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Influx potential and sequestration of CO₂ in Cement based material towards establishing a proactive measure for combatting structural deformations

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Abstract. The effect of carbon dioxide sequestration by Sandcrete structures was investigated as a means of reducing CO₂ emissions. Experiments were conducted to determine the concentration of trioxocarbonate IV (CO₃²⁻) in sandcrete samples as well as its effect on soil pH. The results showed that the presence of CO₃²⁻ in sandcrete samples evidenced the process of carbonation in sandcrete structures and other cement-based material which provides an alternative means of CO₂ Sequestration. Also, the concentration of CO₃²⁻ varied along heights and horizontal distance within sections of the sandcrete structure. The acidic nature of the soil close to some sections of the sandcrete structure experienced varying pHs of 6.14 - 6.35 which gave room for enhanced leaching which seriously undermines the strength of the sandcrete structure. The percentage concentration differential in the horizontal and vertical directions, were found to vary widely from 5-46%. This constitutes a potential danger i.e. cracking and possible collapse of the structure. The results of calculated diffusivity values and concentration gradients of the carbonate ion conformed with results obtained using a predictive model which helped in monitoring the migration patterns of CO₃²⁻ in the sandcrete structure.

Keywords: Carbon sequestration, sandcrete structures, influx potential, structural deformation, CO₃²⁻-migration pattern

1. Introduction

In developing nations such as Nigeria, ninety percent of buildings including industrial structures comprise of sandcrete blocks [1]. Currently, efforts to tackle climate change are on the increase globally [2]. Anthropogenic operations (gas flaring, bush burning and agricultural activities) within such proximities release CO₂ into the atmosphere which affect these structures as well as cause global warming [3-5]. Cement-based structures consists of materials such as sand, stones and gravels which use cement as a binder to create a structure suitable for application in construction works. However, it has been proven that cement-based structures have the potential for atmospheric carbon dioxide sequestration [6]. Upon hydration, cement produces hydroxides that serve as binding agents which help to maintain the strengths of cement based structures [7]. An increase in atmospheric CO₂ leads to



ocean acidification, which may impose drastic consequences in marine ecosystems and according to literature, carbon mitigation is one of the newest strategies of the world's energy transition schemes [8].

Carbon dioxide sequestration is a geo-engineering technique for long term storage of CO₂ and other forms of carbon [6]. CO₂ sequestration includes the storage and capture of carbon/its compounds. This refers to large-scale, permanent artificial capture and sequestration of industrially produced CO₂ using subsurface saline aquifers, reservoirs, ocean water and aging oil fields [9]. Pre- and post-combustion systems for capturing CO₂ were previously discussed [6,10]. Carbonation reactions are exothermic reactions involving simple reactions between binary oxides such as MgO and CaO [11]; carbonates have a lower energy state than CO₂. According to Huntzinger et al. [12], mineral carbonization is one of the only forms of carbon sequestration that is permanent/stable. The disadvantage of other forms such as ocean, geologic and terrestrial sequestration, is largely due to the potential for leakage over a long period of time. This makes the sequestration process ineffective if the trapped CO₂ rapidly escapes back into the atmosphere.

Several models were discussed by Ramezaniapour et al. [13] and Odigure [7] on carbonation of cement-based structures. However, most of these works focused on determining the depth of carbonation, physical as well as chemical changes of the cement-based structures. In an earlier investigation, Odigure [7] presented a diffusion model for determining the diffusion of gaseous pollutants in sandcrete blocks. The shortcoming of the model is that other reactions were not taken into consideration (i.e. the formation of calcium bicarbonate from CaCO₃ and its subsequent leaching from the wall to the ground.

