

Article

An Eco-Friendly Neutralization Process by Carbon Mineralization for Ca-Rich Alkaline Wastewater Generated from Concrete Sludge

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Received: 28 July 2017; Accepted: 11 September 2017; Published: 13 September 2017

Abstract: Waste-concrete recycling processes using wet-based crushing methods inevitably generate a large amount of alkaline concrete sludge, as well as wastewater, which contains abundant Ca ions. The Ca-rich alkaline wastewater must then be neutralized for reuse in the waste-concrete recycling process. In this study, the feasibility of a carbon mineralization process for the neutralization of alkaline wastewater was considered from both environmental and economic perspectives. The optimal reaction time, efficiency of Ca removal and CO₂ sequestration as a function of the CO₂ gas flow rate were assessed. The carbon mineralization process resulted in sequestering CO₂ (85–100% efficiency) and removing Ca from the solution (84–99%) by precipitating pure CaCO₃. Increasing the gas flow rate reduced the reaction time (65.0 down to 3.4 min for 2.5 L of solution), but decreased CO₂ sequestration (from 463.3 down to 7.3 mg CO₂ for 2.5 L of solution). Optimization of the gas flow rate is essential for efficient CO₂ sequestration, Ca removal, CaCO₃ production and, therefore, successful wastewater neutralization following the wet-based crushing process. The method presented here is an eco-friendly and economically viable substitute for dealing with alkaline wastewater. It may also provide a practical guide for the design of carbon mineralization processes for the neutralization of alkaline solutions containing large amounts of Ca.

Keywords: alkaline concrete sludge; carbon mineralization; carbonation; wastewater neutralization; carbon dioxide

1. Introduction

The amount of construction waste produced in Korea increases yearly due to the significant number of aged buildings requiring re-construction. Shortages in raw materials and available landfill sites mean an improvement in construction waste recycling is required [1]. Currently, waste concrete accounts for approximately 63% of construction waste; a total of 198,000 tons generated per day. As a result, more than 95% of waste concrete is used as aggregate material in new concrete [2,3]. In the intermediate-construction waste recycling process, crushing and screening produces a large amount of concrete sludge composed of fine-grained concrete material. Water is used to wash down the concrete surface and separate impurities [4].

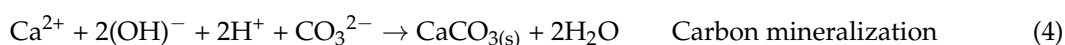
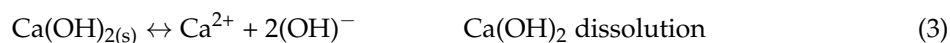
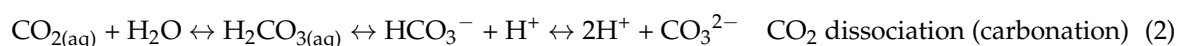
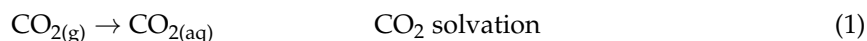
This study assessed the recycling process of a local waste-concrete treatment plant. The wet-based crushing method consisted of crushing/screening, classifying and flocculating concrete fines, as illustrated in Figure 1. Dried sludge, precipitated using cationic-organic polymer coagulants was discarded at a rate of 10 tons/day. The total amount of solid phase included in the concrete sludge from recycled concrete-aggregate production was approximately 7%, increasing up to 40% for high quality aggregate products.



Figure 1. Photographs of a field waste concrete recycling process.

The concrete sludge was strongly alkaline and rich in calcium (Ca) as a result of calcium hydroxide ($\text{Ca}(\text{OH})_2$) dissolution from the concrete particles [5]. The air-dried solids, generated from the solid-liquid separation and flocculation process, were discarded as they were considered to have insignificant economic value. The supernatant from the flocculation process was reused in the wet-based crushing processes, without further treatment. The best practice, from both environmental and economic stand points, should include neutralization for discharge into streams or, ideally, neutralization and reuse to remove cement paste in the wet process of waste concrete production. Treatment using sulfuric acid (H_2SO_4) is commonly employed to neutralize alkaline wastewater, because of the simplicity of the process. However, it does increase the agent cost consumption [6]. This added cost is a significant barrier to the development of the whole waste-concrete recycling process. In addition, neutralization of the wastewater using acids, without Ca removal, is not effective in removing cement paste from the aggregates in the waste-concrete recycling process due to the over-saturation of Ca.

Since the Kyoto Protocol in 1997, countries of the world have been actively working to reduce carbon dioxide (CO_2) emissions, which are emitted from power plants and industry [7,8]. Among possible strategies for reducing atmospheric CO_2 concentrations, carbon capture utilization and sequestration (CCUS) technologies are the most available and widely-applied strategies [9]. The carbon mineralization process is an attractive carbonate-based mineral production technique for industries. In addition, it is also a CO_2 sequestration technique to reduce CO_2 atmospheric levels through the consumption of flue gas [10]. It also neutralizes alkaline solutions [8,11,12] and results in the production of useful carbonate-based minerals such as calcium carbonate (CaCO_3). The carbon mineralization process in a Ca-rich solution occurs according to the following Reactions (1)–(4) [13–17].



