Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2014.884471

54 (2015) 690–698
April

Novel multiple coagulant from Bayer red mud for oily sewage treatment

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Received 31 May 2013; Accepted 11 January 2014

ABSTRACT

Multiple coagulants for the treatment of wastewater were prepared. To obtain the optimum synthetic conditions of coagulants, response surface methodology was used to establish the relationship between parameters and the treatment efficiency. In addition, parameters affecting the coagulation–flocculation process, such as the dosage of attapulgite (ATP), the dosage of poly-dimethyl-diallyl-ammonium chloride (PDMDAAC), and the reaction temperature, were investigated. The optimized parameters to prepare multiple coagulant were determined as the dosage of ATP and PDMDAAC being 4 and 2%, respectively, and the reaction temperature being 60℃. The coagulation performance for oily sewage showed that the optimum dosage of coagulant for the maximum treatment was 100 mg/L and the sedimentation time was 30 min at 60℃. In the coagulation–flocculation process, the COD of the oily water decreased from 534 to 246 mg/L and its turbidity decreased from 124 to 2.0 NTU.

Keywords: Multiple coagulant; Coagulant; Response surface methodology (RSM); Oily sewage

1. Introduction

Inorganic polymeric flocculants (IPF) are widely used as flocculant in water and wastewater treatment [1]. IPF are new class of water treatment agents developed on the basis of the traditional aluminum salts and iron salts after 1960s. IPF are proved more effective than the conventional simple inorganic salts, as their performance is more stable over seasonal changes of the water such as temperature or pH variations [2]. Poly-aluminum-ferric-chloride (PAFC) is among the most efficient inorganic coagulant, which possesses the advantage of both Al and Fe polymers [3].

Researchers are recently focused on the preparation of inorganic–organic coagulants, by combining a cationic inorganic coagulant with an organic polymer, to utilize the effective properties of both components [4]. Although IPF can be used to dispose various kinds of complicated compositions of sewage water, the resulting floc is smaller than those generated by organic polymeric flocculants (OPF). Large dosage is needed for IPF to get satisfactory performance. OPF can just compensate for this shortcoming. Therefore, the combination of the two flocculants might generate

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new flocculants with higher performance. In the traditional wastewater treatment process, the inorganic coagulant and the organic flocculant are needed to be introduced in the wastewater separately, requiring two reagent addition systems [5].

Attapulgite (ATP) is a kind of hydrous magnesium carbonate clay mineral [6]. Its ideal formula is $[(OH_2)_4(Mg, Al, Fe)_5Si_8O_{20}(OH)_2]$ mH_2O, $w(MgO) = 23.83\%$, $w(SiO_2) = 56.96\%$, and $w(H_2O) = 19.21\%$. Since ATP is a porous material, its surface area is high, resulting in high physical adsorption performance [7]. As a result, organic pollutants could be adsorbed into ATP effectively. When disposing the phenol and the ammonia nitrogen, the removal rate by ATP was 85% in the optimized conditions [8,9]. ATP as a flocculant has great potentials in the treatment of wastewater for its high absorption efficiency, low price, non-corrosive nature, and being easy to operate.

Preparation of inorganic–organic coagulant has attracted increasing attention over the last decades. A few researchers [10,11] reported the preparation of a composite coagulant consisting of cationic inorganic coagulant with polymeric ferric aluminum chloride and the cationic organic poly-dimethyl-diallyl-ammonium chloride (PDMDAAC). Similarly, in previous studies, the preparation of PDMDAAC–ATP adsorbents has also been accomplished [12–14]. But there is no report on the preparation of a composite coagulant consisting of PDMDAAC, PAFC, and ATP.