2. Theory

Carbonation is a chemical reaction in which solid products of cement hydrates, primarily calcium hydroxide or calcium silicate hydrates (CSH), calcium aluminate hydrates and calcium sulfoaluminate hydrates (mainly ettringite) in cement-based materials, react with carbonic acid to form carbonates [6]. A technique on how to sequester carbon dioxide was developed by Klaus Lackner [14]. This involves the combination of Carbon dioxide with Fe and Mg-rich serpentine rocks to produce CO₃²⁻ by the reaction in (1). The chemical reactions involved in this process are as given in (2) and (3):



The formed cement hydrates are brought about by hydration in sandcrete structures when they come in contact with acidic gases such as CO₂, SO₃ in the atmosphere. Hydrated cement contains calcium hydroxide, calcium silicate hydrate, calcium aluminate hydrate, hydrated ferrites and calcium monosulphoaluminate. The rate is dependent on the concentration of these gases in the atmosphere and their relative atmospheric humidity. Long exposure of sandcrete structures to aggressive acids, salts and alkalis, enhance their physicochemical as well as their mechanical deterioration. Therefore, this research investigates the possibility of carbon dioxide sequestration in sandcrete structures as well as the effects of carbonation on sandcrete structures. Products arising from carbonation reactions induce important physicochemical changes on these sandcrete structures [9]. One major effect of carbonization in sandcrete structures, is the loss of aesthetic value. Carbonized spots are not localized but spread across the structures of interest. An increase in atmospheric CO₂ contributes significantly to climate changes and global warming as well as the formation of acid rain which enhances corrosion of reinforced steel in concretes and roofing sheets [15,16]. The potential of this process is yet to be exploited, perhaps due to lack of understanding of the nature and possibility of CO₂ sequestration in cement-based materials [7], hence, the motivation for this research which entails the investigation of the mechanism of carbonation of sandcrete structures in relation to the deterioration of sandcrete structures.

Cement hydrates formed as products of hydration in sandcrete structures can react with acidic gases such as CO₂, SO₃ present in the atmosphere. This reaction occurs internally and may have significant adverse effects. The rate of reaction is dependent on the concentration of these atmospheric gases and their relative humidity. Long exposure of sandcrete structures to aggressive media such as acids, salts and alkalis enhance physicochemical and mechanical deterioration of the structures [17,18]. Consequently, this research investigates carbon dioxide sequestration potential and its effects on the chemical and mechanical properties of sandcrete structures at Covenant University by using a model approach. Also, an estimation of the percentage concentration of CO₃²⁻ in sandcrete samples of cracked walls of equal height and depth at a mapped location (Covenant University fence) was carried in order to predict the levels of structural deterioration at such sites. The concentrations of CO₃²⁻ and calcite present in each of the samples as well as the pH of the soil at different sections of the sandcrete structure were also determined.



Acid concentration in pulverized sandcrete was calculated using:

$$C_A V_A / C_B V_B = n_a / n_b \quad (7)$$

$$\text{Concentration of HCl (g/l)} = \text{Molar mass of HCl} * \text{molar concentration of HCl} \quad (8)$$

$$\text{Mass of HCl that reacted with pulverized sandcrete is: volume HCl (l)} 3.65\text{g/l} * \text{concentration} \quad (9)$$

$$\text{Mass of calcite} = \text{mass of HCl used} * \text{molar mass of CaCO}_3 \text{ in g/mol} / (\text{molar mass of HCl}) \quad (10)$$

The mass of CO₂ adsorbed (g/l) calculated mathematically as:

$$m_{\text{CO}_2} = m_{\text{HCl}} * m_{\text{CO}_2} / \text{molar mass of HCl}$$

Where: m_{CO₂} = molar mass of CO₂ (g/mol), molar mass of HCl = 35.5

Equation 6 was used to determine the parameters for the carbonation process.

$$k^2 - Pk - q = 0 \quad (11)$$

where P (concentration gradient) = u / (De) L and u = z/t; z = 0.94 m t = time (h) and q = De / (De)L
ln r.

Solving equation (11) as a quadratic expression gives the possible values of k.

