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PREPARATION, CHARACTERIZATION AND COAGULATION BEHAVIOUR OF A NOVEL INORGANIC COAGULANT – POLYFERRIC(III)-MAGNESIUM(II) SULFATE

A novel coagulant, polyferric(III)-magnesium(II) sulfate (PFMS), was prepared. The preparation technology was optimized and the optimized PFMS product was used to treat actual textile wastewater. The performance of PFMS was evaluated and compared with those of conventional coagulants. X-ray diffraction and infrared spectrometry show that PFMS is a macromolecular composite polymer of ferric and magnesium based on –OH bonds. The coagulation performance of PFMS is better than those of PFS, PAC, and PFC. The coagulation mechanism of PFMS in treating actual textile wastewater is primarily driven by charge neutralization at low dosages and co-precipitation netting at high dosages.

1. INTRODUCTION

Coagulation is an important process in water and wastewater treatment, particularly in destabilization of colloid suspensions as well as in the removal of dissolved and suspended pollutants [1]. Addition of hydrolyzing metal salts based on aluminum or iron can destabilize colloidal impurities and transform small particles into large aggregates to adsorb dissolved organic materials. The aggregates are then removed by sedimentation. Aluminum- and iron-based agents are the most commonly used coagulants in water and wastewater treatment. Conventional inorganic ferric or aluminum salts such as $AlCl_3$, $Al_2(SO_4)_3$, $FeCl_3$, and $Fe_2(SO_4)_3$ hydrolyze rapidly and in an uncontrollable manner. Over the last few decades, inorganic polymeric flocculants (IPFs)

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such as polyaluminum chloride (PACl), polyferric sulfate (PFS), polyferric chloride (PFC), and polyaluminum sulfate (PAS) have been used in water and wastewater facilities as alternative coagulants. IPFs have a high charge neutralization capacity and are more effective at lower dosages than conventional reagents [2]. Recently, the use of IPFs has noticeably increased because of their low cost and wide availability [3]. Such products that are usually prepared by partial neutralization of aluminum or iron salts have been shown to be advantageous over conventional coagulants because of their higher efficiency for removing particulate and/or organic materials, as well as their low sludge production.

The lifetime cumulative aluminum intake reportedly plays a role in the development of neurological disorders, such as Alzheimer's disease [4]. Iron-based agents have been proven to be safe for human consumption in drinking water treatment according to a preliminary toxicity study [5]. Accordingly, researchers focus on development of iron-based agents. However, given their high production cost, iron-based agents are not widely used as an alternative to aluminum-based agents in China. Numerous studies are being conducted to improve IPFs by introducing other components for coagulation. Magnesium salts have been shown to remove impurities or contaminants, and thus act as an effective alternative coagulant for water and wastewater treatments [6–9]. Li [10] used aluminum-magnesium hydroxide mixture to treat the model anionic dye Reactive Brilliant Red K-2BP and obtained satisfactory results. Sang [11] evaluated magnesium as an additive in a novel type of IPF for the post-treatment of landfill leachate at a laboratory scale. In China, solid and liquid magnesium resources abound, with the magnesite resource being the most abundant in the world. These resources enable multipurpose utilization of magnesium such as in water treatment to address the serious problem of water pollution. Thus, maximizing the use of magnesium resources in water and wastewater treatment is an important research direction for environmental workers and researchers.

Textile wastewater generated by various production steps in textile factories has high pH and contains dyes, detergents, oil, suspended and dissolved solids, as well as toxic and non-biodegradable matter, making it very difficult to treat by conventional wastewater treatment methods [12]. Large amounts of dye effluents are produced by the textile industry in China. The presence of dyes in water is aesthetically undesirable because these dyes are highly visible even at very low concentrations. The majority of synthetic dyes are also toxic to some aquatic organisms and may pose serious health threats to humans [13]. Therefore, regulations on dye pollutants are increasingly becoming stringent. Dyes in wastewater have to be efficiently removed before being discharged into receiving waters. Many physicochemical technologies [14] have been developed for textile wastewater treatment such as coagulation, adsorption, membrane filtration, and advanced oxidation. Each technology has its advantages and disadvantages. Advanced oxidation processes can effectively remove most dyes but at a relative high costs. Membrane filtration has some special features advantageous over other

methods but the capital cost and clogging problems limit its application [15]. Active carbon adsorption is also expensive and the regeneration operation is complex, making manufacturers and enterprises unwilling to use it. If low-cost, high-performance adsorbents can be available, adsorption technology would be extensively used [15]. Meanwhile, coagulation is a common process in water and wastewater treatment, including textile wastewater treatment. This process can be applied in a large scale with relatively high operability and cost effectiveness. Direct dyes are widely used and their removal by coagulation has been studied by some researchers [16, 17]. However, the coagulants used in most of these studies are traditional aluminum- and ferric-based salts, or synthesized organic coagulants.

