

STUDY OF VARIABLES IN THE ACID DISSOLUTION OF BRAZILIAN SERPENTINITE FOR CO₂ MINERAL CARBONATION

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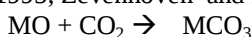
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Abstract. The mineral carbonation is a reaction between silicates rocks (Olivines, serpentines and Wollastonites) and carbon dioxides (CO₂) to form thermodynamically stable inorganic carbonates in the nature. This technology gets to receive more attention, due to that, besides to sequester the CO₂, the carbonates produced could be used as feedstock in others industrial processes. In order to make this technology sustainable, enhance carbonation reaction rates has become a challenge. For that reason were proposed pretreatment methods of silicates rocks. Those methods are called as “activation”, which can be mechanical, thermal and/or chemical. However, chemical activation achieved to enhance substantially the carbonation reaction rates from 70% until 100% so far. Chemical activation is the extraction of metals from silicates rocks (i.e. Mg, Ca and Fe) using acid or bases, in order to obtain an aqueous solution with high concentration of that metals which is carbonated. However, chemical activation with acid solutions shows higher metal extraction percentage. Several studies have used HCl solutions; meanwhile, there is no agreement among these studies about the percentage of extraction. Thereby, there is necessity of more investigations on the metals extraction percentage in function of physical and chemical parameters of silicate rocks. On the other hand, Brazil presents a large availability of serpentinite with a potential use in mineral carbonation process. However, there is scarce information on the metals extraction performance from serpentinite using chemical activation. In this study, Brazilian serpentine from Midwest regions was selected. Firstly, it was characterized to determine the mineralogy composition, elemental analysis, physical structure and thermal behavior. The metals extraction percentage was evaluated in function of factors, such as: temperature, particle size and concentration of HCl solution. A design of experiment (DoE) was used, which considered three factors, three levels using a matrix L9. Each experiment was realized for 2 h of reaction time. The results have shown a carbon sequestration potential of 1,84 t_{SERP}/t_{CO2SEQ}. The chemical activation of Brazilian serpentinite presented a high extraction percentage of Mg achieving a value of 43%. The conditions of this extraction were 70°C, concentration 1M of HCl and particle size around of 256 μm. This extraction percentage of Mg was higher than that founded in the literature, which are between 9% and 40%. Statistical analysis of results showed that the temperature and particle size are more significantly. The high extraction percentage of Mg from Brazilian serpentinites was attributed to low concentration of SiO₂, the crystalline structure of chrysotile and the high reactivity of brucite founded in this serpentine.

Keywords: Brazilian serpentine, chemical activation, HCl solution, CO₂ sequestration, mineral carbonation

1. INTRODUCTION

Mineral carbonation is a process based on a natural phenomenon of carbon dioxide (CO₂) sequestration known as rock disaggregation. This phenomenon played an important role in historical atmospheric CO₂ concentration reduction when the Earth was forming (Sipilä et al., 2008). This process could be useful, since CO₂ emissions must be reduced by 34% in the near future to maintain the balance in the carbon cycle (Arce et al., 2014). Mineral carbonation is a chemical reaction between CO₂ and metal oxides to form solid inorganic carbonates. This is shown in reaction R1 (Lackner et al., 1995; Zevenhoven and Kavaliauskaite, 2004; Olajire, 2013 and Sanna et al., 2014).

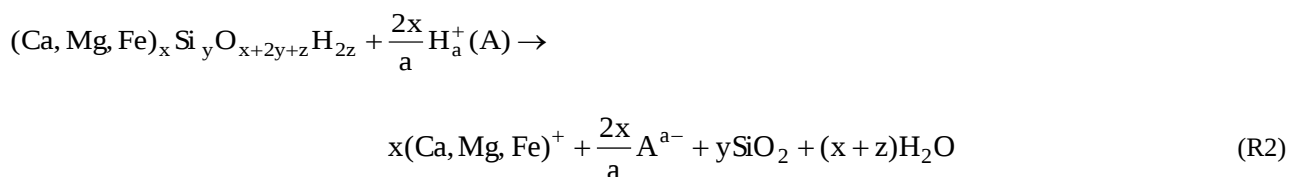


(R1)

According to Chang et al. (2012), mineral carbonation can contribute for CO₂ sequestration when applied near facilities that emit CO₂. The main advantages of carbonation are: (a) it is safe for the environment because the products are stable; and (b) there is a vast amount of material available around the world (Sipilä et al., 2008; Sanna et al., 2014 and Moazzem et al., 2013). In addition, it is possible to use these stable products as raw materials in other applications (Sipilä et al., 2008; zevenhoven and Kavaliauskaite, 2004; Bobicki et al., 2012; Fagerlund et al., 2012; Nduagu et al., 2012).