3. Materials and Methods

3.1. Raw Materials, Equipment and Reagents

Sandcrete samples at sections of a fence at Covenant University, 0.5 M NaOH, 0.1 M (HCl) (Sigma Aldrich), phenolphthalein indicator, distilled water, conical flask, burette, pipette, electronic weighing balance (Stewart systems), spatula, retort stand, 1000 and 50 ml measuring cylinders, laboratory test sieve (300 μm), 250 ml beakers, hammer, measuring tape, ceramic mortar and pestle and pH Meter (Hanna 211 meter).

3.2. Methods

Sandcrete samples were obtained from six sections of a fence at Covenant University, Ota, Ogun state, Nigeria. The samples were obtained from long-exposed sandcrete blocks of the fence by striking a hammer into the blocks that make up the fence at the top (2.82 m section), middle (1.88 m section) and ground level (0.94 m section); the particles falling off were collected in containers. These samples were from six sections of the sandcrete fence at equal height interval and depth (i.e. 20 mm). By means of a measuring tape, horizontal, middle and ground level points of each section of the fence

were measured; the sandcrete fence has aged 14 years. Control samples i.e. five months old samples were obtained from a construction site at Covenant University. Soil samples were also obtained at different wall sections of the sandcrete fence to determine the pH of the soil.

3.2.1. Sample preparation

Each of the sandcrete samples discussed in section 3.2 was ground to fines and sieved with a 300 μm mesh to obtain very fine particles. In order to avoid aeration or absorption of moisture, the samples obtained were stored in transparent airtight moisture-proof containers prior testing.

3.2.2. Estimation of calcite and CO_3^{2-} concentration by quantitative measurement

To estimate the concentration of carbon dioxide and calcite in the samples, back titration was employed. 0.1 M hydrochloric acid was prepared by dissolving 8.33 ml of concentrated HCl to 1 L of distilled water in an Amber bottle. 0.5 M NaOH was prepared by mixing 20 g of Na-pellets in 1 L of distilled water in a reagent bottle. 3 g of the pulverized carbonated sandcrete sample was taken and transferred into a conical flask. 20 mL of 0.1 M HCl was added. The mixture was shaken thoroughly and observed for CO_2 release. 3 drops of phenolphthalein indicator was then added. At the end of CO_2 release, 0.5 M NaOH was measured and titrated against a standard solution of carbonated sandcrete so as to neutralize the HCl therein. Titration of other samples was done thrice in order to abate inaccuracies. The appearance of a pale permanent pink color signified the end point and the titre values were recorded.

3.2.3. Soil-pH at the wall sections of Sandcrete fence

20 g of each soil sample was weighed and put in a beaker. 30 mL of distilled water was added and the mixture was stirred properly. The measuring probe of the pH meter which was connected to an electronic meter was then inserted in the mixture. Data were read off from the panel of the pH meter.

4. Results

At the 0 and 8.3 m points along the horizontal axis of the fence, the concentration of CO_3^{2-} is highest at the middle section of the fence with values being 1.38 and 1.38 g/L respectively (Figure 1). At the 3.8 m point along the horizontal, the highest CO_3^{2-} concentration (1.34 g/L) was observed at the top section of the fence while the ground section at the lowest concentrations of carbonates at the aforementioned sections with values being 1.03, 1.05 and 0.83 g/L at all the 0, 3.8 and 8.3 m sections respectively. At the 10.48, 14.25 and 17.21 m points along the axis of the fence, the CO_2 concentrations were highest at the ground with corresponding values of 0.63, 1.38 and 1.5 g/L respectively with the lowest carbonate concentrations at such points being 0.26 (ground level), 0.22 (at the top level) and 1.43 at the top section of the fence above its horizontal axis. The high, medium and low carbon contents recorded at several sections of the fence are as a result of low influx of and dissolution of carbon in precipitation or moisture in the atmosphere which may have been transported as acid water or trioxocarbonate IV acid via diffusion into the walls of the fence. Also, the ease with which carbonation occurred in the fence is a measure of the type of cement used in the construction of the fence. Also, the manufacturer may have produced blended Portland cement using carbonaceous materials like limestone as additive. This may also result from the difference in the rate of leaching which is connected to porosity of the blocks. Furthermore, the production of CaCO_3 by carbonation can result in the gradual clogging of the pores of the sandcrete [6].

