Available online at www.sciencedirect.com
ScienceDirect
Journal of Hazardous Materials 145 (2007) 162–168
Journal of Hazardous Materials
www.elsevier.com/locate/jhazmat

Optimization of coagulation–flocculation process for pulp and paper mill effluent by response surface methodological analysis

A.L. Ahmad [a,*], S.S. Wong [a], T.T. Teng [b], A. Zuhairi [a]

[a] School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Pulau Pinang, Malaysia
[b] School of Industrial Technology, Universiti Sains Malaysia, 11800 Minden, Pulau Pinang, Malaysia

Received 27 June 2006; received in revised form 6 November 2006; accepted 6 November 2006
Available online 10 November 2006

Abstract

Coagulation–flocculation is a proven technique for the treatment of high suspended solids wastewater. In this study, the central composite face-centered design (CCFD) and response surface methodology (RSM) have been applied to optimize two most important operating variables: coagulant dosage and pH, in the coagulation–flocculation process of pulp and paper mill wastewater treatment. The treated wastewater with high total suspended solids (TSS) removal, low SVI (sludge volume index) and high water recovery are the main objectives to be achieved through the coagulation–flocculation process. The effect of interactions between coagulant dosage and pH on the TSS removal and SVI are significant, whereas there is no interaction between coagulant dosage and water recovery. Quadratic models have been developed for the response variables, i.e. TSS removal, SVI and water recovery based on the high coefficient of determination (R^2) value of >0.99 obtained from the analysis of variances (ANOVA). The optimum conditions for coagulant dosage and pH are 1045 mg L^{-1} and 6.75, respectively, where 99% of TSS removal, SVI of 37 mL g^{-1} and 82% of water recovery can be obtained.

Keywords: Analysis of variances; Water recovery; Sludge volume index; Central composite design

1. Introduction

The pulp and paper mill is a major industrial sector utilizing a huge amount of lignocellulosic materials and water during the manufacturing process. It consumes as high as 60 m^3 of freshwater per ton of paper produced [1]. Common pollutants include suspended solids (SS), chemical oxygen demand (COD), biochemical oxygen demand (BOD), colour, basicity, heavy metals, alkali and alkaline earth metals, phenols, chloro-organics, cyanide, sulphides and other soluble substances [2]. The wastewater can cause considerable damage to the receiving waters if discharged untreated [3–5].

Currently there are 20 paper mills in Malaysia with the total paper production of 1.3 million metric tonnes in the year 2003 [6]. It is estimated that 88 m^3 wastewater is generated for each metric tonne of paper produced. Thus, in the year 2003, 114 million m^3 of wastewater was generated. This implies that the industry must have a high capacity wastewater treatment plant for the treatment of this huge volume of wastewater. The high capacity wastewater treatment plant is usually not economically feasible due to the high energy consumption and high maintenance cost. Today many mills are seeking to reduce operating costs, and water conservation is receiving careful consideration. Consequently, there is a need to reclaim and to recycle the treated wastewater in order to reduce the consumption of freshwater and to lower the capacity of the wastewater treatment plant.

Recently, a review done by Pokhrel and Viraraghavan [7] classified the treatment methods of pulp and paper mill wastewater into three major categories, physicochemical treatment, biological treatment and integrated treatment processes. All of the reported methods have their respective advantages, weaknesses and limitations.

Unlike freshwater, pulp and paper mills wastewater contains fibre and can cause unique solid/liquid separation challenges. Most solid/liquid separation systems have difficulty in operation when the requirements are produce high quality water, remove fine particles, operate continuously and remove high quantities

* Corresponding author. Tel.: +604 593 7788; fax: +604 594 1013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

0304-3894/$ – see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2006.11.008

of fibre. Chemical coagulation followed by sedimentation is a proven technique for the treatment of high suspended solids wastewater especially those formed by the colloidal matters. Research and practical applications have shown that coagulation will lower the pollution load and could generate an adequate water recovery [8–12]. As a result of the smaller load, the wastewater treatment plant might be designed more energy efficiently at a smaller footprint and with lower investment costs [13].

