



# CO<sub>2</sub> storage in cement and concrete by mineral carbonation

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## Abstract

The production of cement is responsible for about 8% of man-made CO<sub>2</sub> emissions. CO<sub>2</sub> fixation by mineral carbonation in Ca- and Mg-rich raw materials such as cement-based concrete in various stages of its lifetime and magnesium silicate-based rocks (e.g. olivine) can provide a significant and long-lasting sink of CO<sub>2</sub>. Carbonated material can be used in novel construction materials, which potentially could even be CO<sub>2</sub> negative. Despite first applications beyond pilot stage, further research is needed to reach economically and environmentally friendly processes. The properties of the novel construction materials, their reaction mechanisms, suitable mix designs, mechanical properties, and durability need to be explored further.

## Addresses

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## Keywords

Cement, Concrete, Carbon dioxide, Carbonation, Climate change.

## Introduction

Concrete is, after water, the most used material in the world and consists in its most simple form of a cementitious binder (i.e. the so-called “Portland cement”), sand, gravel, and water. The reaction of the cementitious binder with the mixing water generates hydrate phases, which cause setting and subsequent hardening of the concrete providing the mechanical properties required for structural applications. So far, no alternative materials to cement and concrete are available to meet the demands needed for the construction of infrastructure.

In 2019, 4.1 Gt of cement were produced worldwide [1]. The production of cement, which is expected to increase significantly in the future [2], accounts for about 8% of the anthropogenic CO<sub>2</sub> emissions [3] and represents the industrial process with the highest CO<sub>2</sub> emissions. As the Paris Agreement targets to limit the average global temperature increase to 2 °C compared to the preindustrial level, it is of very high relevance to significantly reduce the CO<sub>2</sub> emissions related to the production of cementitious construction materials.

Portland cement, mainly based on calcium silicates (tricalcium and dicalcium silicate) and the by far most used type of cement, is produced from a mixture of limestone and clay in rotary kilns at approx. 1450 °C. One ton of the burnt product, the so-called cement clinker, releases approx. 866 kg of CO<sub>2</sub> [4], 60–70% of it originating from the decarbonation of the limestone, 20–30% from the burning of the fuel, and the remaining part from the operation of the cement plant (i.e. electricity) [5,6]. In concrete, the largest share of CO<sub>2</sub> emissions (at least 70%) can be associated to the cement [7].

There are several mitigation strategies to lower CO<sub>2</sub> emissions related to cement production as highlighted, e.g., in [2,6,8]. The most efficient ones are currently the use of alternative nonfossil fuels and the partial replacement of cement clinker by so-called “supplementary cementitious materials” such as slags and fly ashes from industrial processes, limestone powder, or calcined clays [9,10]. However, these approaches have reached a level, where further significant improvement is difficult to achieve. The application of novel technologies such as the use of cementitious binders other than Portland cement [11,12], alternative production technologies of cement [13], or carbon capture and storage/use [14] are currently hampered by factors such as availability of raw materials or costs. Further research is needed to enable a more widespread use of such approaches.

