

Coagulation-adsorption of reactive orange from aqueous solution by freshly formed magnesium hydroxide: mixing time and mechanistic study

Jianhai Zhao, Huanhuan Shi, Meile Liu, Jingfang Lu and Wenpu Li

ABSTRACT

The utilization of magnesium hydroxide was successfully carried out to remove reactive orange by coagulation-adsorption from aqueous solution. The coagulation-adsorption mechanisms and magnesium hydroxide-reactive orange floc property were analyzed through zeta potential, scanning electron microscope (SEM), X-ray diffraction and Fourier transform infrared spectroscopy (FT-IR). Flocculation Index was then discussed with controlled experiments using intelligent Particle Dispersion Analyzer (iPDA) and optimum rapid mixing time of 90 s was obtained for pH 12. The results of this study indicate that charge neutralization and adsorption are proposed to be the main coagulation mechanisms. The FT-IR spectra and SEM showed that reactive orange was adsorbed on the magnesium hydroxide surface during coagulation and adsorption. Freshly generated magnesium hydroxide can effectively remove reactive orange and the removal efficiency can reach 96.7% and 46.3% for coagulation and adsorption, respectively. Adsorption process accounts for 48% of the whole coagulation experiment. The removal efficiency decreased significantly with increasing magnesium hydroxide formation time.

Key words | adsorption, coagulation, magnesium hydroxide, mechanism, reactive orange

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INTRODUCTION

Reactive dyes are being used widespread in textile industry and the unused materials from the processes are discharged amounts of colored effluents (Riera-Torres *et al.* 2010; Verma *et al.* 2012). Typical characteristics of this kind of wastewater include chemical hazards, high pH, high chemical oxygen demand (COD) and strong color (El-Gohary & Tawfik 2009). There are many treatment technologies available for reactive dyes removal, including biodegradation, chemical oxidation, coagulation, electrolysis, and adsorption (Riera-Torres & Gutierrez 2010; Zhou *et al.* 2016). Among these processes, coagulation technology has been carried out for color removal from textile waste effluents due to its low capital cost (Yang *et al.* 2014). Magnesium hydroxide, an environmentally friendly chemical product, has been shown to be an effective alternative to conventional coagulants such as aluminum and iron salts for the removal of textile waste effluents (Tan *et al.* 2000; Semerjian & Ayoub 2003; Wang *et al.* 2009; Zhao *et al.* 2012). The recoverability of magnesium hydroxide may significantly reduce the chemical costs, and formation time of magnesium hydroxide

is very fast. As for alkali wastewater such as reactive dyes waste, magnesium hydroxide coagulation process will be very effective and cheap.

Floc properties and settling characteristics are the main parameters influencing removal efficiency in real industrial scale unit operations (Wang *et al.* 2002; Li *et al.* 2006; Xiao *et al.* 2010). Photometric dispersion technique is useful in monitoring the initial formation of floc after coagulant addition, the effects of mixing conditions on floc formation, growth and the breakage of flocs (Gregory 1985; McCurdy *et al.* 2004; Yu *et al.* 2011). The processes of magnesium hydroxide nucleation and precipitation and then floc formation and growth are important in understanding the coagulation mechanism and the reactive dyes treatment performance (Liu *et al.* 2015; Li *et al.* 2016). Magnesium hydroxide coagulation process is complex and color removal mechanisms are probably as follows: magnesium hydroxide precipitate acts through an adsorptive coagulating mechanism (Leentvaar & Rebhun 1982; Bouyakoub *et al.* 2011), and magnesium hydroxide provides a large

Table 1 | Reactive dye characteristics

Name	Molecular structure	λ_{\max} (nm)
Reactive orange (K-GN)		476

adsorptive surface area and a positive electrostatic surface charge, enabling it to remove the dyes through charge neutralization (Gao *et al.* 2007).

