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ARTICLE

Understanding Contributions of Divalent Cations in Mineral Carbonation Using Artificial Neural Network

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ABSTRACT

The roles played by divalent cations (calcium, magnesium and iron) of rock minerals in the efficiency of mineral carbonation have been investigated. Statistical modeling with Artificial Neural Network (ANN) having configuration ANN[17-4-1] shows that carbonation efficiency largely increases as the quantity of calcium content increases. Averagely, there is approximately 5% rise in the original efficiency for 10% increase in the quantity of calcium. This changes to 3.4% and 1.6% increases in efficiency, relative to the original efficiency for 20% and 30% increases in calcium content, respectively. Iron content of minerals offers clear positive correlation to the carbonation efficiency. From the global average, there is approximately 17% rise in the original efficiency for 10% increase in the quantity of iron. This increases to 29% and 41% over the original efficiency for 20% and 30% increases in iron content, respectively.. The influence of magnesium was found to be mainly negatively correlated to carbonation efficiency, after exceeding an unknown threshold. The global average of the efficiency changes with magnesium content results in original efficiency rising by 2% at 10% quantity increase and then reduces by 3% and 9% for 20% and 30% increase in magnesium quantity, respectively, relative to the original efficiency. Thus, iron compounds are found to be most potent of the divalent cations in carbonation reaction while calcium and magnesium content should maintain a threshold ratio with silica content for improved efficiency.

1. Introduction

The task of stemming the global warming effects on the planet requires the deployment of very effective and capable solutions. The ultimate solutions should be cheap, abundant and simple to apply, considering the scale of greenhouse gases in the atmosphere and the diversity of emission sources, globally. One such solution process is mineral carbonation, which offers the advantage of permanent storage to CO_2, in addition to cheapness and

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ease of application. The world is blessed with plethora of abundant mineral resources to perform the task of mineral carbonation [1]. The abundance of basalt is one strong factor that will positively upgrade the performance of mineral carbonation in the fight against climate change [2]. Mineral carbonation offers the benefits of permanent and safe storage [2] with opportunities for making use of ubiquitous earthly minerals [3], either as whole materials or as mine wastes [4].

To understand and improve the carbonation potentials of mineral, the chemical contents of the minerals must be analyzed to know the roles each component play in the reaction. According to Huijgen and Sanna et al. [5,6], carbonation rate is controlled by the diffusion of Ca^{2+} through the solid Si-rich layers in silicate minerals. Thus, the presence of calcium is significant to determine the performance of mineral carbonation. Mineralogical and chemical composition were of great importance for the mineral carbonation process [7]. Ramli et al. [7] indicates that divalent cations, pH and particle size are important parameters to consider in the carbonation yield. This points to the importance of calcium (Ca^{2+}), magnesium (Mg^{2+}) and others present in the rock minerals. The same concept can also be extended to iron (Fe^{2+}), making the group of common divalent cations. Thus, the optimum conditions for the occurrence of mineral carbonation are greatly dependent on the experimental conditions and material properties [8].

From the above, it is clear that the chemical constituents of the rock minerals are major determinants of the carbonation efficiency. While many factors can be considered, this work focuses on the roles played by divalent cations (calcium, magnesium, iron) in the success of mineral carbonation process with the aid of computational tool- Artificial Neural Network (ANN).

This work aims to demonstrate the feasibility of utilizing ANN to understand the patterns and the conditions of contribution of divalent cations (calcium, magnesium and iron) of rock minerals in the efficiency of mineral carbonation. The study will be among the first set of publications, in the open literature, to utilize the concept of machine learning to predict and forecast the carbonation efficiency of rock materials based on mineral contents.

2. Materials and Methods

The methods of this investigation involved sourcing of reliable data from literature with which different configurations of ANN were trained. The performances of the trained networks (ANNs) were then assessed to arrive at the best-performing ANN configuration. This best-performing configuration was then used to predict and forecast carbonation efficiency based on the influences of mineralogical and chemical constituents of rock materials as well as the experimental conditions.

