Application of polymeric ferric sulfate combined with cross-frequency magnetic field in the printing and dyeing wastewater treatment

Shiguo Gu, Fei Lian, Kejun Yan and Wei Zhang

ABSTRACT

Polymeric ferric sulfate (PFS) was pretreated with a self-made alternating frequency magnetic field for coagulation printing and dyeing (PD) wastewater treatment. The effects of PFS dosage, magnetization intensity, frequency, and time on the removal of chemical oxygen demand (COD), color and turbidity of PD wastewater were investigated. The results indicated that the magnetized PFS significantly improved the removal efficiency in wastewater treatment. When the initial COD, color and turbidity of printing and dyeing wastewater was 464 mg/L, 180 degrees, and 54.8 NTU respectively, the maximum removal rate of COD, color and turbidity was 87.9%, 80.1%, and 95.2% respectively, under the condition of cross frequency magnetic field magnetization PFS. Moreover, the PFS treatment combined with cross-frequency magnetic field could greatly reduce the pollution of iron ions released from iron-based coagulant during wastewater treatment. Characterization of magnetized PFS flocculant by fourier transform infrared spectroscopy, ultraviolet and visible spectrophotometry, and scanning electron microscopy suggested that magnetic crystal with larger size can be formed on the surface of PFS particles.

Key words | alternating frequency magnetic field, characterization, magnetic crystal, polymeric ferric sulfate, wastewater treatment

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INTRODUCTION

With the development of printing and dyeing (PD) industries, PD wastewater has become a serious environmental problem and attracted increasing attention (Liang *et al.* 2018a 2018b). These wastewaters are mainly composed of a large number of contaminants such as polycyclic aromatic hydrocarbon, organic dyes, and heavy metals with relatively high toxicity and poor biodegradability (Domingos *et al.* 2011; Wang *et al.* 2017; Liang *et al.* 2018a 2018b). Moreover, these pollutants frequently combine with other organic or inorganic fractions in wastewater to form large-volume complexes and make conventional wastewater treatments (e.g. coagulation, chemical oxidation, and adsorption processes) less efficient, especially with regards to the color removal (Wu & Wang 2012; Yang *et al.* 2012). Nowadays, a number of emerging strategies have been employed to improve the efficiency of coagulation technology, including combination of filter membrane and aluminum salt (Lee *et al.* 2006), inorganic flocculant, and organic polymer synthesis (Li *et al.* 2017a, 2017b, 2017c). Comparatively, the combination of coagulation and a magnetic field is considered to be one of the most promising methods to enhance coagulation efficiency (Liu *et al.* 2011; Marcin *et al.* 2018).

A magnetic field can greatly affect the physical and chemical properties of both pollutants and coagulants (Liang *et al.* 2018a 2018b; Marcin *et al.* 2018) by transmitting energy and changing their microstructures, and has been widely employed at present in various wastewater treatments (Ozaki *et al.* 2004; Xu *et al.* 2016; Du *et al.* 2017; Li *et al.* 2017a, 2017b, 2017c; Sun *et al.* 2017; Huang *et al.* 2018). Compared with the current pretreatment method, it is simple to use, does not consume any chemicals, has no toxic metal ion residue, and has no ecological toxicity. For example, it was found that the removal of total suspended substances and chemical oxygen demand (COD) increased by 61.1% and 45.9%, respectively, with the combined technique of adsorption and magnetic field for palm oil plant

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wastewater treatment (Mohammed *et al.* 2014). Meanwhile, the removal efficiency of NO^{3-}-N and NH^{4+}-N increased by 19.4% and 36.2%, respectively, after adsorption combined with a magnetic field, compared to the adsorption only method (Zhao *et al.* 2018). Under a magnetic field, the total amount of calcium carbonate precipitates increased and formed in aqueous solution rather than on the tube walls (Alimi *et al.* 2006, 2009). Briefly, a magnetic field can enhance the efficiency of wastewater treatment, compared with conventional methods (Ji *et al.* 2010). Inorganic polymers are among the most widely used coagulants in wastewater treatments, and a majority of them are iron-based polymeric substances (Katsoyiannis *et al.* 2017). As a result, it is likely that iron ions could be released from the iron-based coagulants into the effluent causing color and corrosion problems. To solve the related problems, polymeric ferric sulfate (PFS) has been extensively used in coagulation treatment due to its low price and high flocculation ratio. However, many previous studies found that the concentration of iron ions in effluents cannot be significantly reduced after the PFS treatment (Li *et al.* 2017a, 2017b, 2017c). We hypothesize that the PFS treatment combined with a cross-frequency magnetic field would greatly reduce the 'secondary' pollution (i.e., the release of iron ions) during the wastewater treatment because iron can accelerate corrosion of metal pipelines and water treatment equipment.