Although there are some studies on coagulation mechanism using magnesium hydroxide as a coagulant in reactive dyes system, there have been limited studies on proportion of the adsorptive coagulating mechanism. The adsorptive coagulation mechanism and proportion of adsorption are still not clear and should be further studied. The main objective of this laboratory study was to evaluate the coagulation mechanisms, especially to understand the relationship between adsorption and coagulation process of magnesium hydroxide. Floc and sedimentation will be studied through analysis of floc size, zeta potential and decolourization efficiency. Furthermore, the effects of magnesium hydroxide formation time on adsorption process are also assessed.

MATERIALS AND METHODS

Synthetic water and coagulant

The simulated wastewater samples were prepared by adding 0.25 g of the reactive orange K-GN (Jinan Xinxing Textile Dyeing Mill, Shandong, China) into 1 L of deionized water. The molecular structure and wavelength of maximum absorbance of reactive orange K-GN for this study are shown in Table 1. NaOH solution (0.1 M) was added to each water sample to control the solution pH value of 12 in the experimental process. A pH-meter (PH₅-25 Shanghai Jinke industrial Co.) was used to determine the pH of the solutions. MgCl₂·6H₂O was used to prepare magnesium hydroxide coagulant. All reagents were of analytical grade and used directly. Magnesium ion concentration was analyzed with ion chromatography (ICS-1500, Dionex, USA). The concentration of the K-GN in the solution was analyzed by UV-VISIBLE spectrophotometer (UV2550 Shimadzu, Japan). The measurements were made at the wavelength

$\lambda = 476$ nm, which corresponds to maximum absorbance. Blanks containing no dye were used for each series of experiments. All the experiments were carried out in duplicate.

Jar test procedures

Batch coagulation experiments with 1 L solutions were carried out on a jar-test apparatus with 1-L beakers (ZR4-6, Zhongrun Water Industry Technology Development Co. Ltd, China) at 20 ± 1 °C. The solutions with 0.25 g/L K-GN were stirred rapidly at 250 rpm for 60 s to 120 s during 150 mg/L magnesium ion addition, followed by stirring at 60 rpm for 10 min, and settled for 30 min before the final concentration of reactive orange was measured. As a comparison in the experiment by adsorption, the same procedures were conducted except the sequence of adding reactive orange K-GN. The water samples were just distilled water at pH 12, to which was added magnesium chloride (150 mg/L Mg²⁺) to prepare magnesium hydroxide at rapid mixing period for 60 s to 120 s, then 0.25 g K-GN was added to 1 L suspension. Slow stirring speed was maintained at 60 rpm for 10 min and settled for 30 min. The adsorption time was not enough to reach equilibrium, but it can reflect the ability to adsorb reactive orange of magnesium hydroxide. Throughout coagulation and adsorption periods, an on-line intelligent Particle Dispersion Analyzer (iPDA) was used to monitor the condition of suspensions. The experiment process was similar to that of Liu *et al.* (2015). Flocculation Index (FI) can reflect the changes of flocs aggregation of a suspension. A higher FI value suggests a bigger size of flocs. A schematic diagram of the experimental apparatus is shown in Figure 1.

Floc and sediments characterization

During the slow mixing period, samples of flocs were taken from below the surface of the suspension. The image of flocs in the sample was captured by IX71 digital photomicrography (Olympus, Japan). When sedimentation process was

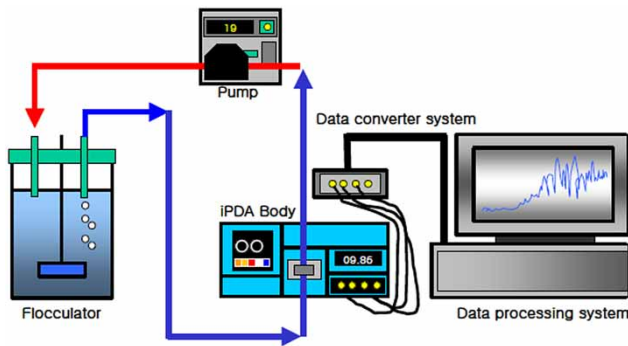


Figure 1 | Experimental apparatus for coagulation-adsorption of magnesium hydroxide.

