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Effect of mixing conditions on coagulation-flocculation process in the dual-coagulant system of magnesium hydroxide and polyacrylamide

Xinge Li, Bo Li, Jianhai Zhao*, Hongying Yuan and Yongzhi Chi
Tianjin Key Laboratory of Aquatic Science and Technology, School of Environmental and Municipal Engineering, Tianjin Chengjian University, 26 Jinjing Road, Xiqing District, Tianjin, 300384, China

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*to whom all correspondence should be addressed: e-mail: jhzhao@tcu.edu.cn

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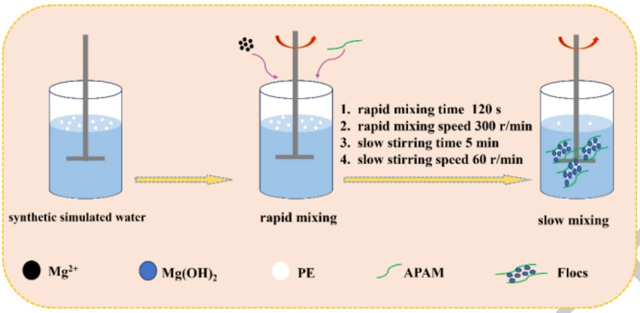
Graphical abstract



Abstract

The effect of mixing conditions on coagulation-flocculation performance and floc characteristics was researched by treating simulated water containing polyethylene with magnesium hydroxide and anionic polyacrylamide as dual-coagulant under alkaline conditions. The floc size distribution and flocculation index were used as the indicators to investigate the floc formation conditions, aiming to discover the relationship between the flocculation effect and floc formation process. The images of flocs were in situ captured using a digital photomicrography to further the knowledge of flocs characteristics, such as floc morphology and settleability. In addition, a single variable was controlled to investigate the effect of different stirring conditions on the experiments by choosing stirring time and stirring speed. The results indicated that under the experimental conditions of rapid mixing for 120 s at 300 r/min or G value of 161.5 s^{-1}, slow mixing for 5 min at 60 r/min or G value of 18.5 s^{-1}, the removal efficiency of polyethylene-contaminated water was more than 84.9% ± 3% (mean ± SD, n = 3). According to the experimental results and coagulation performance of this system, the optimum conditions play an important role in floc growth and PE removal. Furthermore, this work was promising to supply useful recommendations on optimizing coagulation and flocculation.

Keywords: mixing conditions, coagulation-flocculation, magnesium hydroxide, anionic polyacrylamide, polyethylene

1. Introduction

Microplastics (MPs), the microscopic plastics with the size less than 5 mm, were widely used in many industries owing to the stability, strong hydrophobicity, high porosity, large specific surface area and high mobility (Di *et al.* 2025; Parrella *et al.* 2025; Thompson *et al.* 2009). The study, first published in journal of Science in 2004, reported that rising presence of microscopic plastics in the ocean (Richard C. Thompson, 2004). There was subsequently increased concern about the consequences of plastic accumulation in nature, not only in the oceanic environment but also in variety of environmental mediums such as air, soil, and freshwater (Acarer Arat, 2024; Talbot and Chang, 2022; Zhang *et al.* 2022; Zhang *et al.* 2024a). Notably, researchers found a variety of MPs in human blood by analyzing venous blood samples from adult volunteers (Leslie *et al.* 2022). Also, MPs have even been found in the placenta (Ragusa *et al.* 2021). In addition, MPs could easily adsorbed pollutants, containing microorganisms, heavy metals, and organic pollutants, owing to the large surface area and the strong hydrophobic properties of the surface (Zhao *et al.* 2024). MPs could be transferred and enriched along the food chain, thus causing a risk to human health (Yan *et al.* 2024). As a consequence, there is an imperative need to devise sensitive and effective method for removal of MPs in simulated water.

