Original Research Paper

Use of Response Surface Methodology for Optimization of Nickel Adsorption in an Aqueous Solution by Clay

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Abstract: In this study, Scanning Electron Microscopy (SEM), X-ray diffraction and BET surface area (SBET) methods were employed to characterize the adsorbent. Response Surface Methodology (RSM) was consider as a good method to assess Ni adsorption treatment and Box-Behnken design was used to examine simultaneous the effects of different experimental variables (pH, adsorbent dosage, contact time). Experimental results showed high adsorption efficiency. Statistical analysis showed a high correlation coefficient (R^2 value 0.9578). It was showed that adsorption efficiency raised as contact time and adsorbent dosage raised. The best results were acquired with optimum conditions. A contact time of 120 min and an adsorbent dose of 0.5 mg L^{-1} give the highest adsorption capacity. The similarity between observed and predicted values sustains the relevance of the model to estimate the adsorption process. It was suggested that Langmuir and Freundlich isotherm models fitted for Nickel adsorption process (R^2 values ranged between 0.92 and 0.99). The results showed that adsorption efficiency increased as temperature increase. Mostly, Tunisian clay may be assessed as powerful adsorbent for the adsorption of Nickel from aqueous solution.

Keywords: Response Surface Methodology, Clay, Nickel, Optimization, Isotherms

Introduction

The disposal of industrial wastewater and sludge into environment may cause a serious threat to fauna and flora. Industrial effluents contain high levels of heavy metals like Zinc, Copper, Nickel, Lead, Chromium, Cadmium Mercury, etc. The deposal of industrial effluents in the sea and the rivers may cause the biological accumulation and magnifications of heavy metals in the food chain. It is well known that heavy metals had toxic and carcinogenic effect to human health (Rao *et al*., 2018)

Nickel (Ni) existed different forms. It was present in the earth with low level of about 0.009%. Ni was detected in soils and waters. The origins of Ni were coins, earrings, watches, belt buckles, bras, mobile phones, dental and orthopedic implants and cardiovascular stents (Schmidt and Goebeler, 2011).

The exposure to Ni had many effect to human health like dermatitis and carcinogenesis (Zambelli and Ciurli, 2013). The long term exposition to Ni and other heavy metals enhanced cancer impact (Seilkop and Oller, 2003).

Recently, researchers have shown that Ni compounds were responsible to carcinogenicity. It was indicated that metals exposure were responsible to alteration in metabolism cell and cell death (Salnikow and Kasprzak, 2007). It was documented that heavy metals caused a release of toxic pollutants (Lee *et al*., 2012).

The toxic impact of heavy metals on human health and environment had prompted the researchers around the word to discover effective methods for treatment of toxic metal ions from effluent. For this reason, different processes have been worn as precipitation, ion exchange, cementation, coagulation, sedimentation and adsorption (Tovar-Gómez *et al*., 2015). Among these treatment, adsorption was applied successfully to eliminate organic and inorganic compounds (Turki *et al*., 2015). However, this technique had many limitations like costly maintenance and disposal of used adsorbent.

Synthetic adsorbents such as activated carbon have shown to be very expensive. clay was applied with success for the adsorption of heavy metals from water. The use of natural adsorbent like clay had many economic advantages (Vieira *et al*., 2010; Djomgoue *et al*., 2012; Boujelben *et al*., 2015). Clay was used successfully in adsorption process for many reasons. First, the low cost of clay in comparison to others adsorbents. Second, the properties of adsorbent. Clay had a high specific surface area, excellent physical and chemical stability and several other structural and surface properties (Chen *et al*., 2008).

In this study, response surface methodology was performed to analyse adsorption process. Response Surface Method (RSM) is a statistical experimental design technique applied to evaluate the behaviour interaction of various factors and searching for the optimum conditions (Nwabanne and Igbokwe, 2012). Standard techniques do not treat the interaction effect of various parameters concerned in the adsorption treatment (Jia-Hong *et al*., 2012). More these techniques are very expensive and consumed a lot of time. RSM method can be an effective solution to solve the limitation of classical method. The main goal of employing RSM method is to enhance the performance and the cost of experimental study by working less experiments. (Keramat and Zare-Dorabei, 2017; Saini *et al*., 2019).