finished, sediments were withdrawn carefully and then filtered through filter paper and dried naturally for morphology analysis. Crystallinity of the sediments was determined by X-ray diffraction (XRD) with $\text{CuK}\alpha$ radiation (D/MAX-Ultima IV, Japan). Morphology of sediments was observed by scanning electron microscopy (SEM) (FEI, Quanta 200, Czech Republic). The sediment's properties were also analyzed by infrared spectrum analysis (Nicolet iS 10 FT-IR Spectrometer, Thermo Scientific, USA).

RESULTS AND DISCUSSION

Coagulation and adsorption behaviors under different mixing times

Effect of rapid mixing time

As previously found (Liu *et al.* 2015), the optimum coagulant dose of 150 mg/L (magnesium ion) was obtained for pH 12. Based on coagulation performance, the dosage of magnesium ion for the subsequent experiments was chosen as 150 mg/L. Jar test experiments were performed to investigate the effects of rapid mixing time on coagulation and adsorption performance. According to iPDA, a higher FI value means large floc size and significant increases of FI value when aggregation occurs. As shown in Figure 2, the FI values with coagulation are larger than with the adsorption process. The higher FI values, 0.14 and 0.06, were obtained with optimum rapid mixing time of 90 s for coagulation and adsorption, respectively. Magnesium hydroxide coagulation process is similar to the precipitation process, which includes magnesium hydroxide nucleation and combination of reactive orange into flocs. FI values increase first for floc formation and then decrease to a steady-state floc size; there is a dynamic balance between floc growth

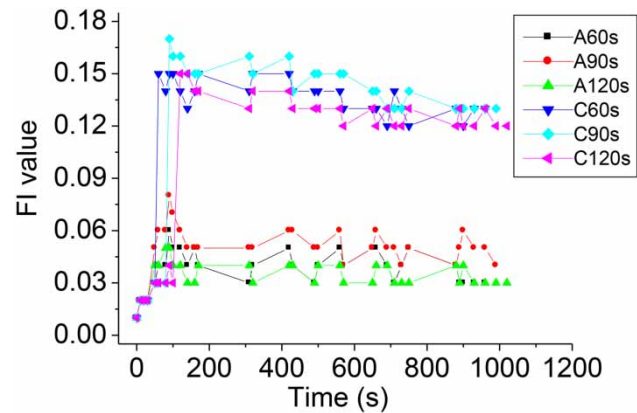


Figure 2 | Effects of rapid mixing time on FI value.

and breakage. In adsorption process, the FI values reach a lower level and remain relatively stable. According to earlier results (Zhao *et al.* 2014), in the early stage of rapid mixing, magnesium hydroxide was formed and particles grew in a very short time. A suitable period of rapid mixing was necessary, and prolonged rapid mixing gave poorer coagulation performance (Yu *et al.* 2011; BinAhmed *et al.* 2015). During the rapid mixing period, voluminous flocs are formed, and thus the increase in floc size primarily results from the enmeshment of particles by flocs. During the slow stir phase, the steady state floc size remains the same.

