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A simple index for optimizing carbon dioxide utilization in recycled concrete aggregates using the wet carbonation method

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ABSTRACT: Securing suitable granular material for earthworks is a recurring challenge. The inevitable need to expand cities and restore infrastructure is expected to augment the stresses on geomaterial resources, accompanied by an unprecedented increase in the generated wastes, including concrete rubble. On the other hand, curbing greenhouse gas emissions has been challenging. Thus, achieving carbon neutrality requires integrating emission reduction efforts while developing innovative carbon dioxide removal solutions. Japan has commenced an initiative to achieve carbon neutrality by 2050. It introduces Artificial Carbon-Utilized Concrete (CUCO) as a promising alternative to natural granular materials. In light of that, developing an efficient artificial carbon fixation technology suited for granular recycled materials is essential to ensure the sustainability of such a system. Generally, artificial CO_2 fixation is categorized based on the carbonation conditions into dry and wet, with the latter requiring a shorter time. However, a simple method for maximizing the fixation efficiency and minimizing operational costs is needed. This study elaborates on the wet carbonation mechanism in the form of precipitated calcium carbonate and divides the governing reactions into hydration, carbonation, and decarbonation. Furthermore, considering the calcium ion concentration in the solution, the carbonation conditions are classified into steady and unsteady states, with the latter requiring about three times longer, which affects the carbonation efficiency. Finally, the optimum carbonation point is proposed as the state at which the decarbonation exceeds the carbonation rate, which can be achieved by continuously monitoring the solution's acidity (pH) and terminating the CO_2 inflow once the pH change rate approaches zero.

KEYWORDS: Carbon neutrality, artificial carbonation, sustainable sources, granular material.

1 INTRODUCTION

Securing suitable granular material for various earthworks is a recurring challenge facing geotechnical engineers. It is reported that sand and gravel are the most used solid materials, with approximately 50 billion tons being extracted annually (UNEP, 2022). Moreover, the inevitable need to expand cities and restore infrastructure is expected to augment the stresses on aggregate resources, accompanied by an unprecedented increase in the generated concrete rubble.

On the other hand, curbing greenhouse gas emissions has been challenging for various engineering sectors, with extensive efforts focusing mainly on developing low-emission technologies (Skocek et al. 2020; Kono & Aoki 2023). However, achieving carbon neutrality relying only on that is far beyond reach. Meanwhile, integrating carbon dioxide removal solutions to eliminate unavoidable emissions is a promising approach (Rosa et al. 2022).

Several initiatives have been commenced, including Japan's commitment to achieving carbon neutrality by 2050. The initiative introduces Carbon-Utilized COncrete (CUCO) as a promising alternative to natural granular materials. In contrast to the recycled concrete aggregate refinement techniques, the introduced technology utilizes the remaining cement portions in the rubble after blasting and segregation to artificially utilize Carbon Dioxide (CO_2) and generate Carbon-Utilized Recycled Concrete Aggregates (CU-RCA). In light of that, developing a reliable and efficient artificial carbon fixation technology suited for granular recycled materials is essential to ensure the sustainability of such a system.

Generally, artificially fixing CO_2 for large quantities of granular materials can be categorized based on the carbonation conditions into wet and dry. The dry method involves inducing carbonation in a sealed environment under a constant CO_2 concentration and temperature for a few weeks. In contrast, wet carbonation involves submerging the aggregates in water while injecting CO_2 gas only for a few hours. Preliminary studies on Portland cement RCAs confirmed the possibility of carbonation using both methods (Mine et al. 2024; Sakamoto et al. 2024), with the wet carbonation requiring a shorter time, thus lowering operational costs. Moreover, it was reported that using the same RCA, finer samples have higher CO_2 fixation capabilities (Hussary et al. 2024; Mine et al. 2024). However, a framework for optimizing the wet carbonation method using a simple index to maximize fixation efficiency and minimize operational costs is lacking. This study investigates the wet carbonation mechanism and proposes a simple approach to optimize the carbonation process using the leached calcium ion (Ca^{2+}) concentration before carbonation and the solution acidity (pH value) during carbonation.

2 THE WET CARBONATION AND THE GOVERNING MECHANISMS

The wet carbonation system includes a carbonation unit comprised of a reactor connected to a CO_2 source, a sample collection unit, and a monitoring unit, illustrated in Figure 1.

