

# FORCED CARBONATION OF RECYCLED CONCRETE AGGREGATES

MARC-PATRICK PFLEGER\*, MARKUS VILL

*University of Applied Sciences Vienna, Favoritenstraße 226 A-1100 Vienna, Austria*

\* corresponding author: marc-patrick.pfleger@fh-campuswien.ac.at

**ABSTRACT.** The production of concrete, especially the contained cement clinker, causes a high proportion of the manmade gaseous air pollution. Studies show that the decarbonation of limestone effects roughly 8% of worldwide CO<sub>2</sub> emissions, which underlines the need for action and research. Due to the present building and infrastructure stocks and their obsolescence, or rather replacement, the amount of demolition material is permanently increasing. In many cases the demolition material consists of high-quality concrete, which could be used again as recycled aggregate in the production process to conserve sources of raw materials. Most of the concrete waste is not carbonated which means that the potential as CO<sub>2</sub> sink can be exploited in the course of the recycling process. Since the recycled and carbonated material is used as a replacement for primary raw material, the state of carbonation itself is irrelevant to any steel corrosion or associated problems. The main objective of this work was to investigate the CO<sub>2</sub> absorption of concrete waste, to investigate and prove its use as a possible carbon sink. At this point, the carbonation velocity had to be optimised by the use of special equipment. First test series show that the ecological footprint of concrete can be significantly reduced if concrete waste is used for carbon storage and admixed as recycled aggregate during the production process. The aim of this study was to point out further possibilities for the ecological optimisation of concrete besides alternative binders, which are the subject of research in various projects worldwide.

**KEYWORDS:** Carbonation, concrete, recycled aggregates.

## 1. INTRODUCTION

The protection of the environment and the global climate is currently a major field of action. Not only changing climate conditions are perceptible, but also rising raw-material demand, land consumption and the pollution of important habitats occur as main problems.

Since every industrial sector has more or less impact on the environment, the question arises where significant entry points for ecological optimisations can be found. Often a close connection can be found between the amount of materials used by a particular business branch and its environmental impact. The construction industry and related trades are associated with an enormous need for raw materials. Further processing uses high amounts of energy, which means that there is a tangible potential for greenhouse gas reduction.

One of the most commonly used materials for civil engineering structures and buildings at present is concrete or rather reinforced concrete. Especially the contained cement clinker is a highly climate relevant substance. According to the current state-of-the-art, structural concrete cannot be adequately replaced, neither technically nor economically. Its robustness, stability and formability, in combination with low material and manufacturing costs, are the main reasons for the continuously rising demand. Air-pollution is increasing analogously to the growing amounts of produced binder or rather the contained

cement clinker. The fact that nowadays about 8% of the worldwide CO<sub>2</sub> emissions (by population) result from producing cement has to be highlighted as absolutely critical.

The mentioned emissions result only partly from combustion processes to provide the needed heat to process clinker out of lime stone. If modern technology is used, over 50% of the overall CO<sub>2</sub> emissions concerning the cementitious binder production result from the lime stone decarbonation, as seen in Table 1.

Emissions due to the raw material decarbonation are process-related and cannot be substituted, if the known characteristics of cement shall not be modified. Especially the corrosion protection of the reinforcement depends on the alkalinity of the hardened concrete.

This paper brings the described problems with greenhouse gas in context with rising amounts of demolition materials, especially concrete recycling products. The majority of concrete waste is not carbonated, in particular the inner sections of structural elements of many civil engineering structures, like bridges with large thicknesses of concrete members.

## 2. BACKGROUND FOR ECOLOGICAL OPTIMISATION OF CONCRETE

As described before, concrete is an indispensable construction material, because of its technical charac-

	Raw material decarbonation	Combustion of fossil fuels	Combustion of biogenic fuels	total
2016	524	260	72	856
2017	517	256	71	844
2018	515	254	75	844

TABLE 1. Specific CO<sub>2</sub> emissions in [kg] per [to] clinker in Austria [1].

teristics. Therefore, optimisations concerning the ecological footprint of concrete can actually only be made if the occurring of actual disadvantages can be consistently avoided. In general, improvement potential can be seen in alternative binder recipes or using high-grade recycling processes for an ideal reuse of concrete waste.

