Optimization of laterites leaching by application of sequential design of experiments

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Abstract: The sequential designed experimentation was applied for leaching of Ni-Fe-Mg containing serpentine ores. The experiments were arranged in two sequences, according to the 2_{IV}^{4-1} fractional factorial experimental design. Six factors were examined, i.e., the material sample, solid-to-liquid ratio (S/L), acid type and its concentration, leaching temperature, and stirring speed. The analysis of variance was used to establish the relation between tested factors, their interactions, and metals recovery. All the derived empirical models were of prime statistical importance.

The obtained results showed that the recovery of Ni was dependent mainly on the material sample, the concentration of acid as well as their interaction, and temperature. The recovery of nickel at the optimal leaching conditions was high (93%). The dissolution of other metals was below 84% (Fe) and 54% (Mg).

Keywords: planned experimentation, fractional factorial experiments, lateritic ores, leaching

1. Introduction

Presently, recovery of nickel from laterite deposits is mostly accomplished with mixed pyro- and hydrometallurgical processes and the suitability of the applied treatment depends on the chemical composition and mineralogy of the Ni-bearing minerals (Arroyo and Neudorf, 2001; McDonald and Whittington, 2008; Guo et al., 2009; Liu et al., 2010; Zuniga et al., 2010; Oxley and Barcza, 2013; Quast et al., 2017). Pyrometallurgical processing is suitable for deposits containing minimum 1.7% of nickel and low content of iron (e.g. nontronite, serpentine, garnierite), whereas low magnesium content is usually preferred when leaching operations are involved (Dalvi et al., 2004; Marshall and Buarzaiga, 2004). Currently, there are two hydrometallurgical processes, i.e., Caron and High-Pressure Acid Leaching (HPAL) that are operated to recover Ni from its ores. The Caron process employs leaching with ammonia and tolerates higher Mg content than the HPAL process, however, this operation is preceded by the calcination and reduction processes. The HPAL process allows eliminating the calcination step; however, it requires expensive autoclaves, and therefore it should be only applied to high-grade ores and in large-scales (Dalvi et al., 2004). The best alternative, to the currently operated processes, seems to be atmospheric leaching, however, its application can be limited, because of ore mineralogy or problems with the further processing of generated leach liquors (McDonald and Whittington, 2008; Wang and Lee, 2017). These solutions typically contain significant concentrations of iron, aluminum, and magnesium, making the downstream processing more challenging, especially to nickel separation (Zhang et al., 2015; Rice, 2016). Other issues are related to an irregularity of the weathering profile (in depth) of the laterites and also due to non-uniform mining. For these reasons, a specific ore sample may contain variable proportions of minerals. The laterite ore feed to a process may thus be characterized by highly variable mineralogy and thus by variable processing characteristics. Therefore, to control the operation under various circumstances, it is necessary to optimize leaching conditions.

Usually, optimization studies are carried out through a traditional experimentation one-factor-at-a-time (OFAT) approach, in which experiments vary only one factor or variable at a time and keep others fixed (Czitrom, 1999). Such experimental approach requires a significant number of runs, and thus is time and material consuming, but costs can be reduced by application of the statistical design. The design of experiments (DOE) is used to change the process variables in specific and patterned ways, according to the factorial treatment structure (Montgomery, 2008). This factorial structure enables finding the effect of factors on either one or more response variables and allows estimation of effects with higher precision than OFAT approach. The secret of the factorial design advantage lies in the idea of hidden replication. It means that factorial approach uses more observations to estimate an effect because each measurement is included in each comparison. The OFAT experimenter must replicate runs to produce equal precision (Leitnaker et al., 1996).

Factorial design offers two additional advantages over OFAT (Anderson and Withcomb, 2007):

- it reveals interactions between factors, i.e., the situation when the effect of one factor depends on the level of another factor,

- it covers a broader area of the factor space from which to draw inferences about a process and consequently improves the prediction of the response in the factor space. Moreover, process optimization is more efficient because the optimal solution is searched for over the entire factor space.

There are many papers which apply the design of experiments (DOE), however, in this approach the number of studied variables is often limited (Li et al., 2010; Poroch-Seritan et al., 2011; Cetintas and Bingol, 2016), and process optimization is performed during a single set of experiments (Kar et al. 2000; Stopic et al., 2003; Milivojevic et al., 2012; Gharabaghi et al., 2012; Gharabaghi and Azadmehr, 2016).

A better approach is a sequential design of experiments, which allows acquiring knowledge about a process gradually. The experience and feedback gained from small-scale tests are used to modify the next run of the experiment, and to improve an original research plan. It allows to introduce new factors or to change levels of the present factors. Therefore, the inference space in a sequential DOE (SDOE) becomes greater. Another advantage of sequential experimentation is that it allows lessening the total number of runs, comparing to the single relative DOE (Box, 1993).

