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Flocculation performance evaluation and flocculation mechanism study of PAC/PMAPTAC composite flocculant in dyeing wastewater

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ABSTRACT

In this paper, the inorganic–organic composite flocculant (PAC/PMAPTAC) was prepared with polyaluminum chloride (PAC) and the self-made PMAPTAC. Then, the flocculation sedimentation experiment was used to evaluate the flocculation performance of single flocculant PAC, PMAPTAC and the composite flocculant PAC/PMAPTAC in dyeing wastewater. At the same time, the flocculation effect of PAC/PDMDAAC was compared with that of PAC/PMAPTAC under the same condition. Finally, the flocculation mechanism was analyzed by zeta potential and floc morphology. The results showed that compared with PAC, the flocculation performance of composite flocculant PAC/PMAPTAC could be markedly improved, resulting in the higher decolorization rate and COD_{Mn} removal rate. Besides, the increase of (η) or content of PMAPTAC in PAC/PMAPTAC could enhance the decolorization rate and COD_{Mn} removal rate simultaneously. Furthermore, the lower dosage was required to achieve the best removal effect, which meant that the cost of the flocculant was reduced. Moreover, the comparison of flocculation performance turned out that the decolorization rate and COD_{Mn} removal rate of PAC/PMAPTAC were higher than those of PAC/PDMDAAC under the same condition. Finally, the zeta potential and floc morphology analysis showed that the flocculation mechanism of the reactive dye simulated wastewater was mainly charge neutralization and adsorption bridging. The above results not only fill the gaps in the research of PAC/PMAPTAC during the treatment of dyeing wastewater, but also provide an experimental basis for its application in dyeing wastewater and expand the application fields of PMAPTAC.

Keywords: PMAPTAC; Polyaluminum chloride; Composite flocculant; Dyeing wastewater; Flocculation mechanism

1. Introduction

Approximately 7 × 10^5 t synthetic dyes are produced around the world each year [1], which cause serious environmental pollution due to their toxicity, mutagenicity and carcinogenicity [2,3]. There are many kinds of dyes, among which reactive dyes are the fastest growing varieties in recent years, accounting for about 30% of the total dye production. Due to the good water solubility of reactive dyes, it is considered to be a kind of printing wastewater which is difficult to handle. The treatment of printing and dyeing wastewater is extremely urgent [4]. Currently, techniques such as adsorption, biodegradation, membrane separation and coagulation/flocculation are commonly used in dye wastewater treatment [5–7]. Among which, the coagulation/flocculation is a commonly used

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process in water and wastewater treatment due to its high efficiency and low cost, and it has been proved effective in dye removal, especially dissolvable dyes in wastewater [8]. In coagulation/flocculation process, coagulants play an important role in target pollutant removal. So far, various coagulant categories have been commercially available including inorganic coagulants, synthetic polymer flocculants and inorganic–organic dual-coagulants [9]. However, flocs produced by inorganic coagulants are tiny and difficult to settle [10], generating high concentration of residual reagent as well as vast amounts of sludge after sedimentation [11]. The amount of sludge can be reduced when using synthetic polymer flocculants and the coagulation performance could be improved by dosing organic flocculants due to the enhanced charge neutralization and adsorption bridging [12–14].

As the organic flocculants, polyquaternary ammonium salt polymers have been widely used in dyeing wastewater treatment [13,15]. Among which, the most widely used cationic monomers are dimethyl diallyl ammonium chloride (DMDAAC), acryloyloxyethyltrimethyl ammonium chloride (DAC) and methacryloxyethyltrimethyl ammonium chloride (DMC). For example, DMDAAC has been widely used in printing and dyeing wastewater because of its advantages of stable structure and wide application range [16–18]. Bian and Gao [19] prepared the inorganic–organic composite coagulant PAC/PDMDAAC with polyaluminum chloride (PAC) and PDMDAAC and one kind of dyeing wastewater from a printing and dyeing factory was treated with PAC/PDMDAAC through coagulation. The results showed that when the dosage of PAC/PDMDAAC was 500~750 mg/L, the decolorization rate and the COD_{Mn} removal rate were 50.4% and 67.4%, respectively. When the dosage exceeded 375 mg/L, the decolorization effect of PAC/PDMDAAC was better than that of PAC and slightly lower than that of PDMDAAC. This study reported that the printing and dyeing wastewater was treated with composite coagulant PAC/PDMDAAC, but its decolorization rate and COD removal rate were both low. Besides, due to the steric hindrance of DMDAAC molecular structure, it is not easy to obtain the PDMDAAC products with higher molecular weights [20], which limits the application of this type of cationic polymer to a certain extent.

The polymers based on DMC and/or DAC have also been widely used in printing and dyeing wastewater [21–24]. Compared with DMDAAC monomer, it is easy to obtain the polymers based on for DAC and DMC monomer with high molecular weights [25,26]. However, because the pendant side chain structure of its monomer molecules contains ester functional groups, the polymer products based on DMC and DAC are easily hydrolyzed when stored and used in the conventional aqueous solutions, resulting in that the polymers cannot fully exert their application effectiveness in certain specific application fields.

Methacryloyl aminopropyl trimethyl ammonium chloride (MAPTAC), with molecular structure similar to those of DAC and DMC, is one kind of straight-chain cationic monomer and also exhibits high polymerization activity. Therefore, the MAPTAC products possess the higher molecular weights [27] when compared with DMDAAC polymers, and it is less affected by pH in the application environment [28,29] when compared with DMC or DAC polymers. Based on these advantages, MAPTAC polymers have been widely used in the fields of medicine [30], papermaking [31], sewage treatment [32], daily chemicals [33] and petroleum extraction [34]. In most literature, the dyeing wastewater were usually treated with the adsorbents, which were usually prepared by combining the monomer MAPTAC with other monomers or inorganic materials [35,36]. However, so far, the dyeing wastewater treated with PMAPTAC, especially the inorganic–organic composite coagulant (PAC/PMAPTAC) has not been reported in the literature.