Coagulation is mainly done with inorganic metal salts, e.g. aluminum and ferric sulphates and chlorides. Polyelectrolytes of various structures, e.g. polyacrylamides, chitosan, polysaccharides, polyvinyl and many more are usually used as coagulant aids to increase the floc density in order to improve the rate of sedimentation. According to Aguilar et al. [14], anionic polyacrylamide when added to ferric sulphate or polyaluminium chloride led to a significant increase in the settling time. In the earlier work done by Stephenson and Duff [15], it was reported that the removal of total carbon, colour and turbidity of up to 88, 90 and 98%, respectively, were observed in the treatment of mechanical pulping effluent using ferric chloride, ferrous sulphate, aluminum chloride and aluminum sulphate. In the earlier work done [16], polyacrylamide was found to be effective for the coagulation–flocculation process of the pulp and paper mill effluent.

Aluminium and iron salts are widely used as coagulants in water and wastewater treatment and in some other applications. Their mode of action is generally explained in terms of two distinct mechanisms: charge neutralization of negatively charged colloids by cationic hydrolysis products and incorporation of impurities in an amorphous hydroxide precipitate so-called sweep flocculation [17]. The relative importance of these mechanisms depends on factors such as pH and coagulant dosage. These two factors were studied by using response surface methodology (RSM) to determine their optimum values and their effects on the treatment of pulp and paper mill wastewater.

Coagulation–flocculation optimization practices in many studies are still reliant, to a very large extent, performed on a trial and error basis using a conventional "change one factor at a time" method. This is an experimentation method in which a single factor is varied while all other factors are kept fixed at a specific set of conditions. The single-dimensional search is laborious, time consuming, and incapable of reaching the true optimum due to neglecting of the interaction among variables [18]. To resolve this problem, response surface methodology (RSM) has been proposed to determine the influences of individual factors and their interactive influences. RSM is a statistical technique for designing experiment, building models, evaluating the effects of several factors and searching optimum conditions for desirable responses and reducing number of experiments [19]. As reported recently, statistical experimental design has been applied by Bacaoui et al. [20] to optimize the preparation of activated carbons for use in water treatment. RSM uses an experimental design such as the central composite design (CCD) to fit a model by least squares technique [21]. Adequacy of the proposed model is then revealed using the diagnostic checking tests provided by analysis of variance (ANOVA). The response surface plots can be employed to study the surfaces and locate the optimum.

The present investigation aims at optimization of the coagulant dosages and pH to achieve highest removal of total suspended solids (TSS), lowest sludge volume index (SVI) and highest water recovery from pulp and paper mill wastewater using alum coupled with cationic polyacrylamide. The optimization is carried out via central composite face-centered (CCFD) RSM experimental design. The interaction between factors influencing TSS removal, SVI and water recovery is established and models describing the effect of the factors on TSS removal, SVI and water recovery are also described.

2. Materials and methods

The experiments were carried out in at laboratory bench scale using a jar test apparatus, and statistically designed experiments were used to optimize two most effective operating variables in the coagulation–flocculation process, namely the coagulant dosage and pH.

The wastewater was collected from the equalization tank of wastewater treatment plant of a paper mill in Penang, Malaysia. An industrial grade alum and coagulant aid with very high molecular weight and low charge density (Organopol 5415) were obtained from Sen Sen Chemicals Sdn. Bhd. and Ciba Speciality Chemicals, respectively. Jar test procedures were performed with the conventional jar apparatus (Stuart Science Flocculator model, SWI) using 500 mL wastewater samples. The coagulant was added in the range 800–1200 mg L^{-1} to the sample and the pH of the sample solution was adjusted in the range 6–8 by addition of H_2SO_4 (the initial pH of pH is 8–9). The coagulant aid of 1 mg L^{-1} was then added to the sample. The sample was stirred rapidly for a period of 2 min at 200 rpm. It was followed by a further slow mixing of 15 min at 40 rpm. The sample was allowed to settle for 30 min.

In all the tests, TSS, SVI and water recovery were measured as the responses. These parameters were measured based on the Standard Methods for the Examination of Water and Wastewater [22]. A pH meter model 320 (WTW, Germany) was used to measure the solution pH. The TSS concentration was determined by filtering a well-mixed sample through a glass fibre filter (Whatman 934AH) and then the residue retained on the filter were weighed after being dried in the oven at 103 °C for 60 min. The settled sludge volume was measured using a 1000 mL Imhoff cone and the SVI and water recovery were calculated accordingly using the value of the settled sludge volume.