In this work, the sequential experimentation approach was used to determine the optimum leaching conditions of high-magnesium nickeliferrous ore. Specifically, our aim was to achieve, simultaneously, an acceptable level of nickel extraction and low magnesium and iron dissolution. The effect of six operation variables, such as the material sample, acid type and its concentration, solid-to-liquid ratio (S/L), temperature and stirring speed were examined. The experiments were arranged in two sequences according to the 2_{IV}^{4-1} factorial design of experiments. The importance of the main and interaction effects was evaluated by the analysis of variance (ANOVA).

2. Materials and experimental methods

A low-grade nickel ore used in this work was derived from Polish serpentine deposit, located in Szklary Massif, SW Poland. The examined material was received in a dry and grounded form. It was averaged using mechanical sample splitter and representative samples were subjected to analyses. The chemical analysis was performed using an EDXRF spectrometer (PANanalytical Epsilon 3x). Before XRF analyses samples were milled manually below 100 μm. The particle size analysis (of aqueous suspensions of samples) was carried out using a Malvern Mastersizer 2000 instrument.

All chemical reagents were of analytical grade. Sulfuric acid (POCh, Poland) and hydrochloric acid (POCh, Poland) were used to prepare leaching solutions.

Leaching experiments were carried out in stirred glass reactors, equipped with a condenser, to avoid rapid vaporization of the leaching solution at higher process temperatures, immersed in a water bath. When the temperature was reached, the leaching slurry was placed into the reactor. During each leaching experiment, samples of the slurry were taken from the reactor, filtrated and then subjected to a metals concentration analysis by using atomic absorption spectroscopy technique (AAS). Each sample was analyzed twice, and the results were presented as a mean of both measurements.

The preliminary experiments, with 1 mol/dm³ H_2SO_4, were run to determine leaching kinetics of the studied metals under different temperatures (20 and 90 °C). For initial tests, the solid-to-liquid

ratio (S/L), stirring speed, and leaching time were kept constant at 1/10, 400 r.p.m. and six h, respectively. The results from preliminary tests were used to set up leaching conditions in the first DOE sequence, and consequently, the results from DOE 1 were used to specify the factors and their levels in the second DOE sequence. Therefore, fixed parameters in the DOE 1 were: the stirring speed (400 r.pm.), temperature (90 °C) and leaching time (3 h), while values of the solid-to-liquid ratio (1/6) and acid type (HCl) were constant in the DOE 2. The variable factors and their levels are given in Table 2 (Section 3.3.) and Table 4 (Section 3.4.), for DOE 1 and DOE 2, respectively.

The densities and viscosities of leaching solutions were obtained using a pycnometer and capillary viscometer, respectively. These physical properties were measured in triplicate for each solution, at 298.15 K.

The leaching efficiencies were evaluated by determining metals leaching recoveries, according to the equation:

$$R_\% = \frac{c \cdot V}{m \cdot \alpha} \cdot 100\% , \tag{1}$$

where:
$R_\%$ - metal leaching recovery, %,
c – metal concentration in the solution, mg/dm^3,
V – leaching solution volume, dm^3,
m – mass of the ore sample, g,
a – metal content in the ore sample, % (g/Mg).

2.1 Design of experiments

A factorial design is widely used to study a random response to a set of k possible factors. The full-factorial design would require 2^k runs if there are k factors, each at two levels. As the number of factors of interest increases, the number of runs required increases rapidly, becoming infeasible from the time and resource point of view. Thus, it is better to use a fractional factorial experiment which uses only a subset of the runs. Fractional factorials are much more advantageous, comparing to full-factorial experiments since the greater number of factors can be examined, keeping the same number of experiments (Montgomery, 2008).

In the presented study, a sequential design of experiments (SDOE) was used in the leaching tests. Based on this approach the leaching tests were arranged in two SDOE series conducted according to the 2^{4-1} fractional factorial experimental design. Such arrangement leads to 2 runs of 8 treatments, what enables to acquire more information on the process than conducting one run of 16 treatments. The statistical significance of experimental results (at $\alpha = 0.10$) was assessed with the analysis of variance. ANOVA is based on F-test and involves subdividing the total variation of a set of data into parts. The F-value in the ANOVA table is the ratio of the model mean square (MS) to the appropriate error mean square. The larger the F-value the more likely that variation (an observed difference) is not by chance (Anderson and Whitcomb, 2007).

The relationship between variables and responses was represented by empirical equations, which were based on the linear regression models. A general model that might describe the response variable Y related to k factors can be written as follows (Montgomery, 2008):

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \ldots \ldots + \beta_k X_k + \epsilon, \tag{2}$$

where, Y is the predicted response, β_0 is the intercept, β_1, β_2, are the model coefficients for the input factors (X_1, X_2, X_k), and ϵ is an experimental error. The residual analysis was performed to examine the goodness-of-fit in ANOVA. The Minitab 17 Software (Minitab Inc.) was used to design the experiments and data analysis.

