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Optimization of the coagulation-flocculation process for pulp mill wastewater treatment using a combination of uniform design and response surface methodology

Jian-Ping Wang [a,b], *Yong-Zhen Chen* [a], *Yi Wang* [b], *Shi-Jie Yuan* [a], *Han-Qing Yu* [a,*]

[a] *Department of Chemistry, University of Science and Technology of China, Hefei, 230026, China*
[b] *Department of Agricultural and Biological Engineering, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA*

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ABSTRACT

Pulp mill wastewater was treated using the coagulation-flocculation process with aluminum chloride as the coagulant and a modified natural polymer, starch-*g*-PAM-*g*-PDMC [polyacrylamide and poly (2-methacryloyloxyethyl) trimethyl ammonium chloride], as the flocculant. A novel approach with a combination of response surface methodology (RSM) and uniform design (UD) was employed to evaluate the effects and interactions of three main influential factors, coagulant dosage, flocculant dosage and pH, on the treatment efficiency in terms of the supernatant turbidity and lignin removals as well as the water recovery. The optimal conditions obtained from the compromise of the three desirable responses, supernatant turbidity removal, lignin removal and water recovery efficiency, were as follows: coagulant dosage of 871 mg/L, flocculant dosage of 22.3 mg/L and pH 8.35. Confirmation experiments demonstrated that such a combination of the UD and RSM is a powerful and useful approach for optimizing the coagulation-flocculation process for the pulp mill wastewater treatment.

1. Introduction

The gross output of paper and paperboard is 79.8 million tons in 2008 in China, and about 22% of them are using straw pulp as staffs. The wastewater generated by the pulp mills is not easy to treat due to the presence of a large amount of chemicals, such as many sodium salts of organic acids. In addition, a large amount of lignin present in the wastewater, which causes colority, turbidity and high COD (chemical oxygen demand), is usually wasted and overburdens the treatment process. Thus, both the lignin reclamation and wastewater treatment are crucial. An efficient and cost-effective process for the treatment of pulp mill wastewater should be pursued. Coagulation-flocculation is a simple and efficient method for wastewater treatment, and has been widely used for the treatment of palm oil mill effluent (Ahmad et al., 2005), textile wastewater (Meric et al., 2005) and abattoir wastewater (Amuda and Alade, 2006), etc. Recently, coagulation-flocculation or flocculation processes have also been extensively used for the treatment of pulp mill wastewater. In such studies, polyaluminium chloride (PAC), chitosan, polymeric phosphate-aluminum chloride, cationic and anionic polyacrylamides (PAMs) and polydiallyldimethylammonium chloride (polyDADMAC) have all been tested as a flocculant in the flocculation process, and various levels of removal efficiency for turbidity and lignin have been achieved (Razali et al., 2011; Renault et al., 2009; Wong et al., 2006; Zheng et al., 2011). In the coagulation-flocculation process, the efficiency is governed by various factors, such as the type and dosage of coagulant/flocculant (Desjardins et al., 2002; Hu

* *Corresponding author*. Fax: +86 551 3601592.
E-mail address: hqyu@ustc.edu.cn (H.-Q. Yu).
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et al., 2005; Nandy et al., 2003; Spicer and Pratsinis, 1996; Wang et al., 2002), pH (Elmaleh et al., 1996; Miller et al., 2008; Rohrsetzer et al., 1998; Syu et al., 2003), mixing speed and time (Gurses et al., 2003; Rossini et al., 1999), temperature and retention time (Howe et al., 2006; Zhu et al., 2004). A proper optimization of these factors could significantly increase its treatment efficiency.

