

## **CHANGES OF THE MATERIAL PROPERTIES BY SELECTIVE CARBONATION OF RECYCLED AGGREGATES FROM CONCRETE**

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### **Abstract**

The aim of this work is to bind a maximum of carbon dioxide in concrete recyclates permanently by means of a selective carbonation. This process is meant to change the properties of the recyclate in such a way that it comes to a decrease in porosity and to an increase in strength. The carbonation process is a well-known procedure. As diffusion-steered process it runs, however, very slowly and takes years. The technical challenge of this work is to accelerate the carbonation reaction in a reactor and thus making it technically usable. A test device was developed for the directed carbonation. It consists of a tubular reactor and several measuring units. The test specimens are continuously subjected to a preset gas mixture in the tubular reactor. The gas flows evenly through the reactor. The first tests were carried out on cement stone at varying CO<sub>2</sub>-concentrations over different time intervals. The results show a correlation of the samples weight increase and their density after staying in the reactor. The CO<sub>2</sub> absorption was confirmed by means of thermal analysis after the carbonation. In further tests a carbonation of concrete recyclates was carried out. The treated and the untreated concrete recyclates as well as the natural aggregate were taken to produce concretes. Then they were characterized as to their fresh concrete properties as well as their hardened concrete properties and both compared and evaluated to one another.

Keywords: selective carbonation, recycled concrete, carbon dioxide

### **1. INTRODUCTION**

Concrete is by far the most deployed building material in the industry. This enormous usage of concrete consequently leads to an increase of crushed concrete in the total waste balance. Since sustainability becomes more and more important in our society and since we are facing a shortage of all resources, the high-quality recycling of demolition waste is gaining increasing significance. For environmental and economic reasons such recycling is strongly required.

There already exist recycling processes for old concrete which aim at separating the used concrete into cement stone and the original aggregate. However, by means of the recycling techniques available on the market, a complete separation is not possible. The resulting

recycled aggregate consists of primary aggregate and adherent cement stone. As a result it comes to an increase of the capillary pore share for the secondary aggregate in comparison to the primary aggregate. That is why the deployment of concrete recyclates as aggregate results in a deterioration of the fresh and hardened concrete properties.

By means of a selective carbonation of the concrete recyclates it should be possible to achieve a decrease in porosity while the raw density as well as the strength increase. Furthermore, a maximum of  $\text{CO}_2$  should permanently be bound in the concrete recyclates. So, a property improvement of recycled concrete can be combined with the positive effect of a permanent bonding of  $\text{CO}_2$ . At the moment is the selective carbonation of the concrete recyclates in practice already in force.

The course of carbonation within the old concrete is a process in which  $\text{CO}_2$  intrudes into the concrete's capillary pore system by means of diffusion.  $\text{CO}_2$  and calcium hydroxide  $\text{Ca}(\text{OH})_2$  dissolve in the pore water and react to calcium carbonate  $\text{CaCO}_3$  which settles in the concrete's capillary pores. Due to the formation of  $\text{CaCO}_3$  it comes to a decrease of calcium ions in the pore water. As a result, further  $\text{Ca}(\text{OH})_2$  can dissolve and the reaction can go on until all the  $\text{Ca}(\text{OH})_2$  is used up. The  $\text{CaCO}_3$ -crystals in the pores continue their growth which leads to a structure densification. The carbonation of the calcium hydroxide in the Portland cement stone leads to a total volume increase of approx. 11 Vol.-% [1]. The total porosity of the concrete decreases and it comes to a strength increase of the concrete recyclates. Both leads to a quality improvement of the old concrete. After a successful carbonation the old concrete can be used as aggregate for new concrete.

## 2. EXPERIMENTAL SET-UP

A test device was developed for the directed carbonation [Fig. 1]. It consists of a tubular reactor and several measuring units. The test specimens are continuously subjected to a pre-set gas mixture in the tubular reactor. The gas flows evenly through the reactor. The  $\text{CO}_2$  concentration of the reaction gas can be varied between 0 Vol. - %  $\text{CO}_2$  and 100 Vol. - %  $\text{CO}_2$ . The gas concentration is taken by means of the  $\text{CO}_2$  – measuring unit located in the inlet. A salt solution in the tubular reactor ensures a continuous relative humidity of 70%. The monitoring of the air humidity and the temperature inside the reactor is done by means of a measuring unit.

