

CO₂ UPTAKE OF CONCRETE RECYCLED FINES CARBONATED USING SOLID-AIR AND AQUEOUS CARBONATION

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ABSTRACT

Mineral carbonation of cement based materials has been investigated as an effective approach to achieve carbon neutrality in the concrete industry. Considering the high levels of concrete consumption, which is still an essential material for shaping the world and supporting societies, recycling of concrete waste is also an important topic to be considered. This experimental research evaluated the carbon uptake potential of real concrete recycled fines, obtained from demolished concrete recycled aggregate production – Recycled Concrete Aggregate Fines (RCAF) – and from dewatered wash-out of returned fresh concrete slurry – Concrete Slurry Waste (CSW). Solid-air and aqueous carbonation methods effectiveness were compared using X-ray diffraction (XRD) and thermogravimetric analysis (TGA). It was confirmed that chemical composition of the material relies greatly on the recycling processes that generate the waste. In addition, XRD and TGA results revealed that, for powdered materials, water content is a factor that significantly affects the carbonation reaction, even more than the reaction time. Also, compared to RCAF, CSW showed a high potential for carbon uptake, reaching more than 20% at optimum conditions – solid-air carbonation with high content of water (w/s>0.50) and aqueous carbonation method.

Keywords: *Solid-air carbonation, aqueous carbonation, CO₂ uptake, concrete recycling.*

1. INTRODUCTION

Currently, there are large numbers of initiatives to achieve carbon neutrality in the construction sector. In the cement industry, combustion and calcination are two inevitable CO₂ emissions processes. Until the past years, the focus was only in reducing these emissions, especially in the production of clinker, and manufacture and shipment of cement. But now, CO₂ capture and utilization have been widely investigated as an efficient approach for carbon neutral in concrete industry.

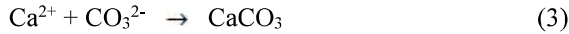
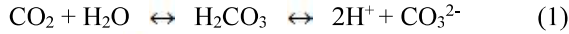
1.1 Concrete recycling

Considering the importance of concrete for society and the aging of structures, concrete waste production is not only difficult to be reduced, but also is likely to increase in the next years. For example, in Japan when it is possible, to demolish and reconstruct a new structure has been considered as a better option than repairing and strengthening a deteriorated existing one. Additionally, natural disasters are common and inevitable, and these also generate a lot of demolished and crushed concrete. Another type of concrete waste is that comes from returned fresh concrete. To prevent cold joint formation and to keep productivity on construction sites, it is affordable to order a little more amount than what is necessary, and then, to pay for the excess of concrete which is returned to the batching plants to be discarded. Appropriate recycling of these wastes produces two types of concrete recycled fines: Recycled Concrete Aggregate Fines (RCAF), which is a by-product of the recycled concrete aggregate production; and Concrete Slurry Waste (CSW), which is a solid waste from liquid wash-out of returned fresh concrete.

1.2 Carbonation and CO₂ uptake

Carbonation is a natural chemical reaction that occurs between cement-based materials, mostly calcium hydroxide and the carbon dioxide from the air. It can be divided in 3 steps: Carbon dioxide diffusion/dissolution (Equation 1); Calcium ion extraction (Equation 2); Calcium carbonate precipitation (Equation 3). It is widely recognized that carbonation in concrete is affected by concrete properties and the environmental conditions, and this phenomenon is also identified as neutralization, because it reduces concrete's alkalinity, breaking down the passivation layer surrounding the reinforcing steel and leaving it vulnerable to corrosion. Although the negative impact for the steel protection, in this reaction CO₂ is absorbed from the environment, producing chemically stable carbonates. So, cement-based materials spontaneously uptake carbon dioxide and

this property has been highlighted as a new technological approach to achieve carbon neutral in the cement and concrete industry.



2. OBJECTIVES

As concrete recycled fines contain basically concrete materials (cement and fine particles from aggregates) and usually are not contaminated by harmful substances, there is a high potential for these materials to be explored as calcium sources and alkaline ingredients for carbonation, and furthermore, to be reused in concrete production. Therefore, the purpose of this research is to evaluate the first part of this double potential of recycled concrete fines by studying solid-air and aqueous carbonation methods as a process to standardize RCAF and CSW and calculating the carbon uptake of these powders.