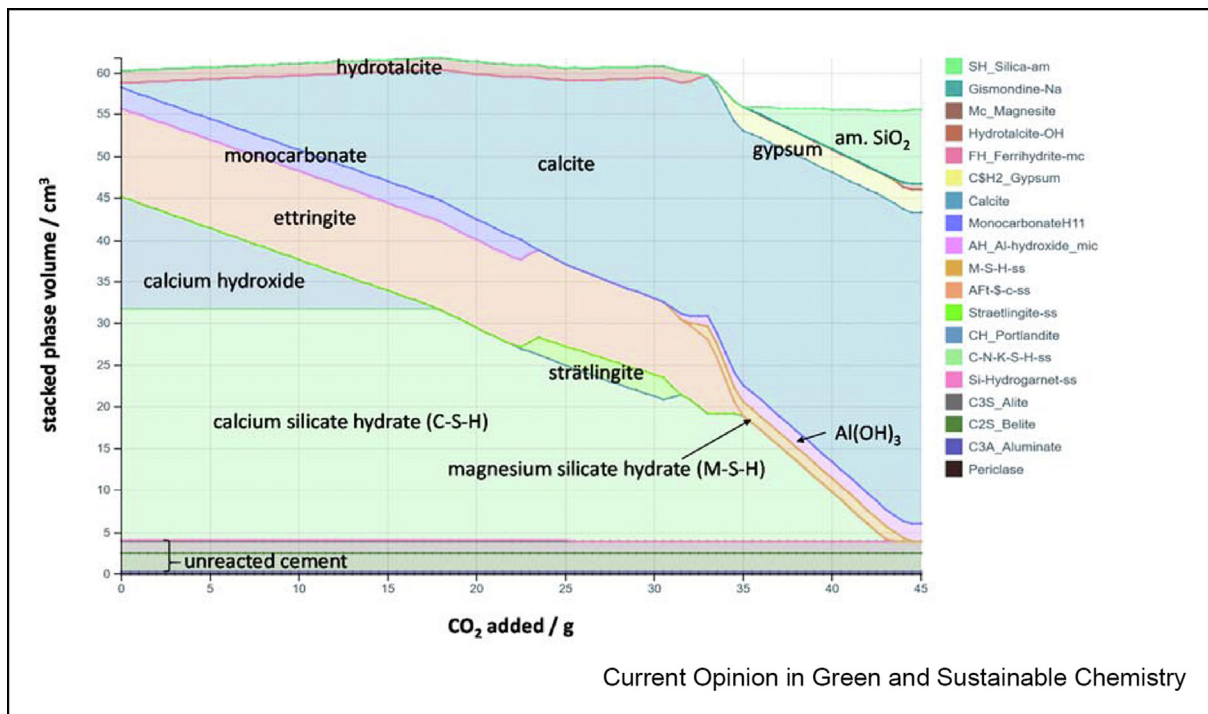
Cement and concrete offer also the possibility of CO<sub>2</sub> storage by accelerated carbonation, i.e., carbonation of old demolished concrete or of other materials containing calcium and magnesium, which could then be incorporated in new concretes. Thus, their CO<sub>2</sub> footprint can be lowered, and even CO<sub>2</sub>-negative construction materials can be achieved in certain cases. In the following, we

will highlight some recent research aspects related to CO<sub>2</sub> storage in cement and concrete.

### Natural carbonation of concrete

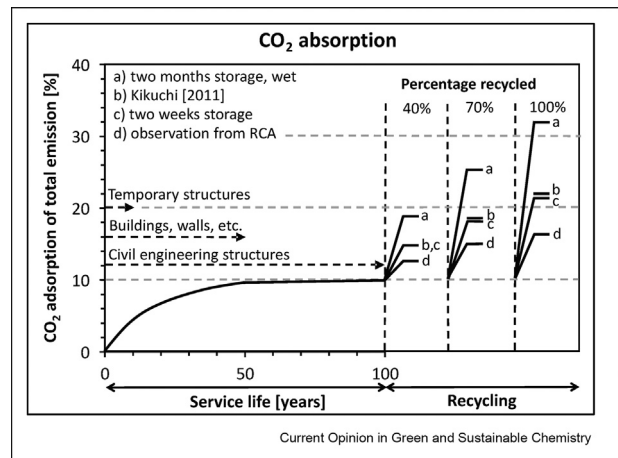
Upon reaction of Portland cement with water (“hydration”), calcium silicate hydrates form as the main strength-building phase (approx. 60 mass-% of the hydration products in a fully reacted cement). Calcium hydroxide, about 20 mass-% of the hydrates, forms as further hydration product. Its presence keeps the pH value of concrete above 12.5, thus preventing the mild steel reinforcement from corrosion. During service life, concrete takes up CO<sub>2</sub> from the environment, with CO<sub>2</sub> concentration, temperature, relative humidity, porosity, and permeability of the concrete, as well as the buffer capacity (i.e. the CaO not already fixed to carbonates or sulfates) playing a major role [15]. The phase changes due to carbonation of hydrated cement pastes can easily be calculated by thermodynamic modeling, e.g., using the cement-specific web-based code CemGEMS [16], see Figure 1. At first instance, calcium hydroxide reacts to calcium carbonate, which may be present as different polymorphs (calcite, aragonite, vaterite, amorphous CaCO<sub>3</sub>) [17]. This causes a drop of pore solution pH to a value of 9. Further carbonation leads to change in other hydrates,