In this study, a novel IPF, polyferric(III)-magnesium(II) sulfate (PFMS), was fabricated and used to treat actual textile wastewater. This work aimed to optimize the preparation technology, measure the species distribution of Fe(III) in PFMS, and investigate the coagulation performance in treating actual textile wastewater based on a comparison with other popular coagulants. The structure of PFMS was explored by X-ray diffraction (XRD) and Fourier-transform infrared (FTIR) spectrometry. The coagulation mechanism was evaluated by measuring the ζ potential of the flocs produced *in situ*.

2. MATERIALS AND METHODS

Preparation of the coagulant. All reagents used for the preparation of PFMS products were analytically pure chemicals and all solutions used for synthesis of the coagulant were prepared with deionized water. PFMS products were fabricated by oxidizing iron in a ferrous state to ferric state, then carrying out a partial prehydrolysis process by the addition of a designated quantity of base ($NaHCO_3$) and followed by a polymerization stage (aging process) to produce stable mixtures of Fe(III)-Mg(II) polymeric species. A typical procedure about the preparation of PFMS coagulant is as follows:

In the first step of oxidation, according to different Fe/Mg molar ratios, 39.72 –47.67 g of $FeSO_4 \cdot 7H_2O$, 7.04–14.08 g of $MgSO_4 \cdot 7H_2O$ and 37 cm^3 H_2SO_4 (20%) were added into deionized water to obtain a mixed solution; the total metallic salt concentration (calculated as [Fe] + [Mg]) in the mixed solution was 2.0 mol·dm^{-3}; and then 2.66–3.19 g $NaClO_3$ according to various Fe/Mg molar ratios was gently added into the mixed solution with continuing vigorous stirring until more than 99% of ferrous ions had been oxidized to ferric ions. Three different Fe/Mg molar ratios (2.5/1, 4/1 and 6/1, respectively) were evaluated in this study. The required oxidation time depends on the residual concentration of ferrous ions in the mixed solution, which should be lower than 0.5 g·dm^{-3} as ferrous ions. The residual concentrations of ferrous ions were detected by the phenanthroline spectrophotometric method.

In the secondary step of partial hydrolysis stage, the temperature of mixed solution was raised and kept at required reaction temperature; 1.0 mol·dm^{-3} of NaHCO$_3$ was added to the Fe(III)-Mg(II) solutions to obtain the desired [OH]/([Fe] + [Mg]) ratio using a 655 Dosimat micro-titration apparatus under vigorous stirring. The reaction temperature and the [OH]/([Fe] + [Mg]) ratio to be evaluated were varied in the range of 40–85 °C and 0.1–0.5, respectively. The [OH]/[Fe]+[Mg] ratio is hereafter denoted as r, where [OH] and [Fe] + [Mg] are the total (molar) amounts of [OH] and [Fe] + [Mg] added to the solution, respectively.

In the followed polymerization stage, performed in a water bath using a HZS-H water bath oscillator, the obtained solution was heated or cooled to the designated aging temperature and kept at this temperature for the designated time. The aging temperature and aging time to be evaluated were varied in the range of 40–85 °C and 1–4 h, respectively.

After the polymerization stage, the PFMS product was prepared. The preparation parameters of different PFMS products are summarized in Table 1.

