [18] have observed that turbidity removal ability could be prohibited in case flocs incorporated PAC into their structure, for reducing the concentration of the PAC particles and the nuclei of flocs.

3.2. Organic carbon removal

As illustrated in Fig. 3(a), all recycling processes could effectively enhance the removal of COD_{Mn}. Moreover, the adsorption of PAC significantly improved the COD_{Mn} removal efficiency. At optimum dosages of 6 mg/L (F process), it was maximally reduce COD_{Mn} from $3.61 \pm 0.17 \text{ mg/L}$ in raw water to $2.53 \pm 0.10 \text{ mg/L}$, which was 17.5% higher than A process ($11.6 \pm 3.55\%$). On the other hand, Fig. 3(b) shows that TOC removal was

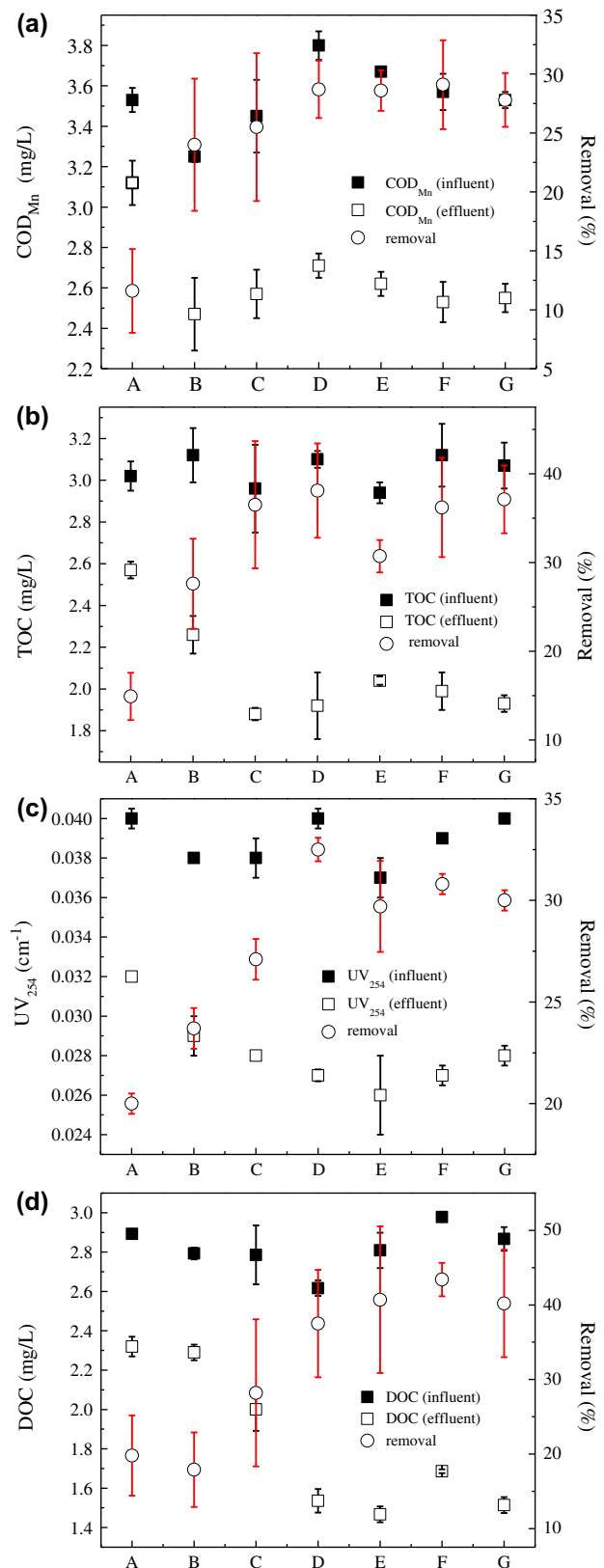


Fig. 3. The organics removal in settled water of different processes (a) COD_{Mn}, (b) TOC, (c) UV₂₅₄ and (d) DOC (Error bars represent one standard deviation, number of samples = 4).

dissimilar with COD_{Mn} removal. Specifically, a higher TOC removal highly resulted from a higher dosage of PAC, processes D could contribute to the equally enhanced removal, which could eliminate TOC from 3.051 ± 0.075 mg/L in raw water to 1.92 ± 0.16 mg/L, with the removal efficiency $38.1 \pm 5.32\%$.