The minimum share of cement clinker in the most commonly used standardised cement mixtures (CEM II) is about 65%. The rest consists, depending on the exact cement type, of fly ashes, blast furnace slag or lime dust. The currently available amount of fly ash and slag is already completely used up, so that no more substitution can be expected. Furthermore, due to the focus of future energy policy, the available quantity of additives, based on fossil combustion processes, will decline. Therefore an extensive amount of research activity is done worldwide to develop alternative binders for concrete applications, like [2], [3]. Commonly the main objective is to reduce or avoid the share of cement clinker in the binder mixture. Problems with durability may occur as a consequence and are not investigated enough in long term tests.

Apart from binder optimisation the rising stream of demolition material has to be involved in high quality recycling. Particularly in urban regions new buildings can only be produced if a site is previously cleared from its original building stock. It can be observed worldwide that a building's life cycle is decreasing, which means that the building stock becomes a more important source of raw material in the sense of "urban mining". Generally demolished concrete structures are processed by one or more crushers to receive a useable concrete granulate.

Depending on the valid standards, this concrete recycling aggregate can be used with a certain percentage as replacement for natural aggregates. Concerning the national documents of Austria, additional differentiation is made for admixing, depending on the grain size of the recycling product [4]. Usually those recycled aggregates have a grain size distribution from 0 to 30 mm or smaller. The maximum grain size in general depends on the desired configuration of the jaw crusher.

### 3. CARBONATION OF CONCRETE

#### 3.1. CARBONATION AS CONCRETE DAMAGE

The carbonation of concrete is usually known as a classic concrete damage and can be described as the

reaction of CO<sub>2</sub> from the surrounding atmosphere with calcium hydroxide, as a product of the hydration of cement clinker. The calcium hydroxide is contained in the pore water of a concrete matrix and its concentration may vary due to the particular binder mixture and the surrounding conditions during hydration. The specific concentration of lye determines the alkalinity of the pore water and thus the capability to prevent the reinforcement from corrosion. Under the absorption of CO<sub>2</sub> this potential declines over time, starting from the concrete surface. Therefore the tolerable carbonation depth is generally limited to the minimum concrete cover of the reinforcement. Concrete technological measures are taken to prevent fast carbonation. Main ambient parameters for the carbonation velocity can be:

- Carbon dioxide level of the surrounding atmosphere,
- Ambient air and concrete temperature,
- Concrete moisture,
- Pressure.

Additional influencing factors result mostly from the material itself such as content of cement, porosity or (micro) cracks which provide an increased surface area for carbon dioxide absorption and reaction [4].

#### 3.2. CARBONATION OF CONCRETE AS CARBON SINK

The inner cross-section parts, or rather the area between the outer reinforcement layers, are usually not carbonated until the demolition of a structure. Emitted CO<sub>2</sub> caused by the calcination of the binder remains in the atmosphere and has a negative impact on the materials ecological footprint.

The question arises as to which surrounding conditions and related time span are required until the carbonation process is completed after a concrete structure is recycled into aggregates. As described before, the carbonation of concrete is a natural process, though natural conditions only lead to a slow reaction process with the actual CO<sub>2</sub> consumption not defined. Nevertheless, there is carbon absorption at any time where the essential conditions, such as concrete moisture, are given. The chemical conversion of calcium hydroxide into calcium carbonate effects that a solution transforms into solid [5]. As a result, the gas permeability of the cement matrix decreases and

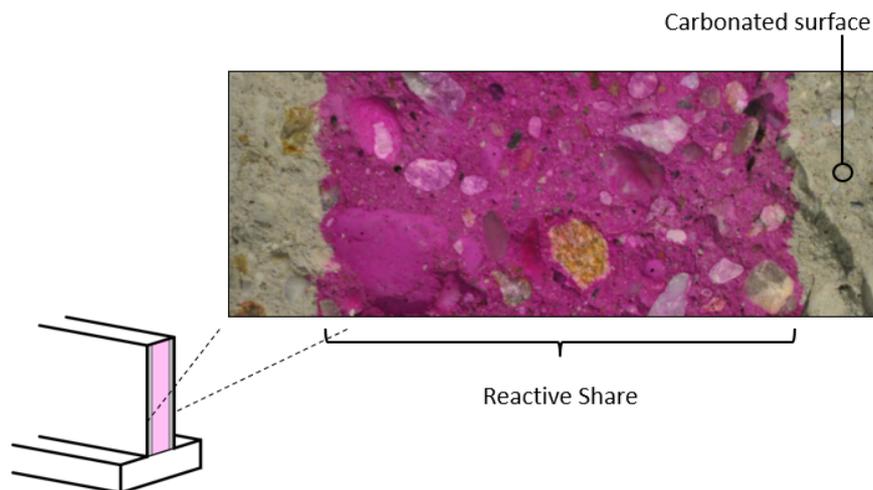


FIGURE 1. Not-carbonated share of a cross-section.

the carbonation velocity asymptotically approaches zero within the grain [6].

