# Improved storage and handling of fine recycled concrete aggregates by accelerated carbonation: towards a quantification method

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Abstract The fines fraction generated by crushing of concrete waste for production of recycled concrete aggregates amounts to 40 - 60 mass-%. Technically, fine recycled concrete aggregates (0 - 2 mm, fRCA) may be employed as a sand substitute in new concrete products. This is, however, hampered by the materials tendency for bridging/caking during storage in open stockpiles or silos. Here we investigated whether the reactivity of fRCA can be decreased and, hence, its storage and handling be improved by accelerated carbonation. Carbonation experiments were conducted in a pilot-scale rotating drum reactor (void volume V0 ~ 75 L) at around unity CO2 partial pressure (re-dosing setpoints - 50 hPa and + 50 hPa in-vessel differential pressure) over reaction times of one hour. Re-dosing cycles and thermogravimetric analysis served to quantify the CO<sub>2</sub> uptake by fRCA. Penetration resistance and sieve passage against control samples served to quantify the effect of carbonation on the bridging tendency of fRCA. Compared to control samples, carbonated fine RCA formed more brittle crusts upon exposure to ambient conditions. Thus bridging/caking of fRCA may be prevented by accelerated carbonation prior to storage. Consequences of the use of carbonated fRCA as a sand substitute in concrete are subject of ongoing investigation while test methods to assess the effects on handling and storage properties are under consideration.

Keywords: construction and demolition waste, recycled concrete aggregates, concrete fines, accelerated carbonation

# 1. Introduction

Concrete is amongst the most used construction products worldwide and one of the most prominent waste streams (Pasquier et al. 2018). The fines fraction generated by crushing of concrete waste for production of recycled concrete aggregates amounts to 40 - 60 mass-% (Quattrone et al. 2014). Technically, fine recycled concrete aggregates (0 - 2 mm, fRCA) may be employed as a sand substitute in new concrete products and thus be kept in the material cycle. Due to its chemical properties and crushing, fRCA has a pronounced reactivity towards CO<sub>2</sub> and is therefore

of interest for carbon capture. In addition to alleviating the product carbon footprint, carbonation could also improve the physical properties of fRCA (in particular a reduction in porosity) and concrete produced therewith (Jang et al. 2016) (Zhang et al. 2015) giving its recycling a double benefit.

Utilization possibilities for fRCA are, however, hampered by the materials tendency for bridging/caking during storage in open stockpiles or silos by associated problems like blockages, occupational safety risks and limitations to material handling (Figure 1).



Figure 1. fRCA aggregate

During storage, the material may be exposed to the ambient  $CO_2$  concentration for weeks. Since storage areas are not always covered, ambient moisture may pave the way for carbonation of RCA. This feeds back into  $CO_2$  uptake since hydration promotes supply hydroxide phases in fRCA enhancing crust formation. Carbonation potential and reactivity are related, among other factors, to the age of the source material since fRCA can be generated from dismantled buildings but also from relatively fresh concrete waste, e.g. from faulty batches or overproduction. The reactivity of fRCA stockpiles may be technically mitigated by accelerated carbonation since, therewith, the

process is speeded up under defined conditions while the material is kept in motion to prevent bridging of particles. Thereby, improved handling properties of fRCA may be expected.

We are not aware of any standardized method for determining the crust formation of fRCA. Accordingly, the aim of this study was to develop a method to determine and quantify the effects of accelerated carbonation on the storage and processing behavior of fRCA.

For this purpose, samples of freshly crushed fRCA exposed to pure  $CO_2$  in a rotating drum reactor. These samples were examined at different time intervals by sieving and penetrometer tests for surface crust formation and particle size distribution.

## 2. Methods

#### 2.1. Carbonation experiments

The accelerated carbonation of the fRCA (0 - 2 mm) was carried out in an axially rotating drum reactor (V = 75 L, Figure 1) in batch operation at ambient temperature. 14.87 kg of the material (dry matter of 94.5 %) were moistened to 11 % water content and exposed to a pure CO<sub>2</sub> atmosphere over one hour at a rotation rate of 5 rpm. Before the start of the experiment, the reactor was purged with CO<sub>2</sub>, followed by CO<sub>2</sub> dosing via a solenoid valve between pressure setpoints of 950 and 1050 hPa. A perforated mixing cage was used to optimize material mixing.

Under these conditions,  $CO_2$  uptake and redosing lead to a sawtooth characteristic of the pressure vs. time curve. We recorded and integrated the in-vessel pressure to quantify the  $CO_2$  uptake. Two batches of the fRCA were carbonated in the rotating drum reactor. One additional batch was subjected to the same moisture condition yet without  $CO_2$  addition and served as a blank.



**Figure 2.** Experimental setup for accelerated carbonation in the rotating drum reactor. (1)  $CO_2$  gas cylinder, (2) solenoid valve, (3) liquid reservoir, (4,5) rotary coupling, (6) circuit, (7) humidification lance, (8) gas-tight rotating drum reactor, (9) pressure switch and sensor with data logger.