In this work, the inorganic–organic composite flocculant was prepared with PAC and the self-made PMAPTAC with different molecular weights. Then, the flocculation sedimentation experiment was conducted to evaluate the flocculation performance of single flocculant PAC, PMAPTAC and the composite flocculant PAC/PMAPTAC in dyeing wastewater. At the same time, the flocculation effect of PAC/PDMDAAC was compared with that of PAC/PMAPTAC under the same condition. Finally, the flocculation mechanism of PAC/PMAPTAC composite flocculant was investigated through the analysis of zeta potential and the size of the floccules. The above results will not only fill the gaps in the research of PAC/PMAPTAC composite flocculant during the treatment of dyeing wastewater, but also provide an experimental basis for its application in dyeing wastewater and expand the application fields of PMAPTAC.

2. Materials and methods

2.1. Materials

In this study, reactive red 3BF was provided by Nantong Shuguang Printing and Dyeing Co., Ltd., China. The PMAPTAC samples with different (η) (used to represent molecular weight) and PDMDAAC were prepared in our lab [20,27]. The (η) of PMAPTAC were 0.75, 1.96 and 2.64 dL/g, and the (η) of PDMDAAC was 2.6 dL/g, which were determined at 30°C ± 0.1°C after dissolved in 1 M NaCl by using an Ubbelohde viscometer [3]. The structures of PMAPTAC and PDMDAAC are shown in Fig. 1.

2.2. Preparation of simulative water and flocculant

Reactive red 3BF was chosen as the representation of reactive dyes. The simulative dyeing solution with the concentration of 50 mg/L was prepared by dosing 0.05 g of the dye power into 1 L of tap water and stirring until dissolved actually.

Polyaluminum chloride (PAC) with Al_2O_3 content (w/w) of 30% was chosen as the coagulant. The concentration of the stock solution was set at 100 g/L. The dosage of PAC was calculated by the content of Al_2O_3. The concentration of PAC used in this work was 10 g/L. The concentration of PMAPTAC solutions used in this work was 1 g/L.

The composite coagulation PAC/PMAPTAC or PAC/PDMDAAC was prepared by adding 40 g of 30% PAC particles into 120 mL tap water and stirred until it's all dissolved. Then certain amount of PMAPTAC or PDMDAAC was added into the 100 g solution of PAC (containing 10 g of Al_2O_3) according to the mass ratio of 5:1, 10:1 and 20:1.

2.3. Flocculation performance

Based on the previous work in our group [37], the procedure of experiment was as follows: 100 mL of 50 mg/L reactive red 3BF dye simulated wastewater were added in 250 mL beakers, then different dosages of flocculant were added, and the flocculation test was carried out on a program-controlled jar test apparatus (TA2-2, Wuhan Hengling Technology Co., Ltd.) at room temperature. The mixture was rapidly stirred at 200 rpm for 25 s and then slowly at 60 rpm for 25 s for the flocs to grow. After 30 min of sedimentation, the supernatant after the treatment at a water depth of 1 cm under the surface was collected for measurement.

The absorbance was measured at the maximum absorption wavelength of the dye solution by a visible spectrophotometer, and the decolorization rate and the COD_{Mn} removal rate were measured and calculated according to the equations:

$$\text{Decolorization rate }\% = \left(\frac{1-A}{A_0}\right)\times 100\% \quad (1)$$

Among them, A_0 is the absorbance of the simulated dyeing wastewater before treatment; A is the absorbance of the supernatant after the flocculation treatment.

$$COD_{Mn}\text{removal rate }\% = \left(\frac{1-B}{B_0}\right]\times 100\% \quad (2)$$

Among them, B_0 is the COD_{Mn} content of the simulated dyeing wastewater before treatment; B is the COD_{Mn} content of the supernatant after the flocculation treatment.

The zeta potential of the flocs in the supernatant was determined according to JS94H micro electrophoresis instrument's manual [37], and the floccules were photographed through a microscope after the precipitation was completed.

2.4. Methods

Firstly, the effect on the decolorization rate and COD_{Mn} removal rate would been studied with PAC or PMAPTAC with different (η) alone. Secondly, the effect of different ratios and (η) of PAC/PMAPTAC on the decolorization rate and COD_{Mn} removal rate in dyeing wastewater was studied. At the same time, the flocculation effect of PAC/PDMDAAC was compared with that of PAC/PMAPTAC under the same condition. Finally, the flocculation mechanism of PAC/PMAPTAC composite flocculant was investigated through the analysis of zeta potential and floc morphology.

3. Results and discussion

3.1. Flocculation performance of PAC or PMAPTAC

The effect on the decolorization rate and COD_{Mn} removal rate were investigated when the PAC or PMAPTAC with different (η) was used alone. The results are shown in Figs. 2 and 3.

It could be seen from Figs. 2 and 3 that the decolorization rate and COD_{Mn} removal rate increased rapidly at the beginning, and then decreased with the increase of dosage. As we all know, both PAC and PMAPTAC had positive charges, while the dyes were negatively charged. Therefore, when added to the activated red simulated dye wastewater, these positive species benefited flocs growing due to charge neutralization causing destabilization of colloids and facilitate the physical or chemical adsorption of the destabilized colloids. As a result, both the decolorization rate and the COD_{Mn} removal rate increased with the increase of the dosage due to the mutual attraction of charges. However, when the dosage exceeded a certain amount, charge repulsion would trigger a slight backmixing phenomenon, resulting in a decrease of the decolorization rate and COD_{Mn} removal rate.

