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## Characteristics and environmental benefits of CO<sub>2</sub> mineralization using a recyclable chelating agent in concrete manufacturing

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Concrete product manufacturing faces two coupled challenges: substantial solid waste generation and CO<sub>2</sub> emissions. While CO<sub>2</sub> mineralization can address both, conventional routes rely on high reagent use and generate wastewater, limiting sustainability. Recyclable chelating agent-assisted CO<sub>2</sub> mineralization offers a more sustainable alternative, yet its cycle-resolved practicality and net environmental benefits remain unclear. This study assessed the characteristics of such CO<sub>2</sub> mineralization processes through experiments using green chelating agent GLDA as the extractant and sludge cake collected from a concrete pole facility as the feedstock. Over ten successive reuse cycles, the efficiency of Ca extraction by the GLDA solution remained stable, yielding a 25% reduction in residue mass. The extracted Ca was selectively carbonated as CaCO<sub>3</sub> after heating the solution to 95 °C, mineralizing 156 g CO<sub>2</sub> per kg of sludge cake. A prospective, gate-to-gate life-cycle assessment (LCA) based on experimental results demonstrates a 16.1% reduction in global warming potential at concrete manufacturing plants. It also reveals a 1.2–10.0% decrease across other key environmental categories, including abiotic depletion potential (fossil fuels) and acidification potential, primarily driven by residue reduction. These findings position recyclable chelating agent-assisted CO<sub>2</sub> mineralization as a scalable option that couples waste minimization with permanent CO<sub>2</sub> storage for the concrete products industry.

**Keywords** Concrete product manufacturing, CO<sub>2</sub> mineralization, Life cycle assessment, Chelating agent

With the rapid advancement of urbanization and the growing demand for infrastructure, the global annual concrete production is projected to increase from 14 billion to 20 billion m<sup>3</sup> by the mid-21st century<sup>1</sup>. However, concrete production poses substantial environmental challenges, including high energy consumption, significant CO<sub>2</sub> emissions, and considerable solid waste generation<sup>2,3</sup>. Among the various stages of concrete production, cement production is the most environmentally detrimental<sup>4</sup>. Numerous technologies developed to mitigate its environmental impact, including renewable energy source integration, energy efficiency enhancements, and recycled material incorporation as raw inputs<sup>5–7</sup>. Nevertheless, large quantities of solid waste and CO<sub>2</sub> emissions are still generated at concrete product manufacturing facilities, such as those producing poles, piles, and civil engineering components, and effective waste management strategies remain limited. Monteiro et al. (2023) reported that in the pole manufacturing industry, over 20,000 tons of waste concrete are produced per million tons of poles manufactured<sup>8</sup>. This waste is often disposed of in landfills without effective recycling or reuse, contributing to the exacerbation of urban waste accumulation, increase in demand for landfill space, and imposition of additional operational burdens on concrete manufacturers.

In recent years, there has been growing interest in utilizing waste concrete materials as feedstocks for CO<sub>2</sub> mineralization<sup>9–12</sup>. CO<sub>2</sub> mineralization is an emerging carbon capture and utilization technology that involves the reaction of CO<sub>2</sub> with Ca- and Mg-rich materials to form stable carbonate compounds, such as calcium carbonate (CaCO<sub>3</sub>)<sup>13</sup>. This approach addresses the problem of managing waste concrete and enables permanent CO<sub>2</sub> storage without requiring complex technologies or infrastructure<sup>14</sup>. The carbonates generated through this process have substantial industrial potential as construction aggregates and fillers in cement and concrete

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production<sup>15,16</sup>. Thus, the implementation of CO<sub>2</sub> mineralization at concrete manufacturing facilities offers both environmental and economic benefits and supports a circular economy in the construction industry.

Many CO<sub>2</sub> mineralization techniques have demonstrated high efficiency in reducing CO<sub>2</sub> in laboratory experiments through pH adjustment using chemical additives<sup>17–19</sup>. However, comprehensive techno-economic-environmental assessments have revealed significant challenges in scaling these technologies. Driver et al. (2024) emphasized that the environmental benefits of many CO<sub>2</sub> mineralization approaches, particularly in terms of net CO<sub>2</sub> emission reduction, are often overstated<sup>2</sup>. Moreover, additional environmental concerns, such as human toxicity and ecotoxicity, must be carefully evaluated before large-scale implementation. To overcome the bottlenecks associated with CO<sub>2</sub> mineralization and enhance process sustainability, research has increasingly focused on developing environmentally friendly and recyclable extraction solutions. These include the use of amino acids and chelating agents for solid dissolution, which can improve CO<sub>2</sub> mineralization efficiency while mitigating potential environmental impacts<sup>20–24</sup>.

Our research group proposed an indirect CO<sub>2</sub> capture and mineralization process that employs a recyclable solution containing chelating agent and concentrated NaHCO<sub>3</sub><sup>25,26</sup>. Under ambient extraction, the chelating agent enhanced Ca leaching from Ca-bearing materials, a subsequent mild heat step (< 100 °C) weaken the Ca-chelating agent complex and drives precipitation of CaCO<sub>3</sub>. The solution can be regenerated after being used for CO<sub>2</sub> capture, ensuring its recyclability. Although the approach has been demonstrated using plastic-incinerator bottom ash<sup>20,25</sup>, its cycle-resolved practicality and net environmental benefits have not been quantitatively established. Moreover, the dominant Ca sources differ between bottom ash and concrete waste, silicate phases in the former versus portlandite and ettringite in the latter<sup>27</sup>, which may alter extraction pathways and impurity co-migration. Therefore, before considering deployment in concrete-product manufacturing contexts, a comprehensive evaluation is required to quantify process performance over repeated reuse and to assess the associated environmental impacts.

In this study, concrete sludge cakes, which are by-products comprising cement, water, and fine aggregates that are commonly generated during concrete product manufacturing, were utilized in the recyclable chelating agent-assisted CO<sub>2</sub> mineralization process. Ten recycling experiments using the same extraction solution were conducted to investigate the behavior of the metal ions during CO<sub>2</sub> mineralization. The results of these experiments, including reductions in sludge cakes, CO<sub>2</sub> emissions, and CaCO<sub>3</sub> production, were subsequently used to assess the potential of the process to reduce environmental impacts at concrete manufacturing facilities using life cycle assessments (LCA). LCA is a tool that allows the identification and quantification of all environmental impacts associated with products, processes, and services throughout their life cycle. Finally, the most significant factors contributing to environmental impact reduction were identified, and operational recommendations were provided to optimize CO<sub>2</sub> mineralization at concrete product manufacturing facilities.