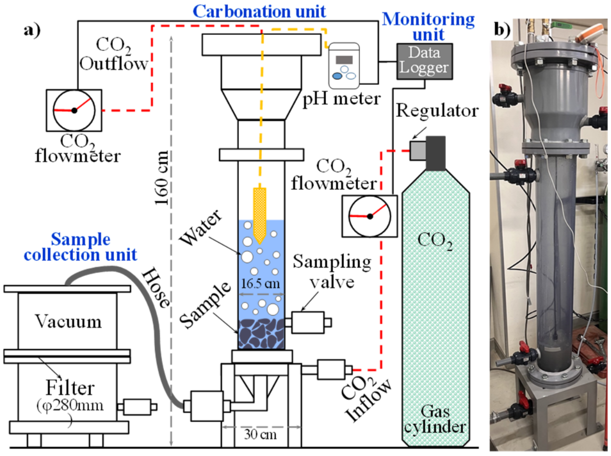

Figure 1. Wet carbonation system. a) Schematic illustration. b) Carbonation reactor.

Initially, 1.5 kg of RCA and 3 L of water are poured into the reactor from the top. The sample is left to stand in water before closing the chamber and injecting CO_2 gas from the bottom at a constant rate of 5 L/min. During carbonation, the solution pH and the CO_2 gas inflow (bottom side) and outflow (top side) are measured continuously. It must be noted that a pH meter equipped with an auto-correction function was used to account for the influence of the temperature. Meanwhile, the calcium ion (Ca^{2+}) concentration is determined every 10 minutes by extracting part of the solution through the valves and measuring with an ion meter. After carbonation, the sample is extracted and separated on a filter in the collection unit.

The RCA's wet carbonation is governed by three primary reactions: hydration, carbonation, and decarbonation, depicted in Figure 2. Initially, the cement hydrates at the surface of the aggregates, including calcium hydrates $Ca(OH)_2$, react with water, Equation (1), releasing calcium ions (Ca^{2+}) and hydroxide (OH^-) into the solution, where the latter induces an increase in the pH value. Once the CO_2 gas enters the solution, the chemical reactions, Equations (2) to (4), occur consecutively, while the released hydrogen ions (H^+) induce a decrease in the pH levels. During carbonation, the carbonate ions (CO_3^{-2}) react with Ca^{2+}, resulting in fixing carbon dioxide in the form of calcium carbonate ($CaCO_3$) that precipitates at the aggregates' surfaces, Equation (5).

$$Ca(OH)_2 \rightleftharpoons Ca^{2+} + 2OH^- \tag{1}$$

$$CO_2 + H_2O \rightleftharpoons H_2CO_3 \tag{2}$$

$$H_2CO_3 \rightleftharpoons HCO_3^{\ -} + H^+ \tag{3}$$

$$HCO_3^{\ -} \rightleftharpoons CO_3^{\ 2-} + H^+ \tag{4}$$

$$Ca^{2+} + CO_3^{\ 2-} \rightleftharpoons CaCO_3 \tag{5}$$

Once the solution becomes acidic due to the release of H^+, decarbonation progresses through the dissolution of the formed $CaCO_3$, Equation (6). It must be noted that during decarbonation, the released bicarbonate (HCO_3^-) has a strong buffering effect that stabilizes the solution's pH value (Tsuruta Kunihiro, 2022).

$$CaCO_3 + CO_2 + H_2O \rightarrow Ca^{2+} + 2HCO_3^{\ -} \tag{6}$$

3 OPTIMIZING THE WET CARBONATION METHOD

Generally, the three primary reactions, shown in Figure 2, occur simultaneously in the reactor. Accordingly, the optimum carbonation is proposed as the point at which the rate of decarbonation exceeds the carbonation rate in the wet carbonation method. A simple method is proposed for achieving the optimum carbonation point using the solution's initial Ca^{2+} concentration and the real-time pH change rate during CO_2 gas injection. RCA material, comprised of ordinary Portland cement mixed with sandstone aggregates, was adopted to elaborate on the proposed optimization method. The concrete rubble was crushed, blasted, and sieved into coarse, medium, and fine aggregates. Medium-sized aggregates with a median diameter (D_{50}) of 0.3 mm, fines content of 2.5%, and specific gravity of 2.6 were used.

3.1 *Steady-state and unsteady-state carbonation*

500g of RCA was mixed in 1 L of distilled water to investigate the Ca^{2+} ion leaching due to hydration before injecting the CO_2 gas. The ion concentration and the pH values were measured at specific intervals, as shown in Figure 3. It was found that the Ca^{2+} ion concentration increased rapidly before converging to a constant value, while the solution remained strongly alkaline with a pH value of around 13. The proposed framework divides the carbonation conditions based on the Ca^{2+} ion concentration into steady-state and unsteady-state prior to injecting CO_2 gas into the solution.