As illustrated in Fig. 3(c), the UV_{254} removal showed the same trend with COD_{Mn} . The raw water had the UV_{254} level of 0.039 ± 0.001 cm^{-1} , it was reduced to 0.027 ± 0.003 cm^{-1} in D process, with the removal efficiency reaching up to $(32.5 \pm 0.58)\%$. As to DOC removal, as compared to 2.820 ± 0.112 mg/L in raw water, $(43.4 \pm 2.24)\%$ of DOC was eliminated in F process with the residual DOC concentration as low as 1.686 ± 0.010 mg/L.

Thus, the recycling sludge combined with AS and PAC exhibited excellent potential for organics removal. The mechanisms to explain the coagulation of OM in this simulated water include charge neutralization, precipitation, bridge aggregation, adsorption and sweep-flocculation. Under different conditions, the different mechanisms or their combination may be dominant [19]. The organics removal in the current research was determined to AS and/or PAC since the zeta potential of suspension solution coagulated by 9 mg/L of PACI was from -0.482 to -0.506 mV and the zeta potential of recycling sludge was from -8.86 to -15.15 mV, which implied that charge neutralization played limited positive role on organic enhancement. In addition, adsorption and bridging of AS could help PACI exert sweep flocculation. Moreover, PAC was one of the most excellent adsorbents for organics removal due to its strong affinity for organic compounds.

3.3. Residual aluminum in settled water

In China, residual alum concentration of finished water is limited to 0.2 mg/L by the regulation of

national drinking water sanitary standard (GB5749–2006). The average value and standard deviation (SD) of residual alum speciation by different processes were summarized in Table 3.

As indicated in Table 3, the Al_T in the raw water was 0.369 ± 0.044 mg/L (alum component in nature colloid and filtered domestic wastewater, as well as in the tap water), the Al_T in the settled water of sludge processes significantly increased and varied according to the following order: $B > D > C > F > G > E$. The increase in residual Al_T was accounted for the enrichment of destabilized alum in recycling flocs, while decreased in C process since the alum enmeshed in flocs was repacked and easily separated. Moreover, the moderate dosage of PAC (process E, 8 mg/L) could further adsorb Al_T and ultimately reduce it to 1.069 ± 0.071 mg/L.

As shown in Table 3, the residual Al_D in all processes was irrelative to the Al_D content in raw water. The Al_D in the raw water was 0.236 ± 0.086 mg/L and that in the effluent kept stable around or marginally above 0.296 mg/L except in the process C (0.08505 ± 0.0068 mg/L).

Residual Al_P was the difference of Al_T and Al_D . Suspended or particulate aluminum could be simply eliminated from the water by coagulation/sedimentation process. Generally, the less quantity of Al_P indicates the lower residual turbidity of treated water. In Table 3, the residual Al_P varied according to the following order: $B > C > D > F > E > G > A$. The introduction of recycling sludge to the destabilized source water subsequently combined with AS and PAC lowered the content of Al_P .

It is worth noting that the residual Al_T or Al_D in the effluent exceeded 0.2 mg/L. This may be ascribed to the higher background in raw water and addition of sludge in recycling process, as well as the alum dissolved out into settled water. Therefore, some pretreatments or subsequent treatments should be further implemented or optimized to avert secondary pollution.

