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Return chemical sludge employed in enhancement of phosphate removal from wastewater

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ABSTRACT

In this study, chemical sludge was recycled by using a coagulation process to improve phosphate removal from wastewater. A pilot experiment was conducted to investigate the appropriate ratio of recycled sludge, the appropriate operating conditions, and the mechanism involved in the enhancement of phosphate removal by return chemical sludge. The results showed that, with sludge recirculation, the process of coagulation for phosphate removal was enhanced by up to 12% compared with no sludge recirculation. Those effects were primarily attributed to the removal of relatively fine particles with a size of 100–310 μm. The sludge settleability was also improved by the addition of chemical sludge, which was mainly attributed to the increase in floc size. The appropriate ratio of recycled sludge was determined to be around 15–20%. Mixing and the aging time of recycled sludge had a significant effect on phosphate removal. Based on these findings, sweep flocculation and/or physical adsorption are expected to play key roles in enhancement of phosphate removal from wastewater.

Keywords: Phosphate removal; Return chemical sludge; Wastewater; Coagulation

1. Introduction

As known, phosphate is an essential nutrient for plants and microorganisms, but if its excess is released to a receiving water body, it could cause eutrophication and many natural water bodies could be affected [1]. Therefore, excess phosphate in domestic and industrial discharges should be controlled using wastewater treatment technology. Generally, several physical, chemical, and biological methods are used in the removal of phosphate from wastewater [2–7]. Physical methods have proved to be either too expensive or inefficient, with regard to phosphate elimination from wastewater [8,9]. The biological method is cheaper but its removal efficiency is low. The chemical composition and temperature of wastewater also strongly affected the phosphate removal efficiency of the process [10,11]. Chemical removal techniques are the most effective and well-established methods for phosphate removal, especially the chemical coagulation method. The removal of phosphate by

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the aid of coagulation is a widely used process. However, the use of fresh coagulant may not be so admirable because of the chemical costs and the treatment and disposal of a large amount of chemical sludge [12–14]. Therefore, it is necessary to find other cost-effective treatment methods and materials.

Chemical sludge is an inevitable byproduct from the wastewater treatment when a metal salt is added as a coagulant. Huge amounts of chemical sludge are produced from water and wastewater treatment plants, annually in China during recent years, and they must be disposed in an environmentally sound manner. However, most chemical sludge is ultimately disposed in a landfill and the disposal method results in the loss of valuable asset and at the same time depletes the capacity of the landfill. So the proper disposal, regeneration, or reuse of recycled chemical sludge have become an increasing environmental and economical concern. It is found that the chemical sludge contains approximately 39% Al by weight after coagulation/flocculation. Some techniques have been developed to regenerate alum from the sludge, such as acidification, membrane separation, and alkaline treatment [15], but these processes have proved to be too expensive. Therefore, it is necessary to find a cheaper alternative to solve the significant environmental issue caused by chemical sludge.

Under these circumstances, the idea of using chemical sludge generated from the coagulation process itself may provide a good solution to solve the above problems. Some researchers investigated the effect of adding alum sludge in primary wastewater treatment on the performance of activated sludge process, anaerobic digestion in wastewater treatment plants and they verified the feasibility of this approach [16–20]. They reported that no adverse effects on overall wastewater treatment plant performance were identified [17]. It was also reported that there was an apparent reduction of scum and sludge bulking problems in the secondary clarifier and that alum sludge addition could increase the gas production in sludge digester and affect the capacity of the centrifuges for sludge dewatering [17,18]. Montgomery and Qasim et al. proposed a cost-effective method to transfer alum treatment works for reuse [21,22]. They suggested controlling the discharge of alum sludge into a sewer system, provided that the sewer system is monitored appropriately during the transfer of the sludge. In order to maximize the benefit gained from the alum sludge reuse, some researches also investigated the appropriate dosage of the alum sludge, appropriate operating conditions, and the possible mechanism responsible for the alum sludge effect. Chu reported that with the use of recycled alum sludge in the coagulation process, lead removal rates increased from 79 to 96–98% with 100–180 mg/L alum sludge, while the required fresh alum dosage was reduced from 175 to 50–12.5 mg/L only [15]. Liu and Liang pointed out that using sludge recirculation, the process of coagulation for the color removal results in 35% enhancement in comparison with no sludge recirculation [23]. Guan et al. reported that the SS and COD removal efficiencies could be improved by the addition of the alum sludge [16]. They postulated that the sweep flocculation and/or the physical adsorption were expected to play key roles in the enhancement of the particulate. Previous works done by some investigators also have showed that the chemical sludge has a considerable adsorption capacity and it can be utilized as a cheaper adsorbent for phosphate removal from wastewaters [24–26].

