Bulletin of Environment, Pharmacology and Life Sciences Bull. Env. Pharmacol. Life Sci., Vol 8 [7] June 2019: 54-63 ©2019 Academy for Environment and Life Sciences, India Online ISSN 2277-1808 Journal's URL:http://www.bepls.com CODEN: BEPLAD Global Impact Factor 0.876 Universal Impact Factor 0.9804 NAAS Rating 4.95

**ORIGINAL ARTICLE** 



# Industrial Alkaline Residue for Carbon Capturing

C.G. Karishma<sup>1,\*</sup>,T.D. Rushendra Revathy<sup>2</sup> and K. Palanivelu<sup>2</sup>

<sup>1</sup>Department of Remote Sensing and GIS, Tamil Nadu Agricultural University, Coimbatore – 641003 <sup>2</sup>Centre for Climate Change and Adaptation Research, Anna University \*Corresponding author:karishmagovindarajan@gmail.com

#### ABSTRACT

Rapid increase of CO<sub>2</sub> concentration in the atmosphere from the pre-industrial values of 280 ppm to the current values of 402 ppm has forced the international community towards adopting a series of actions, e.g., the Kyoto protocol, aimed at reducing anthropogenic emissions of greenhouse gases. Furthermore, as fossil fuels are projected to be a dominant energy resource in the 21st century, technologies for sequestering emissions from fossil fuel combustion in a safe and definitive manner are being developed and implemented. Among the different options, mineral carbonation has been proposed as a possible way for  $CO_2$  sequestration, a technology that could possibly contribute to reduce carbon dioxide emissions either in-situ mineral sequestration (long term geological storage) or by the ex-situ mineral sequestration (controlled industrial reactors). Thus the present work focuses on the study of feasibility of mineral carbonation in the selected alkaline waste residue namely cement kiln dust (CKD). In order to confirm the potential of CKD, characterisation studies were carried out to determine the particle size, pH, EC, ANC, moisture content and loss on ianition. The experiments were carried out via ex-situ process (direct carbonation), by both gas-solid carbonation (dry capture) and aqueous carbonation (wet capture) which involves the reaction of  $CO_2$  with alkaline elements. The experiments were conducted in a stainless steel reactor at varying pressures (1, 2 and 5 bar) and reaction time (15 min to 3 h) at room temperature. The carbonated product obtained was analysed in TOC analyser in order to evaluate the net carbon increase. From the experiments and studies carried out it was inferred that net carbon increase obtained in wet capture was double the amount of carbonation in comparison with dry capture. Thus CKD readily sequesters CO<sub>2</sub> under optimized conditions. Maximum net carbon increase of 5.09 g of CO<sub>2</sub>/kg of CKD was obtained for CKD at 10 bar pressure, contact time of 30 min and S/W ratio of 1:10. It can be concluded that CKD can be effectively utilised for sequestration of carbon dioxide (CO<sub>2</sub>).

Keywords: CO2 sequestration, Cement Kiln Dust (CKD), gas-solid carbonation, aqueous carbonation, TOC analyser

Received 19.01.2019

#### Revised 30.01.2019

Accepted 19.02. 2019

### INTRODUCTION

Over the last century, global average temperature has increased by more than 0.7°C which is the most serious environmental hazard confronting the world. Significant evidence on currently on-going global warming and related climate modifications have been collected and reported by the scientific community in the last couple of decades, in particular by the U.N. International Panel on Climate Change. In order to avoid the potentially devastating consequences of global warming and climate change, the carbon dioxide "CO<sub>2</sub>" emissions caused due to anthropogenic activities must be reduced considerably. CO<sub>2</sub> is naturally present in the atmosphere as a part of the earth's carbon cycle (the natural circulation of carbon among the atmosphere, oceans, soil, plants and animals). Its emissions come from a variety of natural sources while human related emissions may be responsible for the increase that has occurred in the atmosphere since the industrial times, due to combustion of fossil fuels for energy and transportation, certain industrial process and land use changes. If no proactive mitigative actions are taken to limit the magnitude or rate of long-term climate change, the energy related CO<sub>2</sub> emissions are likely to be 40% higher in 2030 than they were in 2000. This can be achieved only through reductions in anthropogenic emissions of greenhouse gases by increasing the capacity of carbon sinks, that is through carbon capture and sequestration and switching to low-carbon energy sources. Mitigating climate change by capturing  $CO_2$  from large point sources such as power plants and industrial sources and subsequently storing it away safely instead of releasing into the atmosphere is an effective option of carbon capture and storage (CCS).