The RSM used in the present study was a CCFD involving two different factors; coagulant dosage and pH. The CCFD contained a total of 13 experiments with the first 9 experiments organized in a factorial design with the experimental trials from 10 to 13 involving the replication of the central point. Repeated observations at the center point were used to estimate the experimental error employed. The ranges and the levels of the variables investigated in the study are given in Table 1. A complete set of the experimental design are shown in Table 2. Panel A in Table 2 shows the levels in terms of coded variables while Panel B represents the conditions used with original units of measurements.

Table 1
Experimental range and levels of the independent variables

Variables (factors)	Range and levels (coded)		
	−1	0	+1
Coagulant dosage, A (mg L^{-1})	800	1000	1200
pH, B	6	7	8

The quadratic equation model [21] for predicting the optimal point can be expressed according to Eq. (1):

$$Y = \beta_0 + \sum_{i=l}^{k} \beta_i X_i + \sum_{i=l}^{k} \beta_{ii} X_i^2 + \sum_{i \le j}^{k} \sum_{j}^{k} \beta_{ij} X_i X_j + \varepsilon \qquad (1)$$

where i and j are the linear and quadratic coefficients, respectively, β the regression coefficient, k the number of factors studied and optimized in the experiment and ε is the random error.

The Design Expert Software (version 6.0, Stat-Ease, Inc., Minneapolis, MN) was used for regression. Analysis of variances (ANOVA) was used for graphical analyses of the data to obtain the interaction between the process variables and the responses. The quality of the fit polynomial model was expressed by the coefficient of determination R^2, and its statistical significance was checked by the Fisher F-test in the same program. Model terms were selected or rejected based on the P value (probability) with 95% confidence level. Three-dimensional plots and their respective contour plots were obtained based on the effects of the levels of two factors (coagulant dosage and pH). From these three-dimensional plots, the simultaneous interaction of two factors on the responses was studied. The optimum region was also identified based on the main parameters in the overlay plot.

3. Results and discussion

Experiments according to the design in Table 2 were carried out and relevant results are shown in Table 3, which lists TSS removal, SVI and water recovery. The results are further analyzed using Design Expert Software. The relationship between two controllable factors (coagulant dosage and pH) and three important operating parameters (TSS removal, SVI and water recovery) for the coagulation–flocculation process is studied. Model terms that are significant are desired to obtain a good fit in a particular model. A CCFD shown in Table 2 allows the development of mathematical equations where each response variable Y is assessed as a function of coagulant dosage (A) and pH (B) and calculated as the sum of a constant, two first-order effects (terms in A and B), one interaction effect (AB) and two second-order effects (A^2 and B^2) according to Eq. (1). The results obtained are then analyzed by ANOVA to assess the "goodness of fit". Only terms found statistically significant are included in the model. The non-significant terms can be reduced by reselect only the significant terms to be included in the model. The model terms with "Prob > F > 0.5" will be eliminated from the model. All the model terms are found to be significant for the TSS removal and SVI except the A, AB and A^2 model terms show non-significant for water recovery. These results clearly show that pH plays an important role to generate high water recovery effluent in the treatment of pulp and paper mill wastewater by coagulation–flocculation process. Therefore, these model terms for water recovery are dropped from the model and then a new ANOVA is performed for the reduced model. The quadratic model is well fitted to the observed data and the following empirical models in terms of coded values are obtained for the TSS removal, SVI and water recovery, respectively:

Table 2
CCFD for the study of two experimental variables in coded units

Run no.	Panel A		Panel B	
	Coagulant dosage, A	pH, B	Coagulant dosage, A (mg L^{-1})	pH, B
1	−1	−1	800.00	6.00
2	1	−1	1200.00	6.00
3	−1	1	800.00	8.00
4	1	1	1200.00	8.00
5	−1	0	800.00	7.00
6	1	0	1200.00	7.00
7	0	−1	1000.00	6.00
8	0	1	1000.00	8.00
9	0	0	1000.00	7.00
10	0	0	1000.00	7.00
11	0	0	1000.00	7.00
12	0	0	1000.00	7.00
13	0	0	1000.00	7.00