3. MATERIALS AND METHODS

This experimental research evaluated the carbon uptake potential of real concrete recycled fines, obtained from concrete recycling plants situated in Tokyo, Japan. RCAF was from demolished concrete recycled aggregate production; and CSW from dewatered (filter press process) wash-out of returned fresh concrete slurry. X-ray diffraction (XRD) and thermogravimetric analyses (TGA) were used to analyze the chemical composition and understand which factors would have more impact in the carbonation reaction of powdered materials.

3.1 Sample preparation

Samples of RCAF and CSW were ground manually and sieved using 150 μm sieve. To remove free water, both samples were dried at 60°C for 1 day.

3.2 Solid-air carbonation

Two important factors were evaluated to optimize carbonation process: water content of the samples and the reaction time. For solid-air carbonation, water-solid ratio (w/s) of the samples was varied from 0.00, 0.25, 0.50, 0.75 to 1.00, by adding distilled water to the dried samples (Fig. 1).

The prepared samples were placed at an accelerated carbonation chamber 5% CO_2 concentration, 60% of relative humidity (RH), 20°C for 3 and 7 days. Samples were manually ground and XRD and TGA were performed before and after the carbonation of the powders.

3.3 Aqueous carbonation

In the aqueous carbonation method, for each powder, a solution 25g/l concentration was prepared using potable water with CO_2 dissolved in it (Fig. 2). The solution was stirred for 120 minutes at atmospheric pressure, filtered using paper filter and the solid material

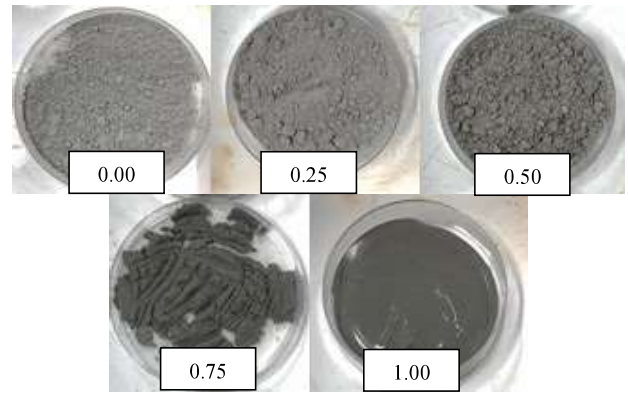


Fig. 1. Samples with different w/s, before solid-air carbonation.



Fig. 2. Solution for aqueous carbonation.

was dried at 40°C. XRD and TGA were performed before and after the carbonation of the samples.

4. RESULTS AND DISCUSSION

4.1 Carbonation methods comparison

Mineral composition of RCAF and CSW are shown in Figure 3a and Figure 3b. For the samples analyzed, before carbonation, RCAF showed high intensity peak for SiO_2 and CaCO_3 . This indicates that fines mostly came from aggregates crushing, or that the original structure had already suffered high level of natural carbonation. Differently, CSW showed a high peak for $\text{Ca}(\text{OH})_2$ and very low peak for CaCO_3 and SiO_2 , indicating a high potential for carbonation. After carbonation, there was no change in intensity peaks for RCAF, indicating that it did not have a significant effect, both for solid-air and aqueous methods. But for CSW, there was a change in the peak from $\text{Ca}(\text{OH})_2$ to CaCO_3 . Especially for aqueous carbonation the intensity peak for calcite became stronger, making clear the effectiveness of this carbonation method for powdered materials.

The TGA results agreed with the XRD results. Calcium carbonate content (%) was calculated based on initial mass weight of each sample. High content of CaCO_3 was identified quantitatively for RCAF before carbonation (26.93%). And even after 7 days in accelerated carbonation chamber or aqueous carbonation, there was an increase of only 2–4 % to in CaCO_3 content

(Fig. 4a). Differently, for CSW it was possible to verify effective carbonation of some samples, which showed a complete change of Ca(OH)_2 to CaCO_3 ($w/s > 0.50$) and as the water-solid ratio increased, solid-air carbonation progressed more than in lower water content samples (Fig. 4b). Another point is that in the first 3 days, carbonation occurred intensively, but even after letting the sample in supply of CO_2 for 7 days, carbonation did not continue for samples with $w/s = 0.00$ and 0.25 , even there was Ca(OH)_2 available for carbonation. This indicates that to achieve an optimum condition for solid-air carbonation, w/s ratio is more important than reaction time.

This relationship between water content available for CO_2 diffusion and reaction time probably is related to the increased contact surface area between CO_2 and the fines, which is greater than in the hardened concrete. In powdered materials, it is not required too much time for CO_2 diffusion to occur, and carbonation does not move progressively inside as occur in solid cementitious matrix (Fig. 5). But in fine powder, the increased contact area also undermines adhesion of H_2O particles to the fines. That is probably the reason why accelerated carbonation at 60% RH was not sufficient for w/s ratios under 0.50 .