There are papers exploring the use of return sludge for the enhancement of lead metal, color, SS, and COD removal, but there was no paper exploring of the use of return sludge for the enhancement of phosphate removal in the coagulation process. If the precipitated chemical sludge is recycled to the flocculation tank, the chemical sludge can be utilized as a coagulant for phosphate removal in the chemical coagulation treatment because the chemical sludge contains a large portion of insoluble aluminum hydroxides. Reusing the chemical sludge may reduce the dosage of fresh coagulant and the time required for floc formation; it may also improve the phosphate removal efficiency of the chemical coagulation treatment. Meanwhile, it can ease the burden of water treatment works relating to sludge treatment and disposal. On the whole, the use of return sludge of chemical coagulation may reduce the high-operational costs incurring from the use of coagulants, and in the treatment and disposal of a large amount of chemical sludge.

Therefore, the recycled sludge was employed for enhancement of phosphate removal through sludge recirculation, in this paper. Specifically, this study examined the feasibility of using return sludge to increase the phosphate removal efficiency in the coagulation process. The appropriate ratio of chemical sludge was then determined and the operating conditions (e.g. the mixing and the aging time of the recycled sludge) were optimized. Finally, possible mechanisms involved in the enhancement of phosphate removal by chemical sludge were investigated.

2. Materials and methods

2.1. Characterization of the feed water, sludge and coagulant

Synthetic feed water prepared using anhydrous KH_2PO_4 was employed in this study. The concentration of phosphate in the feed water was $2.0\,mg\,P\,L^{-1}$, which was considered to be representative of the phosphate concentration of effluent from secondary treatment plants. The sludge was obtained from the coagulation process itself. Unless otherwise specified, the sludge obtained on the day 0 was used. The pH, zeta potential, and phosphate concentration were measured to characterize the synthetic wastewater, while the pH, zeta potential, solid content, and metals content of the sludge samples were measured to characterize the chemical sludge. A novel composite inorganic coagulant, prepared in the laboratory, was selected to investigate the possibility of sludge use. The new coagulant was prepared by mixing red mud and a hydrochloric pickle liquor of bauxite. The characteristics of the new coagulant, which primarily consists of Al salts are shown in Table 1. More information regarding the new coagulant can be found in Zhao et al. [3].

2.2. Coagulation experiments

Coagulation experiments were conducted using pilot-scale tests. Fig. 1 shows a diagram of the system. The bottom part of the column is filled with ganister sand. Wastewater and coagulant are mixed in the bottom part of the column, while coagulation and separation of the flocs from the treated effluent occurs in the same column, as the mixture flows up through it. The coagulation reactor is then backwashed with up-flow water, at appropriate times based on the pressure at the inlet. Finally, the backwash wastewater is drawn out of the system.

2.3. Sampling and analytical methods

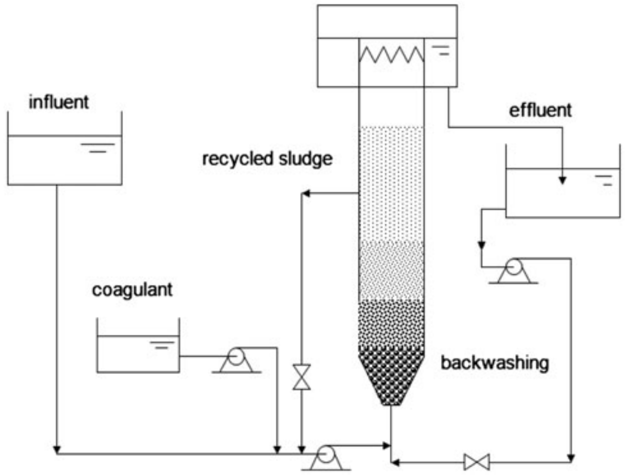

Fig. 1. Diagram of up-flow flocculent reactor that employs recycled sludge.