Therefore, a novel composite inorganic–organic coagulant was prepared by using inorganic flocculants (PAFC) which was laboratory-made from red mud, ATP, and organic flocculants (PDMDAAC) in this study. In order to optimize the synthetic conditions, the effects of the dosage of ATP and PDMDAAC and the reaction temperature on the performance of the coagulation–flocculation were investigated. The response surface methodology (RSM) was employed to investigate the above mentioned parameters rather than a “one-factor-at-a-time” strategy, which is generally a time-consuming and expensive process as a large number of experiments must be carried out [15–18]. Finally, the performance of the coagulant for oily sewage was tested, and the optimum conditions, such as dosage of coagulant, temperature, and the sedimentation time, for oily sewage were determined.

2. Materials and methods

2.1. Materials and instruments

All the mineral materials and organic flocculants used in the experiments were technical grade. The PAFC used in the experiments was laboratory-made from red mud in the form of dried-clay, which was obtained from Shandong Weiqiao Aluminum & Electricity Co., Ltd. Shandong, China. All the solutions were prepared with deionized water. SGZ-2 turbidimeter, Shanghai Yuefeng Instruments & Meters Co., Ltd Export Department, was used to measure turbidity of the treated water; COD was measured by 5B-6 COD reactor and COD-Cr (potassium dichromate as oxidant), Lianhua Tech. Co., China. The morphologies of the coagulants were analyzed by Hitachi S-4800 high-resolution ice emission scanning electron microscopy (SEM).

2.2. Wastewater sample

Oily wastewater was employed in the process of optimizing the performance of the multi-flocculant. The oily water was obtained from Liao he Petroleum Exploration Bureau of China, COD of which was 534 mg/L, the turbidity was 124 NTU, and pH of the oily water was 6.

2.3. Experimental design

Generally, RSM was used to establish the relationship between one or more response variables and a set of quantitative, experimental variables or factors [19]. The relationship between the parameters including the dosage of ATP and PDMDAAC, and the reaction temperature was investigated to get the optimal performance of the flocculant. To establish the combination effect of these parameters, the coagulation–flocculation performance was evaluated at three levels of each factor using the central composite design (CCD). The factor levels were selected that the upper level corresponded to +1, the lower level to −1, and the basic level to zero. The experimental ranges and levels of the independent test variables employed are presented in Table 1. The turbidity and COD removal efficiencies were selected as the response values (dependent variable). The total number of experiments

Table 1
Experimental ranges and levels of the independent test variables

	Range and level/s				
Variables	−2	−1	0	1	2
X_1, ATP (wt.%)	0	2	4	6	8
X_2, PDMDAAC (wt.%)	0	1	2	3	4
X_3, temperature (℃)	20	40	60	80	100

Table 2
Three-factor CCD matrix and the value of response function (residual turbidity (NTU))

	Factors			Responses	
Run	ATP (wt.%)	PDADCCA (wt.%)	Temperature (℃)	Residual turbidity (NTU)	Residual COD (mg/L)
1	0	−2	0	12.1	350.0
2	0	0	0	2.9	271.0
3	−1	−1	1	8.9	316.0
4	1	1	1	6.5	288.0
5	0	0	0	2.9	265.0
6	1	1	−1	4.6	337.0
7	−1	1	−1	8.6	317.0
8	1	−1	1	7.0	305.0
9	−1	−1	−1	10.7	325.0
10	0	0	0	2.7	269.0
11	−1	1	1	7.7	302.0
12	0	0	0	2.2	268.0
13	1	−1	−1	9.0	320.0
14	2	0	0	8.2	311.0
15	0	0	−2	11.6	332.0
16	−2	0	0	11	330.0
17	0	2	0	8.8	319.0
18	0	0	0	2.6	267.0
19	0	0	2	8.4	300.0
20	0	0	0	2.7	264.0

with three variables was 20(= $2^k + 2k + 6$), where k was the number of independent variables. Therefore, the investigation was carried out through 20 experiments as presented in Table 2.

For statistical calculations, the variables X_i (the real value of an independent variable) were coded as x_i (dimensionless value of an independent variable) according to the following equation:

$$x_i = \frac{X_i - X_0}{\delta X} \tag{1}$$

where X_0 was the value of X_i at the center point and X_i represented the step change [20].