From Figs. 2 and 3 we can also know that, when the doses was 15 mg/L for PAC, while 25 mg/L for PMAPTAC with different (η), the highest decolorization rate and COD_{Mn} removal rate were 90.42%, 68.98%, 70.02%, 81.20% and 92.76%, 54.29%, 56.00%, 65.42% for PAC, PMAPTAC with 0.75, 1.96 and 2.64 dL/g, respectively. It was evident that compared with PAC, PMAPTAC exerted poor decolorization rate and COD_{Mn} removal rate when added alone. Besides, for PMAPTAC, the decolorization rate and COD_{Mn} removal rate increased slightly with the increase of (η). This may be because the larger the molecular weight of PMAPTAC, the longer the molecular chain, and the greater the probability of colliding with dye molecules to form flocs [32,38].

3.2. Flocculation performance of PAC/PMAPTAC

3.2.1. Effect of (η) on the decolorization rate and COD_{Mn} removal rate

According to section 2.4, the effect of (η) on the decolorization rate and COD_{Mn} removal rate of the dyeing wastewater were investigated when the PAC/PMAPTAC ratio was 10:1, and the (η) were 0.75, 1.96 and 2.64 dL/g. The results are shown in Figs. 4 and 5.

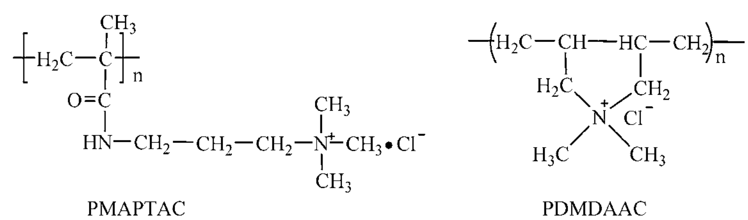

Fig. 1. Structures of PMAPTAC and PDMDAAC.

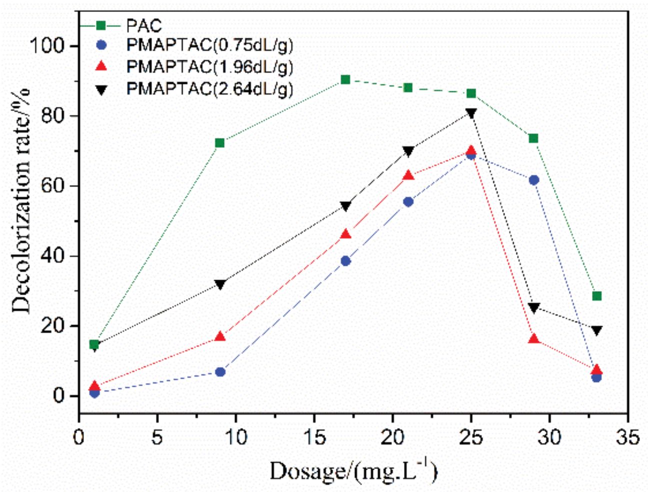

Fig. 2. Effect of dosage on the decolorization rate.

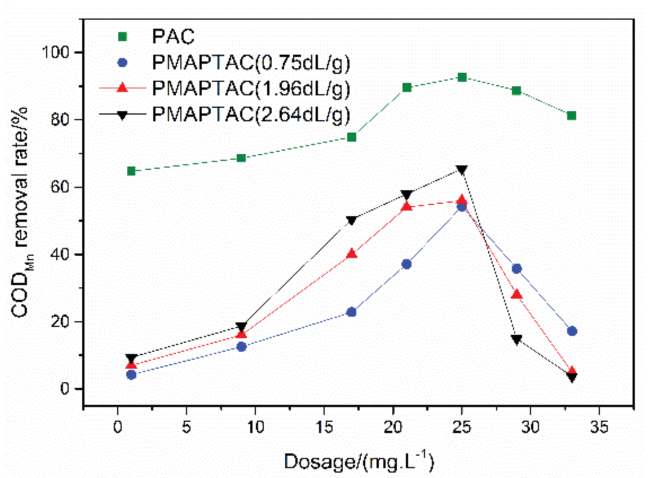

Fig. 3. Effect of dosage on the COD_{Mn} removal rate.

Figs. 4 and 5 show that the decolorization rate and COD_{Mn} removal rate increased rapidly with the increase of dosage at the beginning, then they entered a plateau state in succession, and finally showed a decreasing trend. The composite flocculant with 2.64 dL/g promoted the attainment of the maximum decolorization rate and COD_{Mn} removal rate (99.75% and 94.09%) when the dosage was 13 mg/L. In contrast, the composite flocculant with 0.75 dL/g displayed notable inferior decolorization and COD_{Mn} removal rates (91.85% and 90.83%), indicating that composite flocculants with high (η) were conducive to the removal of the dye particles and COD_{Mn} in solution. Besides, the decolorization rate achieved by PAC/PMAPTAC was significantly superior in comparison with that in PAC coagulation. In addition, the decolorization rate in PAC/PMAPTAC systems were found to attain equilibrium more quickly as compared with that in PAC systems. This phenomenon was particularly obvious in the low dosage range. This may be because PMAPTAC was a cationic polymer with high charge density, which was beneficial in enhancing the neutralization ability of the inorganic flocculant after compounded. And the compounding process may change the morphology and structure of the aggregates in the inorganic flocculant, thereby improving the interaction between the flocculant and the dye molecules. In addition, PMAPTAC itself could also exerted the better bridging performance. The longer the molecular chain, the better the bridging performance, thereby improving the decolorization rate and COD_{Mn} removal rate.