This study has been investigated to found the optimum conditions of the adsorption of Ni by clay. RSM is used to determine most appropriate conditions. RSM analyses the relationship between parameters and response (Lightfoot *et al*., 2017). Box Behnken Design (BBD) was used to examine the impact of various parameters.

Materials and Methods

Characterisation of Adsorbent

The clay used in this study comes from Djebel Sidi Salem Nabeul, which is located in the northeastern region of Tunisia. In order to improve its mechanical resistance and to remove some impurities, Clay was submitted to calcination at 500°C for 24 h. The mineralogical analysis of samples was performed according to processes described by and Holtzapffel (1985). X-ray diffraction techniques was employed for (XRD) analysis. The samples of clay were sieved and air dried at room temperature. The clay was sifted and the < 2 m fraction was used for analysis. The chlorite and kaolinite chlorite and kaolinite after glycolysis for 1 h, solvating and heating for 3 h at 550°C. The logician X'-pert high score was used to quantify different phases. Brunauer, Emmet and Teller method was used to calculate Specific surface area. Scanning Electron Microscopy (SEM) apparatus JSM-IT100 was used to image the Surface morphology.

Response Surface Methodology (RSM): The Box-Behnken Experimental Design

RSM was used to determine optimum condition for the adsorption of Ni by clay (Kataria and Grag, 2018). The most RSM method employed is the Box-Behnken Design (BBD). RSM is a statistical method based on several experiments. The objective of this method is to find the interaction between different variables to obtain optimum conditions (Shahbazi and Zare-Dorabe, 2019). The number of experiments carried out (N) in RSM was calculated according to Eq. (1):

$$n = 2k\left(k-1\right) + c_0 \qquad (1)$$

Where:
k = Number of variables and
c_0 = Centre-point.

In this study, three factors (pH, adsorbent dosage and contact time) were used to determine optimal conditions. Each variable was studied at a three level central composite design (- 1, 0 and + 1). The factor levels are given in Table 1. The lower level variable was designated as "- 1", the central level as "0" and higher level as "+ 1".

Statistical analysis was performed by the licensed software, Design Expert 11.0. Table 1 showed the levels and ranges of the different factors studied. The system was explained according to the Eq. (2):

$$Y = A\sum\nolimits_{j=1}^{3} BXj + \sum\nolimits_{j=1}^{3} C + Xj^2 + \sum\nolimits_{i=1}^{3} D\sum\nolimits_{j=1}^{3} XiXj + \varepsilon \qquad (2)$$

Where:
Y = The predicted response
A = The constant
B = Coefficients of linear effect
C = Coefficients of quadratic effect
D = Coefficients of interaction effect
ε = A random error
Xi and Xj = Dimensionless coded predicted values for the independent factors

The statistical analysis was investigated by Analysis of Variance (ANOVA). The suitability of the response surface models was estimated by determination of the determination coefficient (R^2). Contour plots and surface plots represented the relationship behaviour between parameters and optimal conditions of the system.

Adsorption Isotherms

The impact of temperature on Ni adsorption was determined. Bach experiments were performed using three temperatures (10, 25 and 40°C). The supernatant obtained after withdraw and filtration was used to determine The residual *Ni* concentration employing the spectrophotometer double beam UV-visible spectrophotometer (HITACHI modèle Z-6100). The Ni amount retained in the adsorbent phase was calculated according to Eq. (3) (Arulkumar *et al*., 2011):

$$Qe = \frac{\left(C_o - C_e\right)V}{W} \qquad (3)$$

where, C_0 and C_e are the initial and equilibrium concentrations (mg L^{-1}) of Ni, respectively; V is the volume (L); and W is the weight (g) of the adsorbent. In this study, Langmuir and Freundlich models were tested. The correlation coefficient R^2were calculated to compare the suitability of the isotherm equations.