Table 3
Experimental results

Run no.	TSS removal (%)	SVI (mL g^{-1})	Water recovery (%)
1	94.8	64.0	76.4
2	98.8	39.4	80.5
3	99.4	38.5	81.9
4	95.9	61.3	82.6
5	95.4	62.2	80.2
6	99.2	38.5	82.2
7	99.2	38.5	82.2
8	99.2	38.2	82.3
9	98.0	40.1	76.3
10	99.2	38.5	82.2
11	98.0	38.5	82.2
12	96.9	39.9	76.0
13	97.6	39.5	80.3

- TSS removal:

$$Y_1 = 99.23 + 1.07A - 0.35B - 2.26A^2 - 0.81B^2 - 0.025AB$$

- SVI:

$$Y_2 = 38.41 - 11.60A + 0.48B + 11.55A^2 + 1.40B^2 - 0.35AB$$

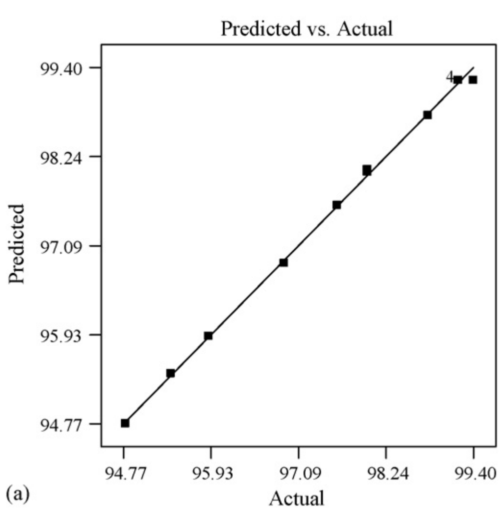
- Water recovery:

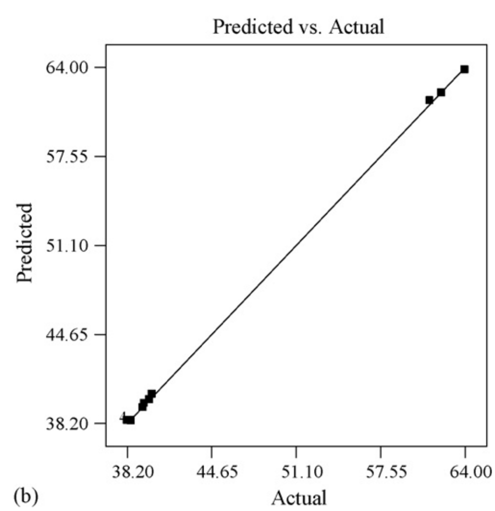
$$Y_3 = 82.23 - 2.05B - 3.95B^2$$

Table 4
Statistical parameters obtained from the ANOVA for the reduced models

Variable	TSS removal	SVI	Water recovery
Significant terms	A, B, AB, A^2, B^2	A, B, AB, A^2, B^2	B, B^2
R^2	0.9986	0.9998	0.9949
R^2 adjusted	0.9976	0.9996	0.9939
Prob > F	<0.0001	<0.0001	<0.0001
Adequate precision	83.5020	190.5510	63.3890
Standard deviation, S.D.	0.0790	0.2000	0.2000
Coefficient of variance, CV	0.0800	0.4400	0.2400
PRESS	0.1500	2.0400	0.6500
Probability for lack of fit	0.7202	0.1206	0.2406

Statistical parameters obtained from the ANOVA for the reduced model of the responses are given in Table 4. ANOVA results of the quadratic models with Prob > F < 0.0001 for all the responses presented in Table 4 indicate that the model equation adequately describes the response surfaces of TSS removal, SVI and water recovery in the interval of investigation. The effect of each variable on the response is the combination of coefficients and variable values as well as contribution of joint effect of variables that cannot be observed by conventional experimental methods. The high R^2 value, close to 1, is desirable and the predicted R^2 must be in reasonable agreement with the adjusted R^2 for a significant model [23]. The values of R^2 for TSS removal, SVI and water recovery are 0.9986, 0.9998 and 0.9949, respectively. In this case it indicates that only 0.02–0.51% of the total variation is not explained by the model. The values of the adjusted R^2 of 0.9976, 0.9996 and 0.9939, respectively, for TSS