#### Carbon capture and sequestration technologies

 $CO_2$  can be captured using the technologies namely post combustion, pre-combustion, oxy-fuel combustion or from industrial process streams. Carbon sequestration can be accomplished through geological storage, ocean storage, terrestrial sequestration, soil sequestration, industrial use and mineral sequestration. Although several carbon sequestration technologies have emerged as potential means to mitigate rising concentrations of  $CO_2$  in the atmosphere, one of these options is mineral carbonation which is carried out in this study.

### Mineral carbon sequestration

Mineral carbon sequestration is a post-combustion technology which involves the process of capturing  $CO_2$  in a mineral form by its reaction with alkaline materials, composed mainly of calcium and magnesium rich oxides and silicates, leading to the formation of solid carbonate products. The main advantage being geochemical stability, reduced transportation cost and storage safety of mineral carbonates. Not only is mineral carbon sequestration a permanent method of  $CO_2$  disposal but the products are environmentally benign and could potentially be sold. Magnesium rich minerals as carbonation feedstock materials for  $CO_2$  sequestration. In which, physical and chemical activation of the mineral, a Cedar Hill serpentine was carried out. The carbonation reaction was conducted at ambient temperature, 20 °C and low pressure, 45 atm and estimated the carbonation efficiency to be at least 53 %.

#### Direct mineral carbonation

In general, ex situ mineral carbonation can be divided into two categories: direct carbonation and indirect carbonation. Direct mineral carbonation is accomplished through the reaction of solid alkaline mineral with  $CO_2$  either in gaseous or in aqueous phase. The reaction of gaseous  $CO_2$  with particulate metal oxides at suitable temperatures and pressures to form carbonates is the basic mechanism of gas-solid carbonation. In aqueous carbonation  $CO_2$  reacts with water to form bicarbonate and a proton, the proton liberates the metal ion from the mineral, and the metal ion reacts with bicarbonate to produce a carbonate precipitate under controlled temperature and pressure in a closed environment. The advantages of direct mineral carbonation include simplicity of the process and minimal use of chemical reagents.

#### Alkaline waste

Of late, considerable and successful efforts have been made to use solid alkaline waste materials as an alternative source of minerals rich in calcium and magnesium. Major advantages of this option in contrary to conventional mineral ores are that no need for extensive pre-processing, the process is cost effective and their widespread availability in industrial areas, i.e. near large point sources of  $CO_2$ . Some of the alkaline waste residues used for mineral carbonation are coal fly ash, municipal solid waste, incinerator ash, steel slag and cement kiln dust. It has been reported that the capacity to sequester  $CO_2$  from these alkaline residues depends directly on the proportion of binary oxides (CaO and MgO) and/or hydroxides (Ca(OH)<sub>2</sub> and Mg(OH)<sub>2</sub>) contained in the waste matrix. Anjali *et al.*, [1] focused on the feasibility of mineral carbonation of C& D waste products. They conducted a series of batch experiments in a self fabricated batch reactor. Sample was exposed to constant  $CO_2$  supply under standardized humidity, temperature conditions (40 °C) and varying w/s ratio. The degree of carbonation achieved in C&D waste, for 16 h at w/s ratio of 0.4 was 38.44 %. This confirms the possibility for utilization of C&D waste for future mitigation of CO<sub>2</sub>.