Table 1

Preparation parameters of various PFMS products

No.		Hydrolysis stage		Polymerisation stage	
	r	Temperature [°C]	Fe/Mg ratio	Duration [h]	Temperature [°C]
A	0.1				
B	0.2				
C	0.3	55			
D	0.4		4/1		
E	0.5				55
F		40		2	
G		70			
H		85			
I	0.3		2.5/1		
J			6/1		
K		55			40
L					70
M			4/1		85
N				1	55
O				4	

Jar test procedures. Coagulation tests were conducted using simulated dye wastewater samples and actual textile wastewater samples. The simulated dye wastewater was studied to determine the optimum preparation technology of PFMS. It was hoped that the use of pure dye solutions would make it possible to obtain information about the colour removal efficiency of different PFMS products. A stock solution con-

taining 10 $g \cdot dm^{-3}$ dye (Direct Blue 15) was first prepared for the simulated dye wastewater. The test water samples were prepared by diluting the stock solutions with tap water to 50 $mg \cdot dm^{-3}$. Distilled water served as a reference, the maximum absorbance wavelength (596 nm) was read using a spectrophotometer (UV-2450). For the simulated dye wastewater, the percentage of colour removal was calculated by comparing the maximum absorbance values of the test water samples after treatment to those of the original dye water samples. The actual textile wastewater was obtained from a textile factory in Nanchang, Jiangxi Province, China. The wastewater was dark blue and mainly contained Direct Blue, additives, and others. The COD and colour of the actual textile wastewater were determined according to *The Manual Standard Methods for the Examination of Water and Wastewater* developed by the China Environmental Protection Agency. Typical characteristics of the wastewater were as follows: COD = 948 $mg \cdot dm^{-3}$, colour = 500 times, pH = 8.3.

Coagulation experiments were performed by jar tests in a six-gang stirrer. Each beak contained 1.0 dm^{-3} of wastewater sample. The jar tests were conducted with 1 min of rapid mixing at 300 rpm and followed by 15 min of slow mixing at 70 rpm. After settling without any agitation for 30 min, the mixed samples were collected from 2.0 cm below the surface of the test wastewater for analysis. Each coagulation test was run in triplicate, and the results reported in this paper are the arithmetic average result of three tests. The ζ potential of fine particles remaining in the supernatant was measured using a Malvern Zetasizer 3000.

Characterization of the PFMS. The PFMS were analyzed by FTIR using a FTIR Spectrophotometer (model Tensor27 Germany). Briefly, a small amount of samples were taken from the freeze-dried PFMS and mixed with certain amount of KBr (5% of the mixture), which resulted in formation of a pellet that was suitable for FTIR analysis. FTIR experiments were then conducted and the spectra were taken in the range of 4000–400 cm^{-1}.

XRD was conducted using an X-ray diffractometer (model D/Max-RC, Japan) to determine the crystalline phases in the solid coagulants with Cu-Ka radiation in the 2θ range of 5.00–79.98° at a scan rate of 4°/min.

Measurement of the degree of iron polymerization. The degree of iron polymerization was measured by the ferron-timed spectroscopy method using an ultraviolet spectrophotometer (UV-2450). Different species of iron are usually classified into three categories, namely, Fe_a (oligomers), Fe_b (polymers), and Fe_c (precipitated species), based on the different reaction times of iron species with ferron reagent (8-hydroxy-7-iodoquinoline-5-sulfonic acid). Ferron reagent can immediately form complexes (within 1 min) with single ferric ions, as well as with monomeric and dimeric species. It can slowlier form complexes with medium- and high-molecular-weight iron polymers (within 180 min), and the precipitated ferric species do not react with the ferron

reagent. These complexes can absorb light at a maximum of 600 nm. Therefore, the absorbance at 600 nm was measured in predetermined time intervals (1 or 180 min), and each absorbance corresponded to the respective ferric species concentration (Fe_a or Fe_b). The concentration of precipitated iron species, i.e., as Fe_c, was calculated by subtracting the two other Fe species from the total iron concentration.