Figure 1



Thermodynamic modeling of the equilibrium phase assemblage during carbonation of 100 g of a white PC (low Fe<sub>2</sub>O<sub>3</sub> content) at water/cement ratio = 0.5, 20 °C and degree of hydration 90%. The calculation was performed using CemGEMS (see <https://cemgems.org>). The undergraded cement paste is shown on the left-hand side, while moving to the right, more and more CO<sub>2</sub> (mass in grams) reacts with the hydrates. Figure modified from [16].

Figure 2



Estimated CO<sub>2</sub> absorption during the service life and recycling phase for the cement production in Switzerland in 2010 after [20]. Total CO<sub>2</sub> emission to which the results are related includes the CO<sub>2</sub> from combustion and calcination.

i.e., partial decalcification of calcium silicate hydrates [18] and decomposition of ettringite (a calcium aluminate sulphate hydrate). Finally, calcite, silica gel,

alumina gel, and gypsum form the major part of a fully carbonated paste.

The carbonation front proceeds with time from the surface to the interior lowering the pH value. When the carbonation front reaches the steel reinforcement, the latter is no longer protected from corrosion, which may result in spalling of the concrete cover in the presence of moisture. Thus, natural carbonation is an unwanted process, and the mix design of a concrete is generally made in a way to limit carbonation as far as possible or needed. In a Scandinavian study [19], it was calculated that roughly 25% of the CO<sub>2</sub>, which can theoretically be chemically bound, is taken up within 100 years, assuming a service life of 70 years and 30 years landfilling of the crushed concrete. The major part of the CO<sub>2</sub> is taken up during the latter period. A similar result was obtained for Switzerland [20], as shown in Figure 2. Although concrete has a huge potential to take up CO<sub>2</sub>, natural carbonation is very slow and incomplete. Cembureau, the European Cement Association, estimates in their targeted scenario for net zero emissions in 2050, a negative contribution of only 51 kg CO<sub>2</sub> per ton of cement by natural carbonation of construction materials in the built environment. The contribution of carbon capture and storage/use to CO<sub>2</sub> reduction including mineral carbonation is targeted to contribute with 280 kg per ton of cement to the net zero emission scenario [1].

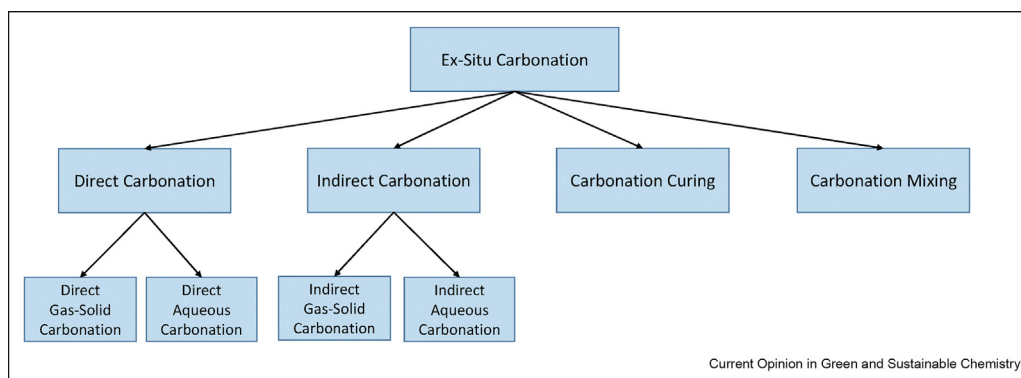
### Accelerated carbonation of concrete

Accelerated carbonation of the cementitious phases of concrete is much more viable to contribute on a short term to mitigation of climate change than natural carbonation. Carbonation to store CO<sub>2</sub> in cement or concrete can be carried out at various stages of concrete service life and beyond as highlighted in Figure 3.

