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Optimization of Nickel Removal from Electroless Plating Industry Wastewater using Response Surface Methodology

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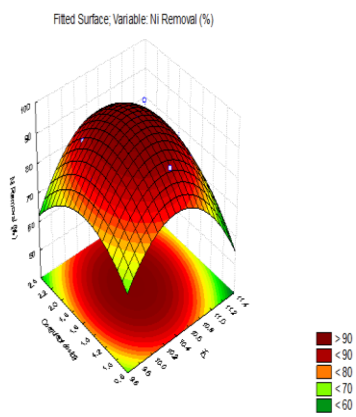
Graphical abstract



Abstract

Optimum pH and coagulant dosage for chemical precipitation in wastewater treatment plants is conventionally obtained through repeated jar test. In this research, optimization of the performance of polyacrylamide in the treatment of industrial wastewater was carried out using response surface methodology. The individual linear and quadratic effect of coagulant dosage and pH on the degree of removals of nickel, total suspended solids, Chemical Oxygen Demand and turbidity were investigated. The optimum pH and polyacrylamide dosage were found to be 10.5 and 1.6 ml/L respectively and the optimum percentage nickel removal was 96.9%. The model used in predicting the precipitation process gave a good fit with the experimental variables and hence the suitability of response surface methodology for the optimization of polyacrylamide performance.

Keywords: Polyacrylamide; nickel; wastewater; chemical precipitation; response surface methodology

1.0 INTRODUCTION

Wastewater from electroless plating industries are characterized with high concentration heavy metal, high total suspended solids (TSS), Chemical Oxygen Demand (COD) and turbidity. Chemical precipitation is an important process in water and wastewater treatment used for the removal of heavy metals such as nickel, TSS, COD and turbidity [1-4]. Most common coagulants are aluminum based salts and iron-based salts. Other coagulant includes polyacrylamide (PAA) and chitosan, a biodegradable, non-toxic linear cationic polymer of high molecular weight [5-10]. PAA, a synthetic polymer derived from acrylamide monomer, was originally introduced for use as a support matrix for electrophoresis in 1959 [11]. However, in the cross-linked form, it is highly water absorbent and can be used for waste water treatment [5]. Water soluble synthetic polymers like PAA have on their chains ligands capable to coordinating sites. In PAA the $–NH_2$ groups serves that purpose. Metals are adsorbed at the polymeric backbone mainly by secondary bonding interactions like hydrogen bonding; coordinate bonding involving the metal ions and the electron donating groups present at the polymer. There are three types of PAA. Anionic PAA has molecules with negative charge, cationic polyacrylamide with positive charge and non-ionic PAA which has molecules with no charge. They are used in very rare instances and special circumstances only particularly in mining.

Optimization plays a key role in environmental engineering parameters since the best system performance mainly is at the optimum point or optimum range. The majority of wastewater treatment processes are multi-variable and optimization through the classical method is inflexible, unreliable and time-consuming. Thus, response surface methodology (RSM), as a very efficient design and widely used technique, can be adapted for parameters optimization of various wastewater treatment processes [12]. Studies have also shown that, the effectiveness of polyacrylamide as a coagulant is affected by dosage, pH, time and temperature. The percentage nickel reduction is reported to increase with increase in

PAA dosage. This is because more active sites become available for binding as the dosage is increased. Also, variation of adsorption with pH can be explained by considering the charge of the ions and the electro–kinetic behavior of PAA. At lower pH values, more protons are available to protonate the amino groups of PAA, therefore, the attractions of both cationic ions decreased. Under strongly basic conditions, the negatively charged phenolic hydroxyl groups become potential active sites (NH_2) and could be attracted by the ammonium groups in the absorbents [13].

There is need to combine these factors appropriately to obtain a high efficiency of treatment. The conventional method to seek the optimal conditions is by trial and error approach using jar tests. This involves changing the levels of one factor and at the same time, keeping the others in constant, running the experiment, observing the results, and moving on to the next factor [10]. This is indeed time and energy consuming. It is also usually incapable of revealing the optimal combination of factors due to ignoring the interaction among them [14-16].