For the last few years, the focus of the investigations was the treatment of the wastewaters mainly by biological, physical, and chemical processes (Kabir *et al.* 2023). Biological treatments, such as microbial degradation, enzyme catalyzed degradation, biofilm technology,

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invertebrate ingestion and phytoremediation, have the advantages of low energy consumption, environmental friendliness, and sustainability, but they also suffer from low efficiency, risk of by-products, and strong environmental dependence (Alfonso *et al.* 2024; Han *et al.* 2024; Huang *et al.* 2024; Thakur *et al.* 2023; Yin *et al.* 2025). Physical treatments remove or reduce microplastics in the environment mainly through mechanical separation, filtration, adsorption, high temperature treatment and other techniques (Bannick *et al.* 2019; Li *et al.* 2023; Rhein *et al.* 2022). Physical treatment methods have the advantages of fast speed, high efficiency and wide applicability, however, there are also disadvantages such as high energy, high cost and incomplete treatment. The main chemical treatment methods are advanced oxidation processes, electrochemical oxidation and coagulation-flocculation (Falco *et al.* 2024; Nguyen *et al.* 2024; Peydayesh *et al.* 2021). Chemical treatments have the advantages of environmental friendliness, wide applicability and high degradation efficiency. Coagulation-flocculation was more often used in water treatment in various industries owing to simple operation, fast flocs settling speed and low capital cost (Fang *et al.* 2024; Panaksri *et al.* 2024; Zaki *et al.* 2023).

In the process of using chemical coagulation to treat simulated water, different mixing conditions will have different effects on the various stages of chemical coagulation, which in turn affects the removal of pollutants by chemical coagulation (Ayoub *et al.* 2014; Nan *et al.* 2016; Yu *et al.* 2011). Limited studies had examined the effects of stirring speed and duration on MPs removal efficiency (Zhang *et al.* 2024b; Zhao *et al.* 2022a). The purpose of this research was to systematically investigate the removal of polyethylene (PE) within an environmentally friendly coagulation system, identifying optimal operating conditions with particular emphasis on the influence of mixing parameters. The parameters, such as coagulant dose of Mg^{2+} and anionic polyacrylamide (APAM), pH and temperature, were measured by the standard jar-test technique. The optimal values of rapid stirring time, rapid stirring speed, slow stirring time and slow stirring speed were determined using the single factor method of proposed optimization method. Furthermore, the effects of different stirring conditions on the coagulation-flocculation performance and floc characteristics of magnesium hydroxide (Mg(OH)$_2$) and APAM were investigated by testing and analyzing the flocculation index (FI) value, removal efficiency and flocs morphology.

The core advantage of Mg(OH)$_2$, as a flocculant, was the outstanding environmental friendliness, coupled with its pH adjustment capabilities and sedimentation efficiency. APAM could offer the core advantages of strong bridging capacity and broad applicability as a flocculant, enabling efficient treatment of diverse water bodies with minimal dosage and controllable costs. Although the coagulation-flocculation treatment of PE was well established, a systematic optimisation scheme for the Mg(OH)$_2$-APAM-PE system remains lacking, and such optimisation was indispensable for practical applications. This research which provides a detailed, systematic dataset for this specific, environmentally relevant application in the PE removal with the "green" coagulant system.

2. Experimental section

2.1. Synthetic simulated water and coagulant

The 50 mg/L simulated water samples were prepared by adding PE (molecular weight 8000 000–10 000 000 Da, Tianjin JiangTian Technology Co. Ltd) to the deionized water. The simulated water samples were adjusted to a pH of 12 with 1 M sodium hydroxide (NaOH, AR, Tianjin Kewei Co. Ltd.) and 1 M hydrochloric acid (HCl, AR, Tianjin Kewei Co. Ltd.), and the solution temperature was maintained at 20 ± 1 °C. Magnesium chloride hexahydrate ($MgCl_2 \cdot 6H_2O$, ≥ 45%, AR, Tianjin Guangfu Fine Chemical Research Institute Co. China) was used for the preparation of Mg(OH)$_2$ coagulant. The reaction was: $Mg^{2+} + 2OH^- \rightarrow Mg(OH)_2(s)$. And anionic polyacrylamide (APAM, molecular weight 8 million, Tianjin Guangfu Fine Chemical Research Institute Co. China) was used as a flocculant in process.