Therefore, the main objective of this study was to improve the utilization of iron in PFS, and reduce COD, color, and turbidity of PD wastewater effectively by magnetic field pretreatment. The effects of magnetization intensity, frequency, dosage of PFS, and time on PD wastewater treatment were evaluated. The magnetized PFS was characterized by Fourier transform infrared spectroscopy (FTIR), scanning electron microscope (SEM), and UV-Vis spectrophotometer to examine the surface properties of PFS particles. In addition, the content of iron ions in the effluent and pH were examined.

EXPERIMENTAL

Magnetization device

The self-made alternating current (AC) frequency electromagnetic device, using frequency controlled power (YTN-11010, Qingdao Yitai Instrument Co, Ltd) as an adjustable power supply, was used to connect the electromagnetic coil and make up the adjustable frequency magnetization device (Figure 1). The magnetic field was determined by adjusting the voltage (0–300 V) and frequency (40–499.9 Hz) of the variable frequency stabilized power supply. The intensity of the magnetic field was measured by digital Gauss meter in the experiment.

Wastewater characterization

The experimental wastewater was taken from two effluents of a PD wastewater treatment plant. Wastewater was sampled from the middle part of the container to examine the water quality; COD was 464 mg/L, color was 180 degrees, turbidity was 54.8 NTU, total iron content was 0.6 mg/L, ferrous ion content was 0.2 mg/L, and pH value was 9.9. Based on the water quality of the wastewater treatment plant, COD, color, and turbidity were chosen as the water quality indexes in this study.

Experimental methods

The PFS solution with a proportion of 5% (w/w) was arranged. The PFS solution was magnetized, treated with 6 experimental circular coagulation cups with 1,000 mL PD

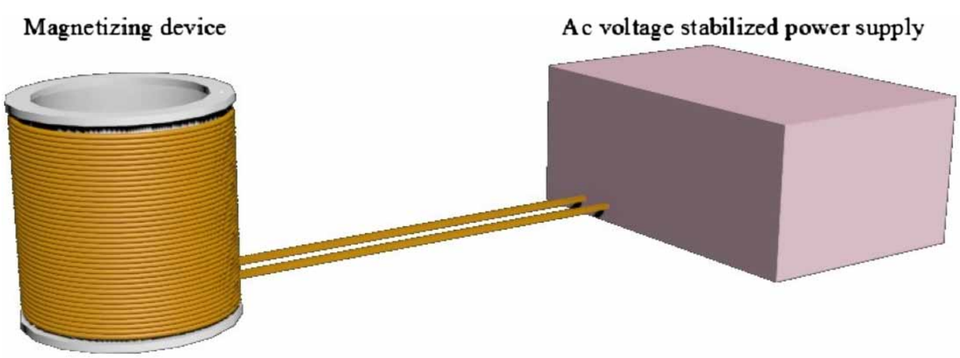

Figure 1 | Homemade frequency magnetization device.

wastewater, respectively. The stirring device was set to a fast stirring intensity of 300 pm for 30 s, then a slow stirring strength of 100 pm for 15 min, and the water sample was allowed to stand for 30 min. An experimental flow chart is shown in Figure 2. The experiment was repeated four times, and the measured data were analyzed with Excel 2013.

Analytical methods

To identify the change of structural components in PFS with or without magnetic field, the morphology of PFS was analyzed by SEM (Quanta-250, FEI, Czech Republic), FTIR (D/max-3c, Japan science corporation) was used to conduct KBr analysis of the structure of PFS in the range of 400–4,000 cm^{-1}, and the PFS density was tracked by UV-vis spectrophotometry (UV-1800, Japan, Shimadzu) at 300–800 nm. The COD value was determined by a potassium dichromate method. Water color was determined using the colorimetric platinum cobalt method, as measured in platinum cobalt units using a photometer (SD9012-A, China). Turbidity was analyzed by a nephelometer (WGZ-200, China) and reported in Nephelometric Turbidity Units (NTU). The pH of the samples was measured by a pH meter (320P-01, USA).