3. Results and discussion

3.1 Material characterization

The investigated nickeliferous ore is very fine grained, poorly crystallized and is characterized by a variable chemical composition and structure (Dubińska et al., 2000). The weathering cover of the rock is composed of iron oxides, oxyhydroxides, and SiO_2 varieties and contains a high number of clay

minerals. In this work, two samples differing in chemical composition (Table 1) and particle size distribution (164 µm - sample 1, and 220 µm - sample 2, Fig. 1) were tested.
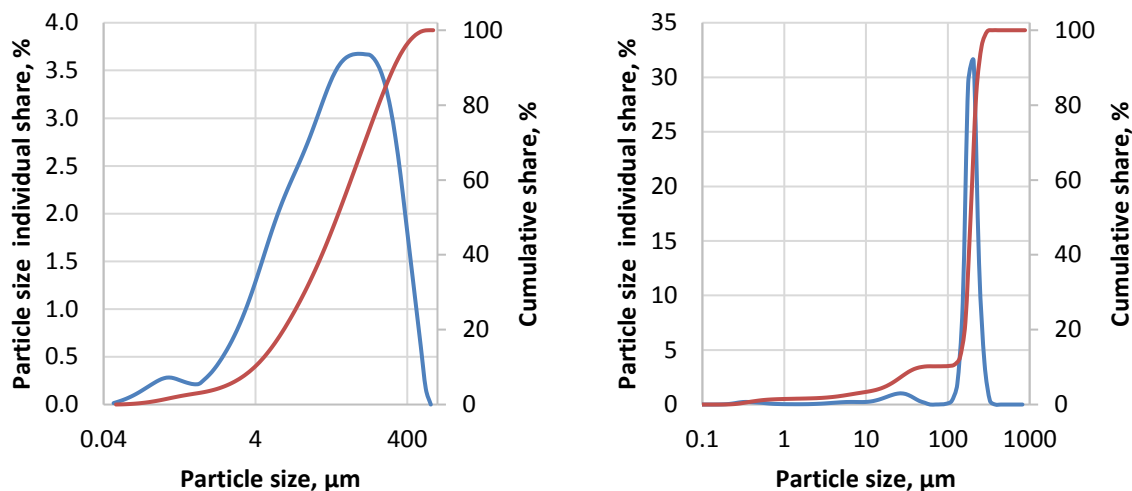

Fig. 1. Granulometric analyses of lateritic material (sample 1 -left and sample 2 -right)

In both samples, nickel-bearing minerals consist mainly of magnesium and aluminum silicates (serpentine minerals, actinolite, vermiculite, schuchardite), nontronite, and pimelite. Other ore minerals are magnetite, chromite, hematite, goethite, and ilmenite. The contribution of gangue and ore minerals differ in both tested samples.

Table 1. Chemical composition of studied samples determined by XRF analysis

Element/content %	Ni	Fe	Mg	C	O	Al	Si	Ca	Mn	Co
Sample 1	0.78	7.50	16.5	1.66	46.0	0.74	25.2	0.95	0.11	0.01
Sample 2	1.10	10.9	11.7	2.68	42.1	2.99	26.4	1.08	0.24	0.03

3.2 Preliminary tests

The preliminary results (Fig. 2) showed that the increase of temperature was beneficial for the leaching kinetics and recovery of all studied metals, particularly of nickel and iron.
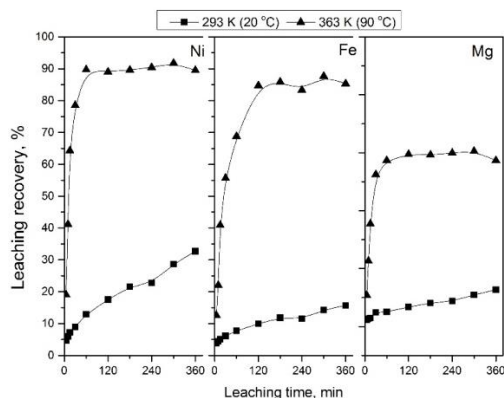

Fig. 2. The kinetics of Ni, Fe, and Mg leaching vs temperature; 1 M H_2SO_4, S/L 1/10, 400 rpm

It can be seen from Fig. 2 that at 90°C Ni recovery quickly reaches plateau at 90%. At the same conditions, recovery of Fe and Mg reach 85 and 60%, respectively. Hence, it was decided to end the process after 3 hours. It was presumed that the recovery of magnesium could be improved by

changing solid-to-liquid ratio or acid concentration. The new values of these factors were expected to alter process responses and, at the same time, to provide adequate slurry density. Considering the oxide nature of the examined material leaching was performed with two acids, i.e., H_2SO_4 and HCl, without the addition of an oxidant. Besides, the choice of acids was dictated by availability and economic reasons.

3.3 The first DOE cycle

Based on the material characteristic and the preliminary results, it was decided to examine the effect of four factors, i.e., the material sample, solid-to-liquid ratio, acid type and acid concentration. Since the number of protons differs in H_2SO_4 and HCl acids, their concentrations were expressed in normality units (N). Further experiments were planned according to the 2^{4-1} fractional factorial design. Table 2 shows the design matrix with results from the DOE 1 cycle.