Figure 3 shows the effects of rapid mixing time on reactive orange removal under different operations. The removal efficiency after coagulation reached 96% under different rapid mixing times from 60 s to 120 s after the 10 min slow mix and 30 min settling. When rapid mixing time is 90 s, the highest removal efficiency is 96.7%. Removal efficiency reaches

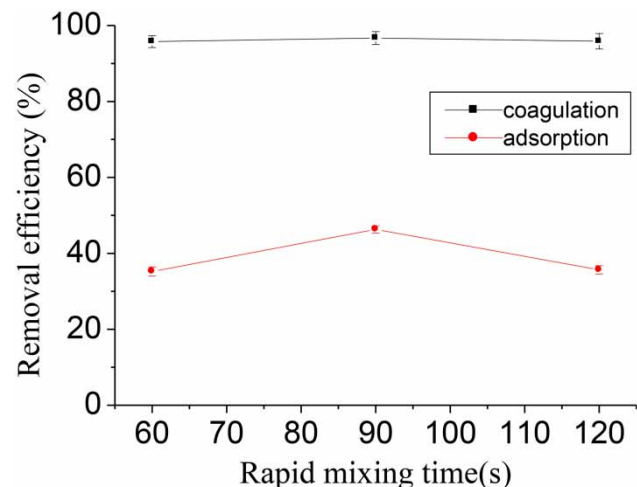


Figure 3 | Effects of rapid mixing time on K-GN removal.

46.3% when rapid mixing time is 90 s in adsorption process. The property of magnesium hydroxide precipitate should play an important role in coagulation and adsorption process. The primary nucleation of magnesium hydroxide also will definitely influence the formation of flocs.

Zeta potential under different slow mixing times

To explore the potential mechanism of reactive orange removal using magnesium hydroxide as coagulant and adsorbent, zeta potential was measured under rapid mixing time 90 s and with slow mixing time 5 min to 20 min. As Figure 4 indicates, zeta potentials were -2.45 mV and -6.04 mV when slow mixing time was 10 min for coagulation and adsorption, respectively. It is likely that higher removal efficiency during coagulation process caused zeta potential near 0 mV. Although slow mixing time increases, the adsorption equilibrium was not achieved; compared with coagulation process, reactive orange was not adsorbed effectively. Based on this observation, it can be reasoned that charge-neutralization is one of the mechanisms for destabilization and removal of reactive orange. This is consistent with the findings of previous study (Zhao *et al.* 2012, 2014). Similar results were also found in the magnesium hydroxide coagulation process, in which magnesium precipitates served as effective coagulant at high pH levels (Semerjian & Ayoub 2003; Ayoub *et al.* 2014). The process of coagulation is complex and may involve several mechanisms. According to removal mechanisms in the literature, charge neutralization and adsorptive coagulating mechanism should be suitable for coagulation process.

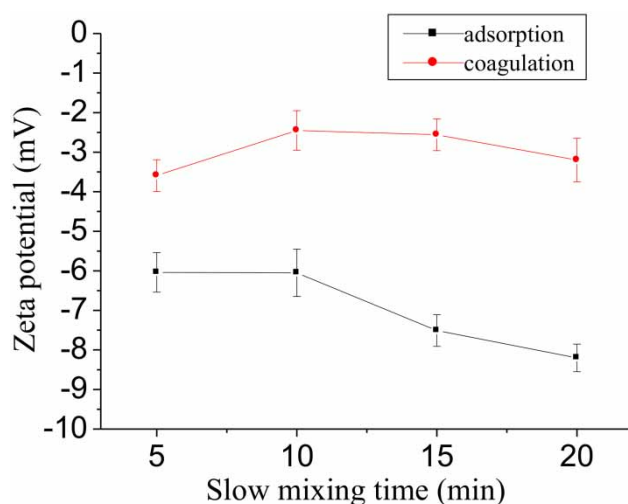


Figure 4 | Zeta potential varying with mixing time.