The phosphate levels were measured using the ascorbic acid method with a Hach DR/4000U spectrophotometer according to the APHA standard methods [27]. A pH meter (Orion) was used to measure the pH of the solutions. A Hach turbidimeter (Model 2100N) was used to measure the turbidity and the results are reported in nephelometric turbidity units. The zeta potentials of the sludge and the feed water were measured with a Zetasizer 2000 (Malvern Instruments Ltd., England). The Al, Ca, Fe, and Mg contents in the sludge were analyzed using an inductively coupled plasma mass spectrometer (Plasma Quad 3). The solid content of the sludge was examined according to the APHA Standard Methods except that a duration of 24 h was employed [28,29]. A Zetasizer 2000 laser scattering particle analyzer (Malvern Instruments Ltd., England) was employed to measure the changes in floc size during the coagulation/flocculation period.

A Hitachi S-3000N scanning electron microscope (SEM) was used to examine the surface and morphology of the sludge before and after adsorption. Specimens were prepared by placing a small amount of sludge on an aluminum holder using double-sided sticky tape, after which they were coated with a gold layer by vacuum evaporation because the red mud was not conductive. Air drying silver was applied to the edges of the gold layer to ground the system. The functional group of the sludge was examined by Fourier transform infrared analysis (EQUINOX-55, Bruker, Ettingen, Germany).

Table 1
Characteristics of the new coagulant

pH (1%sln)	Density (g/cm^3)	Solid content (%)	Basicity (%)	Al_2O_3 (%)	SiO_2 (%)	CaO (%)	Fe_2O_3 (%)
2.1	1.24	41.0	32	12.44	2.95	6.31	3.51

All treatments were conducted in duplicate and here we report the average values. The standard deviation is lower than 5%.

3. Results and discussion

3.1. Characterization of the chemical sludge and feed water

The characteristics of the feed water and the chemical sludge used in this study are listed in Table 2. As shown in Table 2, the chemical sludge had a low solid content indicating that it had not gone through any dewatering processes. Both the feed water and the chemical sludge samples showed a negative zeta potential indicating that charge neutralization will not enhance the phosphate removal [3]. The components in the sludge samples were Al, Ca, Fe, and Mg, which were present at concentrations of approximately 77.64, 29.75, 28.31, and 1.14 mg/g, respectively.

3.2. Effect of ratio of recycled sludge on phosphate removal

For the removal of phosphate by coagulation with sludge reuse, ratio of recycled sludge is considered to be important. In this experiment, ratio of recycled sludge was varied from 0 to 40%, while the coagulant dose was maintained at 20 and 35 mg/L, respectively. The effect of ratio of recycled sludge on the removal of phosphate is in Fig. 2. As seen from Fig. 2, the phosphate removal efficiency first increased as the ratio of recycled sludge increased (≦20%). When 35 mg/L of fresh coagulant was used, the phosphate removal efficiency increased from 65 to 72–79% when 10–20% of the sludge was recycled. Similarly, as the fresh coagulant was further reduced to 20 mg/L, the phosphate removal efficiency increased from 27 to 72–79% when 10%–20% of the sludge was recycled.

Table 2
Characteristics of the wastewater and sludge

Chemical sludge		Synthetic wastewater	
Zeta potential (mV)	−0.24 to −1.42	Zeta potential (mV)	−1.3
pH	6.9	pH	7.3
Solid content (w/w%)	0.82	PO_4^{3-} (mg/L)	2.0
Al (mg/g)	77.64		
Ca (mg/g)	29.75		
Fe (mg/g)	28.31		
Mg (mg/g)	1.14		

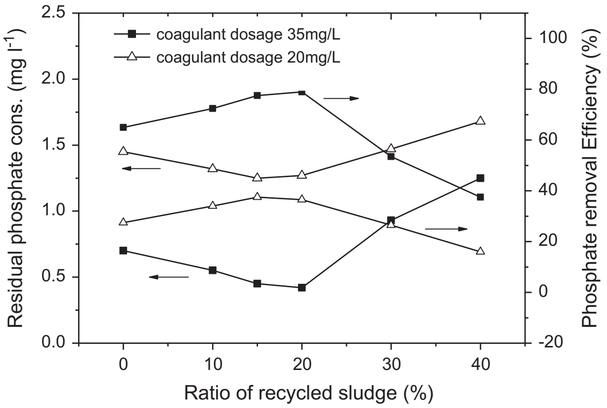

Fig. 2. Effect of ratio of recycled sludge on phosphate removal.