The results were analyzed using the quadratic equation model, which showed the effect of variables in terms of linear, quadratic, and cross-product terms:

$$Y = b_o + \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_{ii} x_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} b_{ij} x_i x_j \tag{2}$$

where Y was the predicted response (turbidity removal efficiency and COD removal efficiency), b_0, b_i, b_{ii}, and b_{ij} were the constant coefficient, the linear coefficients, the quadratic coefficients, and the interaction coefficients, respectively. x_i and x_j were the coded values of the variables.

The regression analysis for the quadratic equation model was investigated using design expert. $\hat{Y}$ was the dependent variables (turbidity removal efficiency and COD removal efficiency), β_o was constant coefficient , β_i was regression coefficient, e was the residual error, and x_i was the values of the independent variables.

$$\hat{Y} = \beta_o + \sum_{i=1}^{n} \beta_i X_i + e \tag{3}$$

Firstly, the feasibility of the quadratic equation model between the test and response variables was established using analysis of variance (ANOVA). Then, F-test and p-values (probability) with 95% confidence level were used to check for the statistical significance of the quadratic equation model and test variables. Secondly, in order to test the model fit, the modeling quality of the model was determined by using the coefficient of determination (R^2). Thirdly, the coefficient of variation (CV), which was a normalized measure of dispersion of a probability distribution, was used to assess the precision and reliability of the experiments. The CV, as the ratio of the standard error of estimate and the mean value of the response for a model, aimed to describe the model fit in terms of the relative sizes of the squared residuals and outcome values, defining reproducibility of the model [21]. As a general rule, a model can be considered reasonably reproducible if its CV is not greater than 10% [22]. Lastly, for investigation of the mutual effects of the test variables on the response value, two-dimensional respective contour was plotted.

3. Results and discussion

3.1. Factors affecting performance of multiple flocculant

3.1.1. ANOVA and model fitting

ANOVA results of the quadratic model for residual turbidity and COD are shown in Tables 3 and 4. Parameters like *F*-value, Probability > *F*, Lack of Fit and R^2 were the measurement of the quality of the fitting.

The model *F*-value of 418.89 and 334.73 for residual turbidity and COD respectively indicated that the models were significant and there was only 0.01% chance that model *F*-values could occur due to noise [23–25]. Values of "Probability > *F*" being less than 0.0500 indicated that model terms were significant. The "Lack of Fit *F*-value" of 0.65 implied that the Lack of Fit was not obviously related to the pure error. There was a 67.88% chance that a "Lack of Fit *F*-value" could occur largely due to noise for turbidity removal. A non-significant Lack of Fit was considered good and was desirable for the fitting. Also, the Lack of Fit *F*-value of 0.30 for COD removal implied the Lack of Fit was not obviously related to the pure error for this matter removal. There was only an 89.37% chance that a Lack of Fit *F*-value could occur due to noise.

The fitting of the model was also expressed by the coefficient of determination R^2, which was found to be 0.9974 and 0.9967 for residual turbidity and COD separately, indicating that 99.74 and 99.67% of the variability in the response could be explained by the model. The high values of adjusted R^2 (0.9950 and 0.9937) implied the model fitted the experimental data well. "Adeq Precision" measures the signal-to-noise ratio, and a ratio greater than 4 was desirable. Therefore, in the quadratic models of residual turbidity and COD, the ratios of 56.355 and 55.410 indicated adequate signals for the models to be used to navigate the design space.

3.1.2. Interactive effect of the ATP, PDMDAAC, and temperature on the residual turbidity

The contour plots of the residual turbidity in Fig. 1 were generated with one variable keeping at its zero level and varying the others within the experimental range.