Floc and sediments characteristics

Images and XRD analysis

The iPDA can tell the growth and breakage of flocs, and the removal efficiency is commonly used to estimate the performance of the coagulation and adsorption process. To gain further insight into the floc and sediment characteristics, image analysis was used to predict the floc properties. Samples of coagulation flocs and sediments for adsorption were withdrawn from sedimentation. The images of flocs and sediments in the sample were captured by IX71 digital photomicrography. As shown in Figure 5, they clearly indicate that the average size of flocs in coagulation system was higher than that in adsorption process. Flocs grew and aggregated to form relatively large particles in coagulation process. The size of some aggregations can reach $15\ \mu\text{m}$. In adsorption process, there is no more aggregation occurring due to the strong

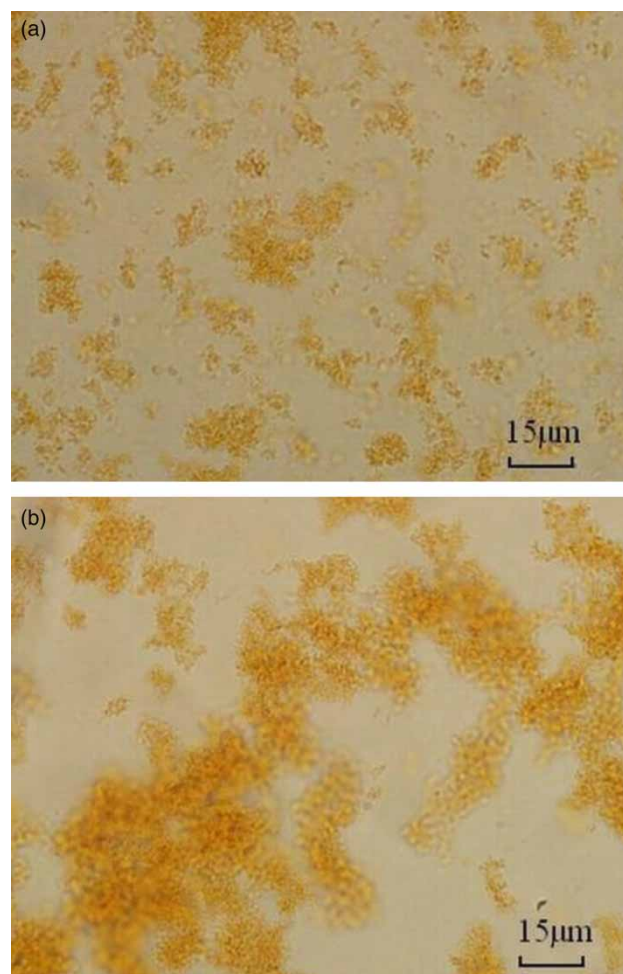


Figure 5 | Floc image analysis: (a) adsorption, (b) coagulation.

repulsion between positively charged particles of magnesium hydroxide. This is consistent with the findings of FI value, which are shown in Figure 2. When magnesium ion is added to a high pH solution, precipitation process of magnesium hydroxide will happen rapidly (Huang et al. 2012). The resulting magnesium hydroxide is a gelatinous precipitate, which was found to serve as an efficient coagulant (Ayoub et al. 2000). To ascertain the precipitation, the crystallographic structure of flocs and sediments was determined by XRD. The XRD patterns of the floc and sediments are presented in Figure 6. The results of XRD proved that the diffraction peaks were almost the same after coagulation and adsorption processes. Both of them indicated the presence of a crystalline structure, evidenced by the appearance of peaks at 19° , 38° and 51° . Figure 6 indicated that magnesium hydroxide crystal was obtained, which is in good agreement with the reported Joint Committee on Powder Diffraction Standards (JCPDS) database.

FT-IR spectral analysis and SEM

In order to confirm the existence of functional groups responsible for coagulation and adsorption of reactive orange, the Fourier transform infrared spectroscopy (FT-IR) spectra of magnesium hydroxide, coagulation flocs and adsorption sediments were shown in Figure 7. In the IR spectrum of the magnesium hydroxide, the peak at $3,700\text{ cm}^{-1}$ was assigned to the free O-H stretching vibration mode of the hydroxyl functional groups, and the peak at $2,360\text{ cm}^{-1}$ was interference peak. The two peaks of the three spectra between $1,650\text{ cm}^{-1}$ and $1,457\text{ cm}^{-1}$ were attributed to the bending vibration of Mg-OH and OH bond in crystal structure, respectively (Wu et al. 2008). The peaks in the $1,488\text{--}1,558\text{ cm}^{-1}$ region were