Yadav et al. [13] demonstrated the applicability of red mud for the sequestration of green house gases with reference to carbon dioxide. They separated the red mud sample into three different size fractions (RM1, RMII, and RMIII) of varying densities (1.5-2.2 g cm<sup>-3</sup>) and carbonation of each fraction of red mud was carried out separately at room temperature using a stainless steel reaction chamber at a fixed pressure of 3.5 bar, time (3.5 h) and liquid to solid ratio (0.5, 0.35 and 0.3). They evaluated the carbonation capacity of RM II to be 5.3 g of  $CO_2/100$  g which had greater sequestration capacity among all the size fractions. Anjali *et al.* [2] studied on exploring an array of alkaline waste residues, procured from diverse sources as means for sequestering maximal CO2. The wastes included cement bag filter dust, finished cement sample, marble dust and baghouse dust. They conducted experiments in a self fabricated carbonation reactor under 80% CO<sub>2</sub> concentration and at 40°C, water: solid (w/s) ratio of 0.2 to 1.2 with relative humidity in the reactor was maintained at 100%. Their experimental investigation demonstrated that 1 ton of CBF, an industrial waste that contains about 51.1 wt. % of lime (CaO), could sequester up to 0.302 kg of CO<sub>2</sub> per kg of CBF with maximum degree of carbonation of 25.84% at the end of 16 h at w/s ratio of 0.4 which had the highest sequestration capacity in comparison with the above mentioned residues. Direct gas-solid carbonation of alkaline residues from air pollution control (APC). They reported that the carbonation of APC residues is a viable process for the storage of  $CO_2$ , yielding almost 60 % conversion of calcium available as calcium oxide or hydroxide and an uptake of around 120 g of CO<sub>2</sub>/kg of residue [3, 4].

Baciocchi *et al.* [4] analysed and compared the results of accelerated carbonation experiments for  $CO_2$  storage carried out on the air pollution control (APC) residues of a waste incineration plant, via both the dry and the wet route. They concluded that two routes achieved a similar maximum calcium conversion to carbonates (around 65 %) corresponding to a potential  $CO_2$  storage capacity of 250 g/kg residues. For the dry route, maximum conversion was achieved in a few minutes at 400 °C under a 10 %  $CO_2$  atmosphere, whereas for the wet route it was obtained in about 10 minutes under a 100 %  $CO_2$  atmosphere, with a liquid to solid ratio of 0.2, at 30 °C and 3 bar, or without water addition at 50 °C. Sahu *et al.* [10] conducted a laboratory study to investigate the ability of neutralization of red mud (RM) using carbon dioxide gas sequestration cycle at ambient conditions. They treated the red mud with  $CO_2$  at 5 mL/min for 5 h in each of the cyclic process and reported that pH and alkalinity of red mud was decreased from ~11.8 to ~8.45 and ~10,789 to ~178 mg/L at the end of the cycle 3, respectively. The permanently captured  $CO_2$  %( w/w) per 10 g of red mud were ~26.33, ~58.01, ~55.37, and ~54.42 in NRM and first, second, third cycles of carbonated filtrate, respectively. They concluded that cyclic carbonation captured more amount of carbon as compared to without cycle. This indicated that caustic filtrate of red mud was more efficient for  $CO_2$  sequestration.

Rendek *et al.* [9] aimed to quantify the volume of  $CO_2$  that could be sequestrated in municipal solid waste incinerator (MSWI) bottom ash. Carbonation was performed with 4 mm-sieved samples in a  $CO_2$  chamber, the  $CO_2$  pressure and humidity of the samples were varied to optimize the reaction parameters and proposed that the volume of sequestrated  $CO_2$  was on average 12.5 L/kg dry matter (DM) for unsieved material and 24 L/kg DM for 4 mm-sieved samples. Their process could reduce incinerator  $CO_2$  emissions by 0.5–1%.

Chang *et al.* [5] considered UF slag, FA slag, and BHC as feedstocks for aqueous carbonation process .The selected feedstocks were alkaline, calcium-rich particles that reacted with  $CO_2$  dissolved in the aqueous slurry to form  $CaCO_3$  coating on the surface of the slag .In their study, the highest conversion of 68.3 %, was achieved by BHC when the aqueous carbonation was conducted at an L/S of 10 mL g<sup>-1</sup>, a  $CO_2$  partial pressure of 700 psig, and a temperature of 160  $^{\circ}$ C, with a reaction time of 12 h in an autoclave reactor. Direct gas–solid carbonation reactions of residues from an air pollution control system (APCr) using different combinations of simulated flue gas to study the impact on  $CO_2$  sequestration. The optimal  $CO_2$  sequestrating temperature of 395  $^{\circ}$ C was easily obtained for APCr using a continuous heating experiment and they achieved a maximum carbonation conversion of between 68.6 % and 77.1 % in a typical flue gas [11].

