

Optimization and Modeling of Leaching Parameters Affecting Nickel Dissolution from Lateritic Ore in Eskisehir (Mihaliccik-Yunusemre) Using Box-Behnken Experimental Design

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Abstract

In this study, optimization and modeling of leaching parameters affecting nickel dissolution from lateritic ore in Eskişehir (Mihalıççık-Yunusemre) were investigated using Box-Behnken design. Stirring speed (100–400 rpm, temperature (40–80°C), acid concentration (0.1–2 M) and dissolution time (30–180 min) were selected as experimental design parameters. 27 experiments were carried out by Box-Behnken experimental design in Minitab 16.0 program. After leaching experiments, the highest Ni dissolution percentages were obtained as 87.85%. The effective parameters and their interactions in nickel dissolution are described with a mathematical model. The results obtained from the experiments were subjected to ANOVA and multiple regression analysis. The R^2 value of the model for nickel dissolution was calculated as 0.980. This showed that the predicted values are in good agreement with the observed values. In addition, three-dimensional response surface and contour graphs of the parameters affecting nickel dissolution efficiency were created and the results were examined.

Keywords: Nickel • Leaching • Box-behnken design • Optimization

Introduction

Nickel is an important and strategic metal and mainly used in modern industrial and metallurgical applications because of its strength and corrosion resistance [1]. Nickel naturally occurs as sulphides and laterites-type ores. Although the lateritic deposits constitute the largest world reserves of nickel, global nickel production has been supplied from sulfide ores due to the challenges of processing laterite compared to sulfide ores [2,3].

The nickeliferous laterite ore deposits are formed by a weathering process. The top of laterite bedrock layer is mainly hematite without a significant nickel content. This is followed by a limonitic zone with 1.5% nickel. Finally there is a saprolitic layer with up to 4.0% nickel content [4,5].

There are several reagents used in lateritic nickel extraction such as hydrochloric acid, sulphuric acid, citric acid, nitric acid, ammonia, oxalic acid and acetic acid [6–15]. Several studies have been performed using Box-Behnken design. Polat and Sayan, studied the struvite precipitation with a Box-Behnken design [16]. Koca et al., studied the evaluation of combined lignite cleaning processes, flotation and microbial treatment, and its modelling by Box Behnken methodology [17]. Ozgen et al., examined the effect of smectite content on swelling to hydrocyclone processing of bentonites with various geologic properties by Box Behnken design [18].

The aim of this study is to optimize leaching parameters affecting nickel dissolution from lateritic ore from Eskişehir (Mihalıççık-Yunusemre) using Box-Behnken design. Stirring speed, temperature, acid concentration and dissolution time were selected as experimental design parameters. The results obtained from the experiments were subjected to ANOVA and multiple regression analysis. In addition, three-dimensional response surface and

contour graphs of the parameters affecting nickel dissolution efficiency were created and the results were examined.

Materials and Methods

Materials

In this study, sample of lateritic nickel ore taken from Mihalıççık-Yunusemre/Eskişehir in Turkey was used (Figure 1). Approximately 100 kg of ore samples were brought to the laboratory and crushed to a size of -3.35 mm. The ore was then ground to size less than 106 μm by a rod mill. Chemical analysis of the sample was done by AAS (GBC SensAA model). The chemical composition of the sample is given in Table 1.

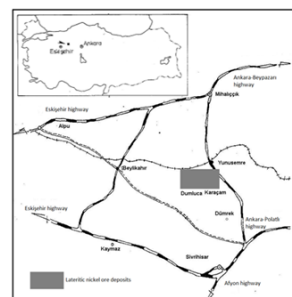


Figure 1. Location of Mihalıççık-Yunusemre/ Eskişehir region in Turkey.

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Received: 02 February 2020; **Accepted:** 11 February 2020; **Published:** 18 February 2020

Table 1. Chemical analysis of lateritic nickel ore sample.

Element	Ni	Fe	Mg	Co
Mass fraction (%)	1.84	23.14	1.58	0.03

Mineralogical investigations were carried out on sample by means of X-ray diffraction (XRD) analysis, X-ray diffraction (XRD) analysis revealed that the main minerals are goethite, hematite and wustite. The gangue minerals of the ore are determined as retgersite, gaspeite, quartz and clay type minerals (Figure 2).

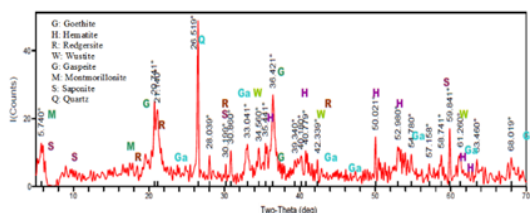


Figure 2. XRD analysis of sample.