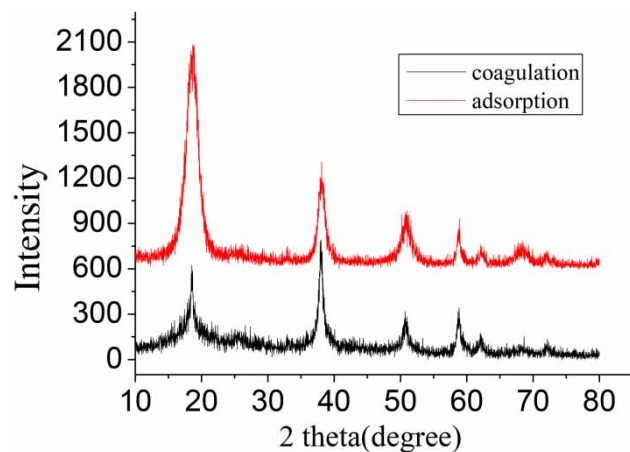


Figure 6 | XRD patterns of the sediments.

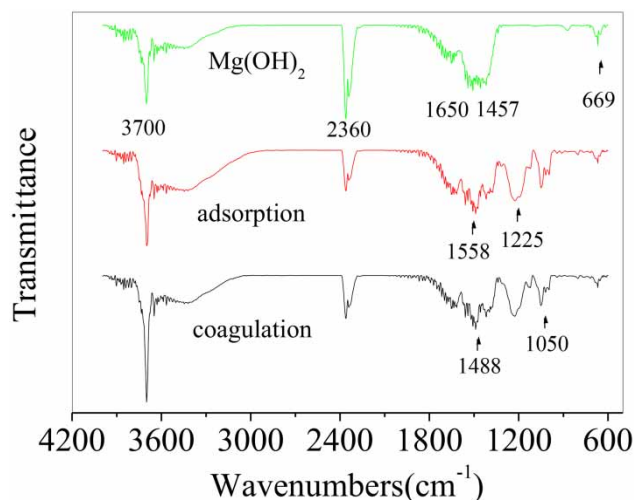


Figure 7 | IR spectrum of $\text{Mg}(\text{OH})_2$ and sediments.

ascribed to the stretching vibration of C=C in the benzene ring and vibration of N=N. The stretching vibration of S=O led to the appearance of the peak at $1,225\text{ cm}^{-1}$ and C-OH led to the peak at $1,050\text{ cm}^{-1}$. The peaks' positions from $1,050\text{ cm}^{-1}$ to $1,558\text{ cm}^{-1}$ are almost the same in coagulation and adsorption processes. The FT-IR spectra showed that reactive orange K-GN was adsorbed on the magnesium hydroxide surface during coagulation and adsorption.

The surface and textural morphology of magnesium hydroxide powder, coagulation flocs and adsorption sediments by SEM images were illustrated in Figure 8. According to the SEM image analysis, the size of $\text{Mg}(\text{OH})_2$ particles was found to range between 1 and $15\text{ }\mu\text{m}$, which is consistent with images shown in Figure 5. Significant changes observed in the surface morphology of dye-loaded adsorbent indicate uneven, irregular surface with molecular cloud of reactive orange. Figure 8(b) indicates that flocs aggregated together in the magnesium hydroxide-reactive orange coagulation system. Since the sediments after settling are mainly determined by the small flocs, the K-GN removal efficiency is only 46.3% in adsorption process. There is no more aggregation occurring due to the strong repulsion between positively charged particles of magnesium hydroxide. The resulting repulsive forces tend to stabilize the suspension and prevent particle agglomeration (Li et al. 2006).