## Materials and methods

### Materials

A 40 wt% N,N-Bis(carboxymethyl)-L-glutamic acid tetrasodium salt (GLDA-Na<sub>4</sub>) solution that purchased from Chelest Corp., Japan was used for experiments. The solution had a pH of ~ 13 with a molar concentration of ~ 1.3 mol/L. NaHCO<sub>3</sub> (> 99.0% purity) was purchased from Kanto Chemical, Japan. The extraction solution was prepared by diluting GLDA-Na<sub>4</sub> and NaHCO<sub>3</sub> with Milli-Q water to the desired concentrations and adjusting the pH with nitric acid (HNO<sub>3</sub>, 60–61%, Kanto Chemical, Japan). High-purity CO<sub>2</sub> (99.995 vol%, Taiyo Nippon Sanso, Japan) was used to adjust the solution pH and compensate for the dissolved CO<sub>2</sub> that was consumed by mineralization.

The concrete sludge cake used in this study was collected from a pole and pile manufacturing plant operated by Tohoku Pole Co., Ltd., Japan. Fresh sludge cakes initially exist as a slurry but harden within 1 day under natural environmental conditions. In this study, the sludge cakes were hardened for more than one month and crushed to a particle size of < 125 µm for use in the experiments. Bulk chemistry was measured using X-ray fluorescence spectrometry (Rigaku, ZSX Primus IV). As shown in Table 1, the results revealed a high CaO content of 51.07 mass%. The mineral composition was determined using X-ray diffraction (XRD; Multiflex, Rigaku, Japan) with Cu-K $\alpha$  radiation (wavelength 1.54 Å), scanned from 5° to 60° (2 $\theta$ ) with a step size of 0.02°, at 40 kV and 15 mA. The results indicated that the hardened sludge cakes primarily comprised portlandite (Ca(OH)<sub>2</sub>, approximately 20 wt%, thereby contributing approximately 30% Ca in the sludge cake), hemicarboaluminate, mono carboaluminate ( $x$ CaO· $y$ Al<sub>2</sub>O<sub>3</sub>· $z$ CaCO<sub>3</sub>· $n$ H<sub>2</sub>O), ettringite (Ca<sub>6</sub>Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>(OH)<sub>12</sub>·26H<sub>2</sub>O), quartz (SiO<sub>2</sub>), and calcite (CaCO<sub>3</sub>) as the main crystalline minerals. In addition, the sludge cakes contained amorphous phases such as calcium silicate hydrate (C-S-H) and calcium aluminate hydrate (C-A-H), which are primary cement hydration products but cannot be conclusively identified by XRD.

| SiO <sub>2</sub> | TiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | Fe <sub>2</sub> O <sub>3</sub> | MnO  | MgO  | CaO   | Na <sub>2</sub> O | K <sub>2</sub> O | P <sub>2</sub> O <sub>5</sub> | LOI  | Total |
|------------------|------------------|--------------------------------|--------------------------------|------|------|-------|-------------------|------------------|-------------------------------|------|-------|
| 22.31            | 0.32             | 5.26                           | 3.68                           | 0.10 | 1.14 | 51.07 | 0.65              | 0.44             | 0.13                          | 14.0 | 99.1  |

**Table 1.** Chemical composition of the sludge cakes as determined using X-ray fluorescence spectrometry (mass%). LOI = Loss on Ignition, representing the percentage decrease in sample mass after heating in a muffle furnace at 950 °C for 2 h.

## Experimental methods

We previously found that a Ca/GLDA molar ratio close to 1 and high concentration of dissolved carbonate ions in the extraction solution, promotes subsequent Ca carbonation<sup>21,26</sup>. Based on preliminary experimental trials (data not shown), a solution containing 3 wt% GLDA-Na<sub>4</sub> (approximately 0.1 mol/L) and 0.8 mol/L NaHCO<sub>3</sub> was selected for this study. The pH was adjusted to 8.0 using HNO<sub>3</sub> solution before being used for the reaction with 25 g/L sludge cake.

The detailed operating conditions are illustrated in Fig. 1. In Step 1, 25 g of sludge cake powder was mixed with 1000 mL extraction solution in a plastic sample bottle, which was placed on a shaker (Reciprocal shaker NR-10, TAITEC, Japan) at 150 rpm to facilitate Ca extraction. This step was conducted at ambient temperature (approximately 20 °C for 15 min). Upon completion of the reaction, the solution and solid components in the bottle were separately collected using a 0.45 µm membrane. In Step 2, the solution collected in Step 1 was heated to 95 °C using a mantle heater stirrer with a digital temperature controller (MS-DMS-10, AS ONE, Japan) and stirred at 100 rpm to initiate Ca carbonation. After 40 min, the slurry was then filtered through a 0.45 µm membrane to separate solids from the liquid. In Step 3, CO<sub>2</sub> was bubbled through the solution until the pH stabilized at 8.0, at which point CO<sub>2</sub> was dissolved as HCO<sub>3</sub><sup>-</sup>. Through Steps 2 and 3, the extraction solution was regenerated and reused in the next cycle.

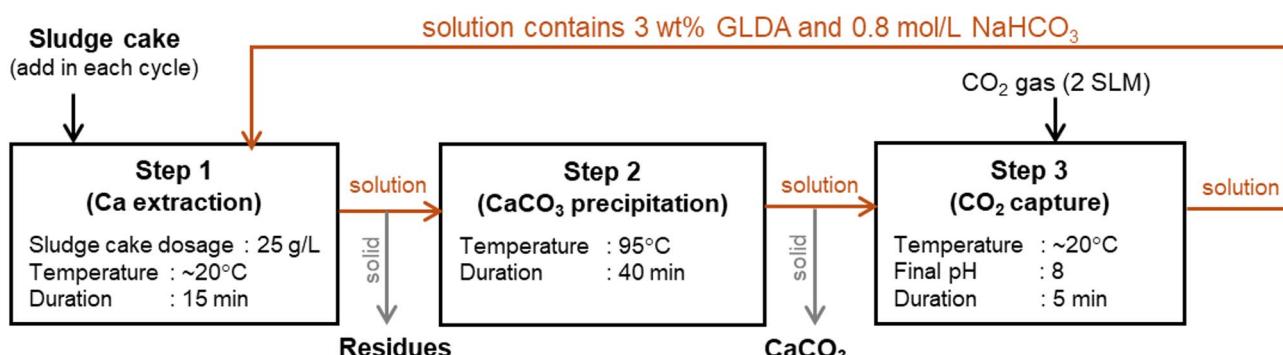
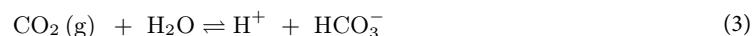
Solution samples collected after each step were analyzed for fluid chemistry by inductively coupled plasma-optical emission spectrometry (ICP-OES, Agilent 5100). Each sample was measured in triplicate to ensure reproducibility, with a relative standard deviation 3%. The elemental extraction ratio was calculated by comparing the concentration of each element in the solution to its initial content in the added sludge cake. Solid samples from Steps 1 and 2 were oven-dried at 50 °C for 24 h in an oven before further characterization using XRD, scanning electron microscopy (SEM, SU-8000, Hitachi, Ltd. Japan), and thermogravimetric (TG) analysis (Thermo plus EVO TG 8120, Rigaku Corporation, Japan). The CaCO<sub>3</sub> generated in Steps 1 and 2 was quantified as follows. In Step 1, CaCO<sub>3</sub> may be produced and mixed with the residual sludge cake. Therefore, TG analysis was employed to determine its weight ratio, which involves heating the sample from 40 to 1000 °C at a rate of 10 °C/min, during which CaCO<sub>3</sub> underwent thermal decomposition at 550–750 °C, releasing CO<sub>2</sub><sup>28</sup>. The corresponding weight loss attributed to CO<sub>2</sub> release ( $W_{CO_2}$  wt%) was used to calculate the CaCO<sub>3</sub> weight fraction. The amount of CaCO<sub>3</sub> generated in Step 1 ( $m_{CaCO_3}$ ) was determined from the change in solid mass and the difference in CaCO<sub>3</sub> weight fractions before and after Step 1.