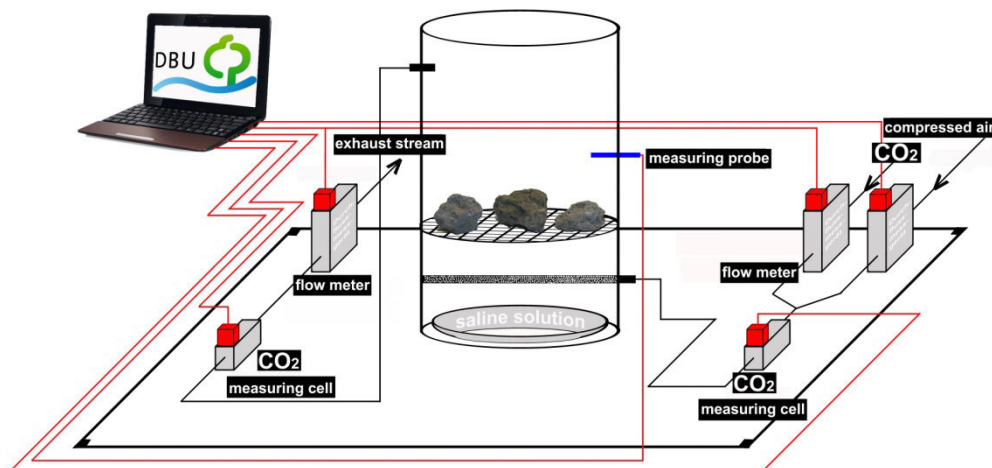


Fig. 1: Schematic illustration of the experimental set-up

### 3. TEST EXECUTION AND RESULTS

#### 3.1 Selective carbonation of cement stone pastes

The first tests were carried out on cement stone blocks. The cement stone blocks had a size of 4 x 4 x 2 cm. The experiments were carried out over varying time intervals at a constant CO<sub>2</sub> concentration. The results shed light on the carbonation depth and the relative weight increase as a function of the w/c ratio of the cement paste prisms. The relative weight increase indicates the carbonation level. As rule of thumb it can be said that at complete carbonation it comes to a weight increase of 30 % as far as cement pastes are concerned.

In further test series investigations were carried out on samples having a grain size of 2/4 mm and a w/c ratio of 0.5. The experiments were carried out at 20, 40, 60, 80 and 100 vol. -% CO<sub>2</sub> at varying weathering times.

The figures 2 to 3 show a part of the test series results with a CO<sub>2</sub> concentration of 20 vol. -%. Figure 2 shows the correlation between the weight increase and density of the samples after their staying in the reactor. A steady increase in density can be observed in dependence of the weight increase. This suggests that a CO<sub>2</sub> absorption with CaCO<sub>3</sub> formation must have taken place. After the weathering a decrease of the calcium hydroxide content and an increase of the calcium carbonate content could be determined by means of thermogravimetry (Fig. 3). For example, the calcium carbonate content in the initial sample was only at 7 %. After the treatment in the carbonation reactor the calcium carbonate content doubled.