Subsequently, maintaining the same RCA to water ratio of 500 g/L, two samples of 1.5 kg to 3 L of water were prepared and left to hydrate for 20 and 100 minutes in the carbonation reactor, corresponding to unsteady-state and steady-state conditions, respectively. Meanwhile, the Ca^{2+} ion concentration for each state was confirmed to have the same values of C-1 and C-2, illustrated in Figure 3. Afterward, the CO_2 gas inflow was commenced, while the solution's pH value was measured continuously using a pH meter immersed directly in the solution inside the carbonation reactor.

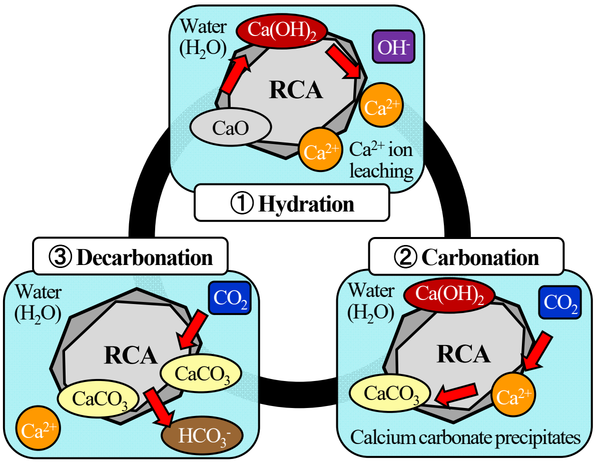

Figure 2. RCA wet carbonation mechanism and precipitation of calcium carbonate.

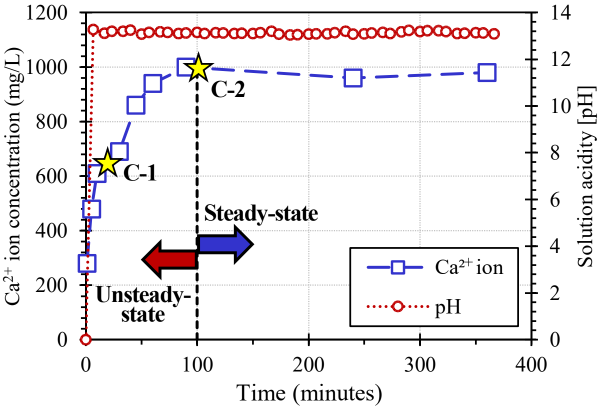

Figure 3. Calcium ion leaching and pH level development with time due to hydration.

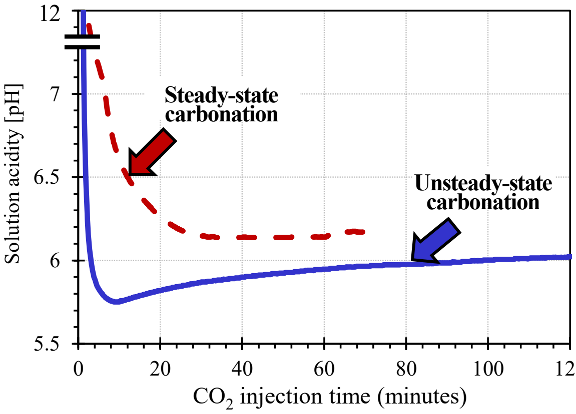

Figure 4. Solution's pH development with time due to carbonation under steady and unsteady states.

The measured pH values under the two conditions, unsteady-state (C-1) and steady-state (C-2), are illustrated in Figure 4. Regardless of the solution's initial Ca^{2+} ion state, it shifted from strongly alkaline to slightly acidic, reaching its minimum value after 10 and 30 minutes for the unsteady and steady states, respectively. Afterward, the pH value increased gradually for the unsteady-state conditions, while it remained constant for the steady-state case. This difference can be associated with the hydration reaction, which occurred mainly before the CO_2 inflow for the steady-state conditions. This reaction induces the dissolution of hydrates such as $Ca(OH)_2$ and OH^-, where the latter stabilizes the pH value of the solution at around six despite the injection of CO_2 gas. On the other hand, for the unsteady-state, the hydration reaction proceeds while the dissolving CO_2 induces the release of H^+, thus reducing the pH value to 5.8 at the early stages. Subsequently, once the carbonation progressed and $CaCO_3$ was formed, the solution's pH value increased gradually, converging to around the same value as that of the steady-state carbonation.