RESULT AND DISCUSSION

Effect of magnetized PFS on quality of wastewater

As described in Figure 3(a), the removal of COD, color, and turbidity of the wastewater treated by magnetized PFS were up to 87.9%, 80.1% and 95.2%, respectively, significantly higher than those without a magnetic field. The average of COD, color, and turbidity indicators in the effluent from the magnetization experiment were reduced by 41.6 mg/L, 33.9 degrees, and 3.6 NTU, respectively, compared to the control. These results indicate that treatment by pre-magnetized PFS solution and re-coagulation can significantly improve the water quality.

The magnetic field cuts the magnetic induction motion of PFS solution back and forth, which causes the energy change in PFS solution (Toledo *et al.* 2008; Cai *et al.* 2009). The energy change is an important contributor to the occurrence of the hydrogen bond (Zaidi *et al.* 2014), which is beneficial for the formation of macromolecules by colloidal molecules and OH groups in wastewater. In addition, the collision between PFS and colloids or suspended particles in wastewater can probably be increased. The PFS solution contains ferromagnetic materials, which are likely to be magnetized and thus increase the proportion of polynuclear complex (Feb) in the hydrolysates of iron salts, while the mass fraction of high polymer (Fec) is significantly reduced (Lei *et al.* 2009). Feb is the most active ingredient in the flocculation process, and Fec is relatively inert in chemical reaction and has a low efficiency in flocculation. The increased Feb is conducive for PFS coagulation performance (Song *et al.* 2006; Harif *et al.* 2012). As a result, more Feb and small molecule particles in wastewater can encounter each other. However, Mohammed *et al.* (2014) found that a high magnetic field (200 mT) effectively reduced the color, total suspended solids (TSS), and COD of palm oil mill effluent. Tao & Zhou (2014) showed that a middle magnetic field of 50 mT can effectively remove total phosphorus and COD. Zieliński *et al.* (2017) also investigated wastewater treatment in an aerobic reactor with activated sludge exposed to a static magnetic field with mean induction of 8.1 mT, where the efficiency of COD removal was about 90%. It is worth noting that only a static magnetic field was used in these studies. We employed an alternating frequency magnetic field combined with PFS to treat PD wastewater in this work, which is more effective to improve water quality and thus needs a lower magnetic field frequency.

With the same amount of PFS, it is shown that the index values in the magnetized experiment effluent are lower than those of the non-magnetized treatment (Figure 3(b)). The average values of COD, color, and turbidity of the effluent

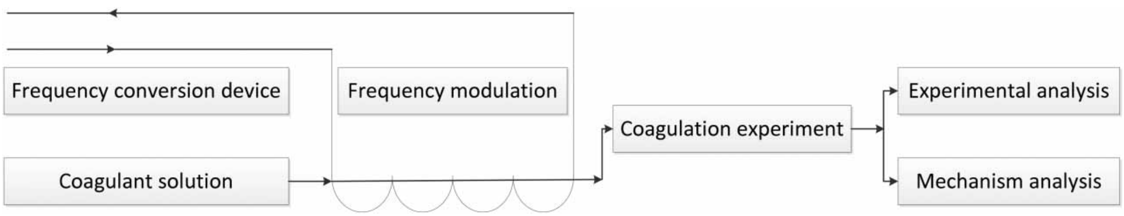

Figure 2 | Cross-frequency magnetic field magnetization and coagulation test flow chart.