Leaching tests

Experiments were carried out in 1 L pyrex leach beaker using 10/500 g/mL (solid-liquid ratio) in a temperature-controlled water bath. The leach solution was provided by Heidolph mark RZR 2021 model mechanical stirrer with a teflon-covered impeller.

Table 2. Experimental factors and their level.

Factors	Description	Units	Low value	Center value	High value
			-1	0	1
X ₁	Stirring speed	rpm	100	200	400
X ₂	Temperature	°C	40	60	80
X ₃	Acid concentration	molar	0,1	0,5	2
X ₄	Dissolution time	min	30	90	180

Box-Behnken design method was applied to determine the effects of nickel dissolution parameters from lateritic ore. The leaching studies were performed according to the full factorial design of experiments. The variables studied were stirring speed, temperature, acid concentration and dissolution time. The variables and levels of full factorial design are presented in Table 2. Each variable was studied at three levels: -1 is for low level, +1 denotes high level, and 0 is used for the midpoint to evaluate the experimental error [16,19].

Box-Behnken designs are based on three-level incomplete factorial designs. Graphical representation of Box-Behnken design matrix for three parameters can be seen in Figure 1 [20]. As seen in Figure 3, experiments are usually performed at center levels of parameters. Design Expert Minitab 16.0 program was used for achieving design matrix and obtaining mathematical models of the response variables.

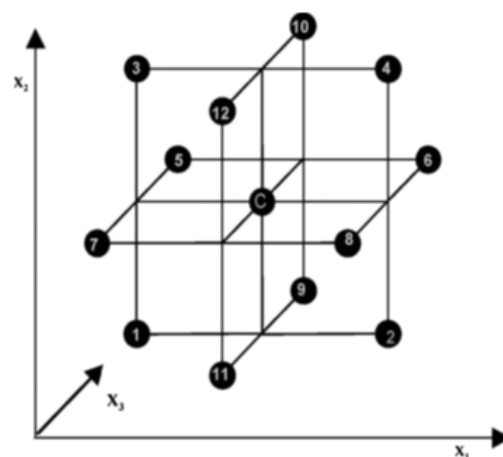


Figure 3. Box-Behnken design with three parameters.

Results and Discussion

Experimental results for the Box-Behnken design experiments

A total of 27 tests were carried out according to Box-Behnken experimental design. Experiments were conducted for three levels of 4 variables. Table 3 shows the experimental and estimated Ni dissolution percentages. The lowest Ni dissolution percentages obtained from experiments was 7.36% and the highest test result was 87.85%. The lowest estimated result and highest estimated result were 6.42% and 81.51%, respectively.

Table 3. Box-Behnken experimental Design table, experimental and estimated Ni dissolution percentages.

Test No	Factors				Test result	Estimated
	X ₁	X ₂	X ₃	X ₄	Ni%	Ni%
1	0	0	0	0	23.23	23.23
2	1	0	1	0	52.81	55.23
3	0	0	1	-1	34.91	38.89
4	0	-1	1	0	45.3	40.38
5	0	0	-1	1	14.33	10.58
6	-1	0	-1	0	9.56	9.76
7	1	1	0	0	42.48	40.97
8	0	1	-1	0	15.83	17.91
9	1	0	-1	0	12.42	11.34
10	0	1	0	1	47.72	49.97
11	0	1	0	-1	24.62	29.16
12	-1	1	0	0	43.27	42.26
13	0	-1	-1	0	8.86	12.36

14	0	0	-1	-1	7.36	6.42
15	0	0	0	0	23.23	23.23
16	-1	-1	0	0	16.25	17.98
17	-1	0	1	0	53.81	57.51
18	1	0	0	-1	14.81	11.73
19	-1	0	0	-1	22.25	17.39
20	0	0	1	1	68.59	69.75
21	-1	0	0	1	29.35	29.59
22	0	-1	0	-1	8.75	9.12
23	1	-1	0	0	17.33	18.56
24	0	0	0	0	23.23	23.23
25	0	1	1	0	87.85	81.51
26	1	0	0	1	32.52	34.54
27	0	-1	0	1	25.24	23.32

ANOVA results

Analysis of variance (ANOVA) based on Box-Behnken experimental design was applied to check the importance and competency of the model coefficient. According to the analysis variance (Table 4), the Fisher's (F-test) with a very low probability value ($P_{Model>F}$) < 0.0001 indicates the model is highly significant. When analyzing ANOVA results, a large value of F with a small value of p (i.e., $p < 0.05$) show that the model is statistically significant, whereas the values greater than 0.1 indicate that the model terms are not significant [21]. It was seen that temperature, acid concentration and dissolution time were significant, whereas Stirring speed, was not significant due to high P-value. P-values for all conditions model are given in Table 4.