Table 2. Experimental matrix and leaching recoveries obtained in the DOE 1, 90 °C, 400 rpm

Run Order	Material sample (A)	S/L ratio (B)	Acid type (C)	Acid conc., N (D)	Recovery, %		
					Ni	Fe	Mg
1	1	1/10	H_2SO_4	3	90.2	78.5	79.3
2	2	1/10	H_2SO_4	4	72.8	64.3	87.2
3	2	1/6	H_2SO_4	3	70.7	59.8	73.1
4	2	1/6	HCl	4	69.9	65.4	74.5
5	1	1/6	H_2SO_4	4	81.2	70.6	45.7
6	1	1/10	HCl	4	79.9	75.4	47.0
7	1	1/6	HCl	3	93.8	84.2	54.3
8	2	1/10	HCl	3	72.6	69.0	58.2

The performed analysis allowed to identify the active factors, which significantly influenced the metal recoveries. The other effects were negligible and were assumed as an experimental noise. The obtained results were statistically analyzed using Minitab software. The ANOVA for Ni, Fe, and Mg recovery (Table 3) together with the suitable factorial plots (Figs. 4-6) summarize the results and illustrate the effects of factors on the leaching recoveries of Ni, Fe, and Mg.
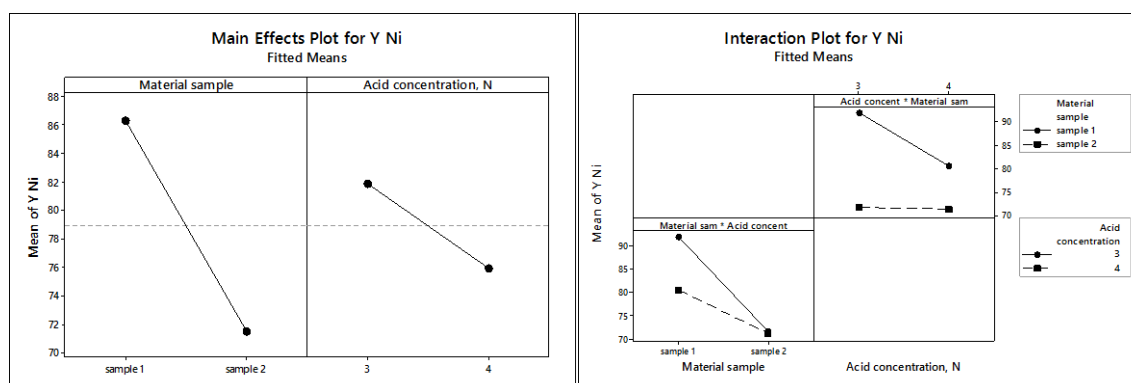

Fig. 3. The main effects and interaction plots for Ni recovery

It was found that only two main factors strongly affected Ni recovery, i.e., the material sample (A) and acid concentration (D). The influence of factor A on the Ni recovery can be attributed to the difference in the particle size of both samples. The smaller the particle size, the greater the surface area, and consequently the higher reactivity of the material. Hence, the recovery of Ni from finer sample 1 was higher of about 16%. Found interaction AD between the factors means that the recovery of Ni was affected by the factor D, but its effect was dependent on the level of factor A.

Table 3. The analysis of variance of Ni, Fe and Mg recoveries in the DOE 1

| Response	Source	DF	Seq SS	Adj MS	F-value	P-value
	ANOVA					
Ni recovery (%)	Main Effects	2	506.98	253.490	77.80	0.001
	A -Material sample	1	436.99	436.986	134.11	0.000
	D- Acid conc.	1	69.99	69.993	21.48	0.010
	2-Way Interactions	1	61.94	61.937	19.01	0.012
	AD	1	61.94	61.937	19.01	0.012
	Error	4	13.03	3.258		
	Total	7	581.95			
Fe recovery (%)	Main Effects	3	399.48	133.159	23.92	0.013
	A- Material sample	1	314.36	314.364	56.47	0.005
	C- Acid type	1	53.93	53.932	9.69	0.053
	D – Acid conc.	1	31.18	31.181	5.60	0.099
	2-Way interactions	1	39.18	39.183	7.04	0.077
	AD	1	39.18	39.183	7.04	0.077
	Error	3	16.70	5.567		
	Total	7	455.36			
Mg recovery (%)	Main Effects	3	895.19	298.40	5.10	0.078
	A- Material sample	1	554.15	554.15	9.46	0.107
	C- Acid type	1	327.39	327.39	5.59	0.054
	D- Acid conc.	1	13.65	13.65	0.23	0.662
	2-Way Interactions	1	634.84	634.84	10.84	0.046
	AD	1	634.84	634.84	10.84	0.046
	Error	3	175.68	58.56		
	Total	7	1705.71			
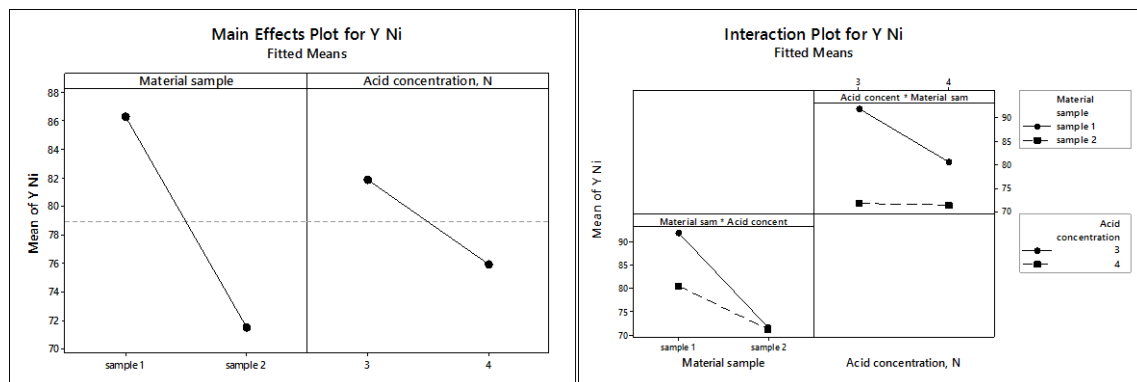
Fig. 3. The main effects and interaction plots for Ni recovery