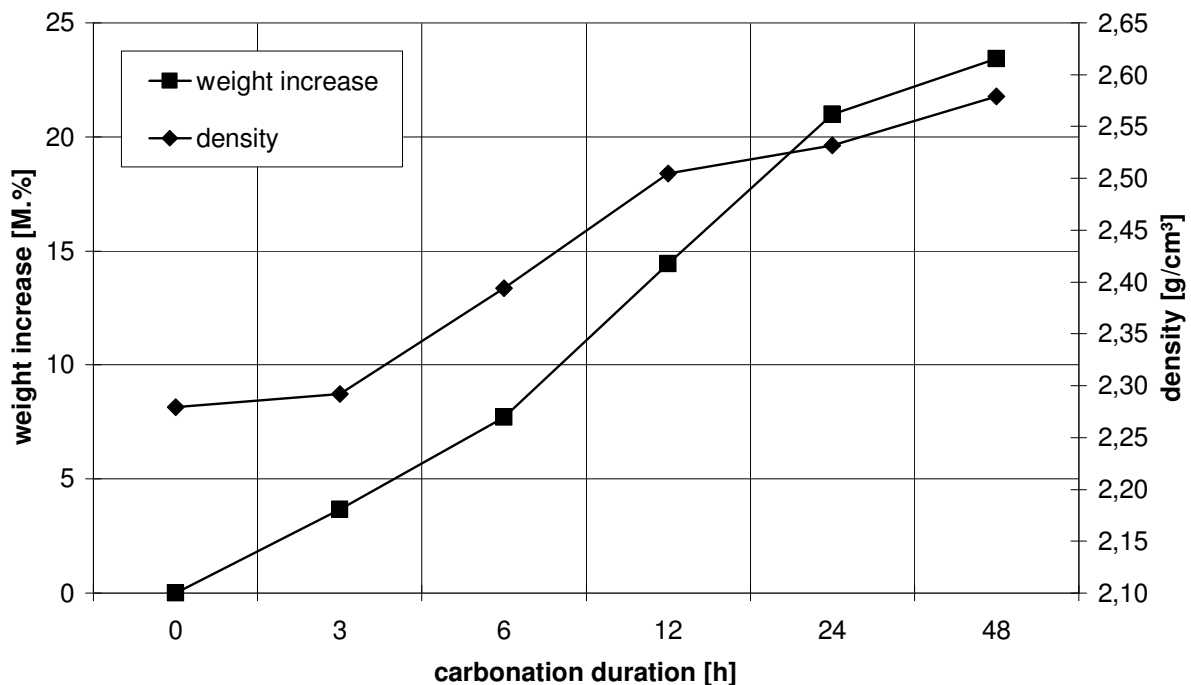


Fig. 2: Increase in weight and density after carbonation

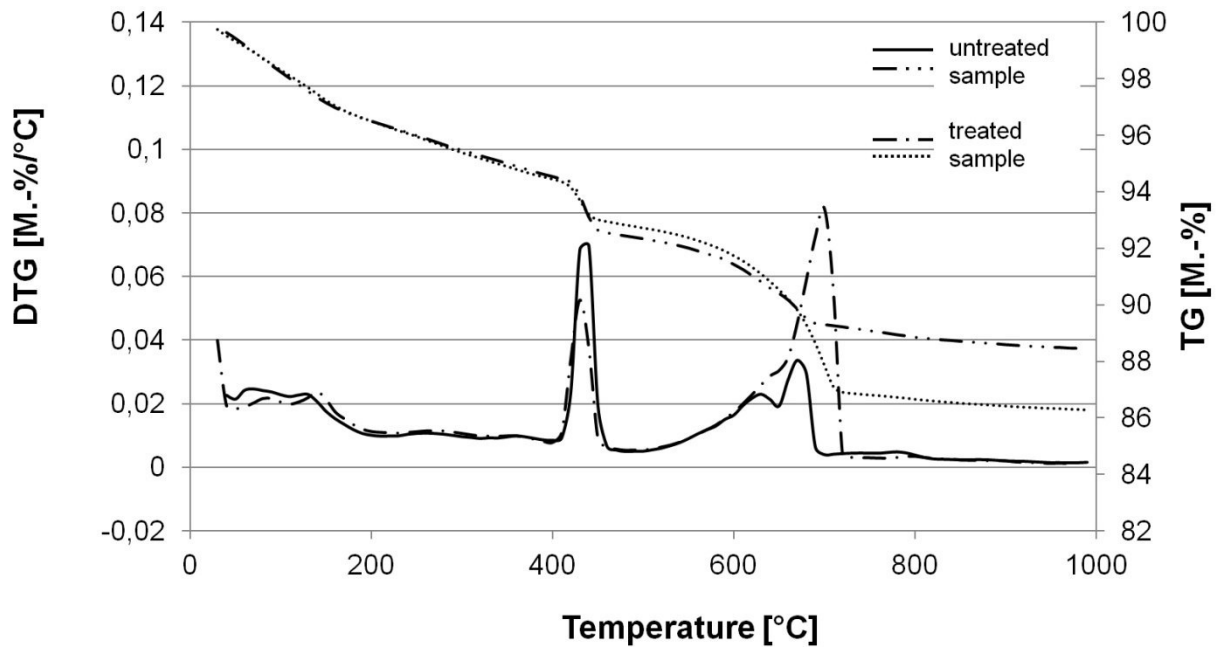


Fig. 3: DTA/TG before and after carbonation

The pore volume and the pore size distribution for the grain size 2/4 were determined by means of mercury intrusion porosimetry. Table 1 and Fig. 4 show the gained results.