Fig. 1(a) presents an elliptic characteristic with the long axis of the ellipse running along the ATP axis. This indicated that PDMDAAC was more influential than ATP in the design range. As Fig. 1(a) showed that at low values of ATP and PDMDAAC, the residual turbidity was remarkably high. With increasing ATP and PDMDAAC, the response value decreased continuously till its lowest level of 2.5 NTU and then gradually increased. The optimum values of ATP and PDMDAAC for residual turbidity were 4.4 and 2.2%, respectively.

The interaction of ATP and temperature is shown in Fig. 1(b). According to the contour plots, there was an elliptic characteristic with the long axis of the ellipse running along the ATP axis. This indicated that temperature was more influential than ATP in the design range. Fig. 1(b) shows that at low values of ATP and temperature, the residual turbidity was remarkably high. With the increase of dosage of ATP and the temperatures, the response values decreased continuously till its lowest level of 2.5 NTU and then gradually increased. The optimized values of ATP dosage and temperature for residual turbidity were 4.4% and 64℃, respectively.

According to Fig. 1(c), the long axis of the ellipse ran along the PDMDAAC axis, pointing out that the influence of temperature was larger than PDMDAAC

Table 3
ANOVA results for residual turbidity

Source	Degree of freedom	Sum of squares	Mean square	*F*-value	*p*-value
Regression	9	206.75	22.97	418.89	<0.0001
Residual	10	0.55	0.055		
Lack of Fit	5	0.22	0.043	0.65	0.6788
Pure error	5	0.33	0.067		
Total	19	207.30			
R^2	0.9974				
R^2 adjusted	0.9950				

Table 4
ANOVA results for residual COD

Source	Degree of freedom	Sum of squares	Mean square	*F*-value	*p*-value
Regression	9	13056.86	1450.76	334.73	<0.0001
Residual	10	43.34	4.33		
Lack of Fit	5	10.01	2.00	0.30	0.8937
Pure error	5	33.33	6.67		
Total	19	13100.20			
R^2	0.9967				
R^2 adjusted	0.9937				

in the design range. The residual turbidity was high while PDMDAAC and temperature were kept at low levels. The response values increased gradually while PDMDAAC increased from 1 to 2.5% and the temperature varied between 40 and 64℃, but decreased as the PDMDAAC and temperature increased further. At PDMDAAC of 2.2% and temperature of 64℃, residual turbidity reached its lowest level of 2.5%.

3.1.3. Interactive effect of the ATP, PDMDAAC, and temperature on the residual COD

The contour plots of residual COD in Fig. 2 were generated with one variable kept at its zero level and varying the others within the experimental range.

The noticeable elongated maxima running along ATP axis were observed in both Fig. 2(a) and (b); the contour plots presented elliptic characteristic with the long axis of the ellipses perpendicular to the respective PDMDAAC and temperature axis. These indicated that PDMDAAC and temperature were more influential than ATP on COD removal rate. The COD removal rate was hardly influenced when the dosage of ATP was more than 5%. These results were consistent with the results of turbidity removal in this study.

The effects of PDMDAAC and temperature on the response were presented at zero level ATP of 4% and are shown in Fig. 2(c). The residual COD was lowest at PDMDAAC of 2.2% and temperature of 67℃. Fig. 2(c) shows that PDMDAAC had obviously impacted the response of COD removal rate. It showed a satisfactory endurance of high pH in a wide range in terms of COD removal; however, the same decreasing tendency of turbidity removal occurred while PDMDAAC exceeded 5%.

With increasing of the amount of PDMDAAC, the treatment effect was getting better, which was due to the fact that PDMDAAC was a positively charged polymer. The introduction of a cationic polymer can improve the charge neutralization of the ferro-aluminum flocculant, thereby enhancing its flocculation capacity [26]. The higher the content of PDMDAAC, the greater the charge neutralization effect, and the adsorption bridging ability are. But when the amount of PDMDAAC increased to a certain extent, the treatment effect deteriorated, mainly due to the reason that PDMDAAC had strong bridging sweeping effect, the inorganic coagulant was precipitated by the strong bridging effect of PDMDAAC before the charge neutralization of inorganic coagulant and the adsorption effect of PDMDAAC fully reached equilibrium. Therefore, the optimum dosage of PDMDAAC for the preparation of the multiple coagulant was determined as 2%.