A better alternative is the use of RSM because it includes the influences of individual factors as well as the influences of their interaction. RSM is a technique for designing experiments, building models, evaluating the effects of several factors, and achieving the optimum conditions for desirable responses with a limited number of planned experiments [16-19]. There are some published RSM studies focusing on the usability of RSM for optimization of various types of wastewater treatment processes [12, 15-16]. It is however observed that works concerning the optimization of the nickel removal using PAA are not readily available. Hence, this work is aimed at investigating the effect of pH and coagulant dosage on nickel, SS, COD and turbidity removal using PAA and optimizing these parameters using RSM in order to obtain the optimum degree of removal.

■2.0 EXPERIMENTAL

2.1 Wastewater Sample and Materials

The wastewater was collected from electroless industry located in Johor Bahru, Malaysia. This is a nickel electroless plating company, which produce a substrate that is use for a memory discs. Aluminum substrate is used as a surface material for nickel to deposit onto it. These aluminum surface needs to undertake nickel electroless plating so as to provide a protective layer between the aluminum surface and the data storage surface and generate amorphous structure with less porosity. Ni-P with > 10% P is non-magnetic and does not interfere with the data storage layer, high hardness and wear resistance and excellent resistance to corrosion. The plating process consists of aluminum substrate, pretreatment and nickel plating. The water quality is shown in Table 1. PAA was purchased in the form of white fine powder from Singaway Fluid Control Pte Ltd.

Table 1 Wastewater characteristics

Parameter	Total nickel	COD	pH	TSS	Turbidity
Values	94.3 mg/L	1320 mg/L	2.6	1780 mg/L	1740 NTU

2.2 Chemical Precipitation

The chemical precipitation using PAA for nickel concentration reduction was conducted on a program–controlled jar tester with six stainless steel paddle blade to simulate the chemical treatment plant. The jar tests were conducted in the 1L graduated glass beaker (90 mm diameter, 150 mm high) fitted with six stirring blade positioned at one-third of the reactor height from the bottom. 1L of the wastewater samples were taken into each of the six beakers and coagulant was added to the samples. The jar test was started with continuous agitation at speed of 250 rpm for 15 minutes. This was then followed by slow mixing at 30 rpm for 30 minutes. The sample was then allowed to settle for 30 minutes. After settling, the top clear phase of supernatant was siphoned with syringe. The supernatant was analyzed to determine the concentration of total nickel, COD, TSS and turbidity. All tests were conducted at an ambient temperature (20–25°C). The above procedure was repeated for 5 times at room temperature (25±1ºC). Similar procedure was repeated at different dosages and pH. The pH adjustment was done by using diluted hydrochloric acid (HCl) and diluted sodium hydroxide (NaOH).

2.3 Design of Experiment

In order to achieve optimum nickel removal, RSM experimental design was used to study the response pattern and to determine the best combination of variables which will give the optimum condition for the experiment. In this study, two variables X_1 (pH) and X_2 (coagulant dosage) were used. The effects of the X_1 (pH) and X_2 (coagulant dosage) at two variables levels are shown in Table 2. For statistical analysis, the relationship between the coded and the actual variables can be expressed by Equation (1).

$$x_i = (X_i - X_o) / \Delta X_i \qquad (1)$$

Where x_i is independent variable, X_i is independent real value; X_o is independent real value on the center point and ΔX_i is change step value. The removal of nickel is taken as the dependent variable or response. Y_i propose model for the response is described by Equation (2) [19-21]:

$$Y_i = \beta o + \beta_1 x_1 + \beta_2 x_2 + + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{12} x_1 x_2 \qquad (2)$$

Where Y_i is the predicted response, βo is the offset term, β_1 and $\beta 2$ are the linear effect terms, β_{11} and $\beta 12$ are the squared effects and β_{12} is the interaction effects. STATISTICA v8.0 computer software was used for the analysis of variance (ANOVA), and multiple regression analysis of the data was obtained. The fit of regression model was checked by coefficient of determination R^2, Fisher's test F and its associated probability P were used to determine the overall model significance. Surface behavior was investigated on the response function (Y_i) by using the regression equation. The fitted polynomial equation expressed as surface plots in sequence to visualize the relationship between the response and experimental level of each factor and to figure out the optimum conditions.