Result and Discussion

Characterisation of Adsorbent

The SBET of clay was determined and the value was about 70.17 m^2 g^{-1}. The high level of SBET showed high adsorption efficiency of the support. The result suggested that clay could be a promising adsorbent used to eliminate Ni from aqueous. SEM technique was used to study the morphology of clay. The aspect of morphology was presented in Fig. 1 (a). It was observed that clay was characterized by porous and rough surface. The figure showed that clay was composed of fine particles which did not uniform in size. The mineralogical aspect of clay was studied by X-ray diffraction. Figure 1 (b) showed different mineralogical phases. The quartz is the most mineral identified (characteristic peaks at 1.81, 2.45, 3.34 and 4.25 Å), with kaolinite (peaks at 7.14 and 3.57 Å). The clay contains other mineral phases such as illite (2.58 Å), Feldspath (3.16°A) and phyllosilicate (4.48 Å).

The RSM Design of Adsorption Study

In this study RSM technique was investigated to discover the interaction between variables and to determine optimal conditions process (Rao *et al*., 2018). Table 1 showed The different variables with their coded and actual values. Twenty-seven trials were investigated according to BBD as shown in Table 2. BBD was applied to study the behaviour interaction effect of three variables (pH, adsorbent dosage and contact time) on Ni adsorption efficiency. As shown in Table 2, twenty-seven experiments were realized.

The Statistical Analysis

Equation 4 presents the empirical model's equations between the quantity of Ni adsorbed and contribution parameters:

$$Y\left(\frac{mg}{g}\right) = 12.53 + 4.88\times 1 + 0.13\times 2 + 2.78\times 3 + 0.05\times 1\times 3 + 2.31\times 1\times 3 - 0.18\times 2\times 3 - 3.16\times 1^2 + 0.20\times 2^2 - 3.57\times 3^2 \quad (4)$$

Where:
X_1 = Represent contact time (min)
X_2 = Represent pH
X_3 = Represent adsorbent dosage (g L^{-1})

ANOVA was applied to study the removal of metal ions. Table 3 showed the statistical analysis of the study. A model is considered significant if the p value <0.05. Statistical tests indicated that the regression was statistically significant at the F-value 134.82 and the value of prob > F was 0.00001 for Ni ions. Statistical results presented the coefficient of determination R^2, adjusted R^2 and predicted R^2. It was observed that the coefficient of determination (R^2 = 0.9578) was slightly higher than adjusted R^2 and predicted R^2, the adjusted R^2 (0.9524) is similar to the predicted R^2 (0.9541) value (Table 3). Figure 2 presented the normal plot of residuals between percentage probability and internally studentized residuals. The figure showed clearly that the data points on the plot were properly distributed close to the straight line. ANOVA test indicated the similarity between the experimental and predicted data. The result confirmed the applicability of the model for predicting the response.

Response Surface Plots

Response surface plots was realized to understand the interaction between different variables and to obtain the optimal condition of adsorption process. The graphical illustration of the adsorbed amount of Ni ions relative to the three variables (pH, contact time, adsorbent dosage) studied are presented in Fig. 3, 4 and 5.

The behaviour of adsorption efficiency relative to variation of pH and contact time was shown in Fig. 3 (a, b). The figure illustrates that adsorption efficiency raised as contact time increased. The optimal adsorption was obtained at contact time 120 min. The lowest amount of Ni adsorbed was observed at contact time 60 min. However, results suggested that that pH had no effect on the adsorption efficiency as shown in Table 2. They are in contrast with results suggested by other studies. Which found that adsorption efficiency increased as both the pH and adsorption time increased up to a point and then decreased (Hameed *et al*. 2008; Ani *et al*., 2019).

Figure 4 (a, b) shows that adsorption capacity desponds on adsorbent dosage and contact time. It is well observed that adsorption efficiency was improved as well as contact time increased and adsorbent dosage increased. These results confirmed the results shown in Table 2. The efficiency of the adsorption increased as adsorbent dosage increased until it reached a point, then it gradually decreased. The initial increase of adsorption process could be due to the uncovered surface area of the adsorbents. With time, the adsorbent sites may be obstructed. That being so, the equilibrium was reached when the totality of surface area was blocked (Ani *et al*., 2019).