2.1 ANN Configuration

In this work, different ANN configurations were trained and evaluated to arrive at the best network for predicting mineral carbonation from rock characteristics and process conditions. The networks were configured with feedforward structure while back-propagation algorithm was employed for the purpose of training. The ANN structure was in the form ANN[X-Y-Z] where 'X' is the number of input variables; 'Y' is the number of neurons in the hidden layer and 'Z' is the number of variables in the output layer. Different ANN configurations were tested using the approaches followed by Hanspal and Abidoye et al. [9,10]. Networks were created using single layer alone but with different number of neurons in each of the layers. The number of neurons was progressively increased for different configuration. The simulation platform was MATLAB (MathWorks, 2016). To implement the simulation procedure in MATLAB, program files were prepared with lines of code to create, train, validate and test the network as well as to generate the goodness of fit parameters of the data points using correlation coefficients (R^2) and mean square error (MSE). In the script, the 'While loop' procedure was used. The criteria of > '0.99' coefficient (R^2) was set for the loop with twenty rounds of training for each configuration. The network with each configuration was subjected to rounds of training until satisfactory performance was obtained. So, if after twenty rounds of training, the criteria were not satisfied, the training was stopped and the training of the next configuration began. The program divides the dataset randomly into 60%, 20% and 20% corresponding to the data for training, validation and testing, respectively. In the training process, epochs of 200 was used as the stopping criterion. Epoch is the maximum number of times all of the training sets are presented to the network. Thus, the training stops if the maximum number of epochs is attained. The network trainings thus stopped when the number of iteration exceeded the stated number of epoch or other criteria is satisfied. At the end of the training, network object is generated with indication of the best validation performance. The result from the training giving the best performance was then selected for each configuration, for comparisons and selection.

2.2 Data Sources and Processing

The data for this work were obtained from the carbonation data contained in the works [1,7]. The work is an experimental investigation of fundamental factors influencing mineral carbonation, using detailed kinetics of the process [1]. In the work, overall, 17 variables were extracted with a total of 4428 data points, but the focus of this study is to understand the roles of divalent cations (calcium, magnesium and iron). The variables and the summary of their statistical variation are shown in Table 1.

2.3 ANN Model Performance Criteria

The criteria used to evaluate the performance of different ANN model configurations listed and explained below. Equations (1) and (2) are mathematical representations of detailed statistical analyses used to evaluate the performances of various ANN configurations that were trained in this work.

Mean squared errors (MSE)

MSE computes the average of the squares of the errors between the observed value (S_{obs}) and the estimated value (S_{cal}).

$$\mathrm{MSE} = \frac{1}{N}\sum_{i=1}^{N}(S_{obs} - S_{cal})^2 \tag{1}$$

where, N = total number of data points predicted, S_{obs} = observed value of relative permittivity, and S_{cal} = calculated value of relative permittivity, .

2.4 Coefficient of Correlation (R2)

The mathematical representation of coefficient of correlation is expressed in Equation (2).

$$R^2 = 1 - \frac{\sum\left(Y_{measured} - Y_{pred}\right)^2}{\sum\left(Y_{measured} - \frac{\sum Y_{measured}}{N}\right)^2} \tag{2}$$

where, *Ypred* is the network prediction value, *Ymeasured* is the experimental response value and *N* is the total number of reading in the data points.

Table 1. Descriptive statistics of the input and output variables used in the machine learning

Variables	Stirrer speed (RPM)	P_{CO2} (bar)	Solid (%)	Time, t (hr)	T (°C)	NaCl (%)	$NaHCO_3$ (%)	Particle size (μm)	Mg (%)	Si (%)	Fe (%)	Al (%)	Cr (%)	Ni (%)	Mn (%)	Ca (%)	pH	Carbonation Yield (%)
Minimum	300	1	0.1	0.50	80	0	0	12.5	1.32	14.05	5.41	0.09	0.67	0.27	0.08	0.11	7	0.58
Maximum	1500	38.6	0.3	27.01	200	2	2	75	27.44	34.97	62.95	2	0.8	0.5	1	15.24	12	79.38
Average	247	27.29	0.12	3.38	159.80	0.89	0.84	28.05	22.53	20.88	15.82	0.48	0.7	0.32	0.27	2.27	7.37	23.45

3. Results

The results of the above investigations are presented and discussed in the following subsections.