Response surface methodology (RSM) is an efficient way to achieve such an optimization by analyzing and modeling the effects of multiple variables and their responses and finally optimizing the process. This method has been widely used for the optimization of various processes in food chemistry, material science, chemical engineering and biotechnology (Granato et al., 2010; Hong et al., 2011; Liu et al., 2010; Singh et al., 2010). In the traditional experimental design approaches used in RSM, such as central composite design, with an increase in experimental factors, the number of coefficients of the quadratic model equation increases exponentially and so does the number of experimental trials (Cheng et al., 2002). To overcome this shortcoming of RSM, uniform design (UD) can be used to investigate more factors with substantially fewer experimental trials, since it determines the number of the experimental trials only by the level of factors, rather than by the number of factors. The UD method was first proposed by Fang (1978). Compared to the traditional experimental design methods, UD is capable of selecting experimental points uniformly in the experimental region and highly representative in the experimental domain; it imposes no strong assumption on the model and may be used when the underlying model between the responses and factors is unknown or partially unknown; it can accommodates the largest possible number of levels for each factor among all experimental designs (Leung et al., 2000; Li et al., 2003; Wen et al., 2005; Zhang et al., 1998). A combination of UD and RSM would be able to achieve the optimization of a complex multivariate process with the fewest multilevel experiments. Therefore, in this study, UD and RSM were integrated to optimize the coagulation-flocculation process for pulp mill wastewater treatment.

The selection of high efficient coagulants and flocculants is essential for a successful coagulation-flocculation process. In this work, on the basis of our previous studies (Wang et al., 2007, 2009), the conventional coagulant $Al(OH)_3$ was chosen as the coagulant, while a novel modified natural polymer, starch-*g*-PAM-*g*-PDMC [polyacrylamide and poly (2-methacryloyloxyethyl) trimethyl ammonium chloride] with both strong charge neutralization and bridging abilities, was used as the flocculant.

The main objective of this work was to treat the pulp mill wastewater using the coagulation-flocculation process, which was optimized using an integrative UD-RSM approach. Removal efficiencies of both supernatant turbidity and lignin, and recovery efficiency of clean water were chosen as the dependent output variables. The compromise optimal conditions for these three responses were obtained using the desirability function approach. The novel optimization strategy used for the pulp mill wastewater treatment process in this study is expected to provide valuable information for other complicated systems in environmental engineering and other fields.

2. Materials and methods

2.1. Chemicals and operation

Aluminum chloride, hydrochloric acid and sodium hydroxide, purchased from Shanghai Chemical Reagent Co., China, were of analytical reagent grade and used without further purification. The flocculant, starch-*g*-PAM-*g*-PDMC, was prepared as follows: starch, (2-methacryloyloxyethyl) trimethyl ammonium chloride and acrylamide were dissolved in water in Pyrex glass vessels, and were heated in a preset water bath using potassium persulphate as the initiator after deoxygenating. Thereafter, the sample solutions were precipitated in acetone and separated by filtration. Homopolymers formed in the reactions were removed using the soxhlet extraction method in ethanol. All the grafted samples were dried in a vacuum oven at 50 °C until a constant weight. The grafting percentage and the cationic degree of the graft copolymer used in this experiment was 269% and 1.96×10^{-3} mol/g, respectively. The point of zero charge of the graft copolymer was measured as 7.80. Its molecular structure is as shown in Fig. 1 and its image is illustrated in Fig. S1 (Appendix).

Pulp mill wastewater was blending black liquor from the primary sedimentation tank of Guoyang Paper and Pulp Mill Co., China. The stuffs of the paper were wheat straw. The initial pH, chemical oxygen demand (COD) and turbidity were 6.99, 1358 mg/L and 1209 NTU, respectively. The average sizes of the colloid particles in the wastewater were 544 nm.

The coagulation-flocculation experiments were carried out using the jar test method in 1-L beakers. After the coagulant (stock solution of 50.0 g/L) was added with a dosage varying from nil to 2100 mg/L, the solution pH was adjusted to 2.5–11.5 by adding 0.1 mol/L HCl or NaOH solutions. Then, the flocculant at a concentration of 1.0 g/L was added with a dosage varying from nil to 48 mg/L. The sample was immediately stirred at a constant speed of 200 rpm for 2 min, followed by a slow stirring at 40 rpm for 10 min; thereafter, a settlement for 5 min was performed. After that, samples were taken from the water level around 2 cm underneath the surface for measuring the turbidity and lignin concentrations of the supernatant. Meanwhile, the volume of produced sludge was calculated directly from the reading on the beakers, and then the volume of the recovered clean water was calculated accordingly.

