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Full Length Research Paper

Optimization of coagulation-flocculation process for printing ink industrial wastewater treatment using response surface methodology

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A coagulation–flocculation process was used to treat water-based printing ink wastewater with aluminum sulphate {$Al_2(SO_4)_3$} as coagulant and with Praestol as flocculant. To minimize turbidity and sludge volume index (SVI), the experiments were carried out using jar tests and response surface methodology (RSM) was applied to optimize this process. A central composite design, which is the standard design of RSM, was used to evaluate the effects and interactions of three factors, that is, coagulant dosage, flocculant dosage and pH on the treatment efficiency. The optimal conditions obtained from the desirable response, chemical oxygen demand (COD) removal, were coagulant dosage of 8250 mgl^{-1}, flocculant dosage of 80 mgl^{-1} and pH 7.25, respectively. The RSM was demonstrated as an appropriate approach for the optimization of the coagulation–flocculation process by confirmation experiments.

Key words: Printing ink industry effluent, chemical oxygen demand (COD) removal, response surface methodology, coagulation-flocculation process.

INTRODUCTION

With the rapid development of industries in Tunisia, the swift increase of variety and dosage of industrial wastewater leads to the increasing complexity of components in effluent and makes polluted water more difficult to be treated than before. For instance, wastewater has the characteristics of large discharge, difficult decolorization and high concentration of organic compounds in biological treatment effluent (Khelifi et al., 2009; Khannous and Gharsallah, 2010). Moreover, most effluent from biochemical process contain lots of toxic and hazardous non-biodegradable substances which have aggregate stability after discharging into water bodies, resulting in potential biological hazards. Therefore, advanced treatment by coagulation-flocculation in treating wastewaters containing complex impurities has a great advantage (Ying et al., 2011; Simate et al., 2012), in which the investigation on multifunction and diversity of coagulants-flocculants has become the focus in the field of coagulation-flocculation process (Ying et al., 2011).

Coagulation–flocculation is widely used for wastewater treatment, as it is efficient and simple to operate (Walsh et al., 2009; Ahmad et al., 2007). In this process, many factors can influence its efficiency, such as the type and dosage of coagulant/flocculant (Nandy et al., 2003), pH (Dominguez et al., 2005), mixing speed and time (Gurse et al., 2003), temperature and retention time (Zhu et al., 2004), etc.

The optimization of these factors may significantly increase the process efficiency. In conventional multifactor experiments, optimization is usually carried out by varying a single factor while keeping all other factors fixed

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Abbreviations: RSM, Response surface methodology; **COD,** chemical oxygen demand; **TSS,** total suspended solid; **VS,** volatile solid; **TS,** total solid; **BOD,** biological oxygen demand; **CCD,** central composite design.

at a specific set of conditions. It is not only time-consuming, but also usually incapable of reaching the true optimum due to ignoring the interactions among variables. Response surface methodology (RSM) has been proposed to determine the influences of individual factors and their interactions.

RSM is a statistical technique for designing experiments, building models, evaluating the effects of several factors, and searching optimum conditions for desirable responses. With RSM, the interactions of possible influencing factors on treatment efficiency can be evaluated with a limited number of planned experiments. RSM is widely used in various fields, for example, preparation of chitosan from beet molasses (dos Santos et al., 2005), synthesis of *N*-carboxybutylchitosan (Ravikumar et al., 2005), and radiolytic degradation of poly-vinyl alcohol (dos Santos et al., 2005). This method has been therefore widely used for the optimization of various processes in food industry, pharmaceuticals industry, material manufacturing, in biotechnology (Granato et al., 2010; Li et al., 2012; Singh et al., 2010) and in environmental science (Xue et al., 2012).

In this study, the RSM was employed for designing the coagulation-flocculation experiments to optimize the factors. The experiments were carried out by jar test which is usually employed to evaluate the treatment process efficiency. In the jar tests, coagulant dosage, flocculant dosage and pH were the factors that needed to be optimized.