Last but not least, excess dosages of coagulants caused resuspension of aggregated particles, resulting in reduction of coagulation efficiency, especially when treated with the composite flocculant with high (η). This may be related to the change of the potential in solution, which would be confirmed in subsequent experiments.

3.2.2. Effect of ratio on the decolorization rate and COD_{Mn} removal rate

According to section 2.4, the experiment was performed to investigate the effect of ratio of PAC/PMAPTAC on the decolorization rate and COD_{Mn} removal rate of dyeing wastewater when (η) was 0.75 dL/g. The results are shown in Figs. 6 and 7.

As shown in Figs. 6 and 7, the decolorization rate and COD_{Mn} removal rate increased rapidly with the increase of dosage at the beginning, and then showed a decreasing trend. The reason may be that for the composite flocculant, the flocculation effect was mainly charge neutralization and adsorption bridging, and the destabilization and restoration of the particles were due to the charge change with the increase of the dosage. The total force between the particles changed from attraction to mutual repulsion due to the reversal of the charge. Therefore, as the dosage increased, the decolorization rate decreased instead and the COD_{Mn} removal rate also exhibited a similar trend.

It can also be seen from Figs. 6 and 7 that the lower the ratio, that is, the higher the PMAPTAC content, the better the decolorization rate and COD_{Mn} removal rate. Additionally, the lower dosage of flocculant were required to make the dye particles in solution reach the agglomerated state, indicating that the cost of dyeing wastewater treatment was saved. The higher the PMAPTAC content in the composite flocculant, the more obvious this effect. Therefore, the increase of PMAPTAC concentration would significantly enhance the charge neutralization ability of the composite coagulant.

3.3. Comparison with the flocculation performance of PAC/PDMDAAC

According to the section 2.4, the flocculation performance of PAC/PDMDAAC was investigated under the condition of the ratio 10:1 and 2.6 dL/g. The results are shown in Fig. 8.

As shown in Fig. 8, the decolorization rate and COD_{Mn} removal rate first increased and then gradually decreased with the increase of dosage. When the dosage of PAC/PDMDAAC composite flocculant was 14 mg/L, the highest decolorization rate and COD_{Mn} removal rate were 92.16% and 72.73%, respectively. In contrast, as shown in Figs. 2 and 3, under the same condition, the highest decolorization

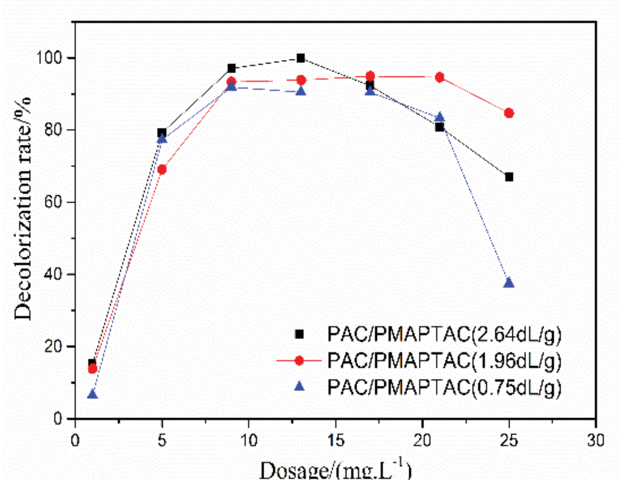

Fig. 4. Effect of (η) on the decolorization rate.

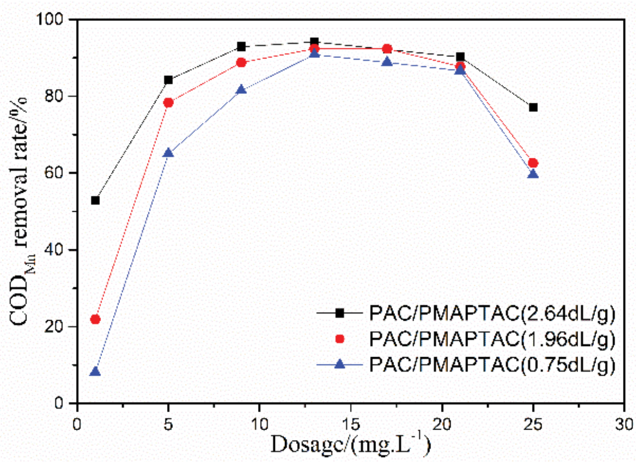

Fig. 5. Effect of (η) on the COD_{Mn} removal rate.

rate and COD_{Mn} removal rate of PAC/PMAPTAC were 99.75% and 94.09%, respectively. It could be concluded that PAC/PMAPTAC showed more excellent color and COD_{Mn} removal efficiency. The reasons may be that under the condition of the same intrinsic viscosity, the charge densities per mole of PDMDAAC and PMAPTAC molecular were basically the same, which meant that they have similar charge neutralization effects when adsorbing negatively charged dyes. Therefore, the difference of flocculation performance between them might mainly come from the adsorption bridging effect. By the analysis of their molecular structure shown in Fig. 1, we can know that PDMDAAC side chain had a five-membered ring, while PMAPTAC had a longer linear side chain containing the amide bond. On the one hand, it might be because PMAPTAC had a longer side chain, which made the adsorption bridging and netting sweeping effect stronger. On the other hand, it was also possible to remove the dye particles through the formation of hydrogen bonds between the dye particles and the amide bond of the PMAPTAC molecule. This two aspects resulted in different adsorption energy

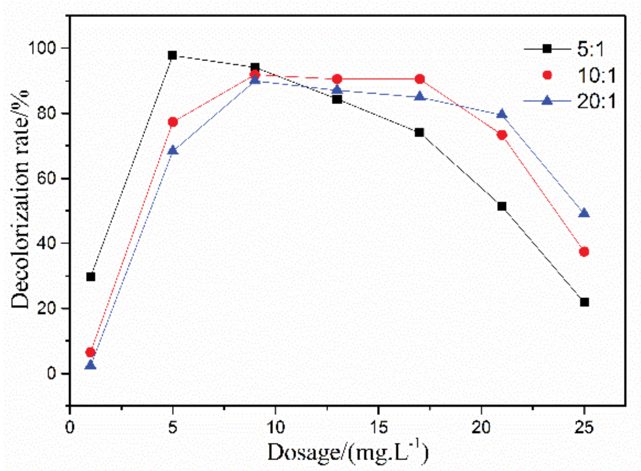

Fig. 6. Effect of the ratio on decolorization rate.