The evolution of adsorption efficiency in function of pH and adsorbent dosage was illustrate in Fig. 5 (a, b). The results showed that Adsorption efficiency increased as the pH and adsorbent dosage increased. Adsorption capacity raised up to a point and then decreased. The optimal adsorbent dosage was 0.5 g L^{-1} after which the adsorption efficiency showed a decrease trend.

Adsorption Equilibrium Study

The most isotherm model applied to analyze adsorption properties are Langmuir and Freundlich isotherm. The interaction between adsorbent- adsorbate was given by equilibrium parameters of empirical model (Liu *et al*., 2011). In this study, different temperatures (10, 25 and 40°C) were used for adsorption equilibrium study. Figure 6 presented the isotherms obtained for Ni adsorption by clay. The Freundlich isotherm model described the adsorption behaviour of Ni ions on the clay (Fig. 7). This isotherm is expressed by Eq. 5 (Boujelben *et al*., 2015):

$$Q = KfCe^{1/n} \tag{5}$$

Where:
K_f : Constant of adsorption capacity
n : Constant of adsorption affinity

The linear form equation is expressed by Eq. (6) (Boujelben *et al*., 2015):

$$\log Qe = \log K_f + \frac{1}{n}\log Ce \tag{6}$$

Where
Qe = The amount of adsorption
k_f = The Freundlich constant related to sorption capacity
$1/n$: = A constant related to energy or intensity of adsorption

The Freundlich isotherm gives the exponential distribution of activated sites and their energy. However, it does not predict any saturation of adsorbent surface. The linear plot of equation determines Freundlich exponent's k_f and $1/n$. The values of the Freundlich constants K_f and $1/n$ are shown in Table 4. The values ranged between 0.64 - 0.75 and 0.8 - 0.9 for K_f and $1/n$, respectively. The heterogeneity of surface was determined by $1/n$ value. It ranged between 0 and 1. The surface is more heterogeneous as its value gets closer to zero (Haghseresht and Lu ,1998). The Freundlich isotherm model showed a deviation from linearity (R^2 ranged between 0.73 and 0.83).

The adsorption behaviour of *Ni* ions on clay were also analysed by the Langmuir model (Fig. 8). In this isotherm the intermolecular forces decrease rapidly with distance (Ghaedi *et al*., 2012). Langmuir isotherm characterized the adsorption attitude of Ni ions on the clay. Linear form is presented according to Eq. (8) (Boujelben *et al*., 2015):

$$\frac{Ce}{Qe} = \frac{1}{Qob} + \frac{1}{Qe}Ce \tag{8}$$

Where,
Ce (mg L^{-1}): Equilibrium concentration;
Qe (mg g^{-1}): Quantity of adsorbate adsorbed per unit mass of the adsorbent at equilibrium condition;
Qo (mg g^{-1}): Highest amount of adsorbate adsorbed forming monolayer;
K_L (L mg^{-1}): LANGMUIR constant associated with the affinity of the adsorption sites;

The adsorption parameters relative to Langmuir isotherm model are presented in Table 4. Results showed that R_L value ranged between 0 and 1. The regression coefficient R^2 ranged between 0.92 and 0.99. Which indicated that Langmuir equation was suitable for Ni adsorption on clay adsorbents. It was observed that when temperature raises from 10 to 40°C, the adsorption efficiency raises from 2.36 mg g^{-1} to 3.43 mg g^{-1}. These results indicated that warming is favourable to improve adsorption efficiency of Ni on clay.