Table 3
Residual aluminum speciation in settled water (Unit: mg/L)

	Influent		Effluent		Influent		Effluent		Influent		Effluent	
	ρAl_T	SD	ρAl_T	SD	ρAl_D	SD	ρAl_D	SD	ρAl_P	SD	ρAl_P	SD
A	0.353	0.010	0.421	0.045	0.286	0.011	0.296	0.031	0.066	0.017	0.125	0.042
B	0.305	0.020	1.679	0.067	0.171	0.0070	0.304	0.0070	0.132	0.13	1.375	0.074
C	0.387	0.088	1.281	0.077	0.226	0.13	0.0851	0.0068	0.162	0.075	1.195	0.070
D	0.377	0.044	1.414	0.011	0.089	0.0049	0.292	0.036	0.027	0.039	1.122	0.025
E	0.357	0.017	1.069	0.071	0.131	0.0064	0.296	0.020	0.226	0.023	0.774	0.091
F	0.403	0.0057	1.145	0.037	0.244	0.018	0.332	0.058	0.159	0.024	0.880	0.094
G	0.408	0.013	1.131	0.011	0.297	0.0076	0.361	0.0097	0.112	0.057	0.770	0.0016

3.4. Characteristics analysis of the flocs

The morphological analysis was applied to study the physical property of the flocs. As we know, the size and number concentration of particles would directly impact the density and sedimentation rate of flocs [20,21]. In addition to particle size, surface morphology of the flocs also affected the behavior of aggregated particles, particularly with regard to collision efficiency and settling velocity. In recent years,

irregular aggregate shapes have been described in terms of two-dimensional fractal dimension [15,16,22]. A higher D_{pf} means a more open or irregular floc structure and the lower effective density. The fractal dimension D_{pf} of derived floc was displayed in Fig. 4.

As indicated, the D_{pf} was slightly decreased to 1.650 in the recycling sludge solely and sharply decreased in the processes that recycling sludge combined with AS or/and PAC as comparison with process A ($D_{pf}=1.732$). The impacts of PAC dosages