Table 2 Independent variables process and their corresponding levels

Independent variables	Symbols		Levels		
	Uncoded	Coded	-1	0	1
pH	X_1	x_1	9.7	10.0	11.2
Dosage (mg/L)	X_2	x_2	1.0	1.6	2.0

■3.0 RESULTS AND DISCUSSION

3.1 Modeling of Chemical Precipitation using PAA

The results of the experimental design are shown in Table 3.

Table 3 Experimental design and predicted responses

Experimental design		Predicted responses			
pH	PAA Dosage (ml/l)	Nickel Removal (%)	COD Removal (%)	TSS Removal (%)	Turbidity Removal (%)
10.5	1.0	90.94	45.53	99.66	99.2
10.5	2.2	88.33	40.66	99.28	98.92
10.0	1.2	86.27	46.55	99.58	99.58
11.0	2.0	90.50	45.32	99.24	99.22
10.5	1.6	96.36	47.88	99.79	99.89
10.5	1.6	96.87	46.90	99.83	99.92
11.0	1.2	82.97	46.66	99.87	99.7
11.2	1.6	80.56	42.76	99.62	99.7
9.8	1.6	85.67	43.43	99.75	99.62
10.0	2.0	89.22	40.53	99.37	99.36

3.2 Model Fitting

The results in Table 3 were used to run ANOVA and Multiple Regression Analysis in STATISCA software using polynomial model Equation (2). This allows the optimum degree of nickel removal and other variables to be predicted. The predicted results are shown in Table 3. The coefficients of the model equation which are used to predict the optimum degree of nickel removal were determined by multiple regression analysis using STATISCA and are shown as Equations 3-6,

$$Y_1 = 96.615 - 2.3117\, X_1 + 1.6972 X_2 - 2.3117 {X_1}^2 - 6.5475\, {X_2}^2 + 2.2900\, X_1\, X_2 \quad (3)$$

$$Y_4 = -2720.09 + 537.36\, X_1 + 7.4800\, X_2 - 26.130\, {X_1}^2 - 20.460\, {X_2}^2 + 5.72\, X_1\, X_2 \quad (4)$$

$$Y_2 = 54.31818 + 7.39654\, X_1 + 8.79456\, X_2 - 0.31250\, {X_1}^2 1.16016\, {X_2}^2 - 0.5250\, X_1\, X_2 \quad (5)$$

$$Y_3 = -271.834 + 54.820\, X_1 + 21.1730\, X_2 - 2.65\, {X_1}^2 - 16.453\, {X_2}^2 + 2.650\, X_1\, X_2 \quad (6)$$

with the variables; nickel removal (Y_1), suspended solid (SS) removal (Y_2), turbidity removal (Y_3) and COD removal (Y_4) and the responses for the tested variables in coded units: pH (x_1) and coagulant dosage (x_2).

The significance of each variable is shown in Table 4 by the F-value of the model at 97.5% confident level. The F-value for nickel, COD, TSS and turbidity removal are 5.79, 23.89, 10.72 and 7.06 with very low probability values (Figures 1-2). This implies a very high significant effect on the performance of PAA. It is reported that regression models with P values less than 0.05 indicates that it is statistically significant [16].

The plots of predicted versus observed variables shown if Figures 1 and 2 yielded straight line with the coefficients of determination (R^2) as 0.87876, 0.92646, 0.87876 and 0.96726 for nickel removal (Y_1), TSS removal (Y_2), turbidity removal (Y_3) and COD removal (Y_4) respectively. The model shows adequate relationship between the observed and predicted results with coefficient (R^2) for all the responses closer to 1. Therefore, the model can be used to predict the nickel removal [21].