Table 1: Box-Behnken design and levels of factors

	Levels of factors				
Variables	Unit	code	Low (-1)	Central (0)	High (1)
Contact time	Min	X1	60	90	120
pH	-	X2	3	5	7
Adsorbent dosage	g	X3	0.1	0.5	1

Table 2: levels and response of Box-Behnken design

	Coded levels			Response Qe (mg/g)		
Runs	X1 (min)	X2	X3 (g)	observed	predicted	Residual
1	60	3	0.1	1.9	0.38	1.52
2	60	3	0.5	2.32	4.49	-2.17
3	60	3	1	2.3	1.68	0.62
4	90	3	0.1	5.23	6.53	-1.3
5	90	3	0.5	14.9	13.72	1.18
6	90	3	1	14.87	14.77	0.10
7	120	3	0.1	5.23	5.42	-0.18
8	120	3	0.5	14.95	13.65	1.30
9	120	3	1	14.92	15.99	-1.07
10	60	5	0.1	1.8	0.44	1.36
11	60	5	0.5	2.24	4.39	-2.15
12	60	5	1	2.25	1.37	0.87
13	90	5	0.1	5.17	6.67	-1.5
14	90	5	0.5	14.81	13.7	1.11
15	90	5	1	14.85	14.53	0.32
16	120	5	0.1	5.2	5.58	-0.38
17	120	5	0.5	14.91	13.65	1.26
18	120	5	1	14.89	15.78	0.89
19	60	7	0.1	2.2	0.91	1.26
20	60	7	0.5	2.42	4.69	-2.27
21	60	7	1	2.39	1.47	0.92
22	90	7	0.1	6.28	7.22	-0.93
23	90	7	0.5	14.92	14.08	0.84
24	90	7	1	14.91	14.71	0.20
25	120	7	0.1	6.3	6.15	0.15
26	120	7	0.5	14.95	14.05	0.86
27	120	7	1	14.9	15.98	-1.08

Table 3: Statistical analysis of model

Std. dev	1.74	R^2	0.9578
Mean	102.11	Adj R^2	0.9524
C.V. %	3.2	Pred R^2	0.9541
Press	152.86	F-value	134.82
Prob >F	0.00001		

Table 4. Langmuir and Freundlich constants of clay at various temperatures

Temperature (°C)	10	25	40		10	25	40
Freundlich constants				Langmuir constants			
K_f (mg g^{-1})	0.73	0.75	0.64	Q_m (mg g^{-1})	2.36	2.75	3.43
1/n (L mg^{-1})	0.8	0.9	0.9	K_L (L mg^{-1})	0.3	0.21	1.25
R^2	0.73	0.83	0.82	R_L (L mg^{-1})	0.04	0.07	0.1
				R^2	0.92	0.99	0.93

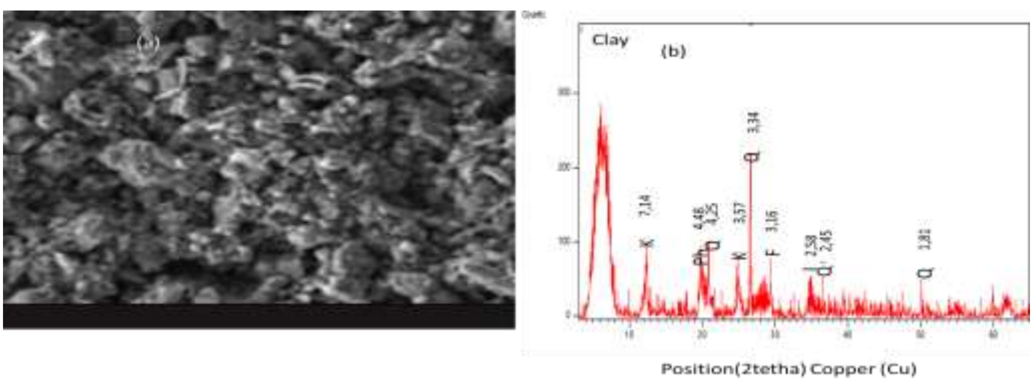

Fig. 1: (a, b). (a) SEM of studied clay, (b) X-ray diffraction pattern of studied clay