4.2 CO_2 uptake

To evaluate the amount of CO_2 absorbed by the material and captured in the solid phase as CaCO_3 , CO_2 uptake was calculated using the Equations (4) and (5):

$$\text{CO}_2 \text{ uptake} = \frac{\text{CO}_2(\text{carbonated}) - \text{CO}_2(\text{initial})}{1 - \text{CO}_2(\text{carbonated})} [\%] \quad (4)$$

$$\text{CO}_2 = \frac{\Delta m \text{CO}_2}{m_{\text{initial}}} [\%] \quad (5)$$

where:

$\Delta m \text{CO}_2$ = CO_2 mass as CaCO_3 (or mass loss due to CaCO_3 decomposition in TGA); and m_{initial} = initial mass sample in TGA.

The calculated CO_2 uptake of the adopted methods are presented in Table 1.

Table 1. CO_2 uptake (%) calculated

Carbonation Method	CO_2 uptake (%)	
	RCAF	CSW
Air-Solid w/s 0.00	1.47	7.23
Air-Solid w/s 0.25	1.02	12.54
Air-Solid w/s 0.50	1.49	20.27
Air-Solid w/s 0.75	2.01	20.96
Air-Solid w/s 1.00	1.09	21.74
Aqueous	1.36	22.14

The maximum value of CSW CO_2 uptake was more than 22.14%, which means that 1 kilogram of the CSW analyzed could absorb than 221.4g of CO_2 in optimum conditions, while RCAF analyzed achieved only 2.01% of CO_2 uptake.

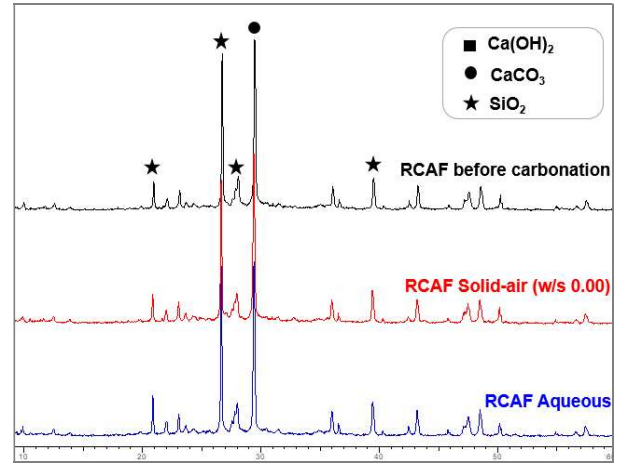


Fig. 3a. XRD pattern of RCAF.

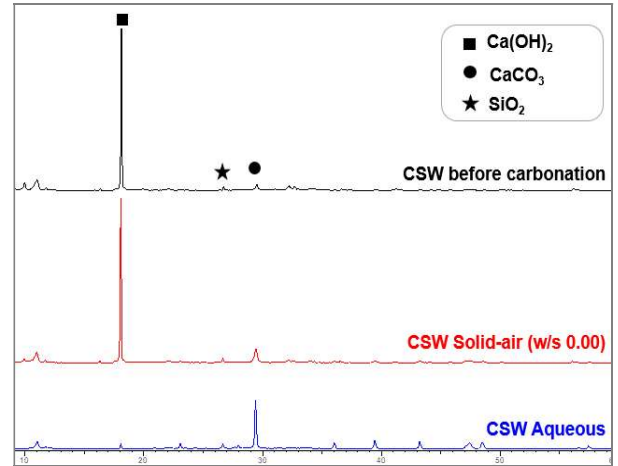


Fig. 3b. XRD pattern of CSW.

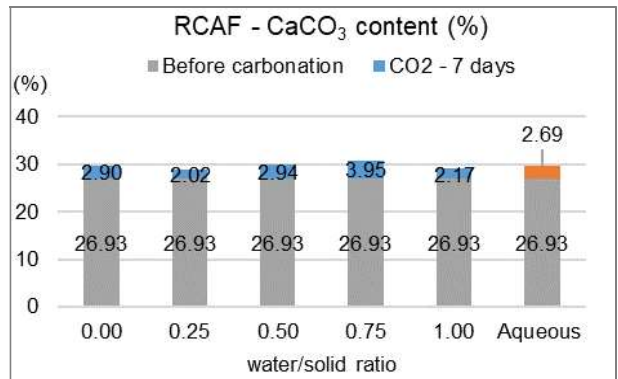


Fig. 4a. CaCO_3 content (%) of RCAF.