2.2. Experimental design and data analysis

UD tables can be described as $U_n(q^m)$, where U, n, q and m stand for the UD, the number of experimental trials, the number of levels and the maximum number of factors, respectively. For a given measure of uniformity M, a uniform design has the smallest M-value overall fractional factorial design with n runs and m q-level factors.

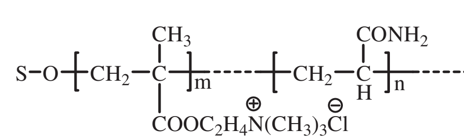

Fig. 1 – Molecular structure of the graft copolymer starch-*g*-PAM-*g*-PDMC (S: Starch).

In this work, coagulant dosage (X_1), flocculant dosage (X_2) and pH (X_3) were chosen as three independent variables in the coagulation-flocculation process. Seven levels for each factor were selected to investigate the influence and interaction of the factors. In order to improve the accuracy, the experiment were carried out using $U_{14}(14^3)$, the range and levels of each factor, the guide for using ($U_{14}(14^3)$) are listed in Tables 1 and 2, respectively.

Efficiencies of turbidity removal, lignin removal and clean water recovery were selected as the dependent variables in order to represent the overall wastewater treatment efficiency. The response variable was fitted by a sufficient model, which is able to describe the relationship between the dependent output variable and the independent variables using the regression method.

$$Y = b_0 + \sum_{i=1}^{j} b_i X_i + \sum_{i=1}^{k} b_{ii} X_i^2 + \sum_{i}^{i<j} \sum_{j} b_{ij} X_i X_j \quad (1)$$

where Y is the response variable to be modeled; X_i and X_j are the independent variables which influence Y_m; b_0, b_i, b_{ii} and b_{ij} are the offset terms, the ith linear coefficient, the quadratic coefficient and the ijth interaction coefficient, respectively. The actual design of this work is given in Table 3 (Fang, 1994).

The parameters of the response equations and corresponding analysis on variations were evaluated using Uniform Design Software 2.1 (http://www.math.hkbu.edu.hk/UniformDesign/software) and MATLAB 6.5, respectively. The interactive effects of the independent variables on the dependent ones were illustrated by three- and two-dimensional contour plots. Finally, two additional experiments were conducted to verify the validity of the statistical experimental strategies.

3. Results

The experimental results are listed in Table 3. The variance trend was discrepant for the three responses. Therefore, the operational conditions have been optimized respectively for different responses.

3.1. *Optimization for supernatant turbidity removal*

The supernatant turbidity removal efficiency listed in Table 3 is an important denotation for the treatment efficiency of the coagulation-flocculation process. The following equation is the regression model using the backward regression method with the experimental results:

$$Y = 29.9 + 22.1X_1 + 11.6X_2 - 2.5X_1^2 - 0.6X_2^2 - 0.7X_3^2 - 1.5X_1X_2 + 1.2X_1X_3$$
$$R^2 = 0.917, \; F = 8.81 \quad (2)$$

Statistical testing of the model was performed with the Fisher's statistical test for analysis of variance (ANOVA). The quadratic regression shows that the model was significant because the value of $F_{statistic}$ (the ratio of mean square due to regression to mean square to real error) of 8.81 was greater than $F_{0.001,7,6}$ (4.27). The value of the correlation coefficient ($R^2 = 0.917$) indicates that only 8.3% of the total variation could not be explained by the empirical model (Leung et al., 2000; Meric et al., 2005).

The p-value ($p = 0.05$) of Eq. (2) also implies that the second-order polynomial model fitted the experimental results well. This is confirmed by Fig. 2a, in which the plots of predicted turbidity removal efficiencies versus measured ones are shown. Most points distributed near to the straight line where the measured and predicted removal efficiencies are the same, indicating that the regression model is able to predict these removal efficiencies.

From Eq. (2), the optimal conditions for the supernatant turbidity removal efficiency were obtained as follows: coagulant dosage of 917 mg/L, flocculant dosage of 33.0 mg/L and pH

Table 1 – Levels of the variable tested in the U7(7^3) uniform designs.