The UNIPACK Paper Cardboard Packing Society from Sfax, Tunisia produces wastewater with high concentrations of organic and inorganic compounds that are not completely removed by preliminary treatments or biological process. The high organic and inorganic load (chemical oxygen demand (COD) more than 20000 mgl^{-1}) of UNIPACK industry represents a major environmental problem. With the establishment of more stringent regulations concerning wastewater treatment, there has been a growing interest in the development of new technologies and procedures for the decontamination of water (de Heredia et al., 2004).

Chemical coagulation followed by sedimentation is a proven technique for the treatment of high suspended solids wastewater especially those formed by the colloiddal matters. Research and practical applications have shown that coagulation will lower the pollution load and could generate an adequate water recovery (Ahmad et al., 2007). The main objective of this work was to optimize the coagulation–flocculation process in order to treat UNIPACK wastewater and investigate the interacttive effects of experimental factors, including coagulant dosage, flocculant dosage and pH. For this purpose, a printing ink wastewater was selected as the target to be treated by the coagulation–flocculation process which was optimized by RSM. The COD removal percentage of treated water was chosen as the dependent output variable. The compromise optimal conditions for the response were also obtained using the desirability function approach.

MATERIALS AND METHODS

Printing ink effluent and physicochemical characteristics determination

The wastewater used in this study was taken from a card board packing industry located in Sfax, (southern of Tunisia). This effluent present a dark gray color mainly caused by the solid ink residue. The industry implanted a wastewater treatment station in 1985. Wastewater treatment plant consisted of a feed tank where industrial effluents are collected and pumped to the coagulation /flocculation tank. The treated effluent was finally collected in decantation tank. Samples were collected within about two months from February to March, 2011. Samples were collected and preserved according to the Standard Methods for the Examination of Water and Wastewater (APHA, 1992).

Physicochemical analyses, for example, pH, total suspended solid (TSS), volatile solid (VS), total solid (TS), turbidity, biological oxygen demand (BOD), and COD of effluent, were carried out immediately after samples arrived in the laboratory according to the standard methods (APHA, 1992). The pH (precision 0.01) and temperature (precision 0.01) of the UNIPACK effluents were measured using a multifunction pH-meter Testo model (ECOMETP25-Korea South). Color of the effluent was noted by visual observation. The remaining effluent characteristics parameter, that is, TSS, VS, TS, turbidity, BOD, and COD were analyzed by volumetrically /titrimetrically as per standard methods (APHA, 1992; Manivasakam, 1996; Trivedi and Goel, 1984) (Table 1), respectively.

The physicochemical analyses were performed in triplicate. The results were expressed as mean value (MV) ± error deviation (ED). The coefficient of variation was calculated as:

$$CV = \frac{ED}{MV} \times 100$$

Before realizing the optimization of the coagulation-flocculation process, an evaluation of the functioning of the actual effluent treatment station during one month was performed. Physicochemical analyses were performed upstream and downstream of the wastewater treatment plant (feed tank and decantation tank). For each analysis, two samples were taken each week during a month from the feed tank and out of the decantation tank. The average values of physicochemical parameters of wastewater recorded during a month were calculated on the eight measurements (2 samples × 4 weeks). This investigation allows the variability of physicochemical characterization of the wastewater before and after the coagulation /flocculation step.

Coagulation-flocculation experiments

Industrial grade aluminum sulfate {$Al_2(SO_4)_3$ $14H_2O$} and praestol® 2500 TR (high molecular, non-ionic polyelectrolyte based on polyacrylamide, density : approx. 650 kg/m^3, viscosity in tap water: 50 mPa*s) were used as the coagulant and as flocculant, respectively. The chemical compounds were obtained from Fisher Scientific UKltd. They were prepared by dissolving powder with distilled water. The coagulation-flocculation experiments were carried out using the jar test method in 500 ml beakers. After the coagulant (stock solution of 50.0 gl^{-1}) was added with a dosage varying from

Table 1. Characterization of industrial wastewaters.