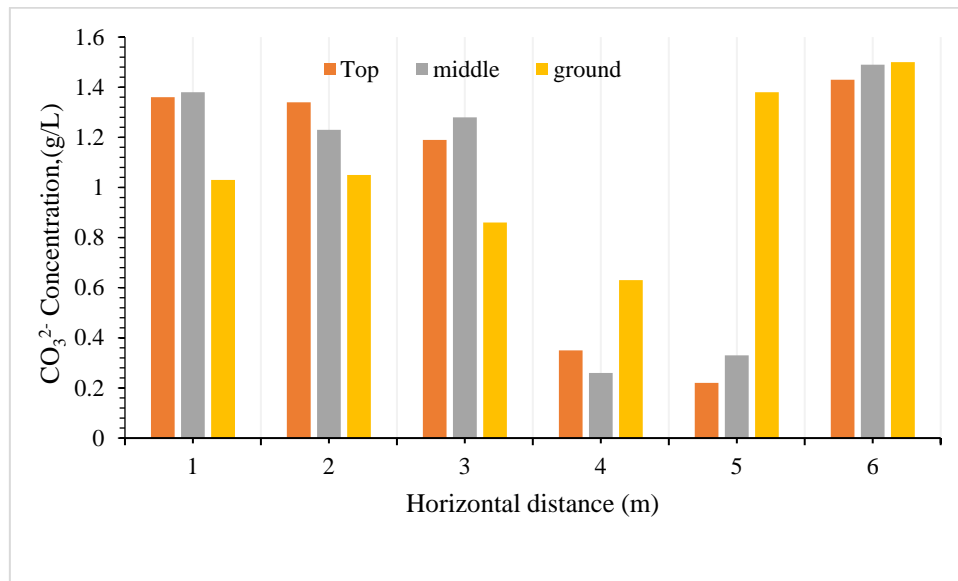


Figure 1. Concentration variation of carbonate at different sections of the fence.
Key codes: 1 = 0 m, 2 = 3.8 m, 3 = 8.3 m, 4 = 10.48 m, 5 = 14.25 m, 6 = 17.21 m along the horizontal axis of the fence

Leaching occurred faster at more porous sections than less porous sections of the fence, hence, the extent of block-porosity influenced the rate of carbonation. Hence, this informs that porosity is not the same at all points in the sandcrete wall.

4.1. Concentration and pH of the soil on which the fence rests

Figure 2 gives an illustration of the soil pH at different sections of the pipe. The highest pH (7.63) was recorded at the 3.8 m section of the soil holding the fence in place. The soil pH was found to be lowest i.e. 6.24 at the 0 and 8.3 m sections, 6.75 at the 17.21 m section, 6.35 at the 10.48 m section and 7.5 at the 14.24 m section. This is an indication that the soil pH was either weakly acidic or weakly alkaline. Since carbonic acid is a weak acid, it justifies the recorded pHs in the acidic range, however, owing to the presence of minerals such as K, Ca, Ph, etc, the salts of these elements may have resulted in the basic pHs recorded above the pH of 7 (neutrality).

