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Review

Carbonation of cement paste: Understanding, challenges, and opportunities


 Branko Šavija^{a,*}, Mladena Luković^{b,1}
^a Microlab, Faculty of Civil Engineering and Geosciences, Delft University of Technology, Stevinweg 1, 2628 CN Delft, The Netherlands

^b Concrete Structures Group, Faculty of Civil Engineering and Geosciences, Delft University of Technology, Stevinweg 1, 2628 CN Delft, The Netherlands

HIGHLIGHTS

- A systematic review of literature related to mechanisms of carbonation in cement paste is given.
- Active uses of carbonation for improvement of cement based materials are also discussed.
- The study provides a starting point for research of carbonation in new cementitious materials.
- It can be of use for development of new active carbonation procedures which utilize and capture CO₂.

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ABSTRACT

Cement paste is known to react with atmospheric carbon dioxide. Carbonation of cement paste has long been recognized as one of the causes of reinforcement corrosion. On the other hand, carbonation causes numerous chemomechanical changes in the cement paste, most notably changes in strength, porosity, pore size distribution, and chemistry. Furthermore, it can cause shrinkage and cracking of the cementitious matrix. The present review summarises the state of the art regarding the understanding and consequences of carbonation of cement paste. Apart from the passive process of reaction of atmospheric CO₂ with cement paste, carbonation is sometimes used on purpose in order to improve certain properties of cementitious materials. This review further summarises recent efforts regarding active use of carbonation as a tool for manipulating certain properties of cement based materials. Possible fields of application include accelerated curing, improvement of fibre reinforced cementitious composites, concrete recycling, and waste immobilization.

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* Corresponding author.

E-mail addresses: b.savija@tudelft.nl (B. Šavija), m.lukovic@tudelft.nl (M. Luković).

¹ Formerly at the Microlab, Faculty of Civil Engineering and Geosciences, Delft University of Technology, Stevinweg 1, 2628 CN Delft, The Netherlands.

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1. Introduction and background

Carbon dioxide from the atmosphere can react with hydrated cement in presence of moisture [1]. This reaction affects both the cement microstructure and durability of (reinforced) concrete. Although the aim of this paper is to systematically review the former, the latter is first briefly discussed.

In the past, a lot of research has been devoted to studying effects of carbonation on durability of reinforced concrete. In concrete, reinforcing steel is protected from active corrosion by a passive film of ferrous oxide which forms on its surface in the highly alkaline concrete pore solution [2,3]. This alkalinity, corresponding to a pH value above 12.5, is provided by dissolution of solid CH (calcium hydroxide, i.e. portlandite) in the pore solution. Carbonation of hydrated Portland cement paste reduces the pH of the pore solution to values lower than 9 (as low as 8.3) [4]. This is lower than the depassivation threshold of reinforcing steel, which is around 9.5 [5,6]. Thus, if the front of low pH reaches the surface of reinforcing steel, the protective oxide film on its surface breaks down. Corrosion then initiates, provided that moisture and oxygen are present [7]. It is generally acknowledged that carbonation results in generalized corrosion, with uniform pressure being exerted on the surrounding concrete and resulting in cracking of the concrete cover [8–12]. Corrosion of reinforcing steel is well recognized as a problem, and is not a focus of this review.

Reaction of carbon dioxide with calcium bearing phases in the cement paste can also cause chemo-mechanical changes in the microstructure. It is commonly assumed that the carbon dioxide diffuses through the concrete cover following a square root of time relation, resulting in a carbonation front [13–15]. However, some studies show that the paste does not become completely carbonated on one side of a sharp reaction front but rather that a zone of partially carbonated material can exist to a depth substantially greater than the depth of full carbonation (i.e. that indicated by phenolphthalein spraying) [16–18]. On the contrary, SEM observations showed (under supercritical carbonation exposure, as explained later) a series of carbonation and dissolution fronts, which progressively translate with time from the outside towards the inside of the sample [17]. They suggested that the affected HCP consists of a carbonated zone, a carbonation front, a dissolution front, and the inner part of the sample (Fig. 1). With increasing exposure, this geometry moves inwards, and relicts of these fronts are observed backwards in the carbonated zone.

Apart from causing the pH drop, most notable changes are changes in porosity, (micro)mechanical properties, and appearance of cracks [19]. These changes can be considered *passive*, because they occur spontaneously when cement paste reacts with carbon dioxide. Therefore, they are commonly considered as deterioration (or ageing) effects, even though some properties of certain cement pastes can be actually improved, as discussed later. On the other hand, there has been a number of recent advances aiming at utilizing the ability of cement paste to react with carbon dioxide to achieve certain economical or environmental benefits (or both). This can be considered as an *active* decision to take advantage of this reactivity.

In short, a passive carbonation process is when carbonation causes unwanted and unplanned changes in the surface layer of a concrete structure or an element when exposed to the environment. Even though passive carbonation can sometimes even result in improvement of certain properties (as elaborated later), it is generally considered as an ageing mechanism. A majority of studies leading to fundamental understanding of chemomechanical mechanisms of carbonation in cementitious materials are based on studying passive carbonation (e.g. using accelerated carbonation experiments with elevated concentrations of CO₂), and are thoroughly reviewed in Sections 4. On the other hand, an active carbonation process results from a procedure designed to intentionally take advantage of the ability of calcium bearing phases of cement paste to react with CO₂ for different applications. Active carbonation assumes controlled exposure of cementitious materials to elevated CO₂ concentrations for certain periods of time, resulting in benefits in terms of mechanical performance or environmental impact of the material. Different procedures which use active carbonation are reviewed in Section 5. Passive and active effects of carbonation are summarized in Fig. 2, and discussed further in detail.

In order to understand the passive and (possible) active effects of carbonation on cement paste, it is important to understand the way that CO₂ reacts with main phases in the cement paste and the factors which influence this process. This is discussed in the following section.

2. Carbonation processes of main cement phases

2.1. Portlandite carbonation

It is commonly assumed that calcium hydroxide (CH, i.e. portlandite) is the hydration product which reacts most readily with CO₂ [1,18]. Carbon dioxide can react with dissolved calcium hydroxide, resulting in precipitation of sparingly soluble calcium carbonate in the pore space [15,20,21]:



Carbonation of CH involves three main steps [20]: (1) dissolution of CH; (2) absorption of carbon dioxide and formation of carbonate ions; and (3) chemical reaction and precipitation.

Chemical reaction of CO₂ with Ca(OH)₂ starts instantly and leads to formation of calcium carbonate [22]. In the early stage of carbonation, CO₂ diffusion through the already carbonated layer (step 2) is the rate-controlling step for carbonation of CH [15,23]. Even though CO₂ diffusivity is significantly higher in gas filled pores, compared to water filled pores, the presence of moisture is necessary for gaseous CO₂ to react with CH [24]. The limiting step changes at later stages of the carbonation process as the rate of CH carbonation decreases due to the formation of a thin layer of calcium carbonate on the surface of CH crystals [16,18,22,25–27]. Calcium carbonate crystals form at the CH surface apparently at random (Fig. 3), but probably originating at dislocations (Fig. 4). Eventually the surface becomes covered as the nucleation sites

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