Variables	Range and levels						
	1	2	3	4	5	6	7
X_1, coagulant dosage (mg/L)	0	350	700	1050	1400	1750	2100
X_2, flocculant dosage (mg/L)	0	8	16	24	32	40	48
X_3, pH	2.5	4.0	5.5	7.0	8.5	10.0	11.5

Table 2 – Guide for selecting columns of generating vectors in $U_{14}(14^3)$.

No. of factors studied	Columns to be used	Discrepancy in uniformity
2	1, 4	0.0957
3	1, 2, 3	0.1455
4	1, 2, 3, 5	0.2091

Table 3 – UD and response results for the study of three experimental variables in coded units.

Run	X_1	X_2	X_3	Factors			Response		
				X_1	X_2	X_3	Turbidity removal (%)	Lignin removal (%)	Water recovery efficiency (%)
1	1	4	7	1	2	4	63.9	36.0	66.0
2	2	8	14	1	4	7	56.9	34.5	50.0
3	3	12	6	2	6	3	95.4	71.6	60.0
4	4	1	13	2	1	7	54.3	24.5	62.0
5	5	5	5	3	3	3	92.7	80.9	68.0
6	6	9	12	3	5	6	92.9	70.9	60.0
7	7	13	4	4	7	2	95.8	74.3	48.0
8	8	2	11	4	1	6	87.8	47.6	68.0
9	9	6	3	5	3	2	92.6	76.2	50.0
10	10	10	10	5	5	5	98.7	82.5	66.0
11	11	14	2	6	7	1	71.2	40.1	50.0
12	12	3	9	6	2	5	95.1	59.5	64.0
13	13	7	1	7	4	1	64.8	45.3	42.0
14	14	11	8	7	6	4	67.2	32.5	64.0

Source: Fang, 1994.

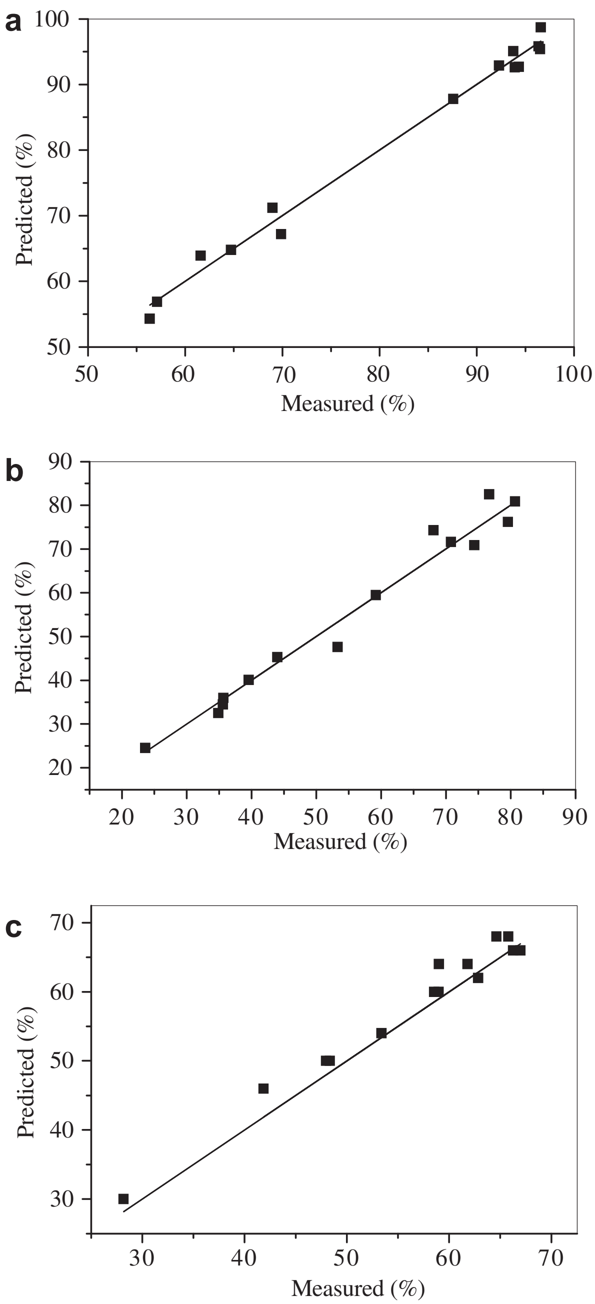

Fig. 2 – Relationship between the predicted and measured (a) turbidity removal efficiency; (b) lignin removal efficiency; and (c) water recovery efficiency.

of 5.67. Under the optimal conditions, the maximal turbidity removal efficiency was estimated to be 99.7%.