Interaction plot (Fig. 3) shows that the increase of acid concentration from 3 to 4 N caused about 10% decrease of Ni recovery from sample 1. The likely cause of this negative influence was the growth of leaching solution viscosity, as a result of the intensive dissolution of minerals present in sample 1. However, further research is necessary to confirm the conclusion. The comparison of viscosity values is presented in Fig. 4. It shows that the viscosities were higher (on average) at higher S/L ratio (1/6), and when sulfuric acid was used as a leaching agent. The change in acid concentration had no effect on Ni recovery from sample 2. These results indicate that acid concentration should be chosen carefully, i.e., it should be low enough to leach Ni-bearing minerals but not too high to dissolve gangue constituents of the ore.
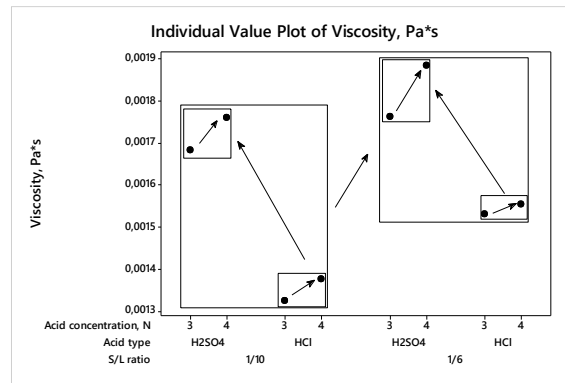
Fig. 4. The dependence of viscosity on factors tested in DOE 1

Extraction of iron was influenced by the same factors that affected nickel recovery (A, D, and AD), and additionally by the acid type (C), (Table 3, Fig. 5). The direction of changes these factors caused was the same, indicating a strong correlation between the recovery of both metals and implying that their separation may be infeasible. However, a way to increase the nickel to iron concentration ratio can be the rational choice of acid type. In contrast to Ni, this factor had an influence on Fe recovery, increasing it by 5%, when HCl was used. The hydrochloric acid is commonly regarded as a more efficient leaching agent than sulfuric acid. It is because chlorides are more soluble than sulfates, and reactions of their formation in leaching processes are significantly faster than reactions of sulfuric acid.
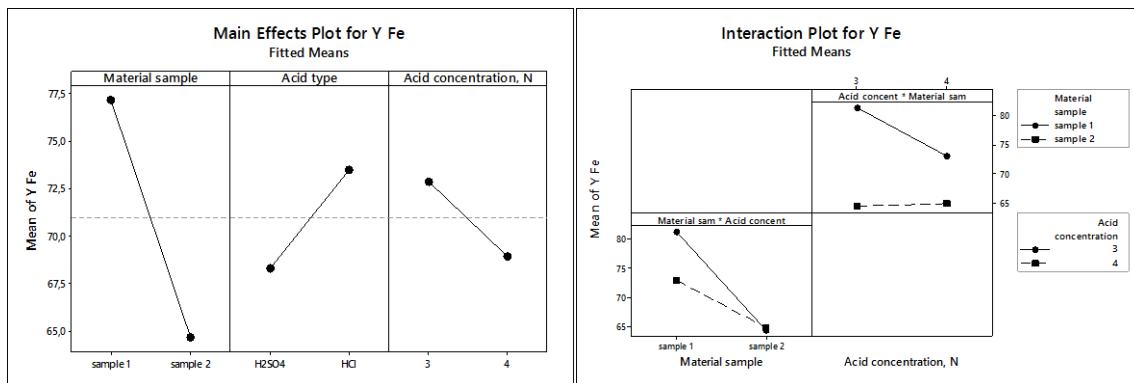