2.2. Indicator detection

An on-line intelligent particle dispersion analyzer (iPDA) (PDA2000, Econovel, Korea) was used to measure the FI of flocs throughout the whole coagulation procedure, in this experimental study. The FI was a quantitative metric for assessing the efficiency of the flocculation process. Its primary function was to evaluate the rate at which suspended particles in water form flocs, their size, and their settling behaviour following the addition of flocculants. During all experiments, the change process of FI value was recorded in real time by collecting data every 5 s. The coagulation-flocculation experimental setup was shown in **Figure 1**. Mastersizer 2000 (Malvern, UK) was employed to generate a measurement of the floc size distribution (FSD). Images of flocs were taken by IX71 digital photomicrography (Olympus, Japan). The turbidity was analysed by a turbidimeter (2100N, Hach, U.S.A.).

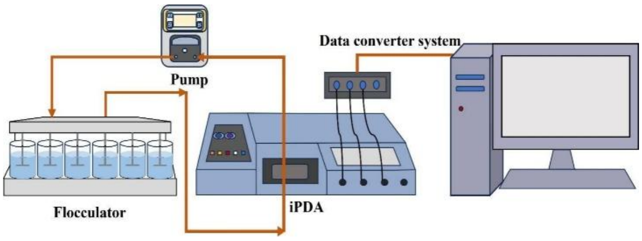


Figure 1. The coagulation-flocculation experimental setup.

2.3. Coagulation-flocculation experiments with jar tests

The Jar Tester with 1-L beakers (ZR4-6, Zhongrun Water Industry Technology Development Co. LTD) was used for coagulation-flocculation experiments. Based on a previous study, 40 mg/L Mg^{2+}, 5 mg/L APAM, 20 ± 1 °C and pH 12 were the optimal experimental conditions for this system (Li *et al.* 2022; Li *et al.* 2025). Base on the experimental conditions, the coagulation stirrer was placed in a thermostatic bath, 4 mL Mg^{2+}solution (10 g/L) was added to the PE simulated water sample after the initial rapid mixing for 30 s. The water samples were then immediately agitated using a hexagonal stirrer. And then, 5 mL APAM (1 g/L) was added to the simulated water before the end of rapid mixing for 10 s. The adding time of APAM was obtained by several comparative experiments of the previous study (Li *et al.* 2022). The optimal coagulant stirring time and stirring speed were determine using the one-factor optimisation method proposed by Cochrane and Cox (M. ROSSINI, 1999). Whilst this approach was suitable for preliminary exploration, it may overlook synergistic interactions between variables. Nevertheless, it was simple and intuitive, straightforward to implement, and places minimal demands on the nature of the function. After stirring, the solution was settled for 5 min to allow the flocs to settle and separate from the solution. After coagulation and sedimentation, the supernatant of the system was collected, then filtered, dried and weighed. In this study, the influences of different stirring conditions on floc growth and PE removal were determined by altering the mixing speed and mixing time.

3. Results and Discussion

3.1. Effect of rapid mixing time

Under the conditions of rapid stirring speed of 250 r/min, slow stirring speed of 60 r/min, and slow stirring for 3 min, the effects of fast stirring time on the formation of flocs were investigated by choosing different times, such as 60 s, 90 s, 120 s, and 150 s, respectively. When the rapid stirring time was controlled for 60 s, as shown in **Figure 2a**, the overall FI value of the floc fluctuates in the range of 2.0 - 4.0, and the maximum value was around 4.4. The rapid stirring time was controlled for 90 s, the FI value of flocs increased significantly, and large number of flocs with FI value above 4.5 appeared. The FI value of the floc further increased when the rapid stirring time was extended to 120 s, at which point the floc FI value was optimal. While extended rapid mixing time up to 120 s, there was a tendency to decrease of the FI value. The results showed that the rapid stirring times of only 30 s and 60 s did not generate enough $Mg(OH)_2$ coagulant to generate enough flocs. The rapid stirring time was extended too long, the high shear force inhibited the nucleation of $Mg(OH)_2$, leading to a reduction in floc number and smaller particle sizes throughout the rapid mixing phase.