3.2 *The solution's pH: An index for optimizing carbonation*

The solution's pH variations during CO_2 injection are deployed to propose a simple index for determining the optimum carbonation point. A series of tests was conducted with the CO_2 inflow terminated once the pH rate changed, and the fixed CO_2 content was measured. The selected points under steady and unsteady carbonation states are illustrated in Figure 5 and Figure 6, respectively.

The loss on ignition method was adopted to estimate the CO_2 content, where around 20 g of RCA was placed in a crucible and heated using a high-temperature furnace. It is reported that the carbonation products, including calcium hydrate and calcium carbonate, thermally decompose at a temperature range of 500 and 900°C (Kanda et al. 2011). The CO_2 fixation ratio was defined as the ratio of the mass difference measured after heating at 500 and 900°C to the sample's original dry mass before ignition. The average of three samples was used for each point.

The measured CO_2 content values corresponding to each case are plotted for the steady-state and unsteady-state in Figure 5 and Figure 6, respectively. It was found that regardless of the Ca^{2+} ion state, the CO_2 content increased gradually with time due to carbonation, reaching a peak of around 3.3%, followed by a reduction in the CO_2 content due to the decarbonation reaction. Although the same peak value (CO_2 content) was achieved, the carbonation time significantly differs, extending for approximately 25 minutes and 60 minutes for the steady and unsteady states, respectively. Thus, under continuous CO_2 inflow conditions, reaching the optimum CO_2 content requires approximately three times longer carbonation and CO_2 gas volume, which strongly affects the efficiency of the wet carbonation method. Moreover, although the RCA might have a higher CO_2 fixation potential (Mine et al. 2024; Sakamoto et al. 2024; Sugimoto & Kono 2024), the decarbonation reaction limits the fixed amount under the tested conditions.

Furthermore, it was found that the pH value remained unstable until reaching the peak (optimum carbonation point), decreasing for the steady-state and increasing gradually after decreasing in the unsteady-state. This unstable behavior can be attributed to the simultaneous hydration and carbonation reactions occurring inside the reactor. On the other hand, exceeding the optimum carbonation point, the unchanging pH value can be associated with the buffering effect of the released HCO_3^-, which stabilizes the solution acidity and reflects that the decarbonation process has become the dominant reaction.

Therefore, this study proposes that the carbonation can be optimized utilizing the change rate of the pH value of the reactor's solution in the wet carbonation method by terminating the CO_2 injection just before the decarbonation becomes the dominant reaction. The pH change rate ($\Delta pH/\Delta t$), over 2-minute intervals, is analyzed for the steady-state and unsteady-state conditions. The pH change rate, focusing on the range extending from just before reaching the minimum pH value and afterward for each state, is plotted in Figure 7. Moreover, the time corresponding to the optimum carbonation point (peak value) is also depicted for the steady and unsteady states. It was found that the optimum carbonation point coincides with the point at which the pH change rate approaches zero, representing the inflection point where the buffering effect becomes dominant due to the HCO_3^- release associated with the decarbonation reaction.

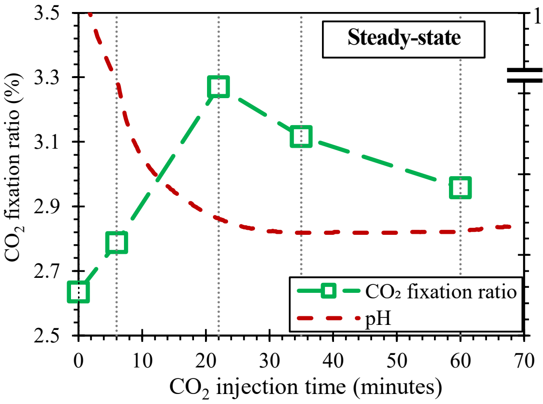

Figure 5. Steady-state fixed CO_2 content and the acidity of the solution.

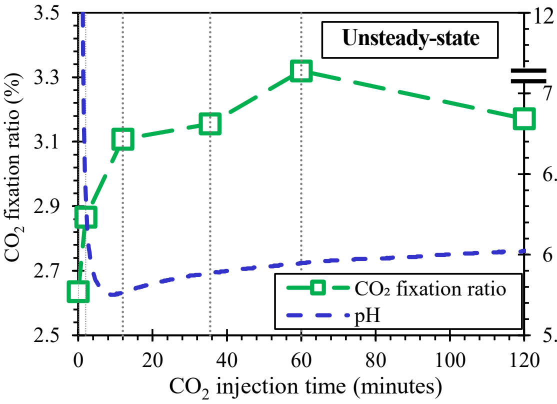

Figure 6. Unsteady-state fixed CO_2 content and the acidity of the solution.