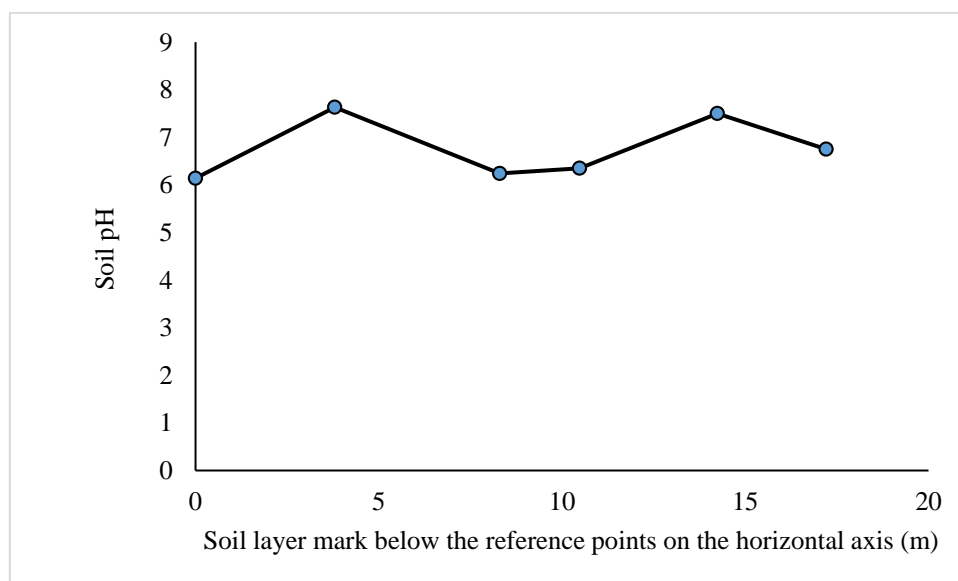


Figure 2. Variation of soil pH at different sections of the fence.

A close look at the pH trend observed at the 0, 7.61, 8.3 and the 10.48 m sections is a clear confirmation that the soil's acidity is a function of the carbonate concentration at those points of the soil holding the fence in place. Owing to the presence of minerals in the soil and conversion of the dissolved carbonate to salt, pH variation possibly set-in which gave rise to soil pH values > 7 . The migration pattern of the CO_3^{2-} horizontally showed that there are sections that tend to allow more transfer of the carbonate to the soil. This might be due to difference in the soil chemistry. If the pH of the surrounding soil is acidic, leaching is enhanced and if it is alkaline, rate of leaching is reduced thereby retaining more of the carbonates in the sandcrete wall.

4.2. Estimated CO_3^{2-} concentration gradient and diffusivity in the sandcrete wall CO_3^{2-} concentration gradient

The estimated carbonate concentration gradients at selected sections/heights above the ground level of the fence are shown in Table 1. The highest carbonate-concentration gradient was obtained at the lowest height above ground level. This shows that, the carbonate concentration gradient or driving force is highest towards the bottom of the fence giving a value of 1.5 g/L.m at the 1.88 m height above ground level/soil holding the fence in place. The lowest carbonate concentration was seen at the highest of all 3 heights considered at the different (6) horizontal sections considered hence, this justifies the fact that carbonate concentration gradient dropped at from the 0-14.24 m points whereas, beyond the 17.21 m point, the value increased significantly. A close look at the extreme values of table 1 shows that the terminals/extremes of the fence stored more carbonate than any other part of the fence because the values are higher at both ends (i.e. the 0 and 17.21 m points) than at any other point on the fence. This is due to those areas being closed ends where carbonate radical diffuses and thus gets stored because there are no extensions/points beyond those regions.

Based on Fick's law of diffusion, concentration gradient is inversely proportional to diffusivity hence, the extreme points of highest concentration gradients have the least diffusivities (Table 2). The extent of carbonation was investigated by calculating diffusivity (De) L values according to the mathematical model. (De)L represents the dispersion of the dissolved CaCO_3 as $\text{Ca}(\text{HCO}_3)_2$ in the block-pore volume. It also represents the effective diffusivity of CO_3^{2-} ions into the sandcrete fence. It was approximately determined from the concentration gradient "P". The estimated negative values are indicative of a decrease in concentration of carbonates in the reference sections of the sandcrete fence. The higher the diffusivity, the lesser will be the expected mechanical strength of the sandcrete fence [7].