$$m_{CaCO_3} = m_{\text{after}} \times W_{CaCO_3, \text{after}} - m_{\text{before}} \times W_{CaCO_3, \text{before}} \quad (1)$$

where  $m_{\text{before}}$  and  $m_{\text{after}}$  (g) and are the dry masses of the solid before and after Step 1, respectively.  $W_{CaCO_3, \text{before}}$  and  $W_{CaCO_3, \text{after}}$  (wt%) are the CaCO<sub>3</sub> weight fractions in the sludge cake before and after Step 1, which are calculated using Eq. 2, where  $M_{CaCO_3}$  and  $M_{CO_2}$  are the molar weights of CaCO<sub>3</sub> and CO<sub>2</sub>, respectively.

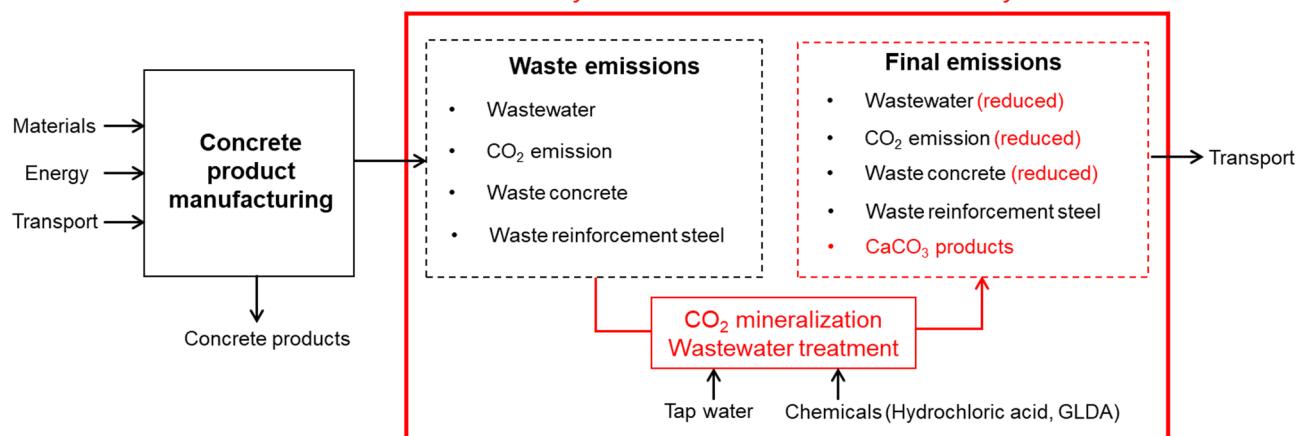
$$W_{CaCO_3} = (W_{CO_2} \times M_{CaCO_3}) / M_{CO_2} \quad (2)$$

The CaCO<sub>3</sub> produced in Step 2 was quantified based on the variation in Ca concentration between Steps 1 and 2, under the assumption that all Ca depletion resulted from the formation of CaCO<sub>3</sub>. The total amount of CO<sub>2</sub> mineralized during one cycle was determined from the cumulative amount of CaCO<sub>3</sub> formed in Steps 1 and 2. Although direct measurement of CO<sub>2</sub> capture in Step 3 was not conducted, the observed decrease in pH was positively correlated with CO<sub>2</sub> dissolution according to Eq. 1. Therefore, it was assumed that when the pH returned to its initial value of 8 after Step 3, the CO<sub>2</sub> concentration in the extraction solution had also returned to its initial level. In other words, the CO<sub>2</sub> consumed during mineralization was considered to be completely replenished in Step 3.



**Fig. 1.** Experimental conditions for recycling experiments. GLDA: *N,N*-Bis(carboxymethyl)-*L*-glutamic acid.

## System boundaries for LCA in this study



**Fig. 2.** System boundaries for LCA involving the environmental impacts associated with waste treatment processes in the pole manufacturing facility.

| Content  | Value   | Unit              |
|--|---------|-------------------|
| Pole production and waste generation           |         |                   |
| Pole production                                | 34,000  | t/y               |
| Wastewater                                     | 63,000  | m <sup>3</sup> /y |
| CO <sub>2</sub> emission                       | 1030    | t/y               |
| Waste concrete*                                | 1100    | t/y               |
| HCl solution (approximately 30%)               | 47      | t/y               |
| Electricity consumption                        | 873,000 | kWh/y             |
| CO <sub>2</sub> mineralization system design   |         |                   |
| Solution volume                                | 40      | m <sup>3</sup>    |
| Waste concrete dosage                          | 25      | g/L               |
| Number of cycles extraction solution is reused | 100     | cycles            |
| Number of operations per day                   | 4       | cycles/d          |

**Table 2.** Pole production, waste emissions, HCl utilization, electricity consumption, and design/operational conditions of the CO<sub>2</sub> mineralization system for LCA analysis. \*Waste concrete refers to the concrete sludge cake in this calculation. LCA: life cycle assessments.