Industrial wastewater	Upstream of the treatment station		Downstream of the treatment station	
	Average value	CV (%)	Average value	CV (%)
pH	7.79 ± 0.79	10.21	6.27 ± 0.43	6.97
COD_s (mgl^{-1})	24046 ± 13306	55.33	944 ± 506.98	53.65
COD_t (mgl^{-1})	38595 ± 20926	54.21	2344.73 ± 1098.45	46.84
BOD_5 (mgl^{-1})	9750 ± 3126	32.06	575 ± 373.83	65.01
TSS (mgl^{-1})	4228 ± 2600	61.50	916.33 ± 848.11	92.555
TS (mgl^{-1})	27555 ± 11488	41.69	5705 ± 3388.07	59.38
VS (mgl^{-1})	16840 ± 9574	56.85	3401.66 ± 3500.24	102.89
COD_t/BOD_5	3.74 ± 1.06	28.39	4.84 ± 2.50	51.69

Table 2. Levels of the variable tested in the 2^3 central composite designs.

Variable	Range and level				
	-2	-1	0	1	2
X1, coagulant dosage (mg l^{-1})	6510	6910	7500	8090	8490
X2, flocculant dosage (mg l^{-1})	60	70	80	90	100
X3, pH	6.00	6.61	7.50	8.39	9.00

6510 to 8490 mgl^{-1}, the solution pH was adjusted to 6.0 to 9.0 by adding 0.1 M hydrochloric acid or 0.1 M sodium hydroxide solutions. Then, the flocculant at a concentration of 1.0 gl^{-1} was added with a dosage varying from 60 to 100 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 of the supernatant.

Experimental design and data analysis

The central composite design (CCD), with nine replicates at the central points was employed to fit a second-order polynomial model and to obtain an experimental error. The CCD was applied with three design factors and three levels. The factors are the coagulant dosage (X_1), the flocculant dosage (X_2) and pH (X_3). The coded levels and the natural values of the factors set in this statistical experiment are shown in Table 2. The central values (zero level) chosen for the experimental design were as follows: Coagulant dosage, 7500 mgl^{-1}; flocculant dosage, 80 mgl^{-1}; and pH, 7.50.

COD removal percentage was selected as the dependent variable. The response variable was fitted by a second-order model in the form of quadratic polynomial equation:

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

Where, y is the response variable to be modeled; X_i, and X_j the independent variables which influence y; 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 used in this work is presented in Table 3.

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 F-test. Model terms were selected or rejected based on the P value (probability) with 99% confidence level. Three-dimensional plots and their respective contour plots were obtained based on the effects of the levels of two factors.

The experimental design, regression and statistical analysis were performed, by *NemrodW®* software (Mathieu et al., 2000). The optimum values of selected variables were obtained by using the desirability function available in *NemrodW®* and also by analyzing the response surface contour plots. 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.

RESULTS AND DISCUSSION

Evaluation of the physicochemical characteristics of the industrial wastewater

The physicochemical characteristics of the UNIPACK effluent showed dark brown in color and is against the standard limit. This may be due to the presence of pigments, binders, carriers and additives (Ma and Xia, 2009). It is noting that the color of the effluent is pH sensitive and the effect of the pH on the color of the effluent is reversible (Devi et al., 2011; Siala et al., 2004). Table 1 shows the pH values of the wastewater measured on the upstream and decantation tanks. The pH values vary from 6.7 to 9.04 in the upstream tank.

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

Run	Factor			Response
	Coagulant dosage (X1)	Flocculant dosage (X2)	pH (X3)	COD removal percentage (Y1)
1	-1	-1	-1	96.90
2	1	-1	-1	97.10
3	-1	1	-1	96.80
4	1	1	-1	96.20
5	-1	-1	1	88.50
6	1	-1	1	96.20
7	-1	1	1	83.90
8	1	1	1	96.90
9	-2	0	0	95.50
10	2	0	0	96.00
11	0	-2	0	90.30
12	0	2	0	95.30
13	0	0	-2	86.00
14	0	0	2	77.10
15	0	0	0	96.90
16	0	0	0	97.10
17	0	0	0	97.00
18	0	0	0	96.90
19	0	0	0	97.00
20	0	0	0	97.10
21	0	0	0	96.90
22	0	0	0	96.90
23	0	0	0	97.00

This could be attributed to the variability of chemical composition of the water discharged. Whereas the treated wastewater collected out of the decantation tank presents a pH value ranging from 5.6 to 6.72. The pH values are sometimes out of the ranges fixed by legislative standards (pH from 5.6 to 5.95) and this variability could be attributed to the alkalinisation step which is arbitrarily realized.