Table 4 Analysis of variance (ANOVA)

Sources	SS	D*f*	MS	F-value	F
Nickel removal					
Regression	218.10	5	43.62	5.79	> 5.52
Residual	30.12	4	7.55		
Total	248.22	9			
COD removal					
Regression	75.14	5	15.03	23.89	> 5.52
Residual	2.52	4	0.63		
Total	76.87	9			
SS removal					
Regression	0.47	5	0.09	10.72	> 5.52
Residual	0.03	4	0.01		
Total	0.47	9			
Turbidity removal					
Regression	265.84	5	53.17	7.06	> 5.52
Residual	30.12	4	7.53		
Total	248.22	9			

3.3 Analysis of Response Surfaces and Pareto Chart

The effects of the independent variables on the dependent variable are elaborated by visualization using response surface plots and pareto charts generated by the STATISTICA software. Figures 3-4 (Pareto charts) for each of the variables show the significance of each variable in Equations 3-6 for nickel, COD, TSS and turbidity removal from their probability (P) values respectively. Figure 3a show the quadratic term of pH (X_1^2), with P value of 0.007 has the highest significance on nickel removal, since P values less than 0.05 indicates that a variable is statistically significant [16,21]. Figure 3a also show that the quadratic term of dosage (X_2^2), the linear terms of dosage (X_2) and pH (X_1) and the interaction between dosage and pH (X_1X_2) with p values of 0.0632, 0.2994, 0.4311 and 0.459 respectively have the least significant effect on nickel removal. However, result in Figure 3b shows that quadratic term of dosage (X_2^2) has the most significant effect on COD removal with P value of 0.00208, followed by the linear term of pH (X_1) with P value of 0.0370, followed by the linear terms of dosage (X_2) with P value of 0.0065. The quadratic term of pH (X_1^2), with P value of 0.1487 and the interaction between dosage and pH (X_1X_2) with p value of 0.459 have least significant effect on COD removal.

More also, Figure 4a shows that the variables with the highest significant effect on TSS removal are the quadratic term of dosage (X_2^2) the linear terms of dosage (X_2) with P values of 0.0064 and 0.0131 respectively. These imply that the removal of SS is most dependent on coagulant dosage and moderately on pH.