3. RESULTS AND DISCUSSION

3.1. EVALUATION OF THE OPTIMUM PREPARATION CONDITIONS OF THE PFMS PRODUCTS

In PFMS products, iron is the major component and magnesium is the assistant component. The species distribution of Fe(III) significantly affects the coagulation performances of different PFMS products. Fe_a and Fe_b induce better coagulation efficiency than Fe_c. Fe_b has been universally acknowledged as the most efficient species for coagulation. Therefore, the effects of different preparation conditions on the species distribution of Fe(III) were investigated by the ferron-timed spectroscopy method. Figure 1 shows the species distribution of Fe(III) in the PFMS products.
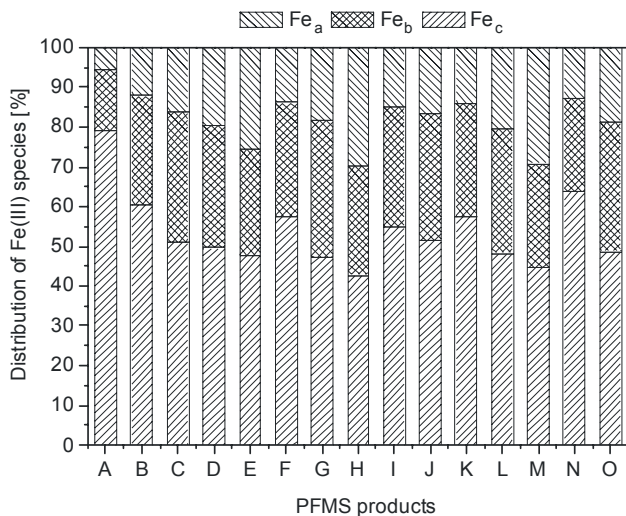

Fig. 1. Species distribution of Fe(III) of various PFMS products

Figure 1 shows that r has a detrimental effect on the species distribution of Fe(III). The proportion of Fe_b species increases with increased r from 0.1 to 0.3, reaching a maximum at $r = 0.3$, and then decreasing at $r > 0.3$. The proportion of Fe_a decreases with increased r, whereas the proportion of Fe_c increases with increased r for all r values. Coagulants F, G, and H were also prepared to evaluate the effect of the reac-

tion temperature. Evidently, with increased reaction temperature, the percentage of Fe_b initially increases within 40–55 °C, almost remains invariant within 55–70 °C, and then decreases at 85 °C. A higher percentage of Fe_b appears at moderate temperatures. For all reaction temperatures, the percentage of Fe_a always decreases and the percentage of Fe_c always increases with increased reaction temperature. To evaluate the effect of Fe/Mg molar ratios on the species distribution of Fe(III), coagulants I and J were prepared. No evident effect of the Fe/Mg molar ratios on the species distribution of Fe(III) is observed. The percentages of three species of Fe(III) almost remain stable with the variation in Fe/Mg molar ratios, also suggesting that no evident effect of magnesium salt on the species distribution of Fe(III). However, the introduction of magnesium can improve the stability of PFMS products and the amount of Fe_b remains at a higher proportion in the storage process than that without magnesium.

An increase in aging temperature at this stage has a direct effect on the polymerization of Fe(III) species (coagulants K, C, L, and M). With increased aging temperature, the percentage of Fe_b initially increases within 40–55 °C and then it decreases at aging temperatures higher than 55 °C, which possibly leads to the breakage of Fe–Fe and Fe–O–Fe bonds. The highest percentage of Fe_b appears at 55 °C. For all aging temperatures, the percentage of Fe_a always decreases and the percentage of Fe_c always increases with increased aging temperature. Within the aging times (coagulants N, C, and O) of 1–2 h, the Fe_b and Fe_c percentages increase whereas the Fe_a percentage decreases with increased aging time. However, with increased aging time longer than 2 h, the amounts of Fe_b and Fe_c slightly increase whereas the amount of Fe_a slightly decreases. In other words, a low tendency of Fe_a transformation to Fe_b and Fe_c during the aging stage of the PFMS products is observed.
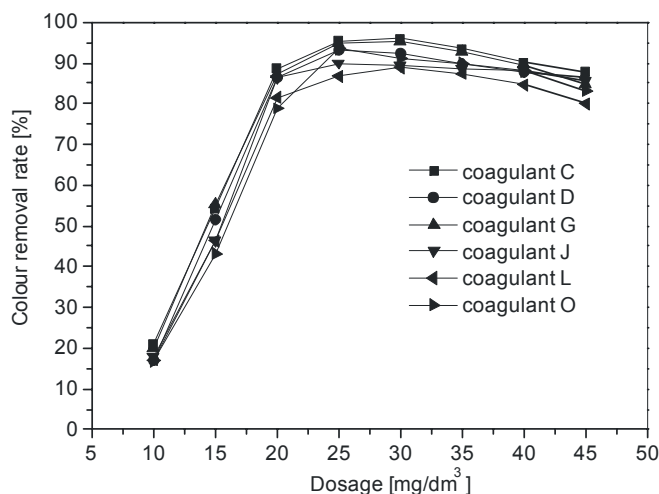

Fig. 2. Colour removal rates of various PFMS products
in treatment of simulated dye wastewater

The coagulation performances in simulated dye wastewater treatment were investigated to evaluate the PFMS capacity. Only the coagulants with a high percentage of Fe_b were used because of the analytical limitation. The PFMS products used were the following: coagulants C, D, G, J, L, and O.