3.2. *Optimization for lignin removal*

Lignin is the main pollutant in the pulp mill wastewater. The efficient removal of lignin from wastewater by the coagulation-flocculation method is crucial to reclaim water and lignin. The following equation (Eq. (3)) is the regression model using the backward regression method with the results of the lignin removal efficiency in the coagulation-flocculation experiments:

$$Y = -45.3 + 35.6X_1 + 24.6X_2 + 8.2X_3 - 4.0X_1^2 - 2.4X_2^2 - 1.3X_3^2 - 1.1X_1X_2$$
$$R^2 = 0.889,\ F = 4.64 \quad (3)$$

The results of $F = 4.64 > F\,(0.05,7,6) = 4.207$ and $R^2 = 0.889$ for the lignin removal efficiency show that the second-order polynomial model was significant and fitted the experimental results well. Fig. 2b shows that the measured versus predicted plot values were distributed evenly near to the straight line. From Eq. (3), the optimal conditions for maximal efficiency of lignin removal were estimated to be: coagulant dosage of 1004 mg/L, flocculant dosage of 25.9 mg/L and pH 5.7. Under these conditions, the maximal lignin removal efficiency was estimated to be 84.1%.

3.3. *Optimization for water recovery efficiency*

The regression model to describe the water recovery efficiency of the coagulation-flocculation experiments was obtained using the total regression method (Eq. (4)).

$$Y = -15.3 - 4.0X_1 + 24.4X_2 + 22.4X_3 - 0.4X_1^2 - 2.4X_2^2 - 1.8X_3^2 + 1.0X_1X_2 + 0.8X_1X_3 - 2.4X_2X_3$$
$$R^2 = 0.935,\ F = 6.35 \quad (4)$$

The regression results, i.e., $F = 6.35 > F\,(0.05,9,4) = 5.999$ and $R^2 = 0.935$, show that the second-order polynomial model was significant and fitted the experimental results for the water recovery efficiency well. Again, the experimental values were distributed near to the straight line (Fig. 2c). From Eq. (4), the optimal conditions for maximal water recovery efficiency were estimated to be: coagulant dosage of 1040 mg/L, flocculant dosage of 20.6 mg/L and pH 8.17, under which the maximal water recovery efficiency was estimated to be 73.4%.

3.4. *Confirmation experimental results*

To confirm the validity of the statistical experimental strategies, additional confirmation experiments were conducted in duplicates. The chosen conditions for the coagulant dosage, flocculant dosage and pH are all listed in Table 4, along with the predicted and measured results. As shown in Table 4, the measured efficiencies of the supernatant turbidity removal, lignin removal and clean water recovery were close to the predicted values using their respective regression models. This demonstrates that the UD-RSM approach was appropriate for optimizing the operational conditions of the coagulation-flocculation process.

3.5. *Multiple-response optimization*

Removal efficiency of supernatant turbidity, removal efficiency of lignin and water recovery efficiency are three individual responses, and their optimizations were achieved under different optimal conditions. Thus, a compromise among the conditions for the three responses is desirable. The desirability function approach was used to achieve such a goal

Table 4 – Measured and calculated values for the confirmation experiments.

Run	Conditions	Parameter	Measured	Calculated
15	Coagulant dosage: 917 mg/L Flocculant dosage: 33.0 mg/L pH: 5.67	Turbidity removal (%)	99.6 ± 0.1	99.7
16	Coagulant dosage: 1004 mg/L Flocculant dosage: 25.9 mg/L pH: 5.7	Lignin removal (%)	88.4 ± 0.3	84.1
17	Coagulant dosage: 1040 mg/L Flocculant dosage: 20.6 mg/L pH: 8.17	Water recovery (%)	74.0 ± 2.0	73.4