Table 4. Analysis of variance for nickel dissolution (Ni%) (ANOVA).

Source	DF	Sum Squares	of Mean Square	F value	P-value Prob>F
Regression	14	10038.5	717.04	41.31	<0.0001*
Linear	4	8852.2	2213.06	127.51	<0.0001*
X ₁	1	0.4	0.37	0.02	0.886
X ₂	1	1634.3	1634.27	94.16	<0.0001*
X ₃	1	6298	6297.96	362.87	<0.0001*
X ₄	1	919.6	919.63	52.99	<0.0001*
Square	4	647.8	161.95	9.33	0.001*
X ₁ ²	1	19.2	6.05	0.35	0.566
X ₂ ²	1	83	169.95	9.79	0.009*
X ₃ ²	1	540.4	447.62	25.79	<0.0001*

X ₄ ²	1	5.2	5.19	0.3	0.595
Interaction	6	538.5	89.75	5.17	0.008*
X ₁ X ₂	1	0.9	0.87	0.05	0.826
X ₁ X ₃	1	3.7	3.72	0.21	0.651
X ₁ X ₄	1	28.1	28.14	1.62	0.227
X ₂ X ₃	1	316.5	316.48	18.24	0.001*
X ₂ X ₄	1	10.9	10.92	0.63	0.443
X ₃ X ₄	1	178.4	178.36	10.28	0.008*
Residual	12	208.3	17.36		
Lack of fit	10	208.3	20.83	624807.17	0.0001*
Pure error	2	0	0		
Total	26	10246.8			

* Significant

Regression results

The results of the regression analysis with Minitab 16.0 software are given in Table 5. The R² value for nickel dissolution was calculated to be 0.980, explaining 98% of the variability in the response. The adjusted R² value was found to be 0.956. The predicted regression coefficients value (R²pred) is in very close agreement with the adjusted regression coefficients value (R²adj), that is because the difference between the two is less than 0.2 [22].

Table 5. Regression values of the optimization study.

Regression Values	Nickel Dissolution Efficiency (%)
Standard deviation	0.0416602
R-Squared	0.98
Adj R-Squared	0.956
Pred R-Squared	0.883

Regression coefficients for nickel dissolution (% Ni) are given in Table 6. From the table, coefficients of acid concentration (X₃), and X₂X₃ seem to be more important for nickel dissolution.

Table 6. Regression coefficients for nickel dissolution (% Ni).

Term	Coefficient	Std. Coefficient	Error T	P-value
Constant	23.2333	2.405	9.659	0.000
X ₁	-0.1767	1.203	-0.147	0.886
X ₂	11.67	1.203	9.704	0.000
X ₃	22.9092	1.203	19.049	0.000
X ₄	8.7542	1.203	7.279	0.000

X_1^2	1.065	1.804	0.590	0.566
X_2^2	5.645	1.804	3.129	0.009
X_3^2	9.1612	1.804	5.078	0.000
X_4^2	-0.9862	1.804	-0.547	0.595
X_1X_2	-0.4675	2.083	-0.224	0.826
X_1X_3	-0.965	2.083	-0.463	0.651
X_1X_4	2.6525	2.083	1.273	0.227
X_2X_3	8.895	2.083	4.270	0.001
X_2X_4	1.6525	2.083	0.793	0.443
X_3X_4	6.6775	2.083	3.206	0.008

Experimental versus predicted

The evaluation between the observed and predicted values from the model is presented in Figure 4. The predicted and actual values of the model are similar and this proved the accuracy of the model.

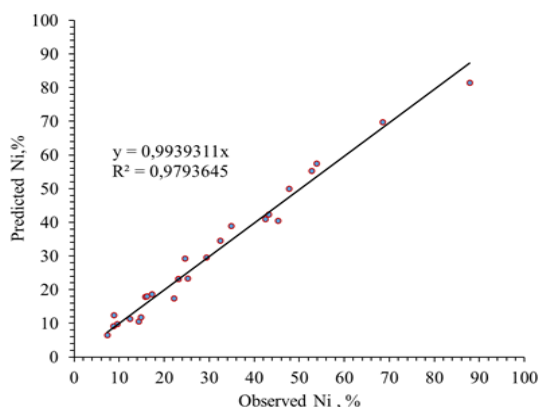


Figure 4. The actual and predicted plot for Ni dissolution recoveries.