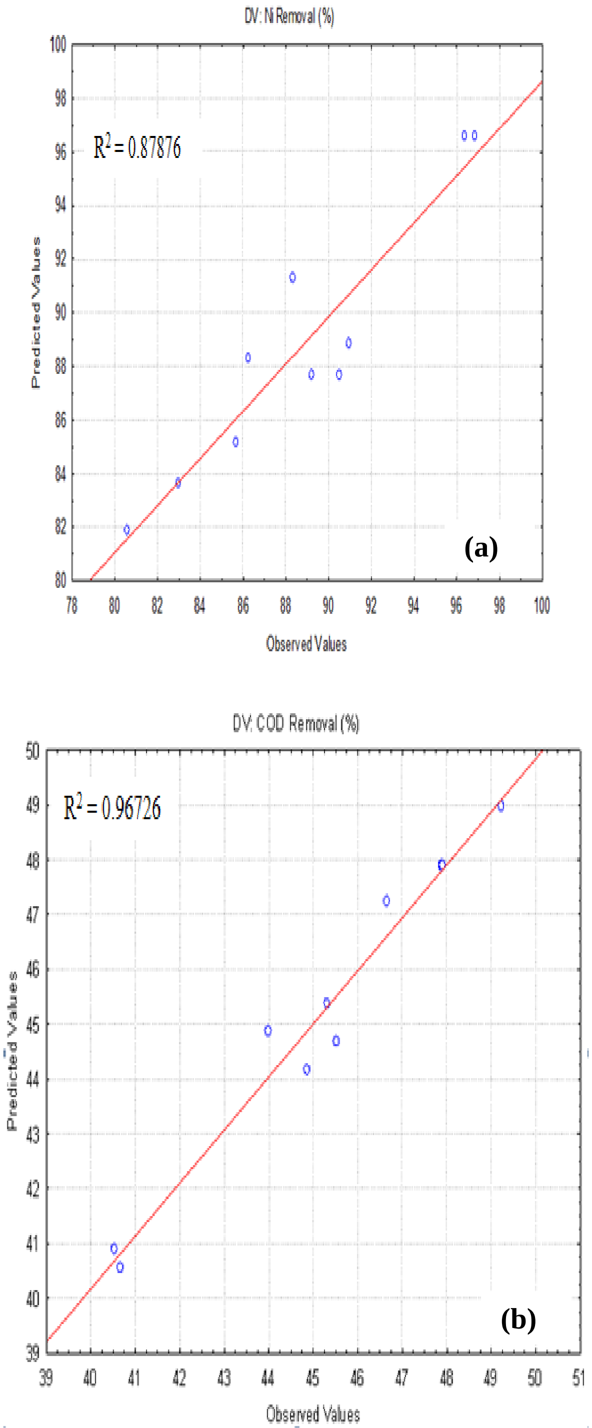


Figure 1 Plots of predicted versus observed variables. (a) Nickel removal and (b) COD removal

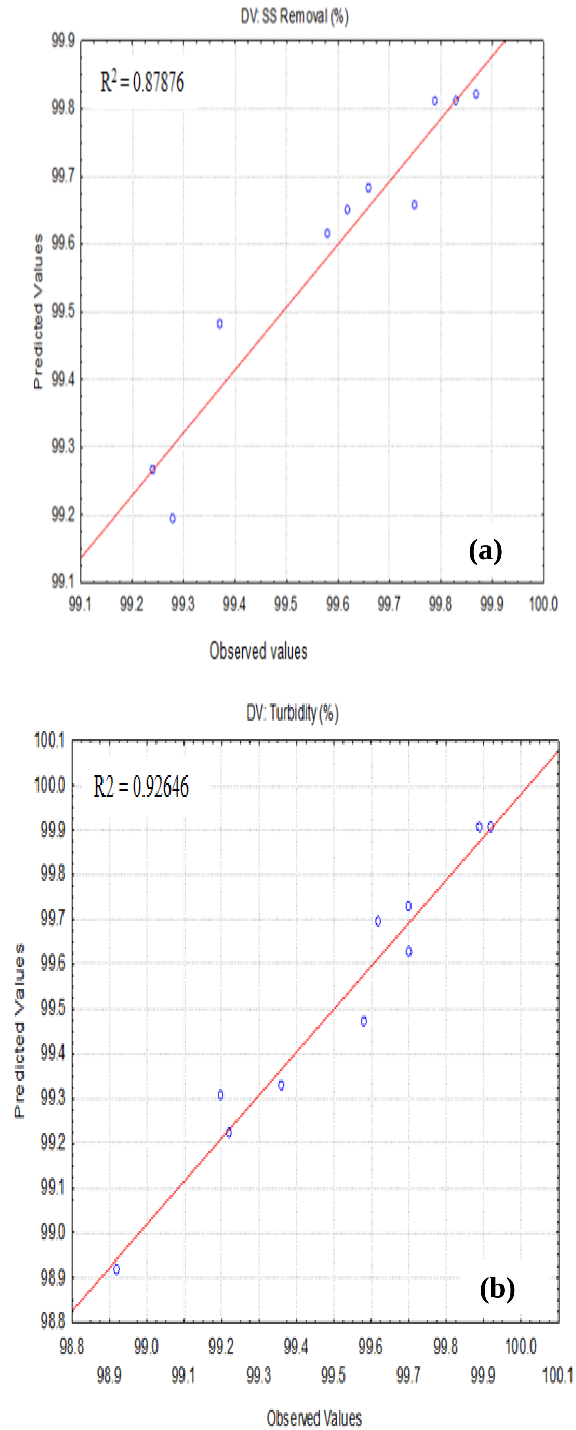


Figure 2 Plots of predicted versus observed variables. (a) TSS removal and (b) turbidity removal