Table 1. Concentrations of CO_3^{2-} at the various sections of the fence.

Vertical Height, m	Estimated concentration gradient g/L.m					
	0 m	3.8 m	8.3 m	10.48 m	14.24 m	17.21 m
2.82	1.36	1.34	1.19	0.35	0.22	1.43
1.88	1.38	1.23	1.28	0.26	0.33	1.49
0.94	1.03	1.05	0.86	0.63	1.38	1.5

Table 2. Estimated diffusivities at the different sections of the pipe.

Vertical Height, m	Estimated Diffusivities * $10^{-6} \text{ m}^2/\text{s}$					
	0 m	3.8 m	8.3 m	10.48 m	14.24 m	17.21m
1.88-2.88	0.36	5.46	-6.56	6.69	5.46	1.09
0.94-1.88	1.73	3.31	1.42	-1.63	-57.46	-0.33

A high rate of carbonate diffusivity is aided by the temperature of the environment, hence, an environment of high temperature increases the rate of diffusion. Also, Sections with high diffusivity gave lower CO_3^{2-} concentration gradients or lower diffusion rates of the carbonate ion. Furthermore, the diffusivity of CO_2 is 10,000 times smaller in water than in air. This means that diffusivity is faster in air than in water. Therefore the rate of carbonation occurs faster in air than water. According to Odigure [14], this parameter gives the extent of reaction between the cement mineral hydrates and CO_2 and their leaching from the structure. D represents the actual values of CO_2 molecules that diffused into and reacted with the cement minerals in the sandcrete matrix. The parameter r represents depth. This varies but for this research work, q was assumed to be zero since samples were collected at equal height and at constant depth of 20 mm.

The depletion of CO_3^{2-} within the sandcrete will lead to a fall in the binding properties of any formed $\text{Ca}(\text{OH})_2$ and may cause a defect or crack in the structure. The observed horizontal changes in carbonate concentration differs with sections of the fence which will exhibit a variation in pH and %composition of its carbon constituents. Furthermore, these changes can lead to internal stress within and along the fence wall which result in crack-formation in the fence structure. Based on the investigation carried out by Odigure [14], the results obtained in this study in terms of calculated diffusivities and concentration gradients of CO_3^{2-} are in close agreement.

Figure 3 is an illustration of the irregular/random variation of soil pH and heights across the fence. At the 1.88- 2.88 m height above the ground, within the soil pH of 6.24-7.5, the diffusivity of CO_3^{2-} is lowest with corresponding values in the range of -6.56-5.46 m^2/s and hence, based on Fick's second law of diffusion, the concentration of the carbonate is highest whereas, it is lowest at pHs of 6.35 and 7.63, the resultant diffusivities are 6.69 and 5.46 m^2/s . However, within 0.94-1.88 m height along the fence, the pHs are 6.35 and 7.5 with corresponding diffusivities of -1.65 and 57.46 m^2/s respectively, which are the least recorded values hence, the concentrations of CO_3^{2-} are expected to be highest here; also, within the measured pHs of 6.14 and 7.63, the estimated diffusivities are 1.73 and 3.31 respectively, which are expected to give the lowest CO_3^{2-} concentrations. Based on the results, the soil pH range is weakly acidic and alkaline with varying values which influence the estimated diffusivities.