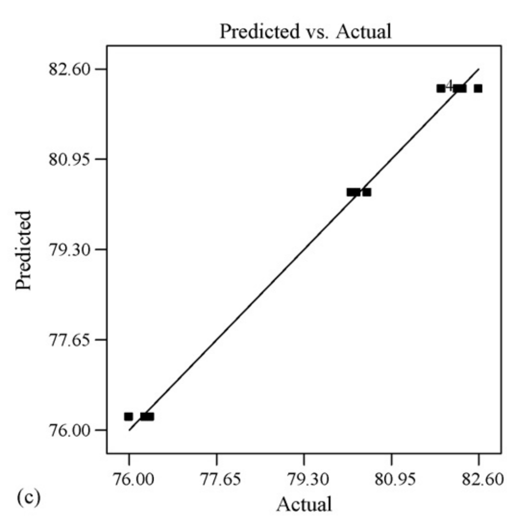


Fig. 1. Design–expert plot. Predicted vs. actual values plot for: (a) TSS removal; (b) SVI; (c) water recovery.

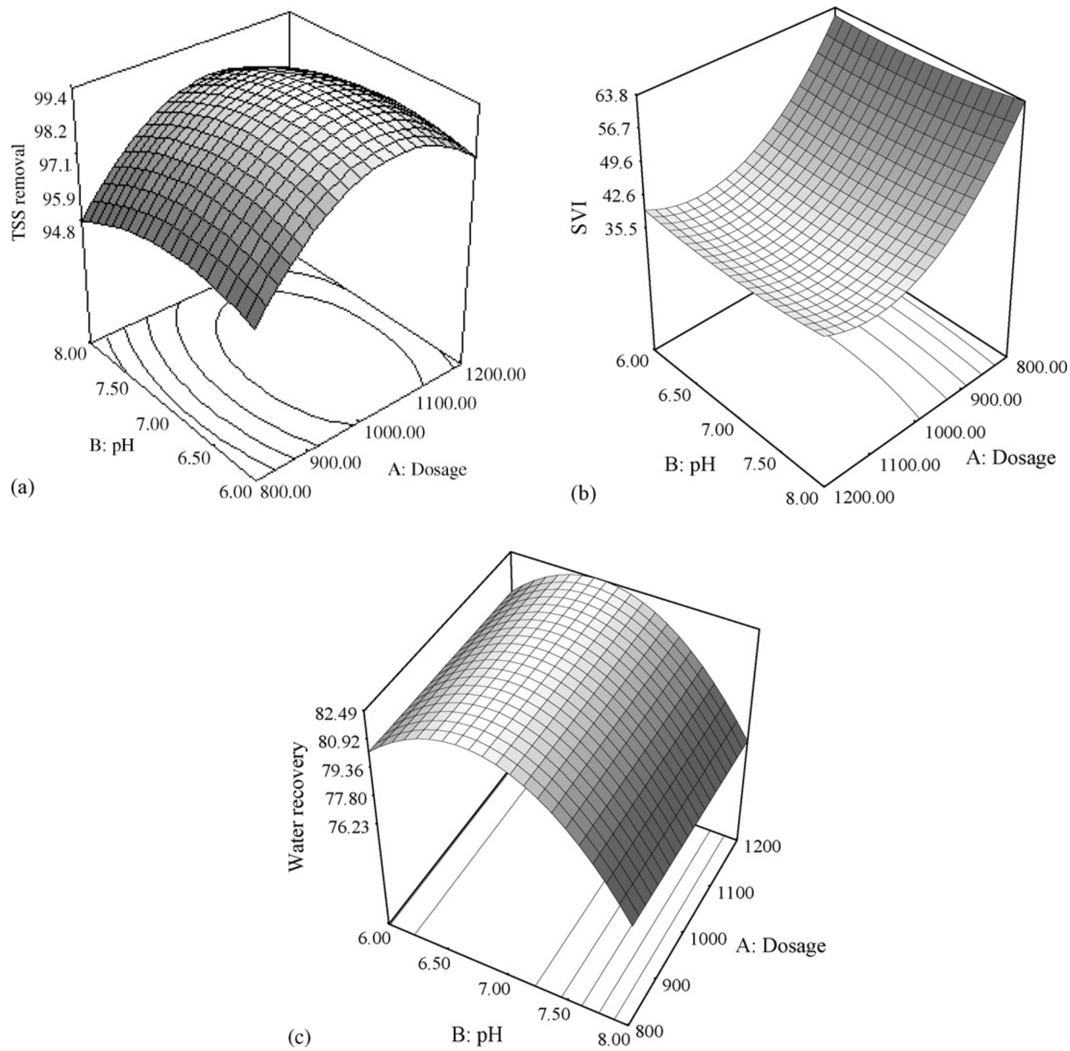


Fig. 2. Design–expert plot. Response surface plot for: (a) TSS removal; (b) SVI; (c) water recovery.