At the stage of fresh concrete, CO<sub>2</sub> can be injected into concrete during mixing and batching [21,22] (“carbonation mixing” in Figure 3). Amorphous CaCO<sub>3</sub>, which is very likely nanocrystalline, forms and accelerates the reaction of the cement and enhances compressive strength. While only a small part of the CO<sub>2</sub> added is stored in the concrete, the enhancement of strength offers the possibility to lower cement content and thus to further reduce the CO<sub>2</sub> footprint of the concrete. A similar approach is the storage of CO<sub>2</sub> in concrete slurry waste, which is generated in the concrete plant during cleaning of concrete mixers and concrete trucks, and the subsequent use of the carbonated slurry as addition during the production of new concrete [23–25]. However, the amount of CO<sub>2</sub> sequestered is limited as well as the availability of this material is only 1–3% of the ready mix concrete production [26].

Carbonation curing is usually associated with strength gain, densification of the pore structure, and mitigation of chemical attack (mainly due to a decrease in permeability); however, the decrease of alkalinity may raise concerns regarding reinforcement corrosion [27]. Carbonation curing of concrete can be applied during the fresh and the hardened state and generally comprises of pre-curing, carbonation, and post-curing steps [26–29]. Pure cement hydrate phases such as calcium silicate hydrates, calcium hydroxide, or ettringite in the form of fine powders carbonate the fastest at high relative humidities [30]. In the case of larger samples or pieces of concrete, porosity, permeability as well as relative humidity, the state of drying and the degree of water saturation inside the concrete pore system play a major role. In addition, other calcium silicate minerals such as  $\gamma$ -dicalcium silicate, rankinite, and wollastonite, which are not reactive with water under alkaline conditions, and therefore not used for conventional cements, can be used

Figure 3



Classification of ex-situ carbonation technologies as summarized in and modified from [34]. Note that in-situ carbonation (injection of CO<sub>2</sub> in silicate-rich geologic formations followed by carbonation) is beyond the scope of this review.

to prepare CO<sub>2</sub>-cured construction materials [31–33]. Accelerated carbonation has the potential to bind substantial CO<sub>2</sub>. However, the extent of the CO<sub>2</sub> binding depends on the amount of CaO available in the cement, which generally has been produced by burning CaCO<sub>3</sub>. Thus, the positive impact of carbonation curing to lower the global warming is mainly due to use of less cement and thus due to the avoided concrete production [34]. The use of such carbonated cured cements and their contribution to lower CO<sub>2</sub> emissions on a global scale seems appears to be a challenging task [35].

Waste concrete derived from demolished buildings makes up a significant source of carbonatable material. Compared to other recycling options for the cementitious part of concrete debris [36], carbonation offers in addition the possibility of long-term storage of CO<sub>2</sub> in mineral phases providing a negative emission value chain as highlighted in [37,38]. Different approaches are described in literature as reviewed, e.g., by [26,34,39–44] using various starting materials such as recycled aggregates (gravel fraction), recycled concrete fines (sand fraction), or separated cement paste. Either a gas-solid or an aqueous carbonation process can be applied in a direct or indirect (at least two process steps) way.

In the gas-solid process at ambient or enhanced relative humidity ( $\leq 95\%$ ), a direct interaction of CO<sub>2</sub> with the cement paste adhering to the recycled aggregates (gravel or fines) via a through-solution mechanism is followed by the precipitation of calcium carbonate on the surface of the aggregates [45]. Recycled aggregates treated in this way were found to show significantly less porosity and less water absorption compared to an uncarbonated aggregate [45]. Thus, the properties of the concrete prepared with the carbonated material can be superior to those with conventional recycled aggregates in terms of mechanical properties and durability [46]. This also enables reducing the cement content in the concrete at equal performance, thus further reducing the CO<sub>2</sub> footprint [47].