In aqueous solution, protons and carbonate ions are generated through dissociation of CO_2 (Equations (1) and (2)). The dissolution of $\text{Ca}(\text{OH})_2$ in concrete by the solubility product constant (K_{sp} of $\text{Ca}(\text{OH})_2 = 5.5 \times 10^{-6}$) results in several alkaline solutions (approximately pH 12.6) and

Ca saturation in solution (approximately 800 mg/L) according to Equation (3). In the field plant, the solution pH and Ca concentration conditions could fluctuate on a daily basis. Protons produced in the carbon mineralization process can be very effective in neutralizing alkaline solution from concrete sludge. In addition, CaCO_3 by-products produced by this process can be used in other industries for cement production, paper filling/coating additives and plastic/paint manufacturing according to purity and varying particle sizes [18,19]. Numerous studies have been undertaken recently on the carbon mineralization processes using Ca-rich alkaline solution from various sources, focusing on the production and characterization of CaCO_3 particles (e.g., rhombic calcite, orthorhombic aragonite, spherical vaterite; $\mu\text{-CaCO}_3$) [15,20–23].

In comparison, this study focuses on the development of an eco-friendly, alkaline wastewater neutralization process without the use of acids. It was designed to sequester CO_2 with CaCO_3 and to recycle the neutralized water through the carbon mineralization process in wet-based crushing procedures. To this end, a laboratory-scale carbon mineralization process was designed to neutralize wastewater generated from concrete sludge during the waste concrete recycling process (Figure 2). The neutralized water, from which Ca ions were removed, is expected to be effectively recycled to remove the cement paste absorbed into aggregate surfaces in the wet-based crushing/screening of the waste-concrete recycling process.

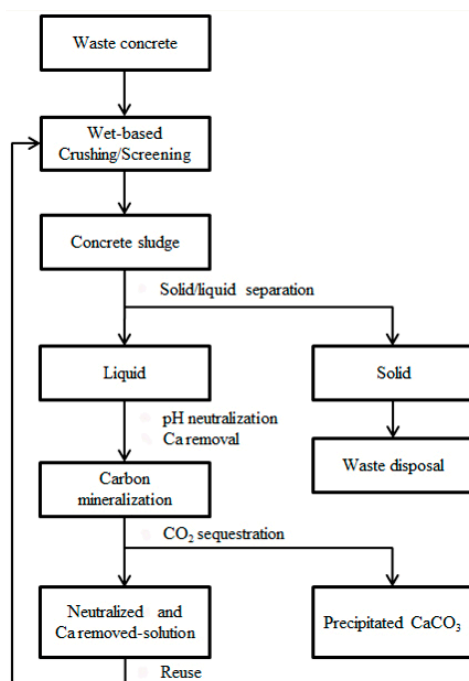


Figure 2. A schematic flow diagram of the neutralization of Ca-rich alkaline wastewater from a waste concrete recycling process by carbon mineralization.

2. Materials and Methods

2.1. Characterization of Concrete Sludge

Approximately 3600 m³ of concrete sludge and wastewater are generated daily through the wet-based crushing/screening process. The concrete sludge sample used in this study was collected from a field construction waste treatment plant (CWTP) in Incheon, Korea. Sludge moisture content was analyzed by weight loss with oven-drying at 105 °C until the weight did not change. The pH and electrical-conductivity (EC) of the sludge were measured using calibrated pH (ORION VSTAR-PH, Thermo Fisher Scientific Inc., Waltham, MA, USA) and EC meters (HQ40d, HACH, Loveland, CO, USA). The sludge particles were separated according to Korean Standard

Testing Method (KS F 2309) based on wet sieving using 70, 100, 200, 325 and 400 mesh, with sludge solution instead of fresh water used to avoid further dissolution of minerals such as $\text{Ca}(\text{OH})_2$. The particle size distribution was determined by weight loss at 105 °C for 24 h after separation by wet sieving. Significant impurities were present in the less than 70 mesh sieve fraction (0.212 mm), so the collected sludge was further separated by vacuum filtration using filter papers with a pore size of 0.45 μm . After filtration, the liquid phase was kept at room temperature to prevent precipitation of natural CaCO_3 through a reaction between Ca in solution and atmospheric CO_2 . The residual solids were completely dried at 105 °C for 24 h. The concentrations of aqueous Ca, K and Na were measured with an inductively-coupled plasma optical emission spectrometer (ICP-OES, Agilent, 720-ES, Santa Clara, CA, USA). The qualitative and quantitative analysis of sludge particle mineralogy was performed by X-ray diffraction (XRD, Philips, X'pert MPD, Almelo, The Netherlands). The chemical composition of the sludge was measured by X-ray fluorescence (XRF, MXF-2400, Shimadzu, Kyoto, Japan). The XRD and XRF analyses were performed on oven-dried samples sieved through 200 mesh (0.075 mm) after vacuum filtration and pulverization with a mortar and pestle.