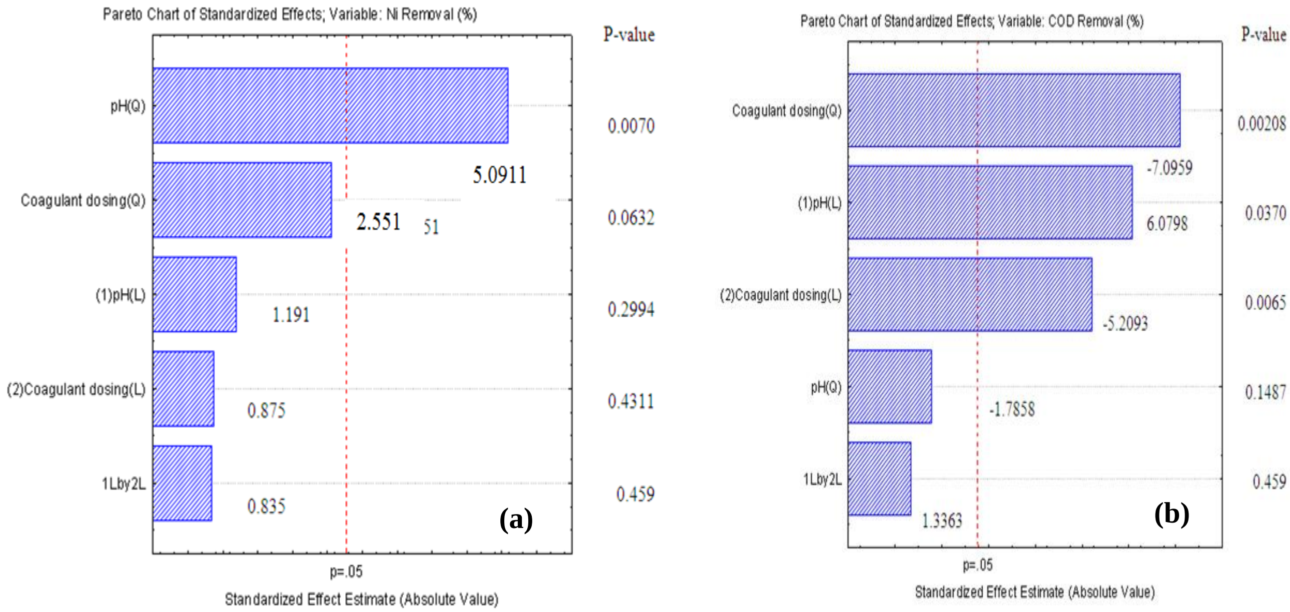


Figure 3 Paretto Chart of (a) Nickel and (b) COD Removal

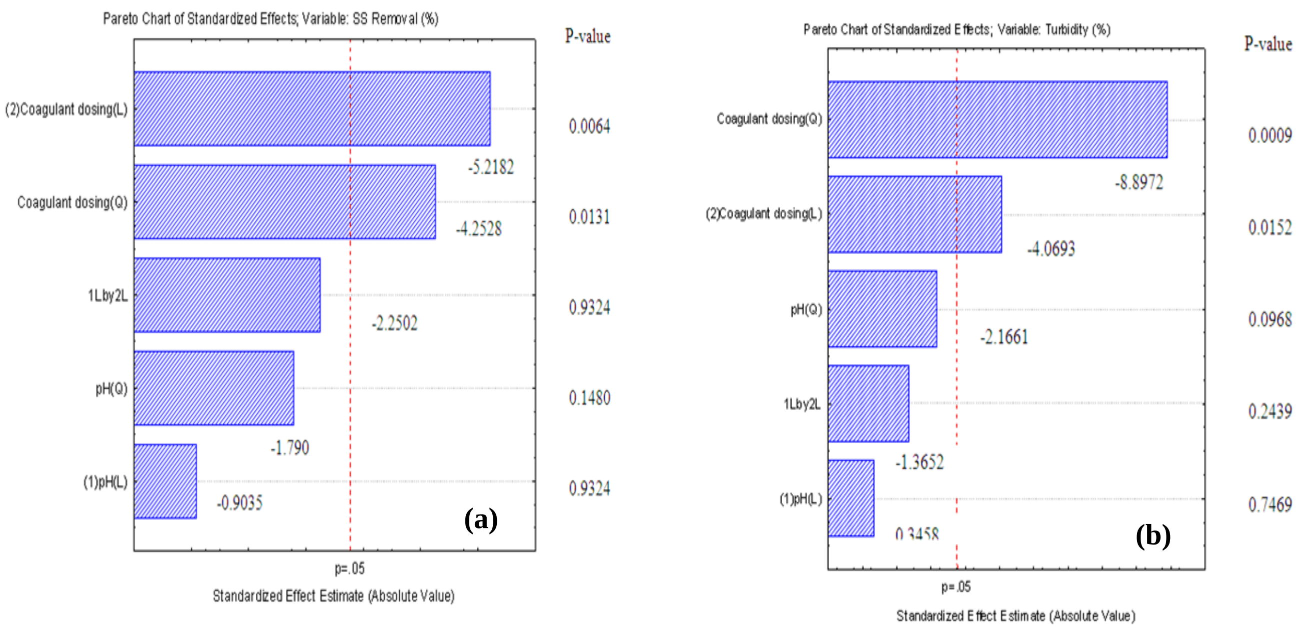


Figure 4 Paretto Chart of (a) TSS and (b) Turbidity Removal

These agree with the result of the optimization of the coagulation-flocculation process for wastewater treatment using polymeric ferric sulfate (PFS)-poly-diallyldimethyl ammonium chloride (PDADMAC) composite coagulant with 89.5% PFS and 10.5% PDADMAC [16]. The interaction between dosage and pH (X_1X_2), the quadratic term of pH (X_1^2), and the linear term of pH (X_1) with P values of 0.9324, 0.1480 and 0.9324 respectively have least effect on the removal of TSS. Figure 4b shows that the variables with the highest significant effect on turbidity removal are the quadratic term of dosage (X_2^2) the linear terms of dosage (X_2) with P values of 0.0009 and 0.0152 respectively. These imply that the removal of turbidity is most dependent on coagulant dosage and moderately on pH. The interaction between dosage and pH (X_1X_2), the quadratic term of pH (X_1^2), and the linear term of pH (X_1) with P values of 0.968, 0.2439 and 0.7469 respectively have least effect on the removal of turbidity.

The combined effect of pH and coagulant dosage on nickel, COD, TSS and turbidity removal are shown Figure 5. The response surface curve were plotted to explain the interaction of the variables and to determine the optimum level of each variable for maximum response. As can be seen from Figures 5a, 5b and 5d, the maximum percent removals of nickel, COD and turbidity were at pH of 10.5 and PAA dosage of 1.6 ml/l. Figure 5c shows that the maximum percentage TSS removal is at pH value of 11.0 and PAA dosage of 1.2 ml/l. In general, the response surface plots indicate that the maximum nickel, COD, TSS and turbidity removal efficiency are located inside the design boundary.