### LCA goal and scope

An LCA was conducted in accordance with the ISO 14,040 and ISO 14,044 standards to evaluate the environmental impact reduction from introducing the recyclable chelating agent-assisted CO<sub>2</sub> mineralization process in concrete product manufacturing facilities. In this LCA, concrete poles were identified as the target products. Therefore, the scope encompassed all stages of pole production, including raw material extraction, transportation, processing, and final product formation<sup>29,30</sup>. However, in line with the objective of evaluating the on-site environmental impact reduction potential of the manufacturing process, the discussion in this study was limited to waste treatment processes, namely recyclable chelating agent-assisted CO<sub>2</sub> mineralization and wastewater treatment. The latter of which is currently implemented at the pole manufacturing facility (Fig. 2). The “waste concrete” in Table 2 specifically refers to the sludge cake, which is the only concrete-related solid waste generated at the site. This term was used to align with the material definitions in the database of SimaPro software (version 9.2.0, Product Ecology Consultants, 2021), which was used for LCA calculation.

The impact assessment during LCA was performed using the CML-IA (baseline) methodology. The evaluation focused on key environmental indicators, including abiotic depletion potential (fossil fuels) (ADP [fossil]), global warming potential (100a) (GWP), ozone layer depletion potential (ODP), human toxicity potential (HTP), fresh water aquatic ecotoxicity potential (FAEP), terrestrial ecotoxicity potential (TEP), acidification potential (AP), and eutrophication potential (EP)<sup>31,32</sup>.

The life cycle inventory analysis process involves obtaining relevant input–output information and data related to processes within the defined system boundaries. Information on raw material consumption and transportation for the production of each functional unit was obtained from peer-reviewed scientific literature,<sup>8</sup> and the Ecoinvent 3.0 database was used to provide data for inventory analysis. Data related to annual pole production, waste emissions, HCl utilization for wastewater treatment, and electricity consumption were provided by Tohoku Pole Co., Ltd. (Table 2; these values are approximate estimates rather than exact figures).

$\text{CO}_2$  emissions were calculated based on the consumption of heavy fuel oil A (380 kL/y) and the standard  $\text{CO}_2$  emission factor per unit of fuel oil A (2.71 kg/L according to the Ministry of the Environment, Japan), which amounts to approximately 1,030 t/y.

Data related to  $\text{CO}_2$  capture and mineralization and sludge cake weight reduction were derived from the experiments conducted in this study. The environmental impacts associated with the production and disposal of the chelating agent GLDA were excluded from this assessment owing to the lack of comprehensive data. However, this omission is expected to have a negligible influence on the overall LCA results because GLDA is a bio-based, readily biodegradable chelating agent with a minimal environmental footprint, produced through energy-efficient manufacturing processes<sup>33,34</sup>. More than 60% of GLDA can degrade within one month<sup>35</sup>. Moreover, its consumption in the  $\text{CO}_2$  mineralization process is also minimal owing to its efficient multicycle recycling system employed. In addition, waste heat is generated from an unavoidable on-site boiler process that would otherwise be vented to the atmosphere. Therefore, according to ISO 14,044 allocation principles, its potential utilization in Step 2 was considered. Consequently, the energy required for  $\text{CO}_2$  mineralization was excluded from the calculation. Finally, the transportation of the resulting  $\text{CaCO}_3$ , which has the potential to be used as an additive in pole production and other commercial applications, was not considered in this study. All inputs, outputs, and operational parameters were normalized to a functional unit, that is, a single concrete pole. Based on the information provided by Tohoku Pole Co., Ltd., approximately 34,000 tons of concrete products (poles and piles) are produced in 1 year (Table 2). Assuming that all products are poles weighing 1,200 kg each, according to Ahmadi et al. (2023), this corresponds to an average annual production of 28,333 poles.

The parameters related to the design and operational conditions of the  $\text{CO}_2$  mineralization system are listed in Table 2. For this assessment, the recycling process was assumed to use a solution with a volume of 40 m<sup>3</sup>, with the extraction solution reused for up to 100 cycles before disposal. This operational strategy was designed to match the sludge cakes generated annually, thereby minimizing the waste generation.

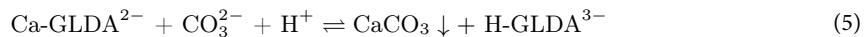
## Characterization of the $\text{CO}_2$ mineralization process

### Element extraction and precipitation

The variations in the Ca concentration over ten recycling cycles are shown in Fig. 3a. The Ca concentration fluctuated periodically, reflecting the extraction of Ca ions from the sludge cake, followed by carbonation during each cycle. After reacting with the sludge cake for 15 min in the first cycle, the Ca concentration in the extraction solution increased to 3150 mg/L. It subsequently decreased to 800 mg/L after carbonation at 95 °C for 40 min. The amount of Ca extracted in each cycle was determined from the difference in Ca concentration before and after Step 1, and the extent of Ca carbonation was quantified based on the difference before and after Step 2. The corresponding results are shown in Fig. 3b. The Ca extraction and carbonation efficiencies did not exhibit observable declining trends, suggesting the high recyclability of the extraction solution and sustainability of the process when utilizing the sludge cake. Over the 10 cycles, the average Ca extraction rate was 27%. The amount of  $\text{CaCO}_3$  generated reaching approximately 235 g/kg of sludge cake, corresponding to 104 g of  $\text{CO}_2$  mineralized per kg of sludge cake in Step 2.