In order to explain the possible mechanisms involved in the enhancement of phosphate removal by chemical sludge, the particle-size distribution in the supernatant of the experiments conducted with or without the addition of recycled sludge was studied. As seen from Fig. 3, the removal of fine particles with sizes ranging from 100 to 310 μm primarily contributed to the increase in phosphate removal efficiency. When no chemical sludge was recycled, particles larger than 310 μm were removed, but when the chemical sludge was recycled, particles larger than 100 μm were removed and smaller particles (100–310 μm) were no longer present in the supernatant. Since the only difference between the two experiments was the addition of the recycled sludge, it is believed that the addition of the recycled sludge resulted in the removal of all particles with a size between 100 and 310 μm.

However, there was a decrease in phosphate removal rate when the ratio of recycled sludge was greater than 20%. To determine the reason, the removal of phosphate using recycled chemical sludge without the addition of a fresh coagulant was investigated. As shown in Fig. 4, the phosphate removal efficiency was −13%, if 15% recycled sludge was used without the addition of fresh coagulant. Since the chemical sludge itself is the only source that may cause water contamination, it is believed that some of the phosphate previously trapped in the recycled sludge will be diffused back into the aqueous phase and cause deterioration of the water quality [15,16,30]. The increase in the phosphate removal efficiency caused by chemical sludge depends on the phosphate removed from the water phase as well as on that released from the chemical sludge. When the return sludge ratio was less than 20%, the phosphate removed by the chemical sludge greatly exceeded the

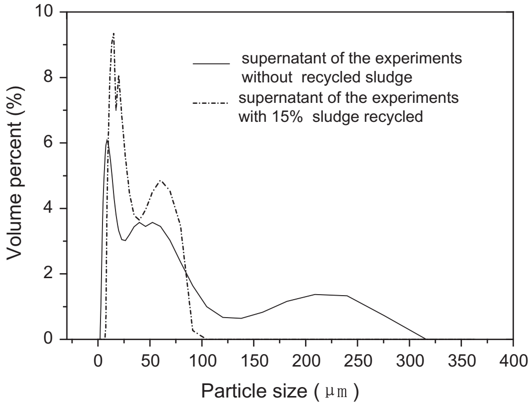

Fig. 3. Particle size distribution in the supernatant of the experiments with or without the addition of recycled sludge.

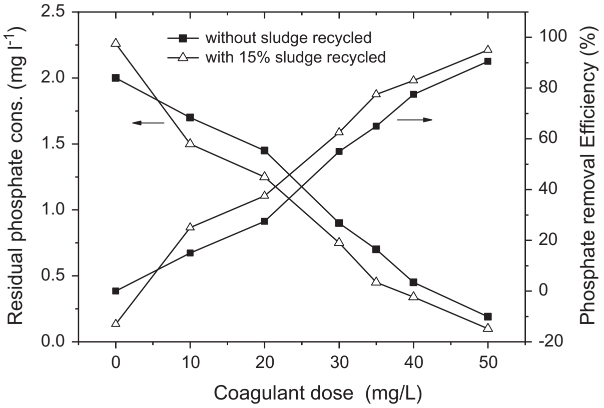

Fig. 4. Effect of coagulant dose on phosphate removal.

phosphate that was released, resulting in an obvious increase in phosphate removal efficiency. Conversely, when the return sludge ratio was more than 20%, the phosphate released by the chemical sludge greatly exceeded the phosphate that was removed, resulting in a marked reduction in phosphate removal efficiency. As the ratio of recycled sludge increased from 15 to 20%, the increase of the phosphate removal efficiency became smaller. 15% was considered as the optimum ratio of recycled sludge for further investigation.