In addition, as shown in **Figure 2b**, the PE removal efficiencies at different fast mixing times varied considerably, 32.76 ± 2.24%, 71.67 ± 0.21%, 84.90% ± 3.00%, and 76.56 ± 3.76%, respectively. When the rapid stirring time was 60 s, the solution did not generate sufficient amount of $Mg(OH)_2$ coagulant, resulting in the final amount of flocs generated was less than the efficient removal of PE, rapid stirring time of 90 s, the removal efficiency increased significantly, but still did not reach the optimal level, the other two conditions of the removal efficiency obtained by the closer, but because of the 150 s too long stirring time, resulting in the flocs that had formed were fragmented, resulting in a slight decrease in the removal efficiency of PE. In conclusion, fast stirring time of 120 s, the best coagulation effect was achieved.

The median particle size of flocs was the smallest, the rapid stirring time of 60 s, only 36.7 µm, as shown in **Figure 2c**. The particle size distributions under the remaining three conditions were extremely close, with median particle sizes of 50.7 µm, 57.2 µm and 50.8 µm, respectively. While the median particle size was optimal at the rapid stirring time of 120 s. In agreement with the conclusions obtained from the above analyses of FI and PE removal efficiencies, the rapid stirring time of 120 s was the optimum time for the coagulation reaction in this experiment.

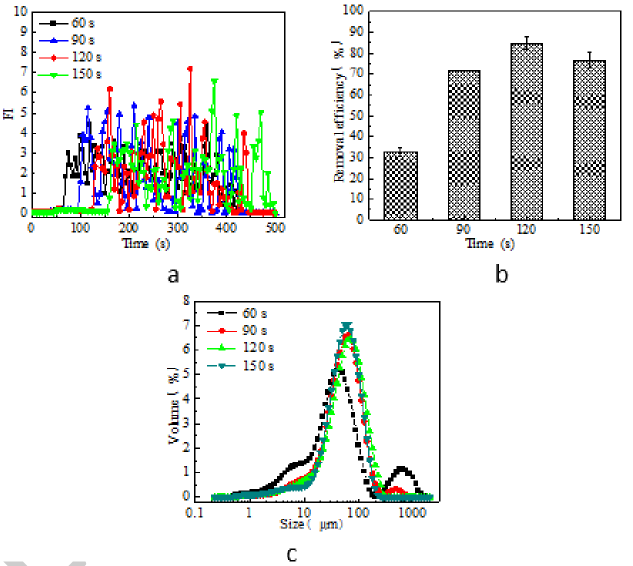


Figure 2. Effect of rapid mixing time on (a) flocculation index (FI), (b) PE removal efficiency and (c) floc particle size distribution.

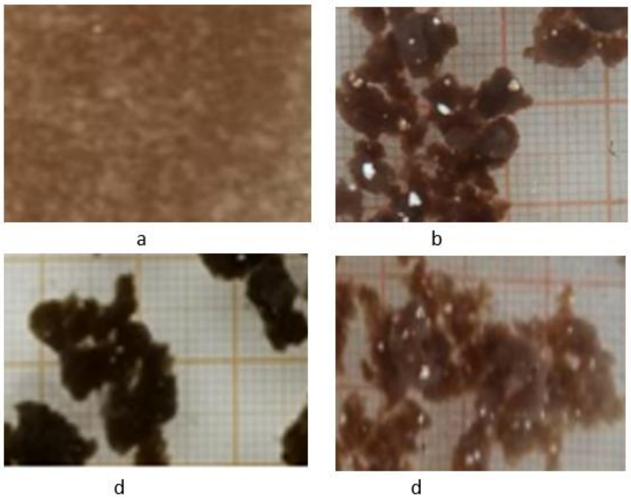


Figure 3. The floc aggregation at different rapid mixing time. (a: 60s, b: 90s, c: 120s, d: 150s)