Based on this knowledge, an artificial method shall be preferred in comparison to carbonation under natural conditions. Controllable parameters and the possibility of measurements during the carbonation ensure a consistent recycling product [7].

#### 4. EXPERIMENTAL SETUP AND METHODS

The test setup was designed to investigate forced carbonation of recycled concrete aggregates, which means that no correlation with natural carbonation could be expected. The first step of the experimental carbonation test series was to determine the exact amount of carbon dioxide which can be bound by a concrete matrix. For this purpose, special equipment was necessary, since standard carbonation chambers do not allow to vary and monitor the reaction conditions in real time as desired [8]. Especially the used experimental setup allowed to monitor and control the exact  $\text{CO}_2$  consumption of the sample material in real time. Further it was planned to find the optimal parameters to fully carbonate a particular sample as fast as possible.

##### 4.1. REACTION PARAMETERS

As basis for the equipment design, the previously defined external parameters were taken into consideration. Thus temperature, humidity as well as the amount of  $\text{CO}_2$  in the sample atmosphere should be varied. Increasing the ambient pressure was planned for future sample series. The range of reaction parameters was defined according to feasible values in large-scale techniques or rather concerning industrial facilities as shown in Table 2.

	From	To
$\text{CO}_2$	0,04%	30%
Humidity	0%	100%
Temperature	20°C	65°C

TABLE 2. Range of reaction conditions.

##### 4.2. EQUIPMENT DESIGN

The equipment consists of a carbonation chamber that can be top-loaded with the sample material. A removable grate is filled with the sample aggregates up to 25cm layer thickness. The container sits hermetically on a catch, so that a forced flow of the ambient atmosphere through the material can be ensured by the integrated fans. The perfusion of the sample container happens from bottom to top.

As shown in Figure 2, all fittings and connections are located at the cover of the carbonation chamber. So only the tempered and humidified granulates are inside the reaction tank which consists of stainless steel. The acryl glass lid is prepared for an attachable steel frame to ensure sufficient stiffness if tests with overpressure will be carried out in the future.

A main requirement was that the  $\text{CO}_2$  absorption of the samples should be measurable at any time of the carbonation process. A standard carbonation chamber does not allow a real-time monitoring of the actual  $\text{CO}_2$  consumption as desired. Further, any implementation of additional custom equipment is easier, dealing with a setup from scratch. It is planned to generate a completely automated process by using artificial intelligence to set the ideal reaction conditions and vary them over the test's duration. Due to the moistened material and the increased humidity inside the chamber it is not possible to obtain reasonable results by measuring the sample's mass during the experiment. The material moisture varies during the test. Additionally, condensation will distort