3.3. Effect of coagulant dose on phosphate removal

Based on the above findings, the use of various coagulant doses for removal of phosphate from wastewater was studied with or without sludge recycling (Fig. 4). When the coagulant dosage was 40 mg/L and no sludge was reused, the residual phosphate concentration was reduced to 0.44 mg/L. Although this level meets the discharge standard (GB18918-2002) for phosphate (⩽0.5 mg/L), use of such a high dose in the treatment process may not be appropriate because of the high chemical and operational costs. To minimize the chemical/operation costs and optimize the phosphate removal efficiency, return sludge was used to increase the phosphate removal efficiency. Various fresh coagulant doses were added to the coagulation system while the ratio of recycled sludge was kept at 15%. As shown in Fig. 4, the use of mixtures of fresh coagulant and sludge had some advantages over the use of fresh coagulant alone. Specifically, when the residue phosphate concentration was kept lower than 0.5 mg/L, the amount of fresh coagulant was reduced from 40 mg/L (without using sludge) to 35 mg/L (with the use of recycled sludge). In addition, the phosphate removal efficiency increased from 65.0 to 77.5% when recycled sludge was used in conjunction with fresh coagulant at 35 mg/L. These results showed that the use of sludge generated from the coagulation process itself is a good method of removing phosphate from wastewater while simultaneously reducing the fresh coagulant dosage.

To further investigate the effects of the addition of recycled sludge on coagulation, the turbidity in the supernatant produced in experiments with or without the addition of recycled sludge was investigated. As shown in Fig. 5, the turbidity decreased as the settling time increased in both experiments; however, the turbidity in the supernatant of the experiments where the recycled sludge was added was higher than that of the supernatant generated in experiments without recycled sludge. It was because the addition of recycled sludge increased the quantities of flocs and these flocs did not settle at the beginning of the experiment. Additionally, the turbidity in the supernatant of the experiments in which recycled sludge was added,

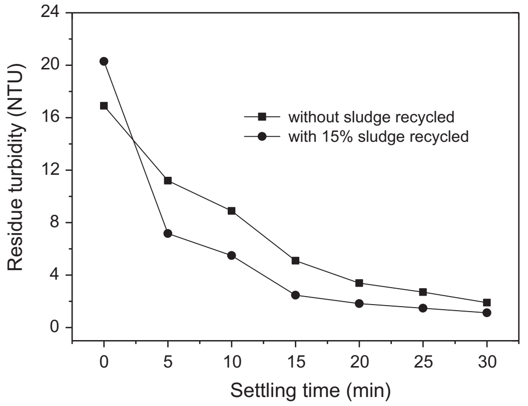

Fig. 5. Residue turbidity in the supernatant of the experiments with and without the addition of recycled sludge.

became lower than that of the supernatant from experiments in which no recycled sludge was added as time increased. This likely occurred because the use of previously settled sludge facilitated floc formation and increased the mass of the settling flocs. The resulting flocs were then heavier than those formed without sludge recycling.

The sizes of flocs in samples collected from systems with and without the recycled sludge are shown in Fig. 6. The addition of recycled sludge increased the floc size in the coagulation experiment. Stokes' Law (Eq. (1)) gives the terminal settling rate of a spherical particle at a low Reynolds number:

$$u_t = g d_{\mathrm{p}}^2 (\rho_{\mathrm{p}} - \rho)/(18\mu) \tag{1}$$

where u_{t} = the terminal settling velocity; g = acceleration of gravity; d_{p} = particle diameter; ρ_{p} = particle density; ρ = density of the surrounding fluid; μ = fluid viscosity.

Stokes' Law shows that at constant particle density, the settling velocity is proportional to the square of the particle diameter. By recycling the sludge, newly formed flocs collided with and adhered to previously formed flocs in the recycled sludge, which resulted in the floc particle size increasing and a subsequent increase in the particle's terminal settling rate.

As previously mentioned, the feed water and the chemical sludge samples showed a negative zeta potential, indicating that charge neutralization will not contribute to the enhanced phosphate removal [16,30]. To explain the improvement of phosphate removal from wastewater through the recycling of chemical sludge, the initial and final pH of wastewater treated with only recycled chemical sludge were measured. The results showed that the pH of the effluent changed little indicating that Al^{3+} ions in the sludge had hydrolyzed completely and ultimately formed metal hydroxides before reuse. This further demonstrated that charge neutralization did not contribute to the enhanced phosphate removal. Taken together, these results indicate that sweep flocculation and/or physical adsorption are responsible for the increased phosphate removal from wastewater.