2.2. Carbon Mineralization

2.2.1. Apparatus of the Carbon Mineralization Processing Reactor

In this study, a carbon mineralization process was designed to neutralize wastewater generated from concrete sludge, and a schematic experimental flow diagram is provided in Figure 3. A batch type acrylic-material based experimental reactor was manufactured with a volume of 3.0 L (Φ : 140 mm, H: 200 mm) and a system created for measuring the solution pH and EC during the carbon mineralization process. In addition, the reactor was sealed in order to measure the volume of venting gas that had not reacted in aqueous solution to the injected gas volume and to prevent the gas leaking from the reactor. The exact volume of CO_2 gas flow was controlled by a mass flow controller (MFC- CO_2) and a mass flow management system (MFM- CO_2). The total volume of gas venting was continuously analyzed using a gas flow and pressure controller (GMC1200, ATOVAC, Yongin-si, Korea).

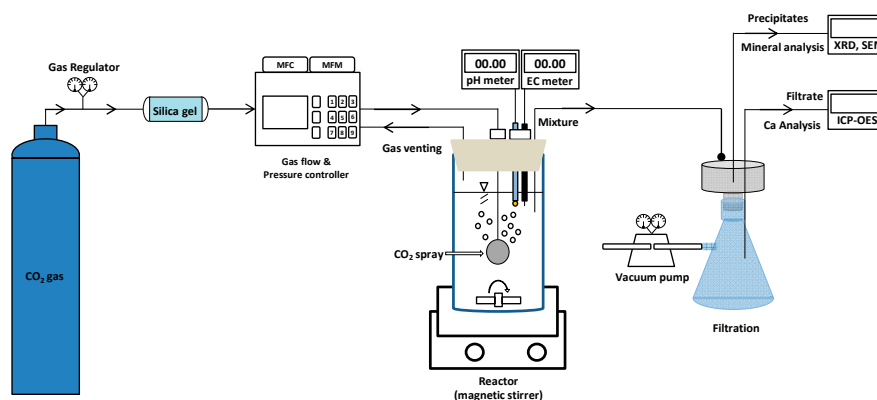


Figure 3. A schematic experimental flow diagram of the carbon mineralization process (MFC: mass flow controller; MFM: mass flow management system; EC: electrical-conductivity).

2.2.2. Carbon Mineralization Experiment

The experimental conditions for all carbon mineralization experiments were as follows: 2.5 L of wastewater were reacted using a magnetic stirrer until the pH dropped to 8.5 at room temperature. The 99.9% CO_2 gas flow rates (Jungang Gas Co., Ltd., Daejeon, Korea) were varied from 8–200 cc $\text{CO}_2/\text{min}/\text{L}$ (0.014–0.360 g $\text{CO}_2/\text{min}/\text{L}$). Solution pH and EC, as well as the accumulated volume of gas venting to the injected volume of gas were measured during the experiments. After the experiments, the solution containing precipitates was immediately passed through a 0.45- μm paper with a vacuum filter press (0.6 MPa). The weight of the residual precipitates was measured after

oven-drying at 105 °C for 4 h. The mineralogy and morphology of the precipitates were analyzed by XRD and scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS, 6380LA, JEOL Ltd., Akishima-shi, Japan), respectively. All experiments were carried out in duplicate for quality assurance.

2.3. Neutralized Water Recycling

In this study, the removal efficiency of the neutralized waste water by carbon mineralization was evaluated in wastewater neutralized by carbon mineralization and subsequently reused in the wet-based crushing of the waste-concrete recycling process. The laboratory-scale experiments using a batch-type stirring vessel were carried out as follows: 15 g of raw-concrete powder (<75 µm; crushed using a jaw-crusher) were added to 150 mL of raw concrete sludge solution and neutralized water by the carbon mineralization processes (8, 40 and 200 cc CO₂/min/L). The result was compared with that of as-received sludge solution. Tap water was also used as a control test. The mixture was mixed using a magnetic stirrer at 300 rpm for 2, 5, 10, 30, 60, 120 and 180 min at ambient temperature under atmospheric conditions. At the specified time, 10 mL of mixture were sampled and then passed through a syringe filter (0.45 µm). Solution pH and Ca concentration were measured from the filtrate, as per Section 2.1.