Fig. 5. The main effects and interaction plots for Fe recovery

In the case of Mg, its dissolution was affected by two main factors: A, C, and one interaction AD (Fig. 6). The analysis showed that the dependencies observed for Mg dissolution differed from those found for Ni and Fe. Significantly more magnesium was leached with H_2SO_4 (about 15%) and from sample 2 (about 35%). The interactions revealed, that to obtain higher magnesium recovery from sample 1, one shall use less concentrated acid, while leaching of sample 2 would require stronger acid.
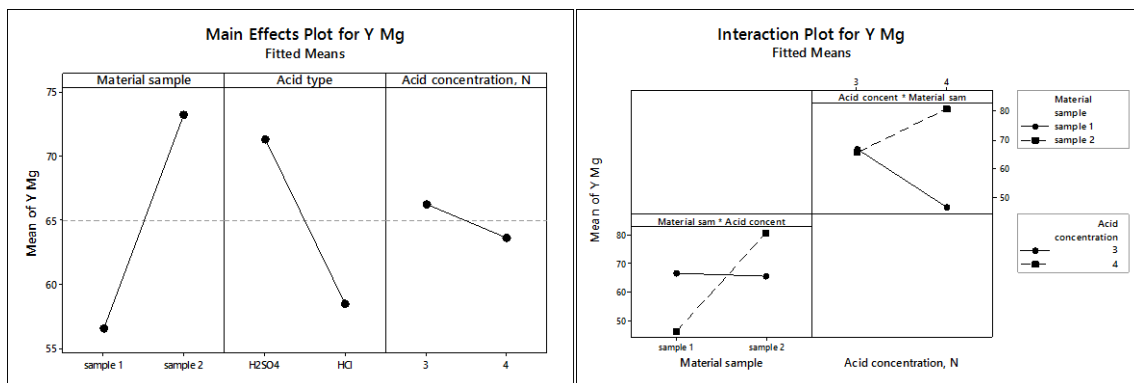

Fig. 6. The main effects and interaction plots for Mg recovery

The overall effect of material sample may be attributed to the differences in the content of host gangue minerals. Several studies on leaching behavior of Ni laterites confirm a strong mineralogy and chemistry dependent behavior (LUO et al., 2010; HUNTER et al., 2013; XU et al., 2013; MACCARTHY et al., 2014a; MACCARTHY et al., 2014b; QUAST et al., 2013).

The following empirical models were found to describe the effect of studied factors on metals recoveries:

$$Ni\ Recovery = 99.59 - 26.87\ A - 5.92\ D + 5.56\ AD \qquad (3)$$
$$R\text{-}Sq = 97.76\%,\ R\text{-}Sq(adj) = 96.08\%,\ R\text{-}Sq(pred) = 91.04\%$$

$$Fe\ Recovery = 84.74 - 21.76\ A + 2.596\ C - 3.95\ D + 4.43\ AD \qquad (4)$$
$$R\text{-}Sq = 96.33\%,\ R\text{-}Sq(adj) = 91.44\%,\ R\text{-}Sq(pred) = 73.92\%$$

$$Mg\ Recovery = 74.1 - 54.0\ A - 6.40\ C - 2.61\ D + 17.82\ AD \qquad (5)$$
$$R\text{-}sq = 89.70\%,\ R\text{-}sq(adj) = 75.97\%,\ R\text{-}sq(pred) = 26.76\%$$

The fitted regression models are of high quality. Small differences between R-squared and R-adjusted statistics indicate that all selected models were not over-fitted. High R-predicted values demonstrate that models calculated for Ni and Fe recovery can very well predict new observations. The correlation 5, obtained for Mg recovery, has lower prediction ability but fairly well reproduces the experimental data.

3.4 The second DOE cycle

When drawing inferences from the designed experiment, it is necessary to consider limitations of conclusions that is all the circumstances affecting the process: the process conditions and tested material. In our case, both samples of Ni – bearing material were leached at constant temperature and constant stirring speed. Therefore, next DOE cycle should explain whether these constant factors can in any way affect the efficiency of metals recovery. The new levels of leaching temperature and stirring speed (50°C, 200 rpm) were set in order to find whether the process can be equally effective as at higher values of these factors. The results from DOE 1 also demonstrated that HCl was slightly better leaching agent and that lower acid concentration was enough to obtain Ni recovery above 90%. Therefore, it was decided to add the new level to that factor and to investigate how 2 N acid will affect the leaching of the examined material.

Thus, in the next series of experiments, the effects of factor A and D (new levels) as well as new factors (temperature – E and stirring speed – F) were examined. The DOE 2 matrix (Table 4) shows new parameters and their levels. Table 5 presents the ANOVA of the metals recovery, whereas Figs. 7 – 9 show the Main effects plots.