The wastewater collected from the upstream of the station presents a COD value varying from 1630 and 6830 mgl^{-1} (CV of 61.5%) (Table1). The high organic concentration of the effluent is essentially due to the presence of pigments during washing pink water process. The COD value measured downstream of the treatment station varied from 1218 to 4042 mgl^{-1} (CV 46.8%) indicating that the organic charge decreased. This value is not conformed to legislative values. The mean values of COD soluble measured on the feed tank and decantation tank were respectively about 24046.5 ± 13306.4 mgl^{-1} and 944.8 ± 506.9 mgl^{-1} with a COD_s/COD_t ratio of 0.04 (Table 1). The high value of the COD was due to TSS present in the wastewaters.

The BOD5 value measured on the downstream of the treatment plant (Table 1) was about 575 ± 373 mg O_2. This value is already higher than the limit standard (400 mgl^{-1}). The high BOD and COD values of the UNIPACK effluents suggest presence of organic and inorganic pollutants in higher quantities (Pandey et al., 2003). Although, the COD/BOD_5 ratio is relatively not constant and fluctuates from 2.4 to 4.9 indicating that the effluent is hardly biodegradable.

The TSS measured on feed tank was about 4228 ± 2600 mgl^{-1} with a CV of 61.5% and reached a value of 100 mgl^{-1} after the treatment station due to the preliminary treatments (screen, sand removal, etc). The TS values ranged from 27555 ± 11488 mgl^{-1} to 5705 ± 3390 mg l^{-1}, upstream and downstream of the station, respectively (Table 1). The VS was about 16840 ± 9574 mgl^{-1} (CV 56.85%) upstream of the station and 3402 ± 3500 mg/l downstream of the station indicating that the TS has an inorganic character.

The analysis of physicochemical parameters of the treated and untreated effluent showed a significant variability of some parameters (Table 1) and particularly the COD (CV = 54.21%). The industrial station of wastewater treatment did not function adequately and the coagulation /flocculation process should be optimized.

A preliminary study on the effect of type and dosage of coagulant, flocculant, mixing speed, time, temperature and pH on the coagulation-flocculation process was

Table 4. Coefficients values and statistical parameters obtained for the model.

Coefficient	Coefficient value	Standard deviation	P-value
b0	96.882	0.028	< 0.01
b1	1.548	0.023	< 0.01
b2	0.257	0.023	< 0.01
b3	-2.670	0.023	< 0.01
b11	0.489	0.021	< 0.01
b22	-0.554	0.021	< 0.01
b33	-4.532	0.021	< 0.01
b12	0.563	0.029	< 0.01
b13	2.638	0.029	< 0.01
b23	-0.362	0.029	< 0.01

Table 5. Statistical parameters obtained from the ANOVA test performed for the model.

Source of variation	Sum of square (SS)	Degree of freedom (ddl)	Average square	Fisher number	Signification	R^2
Regression	525.2591	9	58.3621	7.4736	4.19***	0.838
Residues	101.5183	13	7.8091	-	-	-

carried out in order to determine the most critical factors and their regions of interest.

An experimental study conducted by Adhoum and Monser (2004) showed that the highest COD and color removal efficiencies have been obtained in acidic medium, at pH values in the limits of 4.0 to 6.0. However, very poor removals are found either at low (<2.0) or high (>10) pH. This behavior is attributed to the fact that the amphoteric character of the coagulant used does not precipitate at pH less than 2.0. Therefore, the aim of this study was to optimize pH, coagulant dosage and flocculant dosage in order to keep the optimum of COD removal using statistical analysis.

Analysis of the COD removal efficiency

The RSM have several classes of designs, with their own properties and characteristics. Central composite design (CCD), box-Benkhen design and three-level factorial design are the most popular designs applied by the researchers. The CCD was used to study the effects of the variables towards their responses and subsequently in the optimization studies (Montgomery, 2001). Experiments according to the design were carried out and relevant results are shown in Table 3, which lists COD removal.