As shown in Figure 2, for various PFMS samples, the trends of the colour removal rates of simulated dye wastewaters are similar to the dosage; the colour removal rate initially increases and then decreases. Generally, the treatment of wastewater with coagulants C and G leads to a better coagulation performance than the other coagulant samples. Considering the colour removal and species distribution of Fe(III), the application of coagulant C is suggested to facilitate the preparation of PFMS. Increased reaction temperature requires more energy during the preparation of PFMS, which may be unsuitable for the industrial preparation and application of the coagulant. Therefore, coagulant C was fixed in subsequent studies and the optimum preparation conditions were as follows: $r = 0.3$, reaction temperature – 55 °C, Fe/Mg molar ratio – 4/1, and aging conditions – 55 °C, 2 h.

3.2. STRUCTURE ANALYSIS OF PFMS

The FTIR spectra of the coagulant PFMS after optimization are shown in Fig. 3. The spectra exhibit two characteristic peaks at around 3360 and 1620 cm^{-1} which can be attributed to the stretching vibration of –OH and bending vibration of H–O–H in the coagulants. A strong band appears within 1150–1000 cm^{-1}, and these peaks can be assigned to the stretching vibration of Fe–OH–Fe, Mg–OH–Mg, and/or Fe–OH–Mg. In particular, 1122 cm^{-1} and 1034 cm^{-1} can be attributed to the stretching vibration of Fe–OH–Fe and Mg–OH–Mg, respectively.
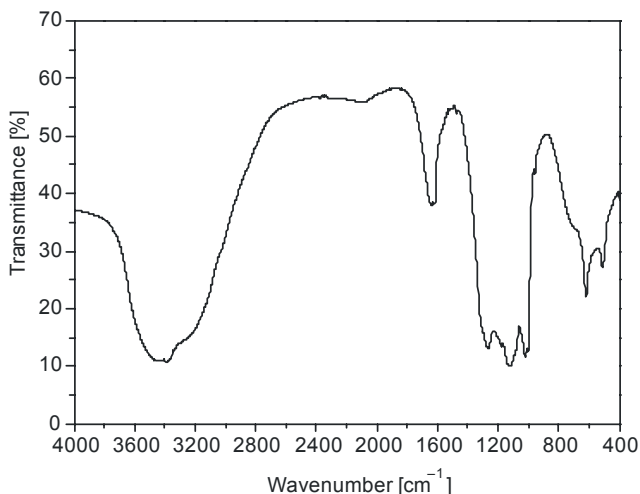

Fig. 3. FTIR spectra of PFMS (coagulant C)