removal, SVI and water recovery, are also high to advocate a high significance of the model [19,24]. The CV as the ratio of the standard error of estimate to the mean value of the observed response (as a percentage) is a measure of reproducibility of the model and as a general rule a model can be considered reasonably reproducible if its CV is not greater than 10% [25]. The CV values obtained for all responses studied are relatively small with none of them exceeding 1% as given in Table 4. This result is reasonable since coagulation–flocculation process is a physicochemical method in which it is more subject to control by simple techniques.

Usually it is necessary to check the fitted model to ensure that it provides an adequate approximation to the real system. Unless the model shows an adequate fit, proceeding with investigation and optimization of the fitted response surface is likely to give poor or misleading results. By applying the diagnostic plots such as the predicted versus actual values plot, the model adequacy can be judged. The predicted versus actual value plot of TSS removal, SVI and water recovery are shown in Fig. 1. The second-order regression model obtained for the operating variables of TSS removal, SVI and water recovery are satisfied as the predicted versus actual value plot approximates along a straight line as shown in Fig. 1.

The response surface plot for TSS removal is shown in Fig. 2(a). The response surface of TSS removal shows a clear peak, suggesting that the optimum condition for maximum TSS removal is well inside the design boundary. There is a clear elongated hill running along the pH axis on the plot of the three-dimensional response surfaces of the quadratic model for the TSS removal. As can be see from Fig. 2(a), the maximum percentage TSS removal of more than 99% is achieved at the coagulant dosage of 1000–1050 mg L^{-1} and between pH 6 and 7. The removal of TSS will decrease with further increase of the coagulant dosage and pH beyond the optimum condition.

Fig. 2(b) shows the response surface for SVI. The curvilinear profile obtained for SVI is in accordance to the quadratic model. The SVI significantly decreases with coagulant dosage and pH. However, the two-dimensional contour plot shows nearly straight line along the pH axis revealing that the pH has little influence on the SVI. The SVI is affected mostly by coagulant dosage. These results observed from the response surface plot are in good agreement with the fitted model for SVI obtained

Table 5
Confirmation experiments for optimum region

Conditions	Responses		
	TSS removal (%)	SVI (mL g^{-1})	Water recovery (%)
Coagulant dosage = 1010 mg L^{-1} (pH 6.5)			
Experimental value	98.82	40.31	81.27
Model response with Cl 95%	99.25	37.98	82.27
Error	−0.43	2.33	−1.00
Coagulant dosage = 1025 mg L^{-1} (pH 7.0)			
Experimental value	99.27	38.91	82.37
Model response with Cl 95%	99.33	37.14	82.23
Error	−0.06	1.77	0.14

earlier. The relative contribution of each factor to each dependent variable (Y_1 to Y_3: TSS removal, SVI and water recovery) was directly measured by the respective coefficient in the fitted model. A positive sign for the regression coefficient (β_2) in the fitted model for SVI, indicates that the ability of the system to achieve low SVI decreases with increase in pH value (B). The SVI is the volume of 1 g of sludge after 30 min settling. A value of 100 mL g^{-1} or less is considered a good settling sludge. Thus, SVI values below 100 are desired to assure that the sludges produced in the coagulation–flocculation process have sufficient settling characteristics. The lowest SVI can be obtained at the coagulant dosage of 1000–1050 mg L^{-1} and pH value of 6–7 as shown in Fig. 2(b).

Fig. 2(c) shows the response surface for water recovery. A clear straight line can be seen along the coagulant dosage axis as shown in the two-dimensional contour plot. This result indicates that the coagulant dosage has no significant effect on the water recovery. The result is in collaboration with the ANOVA results obtained when fitting the model terms for water recovery. The first-order model term of coagulant dosage, the second model term of coagulant dosage and the interaction between coagulant dosage and pH are found to be non-significant and have been eliminated from the model. The maximum percentage water recovery of 82.5% can be obtained at the pH range of 6.5–7 as presented in Fig. 2(c).