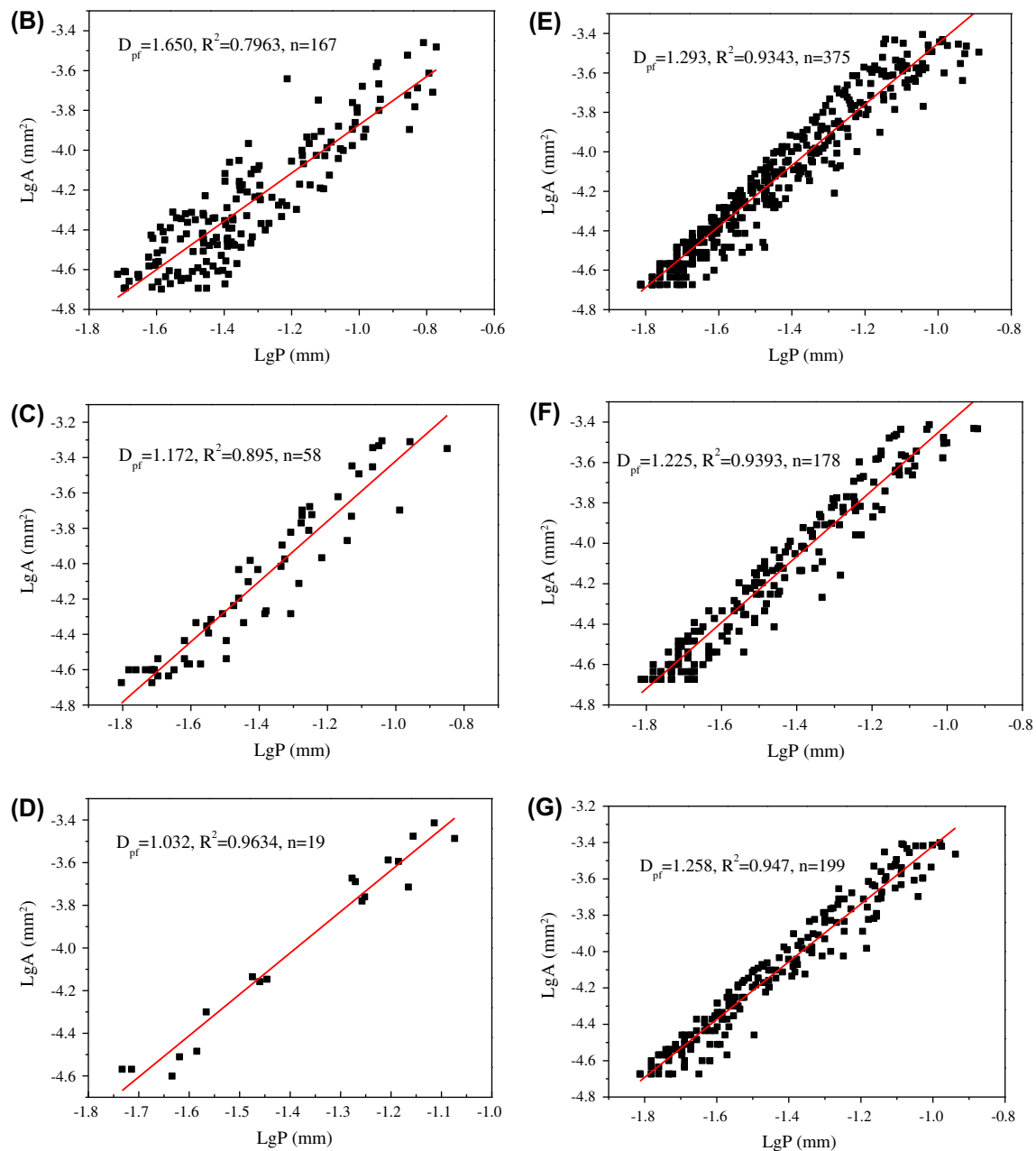


Fig. 4. Fractal dimension of derived flocs (R^2 and n represents correlation coefficient and number of aggregates, respectively).

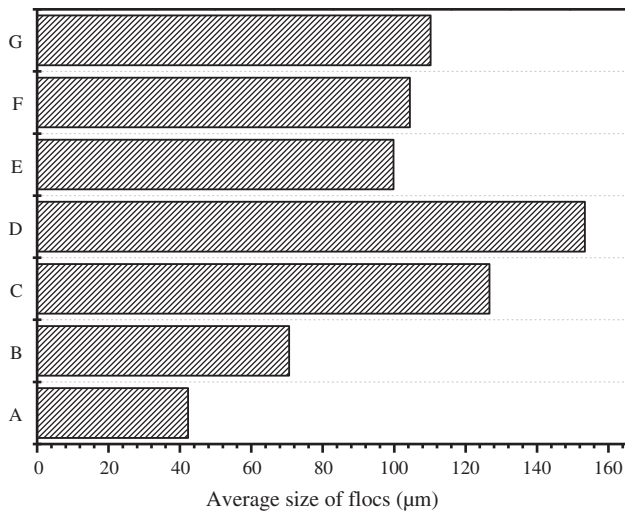


Fig. 5. Average size of the flocs.

on D_{pf} were nearly the same except that in D process, which may be ascribed to the fewer number of aggregates for calculating. Therefore, it can be inferred the flocs structure in process C was more regular than those formed in the processes with PAC. There are two explanations for the relatively irregular floc structures: (1) PAC played an important role in increasing the concentration of the particles and being the condensation nuclei of flocs, with larger flocs incorporating PAC into their structure; (2) the breakage of flocs occurred and the broken flocs may be difficult to recover to their initial size before breakage [20]. The small flocs increased after breakage and recovery, affecting the calculation of D_{pf} according to Eq. (1).