Table 1: Porosity before and after carbonation

sample	porosity [%]
untreated	14.6
treated	10.2

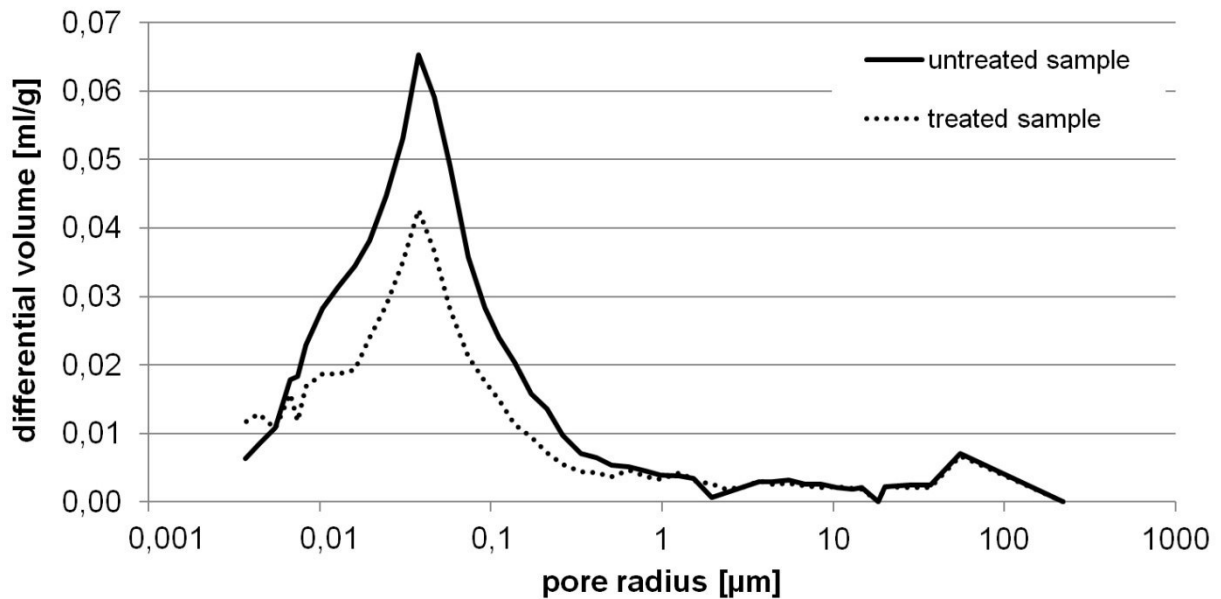


Fig. 4: Distribution of pore radii before and after carbonation

The results gained by means of mercury intrusion porosimetry show a decrease in the total pore volume. The porosity of the cement stone matrix is reduced due to subjecting it to CO<sub>2</sub> from 14.6 % to 10.2 %. The structure of the cement stone becomes denser by the carbonation.

Fig. 4 shows that the decrease in porosity occurred for the most part in the pore radii range of 8 nm to 1 µm. In this range capillary pores exist. That means that, due to the treatment, a reduction of the capillary porosity occurred. This fact has a pronounced effect on the compressive strength of the cement stone since the capillary porosity is an essential parameter for the compressive strength [2].

### 3.2 Concrete Tests

In order to get a better judgement on the effect of the carbonated cement stone matrix on the respective fresh or hardened concrete investigations, several concretes were produced. The concrete composition, i.e. aggregate, cement and water was consciously chosen to be simple in order to reduce additional influences to a minimum. 5 concretes were produced that differ in the composition of their aggregates. Table 2 lists the concretes with the different aggregates. All concretes were produced using a Portland cement CEM I 42.5 N and a w/c-ratio of 0.5. The deployed aggregate was put together according to the particle size distribution curve A/B16 [3]. Only the grain groups 2/8 and 8/16 were replaced by RC-material (45 Vol.-% respectively 100 Vol.-%) according to the DAfStb-guideline [4]. The secondary and the carbonated secondary aggregate were inserted in the grain range 2 to 16 mm of the particle size distribution curve A/B16 in such a way that it came to an even distribution following the primary aggregate.