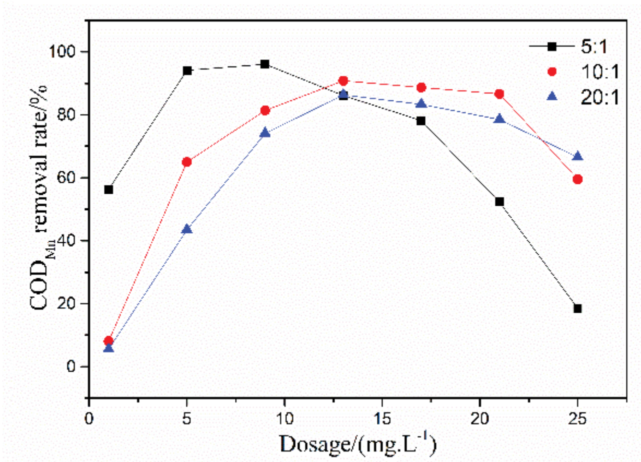

Fig. 7. Effect of the ratio on COD_{Mn} removal rate.

and adsorption configuration when adsorbing dyes, making the adsorption capacity of PMAPTAC higher than that of PDMDAAC under the same condition [39].

3.4. Flocculation mechanism

3.4.1. Zeta potential

Zeta potential is a good indicator of the magnitude of the repulsive interaction between colloidal particles [32]. According to the section 2.4, the zeta potential of the supernatant were determined after the dying wastewater was treated with PAC/PMAPTAC under the condition of ratio 10:1. The results are shown in Fig. 9.

It can be seen from Fig. 9 that the zeta potential increased with the increase of dosage, The zeta potential of the supernatant changed from negative to positive as the dosage increased, indicating that the compressed electric double layer and the adsorption charge neutralization reaction occurred during the flocculation process. Combined with the decolorization rate data in Fig. 4, it can be found that when the zeta potential was less than 0 mV, the decolorization rate gradually increased with increasing dosage, and the decolorization rate reached a maximum at around 0 mV; when the zeta potential was more than 0 mV, the decolorization rate gradually decreased as the dosage increased. This was because when the zeta potential was less than 0 mV, the flocs could completely settle. After the zeta potential was more than 0 mV, the charges in the particles in solution changed from negative to positive and repelled each other, making it difficult to aggregate to form larger flocs, and cannot be completely settling, resulting in a decrease of decolorization rate.

In addition, it was not difficult to find that under the same dosage, the zeta potential of particles treated by PAC/PMAPTAC composite flocculants was significantly higher than that of PAC, which meant that the charge neutralization ability of PAC/PMAPTAC was largely higher than that of PAC. At the same dosage, the higher (η) of PMAPTAC in PAC/PMAPTAC, the higher zeta potential in supernatant was, indicating that the increase of (η) of PMAPTAC in PAC/PMAPTAC could enhance the charge neutralization ability.

3.4.2. The morphology characteristics of flocs

According to the section 2.4, in order to study the adsorption bridging ability of PMAPTAC with different (η), the dying wastewater was treated with PAC/PMAPTAC under the condition of 10:1. The flocs were photographed through electron microscope under the condition of same dosage. The floc morphology are shown in Fig. 10.

It could be seen from Fig. 10 that the floc size of the dyeing wastewater were about 0.1, 0.2 and 0.4 mm after treated with composite coagulant PAC/PMAPTAC ((η) were 0.75, 1.96 and 2.64 dL/g). This was consistent with the results of the decolorization rate. This was because PMAPTAC with high (η) had a relatively high molecular weight and many active sites for adsorbing dye particles. This not only enhanced the charge neutralization effect, leading to the more dye molecules that can be adsorbed, but also provided a strong adsorption bridging between particles, and ultimately formed larger flocs. Therefore, the higher decolorization rate and COD_{Mn} removal rate and larger diameter floccules could been obtained by adding the PMAPTAC with high (η) into composite flocculant.

3.4.3. Flocculation mechanism analysis of dyeing wastewater

The flocculation mechanism of PAC/PMAPTAC composite flocculant is shown in Fig. 11. Lots of "active" sites were provided after adding PMAPTAC in PAC solution, which improved the collision efficiencies in the dye removal. The results of the flocculation experiment show

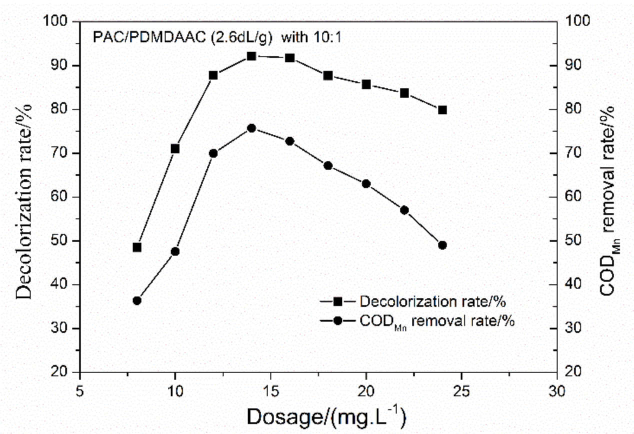

Fig. 8. Effect of dosage on the decolorization rate and COD_{Mn} removal rate.