Increasing the temperature was favored for the dispersion and dissolution of PDMDAAC. But when the temperature was too high, the stability of the composite solution was poor; when the temperature reached 100℃, insoluble flocculent appeared near the surface of the composite solution. The reason might be that as the temperature rose, the PDMDAAC molecular swelling caused mobility variations [27]. Therefore, the optimum reaction temperature was decided as 60℃.

After modification, organic carbon content increased, which resulted in improved hydrophobic properties of ATP. When the multiple coagulant was used to treat wastewater, the long carbon chain on the surface of ATP adsorbed organic molecules in wastewater through hydrogen bonding and van der Waals interactions, which caused aggregation of the fine suspension and colloidal particles, and made the adsorption layer denser. Therefore, the coagulation performance was improved. But when the amount of ATP increased too much, the treatment effect deteriorated, mainly due to the fact that the interaction between the coagulant and pollutant was reduced and thus the coagulant effect was weakened. Consequently, the optimum dosage of ATP for the preparation of the multiple coagulant was determined as 4%.

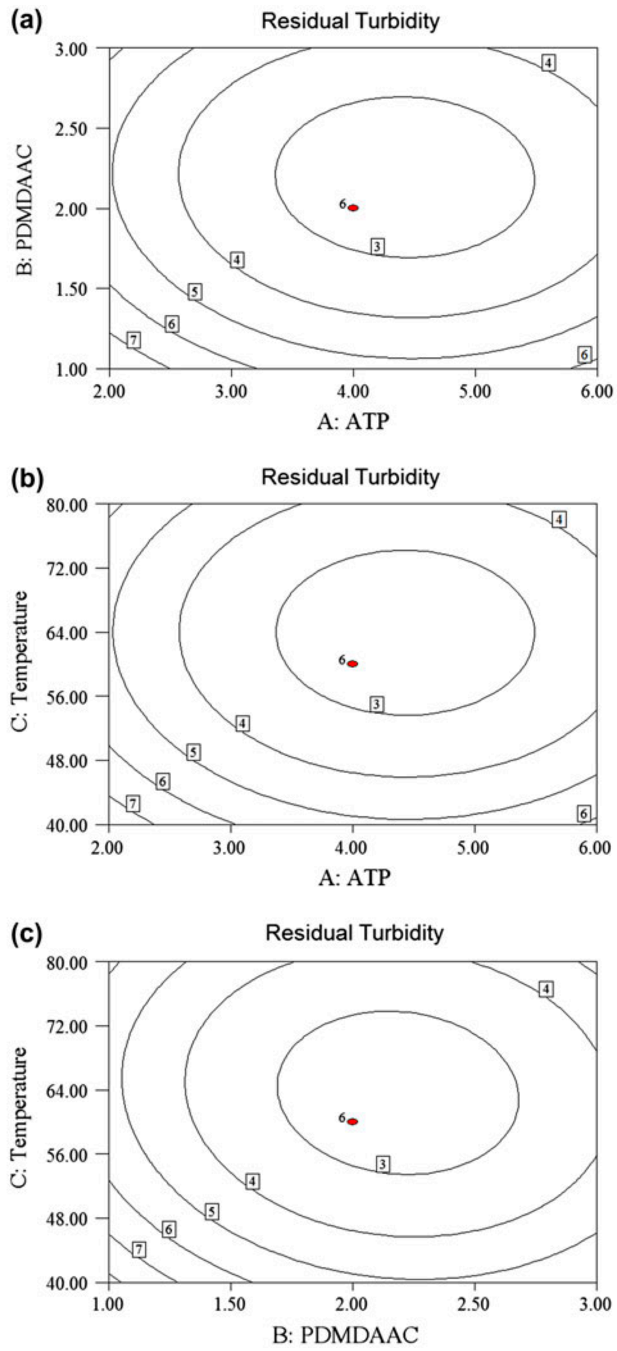

Fig. 1. Contour plots of residual turbidity. Interactive effects of ATP and PDMDAAC on residual turbidity (a); interactive effects of ATP and temperature on residual turbidity (b); and interactive effects of PDMDAAC and temperature on residual turbidity (c).