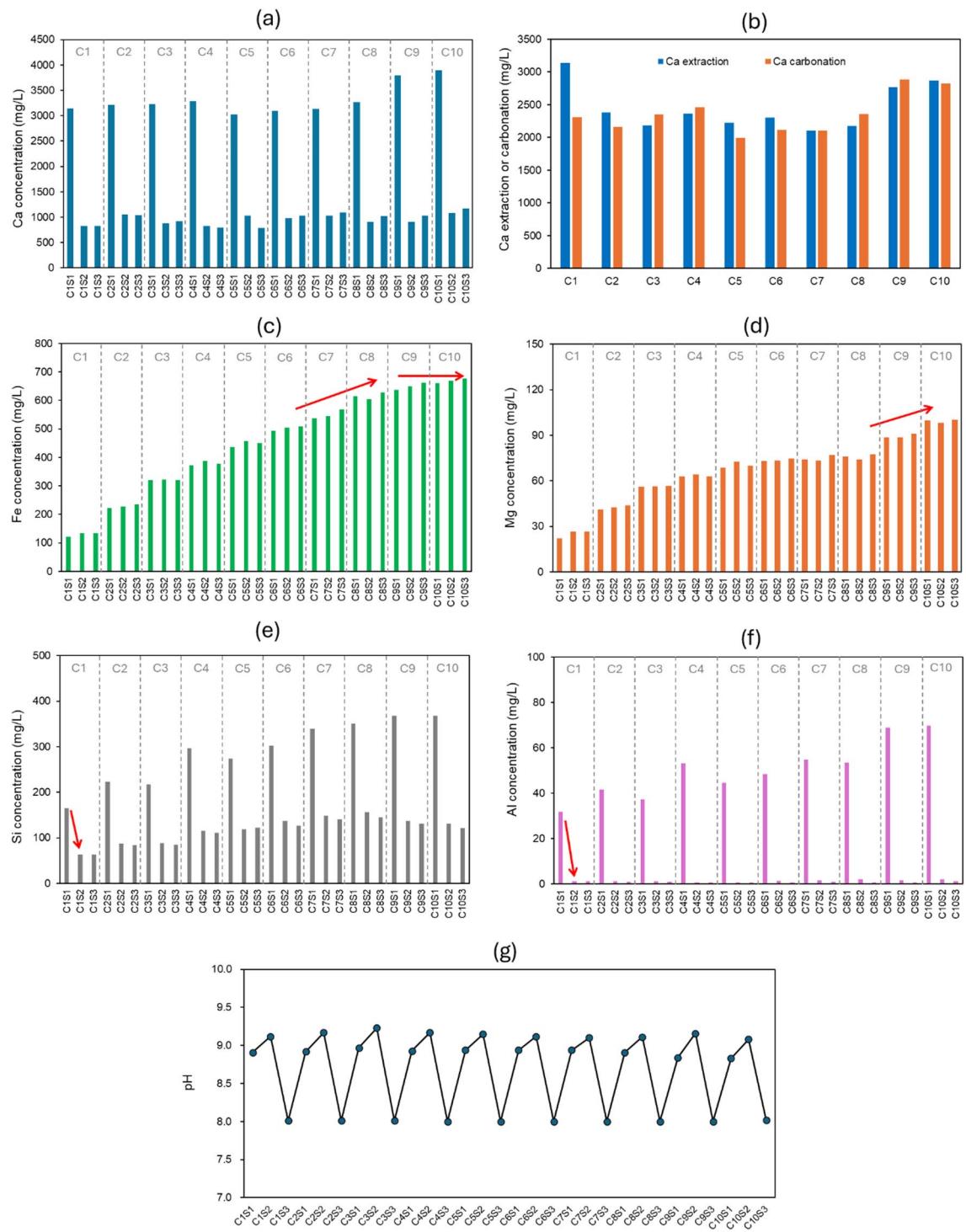
In addition to Ca, Mg and Fe ions also accumulated in the extraction solution during the recycling process (Fig. 3c and d). The concentration of Mg ions increased steadily with prolonged recycling; however, it remained significantly lower than that of Ca ions. Mg ions have lower binding affinity for GLDA compared with Ca ions, making them less competitive in the chelation process<sup>36,37</sup>. The Fe ion concentration, in contrast, tended to stabilize as the number of reaction cycles increased. This stabilization was likely owing to reduced extraction, which may be associated with the formation of low-solubility Fe-bearing minerals, such as  $\text{Fe(OH)}_3$ , which minimally influenced the overall process<sup>20</sup>. The precipitation of Fe is presumed to occur mainly in Step 1 and is estimated to be very limited (< 0.8 wt%), making it difficult to detect in the residue by XRD or SEM analyses.

Consequently, the accumulation of Mg and Fe will not significantly affect the recyclability of the extraction solution, even after multiple recycling cycles. During Step 2, a concurrent decrease in Si and Al concentrations was observed (Fig. 3e and f), suggesting co-precipitation with Ca carbonates. This behavior indicates the possible formation of amorphous silica and silica–alumina hydroxides. The periodic fluctuation of solution pH between 8.0 and 9.2 reflects the interplay of dissolution, precipitation, and  $\text{CO}_2$  adsorption processes. In Step 1, the dissolution of  $\text{Ca(OH)}_2$  and partial formation of  $\text{CaCO}_3$  consume  $\text{H}^+$ , leading to an increase in solution pH (Eqs. 4 and 5). In Step 2, further precipitation of  $\text{CaCO}_3$  continues to raise the pH (Eq. 5), whereas in Step 3,  $\text{CO}_2$  absorption results in a gradual pH decrease (Eq. 3). The consistent amplitude of pH fluctuations observed in each cycle suggests that these reactions reached a steady periodic behavior, indicating overall system stability.



### Characterization of residual sludge cakes and $\text{CaCO}_3$

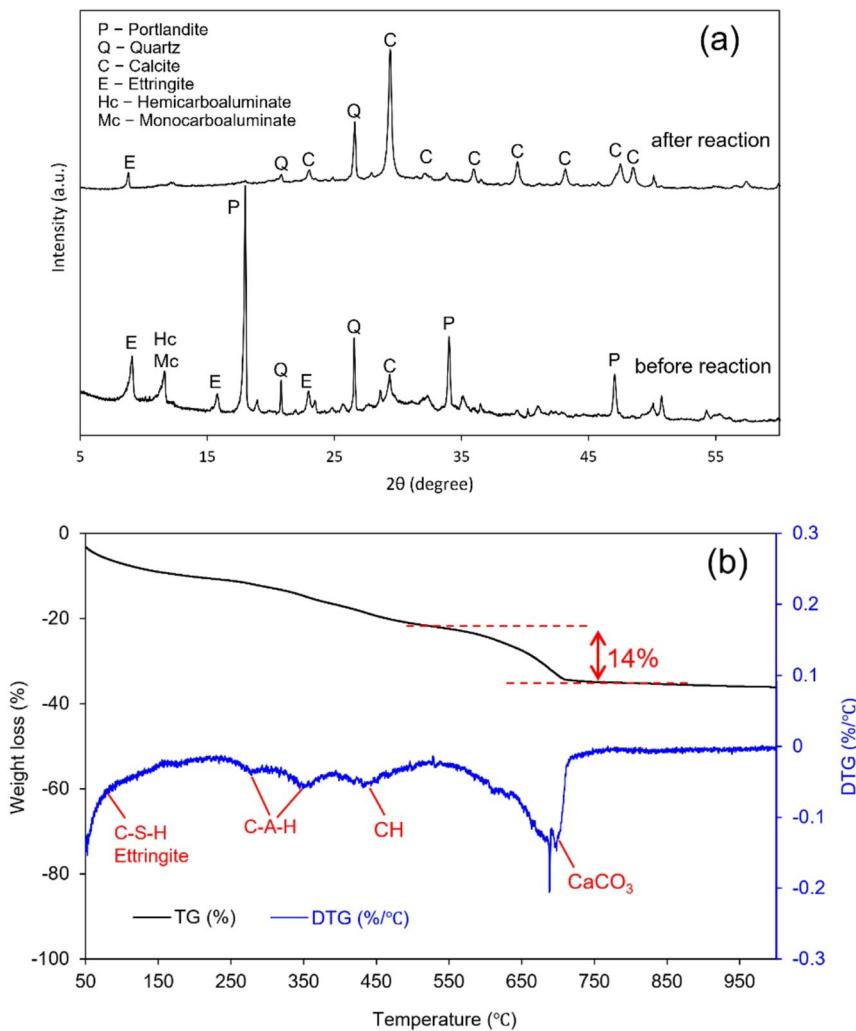
The mass of the sludge cake residue collected after Step 1 was reduced by 25%, indicating significant leaching of elements. Comparison of the XRD patterns of the initial sludge cake and the Step 1 residue revealed the complete dissolution of portlandite, hemicarboaluminate and monocarboaluminate, as well as the partial dissolution of ettringite (Fig. 4a). This suggests that all Ca-bearing crystal minerals except calcite contributed to the release of Ca ions. In contrast, the calcite peaks became more intense than those of the initial sludge cake, suggesting additional calcite formation during Step 1. This could be attributed to the increase in both Ca concentration and pH during dissolution, which promotes the reaction of  $\text{Ca}^{2+}$  with  $\text{HCO}_3^-$ . TG analysis further confirmed that the  $\text{CaCO}_3$  content increased from 6 wt% in the initial sludge cake to 24 wt% in the residue obtained after Step 1 (Fig. 4b). In addition, TG revealed weight losses below 150 °C and between 250 and 370 °C, which are attributed