In order for this process to be efficient, it must use materials with high levels of Ca, Mg, and Fe and low levels of Na and K. This characteristic is important because Ca and Mg are much less soluble than Na and K, which avoids returning CO₂ into the atmosphere. According to Sipilä et al. (2008), there are many materials that could be used in mineral carbonation that are available worldwide. These materials include rocks containing silicates (basalt, feldspar, olivine, pyroxene, serpentine, talc, wollastonite, etc.) and industrial waste (waste from cement production and mining, fly ash from burning lignite, steel slag, etc.).

However, mineral carbonation faces many challenges, such as increasing the reactivity of these materials in order to increase the carbonation reaction kinetic and reduce the energy required in the process (Lackner et al., 1995; Olajire, 2013; Sanna et al., 2014; Wang and Maroto-Valer, 2011; Fricker and Park A-H A, 2013 and Park A-H A and Fan, 2004). For this reason, methods to pre-treat the material have been proposed. These methods are called activation, which can be mechanical, thermal, or chemical. Chemical activation is a leaching process that uses a solvent (weak and strong acids, molten salts, bioleaching, ammonium, base) to extract metals such as Ca, Mg, and Fe (Bobicki et al., 2012, Maroto-Valer et al., 2004, Teir et al., 2007a). In chemical activation, the solvent solution penetrates into the material particle and encounters the surface of metal oxides. Regarding a chemical activation of a silicate rock with a strong acid, at this interface, the extraction expressed in reaction R2 takes places.



According to Kosuge et al. (1995), as the reaction takes place, the pores of the particles are blocked when a layer of amorphous silica hydrate is formed. The internal surface area becomes inaccessible and the effectiveness of Ca, Mg, and Fe extraction is reduced as the process continues (Van Essendelft and Schobert, 2009a). Van Essendelft and Schobert (2009a) and (2009b) carried out studies that assumed that the extraction efficiency of materials was first limited by surface kinetics and then by diffusion in the layers within the material. The diffusion in layers within the material obviously has an important role in the efficiency of chemical activation because of diffusivity. Effective diffusivity for a typical diffusion of a porous material varies from around 9.73×10^{-14} to $3.22 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$. However, free diffusion through aqueous solutions is around $10^{-9} \text{ m}^2 \text{ s}^{-1}$. Effective diffusivity is related to free diffusivity by tortuosity, porosity, surface area, and cross-section of the material. In very porous systems, effective diffusivity can be the same as free diffusivity. However, in systems that aren't very porous, which have dense layers, diffusion can be reduced by three times, making it difficult to extract Ca, Mg e Fe (Van Essendelft and Schobert, 2009b).

2. MATERIALS AND METHODS

Most of the materials pointed out by Sipilä et al. (2008) to be used in mineral carbonation are available in Brazil. Not all of these materials are found in amounts that can be used commercially. However, according to data from the Brazilian Mineral Production Department (Pereira, 2013), Brazil has a reserve of 200 million metric tons of asbestos fiber (Chrysotile). The aim of this paper is to understand the Mg and Fe leaching from Brazilian serpentinite using HCl aqueous solution for the mineral carbonation processes as a CCS technology. These results were compared to Mg leaching of the literature. In this paper it was used serpentinite $Mg_3Si_2O_5(OH)_4$ provided by SAMA S.A. – Minerações Associadas, which, was extracted from Cana Brava mine (located at) Goiás State. This material was named S-GO.

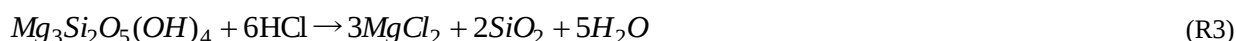
2.1. Carbonation potential (R_{CO_2})

The methodology described by Goff et al. (2000) was used to determine carbonation potential (R_{CO_2}). This method uses Eq. 1 and the total molar fractions of Ca, Mg, and Fe obtained in X-Ray Fluorescence (XRF) analysis of the materials. On the R_{CO_2} calculation, the contents of Ca, Mg, and Fe are given in % w/w for 100 g of material. They are assumed to be totally converted into carbonates. The sum $\Sigma(Ca, Mg, Fe)$ is the sum of the molar fractions of these elements; MM_{CO_2} is the molecular mass of carbonic gas; t_{MP} is the tons of the material; and t_{CO_2SEQ} is the tons of CO₂ that can be sequestered. Note that the lower the R_{CO_2} value, will be the better theoretical carbonation potential of the material in the study.