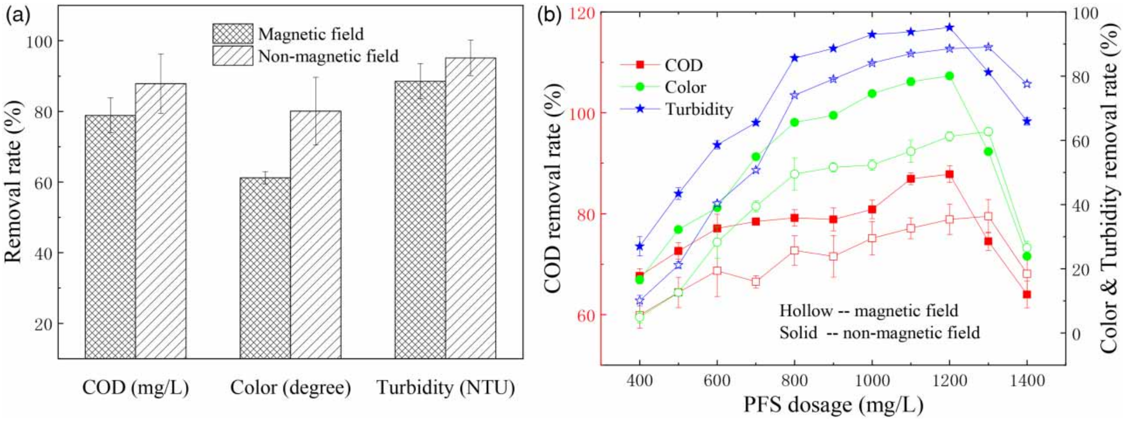

Figure 3 | Effects of frequency of the magnetic field (a) and the amount of PFS on PD wastewater treatment (b).

after magnetization were 27.6 mg/L, 23.5 degrees, and 5.0 NTU, respectively, compared with the non-magnetized effluent. In the magnetization experimental group, the index removal of water samples reached maximum when the dosage of PFS was 1,200 mg/L, and then showed a significant decline. The data indicated that the effluent index is obviously decreased, due to re-stabilization (colloidal particle oversaturation) or charge degeneration with a higher amount of PFS, which is not conducive to the treatment of wastewater (Moussas & Zouboulis 2009; Li *et al.* 2018; Zhou *et al.* 2019). These results indicate that the magnetic field had a positive effect on wastewater treatment.

Effect of magnetization conditions on quality of wastewater

As shown in Figure 4(a), the removal rates of COD, color, and turbidity increased with increasing magnetization intensity at the beginning and then decreased when the magnetization intensity was higher than 12 mT, indicating that an excess high intensity of magnetic field could negatively affect the degradation of organic pollutants in PD wastewater. At relatively high magnetic field intensity, the internal energy of a multi-nucleated iron hydroxyl complex (mainly within Feb) in the PFS solution also increased, resulting in the enhancement of exclusion potential among solution particles, which can increase the diffusion layer thickness and zeta potential of particles (Ofir *et al.* 2007; Harif *et al.* 2012). Thus, the flocculation capacity of the iron hydroxyl complex was improved. The charge neutralization, adsorption bridging, and scavenging effect of the floc could lead to the agglutination of colloids and the formation of polymerization at a higher degree, which lead to the sedimentation of these large agglutinates. Therefore, with increasing magnetic field strength, the iron hydroxyl complex in the PFS solution not only had a relatively high

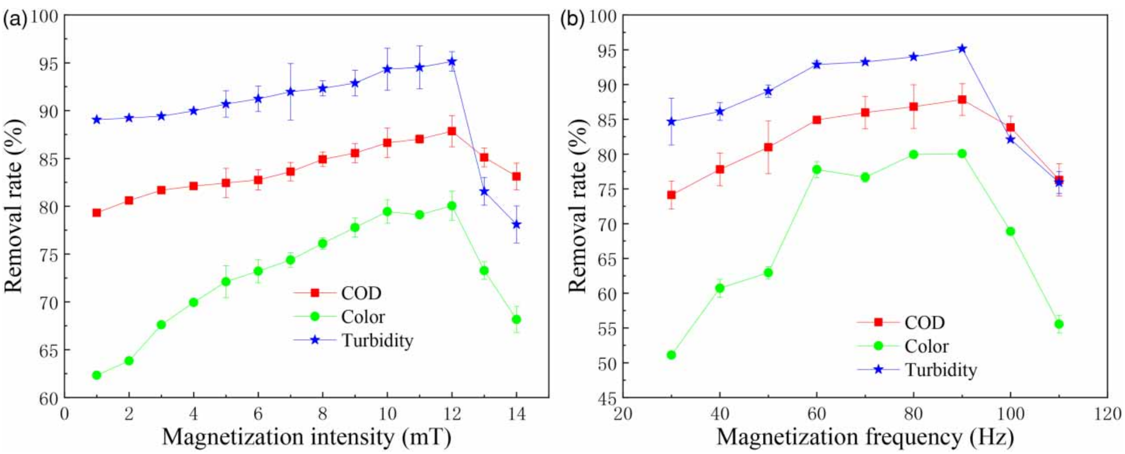

Figure 4 | Effects of magnetization intensity (a) and magnetization frequency on the treatment of wastewater (b).

internal energy and strong exclusion potential among particles, but also enabled a constant high and elevated free state, which makes the PFS more difficult to settle.