Table 2: Designation and meaning of the concretes

<b>Designation</b>	<b>meaning</b>
RFB	Reference concrete with natural aggregate
RB/45	Recycled concrete with 45 Vol.-% recycled aggregates from concrete $\geq 2$ mm
RB/100	Recycled concrete with 100 Vol.-% recycled aggregates from concrete $\geq 2$ mm
RBC/45	Recycled concrete with 45 Vol.-% carbonated recycled aggregates from concrete $\geq 2$ mm
RBC/100	Recycled concrete with 100 Vol.-% carbonated recycled aggregates from concrete $\geq 2$ mm

#### 3.2.1 Fresh Concrete Investigations

By determining the slump directly after the concrete production and then again after 30, 60 und 90 min the consistency course of the produced fresh concretes recorded. The slump directly after production was similar for all 5 concretes. With time progressing the setting of the concretes begins. Fig. 5 it can be observed that the concrete with the carbonated recycle shows a similar behaviour as the reference concrete as far as setting is concerned.

For all mixtures it can be said that there were no big deviations since the w/c-ratio, the cement content and the core humidity of the deployed aggregates were set equal for all concretes.

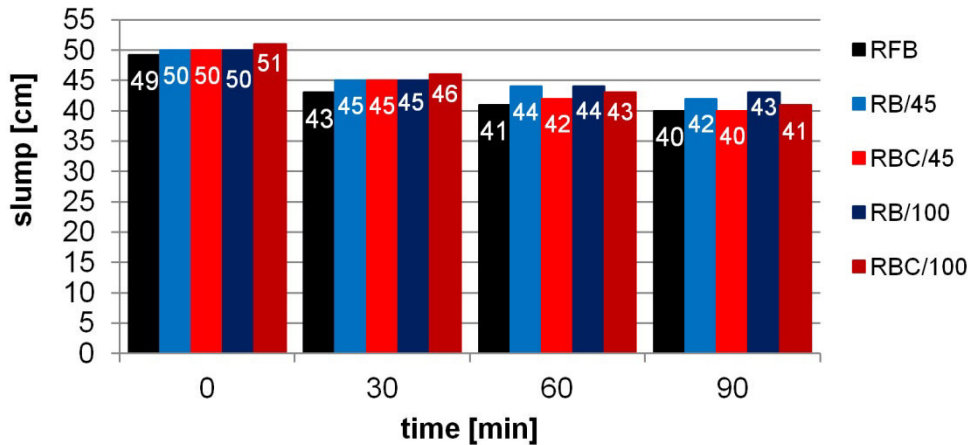


Fig. 5: consistency behavior

### 3.2.2 Hardened Concrete Investigations

The evaluation of the strength development for the 5 concretes was carried out by taking their compressive strengths after 7, 14, 28 and 56 days. The results are given in Figs. 6 – 9.