**Fig. 3.** Changes in the concentrations of (a) Ca, (c) Fe, (d) Mg, (e) Si, and (f) Al and (g) pH in the extraction solution after each step throughout ten recycling experiments, and (b) Ca extraction and carbonation in each cycle calculated based on Ca concentration changes in the extraction solution ( $C_nS_m$ : Step  $m$  in the  $n^{\text{th}}$  cycle;  $C_n$ : the  $n^{\text{th}}$  cycle).

to the dehydration of C-S-H and C-A-H<sup>38</sup>. Therefore, the residual Ca was predominantly present in calcite, ettringite, and amorphous C-S-H/C-A-H phases.

The residue collected after Step 1 was predominantly composed of quartz and calcite, suggesting its potential for reuse either as an additive in concrete production<sup>39</sup> or as a feedstock for  $\text{CO}_2$  mineralization to synthesize high purity  $\text{CaCO}_3$ . Moreover, the total  $\text{CO}_2$  mineralization in Steps 1 and 2 of each reaction cycle was approximately 156 g  $\text{CO}_2$  per kg of sludge cake.



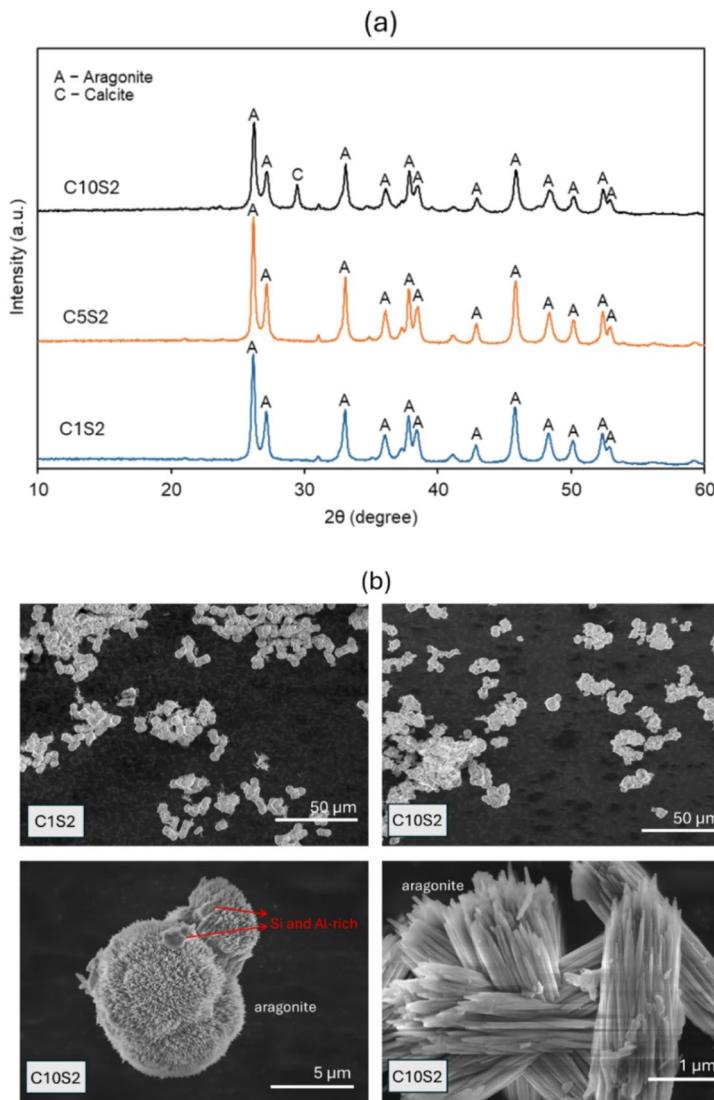
**Fig. 4.** (a) XRD pattern of the sludge cake before and after reaction with the GLDA solution and (b) TG measurement of the sludge cake after reaction with the GLDA solution. XRD: X-ray diffraction, GLDA: *N,N*-Bis(carboxymethyl)-*L*-glutamic acid, TG: thermogravimetric analysis.

The  $\text{CaCO}_3$  generated in Step 2 was characterized using XRD, which revealed that it predominantly consisted of aragonite with a minor presence of calcite (Fig. 5a). The formation of aragonite instead of calcite in the presence of GLDA at elevated temperatures is consistent with the findings of our previous study<sup>25</sup>. SEM analysis of the synthesized  $\text{CaCO}_3$ , as shown in Fig. 5b, indicated that the particle size remained consistent across the ten recycling experiments. The aragonite aggregates measured 10–20  $\mu\text{m}$ , with individual aragonite rods exhibiting widths of < 100 nm.

During the formation of  $\text{CaCO}_3$ , decreases in the concentrations of Si and Al were also observed (Fig. 3e and f), indicating the possible co-precipitation of Si- and Al-containing species with  $\text{CaCO}_3$ . SEM analyses revealed that Si and Al appeared to precipitate as amorphous phases on the surfaces of the  $\text{CaCO}_3$  (Fig. 5b). As the pH increased during  $\text{CaCO}_3$  formation, Si was likely precipitated as amorphous  $\text{SiO}_2$  and Al as  $\text{Al(OH)}_3$ . Based on the variations in Si and Al concentrations in the solution during Step 2 (Fig. 3), the corresponding amounts of  $\text{SiO}_2$  and  $\text{Al(OH)}_3$  formed were estimated. Consequently, the purity of the resulting  $\text{CaCO}_3$  stabilized at approximately 92% throughout the ten cycles. The  $\text{CaCO}_3$  produced in this study is suitable for lower-value applications such as building materials, cement, and soil amendments; however, further purification would be required to meet the specifications for high-grade filler applications<sup>40</sup>.