The decrease in D_{pf} was associated with an increase in floc size. As shown in Fig. 5, the maximum average size of the flocs formed in process D was 2.63 times larger than the process A (42.3 μm), re-growing to 153.4 μm. The results suggested that recycling

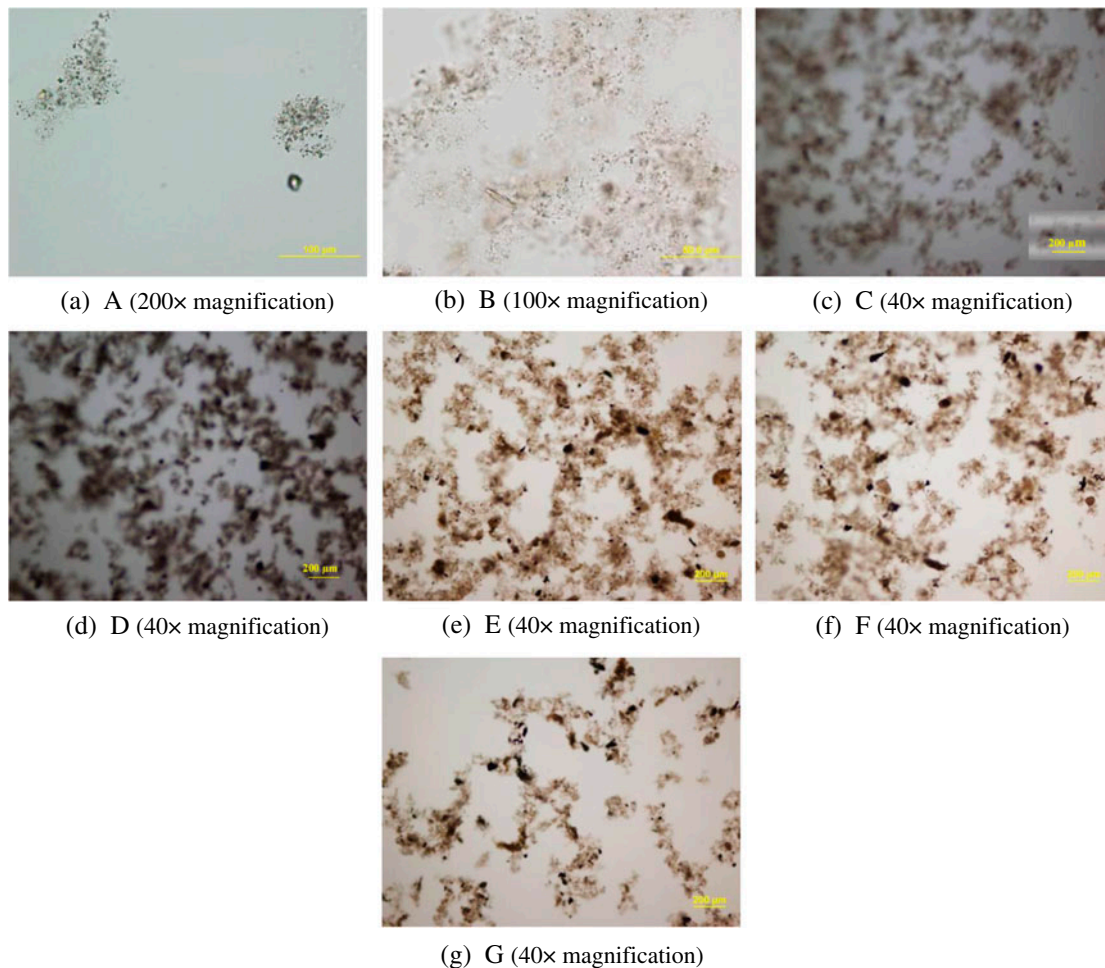


Fig. 6. Microscope images of the flocs.