Effects of magnesium hydroxide formation time on adsorption

By comparison of coagulation and adsorption processes, we can determine that adsorptive mechanism should play an

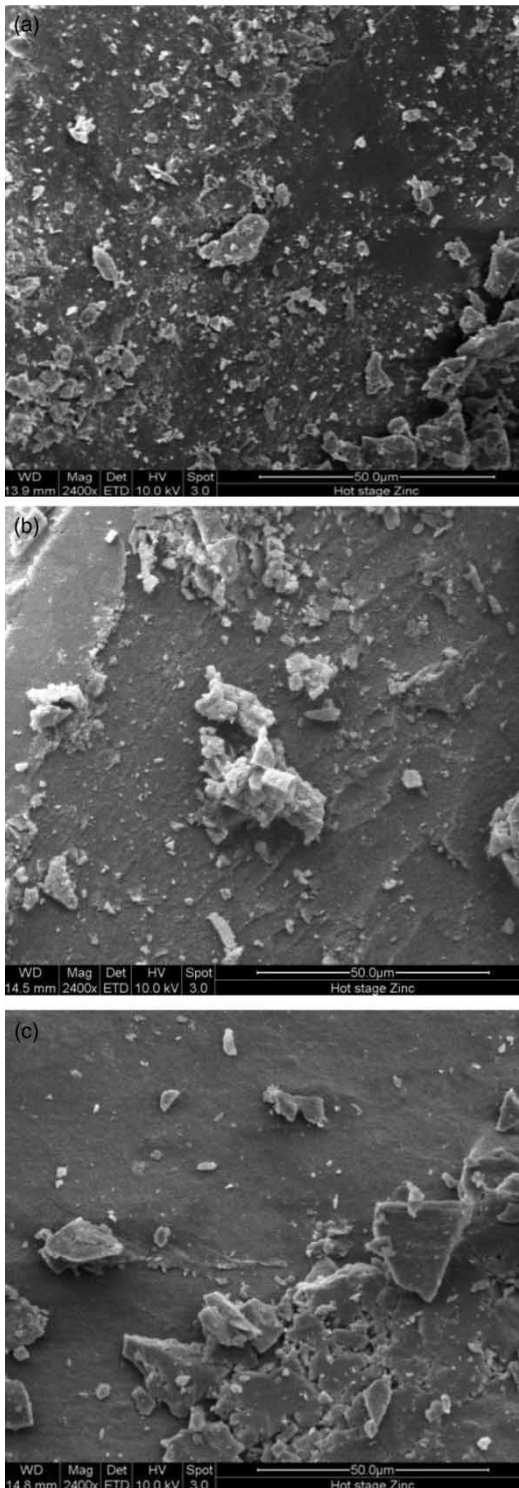


Figure 8 | SEM images of $Mg(OH)_2$ and sediments. (a) $Mg(OH)_2$, (b) after coagulation, (c) after adsorption.

important role in coagulation process. A series of jar tests were conducted in order to investigate the effects of magnesium hydroxide formation time on adsorption process.

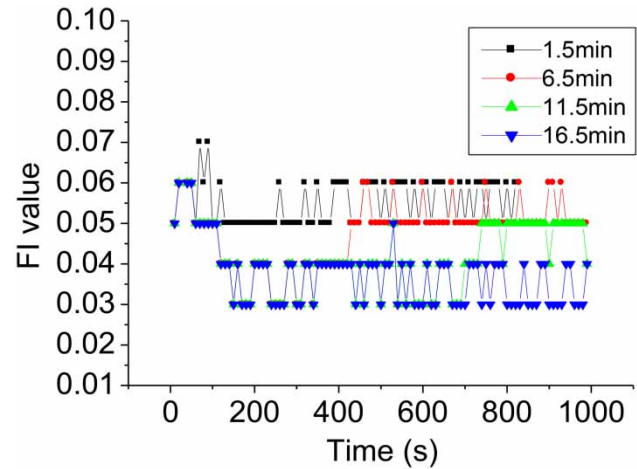


Figure 9 | Effects of formation time on FI value.