The effect of magnetization frequency on the treatment of wastewater was also analyzed, as displayed in Figure 4(b). Similar to the influence of magnetization intensity, the removal of COD, color, and turbidity of wastewater was firstly enhanced with increasing magnetization frequency and then decreased after the frequency was higher than 90 Hz. The increased acting frequency could contribute to the enhancement of both the magnetic induction intensity and kinetic energy of the PFS solution. Thus, the PFS solution is more beneficial to the coagulation of the polynuclear hydroxyl complex, and increases the positive charge of the electrolyte. Furthermore, the coagulant wastewater treatment has negatively charged colloid particles to provide energy. The magnetized PFS solution contains a large number of positively charged particles, which prefer to interact with oppositely charged particles and thus negatively affect the colloid stability. Simultaneously, the probability of collisions between particles increases during their interactions with the frequency of magnetic fields. Finally, a larger floc is formed by condensation and bridging, which includes the aggregation of larger particles by molecular gravitation.

The effect of magnetization time on PD wastewater treatment exhibited a similar trend with magnetization intensity and frequency; that is, the removal rates increased in the beginning stage and then decreased with longer duration (Figure 5(a)). It was indicated that with increasing magnetization time, Lorenz forces could more greatly and constantly destroy the hydrogen bonds among water molecules, resulting in the cracking of large water molecules into numerous small water molecules. The small water molecules consecutively hydrated with Fe^{3+} ions, and meanwhile the proportion of Feb increased, which could increase the coagulation efficiency and more greatly reduce the content of COD, color, and turbidity in the effluent. However, when the magnetization time reached 3 minutes, the hydrates were essentially saturated when approaching its saturation equilibrium point, where the relative content of Feb tended to be stable. With further increasing of magnetization duration, the coagulation effect slowed down and finally almost stagnated. These observations suggested that the pretreatment of PFS with an alternating magnetic field could effectively improve wastewater quality in a relatively short period of time. Similarly, Huang *et al.* (2018) reported that with 30 min pre-magnetization of Fe^0 at 200 mT, the COD removal improved by over 38.4% in the Fe^0-Fenton process. Pan *et al.* (2019) also selected pre-magnetized Fe^0/H_2O_2 using a static and uniform 200 mT magnetic field for 2 min for the degradation and mineralization of antibiotics.

The changes of iron content and pH in the effluent

Figure 5(b) shows that the total iron and ferrous content was 0.4 mg/L and 0.1 mg/L in the effluent, which was 71.7% and 77.3% lower than that of the non-magnetized group, respectively. The results indicated that a magnetic field with alternating frequency could increase the proportion of Feb in the hydrolysate of iron salts. Subsequently, the newly formed active Feb increased the removal efficiency for colloidal impurities through adsorption-bridging and/or sweep-coagulation mechanisms, and hence greatly improved the coagulation effect of PFS and reduced the

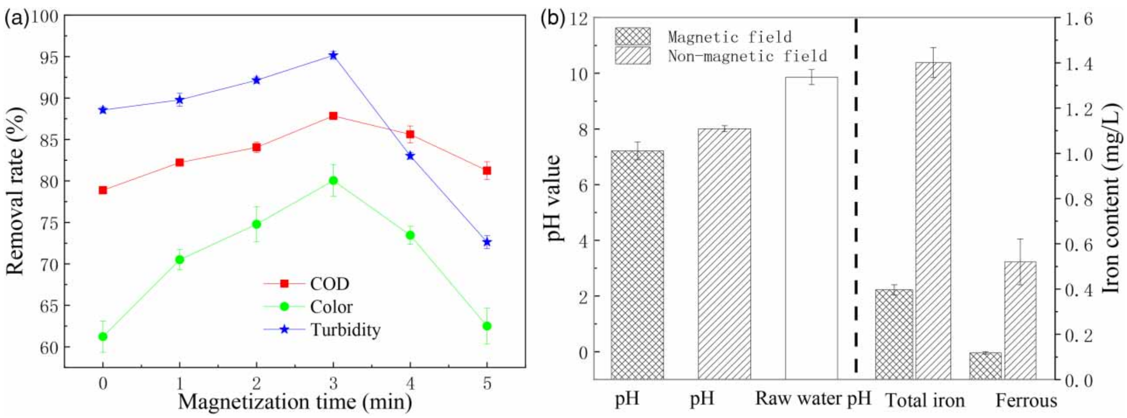

Figure 5 | Effect of magnetization time on the removal of COD, color, and turbidity in the wastewater (a). Alteration of pH in the effluent treated by magnetic field and non-magnetic field (b).