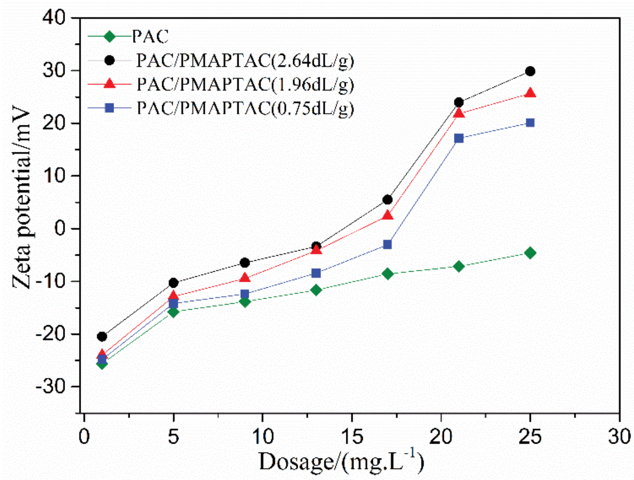

Fig. 9. Effect of dosages on the zeta potential.

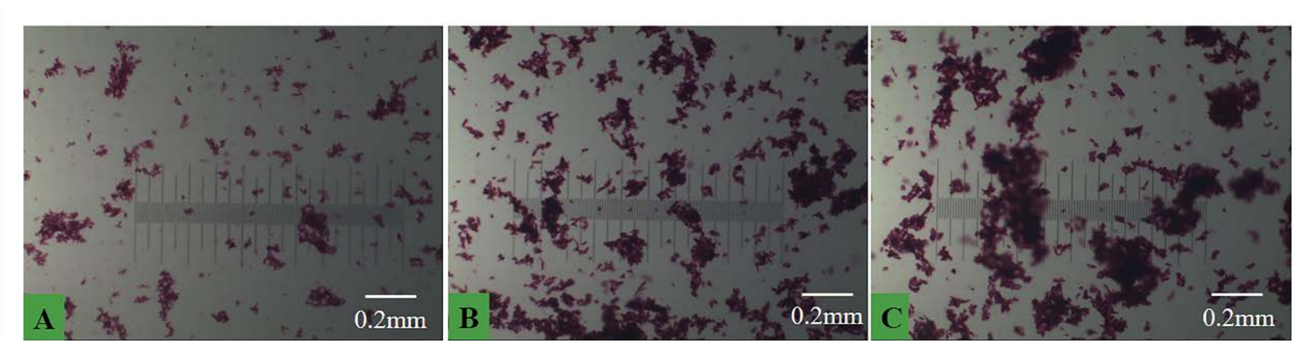

Fig. 10. Floc morphology (A) (η) = 0.75 dL/g, (B) (η) = 1.96 dL/g and (C) (η) = 2.64 dL/g.

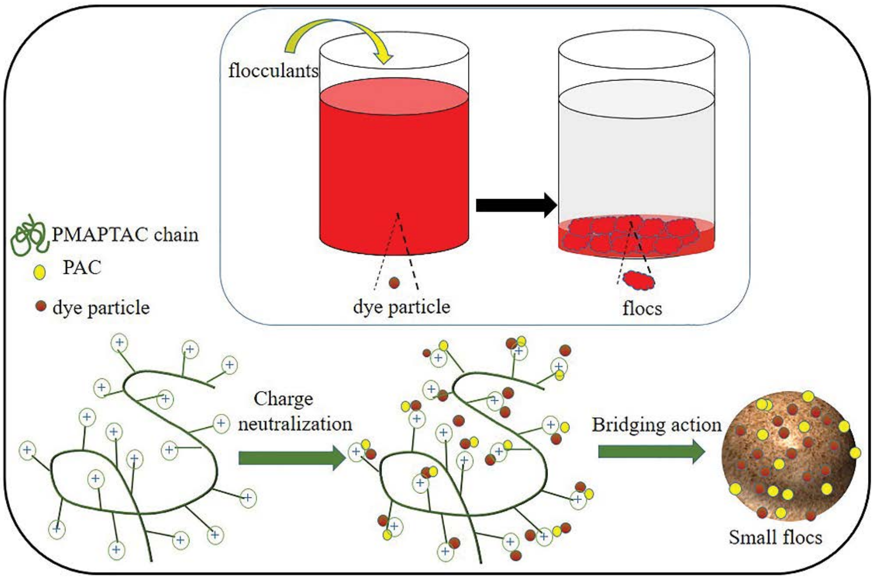

Fig. 11. Flocculation mechanism of composites flocculants for dyeing wastewater.

that the PMAPTAC with high molecular weight showed an excellent color removal effect than that of PMAPTAC with low molecular weight under the same condition. This was because the high molecular weight played a vital role in the aggregation process, not only enhancing the charge neutralization, but also providing a strong adsorption bridging between the particles, which could also be observed from the zeta results and floc morphology.