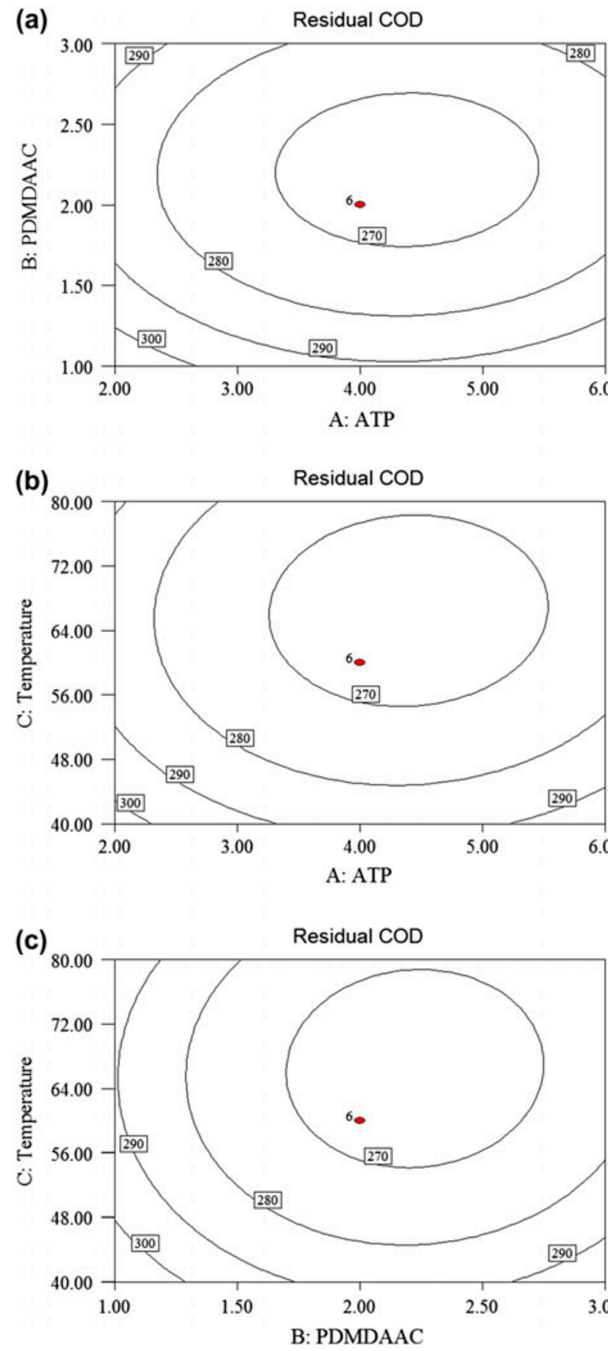

Fig. 2. Contour plots of residual COD. Interactive effects of ATP and PDMDAAC on residual COD (a); interactive effects of ATP and temperature on residual COD (b); and interactive effects of PDMDAAC and temperature on residual COD (c).