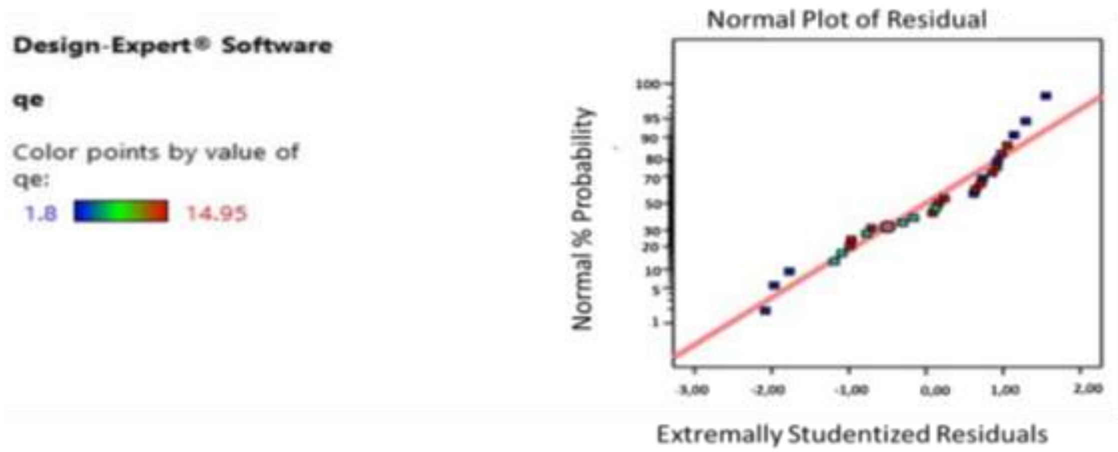

Fig. 2: Normal plot of residuals

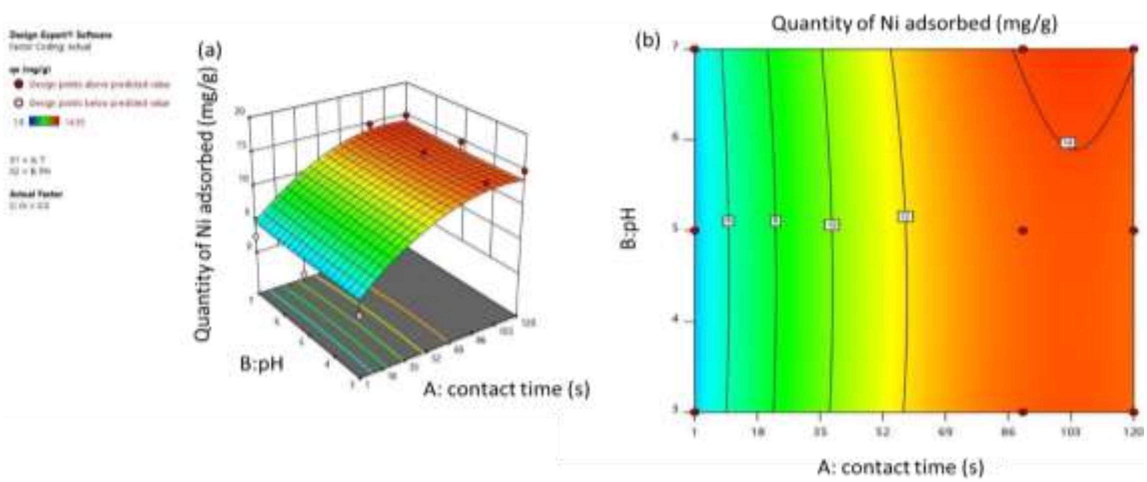

Fig. 3: (a, b). (a) Response surface and (b)contour plots showing interaction between the effect pH (X2) and contact time (X1) on Qe of Nickel, with constant effect of adsorbent dose (X3)