It was found that three main factors, i.e., the material sample (A), acid concentration (D), and temperature (E), were significant for Ni recovery (Table 5, Fig. 7). However, the greatest change of the response produced factor A (19%) and factor E (17%). The increase of acid concentration from 2 to 3 N improved Ni recovery of about 6% indicating that the higher level was the most optimal. The data suggests that stirring speed did not have a marked on Ni leaching recovery.

Table 4. The experimental matrix and the leaching recoveries obtained in the DOE 2

Run Order	Material sample (A)	Acid conc., N (D)	Temp. °C (E)	Stirring speed, rpm (F)	Recovery, %		
					Ni	Fe	Mg
1	Sample 1	2	50	200	69.1	38.6	36.7
2	Sample 1	2	90	400	78.8	66.2	56.0
3	Sample 2	3	90	400	72.4	67.2	60.7
4	Sample 1	3	90	200	86.7	85.2	81.0
5	Sample 1	3	50	400	71.2	52.8	62.1
6	Sample 2	2	90	200	65.4	55.8	53.6
7	Sample 2	2	50	400	45.7	25.5	40.9
8	Sample 2	3	50	200	52.0	35.1	45.1

Table 5. The analysis of variance of Ni, Fe and Mg recoveries in the DOE 2

|				ANOVA		
Response	Source	DF	Seq SS	Adj MS	F-value	P-value
Ni recovery (%)	Main Effects	3	1217.00	405.667	43.36	0.002
	A-Material sample	1	618.22	618.216	66.08	0.001
	D- Acid conc.	1	68.26	68.265	7.30	0.054
	E-Temperature	1	530.52	530.521	56.71	0.002
	Residual Error	4	37.42	9.356		
	Total	7	1254.42			
Fe recovery (%)	Main Effects	3	2683.33	894.44	140.47	0.000
	A-Material sample	1	438.99	438.99	68.94	0.001
	D- Acid conc.	1	368.34	368.34	57.84	0.002
	E-Temperature	5	1876.00	1876.00	294.62	0.000
	Residual Error	4	25.47	6.37		
	Total	7	2708.80			
Mg recovery (%)	Main Effects	2	516.25	258.126	67.80	0.000
	D- Acid conc.	1	49.58	49.58	13.02	0.015
	E- Temperature	1	466.67	466.67	122.58	0.000
	Residual Error	5	19.04	3.807		
	Total	7	535.29			
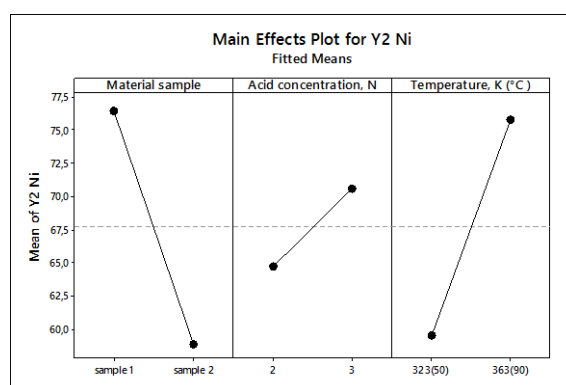
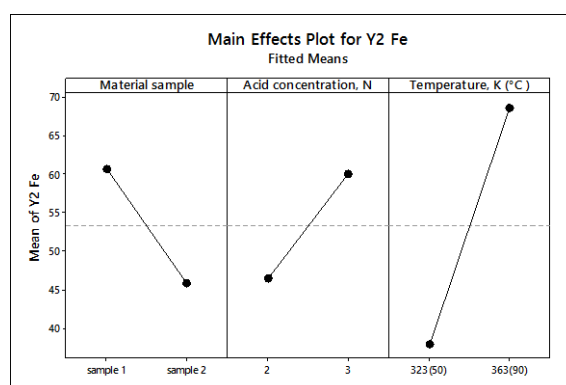Fig. 7. The Main effect plot for Ni recovery

Fig. 8. The Main effect plot for Fe recovery

The correlation between Ni and Fe recovery, revealed during DOE 1, is also evident in the second DOE, and it is confirmed by the same active factors influencing recovery of iron (Fig. 8). Their importance, however, was much more pronounced. For example, the increase of the temperature improved Fe dissolution of ~30%, while the influence of the material sample and HCl concentration was half lower. For both Ni and Fe, their recovery was higher from sample 1.

Two factors were the most influential for the dissolution of magnesium during the DOE 2, i.e., the acid concentration and temperature. Similarly to Ni and Fe, the higher levels of both factors were beneficial for the response, increasing Mg recovery of ~15% and 5%, respectively (Fig. 9).