3.4. Effect of aging on phosphate removal by recycled sludge

When considering the use of recycled sludge for phosphate removal, it is important to investigate the aging effect on phosphate removal. In this study, sludge was analyzed at the day 0 and at three months to investigate the effect of aging on phosphate removal by recycled sludge. The coagulant dose and the ratio of the recycled sludge were kept at 35 mg/L and 15%, respectively. Additionally, coagulation without recycled sludge for phosphate removal was also studied to conduct a comparative analysis. The effects of aging on phosphate removal by recycled sludge are shown in Fig. 7. As shown in Fig. 7, the phosphate removal efficiency increased by ∼10% after applying the sludge obtained on the day 0. Conversely, the phosphate removal efficiency decreased significantly after the sludge collected at three months was applied. As discussed in Section 3.2, the increase or decrease in the phosphate removal efficiency caused by chemical sludge depends on the phosphate removed from the water phase as well as that released from the chemical sludge. The results of the present study revealed that aged sludge cannot collide and adhere to newly formed flocs; therefore, the settling rate was low. This low settling rate combined with the shearing

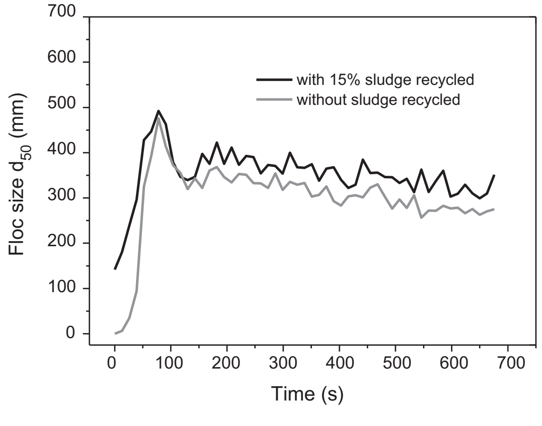

Fig. 6. Floc size in the experiments with and without the addition of fresh sludge.

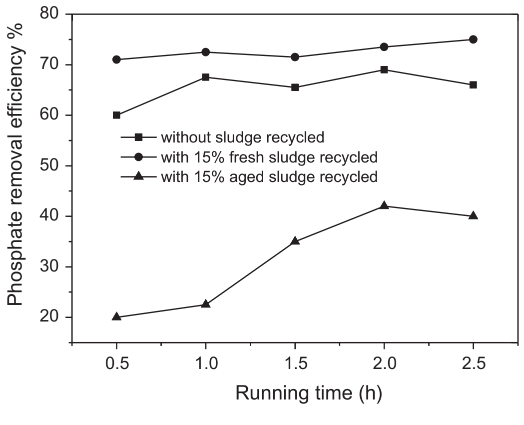

Fig. 7. Effect of aging time of recycled sludge on phosphate removal.

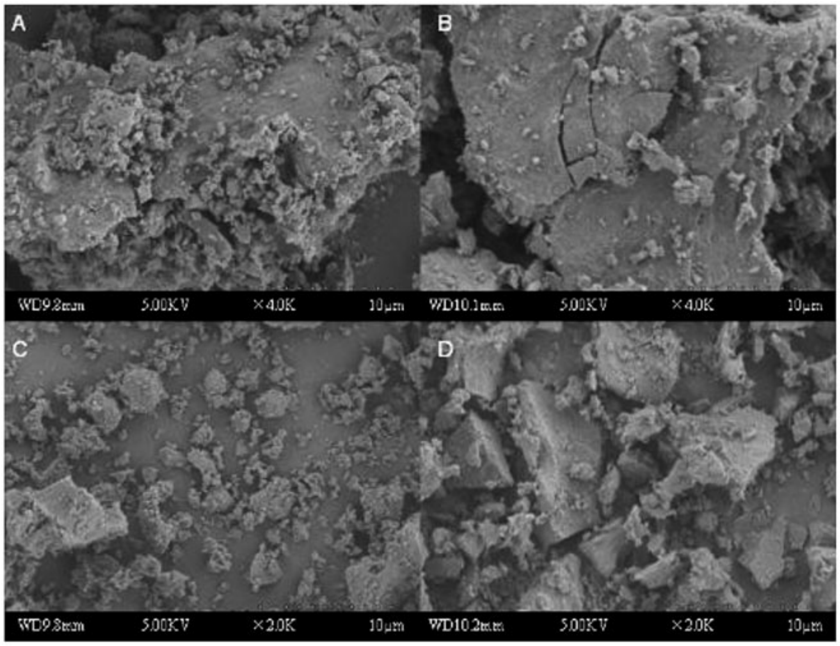

Fig. 8. SEM micrographs of fresh sludge and aged sludge (A and C fresh sludge, B and D aged sludge).

