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STUDY OF THE REDUCTION OF THE SERPENTINITOS PARTICLE SIZE TO INCREASE THE EFFICIENCY OF THE PROCESSES OF MINERAL CARBONATION

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Abstract. Mineral carbonation processes have great potential for use as carbon capture and storage (CCS) technology. Mineral carbonation, using silicate rocks such as serpentinite, for the sequestration of CO_2 through the formation of thermodynamic and environmentally stable products such as carbonates, presents itself as an economically efficient technology. This work presents a study on the leaching process of a serpentinite sample resistant to the leaching process. Experimental planning techniques were applied with the objective to optimize the process and maximize the Mg extraction of the serpentinite sample. The results show that as a result of the reduction of the sample size, a mechanical activation of the material occurred and the extraction of Mg was maximized.

Keywords: Leaching, Serpentinite, optimization, particle size, mineral carbonation

1. INTRODUCTION

Mineral carbonation (CM) processes are considered the most promising since they would allow the capture and fixation of large amounts of carbon dioxide (CO_2) emitted by the energy sector through the production of thermally and thermodynamically stable carbonates. The process of CM involves four steps: 1) Leaching of Mg, Ca and Fe of silicate rocks, 2) purification of the solution by precipitation of Fe and other impurities, 3) basic carbonation of Mg and Ca and finally, 4) Recovery of chemical inputs used (Arce et al., 2017a; ARCE et al., 2015; Azdarpour et al., 2015).

Although this process is promising, it faces major challenges related to the various efficiencies found in the leaching stage of Mg, Fe and Ca, and in some cases was less than 30%, several authors point out that this difference in the extractions efficiencies of Mg, Fe and Ca are due to the various characteristics of the silicate rocks used. In order for this process to be environmentally attractive the process efficiency mainly from step 1 and step 4 should be increased significantly (Dichicco et al., 2015; Hemmati et al., 2014).

Arce et al. (2017a) e (2017b) indicate that silicate rocks within them serpentinites may be more resistant to leaching processes if they had minerals of the pyroxene family such as amphiboles and talc; moreover, coils with Al^{3+} contents greater than 1% inside the serpentine could cause greater resistance to the leaching processes. However, both articles indicate that mechanical activations, i.e reduction of particle size and chemical activation (increase of acid concentration) could reduce this resistance, however, little is known of what would be the energy consumption if a mechanical activation of the material were used, and which particle size would be ideal for significantly increasing efficiency. This work presents a Monte Carlo Simulation Optimization (SMC) model for the prediction and maximization of Mg extraction from the serpentinite rock leaching process. The data for application of the model were obtained through an exploratory process analysis using Taguchi experimental planning and presented by Vieira (2016). For the optimization of the process via monte carlo simulation, Crystal Ball software and the optimizer OPTQUEST were used.

2. MATERIALS AND METHOD

2.1 Samples

In this work a serpentinite sample resistant to leaching was used, supplied by Pedras Congonhas Company, located in Belo Horizonte. The sample was extracted from the Nova Lima mine, located in the Minas Gerais State. In this work the sample will be denominated S-MG. Table 1 shows the chemical composition of the serpentinite sample. Figure 1 shows the sample of S-MG in nature.

Table 1. Chemical composition of material

Elements	Mg	Al	Si	Ca	Cr	Mn	Fe	Ni
%w/w	9.58	0.66	21.74	1.52	0.48	0.17	8.60	0.13
Oxides	MgO	Al_2O_3	SiO_2	CaO	Cr_2O_3	MnO	Fe_2O_3	NiO
%w/w	14.84	1.02	36.39	1.39	0.43	0.13	7.15	0.09

Figure 1. Sample of the rock in natura (S-MG)

2.2 Leaching process

The leaching process was performed using a aqueous solution of HCl 4 molar for extract metals such as Mg, Fe and Ca from S-MG. The reaction system consists of a 250 ml glass reactor, magnetic stirrer with heating controlled and condenser. By the ways, a reaction time of 2 hours, 100°C of temperature and agitation of 600 rpm was used.