In order to further analyse the agglomeration state of flocs under different rapid stirring times, images of flocs generated at 60 s, 90 s, 120 s and 150 s were taken for observation. As shown in **Figure 3a**, with the rapid stirring time of 60 s, the generated flocs like a woven web diffused at the bottom of the beaker, which resulted in a low removal of PE due to the porous surface and extreme fragility. In **Figure 3b**, along with the prolongation of rapid stirring time, the original scattered floc began to agglomerate, and the white PE particles embedded in the floc were clearly visible. In compared with the floc at 120 s

(**Figure 3c**), the floc showed a lower degree of compactness. Futhuremore, the flocs were denser at 120 s to reach the optimum state in this experiment base on the colour of the flocs. Referring to **Figure 3d**, when the rapid stirring time was extended to 150 s, the flocs were fluffy again, because of the fact which that microplastics may be released after a long period of settlement. To further illustrate, 120 s was the optimal fast mixing time in this system.

3.2. Effect of rapid mixing speed

To explore the effect of rapid stirring speed, experiments were conducted with a fixed rapid stirring time of 120 s, slow stirring time of 5 min, slow stirring speed of 60 rpm. As shown in **Figure 4a**, at rapid mixing speed of 200 rpm and 250 rpm, larger FI values were observed during coagulation, but occurred less frequently. Rapid mixing speed at 300 rpm, the FI value of flocs showed a tendency to become better gradually, most of the FI values were above 3.0, which indicated that the coagulation effect was the best under this condition. While extended rapid mixing speed up to 350 rpm, there was a tendency to decrease of the FI value. The majority of FI value fluctuated in the range of 2.0 - 3.0, which indicated that the flocculation effect was poor under this condition.

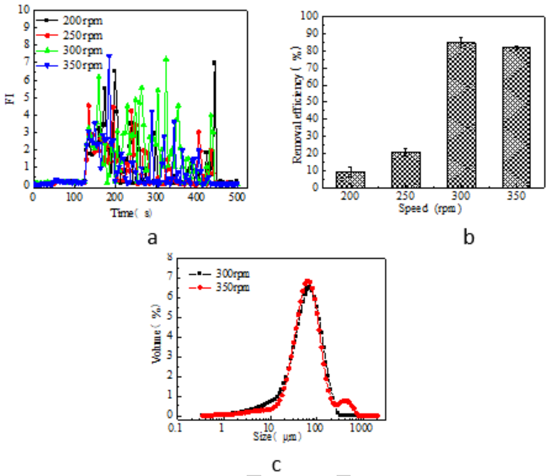


Figure 4. Effect of rapid mixing speed on (a) FI, (b) PE removal efficiency and (c) floc particle size distribution.

The removal efficiency of PE was only 9.11% ± 2.74% and 20.79% ± 1.90% at rapid mixing speeds of 200 rpm and 250 rpm, respectively, as shown in **Figure 4b**. Optimum coagulation was achieved at rapid stirring speed of 300 rpm. The rapid speed was increased to 350 rpm and the removal efficiency was 81.82% ± 0.92% with a slight decrease in the removal efficiency at this condition. The experimental results were in general agreement with the floc particle size results. The median particle sizes of the flocs had slightly different, 70.96 μm and 63.25 μm, respectively, at 300 rpm and 350 rpm as shown in **Figure 4c**.