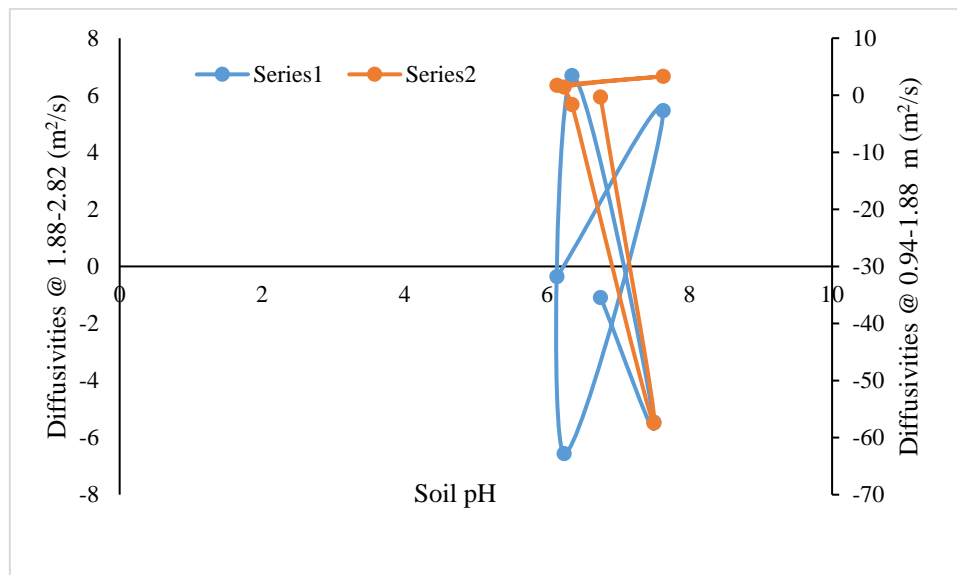


Figure 3. Soil pH and diffusivities at different heights.

5. Conclusion

The presence of CO_3^{2-} in sandcrete samples taken from the fence actually confirm the effect of carbonation in sandcrete structure which gives a clue to the reason for structural failures in fences

caused by carbon sequestration. CO_3^{2-} concentration varied with heights and horizontal distances within sections of the sandcrete fence. pH ranges were judged to be in the weakly acidic and weakly alkaline ranges thus encouraging leaching of the soil/weakening of the fence which resultantly undermines the strength of the fence. The estimated percent carbonate concentration differential in the horizontal and vertical directions were found to vary widely from 5 to 46% which constitute a potential danger to the fence in terms of being subsequently probable to cracking and collapse. The predictive model gave comparable diffusivity values with those obtained in literature. Furthermore, at the 1.88- 2.88 m height above the ground, the concentration of the carbonate was highest whereas, it was lowest at pHs of 6.35 and 7.63, with resultant diffusivities of 6.69 and 5.46 m^2/s respectively while at the 0.94-1.88 m height along the fence, the recorded pHs are 6.35 and 7.5 with corresponding diffusivities of -1.65 and 57.46 m^2/s respectively; these represent the least recorded diffusivities and are expected to be points having the highest concentrations of CO_3^{2-} .

Recommendations

Based on the results of this study, it is necessary to carry out routine CO_2 measurements around Covenant University fences in order to ascertain their deformation potentials; this is aimed at developing adequate carbon capture strategies (adsorption systems/ CO_2 -traps) for CO_2 mitigation. Also, in the year 2019, a collapse of one of the Covenant University fences occurred and this is a clear evidence that the fence had been weakened at some of its sections owing to the influx of acid rain and CO_2 . Furthermore, an investigation of the mechanism and kinetics of the carbonation process of cement needs be carried out in order to clearly understand the sequestration process of cement or sandcrete structures. Analysis involving the use of Scanning Electron Microscopy, Thermal Gravimetric Analysis and X-ray Diffraction can be carried out to investigate the carbonation profile, the mineralogy and morphology of the understudied sandcrete structure. The effect of temperature on cement-carbonation should be investigated. Estimation of Analysis on deposition and penetration rates of other gaseous pollutants such as SO_2 and NO_2 may be considered in order to ascertain their influence on the carbonation processes of sandcrete structures. The carbonation of other cement-based materials such as concrete can be investigated with the results compared with those of these study. The use of Atomic Absorption Spectrophotometer (AAS) is necessary so as to ascertain the types of metal ions especially Ca^{2+} ions in sandcrete samples as this helps to predict the extent of carbonation.

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