4. Conclusions

The inorganic–organic composite flocculants were prepared by compounding PAC and the self-made PMAPTAC with different (η). Then, the flocculation effect of the composite coagulant PAC/PMAPTAC in dyeing wastewater was studied and compared with that of PAC/PDMDAAC under the same condition. Finally, the flocculation mechanism was analyzed by zeta potential and floc morphology. The results showed that firstly, compared with PAC flocculant, the flocculation performance of composite flocculants could be markedly improved by adding PMAPTAC, resulting in higher color and COD_{Mn} elimination efficiencies. Secondly, the increase of molecular weight or content of PMAPTAC in PAC/PMAPTAC could enhance the decolorization rate and COD_{Mn} removal rate simultaneously. What is more, the lower dosage was required to achieve the best removal effect, which meant that the cost of the flocculant was reduced. Besides, the flocculation performance experiments turned out that the decolorization rate and COD_{Mn} removal rate of PAC/PMAPTAC composite flocculant was higher than that of PAC/PDMDAAC. Finally, the zeta potential and floc morphology analysis showed that the flocculation and decolorization mechanism of the reactive dye simulated wastewater was mainly charge neutralization and adsorption bridging.

Therefore, the composite coagulant PAC/PMAPTAC is a potential alternative in treatment of dye wastewater.

Abbreviations

MAPTAC — Methacryloyl aminopropyl trimethyl ammonium chloride
DMDAAC — Dimethyl diallyl ammonium chloride
A^0 — Absorbance of the simulated dyeing wastewater before treatment
A — Absorbance of the supernatant after the flocculation treatment
B_0 — COD_{Mn} content of the simulated dyeing wastewater before treatment
B — COD_{Mn} content of the supernatant after the flocculation treatment

References

[1] W. Song, B. Gao, X. Xu, L. Xing, S. Han, P. Duan, W. Song, R. Jia, Adsorption–desorption behavior of magnetic amine/Fe_3O_4 functionalized biopolymer resin towards anionic dyes from wastewater, Bioresour. Technol., 210 (2016) 123–130.

[2] J. Krishnan, A. Arvind Kishore, A. Suresh, B. Madhumeetha, D. Gnana Prakash, Effect of pH, inoculum dose and initial dye concentration on the removal of azo dye mixture under aerobic conditions, Int. Biodeterior. Biodegrad., 119 (2017) 16–27.

[3] T. Yao, S. Guo, C. Zeng, C. Wang, L. Zhang, Investigation on efficient adsorption of cationic dyes on porous magnetic polyacrylamide microspheres, J. Hazard. Mater., 292 (2015) 90–97.

[4] G. Dogdu, A. Yalcuk, S. Postalcioglu, Application of the removal of pollutants from textile industry wastewater in constructed wetlands using fuzzy logic, Environ. Technol., 38 (2017) 443–455.

[5] R.L. Singh, P.K. Singh, R.P. Singh, Enzymatic decolorization and degradation of azo dyes – a review, Int. Biodeterior. Biodegrad., 104 (2015) 21–31.

[6] J. Zhu, Y. Wang, J. Liu, Y. Zhang, Facile one-pot synthesis of novel spherical zeolite-reduced graphene oxide composites for cationic dye adsorption, Ind. Eng. Chem. Res., 53 (2014) 13711–13717.

[7] M. Dolatabadi, T. Świergosz, S. Ahmadzadeh, Electro-Fenton approach in oxidative degradation of dimethyl phthalate –