3. Results and Discussion

3.1. Physicochemical Characteristics of the Sample

The physicochemical characteristics of the sludge sample are summarized in Table 1. The sludge pH was strongly alkaline (approximately 12.2) due to the hydroxide ions generated from dissolution of Ca(OH)₂ contained in the cement (Equation (3)) [24]. The dissolved Ca ions in the concrete sludge were also sourced from Ca(OH)₂ in the cement. Here, the pH and Ca concentration in solution were determined by the solubility constant of Ca(OH)₂ (K_{sp} : 5.5×10^{-6}) [17,25]. The sludge in this study was at pH 12.2 and contained approximately 330 mg/L of Ca. The high concentration of dissolved Ca²⁺, Na⁺, K⁺ and OH⁻ in the sludge solution increased the EC to approximately 8.9 dS/m [26,27]. Quartz (SiO₂), calcite (CaCO₃) and the feldspar mineral albite (NaAlSi₃O₈) were identified by XRD in dried sludge particles (Figure 4a) [3]. Portlandite (Ca(OH)₂), present in the original concrete (Figure 4b), was not observed in the concrete sludge XRD spectrum, possibly due to dissolution during the wet-based crushing/screening process. In addition, quartz and feldspar, including albite and microcline (KAlSi₃O₈), were derived from the mortar, the concrete production process or both [28,29], and lime had carbonated with the atmospheric CO₂ [30]. Less calcium oxide was present in the concrete sludge compared with the concrete powder due to the dissolution of C₃S, C₂S, C₃A, C₄AF and Ca(OH)₂ (Figure 4b and Table 2).

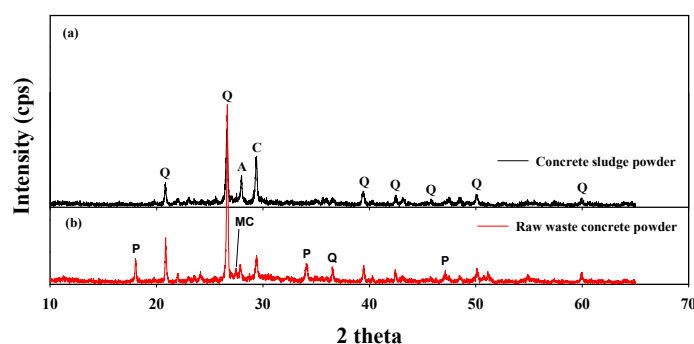


Figure 4. The X-ray diffractogram for concrete sludge powder (a) and raw waste concrete powder (b); peaks are identified for quartz (Q: SiO₂), calcite (C: CaCO₃), albite (A: NaAlSi₃O₈), microcline (MC: KAlSi₃O₈) and portlandite (P: Ca(OH)₂).

Table 1. Physicochemical properties of concrete sludge used in this study.

Parameter	Value
pH	12.2
EC (dS/m)	8.9
ORP ^a (mV)	−108.0
Moisture content (%)	92.5
Particle size distribution (wt %) (KS F 2309 ^b)	
>70 mesh (>0.212 mm)	1.3
70–100 mesh (0.212–0.150 mm)	0.4
100–200 mesh (0.150–0.075 mm)	7.9
200–325 mesh (0.075–0.043 mm)	9.5
325–400 mesh (0.043–0.038 mm)	7.7
<400 mesh (<0.038 mm)	73.1
Elements in aqueous solution (mg/L)	
Ca	333.9 ± 2.0
K	257.1 ± 1.4
Na	317.5 ± 0.0

^a ORP: Oxidation Reduction Potential; ^b KS F 2309: Korean Standard Testing Method.

Table 2. Chemical composition of dried-solid particles in concrete sludge (XRF analysis).

Oxide	Composition (wt %)	
	Dried Concrete Sludge	Raw Waste Concrete
SiO ₂	45.9	38.5
CaO	20.2	30.2
Al ₂ O ₃	9.4	6.9
Fe ₂ O ₃	3.0	4.1
MgO	1.5	1.8
K ₂ O	2.3	1.9
LOI ^a	15.4	14.5
Others ^b	1.8	1.6

^a Loss of ignition; ^b Na₂O, TiO₂, MnO and P₂O₅ included.