3.2. Application of the multiple coagulant in oily sewage

3.2.1. Effect of dosage of coagulant

Coagulant dosage is an important factor affecting the treatment effect. Fig. 3 illustrates the effect of coagulant dosage on residual turbidity and COD. With the increase of coagulant dosage, the residual turbidity and COD decreased noticeably until the coagulant dosage reached 100 mg/L. In addition, as the dosage increased over 100 mg/L, there was an obvious

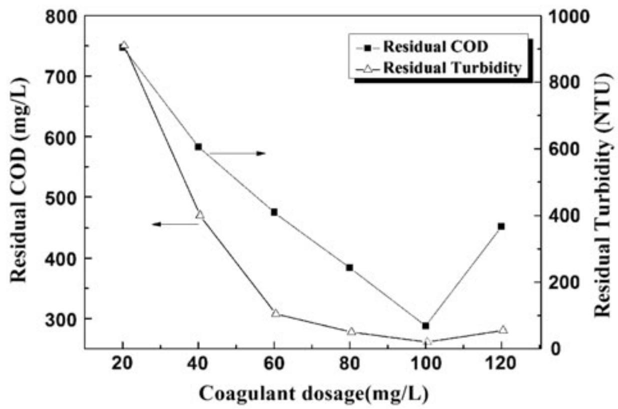

Fig. 3. Effect of coagulant dosage on turbidity removal and COD removal for oily sewage.

increase in the residual COD and a little increase in the residual turbidity. Too high or too low coagulant dosage will decrease coagulant stability and its overall performance. If coagulant dosage was too low, charge neutralization and adsorption bridging would not be achieved. If the amount was too high, the micelle surface will be saturated, causing the increase of the zeta potential and thus the repulsive force between the small particles [28,29]. Then, the formed floc would re-disperse into a stable colloidal and coagulant effect would decrease. Therefore, the optimum treatment efficiency was achieved at the dosage of 100 mg/L, where the residual turbidity and COD was 2.5 NTU and 286 mg/L, respectively. The treatment efficiency of turbidity and COD were 98.0 and 46.4%.

3.2.2. Effect of temperature

In order to study the effect of temperature, other parameters for the treatment of oily sewage were kept constant while varying the reaction temperatures from 25 to 100℃. The temperature effect on residual turbidity and COD is shown in Fig. 4. It showed that with the increase of coagulant dosage, the residual turbidity and COD decreased noticeably until the temperature reached 60℃. In addition, as the temperature was higher than 60℃, there was an obvious increase in the residual turbidity and COD. When the reaction temperature was too high, the chemical reaction was accelerated, and the hydration of the formed little sized floc increased. So, some floc was partly dissolved, which was disadvantageous for the settlement of the flocculant. When the water temperature was too low, the coagulant hydrolysis reaction was slowed, and the hydrolysis time was lengthened. In addition, at low temperatures, the viscosity of the wastewater would increased, which was also disadvantageous for the settlement of the flocculant [30]. Therefore, the optimum treatment efficiency was achieved at the temperature of 60℃, where the residual turbidity and COD were 2.0 NTU and 246 mg/L, respectively. Under these optimized conditions, the turbidity and COD removal efficiency of the prepared coagulant were 98.4 and 53.9%, respectively.

3.2.3. Effect of sedimentation time

In order to obtain the optimized sedimentation time, other parameters for the treatment of oily sewage were kept constant while varying the sedimentation time from 10 to 60 min. The effect of sedimentation time on residual turbidity and COD is shown in Fig. 5. It showed that with the increase of sedimentation time, the residual turbidity and COD decreased noticeably until the sedimentation time of 30 min. In addition, as the sedimentation time increased above 30 min, there

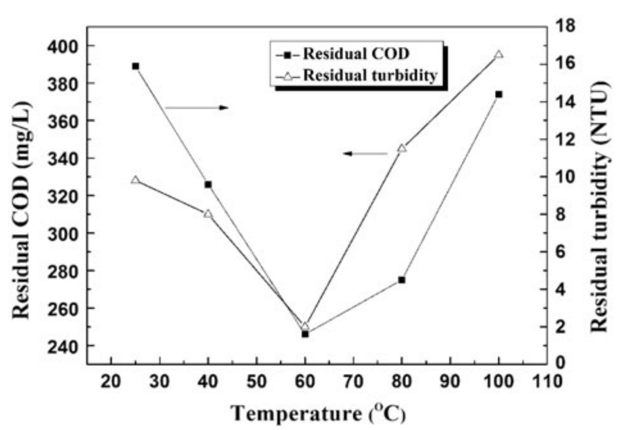

Fig. 4. Effect of temperature on turbidity removal and COD removal for oily sewage.