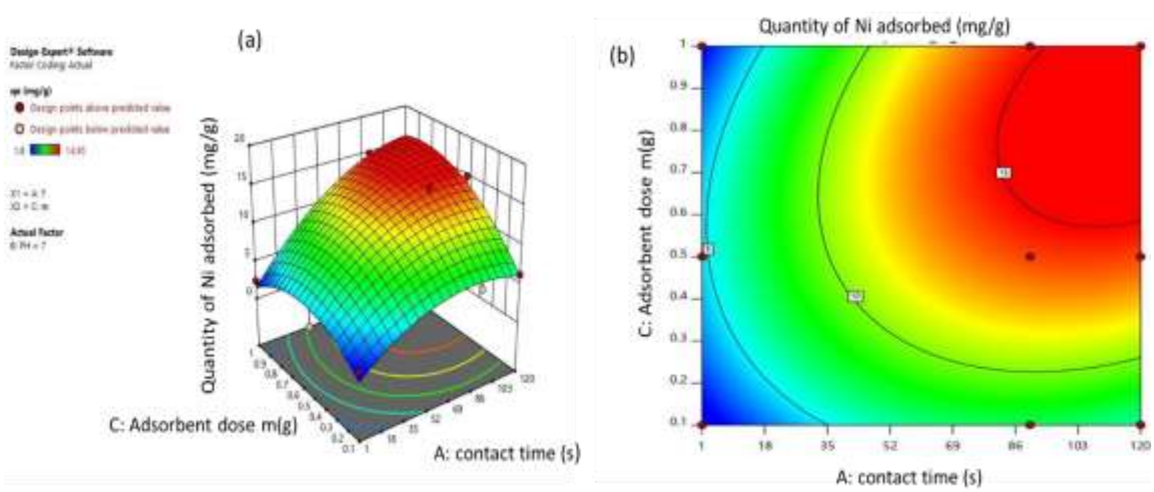

Fig. 4: (a, b). (a) Response surface and (b) contour plots showing interaction between the effect contact time (X1) and adsorbent dose (X3) on Qe of Nickel, with constant effect of pH (X2)

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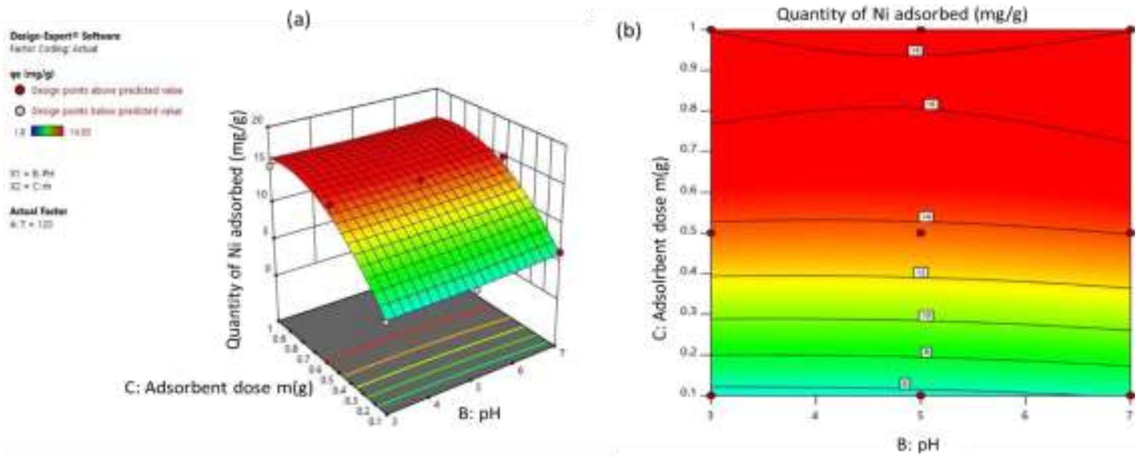

Fig. 5: (a, b). (a) Response surface and (b)contour plots showing interaction between the effect of adsorbent dose (X3), pH(X2) on Qe of Nickel, with constant effect of contact time (X1)