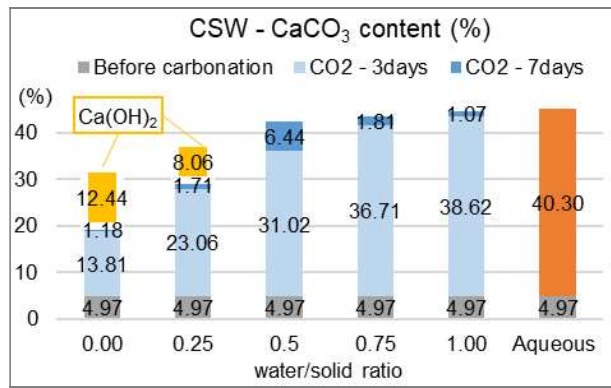


Fig. 4b. CaCO₃ content (%) of CSW.

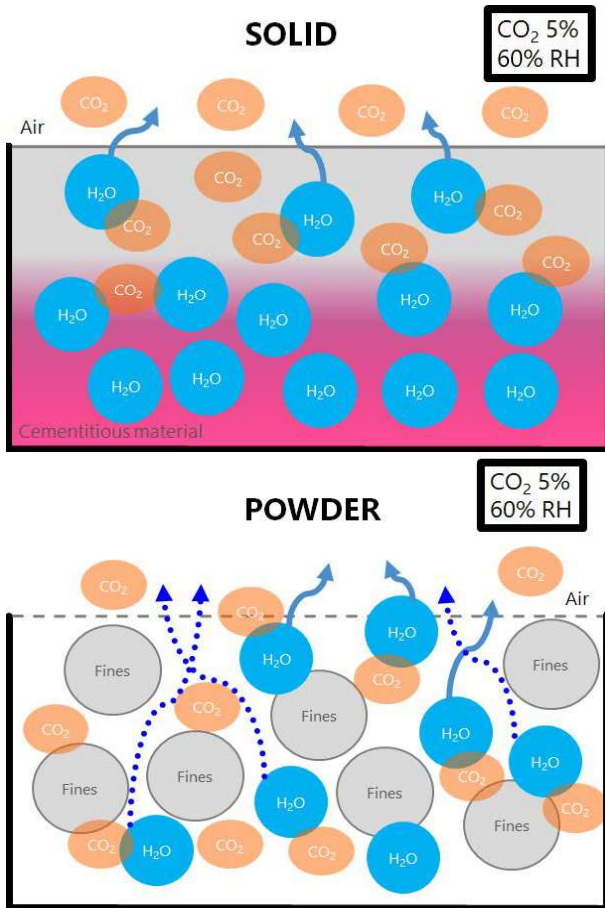


Fig. 5. Comparison between H₂O adhesion at accelerated carbonation chamber for solid and powder cementitious materials.

5. CONCLUSIONS

In this research, it was confirmed that chemical composition of the concrete recycled fines (RCAF, CSW) relies greatly on the concrete recycling processes that generate the waste, so to expand the application of these materials, especially for concrete production, it is necessary an adequate management of the material, and carbonation showed to be a successful approach for it.

About accelerated carbonation of powdered materials, water content revealed to be a factor that significantly affects the reaction, even more than the reaction time. About the carbonation method, aqueous

carbonation had a high efficiency to promote CO₂ absorption.

Comparing the materials from the batches analyzed, it was verified that RCAF can be a material which has already been carbonated naturally and, thus, did not showed good performance for carbon uptake in the accelerated carbonation environment (maximum of 2.01%). A methodology to quantify the carbon uptake of natural carbonation that occurred in a concrete structure during its service life is another topic that needs to be investigated. Also, samples of RCAF from different batches will be tested in further projects to verify the potential of RCAF for carbon uptake. Differently from RCAF, CSW indicated a high potential as a carbon uptake material, reaching more than 20% for solid-air carbonation with $w/s > 0.50$, and for aqueous carbonation method.

For future research, the effect of carbonation on RCAF and CSW will be evaluated using the material for concrete production.

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NOMENCLATURE

- RCAF : Recycled Concrete Aggregate Fines
- CSW : Concrete Slurry Waste
- w/s : water-solid ratio

PHOTOS AND INFORMATION



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