'secondary pollution' of ions to the effluent (Shi *et al.* 2004; Yang *et al.* 2004).

The final pHs in effluent under different treatments were determined as shown in Figure 5(b). It was found that the magnetization pre-treatment could make the wastewater have a tendency to be more neutral. In fact, the results revealed that the pH in effluent treated by magnetization was lower than that of the non-magnetized group (where the pH was alkaline). For the hydrolysis of PFS, a large number of $[Fe_4(H_2O)_6]$, $[Fe_2(H_2O)_6]$, $[Fe(OH)_2]$ and other polynuclear complexes were produced. Moreover, the PFS solution increased the proportion of polynuclear polymer (Feb is the most active component in the flocculation process) in the hydrolyzed products under the influence of an alternating magnetic field (Lei *et al.* 2009). During the coagulation process, PFS (mainly Feb) and colloidal particles in wastewater coagulated to form precipitation through adsorption, bridging, and cross-linking. Moreover, the influence of the alternating magnetic field is advantageous to the precipitation of Fe^{2+} and Fe^{3+}, and the enhancement of H^+ concentration. The reaction mode is as follows:

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3(\text{colloid}) + 3H^+ \quad (1)$$

$$Fe^{2+} + 2H_2O \rightarrow Fe(OH)_2 + 3H^+ \quad (2)$$

Characterization of magnetized and non-magnetized PFS

The chemical structure of magnetized and non-magnetized PFS was analyzed with FTIR and UV-vis, respectively, as displayed in Figure 6. The peak at 850–880 cm^{-1} is the bending vibration absorption of Fe-OH-Fe, The surface and global Fe-OH vibration absorption was approximately at 1,000 and 650 cm^{-1}, respectively. The results showed the existence of the polymerization structure, iron, and hydroxyl groups (Fu *et al.* 2007; Ma *et al.* 2018). The metal coordinated SO_4^{2-} has the characteristic absorption peak in the range of 900–1,250 cm^{-1}. Peaks around 2,360 and 2,150 cm^{-1} could be the absorption of HSO_4^- and that of 1,230 cm^{-1} is the symmetric expansion vibration of SO_4^{2-}. In addition, 1,140 and 1,130 cm^{-1} are the characteristic frequencies of SO_4^{2-} (Jiang *et al.* 2004; Fu *et al.* 2007; Ma *et al.* 2018). The FTIR results suggest that polyhydroxy sulfate might be formed on the surface of PFS, and the UV-vis results showed that the magnetized PFS has a larger aggregation state, indicating that larger aggregates can be formed on the surface of magnetized PFS. Hence, it is concluded that the 'magnetic crystallization effect' can be derived from the larger crystals of PFS particles.

SEM was employed to analyze the morphology and structure of magnetized and non-magnetized PFS, respectively, as shown in Figure 7. In the magnetization experiment, the magnetic field affects the crystallization kinetics of small molecules (Kimura 2003) and alters the nucleation rate and crystal size of small molecules, thus the 'magnetic crystallization effect' emerges. The magnetized PFS crystal particles resembles polyphosphate ferric sulfate (PPFS) as was apparent during the formation of new hydroxyl bond compounds (Wang *et al.* 2016). As shown in Figure 7, under high magnification, the particles of magnetized and non-magnetized PFS are amorphous. The predominant difference between the two types of PFS was that the size of magnetized PFS crystals (106.6 ± 15.6 um) was significantly larger than the non-magnetized PFS