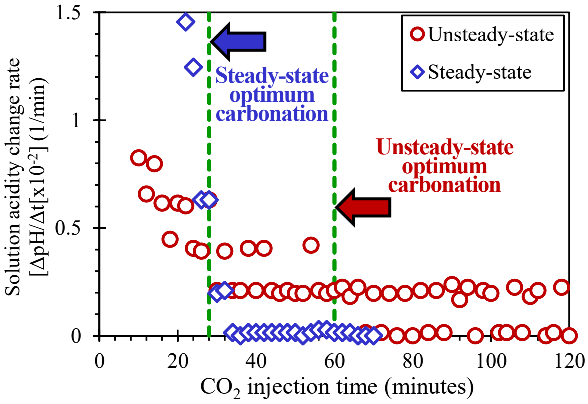

Figure 7. Solution's acidity change rate and the optimum carbonation point under steady and unsteady states.

3.3 Post-carbonation: Influence of the reactors' solution on the fixed CO_2 content

Generally, using the wet carbonation system, extracting the sample from the device after carbonation extends for several minutes. The influence of the reactor's solution on the fixed CO_2 content after terminating the CO_2 inflow was investigated by leaving three different carbonated samples immersed in the reactor's solution for 10, 30, and 60 minutes. Two types of samples, carbonated to the optimum carbonation point and until the solution pH value reached 7, were used. The changes in the fixed CO_2 content with time for the two tested conditions are summarized in Figure 8. It was confirmed that the fixed CO_2 content remained unchanged regardless of the time, suggesting that the wet carbonation treatment is not affected by the post-carbonation reaction between the carbonated aggregates and the reactor's solution.

Finally, it can be concluded that the optimum carbonation point can be practically achieved by continuously monitoring the solution's acidity (pH value) and terminating the CO_2 injection once the pH change rate approaches zero. Meanwhile, the influence of the post-carbonation reaction between the carbonated aggregates and the reactor's solution, during the extraction of aggregates, on the carbonation efficiency is small and can be neglected. Moreover, allowing the RCA to stand in the solution until the Ca^{2+} ion concentration converges to a steady state before injecting CO_2 gas significantly shortens the carbonation time, thus enhancing the efficiency.

4 CONCLUSIONS

A framework for optimizing carbon dioxide utilization in recycled concrete aggregates using the wet carbonation method is established. Initially, the study elaborates on the carbonation mechanism in the form of precipitated calcium carbonate and divides the governing reactions into hydration, carbonation, and decarbonation. Moreover, the optimum carbonation point is introduced as the state at which the decarbonation exceeds the carbonation rate in the wet carbonation method.

Considering the calcium ion concentration before injecting CO_2 gas into the solution, the carbonation conditions are classified into steady and unsteady states. Furthermore, a simple method is proposed for achieving the optimum carbonation point using the real-time pH change rate (during CO_2 injection). It was found that the point at which the pH change rate of the solution approaches zero corresponds to the optimum carbonation point. Moreover, injecting CO_2 for longer durations induces the dissolution of the formed $CaCO_3$, accompanied by the release of HCO_3^-, with its buffering effect stabilizing the solution's acidity.

Finally, it is proposed that effectively deploying the wet carbonation method in practice can be achieved by continuously monitoring the solution's acidity (pH value) and terminating the CO_2 inflow once the pH change rate approaches zero. However, pH meters equipped with a temperature correction function are recommended to detect the optimum carbonation point distinctly. Moreover, allowing the aggregates to stand in the solution until the Ca^{2+} ion concentration converges to a steady state before injecting CO_2 gas significantly shortens carbonation time.

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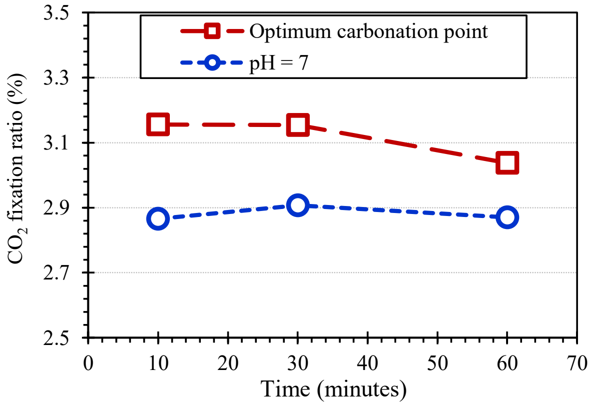

Figure 8. Influence of the reactors' solution on the fixed CO_2 content [Post-carbonation].

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