Mathematical model and ANOVA analysis

$Y = 96.88 + 1.54\ X_1 + 0.25\ X_2 - 2.67\ X_3 + 0.48\ (X_1^2) - 0.55\ (X_2^2) - 4.53\ (X_3^2) + 0.56\ (X_1\ X_2) + 2.63\ (X_1\ X_3) - 0.36\ (X_2\ X_3)$. Coefficient values and statistical parameters obtained for the model are given in Table 4.

The results obtained are then analyzed by F- statistical test for analysis of variance (ANOVA) to assess the "goodness of fit".

The result of the ANOVA analysis for the model is shown in Table 5. The model equation adequately describes the response surfaces of COD removal in the interval of investigation. The models are found to be significant at 95% confidence level by the *F*-test as shown in Table 5, with all *p*-values of regression ≤0.05 (data not shown)). In fact, the value of $F_{statistic}$ (the ratio of mean square due to regression to mean square to real error) of 7.4736 was greater than $F_{0.001,9,13}$ (4.19). In addition, the models do not exhibit lack of-fit (p > 0.05). The lack-of-fit test measures the failure of the model to represent data in the experimental domain at points that are not included in the regression. If a model is significant, meaning that the model contains one or more important terms, and the model does not suffer from lack-of-fit, does not necessarily mean that the model is a good one (Mathieu et al., 2000). If the experimental environment is quite noisy or some important variables are left out of the experiment, then it is possible that the portion of the variability in the data not explained by the model, also called the residual, could be large. Thus, a measure of the model's overall performance referred to as the coefficient of determination and denoted by *R*2 must be considered.

At the same time, adjusted *R*2 (0.726) allowing for the degrees of freedom associated with the sums of the