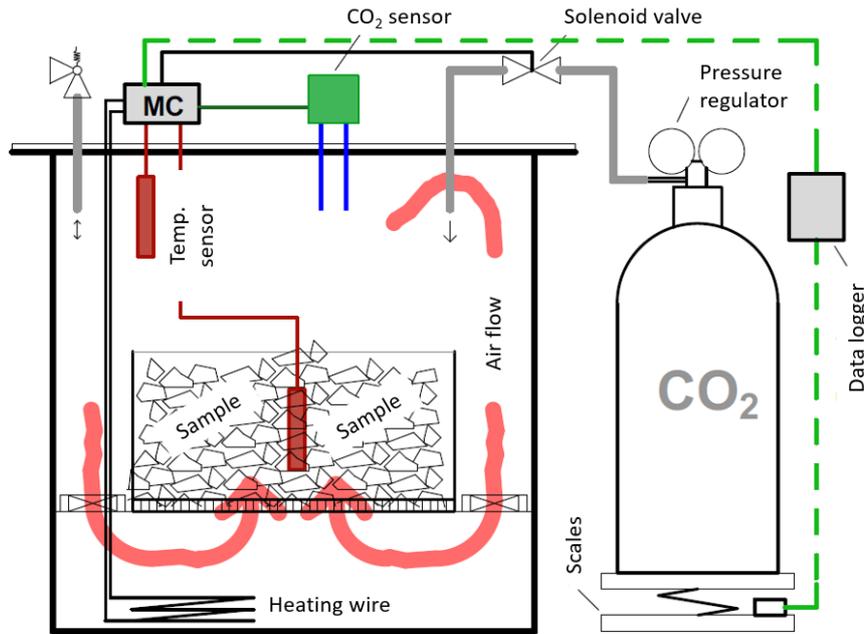


FIGURE 2. Simplified scheme of the test set-up.

any weighing process inside the chamber.  $\text{CO}_2$  reacts with the calcium hydroxide contained in any concrete waste to  $\text{CaCO}_3$ , so that a direct conclusion can be drawn about the absorbed carbon. Therefore the increasing weight of the aggregates shall be proved by other boundary parameters such as the decreasing gas bottle mass, the opening time of the valve and the flow volume of  $\text{CO}_2$ . All these three parameters were monitored and compared to validate the accuracy of the data. The measured values are documented and analysed in real time, which allows statements about the actual  $\text{CO}_2$  consumption according to current reaction conditions. At the end of the experiment dry mass of the sample can be weighed and compared with the initial mass in order to verify the results derived from the experiment.

Data-logging and the real-time analysis is physically located in an extra housing where also the  $\text{CO}_2$  probe was placed. Gas from the reaction chamber is extracted, analysed and pumped back. Since only carbon consumption was expected here, no precautions were taken to reduce the carbon dioxide share of the test atmosphere. Due to the more or less constant reaction of  $\text{CO}_2$  the valve is only opened when necessary to keep the concentration just as desired. Ideally the reaction conditions are varied during the carbonation process to achieve more efficiency. Furthermore, initial tests show that varying the reaction conditions according to different cement types gives better results.

#### 4.3. SAMPLES

Granulated concrete with accurately known recipe was found to be a suitable sample material for the first test series, which also should serve for data ver-

ification. Several concrete test cubes with 15cm edge length were produced and stored in the air for 60 days under standardised conditions for 60 days exposed to air.

### 5. TEST SERIES

#### 5.1. SAMPLE CONCRETE AGGREGATES

All experiments carried out with crushed concrete aggregates were done by using the same concrete mixture to achieve ideal comparability. The determined samples were made from C25/30 containing CEM II B with a maximum grain size with 22mm in the matrix. The binder portion was defined as 15% of the wet concrete mass. The water-cement ratio was kept at 0.5.

#### 5.2. PREPARATION OF THE SAMPLE MATERIAL

The first step of each test series was the determination of the samples dry mass. To extract residual moisture the grains were put in a dry kiln until the dry weight was proven by mass stability. As basis for all test series the aggregate mixture was separated into grain fractions 0 to < 8mm and 8 to 22mm. The fraction <8mm had 26.5% of the total mass, thus 73.5% were weighed with 8-22mm.

Depending on the particular experiment the samples were moistened with a defined amount of water to exploit the influence of material moisture to the carbonation velocity. In fact, the ideal material moisture cannot be expressed with a simple percentage, since the porosity, in particular the pore diameter, is a key factor for the amount of water needed to moisten the pore walls [5].

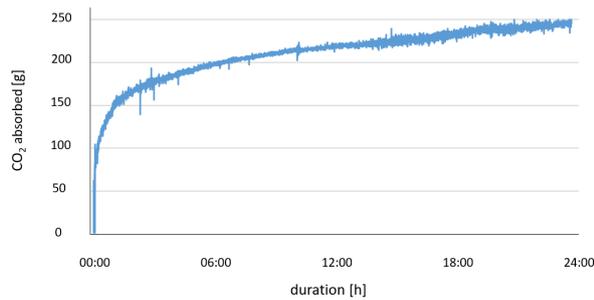


FIGURE 3. Carbonation results for C25/30, CEM II-B, grain size 0 – 8mm.