sludge combined with AS or/and PAC contributed to formation of stronger flocs. Therefore, the average size of flocs in CDEFG processes was much larger than that in process A. Furthermore, PAC in recycling sludge and fresh “preloads” played a negative role on flocs structure, but a limited positive role on flocs formation. This was probably because the new formed hydroxide precipitates were distinct in surface characteristics from the aged precipitates. It is likely that there were more active groups on the fresh precipitate particles, which were capable of binding the aged precipitates together. Thus, the adsorption of newly formed soluble Al species on the surface of the aged flocs gives improved adhesion [23]. Besides, the adsorption and bridging of AS could further bind the new and aged flocs together.

The images of flocs obtained by optical microscopy are shown in Fig. 6. The surface image of flocs in process A was finer and looser, having an adverse effect on subsequent sedimentation unit. The process B in Fig. 6(b) produced more compact and denser flocs, which was benefit to increase the settling rate and also improve the water. As to process C, the flocs was even larger and more readily to settle, and the flocs characteristics were improved due to the adsorption and bridging of AS. As to the PAC effect, as shown in Figs. 6(d-g), the flocs were larger and more compact than those without PAC (A/B/C processes). Regrettably, larger flocs could incorporate PAC into their structure to reduce the efficiency of mixing, and adsorption capacity sites for organics. Consequently, the highest concentration of PAC (10 mg/L) did not necessarily indicate the most excellent contaminates removal, which was consistent with the previous results.

4. Conclusion

The main conclusions of this work were as follows: Recycling sludge combined with AS exhibited excellent potential for turbidity and particulate removal and the number of fine particles ($<2\mu\text{m}$) was greatly decreased. PAC addition was significantly further improving organic matters elimination. The residual Al_T varied according to the following order: $B > D > C > F > G > E > A$. Besides recycling sludge combined with AS exhibited the least concentration of residual Al_D , followed by processes DEABFG (in increasing order). The residual Al_T or Al_D exceeded 0.2 mg/L. Therefore, to limit the content of aluminum under control, some pretreatments or subsequent treatments should be carried out. Applied during the coagulation process, recycling

sludge combined with AS and/or PAC could be significant for the low-turbidity source water treatment.

Acknowledgments

This research is jointly funded by the National Natural Science Foundation (Grant No. 51,278,005) and Beijing Natural Science Foundation (Grant No. 8,132,007) of China, and the 11th post-graduate sci-tech innovation foundation of Beijing University of Technology (Grant No. ykj-2012-8,728). We would like to give our sincere thanks to the peer-reviews for their suggestions and discussions.