(Zhang et al., 1998). The regression equation of the compromise was obtained as follows:

$$Y = 14.401 + 0.0918X_1 + 0.923X_2 + 11.084X_3 - 0.0000395X_1^2 - 0.0463X_2^2 - 0.818X_3^2 + 0.000401X_1X_2 - 0.00759X_1X_3 + 0.156X_2X_3 \quad (5)$$

The optimal conditions calculated from the regression equation were as follows: coagulant dosage of 871 mg/L, flocculant dosage of 22.3 mg/L and pH 8.35, respectively. The corresponding removal efficiency of turbidity, removal efficiency of lignin and water recovery efficiency were 95.7%, 83.4% and 72.7%, respectively. The overlay plot for the optimal region is presented in Fig. 3. The shaded portion gave the permissible values of the two variables by defining the desired limits of removal efficiency of supernatant turbidity, removal efficiency of lignin and water recovery efficiency.

A confirmation experiment under the compromised conditions was carried out in triplicates, and the average removal efficiencies of turbidity and lignin, and water recovery efficiency were obtained as 95.0%, 83.5% and 72.0%, respectively (Fig. S4 in Appendix). These results were in good agreement with the model predictions. After flocculation, the aluminum ion concentration in the supernatant was measured as 6.8 mg/L, and the final pH was 6.2.

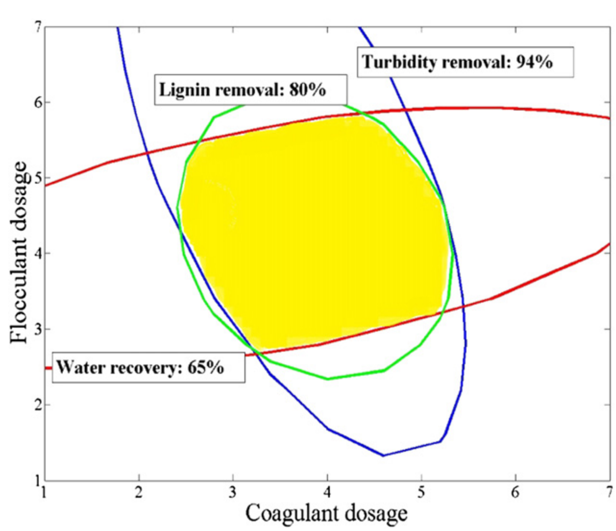

Fig. 3 – Overlay plot for the optimal region.

An optimal pH of 8.35 was predicted for multi-response optimization purpose, at which the optimal compromised removal efficiencies were obtained. However, it is worthwhile noting that, decent efficiencies can be obtained at neutral pH (the initial pH of the wastewater) based on our experiment results and the prediction by Eq. (5). Thus, in the practical application, this wastewater could be treated without pH adjustment when the treatment efficiency is fulfilled and the cost for pH adjustment is a big concern. This is a tradeoff between the efficiency of the process and the cost for pH adjustment.

4. Discussion

4.1. *Supernatant turbidity removal*

With the turbidity removal efficiency as the response, the response surfaces of the quadratic model with one variable kept at the optimal level and the other two varying within the experimental ranges are shown in Fig. 4. The obvious peak in the response surfaces indicates that the optimal conditions were exactly located inside the design boundary. In other words, there were significant interactive effects on turbidity between coagulant dosage and the flocculant dosage, coagulant dosage and pH, as well as flocculant dosage and pH.

The turbidity removal efficiency was high when the coagulant dosage and the flocculant dosage were within the range of 700–1400 mg/L and 16–48 mg/L, respectively, at the optimal pH 5.3 (Fig. 4b). Charge neutralization and sweep-floc were the two main mechanisms leading to the aggregation of particles in the coagulation process. In general, the appropriate pH for the charge neutralization in the coagulation process is in a range of 4.0–5.5 (Chang et al., 1993). When the aluminum ions is used as a coagulant, pH 6.0–8.0 is suitable for the formation of amorphous $Al(OH)_3$, which removes organic matters by adsorption on the precipitation of $Al(OH)_3(s)$ through the sweep-floc mechanism (Chang et al., 1993). Thus, pH 5.3 is favorable only for the charge neutralization. However, the acidic condition (pH 5.3) was in favor of the improvement of cationic charge density as well as the extension of the grafting chain in the solution. In this case, both the charge neutralization ability and the sweep-floc ability were improved. Taking into account the two factors, pH 5.3 was appropriate for the turbidity removal of the pulp mill wastewater.