the treatment of aqueous leachate from landfills, Sci. Total Environ., 772 (2021) 145323, doi: 10.1016/j.scitotenv.2021.145323.
[8] B.-Y. Gao, Q.-Y. Yue, Y. Wang, W.-Z. Zhou, Color removal from dye-containing wastewater by magnesium chloride, J. Environ. Manage., 82 (2007) 167–172.
[9] T. Chen, B. Gao, Q. Yue, Effect of dosing method and pH on color removal performance and floc aggregation of polyferric chloride–polyamine dual-coagulant in synthetic dyeing wastewater treatment, Colloids Surf., A, 355 (2010) 121–129.
[10] K. Lal, A. Garg, Physico-chemical treatment of pulping effluent: characterization of flocs and sludge generated after treatment, Sep. Sci. Technol. (Philadelphia, PA, U.S.), 52 (2017) 1583–1593.
[11] F.H. Mohd. Yunos, N.M. Nasir, H.H. Wan Jusoh, H. Khatoon, S.S. Lam, A. Jusoh, Harvesting of microalgae (*Chlorella* sp.) from aquaculture bioflocs using an environmental-friendly chitosan-based bio-coagulant, Int. Biodeterior. Biodegrad., 124 (2017) 243–249.
[12] B.-Y. Gao, Y. Wang, Q.-Y. Yue, J.-C. Wei, Q. Li, Color removal from simulated dye water and actual textile wastewater using a composite coagulant prepared by polyferric chloride and polydimethyldiallylammonium chloride, Sep. Purif. Technol., 54 (2007) 157–163.
[13] H. Wei, B. Gao, J. Ren, A. Li, H. Yang, Coagulation/flocculation in dewatering of sludge: a review, Water Res., 143 (2018) 608–631.
[14] V. Vajihinejad, S.P. Gumfekar, B. Bazoubandi, Z. Rostami Najafabadi, J.B.P. Soares, Water soluble polymer flocculants: synthesis, characterization, and performance assessment, Macromol. Mater. Eng., 304 (2019) 1800526, doi: 10.1002/mame.201800526.
[15] W. Jaeger, J. Bohrisch, A. Laschewsky, Synthetic polymers with quaternary nitrogen atoms: synthesis and structure of the most used type of cationic polyelectrolytes, Prog. Polym. Sci., 35 (2010) 511–577.
[16] Q. Liu, N. Xia, W. Wan, Y. Gao, S. Zhu, Selective capture of toxic anionic dyes of a novel prepared DMDAAC-grafted chitosan/genipin/cellulose hydrogel beads with antibacterial activity, Int. J. Biol. Macromol., 189 (2021) 722–733.
[17] Q. Su, Y. Wang, W. Sun, J. Liang, S. Meng, Y. Wang, Z. Li, Synthesis and characterization of cyclodextrin-based acrylamide polymer flocculant for adsorbing water-soluble dyes in dye wastewater, R. Soc. Open Sci., 7 (2020) 191519, doi: 10.1098/rsos.191519.
[18] Q. Xu, J. Peng, W. Zhang, X. Wang, T. Lou, Electrospun cellulose acetate/P(DMDAAC-AM) nanofibrous membranes for dye adsorption, J. Appl. Polym. Sci., 137 (2020) 48565, doi: 10.1002/app.48565.
[19] L.-f. Bian, B.-y. Gao, Study on dyeing wastewater by new composites coagulant PAV-PDMDAAC, Jiangsu Huagong, 33 (2005) 54–56.
[20] X. Jia, Y. Zhang, Preparation and characterization of poly(dimethyldiallylammonium chloride) with high molar mass using high purity industrial monomer, J. Appl. Polym. Sci., 118 (2010) 1152–1159.
[21] B. Zhou, Y. Tang, L. Zhao, L. Guo, J. Zhou, Novel Fe_3O_4-poly(methacryloxyethyltrimethyl ammonium chloride) adsorbent for ultrafast and efficient removal of anionic dyes, RSC Adv., 11 (2021) 1172–1181.
[22] S. Wei, W. Chen, Z. Tong, N. Jiang, M. Zhu, Synthesis of a functional biomass lignin-based hydrogel with high swelling and adsorption capability towards Acid Red 73, Environ. Sci. Pollut. Res., 28 (2021) 51306–51320.
[23] T. Wang, X. Tang, S. Zhang, J. Zheng, H. Zheng, L. Fang, Roles of functional microbial flocculant in dyeing wastewater treatment: bridging and adsorption, J. Hazard. Mater., 384 (2020) 121506, doi: 10.1016/j.jhazmat.2019.121506.
[24] S. Khan, H. Zheng, Q. Sun, Y. Liu, H. Li, W. Ding, A. Navarro, Synthesis and characterization of a novel cationic polyacrylamide-based flocculants to remove Congo red efficiently in acid aqueous environment, J. Mater. Sci.: Mater. Electron., 31 (2020) 18832–18843.
[25] T.T. Chen, Y.J. Zhang, Research progress in the synthesis of cationic polyacrylamide P(DACAM), J. Chem. Eng. Chin. Univ., 33 (2019) 1025–1036.
[26] X.Q. Fu, Y.J. Zhang, Research progress in synthesis and application of methacryloyloxyethyltrimethyl ammonium chloride polymers, Fine Chem., 37 (2020) 657–664.
[27] Y. Zhang, Y. Wang Preparation Method of Poly(Methacrylamidopropyltrimethylammonium) Chloride with Serialized Characteristic Viscosity, CN109912739A, Chinese Patent, 2019.
[28] X. Li, H. Zheng, B. Gao, Y. Sun, X. Tang, B. Xu, Optimized preparation of micro-block CPAM by response surface methodology and evaluation of dewatering performance, RSC Adv., 7 (2017) 208–217.
[29] X. Li, H. Zheng, B. Gao, C. Zhao, Y. Sun, UV-initiated polymerization of acid- and alkali-resistant cationic flocculant P(AM-MAPTAC): synthesis, characterization, and application in sludge dewatering, Sep. Purif. Technol., 187 (2017) 244–254.
[30] P. Ilgin, H. Ozay, O. Ozay, Synthesis and characterization of pH responsive alginate based-hydrogels as oral drug delivery carrier, J. Polym. Res., 27 (2020) 251, doi: 10.1007/s10965-020-02231-0.
[31] Z. Long, Z. Wu, R. Yang, Water-Resistant and Moisture-Proof Agent for Paper and Preparation Method Thereof, CN111424462A, Chinese Patent, 2020.
[32] Y. Guo, X. Li, J. Sun, Y. Liu, H. Wang, J. Ding, L. Chen, X. Tian, Y. Yuan, Physicochemical characterization and flocculation performance evaluation of PAC/PMAPTAC composite flocculant, J. Appl. Polym. Sci., 139 (2022) 51653, doi: 10.1002/app.51653.
[33] N.V. Nguyen, J. Singer, X. Zhou, J. Haghpanah, C. Shaw, Cosmetic Compositions Containing Oxazoline Functionalized Polymers and Carboxyl Group-Containing Polymers, US20200069557A1, United States, 2020.
[34] Y.J. Wang, Y.J. Zhang, Research progress in synthesis and application of 3-methacrylamide propyl trimethyl ammonium chloride and its polymers, Fine Chem., 38 (2021) 1333–1341.
[35] B. Valley, B. Jing, M. Ferreira, Y. Zhu, Rapid and efficient coacervate extraction of cationic Industrial dyes from wastewater, ACS Appl. Mater. Interfaces, 11 (2019) 7472–7478.
[36] J. Cui, F. Li, Composite Decolorant for Printing and Dyeing Wastewater, and Preparation Method Thereof, CN111392806A, 2020.
[37] L. Yuan, C. Chen, X.L. Zhao, Research on the flocculation decolorization effects of P(DMC-AM) on reactive dye wastewater, Ind. Water Treat., 37 (2017) 78–82.
[38] K. Guo, B. Gao, W. Wang, Q. Yue, X. Xu, Evaluation of molecular weight, chain architectures and charge densities of various lignin-based flocculants for dye wastewater treatment, Chemosphere, 215 (2019) 214–226.
[39] B. Ren, F. Min, J. Chen, F. Fang, C. Liu, Adsorption mechanism insights into CPAM structural units on kaolinite surfaces: a DFT simulation, Appl. Clay Sci., 197 (2020) 105719, doi: 10.1016/j.clay.2020.105719.