Three-dimensional graphics (3D) response surface

The 3D response surface plots are a graphical representation can be prepared as a function of two factors, keeping all other parameters at constant levels, and these graphs are useful for understanding the relationship between the response and experimental levels of each variable and the interaction effects between the variables [16,23-25].

Figure 5 shows the 3D response surface relationship between temperature (X_2) and acid concentration (X_3) with recovery of nickel. It is obvious that the highest recovery could be achieved with the maximum level of temperature (X_2) and acid concentration (X_3). Figure 6 shows the 3D response surface relationship between dissolution time (X_4) and temperature (X_2) with recovery of nickel. It is clear that the highest recovery could be achieved with the maximum level of temperature (X_2) and center level of time dissolution (X_4). Figure 7 shows the 3D response surface relationship between temperature (X_2) and stirring speed (X_1) with recovery of nickel. It is appear that the highest recovery could be achieved with the maximum level of temperature (X_2) and center level of stirring speed (X_1). Figure 8 shows the 3D response surface relationship between time dissolution (X_4) and acid concentration (X_3) with recovery of nickel. The highest recovery was obtained at maximum level of acid concentration (X_3) and center level of time dissolution (X_4). Figure 9 shows the 3D response surface relationship between stirring speed (X_1) and acid concentration (X_3) with recovery of nickel. It is seen that the highest recovery could be achieved with the maximum level of acid concentration (X_3) and center level of stirring speed (X_1). Figure 10 shows the 3D response surface relationship between stirring speed (X_1) and time dissolution (X_4) with recovery of nickel. It is seen that that the highest recovery could be achieved with the center level of stirring speed (X_1) and dissolution time (X_4). Three-dimensional response surface and contour graphs showed similar results and they supported each other.

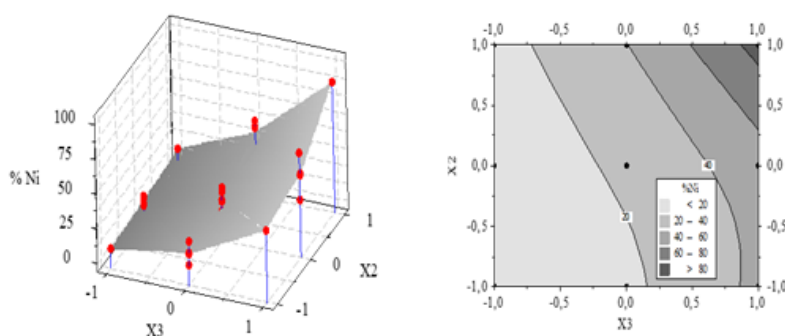


Figure 5. Effects of temperature (X_2) and acid concentration (X_3) on recovery of nickel.

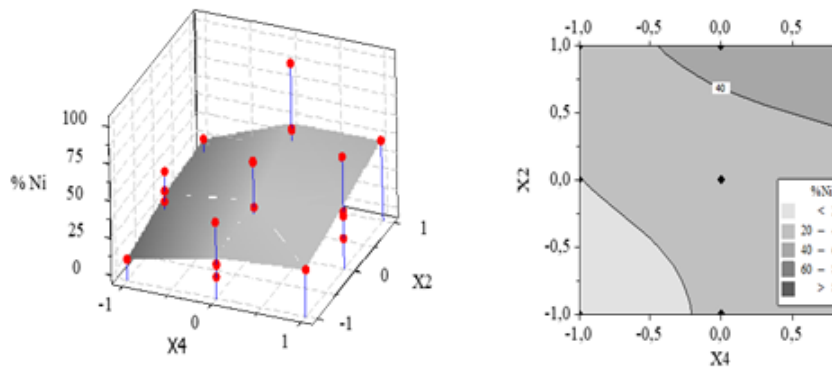


Figure 6. Effects of dissolution time (X_4) and temperature (X_2) on recovery of nickel.

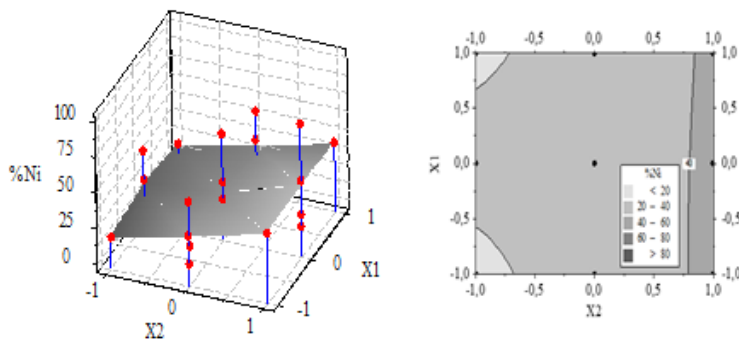


Figure 7. Effects of temperature (X_2) and stirring speed (X_1) on recovery of nickel.