Likewise, at the optimal flocculant dosage, the coagulant dosage was within the range of 350–1050 mg/L and pH was 2.0–8.5 (Fig. 4b). Under these conditions, the coagulant $AlCl_3$ exhibited good charge neutralization and sweep-floc abilities, and thus the flocculant, in addition to the two abilities above, had an improved adsorption bridging ability.

In addition, at the optimal coagulant dosage, the acidic condition and flocculant dosage within the range of 24–48 mg/L were favorable for the supernatant turbidity removal. As mentioned above, for a given coagulant dosage, the acidic condition was appropriate for the removal of turbidity with the graft copolymer used as the flocculant.

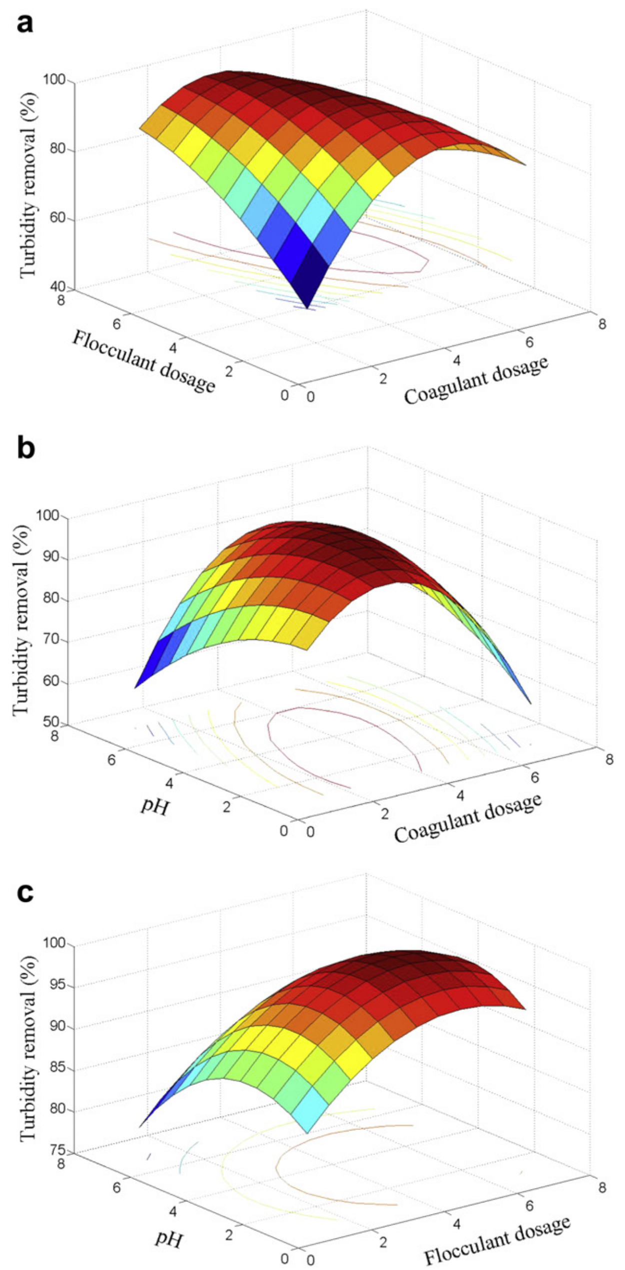

Fig. 4 – 3D surface graphs and contour plots of turbidity removal efficiency showing the effect of variables: (a) X_1–X_2; (b) X_1–X_3; and (c) X_2–X_3.

4.2. *Lignin removal*

When the lignin removal efficiency was selected as the response, the response surfaces of the quadratic model with one variable kept at the optimal level and the other two varying within the experimental ranges are illustrated in Fig. S2 (Appendix). The peak in the response surfaces indicates that the optimal conditions were exactly located inside the design boundary. Furthermore, there were significant interactive effects on turbidity between coagulant dosage and flocculant dosage, coagulant dosage and pH, as well as flocculant dosage and pH.