A characteristic peak at around 655 cm^{-1} can be observed, assigned to the stretching vibration of Fe–OH. However, the peak at 510 cm^{-1} in the case of PFMS is assigned to the stretching vibration of Mg–OH. These experimental findings indicate that the coagulant PFMS is a complex polymer of ferric and magnesium formed by –OH bonds.
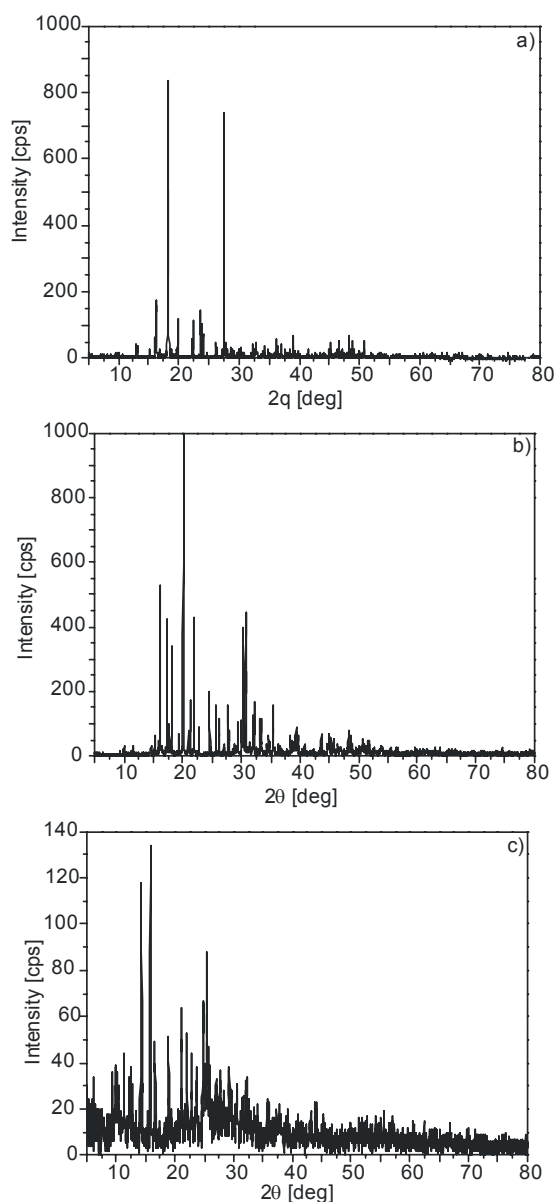

Fig. 4. XRD patterns of two precursor materials:
a) FesO$_4$·7H$_2$O, b) MgSO$_4$·7H$_2$O and c) PFMS (coagulant C)

The XRD spectra of PFMS and two precursor materials, $FeSO_4 \cdot 7H_2O$ and $MgSO_4 \cdot 7H_2O$, evidently different, are shown in Fig. 4. The spectra of PFMS present semi-amorphous shapes, some crystal shapes, and some amorphous shapes. On the other hand, the feature peaks of the XRD spectra of the two precursor materials disappear. These findings demonstrate that PFMS is a novel polymer different from the precursor materials. The disappearance of the crystalline property is further evidence that PFMS is a complex polymer of iron and magnesium formed by –OH bonds. The loss of the crystalline property indicates that novel polymer morphologies differing from the precursor materials are produced during the preparation of PFMS. The above analysis shows that some reactions occur between the two precursor materials, Fe^{3+} and Mg^{2+}, resulting in the formation of new substances with better coagulation performance.

3.3. COAGULATION PERFORMANCE OF PFMS
IN THE TREATMENT OF ACTUAL TEXTILE WASTEWATER

The optimized PFMS was used in the treatment of actual textile wastewater to study its coagulation performance and compare with those of three commercial coagulants.
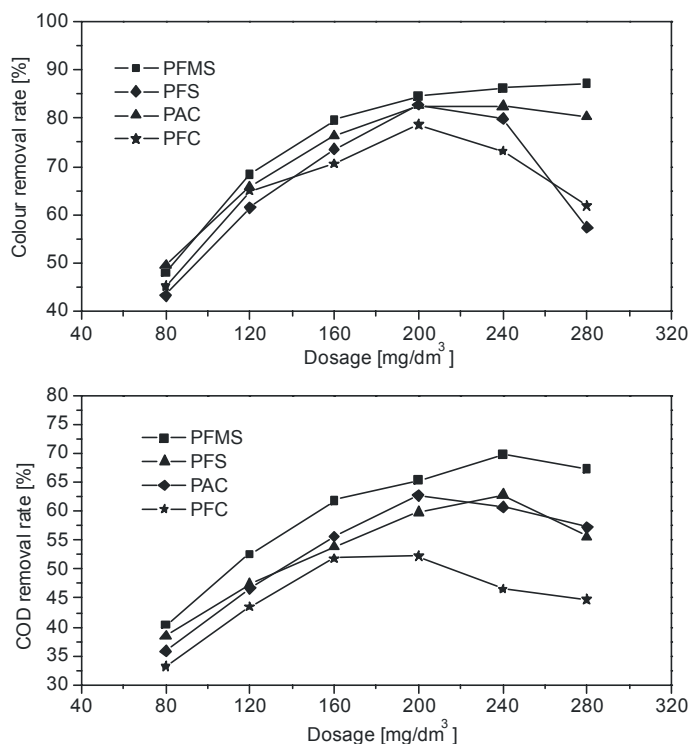