3.3. Effect of slow stirring time

A range of jar tests were performed to explore effects of slow stirring time. Fixed rapid stirring time of 120 s, rapid stirring speed of 300 rpm, slow stirring speed of 60 rpm, investigate the effect of different slow stirring time on flocculation effect of 1 min, 3 min, 5 min and 7 min. As shown in **Figure 5a** slow stirring flocculation time of 1 min or 3 min, the FI value shows poor performance, which indicating that there was no larger particles flocs during the coagulation process. The short flocculation time was not conducive to the growth of floc agglomeration, resulting in small floc size, which in turn leads to the failure to achieve the optimal sedimentation rate. From the trend of FI values, it could be seen that the slow stirring flocculation time was adjusted to 5 min and 7 min, the frequency of high FI values increased, especially in the slow stirring time of 7 min. However, it was obvious that the number of larger FI values was less than the number of FI values in 5 min slow stirring time, which indicates that long slow stirring time was not beneficial to floc agglomeration and may break up the already agglomerated flocs. The condition of 5 min slow stirring time was more concentrated in the larger FI values, indicating the existence of a continuous process of generating flocs with large particle sizes. Therefore, 5 min was chosen as the optimal slow stirring time in this experiment.

As shown in **Figure 5b**, there was not much difference in PE removal between the two conditions was concerned, 72.44% ± 0.59% and 79.42% ±1.17%, respectively, which was consistent with the direction of the FI value at the slow stirring time of 1min and 3 min. At the slow stirring time of 5 min, the highest PE removal efficiency was also achieved under this condition in this study due to APAM played the role of adsorption and bridging more fully in the rapid stirring stage of the generation of small particle size floc agglomeration. When the slow stirring time was continued to be increased, the removal of PE was less effective, only 59.67% ± 4.00%. It was assumed that the long slow stirring time released the already encapsulated PE particles, resulting in a drastic decrease in the removal efficiency of PE.

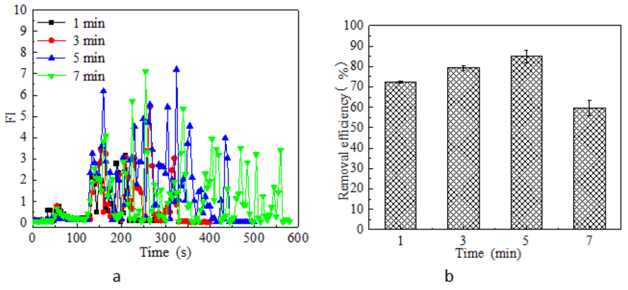
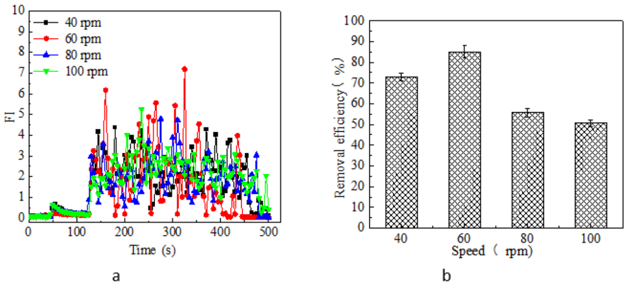


Figure 5. Effect of slow stirring time on (a) FI and (b) PE removal efficiency.

3.4. Effect of slow stirring speed

As shown in **Figure 6a**, there was a good trend of FI values for different slow stirring speeds of 40 rpm, 60 rpm, 80 rpm and 100 rpm. The presence of large FI values at different slow stirring speeds indicated the presence of large flocs. FI values were mostly concentrated in the range of 1.5 - 4.0 under the four rotational speed conditions, proving that no very small floc particles were present overall. However, the PE removal efficiency at the four slow stirring speeds varied considerably. As shown in **Figure 6b**, the slow stirring speeds of 40 rpm and 60 rpm were 72.9% ± 1.93% and 84.9% ± 3%, respectively. The difference in PE removal efficiencies between the two conditions was not significant, which was consistent with the behavior of the FI values in the above graph. After increasing the slow stirring speed to 80 rpm and 100 rpm, the PE removal efficiency decreased significantly to only 55.7% ± 2.10% and 50.6% ± 1.65%, respectively. In summary, it was clear that flocs with larger particle sizes were formed due to the use of APAM during the slow churning process. As the mixing speed increases, the long slow churning process provided a constant high shear force, resulting in the flocs being broken up in the process and the already encapsulated PE particles being released. Therefore, the slow stirring speed of 60 rpm was chosen for this experiment.



a b

Figure 6. Effect of rapid mixing time on (a) FI and (b) PE removal efficiency.