Ramme *et al.* [8] investigated the sequestration of  $CO_2$  through mineralization into carbonate compounds. High-surface area, calcium and alkali rich by-product materials which included sources of cement-kiln dust, blast furnace slag, fly ash, lime-kiln dust, recycled concrete fines were used to sequester  $CO_2$  from ambient atmospheric gas,  $CO_2$  gas, and coal-combustion flue gas. They utilized a foam generator with a foaming agent and various  $CO_2$  containing gases to produce a lightweight adequate strength, material with readily available increased surface area from by-product materials, and the available  $CO_2$  for rapid carbonation. Then the carbonated product was crushed into a carbonate-rich aggregate suitable for a variety of construction uses.

Anjali et al. [1] examined the effect of temperature and varying proportions (0 %, 25 %, 50 %, and 75 % soil content) of soil-waste mixture on the potential of CO<sub>2</sub> mineral sequestration process in limiting the overall CO<sub>2</sub> production in landfills. They focused on alkaline wastes like construction and demolition (C&D) waste and hospital incineration (HI) ash. They carried out Experiments in a self fabricated batch reactor at 0.4 water-to-solids (w/s) ratio, relative humidity of 60-80 %, and flow rate of CO<sub>2</sub> at 10 L/min for 2 h for temperature variation from 25 °C to 40 °C at intervals of 5 °C and found out the optimum temperature to be 30 °C. They obtained maximum CO<sub>2</sub> uptake of 10.56 % and 9.14 % for C&D waste and HI Ash respectively at 0 % soil content. They concluded that soil-waste mixture of 0 % and 25 % soil content was established to be best suited for CO<sub>2</sub> sequestration. Accelerate the carbonation of municipal solid waste incineration bottom ash for CO<sub>2</sub> sequestration. Their optimum condition for CO<sub>2</sub> sequestration was an L/S ratio of 0.2, a temperature of 20 °C and a CO<sub>2</sub> concentration of 20 %. They suggested that this can be utilized directly at the CO<sub>2</sub>-emitting incinerator, bottom ash generated by water-cooling accounts for 15–25 % of the water content, and CO<sub>2</sub> emission incinerators account for 5–20 % of CO<sub>2</sub> concentration [7].

#### Cement kiln dust

The cement manufacturing process produces millions of tons of CKD each year as a by-product. It is a fine-grained mixture of partially calcined and unreacted raw feedstock, which is removed from cement kiln exhaust gas by air pollution control devices like electrostatic precipitators, bag house etc. CKD has the potential to be used for mineral carbon sequestration because of its high mass fraction of CaO (20 to 70%), finely divided particle size and freely available in large quantities at the point source. Huntzinger *et* 

*al.* [6] presented the first study examining the feasibility of carbon sequestration in cement kiln dust (CKD), a by-product generated during the manufacturing of cement. They conducted a series of column experiments on segments of intact core taken from landfilled CKD and operated the columns(A,B,C,D) under varying conditions to determine the impact of humidity and  $CO_2$  concentration on the extent of carbonation or sequestration. The overall extent of carbonation/sequestration was greater in columns with lower water contents. They obtained a maximum carbon sequestration of 34.6 g of  $CO_2/100$  g of CKD.

#### MATERIALS AND METHODS

In general there are large quantities of industrial alkaline residues generated all over the country from various sources as listed in the table 1 below, from which CKD was selected as one of the potential residue for sequestration of  $CO_2$  among the identified wastes. The cement kiln dust used in the present study is a by-product generated from a cement industry (ACC cements) situated in Coimbatore, Tamilnadu. CKD is a dry fine-grained material that is captured in the air pollution control equipment (baghouse). The CKD sample was received dry and stored at room temperature in an air tight container. The  $CO_2$  gas used in the study was 99.9% purity analytical grade gas. All the chemicals used in the study were analytical grade reagents.

### Characterization

#### Particle size:

Particle size of the sample was determined by sieving method using IS standard sieves either horizontally or vertically. The sieving causes a relative movement between the particles and the sieve, depending on their size the individual particles either passes through the sieve mesh or is retained on the sieve surface.

# Moisture content:

Moisture content of CKD was measured by determining the weight loss of the sample due to heating. 1 g of sample was weighed and dried at 105 °C in the hot air oven for 2 h, after which the sample was placed in a desiccator and then weighed. Thus the moisture content of the sample was determined by measuring the differences in the initial and final weight.