In the aqueous carbonation process a wide range of process conditions (i.e. water/solid ratio, CO<sub>2</sub> concentration, pressure, temperature, reagents used) are reported, see e.g., [39,48–51]. Aqueous carbonation is more complete than under dry conditions, as calcium ions are leached out from the crushed concrete. Besides portlandite, other phases, i.e., calcium silicate hydrates and the calcium aluminate hydrates, are also subjected to carbonation like in natural carbonation. However, the degree of carbonation is more pronounced with a complete decalcification of calcium silicate hydrate [18]. As a result, not only CaCO<sub>3</sub> but also an increasing amount of silica and alumina gel is formed compared to carbonation at natural CO<sub>2</sub> concentrations. These gels were found to act as a pozzolan [50], when the

carbonated material is later used a mineral addition in newly produced concrete, thus allowing to replace a portion of the cement. Accelerated carbonation of waste concrete offers thus a large potential of long-term storage of CO<sub>2</sub> on global scale in comparison with natural carbonation occurring during landfilling, where only 25% of the CO<sub>2</sub> binding potential is exploited [19,20].

### Mineral carbonation of other materials and their use in construction products

Besides (recycled) concrete, other materials can also be used in mineral carbonation processes. Potential sources from waste streams are Ca compounds prone to carbonation such as cement kiln dust [25] as well as slags and ashes from various processes [52] including steel slags [53]. Other suitable materials for mineral carbonation are natural Ca or Mg minerals such as wollastonite [54] and Mg silicates such as forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) or serpentine (Mg<sub>6</sub>Si<sub>4</sub>O<sub>10</sub>(OH)<sub>8</sub>) [55,56]. Especially, Mg silicates are available in large quantities, which may be used for binder production in regions where limestone and clay are scarce [57]. Furthermore, other Mg sources may be used such as rejected brines from desalination plants, which can provide concentrated Mg solutions for carbonation [58–61]. A recent example of a successful carbon capture and utilization process was the carbonation of Mg-rich brines to produce plaster-like products from thermally activated nesquehonite (MgCO<sub>3</sub>·3H<sub>2</sub>O) showing self-cementing properties when mixed with water [60].

In general, such created magnesium carbonates can be either used as permanent CO<sub>2</sub> sink or as raw material to produce MgO. Conversion of Mg-silicates and Mg-rich brines through carbonation and subsequent calcination to MgO is a process, in which no fossil-bound CO<sub>2</sub> (excluding potential combustion of fossil fuel for energy production) is released. Thus, binders based on magnesium oxide derived from magnesium silicates (MOMS) offer a possibility to produce raw material for a binder with a reduced carbon footprint [12]. Moreover, due to the high CO<sub>2</sub> absorption ability of MgO (1.09 metric tons per metric ton of MgO [62]), it is theoretically possible to produce a construction material, whose CO<sub>2</sub> uptake outweighs CO<sub>2</sub> emissions produced during binder manufacture.

MgO can be utilized as raw material for several Mg-based binders; the most prominent being Mg phosphate cements (MPC), Mg oxysulfate cements (MOS), Mg silicate hydrate (M-S-H) cements, and Mg oxychloride (MOC) cements [63,64]. A Mg binder developed under the former tradename Novacem® [62] was perhaps the first attempt to create “carbon-negative” cements by a low-carbon raw material production route and the subsequent natural carbonation of the hydrated binder during its service time. The product

contained MgO and basic Mg carbonates, which hydrate to a mixture of brucite and amorphous magnesium carbonate hydrates [65,66]. Upon natural or accelerated carbonation, brucite is converted into various hydrated magnesium carbonates depending on carbonation conditions and minor additions present [67,68].

Such binder compositions may reach a compressive strength almost similar to the one of Portland cement (Figure 4). The binder formulation was later extended to include other mineral additives such as SiO<sub>2</sub> (M-S-H cement) and/or Al<sub>2</sub>O<sub>3</sub> bearing industrial by-products or natural inorganic materials, thus providing higher mechanical properties. However, economical production remains the key obstacle for implementation of such novel binders to this day [64].