Above observations reflected a strong mineralogy- and temperature-dependent leaching behavior. The intensive Ni release at higher temperature co-occurred with a significant increase in the leaching of accompanying impurity metals - Fe and Mg.
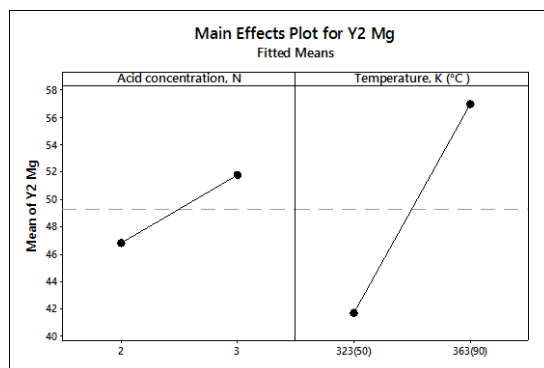

Fig. 9. The Main effect plot for Mg recovery

The regression analysis of experimental results leads to the correlations presented below:

$$Ni\ recovery = 24.56 - 8.79A + 5.84D + 0.41E \tag{6}$$
$$R\text{-}Sq = 97.02\%,\ R\text{-}Sq(adj) = 94.78\%,\ R\text{-}Sq(pred) = 88.07\%$$

$$Fe\ recovery = -34.23 - 7.41A + 13.57D + 0.77E \tag{7}$$
$$R\text{-}Sq = 99.06\%,\ R\text{-}Sq(adj) = 98.35\%,\ R\text{-}Sq(pred) = 96.24\%$$

$$Mg\ recovery = 10.14 + 4.98D + 0.38E \tag{8}$$
$$R\text{-}Sq = 96.44\%,\ R\text{-}Sq(adj) = 95.02\%,\ R\text{-}Sq(pred) = 90.90\%$$

All correlations derived in the second DOE cycle fit the data very well. It is confirmed by the goodness-of-fit statistics, especially high values of the R-squared, predicted R-squared and adjusted R-squared parameters. The P-value in correlations (6), (7) and (8) is lower than 0.01, meaning they are significant at the 99% confidence level.

3.5 The confirmation tests

To evaluate the correctness of the obtained statistical models, we carried out the experiments under optimal conditions with goals stressed to maximize Ni recovery and minimize Fe and Mg recovery. All response predictions were determined with help of Minitab software.

According to Table 6, the results of confirmatory tests were within low and high predicted values in 95% confidence intervals (CI). The results indicated that the experimental responses were quite close to the predicted values, confirming the validity of the models. From the four presented models, the highest Ni recoveries were obtained for sample 1 leaching, at 90 °C, with 3 N sulfuric acid. The dissolution of Fe and Mg was below 84 and 55%, respectively. The remaining model configurations allowed for a noticeable decrease of Fe and Mg recoveries, although Ni recovery was not satisfactory.

4. Conclusions

This work demonstrates that application of sequential experimentation is extremely beneficial in establishing the cause and effect relation between process variables and the measured responses. The arrangement of designed experiments in two sequences allowed gathering a lot of valuable data at the lowest number of tests.

The data showed that leaching recoveries of Ni, Fe and Mg depended primarily on the material sample, temperature and acid concentration, indicating a strong mineralogy/chemistry and temperature dependent behavior. These dependencies were supported by the mathematical models, which gave correlations of great statistical importance. The conducted confirmation tests proved high

prediction ability of the derived models and showed that sample 1 leaching at 90 °C, with 3 N sulfuric acid led to high Ni recoveries (~ 93%) and reduced dissolution of iron ~(~83%) and magnesium (55%).

We conclude that the proposed method can be used to find optimal conditions for ore samples containing a variable proportion of minerals and can significantly shorten the time for experimentation.

Table 6. The predicted responses with confidence levels and corresponding responses from confirmation tests

Response	Fit	SE Fit	95% CI	95% PI	Confirmation test
Response prediction for DOE1: sample 1, 3N H_2SO_4					
Y Ni	92.02	1.28	(88.47; 95.56)	(85.88; 98.16)	92.8
Y Fe	78.78	1.87	(72.84; 84.72)	(69.21; 88.35)	83.2
Y Mg	73.21	6.05	(53.95; 92.46)	(42.16; 104.25)	54.7
Response prediction for DOE1: sample 2, 3N HCl					
Y Ni	71.67	1.28	(68.13; 75.22)	(65.53; 77.81)	70.44
Y Fe	67.01	1.87	(61.07; 72.95)	(57.44; 76.58)	63.76
Y Mg	59.24	6.05	(39.99; 78.49)	(28,20; 90,29)	45.09
Response prediction for DOE 2: sample 2, 3N HCl, 86°C					
Y2 Ni	68.39	2.07	(62,65; 74,13)	(58,14; 78,64)	67.22
Y2 Fe	65.06	1.71	(60,33; 69,80)	(56,61; 73,52)	62.58
Y2 Mg	57.99	1.12	(55,10; 60,88)	(52,20; 63,78)	59.63
Response prediction for DOE 2: sample 1, 2N HCl, 90°C					
Y2 Ni	81.68	2.16	(75.68; 87.69)	(71,28; 92,08)	85.5
Y2 Fe	69.22	1.78	(64,27; 74,18)	(60,64; 77,81)	77.5
Y2 Mg	54.47	1.19	(51,40; 57,54)	(48,59; 60,35)	53.7

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