3.3 Optimization

Models obtained in this study were utilized for each response in order to determine the specified optimum conditions and the optimum conditions for prediction of nickel removal were at the pH and coagulant dosage of 10.46 and 1.64 mg/L respectively as tabulated in Table 5. These conditions corresponded to 96.7% of predicted removal of nickel. Confirmatory experiments were run with these optimum values of pH and dosage. The observed value of nickel removal was 96.9% indicating 0.21% error between observed and predicted value as shown in Table 6. This error is considered small as the observed value is within the 5% of significance level.

Table 5 Predicted analysis on reduction of nickel at optimum conditions

Factor	Observed minimum	Critical values	Observed maximum
pH	9.79	10.46	11.21
Coagulant dosage	1.03	1.64	2.16

Table 6 Predict and observed values of the reduction of nickel at optimum process condition

Response	Predicted value	Observed value	Error (%)
Reduction of nickel	96.7	96.9	0.21

■4.0 CONCLUSION

The modeling and optimization of chemical precipitation of nickel from wastewater using PAA shows that the highest nickel, COD, TSS and turbidity reduction were obtained at PAA dosage of 1.6 ml/l and pH 10.5. The optimum percentage nickel removal was 96.9%. The coefficients of determination (R^2) are 0.87876, 0.92646, 0.87876 and 0.96726 for nickel removal, suspended solid (SS) removal, turbidity removal and COD removal respectively. This model used in predicting the precipitation process shows a good fit with the experimental variables and hence the suitability of RSM method for the optimization of PAA performance.