$$R_{CO_2} = \frac{100}{(\sum Ca, Mg, Fe)MM_{CO_2}} \left[\frac{t_{MP}}{t_{CO_2}} \right] \quad [1]$$

2.2. Chemical Activation with HCl

To assessment to chemical activation with HCl for Mg, Fe leaching from S-GO it was considered a main reaction as shown in the reaction R3:



In this reaction were identified three factors that could influence the Mg and Fe extraction. These factors are: particle size (μm), concentration of acid aqueous solution (M), and temperature of the process ($^{\circ}C$). Then, for each factors, there are three levels (Tab. 1).

Table 1. Levels each factor identified to chemical activation with HCl.

Factor	Level		
	1 - Low	2 - Medium	3 - High
A – particle size (μm)	64	115	256
B - Concentration (M)	1	2	4
C – Temperature ($^{\circ}C$)	30	50	70

To evaluate the levels of the factors that could influence on the chemical activation it was used a L_9 matrix (Tab. 2). This matrix gives the experimental conditions for each test.

Table 2. Design of Experiment – Orthogonal arrays for L_9 .

Test	Factor Codified			Numerical Factor		
	A	B	C	Φ μm	C_{HCl} [M]	T $^{\circ}C$
1	1	1	1	64	1	30
2	1	2	2	64	2	50
3	1	3	3	64	4	70
4	2	1	2	115,5	1	50
5	2	2	3	115,5	2	70
6	2	3	1	115,5	4	30
7	3	1	3	256	1	70
8	3	2	1	256	2	30
9	3	3	2	256	4	50

2.4. Taguchi Methodology

The robustness data analisys have two important tools, the Noise Signal (S/N) and the effect of factor (m_{ij}) (Taguchi, 2005). The response variable in this paper is Mg and Fe extraction, being this the main parameters of the mineral carbonation processes, because the higher Mg and Fe extraction will obtain higher efficiency on chemical reaction involved. So, The (S/N) ratio used was “the higher is the better” (Eq. 4)

$$(S/N) = -10 \log \left(\frac{1/y^2}{N} \right) \quad [4]$$

being “y” a response variable and N represents how many times the level is repeated into the matrix L_9 . Table 6 and Table 7 were built using the Eq. 4 – 6.

$$(m_{ij}) = \left(\frac{\sum_{j=1}^n y_j}{n} \right) \quad [5]$$

$$(S/N)_{i,j} = \left(\frac{\sum_{j=1}^n (S/N)_j}{n} \right) \quad [6]$$

being “ m_{ij} ” the average of the response variables for each factor (i) in its level (j), “ y_j ” the response variables for each level (j), $(S/N)_{i,j}$ average of (S/N) values for each factor (i) in its level (j), respectively.

3. RESULTS AND DISCUSSIONS

3.1. Material characterization

The main mineral found in the S-GO sample was chrysotile, which according to Monterrubio (1991) is low-grade serpentine asbestos (this mineral generally has an average MgO content of around 43.63%). In the same sample, it was noted, that brucite and magnetite were also present. It should be pointed out that this sample contained the lowest level of lizardite. Sequential XRF analysis were carried out to determine the chemical composition of the materials. Chemical compositions of the S-GO determined using XRF was used in equation 1 to calculate the carbonation potential (R_{CO_2}). The BET area was determined using N₂ adsorption technique (Tab. 3).

Table 3. Chemical composition and BET area of S-GO.

Material	SiO ₂	MgO	CaO	Fe ₂ O ₃	Al ₂ O ₃	MnO	Cr ₂ O ₃	S _{BET} (cm ³ /g)	R _{CO2} t _M /t _{CO2}
S-GO	40,64	43,33	0,10	12,61	1,17	0,20	1,02	5,1	1,84

3.2. Performance of Mg and Fe leaching on Chemical Activation using HCl

The experimental test was realized according to Table 2. After the experimental test, the final solution (FS) was analyzed using ICP-OES technique to determine Mg, Fe. Table 4 shows the concentration value for each cations: Mg and Fe (ppm). This concentration was used to determine the extraction percentage of based on the initial weight of Mg and Fe (Tab. 5).