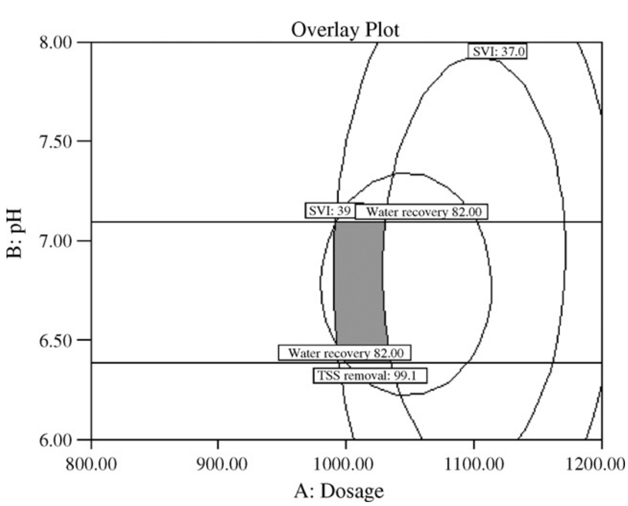


Fig. 3. Design–expert plot. Overlay plot for optimal region.

The optimum condition can be visualized graphically by superimposing the contours for the various response surfaces in an overlay plot. By defining the limits of the TSS removal, SVI and water recovery desired, the shaded portion of the overlay plot, as shown in Fig. 3, defines the permissible values of the dependent variables. The optimum region is made by considering TSS removal and water recovery greater and SVI less than the values mentioned in the overlay plot. Based on the overlay plot, the optimum conditions for coagulant dosage and pH are 1045 mg L^{-1} and 6.75, respectively. A confirmation of the results applying the coagulant dosage and pH for those fall in the optimum region is accomplished by repeating two additional experiments. As shown in Table 5, the TSS removal, SVI and water recovery obtained experimentally are closed to those estimated using the model.

4. Conclusion

Optimization of coagulation–flocculation process with respect to TSS removal, SVI and water recovery for the treatment of pulp and paper mill wastewater has been investigated. Response surface methodology using CCFD was applied to determine the optimum operating conditions for maximum TSS removal, lowest SVI and maximum water recovery. The coagulant dosage and pH are both significant terms to yield higher removal of TSS and minimum SVI. Coagulant dosage is not an important factor influencing water recovery. The reduced quadratic models developed using RSM for TSS removal, SVI and water recovery can be used for prediction within the ranges of the factors investigated. By applying RSM, the optimum region for the coagulation–flocculation process operation is located. The optimum conditions for coagulant dosage and pH are 1045 mg L^{-1} and 6.75, respectively, where 99% of TSS removal, SVI of 37 mL g^{-1} and 82% of water recovery can be obtained.

Acknowledgment

The authors would like to acknowledge the financial support from the Graduate Assistant Scheme of Universiti Sains Malaysia.

References

[1] G. Thompson, J. Swain, M. Kay, C.F. Forster, The treatment of pulp and paper mill effluent: a review, Bioresour. Technol. 77 (2001) 275–286.

[2] J.D. Achoka, The efficiency of oxidation ponds at the Kraft pulp and paper mill at Webuye in Kenya, Water. Res. 36 (2002) 1203–1212.

[3] M. Ali, T.R. Sreekrishnan, Aquatic toxicity from pulp and paper mill effluents: a review, Adv. Environ. Res. 5 (2001) 175–196.

[4] S. Lacorte, A. Latorre, D. Barceló, A. Rigol, A. Malmqvist, T. Welander, Organic compound in paper-mill process waters and effluents, Trends Anal. Chem. 22 (2003) 725–737.

[5] D. Berryman, F. Houde, C. DeBlois, M. O'Shea, Nonylphenolic compounds in drinking and surface waters downstream of treated textile and pulp and paper effluents: a survey and preliminary assessment of their potential effects on public health and aquatic life, Chemosphere 56 (2004) 247–255.

[6] A.S. Masut, The status and market information for pulp and paper industry in Malaysia for 2003, Malaysia Pulp Paper Manuf. Assoc. (2004) 1–30.