Under the experimental conditions, the lignin was indiscerptible and suspended in the wastewater as colloids (about 500 nm). Thus, the coagulation-flocculation method was appropriate for lignin removal. Acidic condition was in favor of the precipitation of lignin and thus its removal. However, the lignin removal efficiency was less than the turbidity removal efficiency under their respective optimal conditions. This is attributed to the fact that there is a great amount of soluble phenols in the wastewater and phenols could not be efficiently removed by the coagulation-flocculation method.

4.3. *Water recovery efficiency*

The response surfaces of the quadratic model with one variable kept at the optimal level and the other two varying within the experimental ranges, with the water recovery efficiency as the response, are shown in Fig. S3 (Appendix). The elliptical contour plots indicate that there were significant interactive effects between coagulant dosage and flocculant dosage, coagulant dosage and pH, as well as flocculant dosage and pH, even the water recovery efficiency percentage increased at the center of the three regions. This was evidenced by the obvious peak in the response surfaces, in which the optimal conditions were exactly located inside the design boundary.

In the coagulation-flocculation process, the coagulant was dispersed in the wastewater to destabilize the colloidal particles and the flocculant was used to agglomerate the destabilized colloidal particles into large particles and then precipitates. The flocculant used in our experiment was a modified natural polymer, which was synthesized by grafting two monomers onto starch backbone in order to improve its charge neutralization and bridging ability (Wang et al., 2009). The flexible grafting chain grafted onto the rigid starch backbone increased the chances for the flocculant to approach to the contaminant particles in the wastewater. Therefore, the graft copolymer used as the flocculant would conduce to the formation of larger and denser flocs, which are readily separated from the wastewater. As a result, better quality and higher water quantity could be obtained.

4.4. *Significance of the integrated UD-RSM approach*

A combination of UD and RSM applied in this work has a rational statistical basis, and is demonstrated to be a powerful approach for the optimization of the coagulation-flocculation process for pulp mill wastewater treatment in this study. With the UD method, the selected experimental points were distributed uniformly in the factor space for all the three key factors influencing the efficiency of this process, i.e., coagulant dosage, flocculant dosage and pH. This facilitated the acquisition of most response information through the fewest numbers of experiments. In addition, the application of number theory in the experimental design facilitates the computer statistical modeling and the subsequent regression analysis in the UD software, such as linear regression, non-linear regression and quadratic regression, etc. In addition, the number of the experimental trials in UD is determined only by the level of factors, not by the number of factors. The UD method can also be used when the levels of factors are different. These are the advantages of UD over other experiment design approaches.

A combination of UD and RSM provided a straightforward way to evaluate the individual effects and interactions of the experimental factors for desirable responses. Following the RSM optimization, a desirability function approach could be employed to obtain the compromise optimal conditions. This study demonstrates that this integrated approach could optimize the coagulation-flocculation process effectively using few data sets, which becomes very attractive for the processes where data are obtained costly. Thus, this integrated optimization approach can be useful for other complex wastewater treatment processes and multivariate systems in other fields.

5. Conclusions

A coagulation-flocculation process with aluminum chloride as the coagulant and a modified natural polymer, starch-*g*-PAM-*g*-PDMC [polyacrylamide and poly (2-methacryloyloxyethyl) trimethyl ammonium chloride], as the flocculant was employed for pulp mill wastewater treatment. A novel approach combined response surface methodology (RSM) and uniform design (UD) was used to optimize the process and evaluate the effects and interactions of three main influential factors, coagulant dosage, flocculant dosage and pH, on the treatment efficiency in terms of the supernatant turbidity and lignin removals as well as the water recovery. An optimal condition of coagulant dosage 871 mg/L, flocculant dosage 22.3 mg/L and pH 8.35 was obtained from the compromise of the three desirable responses, i.e. supernatant turbidity removal, lignin removal and water recovery efficiency. Further confirmation experiments demonstrated that such a combination of the UD and RSM is an effective and powerful approach for the optimization of the coagulation-flocculation process for pulp mill wastewater treatment.

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Appendix. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.watres.2011.08.023.

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