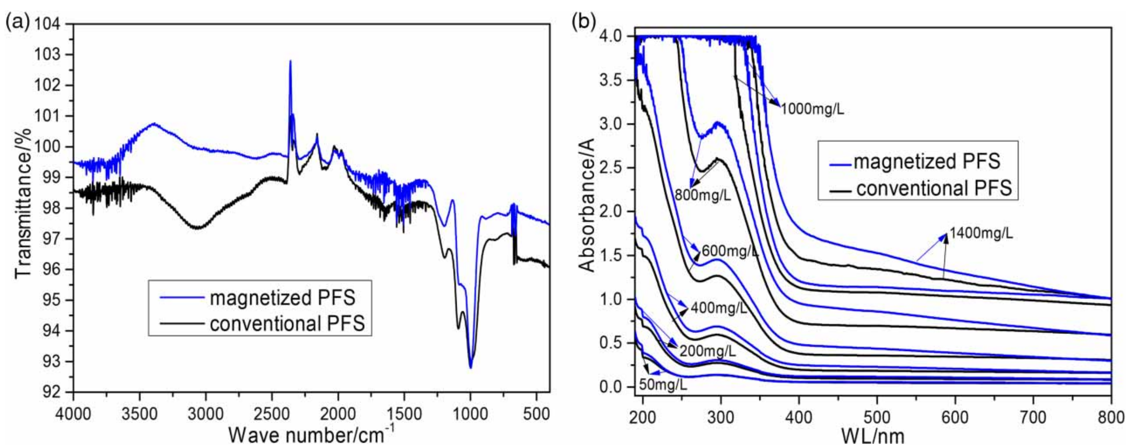

Figure 6 | FTIR (a) and UV-vis (b) diagram of magnetized and non-magnetized PFS.

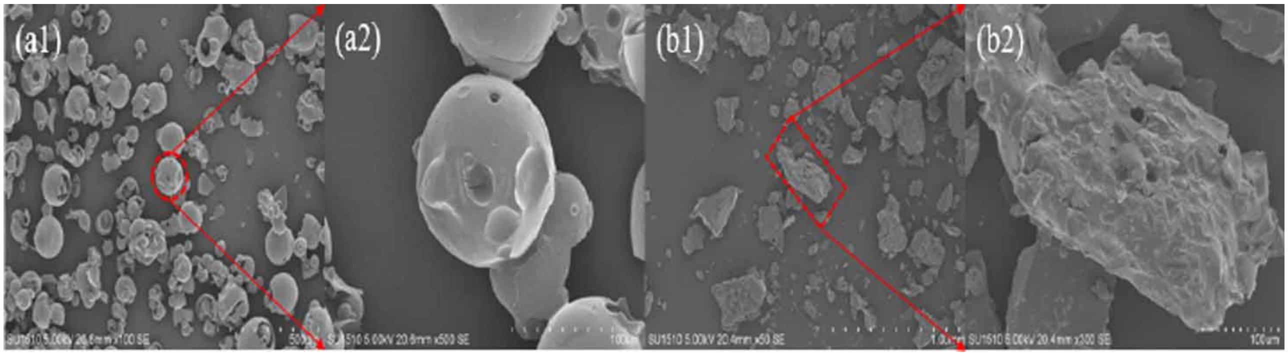

Figure 7 | The SEM images of non-magnetized (a1–a2) and magnetized PFS (b1–b2).

crystals (506.5 ± 36.2 um). The PFS flocculant exhibited a reticular and flaky structure. This special network structure enables PFS to have unique characteristics including a large surface area and strong adsorptive force. In addition, the development of the flake structure makes the flocculation ability of PFS more effective.

CONCLUSIONS

PFS treatment combined with a cross-frequency magnetic field (intensity12 mT, frequency 90 Hz, and time 3 min) could significantly increase the removal efficiency for wastewater treatment. At this time, the removal efficiency of COD, color, and turbidity of the effluent was obviously superior to that of the treatment without magnetization. The COD, color, and turbidity of the effluent were reduced by 41.6 mg/L, 33.9 degrees, and 3.6 NTU respectively, in comparison to the non-magnetized PFS. Moreover, the total iron, ferrous iron content, and pH of the effluent were lower than the PFS components in the unmagnetized treatment.

The results of SEM, FTIR, and UV revealed that active substance of polyhydroxy sulfate can be formed on the magnetized PFS surface, which could produce a magnetic crystallization effect and contribute to enlargement of the crystal size of PFS particles. Henceforth, PFS is beneficial to the coagulation performance with the assistance of an alternating magnetic field.

The results of this study allow the conclusion that the combination of alternating frequency magnetic field and coagulation process has a vital role in treating PD wastewater. The magnetic effect greatly increases the removal efficiency of COD, color, and turbidity. However, the related mechanisms are largely unknown and need more research work in the future.

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