### LCA analysis

Recycling experiments showed that Fe and Mg had minimal effects on Ca extraction and carbonation, while the concentrations of Si and Al returned to their initial levels after each cycle, that neither impurity accumulation nor progressive chelate deactivation occurred within the tested range. Therefore, in the LCA model, the GLDA solution was assumed to be reused up to 100 cycles to represent a long-term operational scenario. Moreover, as the environmental impacts of GLDA production and treatment were excluded from the system boundary, this assumption has minor influence on the LCA results.



**Fig. 5.** (a) XRD patterns and (b) SEM images of the CaCO<sub>3</sub> formed at Step 2 of the 1st, 5th, and 10th cycles (C<sub>n</sub>Sm: Step *m* in the *n*th cycle). XRD: X-ray diffraction, SEM: scanning electron microscopy.

| Life cycle inventory      | Current process | With CO <sub>2</sub> mineralization | Unit           |
|---------------------------|-----------------|-------------------------------------|----------------|
| Input                     |                 |                                     |                |
| HCl (30%)                 | 1.659           | 1.659                               | kg             |
| Tap water                 | 0.107           | 0.111                               | m <sup>3</sup> |
| Output                    |                 |                                     |                |
| Pole                      | 1               | 1                                   | item           |
| Waste concrete            | 38.824          | 29.118                              | kg             |
| Waste steel reinforcement | 114.300         | 114.300                             | g              |
| Wastewater                | 2.223           | 2.227                               | m <sup>3</sup> |
| CO <sub>2</sub> emission  | 36.353          | 30.353                              | kg             |

**Table 3.** Waste treatment input–output data for the pole manufacturing facility used for LCA analysis (Functional unit: one pole weighing 1200 kg).

The input–output data related to waste treatment in the conventional and modified processes incorporating CO<sub>2</sub> mineralization are summarized in Table 3. The introduction of the GLDA solution resulted in a slight increase (approximately 4%) in tap water consumption and a corresponding increase in wastewater discharge. The experimental results suggest that the CO<sub>2</sub> mineralization process reduced the overall mass of the sludge

cake emissions by 25%. For each concrete pole produced, the sludge cake emissions decreased from 38.824 to 29.118 kg. Additionally, 6.0 kg of CO<sub>2</sub> was successfully mineralized, leading to a reduction in CO<sub>2</sub> emissions from 36.353 to 30.353 kg per pole. Additional input–output data related to materials, energy consumption, transportation, and waste emissions during pole production were obtained from Monteiro et al. (2023)<sup>8</sup>.

These data were used to evaluate the environmental impacts of the pole manufacturing facilities, with the values for each category summarized in Table 4 and reductions in each category presented in Fig. 6a. The implementation of the CO<sub>2</sub> mineralization process resulted in a decrease across all impact categories, including GWP, with reductions ranging from 1.2% to 10.0%. The fractions of each inventory component are shown in Fig. 6b,c. Regardless of the implementation of CO<sub>2</sub> mineralization, CO<sub>2</sub> emissions remained the primary contributor to the environmental impact of GWP at the concrete manufacturing facility. However, for the other environmental impact categories, the main contributors were wastewater and HCl used for wastewater treatment, followed by waste concrete, tap water, and waste steel reinforcement.

For the GWP, the application of CO<sub>2</sub> capture and mineralization processes is expected to achieve an approximate 16.1% reduction, which aligns with the CO<sub>2</sub> utilization efficiency observed in the experiments. Consequently, producing a single concrete pole (weighing 1200 kg) results in a reduction of 6.08 kg CO<sub>2</sub> equivalent, corresponding to a decrease of 156.6 kg CO<sub>2</sub> equivalent per ton of waste concrete. This reduction is relatively higher than GWP reduce reported for alternative CO<sub>2</sub> mineralization technologies, which are typically less than 4.3 kg CO<sub>2</sub> equivalent per ton of solid wastes<sup>41–43</sup>. This indicates that the recyclable chelating agent-assisted CO<sub>2</sub> mineralization process exhibits competitive environmental performance due to its high CO<sub>2</sub> conversion efficiency and reagent reusability. The ADP (fossil) decreased from 18.659 to 16.796 MJ per pole, representing a 10.0% reduction, which was the most significant decrease among all environmental impact categories. ADP (fossil) is associated with sulfuric acid production, and this reduction is primarily attributed to the decrease in sludge cake emissions, which accounted for 34.2% of the total ADP (fossil) impact (Fig. 6b). The other environmental impact categories were reduced by 1.2%–6.2%.

The LCA conducted in this study demonstrated that the implementation of recyclable chelating agent-assisted CO<sub>2</sub> mineralization can significantly reduce GWP and substantially mitigate the various other environmental impacts of concrete production sites. The recycling of treated solid waste should be considered to maximize the environmental benefits of CO<sub>2</sub> mineralization. In this study, the treated sludge cake was found to contain calcite and quartz, with very low concentrations of heavy metals that could pose environmental concerns (not shown), demonstrating its potential for safe disposal as soil or reuse as fine aggregates in concrete manufacturing<sup>44,45</sup>. If such utilization is achieved, the environmental impact can be further reduced in all categories. The ADP (fossil) of pole manufacturing facilities is expected to be reduced by 44%.

Subsequently, this study verified the technical and environmental feasibility of implementing a recyclable chelating agent-assisted CO<sub>2</sub> mineralization process at concrete manufacturing facilities. This process exhibits strong sustainability because a stable amount of CO<sub>2</sub> is mineralized into aragonite. Among the three polymorphs of CaCO<sub>3</sub> (calcite, aragonite, and vaterite), aragonite is considered the most suitable filler material for the paper and plastic industries and is a valuable additive in biomedical applications<sup>46</sup>. These results highlight the potential economic benefits of the proposed process.