# pH:

A pH meter provides a value as to how acidic or alkaline a liquid is. The basic principle of the pH meter is to measure the concentration of hydrogen ions. Acids dissolve in water forming positively charged hydrogen ions (H<sup>+</sup>). The greater this concentration of hydrogen ions, the stronger the acid is. Similarly an alkali dissolves in water forming negatively charged hydroxyl ions (OH<sup>-</sup>). The stronger a base is the higher the concentration of negatively charged hydroxyl ions there are. The amount of these hydrogen ions present in solution is dissolved in some amount of water determines the pH.

To measure the pH of CKD, 20 g of sample was taken in a 50 ml beaker and 100 ml of reagent water was added to it. The beaker was covered and the suspension was continuously stirred for 5 min and placed aside for about 15 min to allow most of the suspended waste to settle out from the suspension, otherwise it could be filtered or centrifuged from the aqueous phase to measure the pH .The electrodes were washed thoroughly with distilled water and the system was allowed to stabilize before making the final reading.

#### Electrical Conductivity (EC):

Electrical conductivity measurement gives the total amount of soluble salts present in a solution and it is expressed as mS<sup>-1.</sup> An electrical conductivity meter (EC meter) measures the electrical conductivity in a solution it employs a potentiometric method and consists of four electrodes. It is used to monitor the amount of nutrients, salts or impurities in the water.

#### Acid Neutralising Capacity (ANC):

ANC applies to the acid neutralising capacity of solutes plus particulates in an unfiltered water sample reported in equivalents per liter (or milliequivalents or microequivalents). (The ANC value would be equivalent to alkalinity for samples without titratable particulate matter.) i.e. the equivalent sum of all bases or base producing materials in an aqueous system that can be titrated with a strong acid to an equivalence point.

A measure of 1:5 ratio of sample and distilled water was added respectively to prepare the water slurry. To measure the ANC capacity of CKD, approximate volume of sample (25 ml) was pipetted into the beaker and the beaker was placed on the stirrer. Then the electrodes and temperature sensors were inserted into it, away from bottom and sides and gently stirred. The concentration of HCl taken in the burette was 0.1 N. The initial pH and the burette reading was recorded. Then the titrant was added and stirred for 15-20 s, the pH was recorded. The same procedure was repeated until a pH of slurry solution drops to 4. The ANC value was computed with a pH equivalence of 8.3 for the residue. *Loss on ignition:* 

A known weight of solid ignited at 550  $^{\circ}$ C and the loss in weight due to destruction of organic matter, CaCO<sub>3</sub>, MgCO<sub>3</sub> and loss of moisture is reported as loss on ignition. To determine the loss on ignition of CKD, the cruicible was weighed then 0.5 to 5 g of dried sample was weighed to the nearest 1 mg into the cruicible and heated in the furnace at 550  $^{\circ}$ C ± 25  $^{\circ}$ C for at least 60 min. After the specified time the hot cruicible containing the residue on ignition was placed in the desiccator and allowed to cool. After cooling to ambient temperature, the cruicible was weighed to the nearest 1 mg. Loss on ignition of the solid sample was expressed in %.

#### Dry capture:

Dry capture studies were conducted in a stainless steel reactor vessel with pure  $CO_2$ .  $CO_2$  was purged inside the reactor containing a measured quantity of CKD (10 g) and under varying pressures (1, 5 and 10 bar) and reaction time (15 min to 3 h) was studied. The carbonated samples were analysed for net carbon increase using TOC analyser.

#### Wet capture:

Experiments were conducted in a stainless steel reactor vessel with pure  $CO_2$  and CKD water slurry (1:10 solid/liquid ratio). CKD water slurry was taken in the reactor and  $CO_2$  was purged into it. The reactor was closed and then placed on a magnetic stirrer in order to stir the slurry at 500 rpm for the given reaction time. The effect of initial  $CO_2$  partial pressure (1, 5 and 10 bar) and reaction time (15 min to 3 h) on carbonation was studied. On completion of the experiment, the CKD water slurry was filtered through whatman filter paper no.42 using vacuum pump. All the experiments were conducted in triplicates to ensure reproducibility. The solid obtained was analysed in a TOC analyser to test its net carbon increase. **TOC analyser:** 

TOC analyser used is analytic jena multi N/C 2100S which converts the carbon in the sample to carbon dioxide by catalytic combustion in a horizontal combustion chamber at a temperature range of 1150  $^{\circ}$ C in a oxygen-rich environment. The carbon dioxide generated by oxidation is detected using non-dispersive infrared (NDIR) detector. The amount of CO<sub>2</sub> in a sample is directly proportional to the concentration of carbonaceous material in the sample. The analyser was calibrated using calcium carbonate (120 g/kg of carbon content) by varying weight (50,100,150,200 and 250 mg) with the fixed concentration. The instrument had a reproducibility of 3-4%.