3.2. Carbon Mineralization

3.2.1. Changes in Solution pH and EC

Figure 5 tracks the changes in solution pH and EC with gas flow rate during the carbon mineralization process. Protons generated by the carbonation reaction (Equations (1) and (2)) decreased the solution pH while consuming OH[−] ions [31]. The total reaction time required to neutralize alkaline solution pH was strongly dependent on the amount of CO₂ gas injected [15]. These trends can be explained by the difference in the amount of protons generated from bicarbonate and carbonate reactions, following the dissolution of CO₂ gas in aqueous solution. Azdarpour et al. [32] also reported that an increase in CO₂ gas pressure (i.e., an increase in CO₂ gas injection) enhanced the carbonation efficiency. In this current study, the protons that led to the decrease in solution pH were generated by the formation of CO₃^{2−}, not HCO₃[−], above a solution pH of 8.5 [33]. The generation of CO₃^{2−} cannot be accelerated under a solution pH of approximately 8.0 due to the equilibria of the carbonate system (i.e., acid dissociation constant: pK_a). This phenomenon can be explained by the changes in solution EC during the carbon mineralization process and used to indirectly understand the changes in conductive ions in solution. Interestingly, solution EC rapidly decreased with the consumption of Ca ions due to the precipitation of CaCO₃. Finally, solution EC was maintained at approximately 2.90 dS/m in all experiments due to the remaining conductive ions, including Na and K in solution, except where the gas flow rate was 200 cc CO₂/min/L. Jo et al. [34] also observed that solution pH and EC simultaneously decreased with reaction time during the carbonation process in Ca-rich solutions. They reported that the decrease in solution EC was terminated due to the consumption of Ca ions through CaCO₃ precipitation. In the 200 cc CO₂/min/L experiment of the current study, however,

the solution EC rapidly increased from 3.07 dS/m (the lowest EC value) to 3.47 dS/m at the end of the reaction. Han et al. [35] reported that CaCO_3 produced by CO_2 injection in the carbon mineralization process was converted to a soluble form of $\text{Ca}(\text{HNO}_3)_2$ at solution pH below 8.3, hence the slight increase in solution EC. This phenomenon might be interpreted as a stabilization of the final solution pH around 8.0, due to continuous proton production by a significant amount of un-solvated CO_2 gas, even though the gas injection was stopped at solution pH 8.5. Chang et al. [36] reported that a higher CO_2 flow rate decreased carbonation conversion due to poor CO_2 mass transfer between gas and liquid phases; hence, the increase in Ca ions in solution with the dissolution of CaCO_3 at a pH of approximately 8.0 [37]. Consequently, under high gas flow conditions, gas injection control is crucial for targeting the final solution pH to avoid re-dissolution of CaCO_3 .

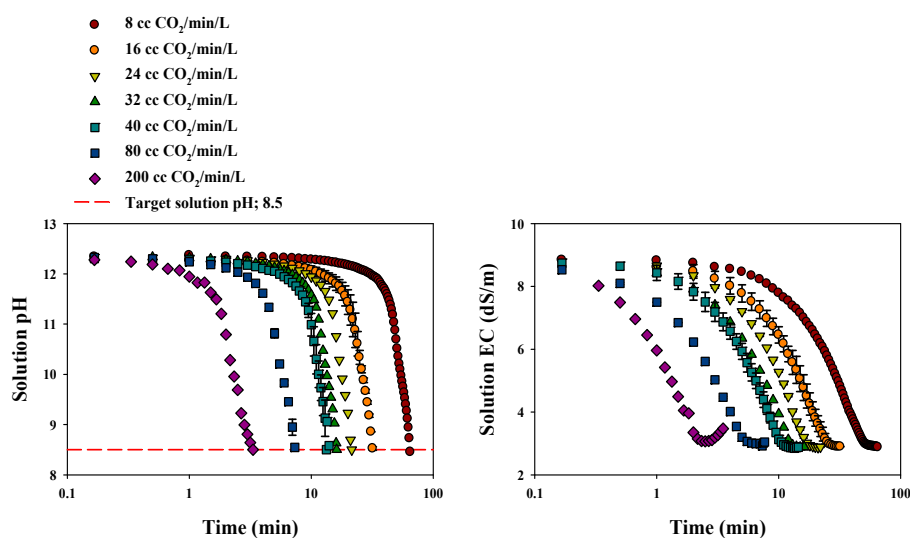


Figure 5. Changes in solution pH and EC during the carbon mineralization processes.

3.2.2. CO_2 Sequestration

The overall results of carbon mineralization experiments, with changes in the CO_2 gas flow rate, are presented in Table 3. The consumption of the CO_2 gas was calculated as in the following equation:

$$\text{CO}_2 \text{ sequestration} = \left(1 - \frac{V_{\text{out-accumulated}}}{V_{\text{in-accumulated}}}\right) \quad (5)$$

where $V_{\text{in-accumulated}}$ and $V_{\text{out-accumulated}}$ refer to the total volume of accumulated CO_2 gas, injection and venting, respectively. The amounts of total CO_2 injection and venting were calculated based on conditions at atmospheric pressure and a temperature of 20 °C.

Table 3. Overall results of carbon mineralization processes with changes in CO_2 gas flow rates (2.5 L of solution).