References

- [1] A. Gottfried, A.D. Shepard, K. Hardiman, M.E. Walsh, Impact of recycling filter backwash water on organic removal in coagulation–sedimentation processes, *Water Res.* 42 (2008) 4683–4691.
- [2] J.J. Klemeš, P. Stehlik, E. Worrell, Waste treatment to improve recycling and minimise environmental impact, *Resources, Resour. Conserv. Recycl.* 54 (2010) 267–270.
- [3] N.J. McCormick, M. Porter, M.E. Walsh, Disinfection by-products in filter backwash water: Implications to water quality in recycle designs, *Water Res.* 44 (2010) 4581–4589.
- [4] C. Rodriguez, K. Linge, P. Blair, F. Buseti, B. Devine, P. Van Buynder, P. Weinstein, A. Cook, Recycled water: Potential health risks from volatile organic compounds and use of 1,4-dichlorobenzene as treatment performance indicator, *Water Res.* 46 (2012) 93–106.
- [5] C. Chen, X.J. Zhang, L.X. Zhu, J. Liu, W.J. He, H-d Han, Disinfection by-products and their precursors in a water treatment plant in North China: Seasonal changes and fraction analysis, *Sci. Total Environ.* 397 (2008) 140–147.
- [6] J.C. Bourgeois, M.E. Walsh, G.A. Gagnon, Treatment of drinking water residuals: comparing sedimentation and dissolved air flotation performance with optimal cation ratios, *Water Res.* 38 (2004) 1173–1182.
- [7] W. Chu, Dye removal from textile dye wastewater using recycled alum sludge, *Water Res.* 35 (2001) 3147–3152.
- [8] X.H. Guan, G.H. Chen, C. Shang, Re-use of water treatment works sludge to enhance particulate pollutant removal from sewage, *Water Res.* 39 (2005) 3433–3440.
- [9] D.A. Cornwell, R.G. Lee, Waste stream recycling: Its effects on water quality, *J. AWWA*, 86 (1994) 8650–8663.
- [10] M. Yan, D. Wang, J. Qu, J. Ni, C.W.K. Chow, Enhanced coagulation for high alkalinity and micro-polluted water: The third way through coagulant optimization, *Water Res.* 42 (2008) 2278–2286.
- [11] M. Campinas, M. João, Removal of microcystins by PAC/UF, *Sep. Purif. Technol.* 71 (2010) 114–120.
- [12] T. Lebeau, C. Lelivre, H. Buisson, D. Cret, L.W.V.D. Venter, P. Ct, Immersed membrane filtration for the production of drinking water: combination with PAC for NOM and SOCs removal, *Desalination* 9164 (1998) 219–231.
- [13] Y. Zhang, J. Tian, J. Nan, S. Gao, H. Liang, M.L. Wang, G.B. Li, Effect of PAC addition on immersed ultrafiltration for the treatment of algal-rich water, *J. Hazard. Mater.* 186 (2011) 1415–1424.
- [14] L.H. Sun, M. Lv, Y.L. Yang, L.L. Zhou, G.B. Li, Enhanced treatment of water with low turbidity: Combined effects of permanganate, PAM and recycled sludge, *J. Harb. Inst. Technol.* 6 (2009) 863–868.
- [15] F. Xiao, Ju-Chang H. Huang, B.J. Zhang, C.W. Cui, Effects of low temperature on coagulation kinetics and floc surface morphology using alum, *Desalination* 237 (2009) 201–213.

- [16] F. Xiao, J. Ma, P. Yi, Ju-Chang H. Huang, Effects of low temperature on coagulation of kaolinite suspensions, *Water Res.* 42 (2008) 2983–2992.
- [17] L. Qi, H. Liang, Y. Wang, G.B. Li, Integration of immersed membrane ultrafiltration with the reuse of PAC and alum sludge (RPAS) process for drinking water treatment, *Desalination* 249 (2009) 440–444.
- [18] Z.W. Zhou, Y.L. Yang, X. Li, W. Gao, H. Liang, G.B. Li, Coagulation efficiency and flocs characteristics of recycling sludge during the treatment of low temperature and micro-polluted water, *J. Environ. Sci.* 24 (2012) 1014–1020.
- [19] J. Duan, J. Gregory, Coagulation by hydrolysing metal salts, *Adv. Colloid Interface Sci.* 102 (2003) 475–502.
- [20] W. Yu, J. Gregory, L.C. Campos, Breakage and re-growth of flocs formed by charge neutralization using alum and poly-DADMAC, *Water Res.* 44 (2010) 3959–3965.
- [21] A. Vahedi, B. Gorczyca, Application of fractal dimensions to study the structure of flocs formed in lime softening process, *Water Res.* 45 (2010) 545–556.
- [22] J. Nan, W.P. He, Z.J. Zhang, Research on fractal characteristics of particles size distribution in dynamic flocculation process, *Wuhan Univ. J. Natural Sci.* 14(6) (2009) 511–517.
- [23] T. Liu, Z.L. Chen, W.Z. Yu, J.M. Shen, J. Gregory, Effect of two-stage coagulant addition on coagulation-ultrafiltration process for treatment of humic-rich water, *Water Res.* 45 (2011) 4260–4268.