#### **RESULTS AND DISCUSSION**

#### Characterisation

The experiment carried out to determine the particle size of CKD revealed that the particle size was 500 to 63  $\mu$ m. Moisture content studies showed that there was no presence of moisture content in the sample. The pH was measured to be 10.5 which indicated that the residue was highly alkaline in nature and thus suitable for CO<sub>2</sub> sequestration. Electrical conductivity of CKD was found to be 1818  $\mu$ s/cm. loss on ignition of the residue was determined to be 10.5 %.



Figure 1. Cement klin dust (CKD)





Figure 3. Net carbon increase of CKD at varying CO<sub>2</sub> partial pressure (Reaction time -1 h, Room temperature)



Figure 4. Net carbon increase of CKD at varying reaction time (CO<sub>2</sub> partial pressure-10bar, Room temperature)







Figure 6. Net carbon increase of CKD at varying reaction time (Stirring speed-500rpm, Slurry ratio-1:10, Room temperature, Initial CO<sub>2</sub> partial pressure-10bar)





Alkaline residue	Source
Rice husk ash	Rice mill
Cement kiln dust(CKD)	Cement industry
Fly ash	Power plants
Red mud	Aluminium plants
Iron slag	Iron or steel plants
Lime sludge	Paper mills
Lime sludge	Sugar mills

# Table 1. Identified industrial alkaline residues

# Table 2. Observed values for ANC

S.No	Volume of HCl added (mL)	Total volume of HCl added (mL)	meq	рН
1	0	0	0	9.57
2	0.1	0.1	0.04	9.05
3	0.1	0.2	0.08	8.64
4	0.1	0.3	0.12	8.31
5	0.1	0.4	0.16	8.23
6	0.1	0.5	0.2	8.1
7	0.1	0.6	0.24	7.75
8	0.1	0.7	0.28	7.69
9	0.1	0.8	0.32	7.46
10	0.1	0.9	0.36	7.4
11	0.1	1	0.4	7.36
12	0.1	1.1	0.44	7.24
13	0.1	1.2	0.48	7.12
14	0.1	1.3	0.52	6.96
15	0.1	1.4	0.56	6.9
16	0.1	1.5	0.6	6.88
17	0.1	1.6	0.64	6.84
18	0.1	1.7	0.68	6.83
19	0.1	1.8	0.72	6.83
20	0.2	2	0.8	6.7
21	0.2	2.2	0.88	6.58
22	0.5	2.7	1.08	6.46
23	1	2.8	1.12	6.18
24	1	2.9	1.16	6.3
25	1	3	1.2	6.09
26	1	4	1.6	6.03
27	2	6	2.4	5.9
28	3	9	3.6	5.87
29	5	14	5.6	5.84
30	5	19	7.6	5.61
31	10	29	11.6	5.48
32	10	39	15.6	5.42
33	10	49	19.6	5.36
34	10	59	23.6	5.39
35	10	69	27.6	5.3

### ANC

ANC results are tabulated in the table 2. It indicates that there occurred a stedy drop in the pH value with the addition of 0.1N HCl and the millequivalence was computed using the formula. Thus the meq value corresponding to pH of 8.3 was considered as acid neutralising capacity of CKD. The ANC behaviour of unreacted CKD residue is shown in figure-2. Based on the acid neutralization capacity (ANC) test results, for the reduction of CKD pH to a value of pH = 8.3. The reduction occurred at 0.08 meq HCl/g CKD. Finally the pH of the system dropped progressively.