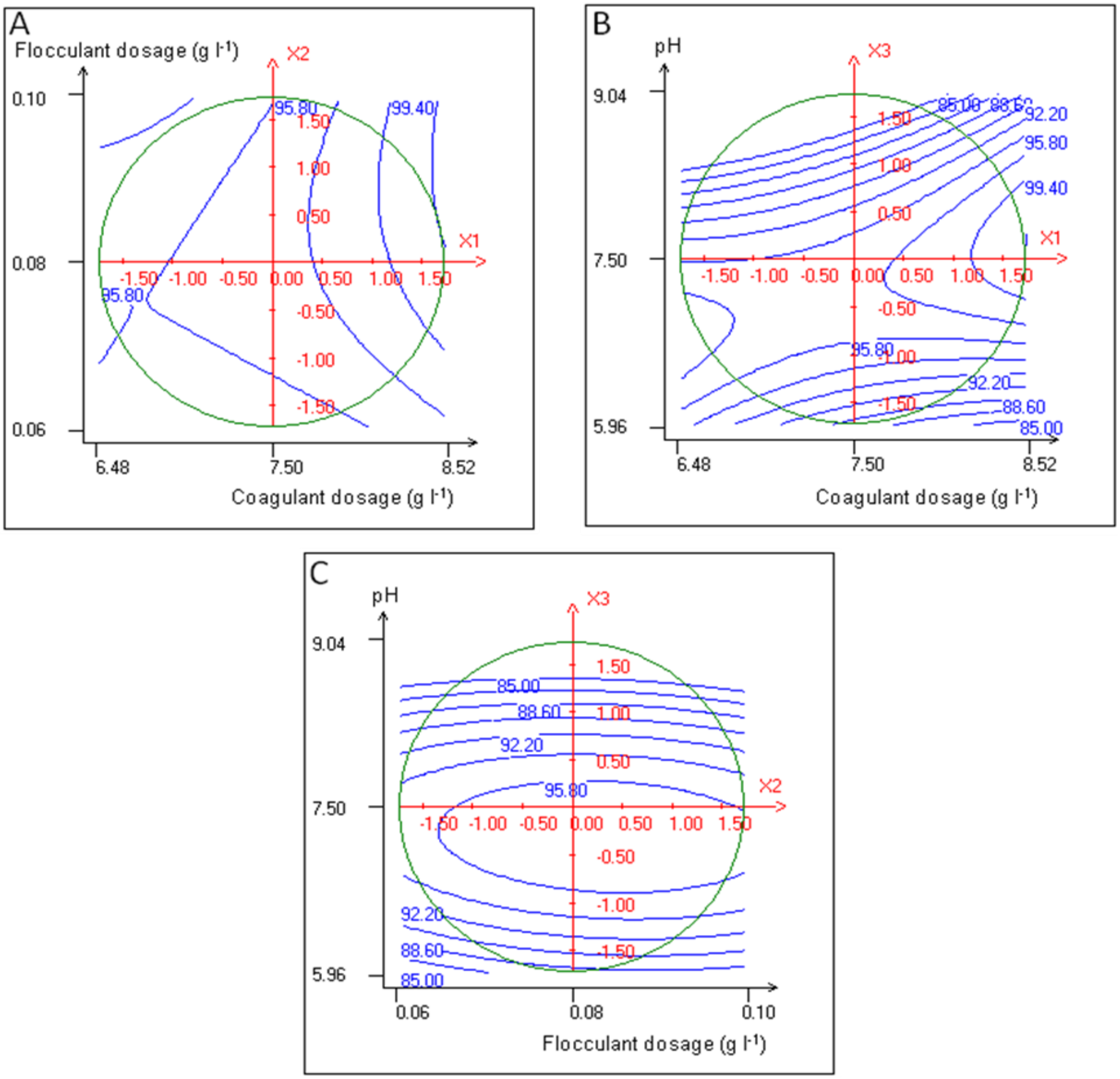

Figure 1. Contour plots of response Y (COD removal) showing the effect of variables: A, Effect of coagulant and flocculant dosage; B, effect of pH and coagulant dosage; C, effect of pH and flocculant dosage.

squares is also considered in the lack-of-fit test, which should be an approximate value of $R2$ (0.838). This latter value of the correlation coefficient indicates that only 16.2% of the total variation could not be explained by the empirical model (Ahmad et al., 2005).

Optimization of the COD removal efficiency

With COD removal as the response, the response surfaces (3D) and the contour plots (2D) of the quadratic model with one variable kept at central level and the other two varying within the experimental ranges are respectively shown in Figures 1 and 2. The obvious trough in the response surfaces indicates that the optimal conditions were exactly located inside the design boundary.

The corresponding two-dimensional contours showed a considerable curvature in contour curves, implying that these three factors were interdependent (Figure 1). There were significant interactive effects on COD removal between coagulant dosage and flocculant dosage (Figure 1A), coagulant dosage and pH (Figure 1B), as well as flocculant dosage and pH (Figure 1C). The contour plot of coagulant dosage versus pH showed that the optimal conditions for the response were located in the region, where coagulation dosage ranged from 8200 to 8300 mg/l and pH from 7.2 to 7.3. In the coagulation process, the aggregation of colloidal particles occurred through charge neutralization and sweep-floc effects (Aguilar et al.,