ANN Models

As stated earlier, different configurations of the ANN models were tested to effectively and efficiently predict the carbonation efficiency of mineral rocks based on the mineralogical and chemical compositions of the rock materials. This testing of different configurations is necessary to ensure that the most reliable ANN structure is applied to learn the trends and relationships in the range of data used. The well-trained ANN model, having the best performance criteria, can then be used to predict the carbonation efficiency values applicable to the cases and conditions of interest. Therefore, this subsection presents the results of the training, validation and testing, as well as the performances of the different ANN model configurations. Out of all the configurations tested in this study, the comparisons of the coefficient of correlation (R^2) and Mean Squared Error (MAE) show that the ANN model with ANN[17-4-1] has the best performance. This model has only 4 neurons in the single hidden layer. The procedure followed in these comparisons was similar to the one described in the work [11].

In this work, discussion is limited to the best-performing configuration (ANN-[17-4-1]) to save space and time of the readers. The performances in training, validation and testing as well as the post-training regression for the best-performing ANN configuration is shown in Figures 1 and 2. As shown in figure 1, the performance during the training shows gradual learning of the trend in the data and the effective prediction of the output, resulting in gradual reduction in error (MSE) as the epoch number increases. The validation process shows that the network has grabbed the relationship among the data and the output without tending to overfit. This culminated in the optimal error value of 8.73×10^{-3} at the 23rd epoch.

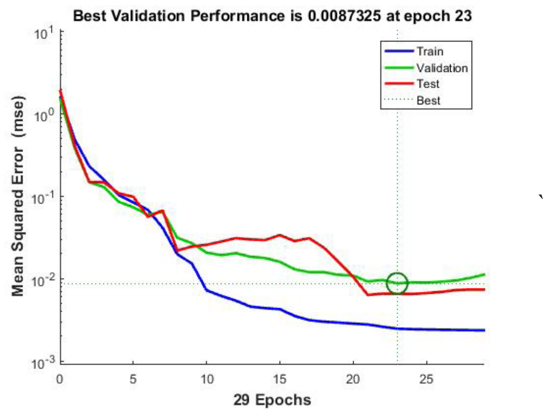

Figure 1. Performance of the ANN[17-4-1] model during Training, Testing and Validation processes.

The effect of good training and learning is further reflected in the regression output of the network. This is shown Figure 2, where the correlation coefficient is greater than 99%. This points to the fact the network really learns and adapts the data well. Thus, the ANN[17-4-1] is suitable for predicting and forecasting the carbonation efficiency of mineral rocks based on the mineralogical and chemical compositions of the rock materials as well as the other experimental parameters. Other configurations perform less in terms of the criteria of evaluation (MSE and R^2). Therefore, this well-trained network was used to predict the influences of different parameters on the carbonation efficiency.

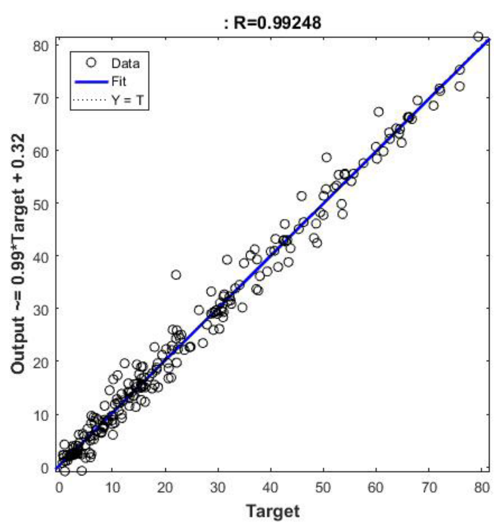

Figure 2. Regression analysis of the predicted and targeted outputs

Carbonation Efficiency

Figure 3 shows the prediction of the experimental outputs by the best-performing ANN configuration (ANN[17-4-1]). The figure shows good matches of the original experimental outputs (carbonation efficiencies) at most of the points. In fact, in most of the points, the predicted outputs wholly overlayed the original experimental outputs. In the remaining points, it is visually evident that the ANN predictions provide over 99% coverage of the original experimental output values. This corresponds to the correlation coefficient of above 0.99, shown in Figure 2. Thus, the network model used in this work is a highly efficient one to predict the output of carbonation process. As stated earlier, this study will be among the first set of publications, in the open literature, to utilize the concept of machine learning to predict and forecast the carbonation efficiency of rock materials based on the material contents and experimental conditions.