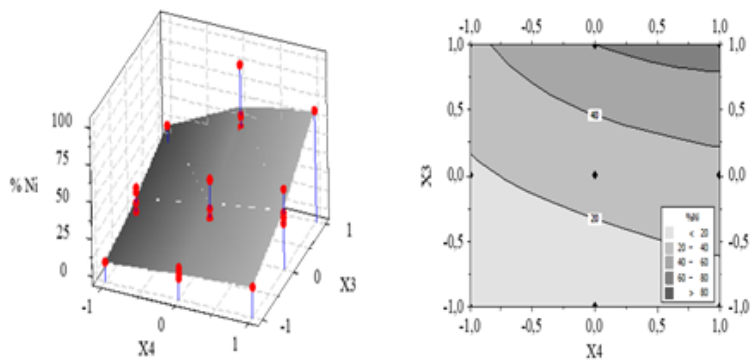


Figure 8. Effects of dissolution time (X_4) and acid concentration (X_3) on recovery of nickel.

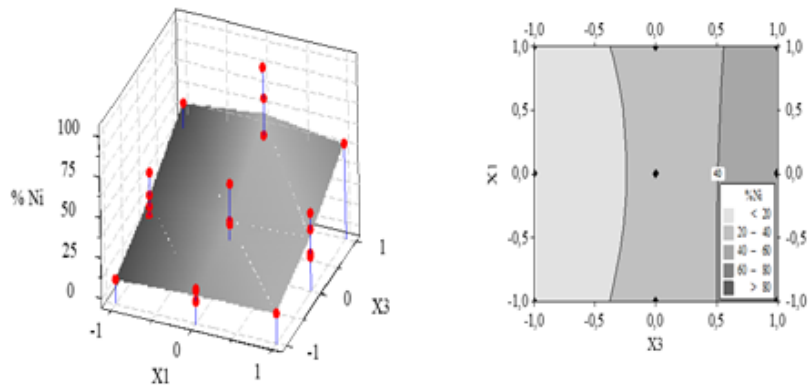


Figure 9. Effects of stirring speed (X_1) and acid concentration (X_3) on recovery of nickel.

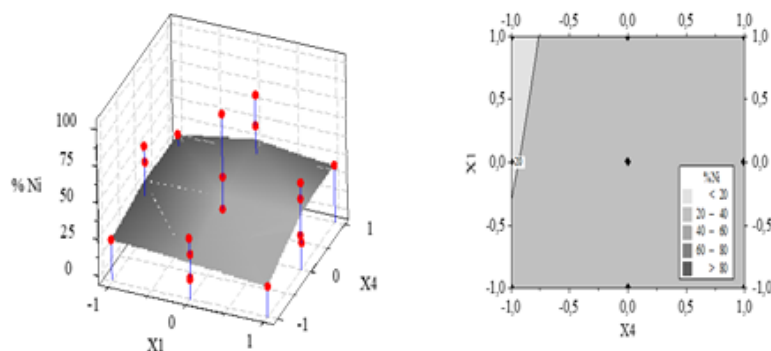


Figure 10. Effects of stirring speed (X_1) and dissolution time (X_4) on recovery of nickel.

Conclusion

In this study, a Box-Behnken design was applied for modeling and optimization of some operating variables on the leaching of nickel from lateritic ore from Eskişehir (Mihalıççık-Yunussemre). The variables studied were stirring speed, temperature, acid concentration and dissolution time. After leaching experiments, the highest Ni dissolution percentages were obtained as 87.85%. The results obtained from the experiments were subjected to ANOVA and multiple regression analysis by using Minitab 16.0 software. The R^2 value of the model for nickel dissolution was calculated as 0.980. This showed that the predicted values are in good agreement with the observed values. In order to gain a better understanding of the effect of the variables on nickel dissolution, three dimensional response surface and contour graphs were created and the results were examined.

Acknowledgements

The author Mohamed Taha Osman wishes to express sincere thanks to his supervisor in Konya Technical University, Department of Mining Engineering (Faculty of Engineering) Associate Professor Dr. Tefvik Agacayak for his honest help and kind support.

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How to cite this article: Tevfik Agacayak and Ahmed Mohamed Taha Osman Abdelraheem. "Optimization and Modeling of Leaching Parameters Affecting Nickel Dissolution from Lateritic Ore in Eskisehir (Mihaliccik-Yunusemre) Using Box-Behnken Experimental Design". *J Environ Anal Chem* 7 (2020).