### 5.3. STRUCTURE OF THE GRAIN FRACTIONS

After separating the grain fractions visual inspection alone showed that most of the cementitious parts of the crushed down concrete find themselves mainly in small grains. This means that also most of the potential carbon sink was initially expected in small grain fractions.

### 5.4. INFLUENCE OF THE LAYER THICKNESS

Several carbonation tests were done with mainly small grain sizes from 0 to 8mm. As expected, the permeability of a sample layer consisting of only small grain fractions is poor. The effect is seen when checking the carbonation of grains from the centre of the bulk. When testing coarse aggregates, a more consistent carbonation through the whole layer depth was verified, whereas aggregate mixtures do not ensure enough permeability as well as recycling sands. The issue with poor permeability can be obviated by using a non-static process.

## 6. RESULTS

### 6.1. CONCRETE AGGREGATES

The following results show the total amount of bound carbon dioxide in the samples over the test duration. Ideally the mass increasing shall not be compared directly with the total mass at the experiment's start, because of the contained natural aggregates in the concrete matrix, which are not reactive. When the ecological improvement is calculated, only the binder or cement clinker share shall be considered.

#### 6.1.1. GRAIN SIZES UNDER 8MM

Figure 3 shows exemplarily how carbon absorption proceeds at grain fractions below 8mm. The experiments duration was 24 hours with 20% CO<sub>2</sub> in the carbonation chamber, heated to 40°C. The carbon dioxide concentration as well as the temperature was constant during the whole test. The sample's dry mass was determined to 11.289g at the beginning.

It can be seen that the CO<sub>2</sub> consumption is very high at the beginning, which can be explained by the rapid carbonation of the surface areas of the crushed

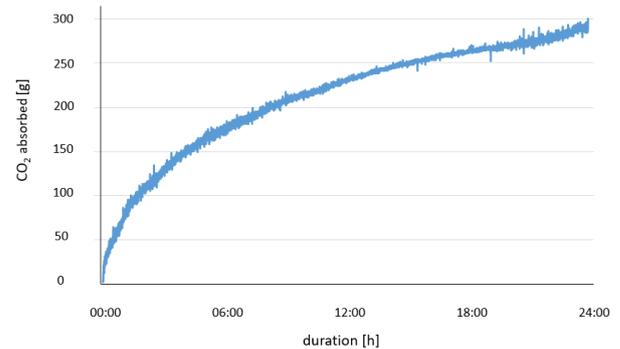


FIGURE 4. Carbonation results for C25/30, CEM II-B, grain size 8 – 22mm.

down concrete and the high share of cement of the recycling sands.

The further course of the graph varies visibly over time. In this case this phenomenon cannot be explained by the saturation of the aggregate. When the test was completed it was seen that poor carbonation results were achieved in the centre of the bulk. This can be explained by too little gaseous exchange due to the high packing of the small grains. The test ends with 238g CO<sub>2</sub> bound in the sample, resulting in 21g CO<sub>2</sub> per 1kg recycled aggregate (grain size < 8mm).

#### 6.1.2. GRAIN SIZES OVER 8MM

Figure 4 shows exemplarily how carbon absorption proceeds at grain fractions over 8mm. The outer parameters of the experiment were exact the same as mentioned in 6.1.1., which means: 24 hours of carbonation with 20% CO<sub>2</sub> at 40°C in the test setup. Both the carbon dioxide concentration and the temperature were stable during the whole test. The sample's dry mass was determined to 11.385g at the start.

Again it can be seen that CO<sub>2</sub> consumption is high at the beginning, whereas the gradient is less prominent when smaller fractions are analysed. The graph shows a clearly more flat carbonation curve at the start due to the smaller surface to volume ratio.

Furthermore, the reaction velocity decreases permanently over time and approaches zero. After the removal of the sample material it was seen that constant carbonation took place in all layers of the bulk. The test ends with 284g CO<sub>2</sub> bound in the sample, resulting in 25g CO<sub>2</sub> per 1kg recycled aggregate (grain size 8-22mm).

The test series show that CO<sub>2</sub> can be absorbed with 2.1% to 2.5% of the sample mass in a static process. That mass increase is accompanied by the formation of lime in the concrete matrix which means that simultaneously the porosity of the samples must decrease. This phenomenon was proved by an additional saturation experiment before and after the carbonation process.