The product of the dissolution reaction is a slurry (liquid + suspended solids). Through the filtration process a rich aqueous solution of Mg, Fe and Ca is obtained. The concentrations of Mg in the solution obtained were determined by atomic absorption analysis. Figure 2 shows the leaching process.
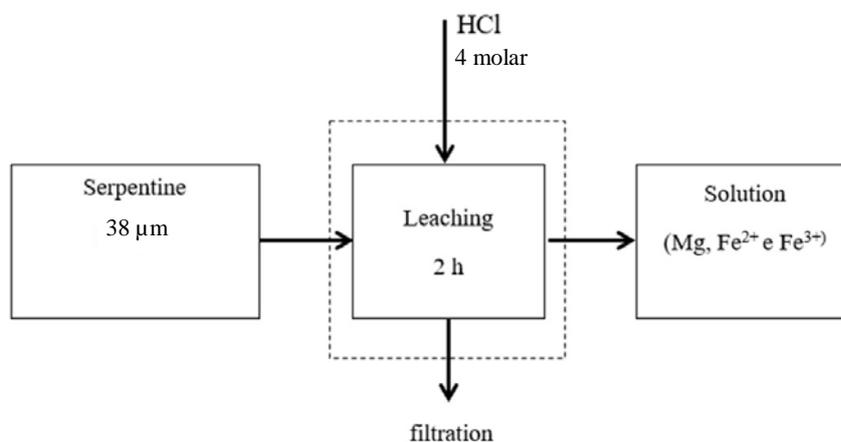

Figure 2. leaching process.

2.3 Simulation optimization of Monte Carlo and problem modeling

Monte Carlo simulation was used to optimization of mg extraction. This simulation is an adequate tool to predict the behavior of variations in problems involving uncertainty. For the application of the model of optimization and simulation of monte Carlo it is necessary to know the process to define the limits of uncertainty (Baudry et al., 2018). The simulation technique of monte Carlo allows to explore the uncertainties in stochastic problems and allows to estimate values for process factors (Li and Li, 2009; Rezaie et al., 2007).

The simulation was realized using the results of the exploratory research of the leaching process presented by Vieira (2016) and shown on Table 2. Table 2 presents data on the serpentinite leaching process (S-MG) under different conditions. The temperature (X_1), HCl concentration (X_2) and the particle size of material (X_3) are the process independent variables (i.e. the inputs data base). The extraction rate of Mg (Y) is the variable response that one wishes to maximize.

Table 2. Leach process input and output matrix.

X_1 (T°C)	X_2 (C_{HCl})	X_3 (μm)	Y (% Mg)
30	1	69	2
30	2	328	1
30	4	550	4
50	1	328	4
50	2	550	5
50	4	69	16
70	1	550	17
70	2	69	32
70	4	328	30

Source: Adapted from (Vieira, 2016)

3. RESULTS AND DISCUSSION

Through the data presented by Vieira (2016), a mathematical model of linear regression was described to describe the extraction of Mg in the leaching process. The R^2-adjusted model was 88%, which indicates a strong correlation between the X and Y variables and the value-p values were lower than the 5% significance level.

In Table 3 we can see that the simulation predicted a 51% Mg extraction by adjusting the dependent variables at 100 ° C (X_1), 4 M of HCl concentration and 38 μm of particle size. Arce et al., (2017b) presents a study on the influence of physico-chemical properties on the leaching process of Brazilian serpentinites, for the same material used in this work. The experiment performed by Arce et al. (2017b) obtained a 34% Mg extraction for the S-MG sample with particle size of 300 μm, when the authors reduced the particle size until 69 μm the efficiency increased only 2%. In this work we explore serpentinite with 38 μm particle size, as provided for in monte Carlo model. Interestingly, it was observed that the resistance to leaching of S-MG rock could be minimized, reaching a Mg extraction of 86%, higher than those reported in Vieira (2016) and Arce et al. (2017b).

Table 3 shows the results of extraction of mg obtained by Arce et al. (2017b), the forecast of extraction obtained in the model of simulation of monte Carlo and the experimental result obtained from the conditions of optimization proposed by the model of monte Carlo. It can be observed that when reducing the particle size of the serpentinite to 38 μm the extraction of Mg increased by 35%, when compared to the results obtained by Arce et al. (2017b).

Table 3: Experimental result.

Test	X_1 T(˚C)	X_2 C_{HCl}	X_3 μm	Y Mg (%)
S-MG[a]	100	4	300	34
S-MG[b]	100	4	69	51
S-MG[a]	100	4	69	36
S-MG	100	4	38	86

[a]from Arce et al. (2017b), [b]condition predicted by Monte Carlo Simulation

4. CONCLUSIONS

This work evaluated the leaching process of a Brazilian serpentinite. From a simulation of monte Carlo was determined the experimental condition that maximizes the extraction of Mg of the material. In the experimental tests, it was verified that the granulometry proposed by the simulation model reduced the resistance to leaching of the S-MG rock, increasing the Mg extraction when compared to other works in the literature. With the increase of Mg extraction, the use of this material in processes of mineral carbonation becomes viable. The use of computational tools such as monte Carlo simulation is an important tool for estimating experimental results when the process studied is already known.

The next steps of this research is to carry out a study on the energy consumption to activate and to maximize the extraction of Mg.

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