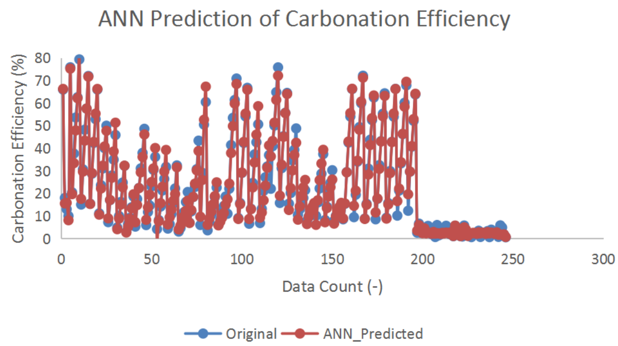

Figure 3. Prediction of the experimental efficiency data by the ANN

Effects of Calcium

Figure 4 shows the pattern of carbonation efficiency with changing percentages of calcium in the mineral. Largely, the percent of calcium correlates positively with the carbonation efficiency. Maximum effect of calcium was found to be 344% rise in carbonation efficiency at 10% increase in the percentage of calcium. This occurs at data count of 222 in Figure 4. Further increase in the amount of calcium to 20% and 30% reduces the effect on carbonation to 318% and 286%, respectively. Similar behaviour was noticed at the data count of 237, where the maximum effect of calcium on efficiency was found to be 248% at 10% increase in calcium amount. This reduces to 246% and 243% upon further increase in calcium amount.

The above goes to show that there is optimum level of increase in the calcium amount to yield increased carbonation efficiency. Though, further increase in the calcium also yielded higher efficiency, with reference to the original level of the calcium in the mineral, but the marginal increase continues to decrease with higher % of calcium.

Li et al. [12] emphasized that the release of calcium or magnesium from the silicate minerals will serve a great effect on carbonation yield. This idea emanated from the natural carbonation process which involves the weathering of magnesium, calcium and iron oxide-based silicate minerals, which then transforms atmospheric CO_2 into carbonate minerals [13]. The abundance of Mg/Ca-silicates on Earth offers enormous capacity for sequestering CO_2 [14]. Thus, calcium component of the minerals has great and positive influence on the efficiency of the carbonation.

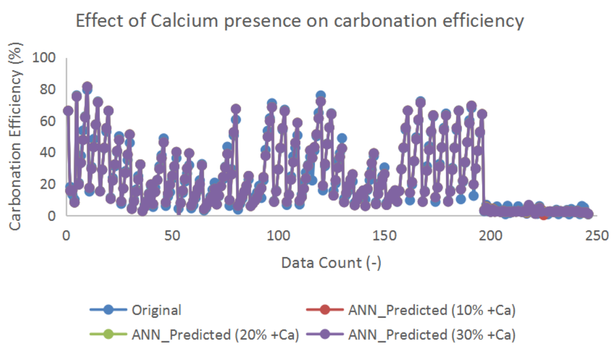

Figure 4. Carbonation efficiency with change in % of calcium.

Effect of iron

The presence of iron bears strong positive correlation with carbonation efficiency in most of the cases considered in this study. This is shown in Figure 5. Also, there is consistent increase in the efficiency as the iron content of the mineral increases. The highest predicted performance was recorded as 1541% at 30% increase in iron content. This has exceeded the performance of all other mineral contents. At 10% and 20% rises in calcium content, 526% and 1039% changes in efficiency were recorded, respectively. This occurred at data count 217 in Figure 5. What can be inferred from the results are the complex interplay of fractional composition of the mineral contents. For example at data count 221 in Figure 5, where conditions are similar to that at the count 217, the highest performance recorded was 674% at 30% rise in iron content. Upon close inspection, the discrepancy was attributed to the relatively higher level of magnesium in the former (data count 217). This seems to reveal the complex interrelationship between the fractional composition of mineral content and carbonation efficiency. Ramli et al. [7] found that iron mining waste was influential in determining carbonation efficiency.