CO_2 Gas Flow Rate (cc/min/L)	Time (min)	Final EC (dS/m)	Total CO_2 Injection (g)	Total CO_2 Venting (mg)	CO_2 Sequestration ($1 - (V_{\text{out}}/V_{\text{in}})$)	Conc. of Ca (mg/L)	Precipitated CaCO_3 (g)
8	65.0	2.9	2.38	7.33	1.00	2.64	2.69
16	32.0	2.9	2.09	13.74	0.99	3.00	2.60
24	22.0	2.9	2.23	59.52	0.97	5.76	2.64
32	16.2	2.9	2.33	120.87	0.95	6.58	2.58
40	13.3	2.9	2.35	129.11	0.95	7.55	2.66
80	7.5	3.0	2.72	212.44	0.92	23.13	2.50
200	3.4	3.5	3.05	463.33	0.85	53.21	1.78

More than 85% of the CO₂ gas injected into the solution was consumed in neutralizing processes. Despite the reduction in the total reaction time required for neutralizing the alkaline solution pH, the increase in CO₂ gas flow accelerated gas venting to the atmosphere instead of solvation within the aqueous solution. The continuous injection of CO₂ gas resulted in a gradual increase in the CO₂ gas venting due to saturation of carbonate ions with the decrease in Ca concentration in solution (Figure 6). Jo et al. [38] reported that the increase in carbonation time (i.e., with a lower CO₂ gas flow injection rate) enhanced CO₂ sequestration by slowing the decrease in solution pH. Furthermore, the increase of CO₂ gas flow accelerated the gas venting with higher residual Ca concentrations in solution; thus, the amount of precipitated CaCO₃ was reduced. This indicates that excessive CO₂ gas flow (200 cc CO₂/min/L) favors the precipitation of vaterite, due to the lack of reaction time [14,15]. Hence, Ca was re-extracted into solution from the vaterite with the decrease in solution pH [34]. This appears to agree with the rapid increase in solution EC from 3.1 dS/m–3.5 dS/m and the increase in Ca ions in solution.

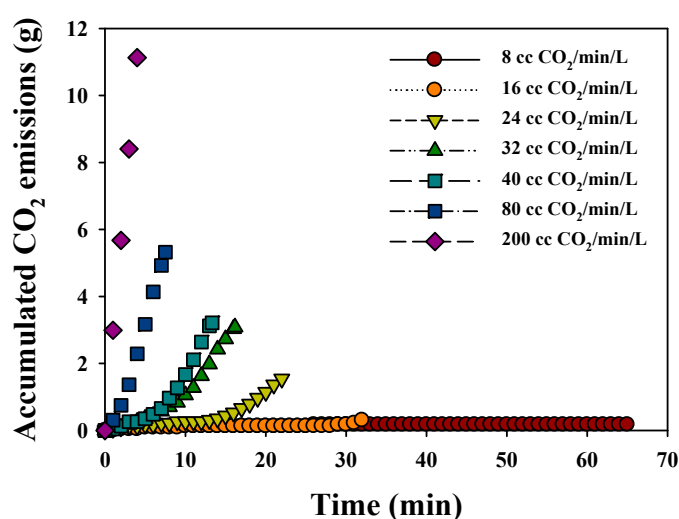


Figure 6. Accumulated CO₂ venting to the atmosphere during carbon mineralization processes.

An efficient carbonation process requires optimal neutralizing conditions to balance CO₂ sequestration, reaction time and dissolved Ca concentration remaining in solution. The relationships between these parameters are graphed in Figure 7. The fitting results reveal a good correlation between CO₂ sequestration ($R^2 = 0.9996$), reaction time ($R^2 = 0.9317$), Ca concentration remaining in solution ($R^2 = 0.9928$) and CO₂ gas flow rates. According to equations calculated from each correlation result, the CO₂ sequestration had to be 97.7% per total CO₂ injection to achieve both pH neutralization and over 99% removal of Ca from the solution. The required reaction time for this CO₂ sequestration was calculated at 32.4 min.

The amount of concrete sludge generated from the field treatment plant was approximately 3600 m³/day. The amount of CO₂ gas sequestered by pH neutralization in the field plant was calculated for each gas flow rate selected in this study. Based on high purity CO₂ gas (99.9%), the expected amount of CO₂ gas sequestered was between approximately 21.3–26.7 and 0.1–4.0 tons CO₂/day, at gas injection rates of 8–200 cc CO₂/min/L, respectively. In comparison, based on the low CO₂ content (15 vol %) of the flue gas discharged from coal-fired power plants, the amount of flue gas sequestered and vented would be calculated between approximately 142.3–177.7 and 0.5–27.0 tons flue gas/day, respectively. In addition to the CO₂ sequestration benefits of the carbon mineralization process, approximately 99.2% purity CaCO₃ material could be produced. Applying the carbon mineralization process to the field-scale treatment plant (3600 m³/day), it was calculated to produce between approximately 2.6 and 3.9 tons CaCO₃/day. At a higher rate of CO₂ gas injection, spherical vaterite formed within the shortened reaction time (Figures 8 and 9). In contrast, a longer application of the carbon mineralization

process, with a lower gas flow rate, resulted in the vaterite transforming to more stable calcite crystals by reacting with water in the aqueous phase under atmospheric pressure conditions [18,39].