[7] D. Pokhrel, T. Viraraghavan, Treatment of pulp and paper mill wastewater—a review, Sci. Tot. Environ. 333 (2004) 37–58.

[8] M.H. Al-Malack, N.S. Abuzaid, A.H. El-Mubarak, Coagulation of polymeric wastewater discharged by a chemical factory, Water Res. 33 (1999) 521–529.

[9] M.I. Aguilar, J. Saez, M. Llorens, A. Soler, J.F. Ortuno, Nutrient removal and sludge production in the coagulation–flocculation process, Water Res. 36 (2002) 2910–2919.

[10] P.K. Holt, G.W. Barton, M. Wark, C.A.A. Mitchell, Quantitative comparison between chemical dosing and electrocoagulation, Colloids Surf. A 211 (2002) 233–248.

[11] D. Georgiou, A. Aivazidis, J. Hatiras, K. Gimouhopoulos, Treatment of cotton textile wastewater using lime and ferrous sulfate, Water Res. 37 (2003) 2248–2250.

[12] N.Z. Al-Mutairi, M.F. Hamoda, I. Al-Ghusain, Coagulant selection and sludge conditioning in a slaughterhouse wastewater treatment plant, Bioresour. Technol. 95 (2004) 115–119.

[13] A.F. Van Nieuwenhuijzen, A.R. Mels, P. Piekema, C.E. Brandt, C.A. Uijterlinde, The fate of particles and flocculants in pre-sedimentation tanks, in: Chemical Water and Wastewater Treatment VIII, IWA Publishing, London, 2004, pp. 49–57.

[14] M.I. Aguilar, J. Saez, M. Llorens, A. Soler, J.F. Ortuno, V. Meseguer, A. Fuentes, Improvement of coagulation–flocculation process using anionic polyacrylamide as coagulant aid, Chemosphere 58 (2005) 47–56.

[15] R.J. Stephenson, S.J.B. Duff, Coagulation and precipitation of a mechanical pulping effluent. I. Removal of carbon, colour and turbidity, Water Res. 30 (1996) 781–792.

[16] S.S. Wong, T.T. Teng, A.L. Ahmad, A. Zuhairi, G. Najafpour, Treatment of pulp and paper mill wastewater by polyacrylamide (PAM) in polymer induced flocculation, J. Hazard. Mater. 135 (2006) 378–388.

[17] J. Duan, J. Gregory, Coagulation by hydrolyzing metal salts, Adv. Colloid Interface 100–102 (2003) 475–502.

[18] R.L. Mason, R.F. Gunst, J.L. Hess, Statistical Design and Analysis of Experiments with Applications to Engineering and Science, 2nd ed., John Wiley and Sons, New York, 2003.

[19] A.I. Khuri, J.A. Cornell, Response Surfaces, Design and Analyses, 2nd ed., Marcel Dekker Inc., New York, 1996.

[20] A. Bacaoui, A. Dahbi, A. Yaacoubi, C. Bennouna, F.J. Maldonado-Hodar, J. Rivera-Utrilla, F. Carrasco-Marin, C. Moreno-Castilla, Experimental design to optimize preparation of activated carbons for use in water treatment, Environ. Sci. Technol. 36 (2002) 3844–3849.

[21] D.C. Montgomery, Design and Analysis of Experiments, 5th ed., John Wiley and Sons, New York, 2001.

[22] APHA, Standard Methods for the Examination of Water and Wastewater, 20th ed., American Public Health Association, Washington, DC, 2000.

[23] M.Y. Nordin, V.C. Venkatesh, S. Sharif, S. Elting, A. Abdullah, Application of response surface methodology in describing the performance of coated carbide tools when turning AISI 104 steel, J. Mater. Process. Technol. 145 (2004) 46–58.

[24] A.L. Ahmad, S. Ismail, S. Bhatia, Optimization of coagulation–flocculation process for palm oil mill effluent using response surface methodology, Environ. Sci. Technol. 39 (2005) 2828–2834.

[25] M. Ahmadi, F. Vahabzadeh, B. Bonakdarpour, E. Mofarrah, M. Mehranian, Application of the central composite design and response surface methodology to the advanced treatment of olive oil processing wastewater using Fenton's peroxidation, J. Hazard. Mater. 123 (2005) 187–195.