Table 4. Mg and Fe extracted after 2 h of leaching reaction (%).

Test	Numerical Factor			Response Variables (ppm)			V _{FS} (L)	Response Variables (% of extraction)		
	Φ μM	C _{HCl} (M)	T °C	Mg		Fe		Mg (%)		Fe (%)
1	64	1	30	2907,50		849,00	0,169	17,83		23,46
2	64	2	50	25888,00		5038,00	0,073	34,30		30,07
3	64	4	70	44793,00		8932,50	0,093	37,81		33,96
4	115,5	1	50	12108,00		2210,50	0,078	34,27		28,19
5	115,5	2	70	23844,00		4397,50	0,076	32,89		27,33
6	115,5	4	30	28992,50		6870,00	0,120	31,58		33,7
7	256	1	70	12729,50		2213,00	0,094	43,42		34,01
8	256	2	30	14141,00		2838,00	0,110	28,24		25,53
9	256	4	50	45285,50		9436,50	0,093	38,22		35,88

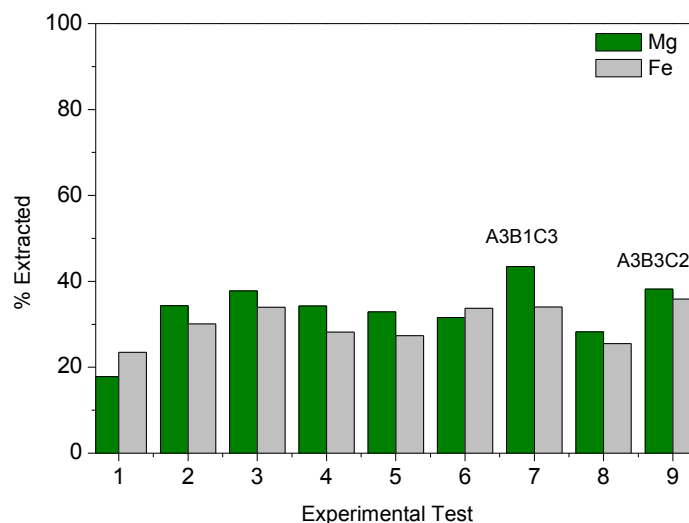


Figure 1. Performance of Mg and Fe leaching.

The best condition for Mg and Fe leaching was obtained on the experimental test number 7° and 9° respectively (Tab. 4 and Fig. 1). Where the conditions were: A (Φ) = 256 μ m, B (C_{HCl}) = 1M e C (T) = 70°C (Mg Extraction) e : A (Φ) = 256 μ m, B (C_{HCl}) = 4M e C (T) = 50°C (Fe Extraction).

In the literature there is efficiency on Mg extraction of serpentinite rocks, which, are reported in the Tab. 5. As shown on Tab. 5 higher Mg Extraction is reached using Brazilian serpentinite when compared to other serpentinites around the world. According to Van Essendelf & Schobert (2009b) and Daval et al. (2013), metal extraction in chemical activation depends on the structure of the serpentine (BET surface area) and the percentage of mass of SiO_2 it contains. Daval et al. (2013) carried out chemical activation with HCl solutions. Their Mg extraction percentage was lower than that of Teir et al. (2007a) and (Teir et al., 2007b) (Tab. 5). This difference was attributed to the physical structure of the material, specifically the BET surface area. Daval et al. (2013) reported a BET surface area four times smaller than of Teir et al. (2007a) and (2007b). However, Tab. 5 also shows that the experiment carried out by Park et al. (2003) was more efficient at extracting Mg. The BET surface area reported by Park et al. (2003) is almost half of that reported by Daval et al. (2013). However Mg extraction was much higher than reported by Teir et al. (2007a), who used a material with BET surface area seven times greater. It can be claimed that BET surface area has little influence on chemical activation, and that other parameters have to be taken into account, such as percentage of SiO_2 , reaction temperature, and reaction time (see Tab. 5).

Table 5. Physical and chemical properties of serpentinite and leaching process conditions used in chemical activation using HCl in different studies.