In addition to pressure-based CO<sub>2</sub> uptake quantification, native and carbonated fRCA samples were dried, ground

and subjected to TG analysis (thermogravimetric analyser NETZSCH TG 209 F1 Iris, NETZSCH Proteus Analysis Version 8.0.3) to investigate changes in the carbonate content.

#### 2.2. Testing of crust formation

Carbonation-induced changes in the material properties were tested using two methods.

1. Penetration resistance

To evaluate carbonation effects on crust formation, 300 g of native and carbonated fRCA were filled into PVC rings (inner diameter 12 cm) and compacted with a plunger. The specimens were then exposed to ambient conditions for 1, 2, 3, 7, and 14 d. At each sampling time the penetration resistance of the samples a was recorded in two- to threefold replication using a penetrometer.

#### 2. Sieve passage

To test the stability of the agglomerates and crusts formed in the carbonated and control samples, the cylindrical samples used in the penetrometer tests were carefully shuttered onto the coarsest sieve of a sieve tower (mesh sizes 4, 2.8, 2, 1, 0.5, 0.25, 0.125) such that damage to observable surface crusts was minimized. The material was then sieved for 20 s at the lowest intensity (stage 1, FRITSCH analysette 03502).

In addition, grading curves of the native and carbonated material were prepared according to DIN EN 12620 to determine a potential grain size shift due to carbonation of the fRCA.

#### 3. Results and Discussion

#### 3.1 Carbonation

Based on the pressure course, the carbonation tests resulted in a  $CO_2$  uptake of 2.6 g  $CO_2$  and 2.5 g  $CO_2/100$  g dry matter in the consecutive batch treatments. This indicates a good reproducibility of the process.



Figure 3. Specific cumulative CO<sub>2</sub> uptake of fRCA

According to the TGA data and, specifically, based on the mass loss between  $600 - 800^{\circ}$  C (Scrivener et al. 2018) the

native fRCA already contained about 6.4 g CaCO<sub>3</sub>/100 g dry matter, indicating that it had already undergone undeliberate carbonation. However, this value could be increased to more than 9 g/100 g within one hour in the rotating drum reactor.

## 3.2 Penetration resistance

The penetration resistance of the carbonated samples was always around 0,5 MPa over the 14-d test period. The control samples had somewhat lower values and showed a non-systematic scatter between curation time-variants. This may have been due to an uneven distribution of reactive components in the samples.



**Figure 4.** Development of the penetration resistance [MPa] in the control (purple) and carbonated (yellow) fRCA over time

The edges the penetrometer marks in the samples (Figure 5) show clear differences between carbonated and control fRCA. While the control penetrometer mark had just the same diameter as the instrument, the carbonated sample (right) had a larger mark diameter. This suggests that the carbonated sample was more brittle. Also, the surface of the control material seemed more compact, smoother and was slightly darker than that of the carbonated material.



**Figure 5.** Surface of compressed fRCA specimens after penetrometer tests (left: control, right: carbonated)

#### 3.3 Sieve passage

When the ring samples were stripped onto the sieve tower, there was a clear difference between the carbonated and the native material. The carbonated material formed a coherent cylindrical sample that enclosed almost the entire material, whereas the control formed a crust of about 0.5 cm on the surface (Figure 6). The underlying native fRCA was loose. This effect could be because the CaCO<sub>3</sub> is evenly distributed in the carbonated fRCA, whereas in the control sample it forms a barrier on the exposed surface, which adheres more strongly and makes it difficult for CO<sub>2</sub> to diffuse into the lower layers of the sample.



Figure 6. Stripped samples (left: control, right: carbonated)

Sieving the samples highlighted further differences and confirmed the assumptions on the effects of fRCA carbonation. The carbonated material crumbled completely within 20 s of sieving although it seemed completely cured. The crust of the control fRCA, which was significantly thinner, remained stable and only broke into slightly smaller clumps during sieving. Other granulometric effects of the carbonation procedure were visually discerned (Figure 7).



**Figure 7.** Comparison of sieved control (top) and carbonated (bottom) samples.

During the one-hour carbonation process, granules with a diameter of > 4 mm formed. The control material broke into clods with a diameter of 2 - 3 cm. When comparing the next smallest fraction, a change in colour and shape due to exposure to CO<sub>2</sub> (lighter hue, rounder grains) was recognized, which may have been due to granulation and coating with CaCO<sub>3</sub>.

The sieve lines (Figure 8) show a shift between the control and carbonated fRCA, especially in the range < 1 mm. Apparently carbonation caused a decrease in fine particles due to agglomeration.



**Figure 7.** Sieve line of control (purple) and carbonated (yellow) fRCA

#### 4. Conclusion and Outlook

This study aimed at elucidating the effect of fRCA carbonation on its handling and storage characteristics. Within one hour, the material took up 2.6 m-% CO<sub>2</sub>/100 g dry matter. Upon carbonation the material appeared lighter in colour and exhibited discernible granulation. The dried carbonated samples were more friable than the control fRCA. This supports the hypothesis that accelerated carbonation may reduce the tendency of fRCA crust formation. This was, however, not mirrored by penetration resistance testing.

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