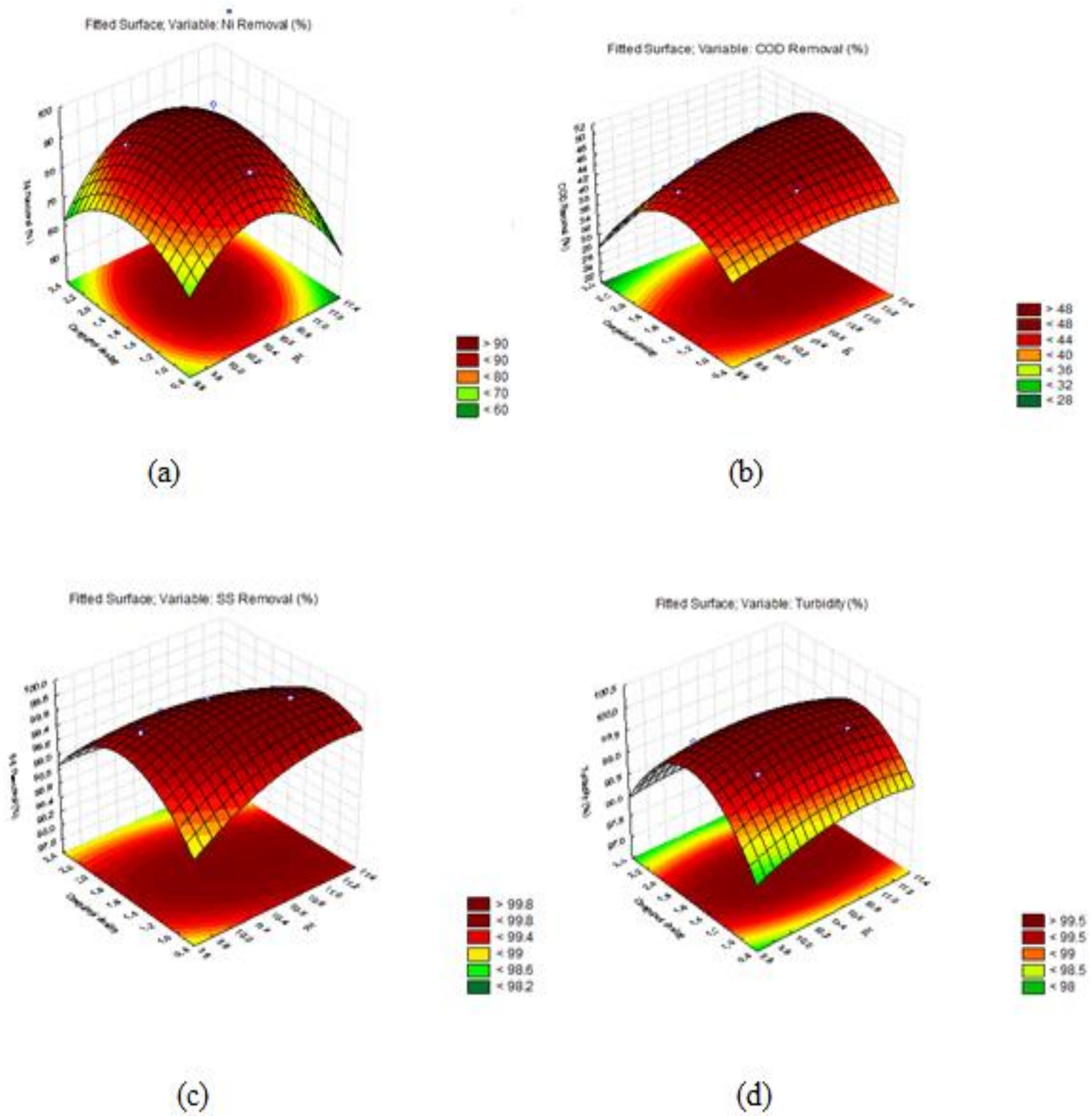


Figure 5 3D response surface plots of (a) nickel (b) COD (c) TSS and (d) turbidity removal versus PAA dosage and pH

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