Mineral	BET m ² /g	SiO ₂ %	MgO %	Mg Extraction%	Acid	Condition
Serpentinite ¹	27,9	40,3	40,0	17	HCl	1M, 20°C, 1h
Serpentinite ²	7,2	42,9	40,5	9	HCl	1M, 70°C, 2h
Serpentinite ³	4,2	39,7	38,8	40	HCl	1M, 70°C, 5h
Serpentinite ⁴	n.r.	n.r.	41,0	34	HCl	1M, 25°C, 2h
Serpentinite*	5,1	40,6	43,3	43	HCl	1M, 70°C, 2h

⁽¹⁾Teir et. al., 2007 a,b. ⁽²⁾Daval et. al., 2013, ⁽³⁾Park et al., 2003; ⁽⁴⁾Alves et al., 2013.

(*) This paper

n.r. = unreported

The higher Mg Extraction was obtained using Brazilian serpentine (Tab. 5) with approximately 43%. Similar percentage of Mg extraction was obtained by Park et al (2003). But the time of reaction used by Park et al (2003) was higher than that of the time used in this paper.

Considering the reaction conditions (1M, 70°C e 2h), similar experiment was conducted by Daval et al (2013). But Mg Extraction was 5 times lower than the Mg extraction obtained in this paper. This could be due to the high content of SiO_2 in the serpentinite bearing lizardite type and magnetite from Daval et al (2013), but on the other hand, the high content of Mg from brazilian serpentinite (bearing chrysotile type, brucite and magnetite) could be the cause of the best extraction on chemical activation due to its 20% of brucite ($Mg(OH)_2$), increased Mg content and decreased Si content.

3.2. Taguchi Methods on Acid Dissolution

Using the data obtained during the leaching of Mg and Fe, it was applied statistical analyses using Taguchi methods. With the data from Tab. 4 it were calculated the effect of the factors and (S/N) ratio using Eq. [4], [5] and [6]. The results are shown on Tab. 6 and 7 respectively.

Table 6. Effect of factors and Noise Signal of Mg extraction.

Test	Factors Codified			Numerical Factors			Response Variable	y^2	S/N	Effects			
	A	B	C	ϕ (μm)	C_{HCl} (M)	T ($^{\circ}\text{C}$)	y Mg(%)			Factors (m_{ij})		Noise Signal ($S/\bar{N}$) _{i,j}	
1	1	1	1	64	1	30	17,83	317,91	25,02	m_{A1}	29,98	S/N_{A1}	29,09
2	1	2	2	64	2	50	34,3	1176,49	30,71	m_{A2}	32,91	S/N_{A2}	30,34
3	1	3	3	64	4	70	37,81	1429,60	31,55	m_{A3}	36,63	S/N_{A3}	31,14
4	2	1	2	115,5	1	50	34,27	1174,43	30,70	m_{B1}	31,84	S/N_{B1}	29,49
5	2	2	3	115,5	2	70	32,89	1081,75	30,34	m_{B2}	31,81	S/N_{B2}	30,02
6	2	3	1	115,5	4	30	31,58	997,30	29,99	m_{B3}	35,87	S/N_{B3}	31,06
7	3	1	3	256	1	70	43,42	1885,30	32,75	m_{C1}	25,88	S/N_{C1}	28,01
8	3	2	1	256	2	30	28,24	797,50	29,02	m_{C2}	35,60	S/N_{C2}	31,02
9	3	3	2	256	4	50	38,22	1460,77	31,65	m_{C3}	38,04	S/N_{C3}	31,55
Average values											33,17		30,19

Table 7. Effect of factors and Noise Signal of Fe extraction.

Test	Factors Codified			Numerical Factors			Response Variable	y^2	S/N	Effects			
	A	B	C	ϕ (μm)	C_{HCl} (M)	T ($^{\circ}\text{C}$)	y Fe (%)			Factors (m_{ij})		Noise Signal ($S/\bar{N}$) _{i,j}	
1	1	1	1	64	1	30	23,46	550,37	27,41	m_{A1}	29,16	S/N_{A1}	29,20
2	1	2	2	64	2	50	30,07	904,20	29,56	m_{A2}	29,74	S/N_{A2}	29,43
3	1	3	3	64	4	70	33,96	1153,28	30,62	m_{A3}	31,81	S/N_{A3}	29,96
4	2	1	2	115,5	1	50	28,19	794,68	29,00	m_{B1}	28,55	S/N_{B1}	29,01
5	2	2	3	115,5	2	70	27,33	746,93	28,73	m_{B2}	27,64	S/N_{B2}	28,81
6	2	3	1	115,5	4	30	33,7	1135,69	30,55	m_{B3}	34,51	S/N_{B3}	30,76
7	3	1	3	256	1	70	34,01	1156,68	30,63	m_{C1}	27,56	S/N_{C1}	28,70
8	3	2	1	256	2	30	25,53	651,78	28,14	m_{C2}	31,38	S/N_{C2}	29,89
9	3	3	2	256	4	50	35,88	1287,37	31,10	m_{C3}	31,77	S/N_{C3}	29,99
Average values											30,24		29,53

Considering Mg and Fe leaching, in the Fig. 2, it can be inferred that low and medium levels do not have influence (insignificants) on A and B factors (particle size and HCl concentration). For the other site, on C factor only the low level is insignificant, so, medium and high level have influence on Mg and Fe extraction.