Fig. 5. Coagulation efficiencies with changes in the dosage of the coagulants:
a) colour removal, b) COD removal

The pollutants in actual textile wastewater were evaluated in terms of the COD and colour. The coagulation efficiencies of PFMS and other three commercial coagulants were investigated, and the results are shown in Fig. 5. A comparison of the coagulation efficiencies of the four coagulants reveals that the COD removal rate of PFMS is higher than those of the other coagulants under the experimental conditions. The highest removal rate is 70% at the dosage of 240 mg·dm^{-3}. PAC and PFS also exhibit good coagulation performance, and the COD removal rates are over 50%. However, the coagulation efficiency of PFC is low and the COD removal rate is about 40%. Regarding the colour removal, the coagulation efficiency of PFMS is also higher than those of the other coagulants. Within the range of dosages tested, PFMS can typically increase the colour removal rate from 10% to 20% compared with the other coagulants. Overall, the higher performance of PFMS than the other coagulants indirectly demonstrates the role of magnesium salts in PFMS.

During the coagulation of PFMS, adsorption and charge neutralization initially occur when various species of iron and magnesium with positive charges come into contact with the negatively charged colloidal particles in water and wastewater. Certainly, some species transformation of iron and magnesium simultaneously occur, including certain complex reactions between various species and hydroxyl or other functional groups of colloidal particles. These transformations indicate the generation of new copolymers based on further hydrolysis polymerization using colloidal particles as the matrix. These changes likely result in the combination of species of iron and magnesium with colloidal impurities. Therefore, both positive and negative charges are neutralized to some degree, and the stability of the colloidal impurities declined or even completely eliminated, which is dependent on the characteristics as well as amount of colloidal impurities and coagulant dosage. If the colloidal impurities in wastewater have a certain size and possess the ability to self-aggregate, large flocs easily form with the added coagulant, which results in good sedimentation. However, if the colloidal impurities are relatively small or do not form large flocs, the coagulant dosage must be continuously increased. Under some conditions, when the dosage increases to a certain amount, colloidal particles and various species of iron and magnesium adhere to each other and form much larger flocs. During the precipitation of these flocs, the partially stabilized particles are netted and removed. At this time, co-precipitation netting may play a role in the removal of colloidal impurities.

To determine the coagulation mechanism of PFMS, the COD removal rate and ζ potential of actual textile wastewater at various dosages are measured, and the results are shown in Fig. 6. The COD removal rate initially increases to the peak value and then gradually decreases with the increasing dosage. The ζ potential of colloidal particles in the effluent of actual textile wastewater gradually increases from –25 mV to –8 mV. This indicates that the negative charges can be decreased by adding of PFMS. However, the ζ potential of the effluent after coagulation is relatively high and

not close to zero, which indicates that the negative charges are not completely neutralized by the added PFMS.
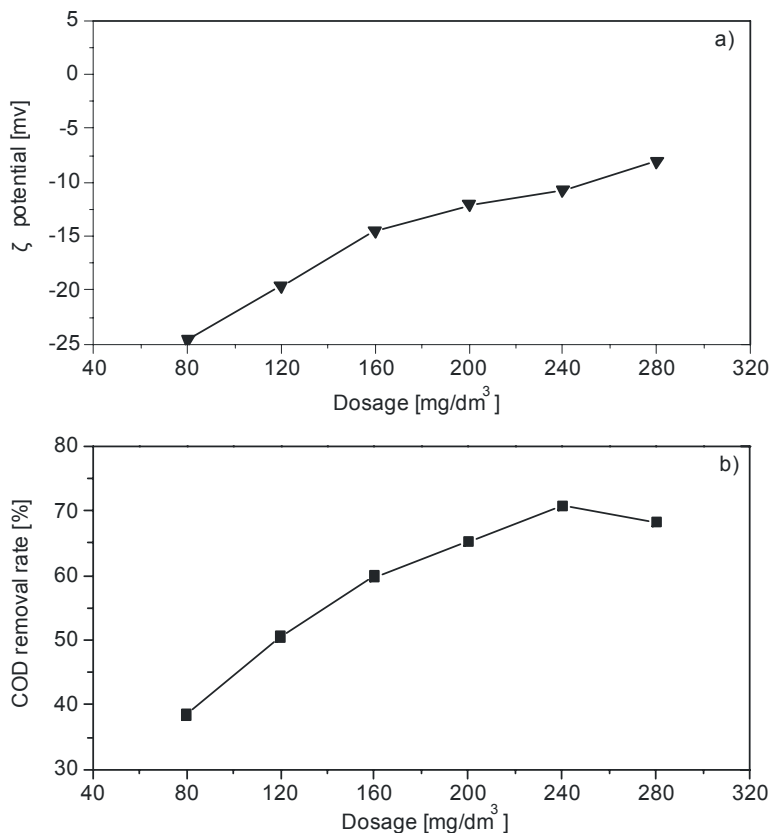