3.5. *Effect of G value during mixing period*

The flocculation effect could be characterized by the value of the velocity gradient G. G value was really the concept of energy consumption, which could be calculated by the power consumed by a unit volume of fluid that is instantaneously twisted by shear (Ayoub *et al.* 2014).

$$G = \sqrt{\frac{P}{\mu V}} \tag{1}$$

Where G was the velocity gradient, s^{-1}, p was the power consumed per unit volume of fluid, W/m^3, μ was the dynamic viscosity of water, Pa·s, V was the volume of the beaker/jar.

Theoretically, the larger the G value, the higher the collision rate of particles and the better the locculation effect (Zhao *et al.* 2022b). However, the increase of G value made the shear force also increase. In practice, it is also found that the shear force was too large to break the flocs. Therefore, it was necessary to choose a suitable G value. This revealed the equilibrium issues at solid-liquid interfaces that were commonly encountered in adsorption/coagulation processes: sufficient fluid shear stress was essential for mass transfer, yet excessive stress could disrupt formed aggregates (Bai *et al.* 2022). As shown in the **Table 1**, the rapid mixing speeds were 200 rpm, 250 rpm, 300 rpm and 350 rpm, which corresponded to a G-value of 93.6 s^{-1}, 126.3 s^{-1}, 161.5 s^{-1} and 201.3 s^{-1}, respectively. Combined with the actual effect of coagulation experiments, a small G value, the collision rate between particles in the solution was low, which could not promote the growth of flocs. While a large G value affected the growth of $Mg(OH)_2$ and the adsorption of humic acid through the rapid stirring, which inhibits the growth of flocs. Therefore, the optimal G value selected in the rapid stirring stage of this experiment was 161.5 s^{-1}.

Table 1. Relationship of rapid stirring speed and G.

Rapid stirring speed (rpm)	**200**	**250**	**300**	**350**
G (s^{-1})	93.6	126.3	161.5	201.3

Table 2. Relationship of slow stirring speed and G.

Slow stirring speed (rpm)	**40**	**60**	**80**
G (s^{-1})	10.3	18.5	27.3

The flocs grow better together in the slow stirring stage, so it was necessary to select a smaller and suitable G value. The relationship between G value and slow stirring speed in the experiment was shown in the **Table 2**. The slow stirring speed of 40 rpm resulted in a small G value of 10.3 s^{-1}, which was not favourable for particle collision. The slow stirring speed of 80 rpm, the G value of 27.3 s^{-1} was relatively large. Therefore, the best G value for the slow stirring stage was 18.5 s^{-1}, which was the best value for the slow stirring stage in combination with the actual effect of coagulation and experience.

4. Conclusions

In this research, the effects of mixing conditions on $Mg(OH)_2$ and APAM coagulation performance and flocs properties were investigated. The mixing speed played a significant role in the flocs formation and growth process. The role of the rapid mixer was to supply a site for the rapid nucleation and precipitation of $Mg(OH)_2$. Slow stirring allows the adsorbed particles of $Mg(OH)_2$-APAM flocs formed during the rapid stirring phase to increase in size in order to net trap and sweep up more colloidal particulate matter. The optimal stirring conditions for the coagulation performance of $Mg(OH)_2$ at the dosage of 40 mg/L Mg^{2+} were as follows: rapid stirring for 120 s at a rotational speed of 300 r/min or a speed gradient G value of 161.5 s^{-1}, followed by slow stirring for 5 min at a rotational speed of 60 r/min or a G value of 18.5 s^{-1}. By adjusting the optimal mixing conditions, the removal efficiency of PE can reach more than 84.9% ± 3%.

5. Acknowledgement

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6. Statements and Declarations

The data used or analyzed during the study are available from the corresponding author on reasonable request.

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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