Figure (2) showed the effect of factor (A,B and C) on the average value (dotted line) considering high, medium and low levels (3,2 and 1) in the Mg e Fe extraction. According to Fig. (2) the best Mg extraction would be achieved with the condition ($A_3B_3C_3$), even if this condition was not included in the L_9 Matrix. In the Fe extraction the best condition would be achieved with the condition ($A_3B_3C_2$) and ($A_3B_3C_3$), even if the first condition was included in the L_9 matrix attaching 38% of Fe extraction, but, the second condition was not included in the L_9 matrix.

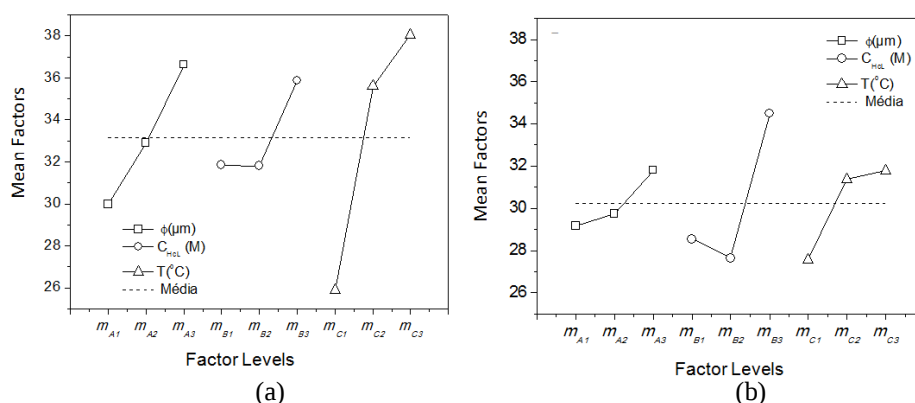


Figure 2. Effect of factors on the average of the extraction: (a) Magnesium e (b) Iron.

4. CONCLUTIONS

In this paper it was evaluated the performance of Mg and Fe extraction on S-GO, on chemical activation by leaching processes using HCl. The S-GO had the best efficiency when compared to the literature.

The higher efficiency found in this paper of Mg and Fe extraction could be due to the mineralogy of serpentinite rock (S-GO), meaning that the 20% of brucite contained in the serpentinite rock favored the extraction of Mg and Fe.

However, statistical analyses have showed that it could be attached higher Mg and Fe extraction on the following conditions: particle size (256 mm), acid concentration (4M), temperature (70 °C). Actually, these Conditions are being evaluated.

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7. RESPONSIBILITY NOTICE

The following text, properly adapted to the number of authors, must be included in the last section of the paper:
The author(s) is (are) the only responsible for the printed material included in this paper.

NOMENCLATURE

R_{CO_2} = Carbonation potential

$\Sigma(Ca, Mg, Fe)$ = sum of the molar fractions of these elements

MM_{CO_2} = molecular mass of CO₂

$\left[\frac{t_{MP}}{t_{CO_2}} \right]$ = units of R_{CO_2} (tons of the material used for each CO₂ tons sequestered)

S_{BET} = Surface area

$A = \Phi$ = Particle size (μm)

$B = C_{HCl}$ = Concentration of acid aqueous solution (M),

$C = T$ = Temperature of the process (°C)

1 = Low Level

2 = Medium level

3 = High level

(S/N) = Noise Signal

y = Response variable or dependent variables.

i = factors, such as: A, B, C.

j = Levels, such as: 1, 2, 3.

$N = n$ = Times the levels (j) of each factor (i) are repeated into matrix L_9 .

m_{ij} = average of the response variables (y) for each factor (i) in its level (j)

$(\overline{S/N})_{i,j}$ = average of the (S/N) values for each factor (i) in its level (j)

V_{FS} = Volume of the final solution.