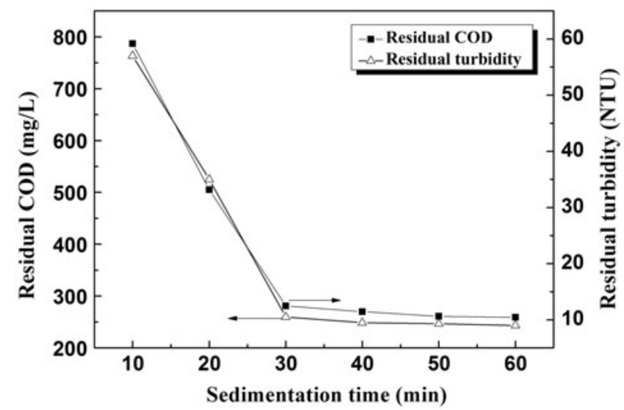

Fig. 5. Effect of sedimentation time on turbidity removal and COD removal for oily sewage.

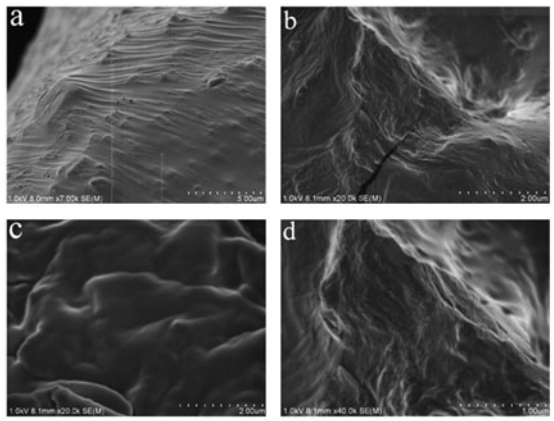

Fig. 6. The SEM of multiple flocculant.

was no obvious decrease in the residual turbidity and COD. Also, the color of the water did not change significantly. Therefore, the optimum treatment efficiency was achieved at the sedimentation time of 30 min, where the residual turbidity and COD were 10 NTU and 280 mg/L, respectively. The turbidity and COD treatment efficiency by the coagulant were 91.9 and 47.6%, respectively.

3.3. Characterization of multiple flocculant

In order to further analysis the above results, the morphologies of the multiple flocculent in the micrometer range were observed with SEM and shown in Fig. 6. It displayed rough surfaces covered with ridges and wrinkles. The rough surface morphology might result from a larger degree of dehydration during the preparation of the SEM sample in vacuum. It requires further studies for the sufficient proof.

4. Conclusion

In this research, results of an experimental study on multiple coagulants as a modified coagulation reagent for the treatment of wastewater were presented. During the process of preparing multiple coagulant, parameters impacting the synthesis process were experimentally studied. The results showed the optimized parameters to prepare multiple coagulant were determined as the dosage of ATP and PDMDAAC being 4 and 2%, respectively, and the reaction temperature being 60℃. Also to obtain the optimum synthetic conditions, RSM was used to establish the relationships between the above parameters and the treatment efficiencies. In addition, parameters affecting the coagulation–flocculation processes using multiple coagulant, including the dosage of ATP and PDMDAAC and reaction temperature, were investigated by testing the COD and turbidity of the treated water.

The performance tested with oily sewage showed that the optimum coagulant dosage for the maximum treatment efficiency was 100 mg/L, the reaction temperature was 60℃, and the sedimentation time was 30 min. The high performance of multiple coagulant confirmed that a new material for the effective treatment of wastewater was prepared.

Acknowledgements

This research was jointly supported by National High Technology Research and Development Program (863 Program 2012AA06A109) of China, the Fundamental Research Funds for the Central Universities (2010ZD08).

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