Effect of magnesium

Figure 6 shows the effects of quantitative changes in magnesium on the carbonation efficiency. It can be observed that the efficiency drops slightly as the magnesium content increases. The reason for this is not obvious. This is unlike the cases with iron and calcium, where the efficiency rises with an increase in their quantities. For example, at data count 15, the carbonation efficiency decreases from the original efficiency of 72% to 65%, 55% and 51% for 10%, 20% and 30% rises in magnesium quantity, respectively. Similarly, it was observed at data count 115, original efficiency at 36.9% falls to 35%, 29% and 22% for 10%, 20% and 30% rises in magnesium quantity, respectively. Also, at data count 200, original efficiency at 4% falls to 2.34%, 2.32% and 2.31% for 10%, 20% and 30% rises in magnesium quantity, respectively. This consistent behaviour shows that there is a maximum amount expected of magnesium in carbonation minerals, unlike the iron and calcium.

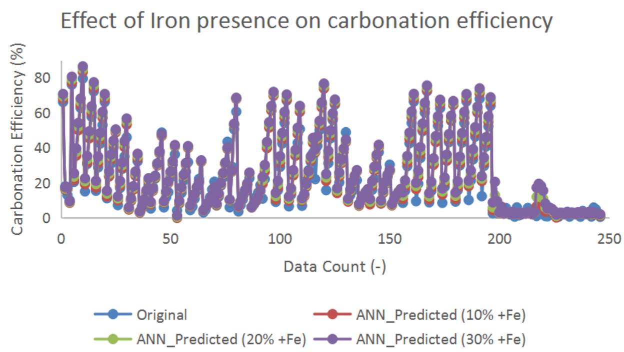

Figure 5. Carbonation efficiency with change in % of iron

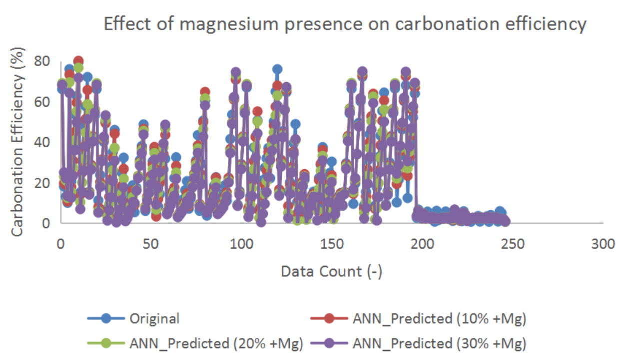

Figure 6. Carbonation efficiency with change in % of magnesium

4. Conclusions

This work has successfully demonstrated the clarity of influences played by various divalent cations in the efficiency of mineral carbonation. By training different configurations of Artificial Neural Network (ANN) to understand the complex roles of mineral constituents in the carbonation process, statistical appraisal shows that ANN[17-4-1] possesses the best performance criteria. This best-performing network was now employed to study the roles of divalent cations (calcium, magnesium and iron) of rock minerals in the efficiency of mineral carbonation. It was found that carbonation efficiency largely increases as the quantity of calcium content increases. Averagely, there is approximately 5% rise in the original efficiency for 10% increase in the quantity of calcium. This reduces to 3.4% and 1.6% rises over the original efficiency for 20 and 30% increases in calcium content, respectively. Iron content of minerals offers clear positive correlation to the carbonation efficiency. From the global average, there is approximately 17% rise in the original efficiency for 10% increase in the quantity of iron. This changes to 29% and 41% rises over the original efficiency for 20% and 30% increases in iron content, respectively. This goes to show iron as a great contributor to the mineral carbonation among other divalent cations. The influence of magnesium was found to be mainly negatively correlated to carbonation, after exceeding an unknown threshold. The global average of the efficiency changes with magnesium content results in original efficiency rising by 2% at 10% quantity increase and then falls by 3% and 9% for 20% and 30% increase in magnesium quantity, respectively. The study shows that, unlike iron, there is optimum level of increase in the calcium and magnesium contents to yield increased carbonation efficiency, following which the marginal increase in % of the chemicals continue to result in decrease of efficiency.

Conflict of Interest

There is no conflict of interest.

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