force of the up-flow of the wastewater caused the sludge to break up. As a result, aged sludge that has been broken up is lost via the outlet flow, decreasing the quality of the effluent. The surface and morphology of the fresh and aged sludge were examined by SEM (Fig. 8). The SEM images of the fresh and aged sludge clearly show that their surfaces differed. For the fresh sludge, a porous and amorphous structure was observed, while the surface of the aged sludge was much smoother, indicating that its surface area was lower than that of the fresh sludge. Since most of the surface area is located in the interior of the chemical sludge particle, it has a direct impact on phosphate removal. Accordingly, the decreased surface area of the aged sludge may also explain the decreased phosphate removal efficiency. In a study of P-adsorption on alum hydrolysis products, phosphate removal efficiency was found to decrease as the aging time of the sludge increased [24]. Specifically, a transformation of amorphous aluminum hydroxide to the crystalline form was accompanied by a decrease in surface area over the aging time. Georgantas and Grigoropoulou provided this evidence on XRD monitoring of aluminium hydroxide during aging. It has been demonstrated that the characteristic diffraction band of gibbsite appeared during aging and this diffraction band becomes sharper with time [31].

3.5. Effect of mixing recycled sludge on phosphate removal

To further minimize the chemical/operational cost and optimize the phosphate removal, the recycled sludge was mixed rapidly before it was used to assist with phosphate removal. It was believed that reusing the rapidly mixed sludge could increase the chances of collision between the chemical sludge and the phosphate in the wastewater and reduce the demand for fresh coagulant. Therefore, the effects of various return sludge ratios in conjunction with raw or rapidly mixed recycled sludge on the phosphate removal efficiency were investigated (Fig. 9). The phosphate removal efficiency increased from 62–75% to 67–81% after applying rapidly mixed sludge compared with application of the same dose of raw chemical sludge. These results indicated that reusing the rapidly mixed sludge had better performance with respect to phosphate removal, than reusing the raw chemical sludge. An explanation for this was that the floc size of the chemical sludge after rapid mixing became smaller than that of the raw chemical sludge, because the rapid mixing induced the temporal detachment of particles from the sludge nucleus. Comparing to the raw sludge, more flocs were released from the rapidly mixed sludge and the specific surface area was increased [32]. Therefore, more nucleation sites for aggregating and adsorbing flocs were created and the phosphate removal efficiency was eventually

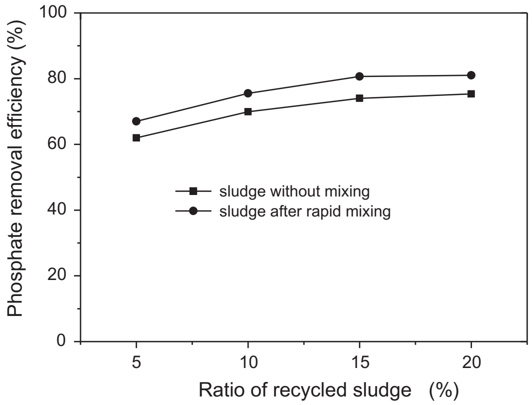

Fig. 9. Effect of rapid mixing of sludge on phosphate removal.

increased after the return sludge was rapidly mixed. Guan et al. found that reducing the floc size of the alum sludge enhanced the SS and COD removal, because the dispersed alum sludge could remove smaller particles than the non-dispersed alum sludge [16]. Our experiment result was consistent with this report.

4. Conclusion

To conclude, we saw that recirculation of sludge during chemical coagulation was employed to enhance the phosphate removal from wastewater. The recycled chemical sludge enhanced the phosphate removal efficiency when the appropriate ratio of recycled sludge (15–20%) was used. The sludge settleability was improved by addition of the chemical sludge. Additionally, rapid mixing of the chemical sludge further enhanced the phosphate removal efficiency. The aging time of recycled sludge also had a significant effect on phosphate removal. It is believed that sweep flocculation and/or physical adsorption play key roles in the enhancement of phosphate removal from wastewater.

Acknowledgments

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