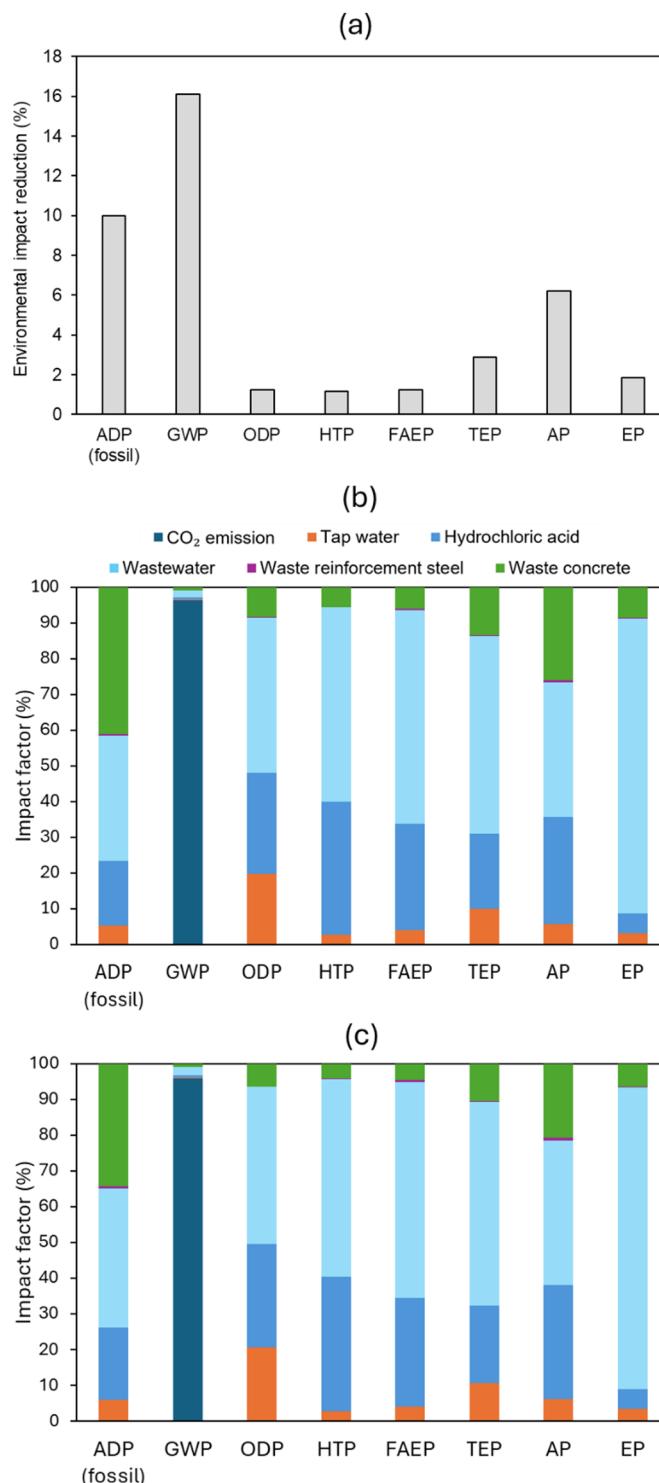
However, the LCA presented in this study remains preliminary, as it does not account for the environmental impacts associated with the production and treatment of GLDA, nor does it consider the energy requirements for the triggering of CO<sub>2</sub> mineralization. Future studies should address these aspects by incorporating specific technical parameters and economic considerations to enable a more comprehensive and accurate assessment. Moreover, this study suggests the reuse of sludge cake residues as fine aggregates in concrete production. However, further evaluation of this reuse potential requires systematic studies, including leaching and toxicity tests as well as mechanical performance assessments, which will be conducted in future work.

## Conclusion

To address the environmental limitations of conventional CO<sub>2</sub> mineralization routes, our research group developed a more sustainable CO<sub>2</sub> mineralization process that utilizes recyclable chelating agents and waste-derived calcium sources. This study, for the first time, demonstrates both the technical feasibility and the

| Impact category                                 | Impact          |                                     | Unit                     |
|---|-----------------|-------------------------------------|--------------------------|
|   | Current process | With CO <sub>2</sub> mineralization |                          |
| Abiotic depletion (fossil fuels) (ADP (fossil)) | 18.659          | 16.796                              | MJ                       |
| Global warming (GWP)                            | 37.813          | 31.726                              | kg CO <sub>2</sub> eq    |
| Ozone layer depletion (ODP)                     | 0.088           | 0.087                               | mg CFC-11 eq*            |
| Human toxicity (HTP)                            | 2.594           | 2.563                               | kg 1,4-DB eq**           |
| Fresh water aquatic ecotoxicity (FAEP)          | 1.286           | 1.270                               | kg 1,4-DB eq             |
| Terrestrial ecotoxicity (TEP)                   | 9.496           | 9.221                               | g 1,4-DB eq              |
| Acidification (AP)                              | 7.214           | 6.768                               | g SO <sub>2</sub> eq     |
| Eutrophication (EP)                             | 5.206           | 5.110                               | g PO <sub>4</sub> --- eq |

**Table 4.** LCA results for the pole manufacturing facility with and without chelating agent-assisted CO<sub>2</sub> mineralization implementation (Functional unit: one pole weighing 1200 kg). \*CFC-11: trichlorofluoromethane (CCl<sub>3</sub>F). \*\*1,4-DB: 1,4-dichlorobenzene (C<sub>6</sub>H<sub>4</sub>Cl<sub>2</sub>).



**Fig. 6.** (a) Reductions in the environmental impact categories for the concrete pole manufacturing facility following CO<sub>2</sub> mineralization implementation and fractions of each inventory component (b) without and (c) with CO<sub>2</sub> mineralization. Abiotic depletion (fossil fuels): ADP[fossil], GWP: Global warming, ODP: Ozone layer depletion, HTP: Human toxicity, FAEP: Fresh water aquatic ecotoxicity, TEP: Terrestrial ecotoxicity, AP: Acidification, EP: Eutrophication.

environmental advantages of implementing this process at a concrete product manufacturing facility, based on multi-cycle experiments combined with a comprehensive LCA assessment. The use of sludge cake from concrete pole manufacturing renders this process highly sustainable. Ten recycling uses of the extraction solution demonstrated that the process reduced the sludge cake by approximately 25% and mineralized 156 g of CO<sub>2</sub> per

kg of sludge cake into  $\text{CaCO}_3$ . The LCA results further indicated that the implementation of this process could reduce the GWP at a concrete pole manufacturing facility by 16.1% and decrease other environmental impact categories by 1.2–10.0%. These findings highlight the potential of the  $\text{CO}_2$  mineralization process to effectively mitigate the environmental impact of the concrete manufacturing industry.

## Data availability

Data will be made available upon request from the corresponding author.

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## Author contributions

Conceptualization: J.W., N.T., and N.W.; Methodology, Data curation and Formal analysis: R.M. and V.N.A.; Investigation: J.W., R.M., and N.W.; Visualization: J.W. and R.M.; Funding acquisition: J.W. and N.W.; Project administration: J.W., K.S., M.S., and N.W.; Supervision: J.W., K.S., M.S., N.T., and N.W.; Writing-original draft: J.W.; Writing-review and editing: R.M., V.N.A., K.S., M.S., N.T., and N.W.

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## Declarations

### Competing interests

Jiajie Wang, Noriaki Watanabe, Noriyoshi Tsuchiya have patent #JP7345791 licensed to Tohoku University.

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