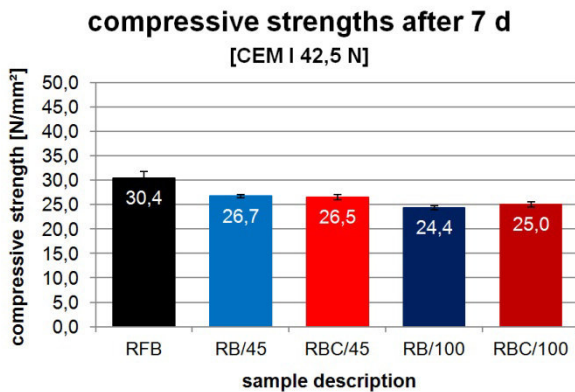


Fig. 6: compressive strengths after 7 d

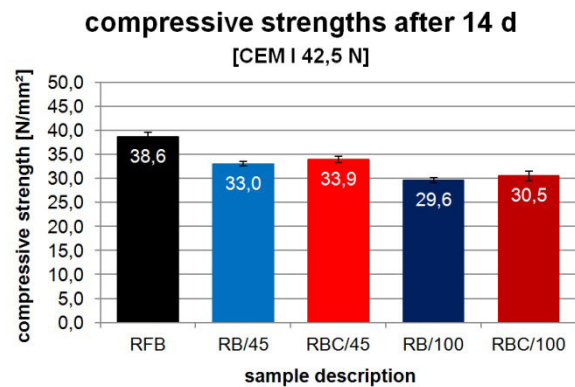


Fig 7: compressive strengths after 14 d

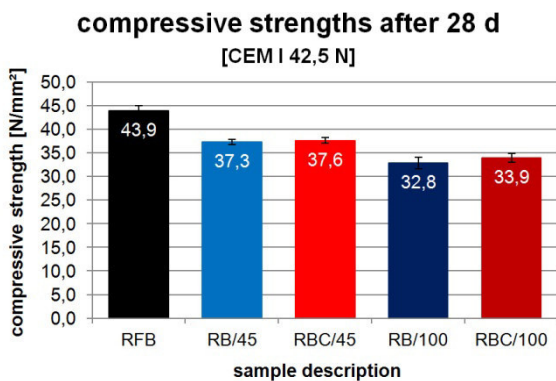


Fig. 8: compressive strengths after 28 d

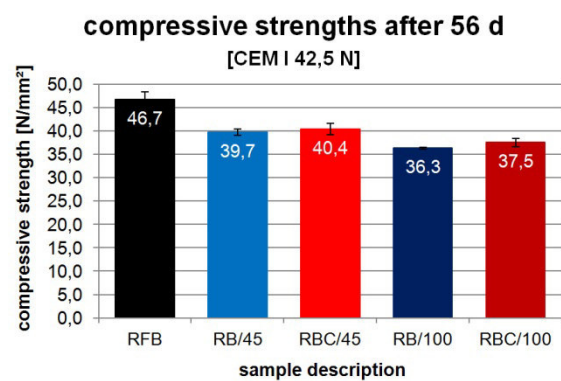


Fig. 9: compressive strengths after 56 d

The Figs. 6 to 9 illustrate that with the hydration time progressing an increase in compressive strength can be observed for all concretes. When one compares the concretes with secondary and carbonated secondary aggregate, one can see over the different hydration levels that it

comes to a slight but nevertheless significant increase in strength as far as concretes with carbonated secondary aggregate are concerned. Only in case of early strength after 7 days no strength increase can be observed yet when carbonated material with a recycle share of 45 Vol.-% was used. The higher the content of recycled aggregate in the concrete, the more pronounced is the strength increasing effect of carbonation.

### **3. CONCLUSION**

The test results show that a structure densification in the Portland cement stone by means of a directed carbonation in a lab reactor is possible. It comes to a bonding of CO<sub>2</sub> and an improvement of the material properties. By means of a directed carbonation of the cement stone matrix it comes to an increase in saturated density and a decrease in porosity. Measurements show that the CO<sub>2</sub>-absorption by the cement stone can be significantly accelerated under the chosen test conditions. The time scale for the carbonation of buildings which is usually measured in years is shifted to the hour scale. The set CO<sub>2</sub>-concentration of 20 Vol.-% used in the tests is comparable to real concentrations in power plant exhaust fumes.

In the next step concretes were produced under usage of concrete recyclates that had undergone the CO<sub>2</sub>-treatment, then these concretes were investigated. In these investigations concretes with treated recyclates were compared and evaluated with concretes that had been produced with untreated recyclates or natural aggregates. After the compressive strength tests, tests on the concrete durability were carried out.

Looking at the determined fresh concrete parameters no significant differences could be found regarding the usage of carbonated secondary aggregates. Due to the deployment of carbonated recyclates it comes to an increase in the compressive strength in comparison to concretes with non-carbonated recycled aggregates. In case of a carbonated recycle share of 45 Vol.-% the strength increase after 56 days is at 1.8 % compared to concretes with untreated recyclates. When one increases the recycle share to 100 Vol.-% it comes to a strength increase of 3.3 % after 56 days.

Summarising it could be shown that a directed carbonation of adhering cement stone at the secondary aggregates there is a denser material structure and thereby the hardened concrete properties can be improved. This result puts the demolition material concrete once again into the focus of the recycling and resource management industries and can contribute to close the cycle of materials.

### **ACKNOWLEDGEMENT**

The authors thank the German Federal Foundation for Environment (DBU) for the support in the context of this research project.

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