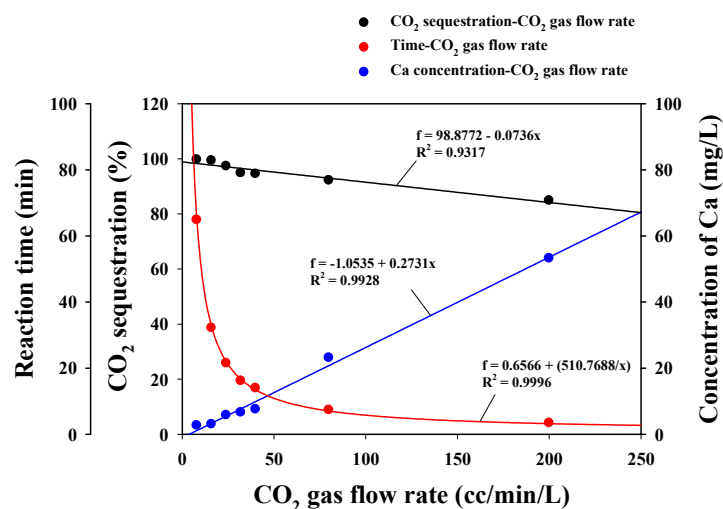


Figure 7. Correlation between CO₂ sequestration, reaction time and residual Ca concentration and CO₂ gas flow rate during the carbon mineralization processes.

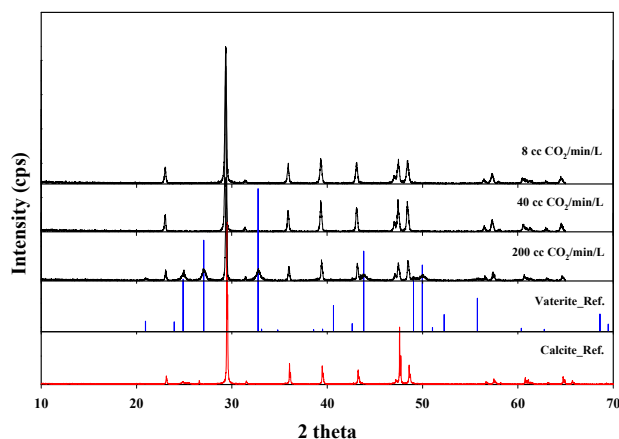


Figure 8. The XRD spectra of particles precipitated during the carbon mineralization processes at 8, 40 and 200 cc CO₂/min/L.

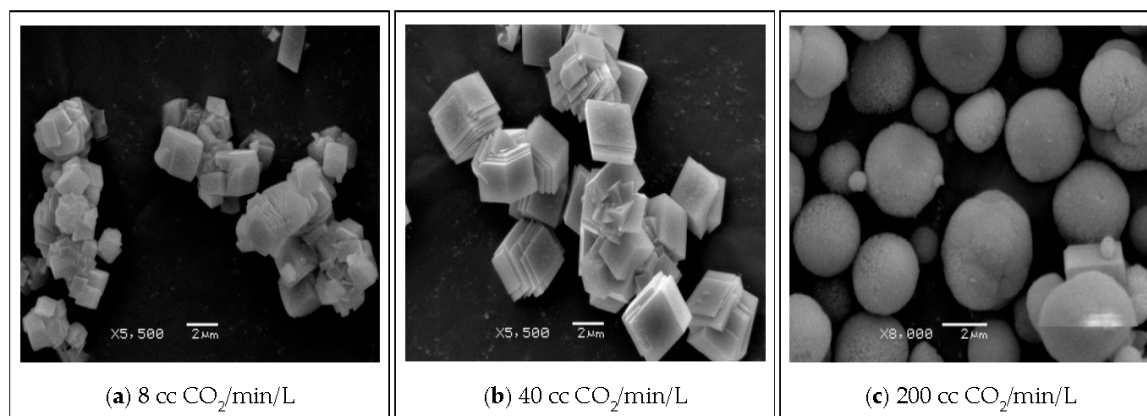


Figure 9. (a–c) The SEM images of CaCO₃ particles precipitated through the carbon mineralization processes (8, 40 and 200 cc CO₂/min/L).

The increase in CO₂ gas flow rate may have a positive effect on operating time and power consumption for the field-scale neutralization of alkaline wastewater. However, the high gas flow rate could cause problems such as a reduction in the amount of precipitated-CaCO₃ and an increase in gas venting. Thus, when neutralized-water with a high Ca content is re-used, it could lead to a reduction in the efficiency of the wet-based crushing method in the waste-concrete recycling process. This study focused on the feasibility of neutralizing and re-using wastewater generated from concrete sludge without using acids; a detailed field-scale investigation is required to understand the overall economy of the process.