## 6.2. EXEMPLARY PRESENTATION OF THE ECOLOGICAL BENEFIT

Based on the results from 6.1. the ecological advantage of recycling concrete with forced carbonated can be pointed out. Due to current Austrian national standards, the admixture of up to 50% recycled which is explained by unpredictable increased water needs [4].

The following example states, that 50% of the natural aggregates are substituted by a forced carbonated recycling product. It is expected that a concrete strength of C25/30 with CEM II shall be achieved. It can be assumed that 900kg of carbonated aggregates are admixed while concrete production. Thereby 22.5kg carbon dioxide can be credited per m<sup>3</sup> fresh concrete if the ecological footprint of the material is calculated. In comparison to the carbon emissions due to the binder production with approximately 170kg per m<sup>3</sup>, an improvement of about 13% can be achieved.

## 7. CONCLUSION

The exemplary stated test series show the potential of forced carbonation of recycled concrete aggregates in principle. An absorption of 2.1% to 2.5% of the sample's mass was achieved by the test setup. As the cement clinker production increases permanently worldwide and thereby the carbon dioxide emissions rise, ways for ecological optimisation must be found. The experiments show that forced carbonation is suitable to utilise existing carbon sink potentials of demolished concrete. If the carbonation of recycled concrete aggregates can be implemented in current processes with high carbon dioxide emissions, such as cement plants, the ecological footprint of concrete actually can be reduced.

Currently, the life-cycle assessment of concrete does not take recarbonation into account. In practice carbonation of structural concrete elements is generally present. However, only the surface areas can be practically considered during a buildings period of use. At the end of a structure's life-cycle, a demolished concrete is stored in piles and may be used as recycled aggregates for mixing concrete or technical backfill material. As the test showed an increased natural carbonation cannot be expected, because of insufficient air flow or rather gas exchange through the

stored material. A forced carbonation process can efficiently use the crushed down material as carbon sink in a much more predictable period of time. For this reason an uninterrupted recycling process leading to the final product is possible.

The further purpose is to show if the expected differences in the carbonation behaviour of different cement types will occur during the tests. If decisive differences are seen, the findings can simplify a possible automation for the adjustment of ideal carbonation parameters.

## REFERENCES

- [1] G. Mauschitz. Emmissionen aus Anlagen der österreichischen Zementindustrie, Berichtsjahr 2018. Technical University of Vienna, 2019. [https://www.zement.at/downloads/downloads\\_2019/Emissionen\\_2018.pdf](https://www.zement.at/downloads/downloads_2019/Emissionen_2018.pdf).
- [2] T. Higuchi, M. Morioka, I. Yoshioka, et al. Development of a new ecological concrete with CO<sub>2</sub> emissions below zero. *Construction and Building Materials* **67**:338-43, 2014. <https://doi.org/10.1016/j.conbuildmat.2014.01.029>.
- [3] J. Juhart, C. Baldermann, G-A. David, et al. Erescon - Neue Betone für den Infrastruktur-Bereich, Energie- und Ressourcenoptimiert. eigen, 2015. <https://www2.ffg.at/verkehr/studien.php?id=1112&lang=de&browse=program>.
- [4] ÖNORM B 4710 - 1:2017-06-15. Austrian Standards Institute, Vienna, 2017.
- [5] H. Grube, J. Krell. Zur Bestimmung der Carbonatisierungstiefe von Mörtel und Beton. *Beton* **36**(3), 1986.
- [6] J. Gajda, F. M. Miller. Concrete as a Sink for Atmospheric Carbon Dioxide: a Literature review and estimation of CO<sub>2</sub> absorption, *Portland Cement Concrete: Portland Cement Association*, 2000. .
- [7] C. Gehlen. Probabilistische Lebensdauerbemessung von Stahlbetonbauwerken. DAFStb (= DAFStb-Heft. Band 510). Beuth, 2000.
- [8] M. Seidemann, A. Müller A, H. Ludwig. Weiterentwicklung der Karbonatisierung von rezyklierten Zuschlägen aus Altbeton (2. Phase: Prozessoptimierung im Labormaßstab und Technologieentwurf), 2015. [https://www.dbu.de/OPA/C/ab/DBU-Abschlussbericht-AZ-23689\\_02.pdf](https://www.dbu.de/OPA/C/ab/DBU-Abschlussbericht-AZ-23689_02.pdf).