## Conclusions and outlook

Mineral carbonation offers a possibility to sequester large quantities of CO<sub>2</sub> in basic calcium and magnesium compounds. Possible resources are fresh concrete (carbonation curing), recycled concrete from demolished buildings, by-products from industry such as cement kiln dust, steel slags, or municipal waste incineration slags and ashes or naturally occurring rocks. The use of the carbonated materials in construction products (i.e. cement and concrete) offers a huge sink to store CO<sub>2</sub> and a significant contribution to lower their CO<sub>2</sub> emissions and the circularity of their constituents. The CO<sub>2</sub> storage by mineral carbonation is stable on a long

term as demonstrated by all the carbonatic rocks occurring on Earth since many millions of years.

In order to mitigate climate change fast enough, accelerated carbonation is highly favorable over natural weathering of, e.g., concrete or rocks. Currently, various technologies related to different materials and different processes are increasingly explored, which is on the one hand depicted by the strongly increasing numbers of scientific papers in this field. On the other hand, the first industrial applications are beyond pilot stage, for instance [70,71] in Switzerland. The feasibility and environmental impact of the various approaches are projected differently depending on the literature source. For instance, in a meta-study on life cycle assessment [34], direct aqueous carbonation, carbonation mixing, and carbonation curing were found to be most promising, while [26] highlights a holistic approach of the various carbonation technologies in combination with concrete recycling in the context of circular economy. Such an approach can provide a significant sink for CO<sub>2</sub> in the future [37]. Thus, detailed life cycle analyses (cradle to grave/cradle to cradle) as well as economic assessments of the various technologies are crucially needed.

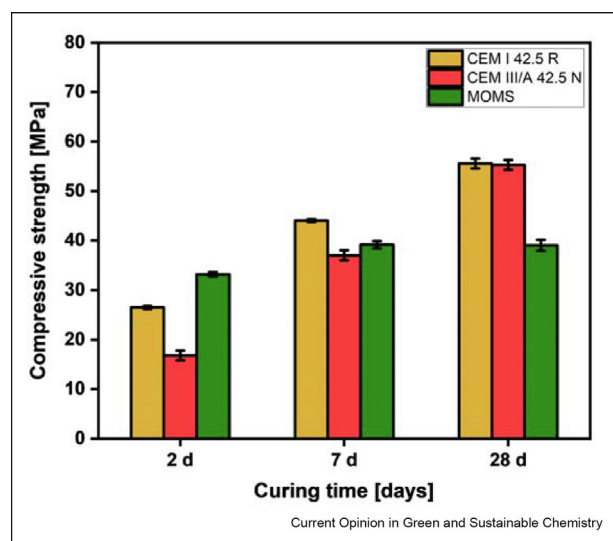
Mineral carbonation of magnesium silicate-based rocks has a significant potential, as available olivine resources are sufficiently abundant to sequester all anthropogenic emissions during the next >1000 years [57]. However, this would require huge mining activities, as the carbonation of approx. 100 billion tons of olivine would be needed annually. This amount is approximately three times higher than even the annual worldwide concrete production. Nevertheless, the carbonation of magnesium-based rocks and brines do not only offer the sequestration of large amounts of CO<sub>2</sub> but also the possibility to manufacture novel, CO<sub>2</sub>-negative construction materials, which could replace the conventional Portland cement in certain applications. For this purpose, large-scale industrial carbonation processes are needed, which are economically viable.

For all the novel materials incorporating sequestered CO<sub>2</sub>, i.e. concretes containing carbonated materials and especially the radically new binders based on magnesium carbonates, further fundamental research is needed to enable a widespread application. Such research includes not only the carbonation processes but also the properties of the novel construction materials in terms of hydration/carbonation mechanisms, concrete mix design, mechanical properties (such as strength, elastic modulus, shrinkage, creep), and durability (i.e. reinforcement corrosion).

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships

Figure 4



Compressive strength of CEM I, CEM III, and MOMS mortars after 2, 7, and 28 d of curing in water. Mortars were cast following EN 196-1 using a water-to-cement ratio of 0.5. Unhydrated MOMS binder contained 90 mass-% reactive MgO/10 mass-% hydromagnesite ( $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$ ), and a polycarboxylate ether superplasticizer was used to achieve a workable mortar. Unpublished data obtained at Empa within the PhD thesis of A. German [69].

that could have appeared to influence the work reported in this paper.

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\* of special interest

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