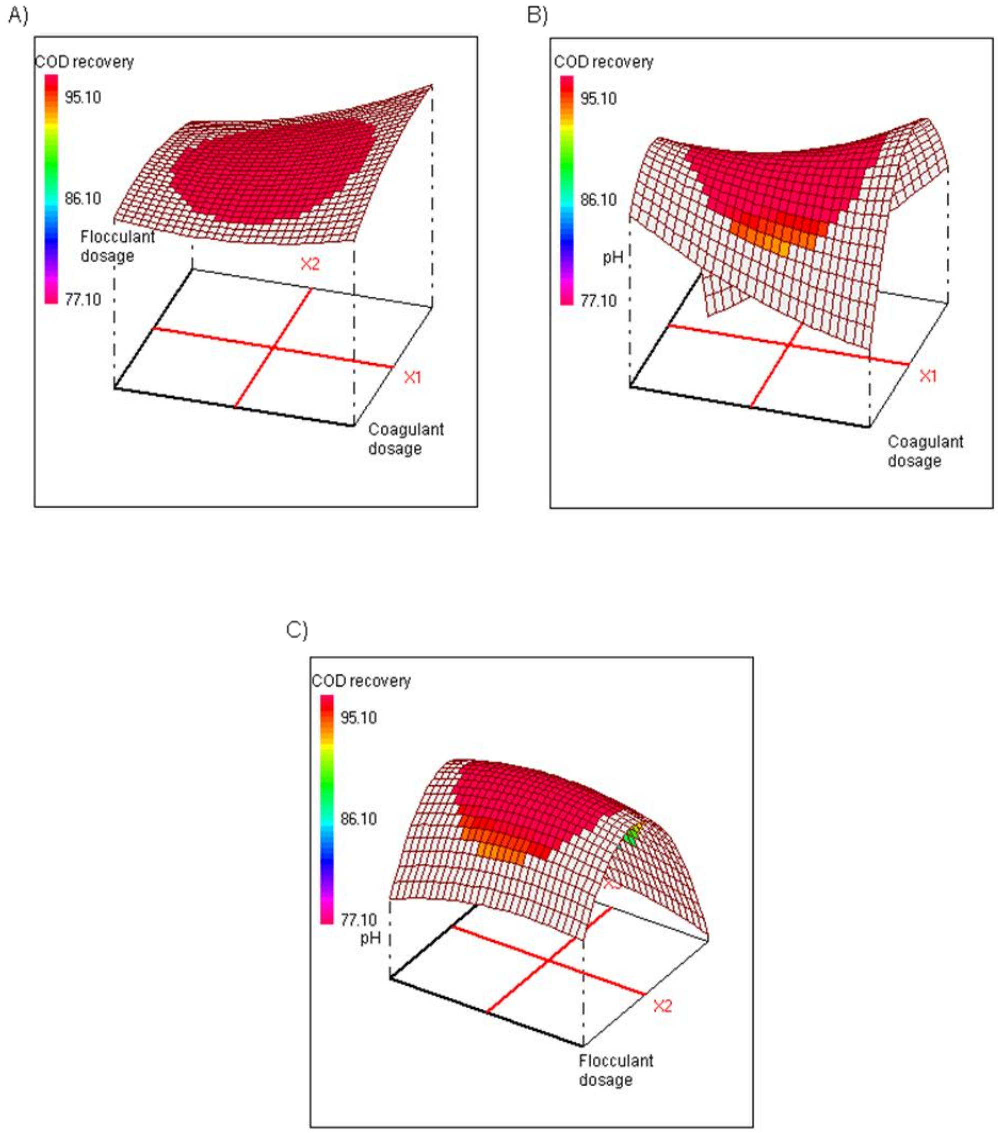

Figure 2. Surface graphs of response Y (COD removal) showing the effect of variables: A, Effect of coagulant and flocculant dosage; B, effect of pH and coagulant dosage; C, effect of pH and flocculant dosage.

2003). The contour plot of flocculant dosage versus pH showed that the optimal conditions for the response were located in the region, where flocculation dosage were ranged from 70 to 90 mg/l and pH from 7.2 to 7.3. These conditions showed a remarkable performance on total COD removals with average treatment efficiencies between 96 and 97% (Figure 2).

Conclusion

The physicochemical process that is known as coagulation–flocculation is common and necessary in pink printing industrial effluent treatment. This work has demonstrated the application of RSM in seeking optimal conditions for this process with respect to the COD removal. RSM using CCD was applied to evaluate effects of coagulant dose, flocculant dose and pH on the coagulation–flocculation effectiveness, and then determine the optimum conditions. The results showed that the three factors considered in this study played an important role on removal efficiency of COD. The optimum conditions obtained for coagulant dose, flocculant dose and pH were 8250, 80 and 7.25 mg/l, respectively. Under these optima conditions, about 97% COD removal was obtained. The results of the confirmation experiment agreed with predictions. This demonstrates that RSM can be successfully applied for modeling and optimizing the coagulation–flocculation process and it is the economical way of obtaining the maximum dosage of information in a short period of time and with the least number of experiments.

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