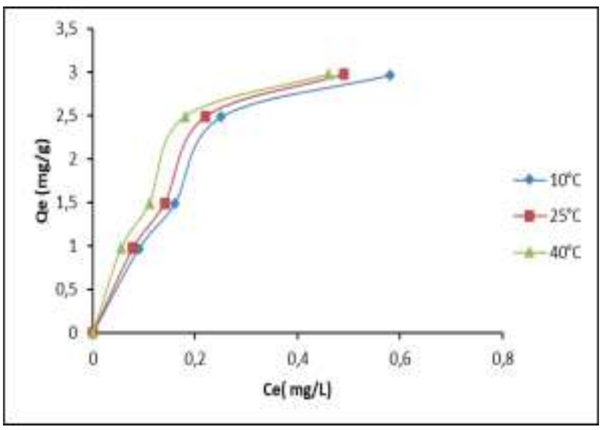

Fig. 6: Equilibrium isotherms for adsorption of Ni ions on clay for 0.5 g adsorbent/25 mL effluent, various temperatures (10, 25 and 40°C) and at pH 7

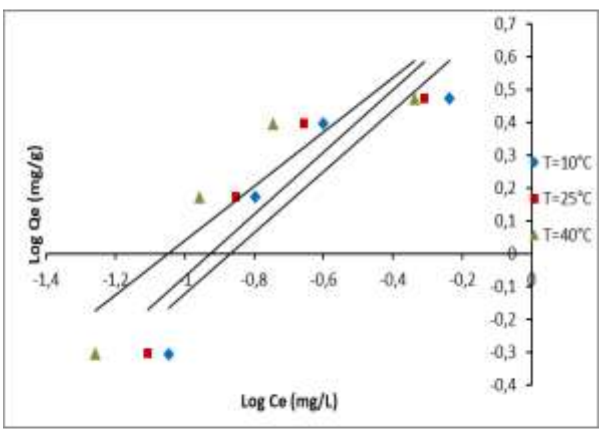

Fig. 7: Freundlich isotherms for the adsorption of Ni on clay at different temperatures (10, 25 and 40°C)

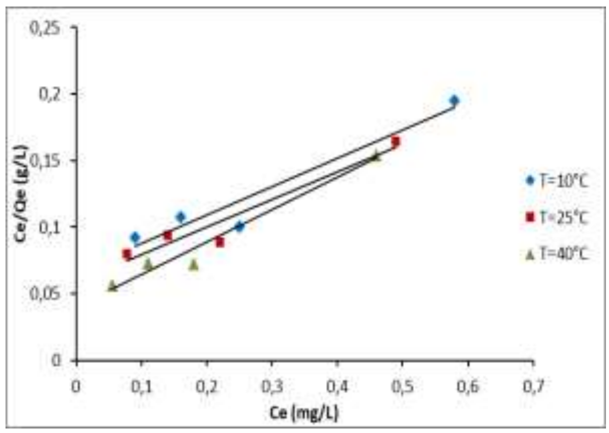

Fig. 8: Langmuir isotherms for the adsorption of Ni on clay at different temperatures (10, 25 and 40°C)

Conclusion

In this study, response surface methodology using Box–Behnken design seems to be a good alternative to found optimal conditions for the adsorption of Ni onto clay. Three independents parameters (pH, contact time and adsorbent dosage) were investigated to understand the behaviour of adsorption process. Graphical illustrations were realized to predict the interactive effect of the variables studied. The selected quadratic model was used to estimate response under the same conditions of trials. Statistical analysis showed that clay was efficient in the adsorption of Ni for a contact time of 120 min and at a dosage of about 0.5 g L^{-1}. It was also documented that Adsorption efficiency increased as temperature raises. The results of this study clearly demonstrates that the use Tunisian clay as adsorbent could be an efficient support for environmental protection.

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Author Contributions

Nesrine Turki: Participated in all experiments and contributed to the writing of the manuscript.

Nesrine Boujelben: Organized the plan of the work.

Zaineb Bakari and Jalel Bouzid: Coordinated the work.

Ethics

This article is original and contains unpublished material. The corresponding author confirms that all of other authors have read and approved the manuscript and no ethical issues involved.

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