3.3. Evaluation of Neutralized Waste Water

The removal efficiency of cement paste from aggregates was evaluated using raw concrete sludge solution before and after the suggested neutralization processes; it was also compared with the tap water control. The changes in solution pH and Ca concentration, with time, are presented in Figure 10. The waste water neutralized by carbon mineralization showed much lower Ca content as it was consumed in CaCO₃ production. Solution pH and Ca concentration rapidly increased initially for tap water and neutralized water due to the dissolution of Ca(OH)₂ in cement. However, they then slightly decreased with time, which means that a natural CaCO₃ precipitate was produced through the reaction between atmospheric CO₂ and Ca in solution. In the case of raw sludge solution, however, pH did not increase significantly, and Ca saturation was observed. In addition, the maximum solution pH and Ca concentrations were, for the most part, constant across the treatments due to Ca saturation of the solution. This indicates that with a higher gas flow rate the Ca remaining in water neutralized by carbon mineralization and raw sludge solution may have a negative effect on the removal Ca(OH)₂ in cement from aggregates. Therefore, it should be possible to significantly manipulate the gas flow rate in the carbon mineralization process to enhance the removal of cement paste from the wet-based crushing process during the waste-concrete recycling process.

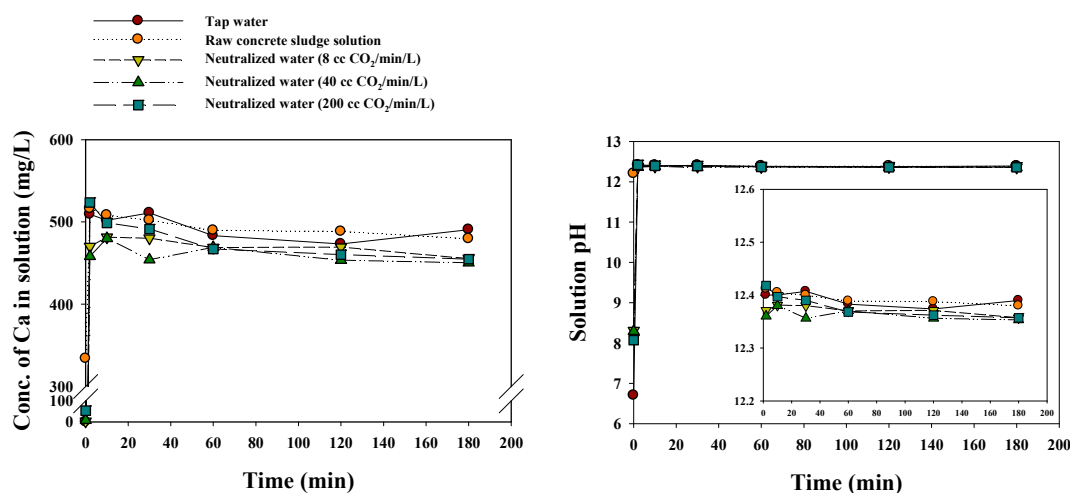


Figure 10. Changes in Ca concentration and solution pH from raw concrete powder dissolution using tap water, raw concrete sludge solution and neutralized water by carbon mineralization (8, 40 and 200 cc CO₂/min/L).

4. Conclusions

In this study, the carbon mineralization processes were investigated as a function of CO₂ gas flow rates (8–200 cc CO₂/min/L solution) to neutralize (target solution pH of 8.5) the Ca-rich alkaline concrete sludge solution generated from the recycling of waste concrete using a wet-based crushing process. Higher gas flow rates accelerated the drop in solution pH and EC; protons were generated by the carbonation reaction, while Ca ions precipitated out of solution as CaCO₃. However, a high

gas flow could lead to problems such as CO₂ venting to the atmosphere and an increase in soluble Ca, as CaCO₃ dissolved with a solution pH drop below the target pH of 8.5. In addition, water neutralized by a carbon mineralization process with a high gas flow rate, which remained Ca enriched, may not be efficient in the removal of cement paste (Ca(OH)₂) from the wet-based crushing process of the waste-concrete recycling process. Therefore, the control of gas flow into the solution should be systematically manipulated in the carbon mineralization process to neutralize alkaline solutions containing large amounts of Ca. This process could make significant environmental and economic contributions by:

- (1) Providing an eco-friendly neutralization process for the treatment of alkaline wastewater.
- (2) Improving the cement paste removal process through the reuse of neutralized wastewater in the wet-based crushing process.
- (3) Sequestering CO₂ in CaCO₃.
- (4) Producing, as a by-product, a commercially viable pure source of CaCO₃.

Acknowledgments: This research was supported by the Basic Research Project (17-3422) of the Korea Institute of Geoscience and Mineral Resources (KIGAM).

Author Contributions: Heeyoung Shin conceived and designed the experiments; Jongchan Yoo performed the experiments and analyzed the data; Heeyoung Shin and Sangwoo Ji contributed reagents/materials/analysis tools; Jongchan Yoo wrote the paper.

Conflicts of Interest: The authors declare no conflict of interest.

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