#### Gas –solid carbonation

The reaction of gaseous  $CO_2$  with particulate metal oxides at suitable temperatures and pressures to form carbonates is the most basic form of direct mineral carbonation. The reaction equations are as follows.

$$CaO + CO_2 \rightarrow CaCO_3$$
  
MgO + CO\_2 \rightarrow MgCO\_3  
CaSiO\_3 + CO\_2 \rightarrow CaCO\_3 + SiO\_2

 $Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2$ 

Dry capture of CKD at varying pressures indicated that there occurred maximum capture at 10bar in comparison with 1 and 5 bar studies carried out, with a fixed reaction time of 1 h as depicted in figure-3. Net carbon increase at 10 bar was  $2.65 \pm 0.03$  g of CO<sub>2</sub>/kg of CKD which was the highest in comparison. The impact of contact time on CKD was assessed by varying the time from 15 min to 3 h, with constant CO2 partial pressure. From the figure-4 it was observed that carbon capture increased with increasing reaction time up to 1 h, after which not much difference was noticed. The reaction stabilised at 60 min (1 h) in which the maximum increase occurred was  $2.65 \pm 0.08$  g of CO2 /kg of CKD.

#### Aqueous carbonation

Direct aqueous mineral carbonation involves the reaction of  $CO_2$  with alkaline residues in an aqueous suspension in a single stage.  $CO_2$  reacts with water to form bicarbonate and a proton, the proton liberates the metal ion from the mineral, and the metal ion reacts with bicarbonate to produce a carbonate precipitate.

 $\begin{array}{c} \text{CO}_2 \texttt{+} \text{H}_2 \text{O} \rightarrow \text{H}_2 \text{CO}_3 \rightarrow \text{H} \texttt{+} \rightarrow \text{HCO}^{-}_3 \\ \text{Mg}^{2\texttt{+}} \texttt{+} \text{HCO}^{-}_3 \rightarrow \text{MgCO}_3 \texttt{+} \text{H}^{+} \end{array}$ 

The carbon content increased with increasing  $CO_2$  partial pressure in the range of 1 to 10bar (Figure 5.). Among these pressures maximum carbon increase was evident at 10 bar 4.23 ± 0.1 g of  $CO_2$  /kg of CKD because the higher pressure brought forth maximum dissolution. This indicated that carbon capture was greatly influenced by high pressure. Obviously, the contact time increased the reaction kinetics was elevated with a constant pressure of 10 bar, with continuous stirring that enhanced the easy dissolution of  $CO_2$  in the sample. Thus among the considered time periods, at 30 min maximum increase of 5.09 ± 0.1 g of  $CO_2$  /kg of CKD was obtained (Figure 6.).

Comparison of the carbonation routes

The net carbon increase in CKD through *gas–solid* as well as aqueous carbonation routes. It can be clearly seen that in aqueous carbonation route the net carbon increase in CKD has increased by two folds (Figure 7.).

# CONCLUSION

Carbon capture and storage is gaining in popularity as a means to combat climate change as it allows carbon emissions to be managed during fossil fuel dependence while sustainable infrastructure is established. Mineral carbon sequestration is the only known form of permanent carbon storage and offers the possibility of carbon capture and storage in a single step. The direct carbonation process appears the most promising one. This paper demonstrates the feasibility of mineral carbonation in CKD through direct and aqueous carbonation at varying  $CO_2$  partial pressures (1 to 5 bar) and contact time (15 min to 3 h). From the experiments and studies carried out it was inferred that net carbon increase obtained in wet capture was double the amount of carbonation in comparison with dry capture. Thus CKD readily sequesters  $CO_2$  under optimized conditions. Maximum net carbon increase of 5.09 g of  $CO_2/kg$  of CKD was obtained for CKD at 10 bar pressure, contact time of 30 min and S/W ratio of 1:10. It can be concluded that CKD can be effectively utilised for sequestration of carbon dioxide ( $CO_2$ ).

# ACKNOWLEDGEMENT

Author very thankful to <sup>1</sup>Department of Remote Sensing and GIS, Tamil Nadu Agricultural University, *Coimbatore – 641003 India* provide all the necessary inputs/information to complete this work.

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#### **CITATION OF THIS ARTICLE**

C.G. Karishma,T.D. Rushendra Revathy And K. Palanivelu. Industrial Alkaline Residue for Carbon Capturing. Bull. Env. Pharmacol. Life Sci., Vol 8 [7] June 2019: 54-63