Fig. 6. Variation in the relationship of ζ potential (a) and the COD
removal rate (b) with changes in the dosage of PFMS (coagulant C)

At low dosages (<80–160 mg·dm^{-3}), the amount of flocs formed in the coagulation system is not sufficient to enable adsorption bridging. Certainly, the flocs cannot possibly play a role in co-precipitation netting. The coagulation efficiency primarily depends on the self-aggregation that occurs after the neutralization of the surface negative charges of colloidal particles in the actual textile wastewater by the positive charge of the coagulant. The smooth increase in the ζ potential, which is in line correlation with increase of dosage, confirms this finding.

If the charge neutralization is the only path for coagulation, the ζ potential should be highly correlated with the coagulant dosage. A high COD removal rate is achieved when the ζ potential is close to zero [18]. At high dosages (>200–240 mg·dm^{-3}), the ζ potential of the colloidal particles is always negative and the COD removal rate of

the system is optimal within a wide dosage range. At the pH of experiment, there is a trend for the formation of flocculent amorphous precipitates of $Fe(OH)_3$ and $Mg(OH)_2$ with few charges in the coagulation system regardless of the species distribution of iron and magnesium. The increasing amplitude of the ζ potential of the colloidal particles is small within a wide dosage range because of the reduced charge. The flocculent amorphous precipitate of $Fe(OH)_3$ has a large specific surface area and the precipitate of $Mg(OH)_2$ has good adsorption ability. During the formation of the two precipitates, the colloidal particles are attached and netted onto the precipitate, and subsequently removed by co-precipitation. Therefore, the coagulation mechanism of PFMS for the treatment of actual textile wastewater is co-precipitation netting, which can be realized by improving the coagulation conditions.

Generally, the new coagulant PFMS displays superiority over commercially available coagulants. First, at the same dose of key elements, the coagulation efficiency of PFMS is higher than that of the commercially available coagulants. Second, the optimum dosage range of PFMS is lower than that of the commercially available coagulants, thus the new coagulant has the obvious advantage of low cost in the long run. Finally, the raw material price of magnesium salts in PFMS is much lower than that in conventional coagulants comprising ferric or aluminum salts, and the preparation process is simple.

4. CONCLUSIONS

- Certain parameters affect the coagulation performances of the novel coagulant PFMS. The optimum preparation conditions are as follows: $r = 0.3$, reaction temperature = 55 °C, Fe/Mg molar ratio = 4/1, and aging conditions = 55° C, 2 h.
- The species distribution of different PFMS products characterized by the ferrontimed spectroscopy method shows that PFMS under these conditions has a higher degree of polymerization and exhibits superior coagulation efficiency over other prepared samples in the treatment of simulated dye wastewater.
- XRD and infrared spectrometry reveals that different types of polymerization occur during the preparation of PFMS, which probably generates novel chemical bonds and leads to the synthesis of a composite polymer of iron and magnesium that is based on –OH bonding. These properties are the main reasons for the better coagulation efficiency of PFMS than other commercial coagulants.
- Under the same conditions, the COD and colour removal rates of PFMS for the treatment of actual textile wastewater are generally higher than those of the conventional coagulants. The COD and colour removal rates using PFMS reach 71% and 88%, respectively. The coagulation mechanism of PFMS for the treatment of actual textile wastewater is primarily charge neutralization at low dosages and co-precipitation netting at high dosages. Charge neutralization is primarily determined by

the inherent properties of the PFMS, namely, the species distribution of ferric and magnesium. Co-precipitation netting can be realized by improving the coagulation conditions.

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