The water samples were just distilled water to which was added magnesium chloride in advance to prepare magnesium hydroxide at first and then the reactive dyes were added to the suspension. Stirring speed was maintained at 250 rpm for 90 s and then at a slow stirring speed of 60 rpm for 0 min to 15 min, to allow magnesium hydroxide flocs to grow. In other words, magnesium hydroxide formation time is from 1.5 min to 16.5 min coupled with rapid mixing time 90 s. After this, reactive orange was added to the suspension, followed by stirring at 60 rpm for additional 10 min and settled for 30 min. In this process, magnesium hydroxide acted as adsorbent to remove reactive dyes. The working behavior of the iPDA is generally discussed here. As shown in Figure 9, the higher FI values were obtained with shorter magnesium hydroxide formation time. All the FI values were lower than 0.07 and remained stable with the formation time from 1.5 min to 16.5 min. Just as mentioned above, the variation of the iPDA output FI reflects actually the change in floc size. Different formation time will cause different characteristics of magnesium hydroxide including surface properties, size distribution and zeta potential (Zhao *et al.* 2014). In the early stage of the rapid mixing, large numbers of magnesium hydroxide nuclei were created rapidly and grew fast (Wei *et al.* 2014; Liu *et al.* 2015). However, during the slow mixing period, magnesium hydroxide particles did not grow and remained a stable floc size.

The changes of zeta potential and removal efficiency with different magnesium hydroxide formation times are summarized in Table 2. It can be found that the removal efficiency decreased with increasing magnesium hydroxide formation time. Formation time of 0 min means coagulation process with rapid mixing time 90 s. The highest removal

Table 2 | Coagulation-adsorption performance under different Mg(OH)₂ formation times

Formation time (min)	0	1.5	6.5	11.5	16.5
Removal efficiency (%)	96.71	46.33	45.12	29.71	22.34
Zeta potential (mV)	-2.45	-6.04	-6.55	-14.03	-15.04

efficiency of 96.71% was achieved and zeta potential was only -2.45 mV.

When magnesium hydroxide formation time was increased from 1.5 min to 16.5 min, zeta potential changed from -6.04 mV to -15.04 mV and removal efficiency of reactive orange decreased from 46.33% to 22.34%. Freshly generated magnesium hydroxide can effectively remove reactive orange by adsorption.

Magnesium hydroxide has a positive superficial charge, which attracts the negatively charged reactive dye in both coagulation and adsorption processes. Although it is not easy to distinguish between the charge neutralization and adsorption in coagulation process, through these adsorption experiments we can determine which mechanisms are dominant. The coagulation behavior indicates that charge-neutralization and adsorption must be the main mechanisms. The results in Table 2 also indicate the proportion of adsorption. As mentioned above, formation time 0 min means coagulation process and removal efficiency is 96.71%, while in adsorption process with formation time 1.5 min as comparison, removal efficiency is only 46.33%. It was calculated that the proportion of adsorption in coagulation process accounted for 48%.

CONCLUSIONS

In this research, freshly formed magnesium hydroxide coagulation and adsorption mechanisms and floc characteristics under different mixing times were investigated. The higher the FI values of the system, the higher removal efficiency was obtained. Reactive orange removal efficiency reached 96.7% and 46.3% for coagulation and adsorption, respectively. Flocs formed at rapid mixing time 90 s were relatively large and aggregated together in coagulation process. The smaller particles were obtained when increasing slow mixing time in adsorption process. During the coagulation process, magnesium hydroxide-reactive orange flocs were aggregated mainly through charge-neutralization and adsorptive mechanisms. The proportion of adsorption accounted for 48% and played an important role in the

process. Reactive orange K-GN was adsorbed on the surface of magnesium hydroxide in both coagulation and adsorption processes.

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