



Carbon sequestration in concrete sidewalk samples



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HIGHLIGHTS

- Levels of CO₂ sequestration in interior portions of concrete pavements.
- Spatial distribution of carbonation levels.
- For 120 mm thick sidewalk samples, carbonation percentage = 25.4% for interior specimens.
- For 160 mm thick sidewalk sample, carbonation = 28.2% for interior specimens.

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ABSTRACT

Concrete can sequester CO₂ from the air in a reaction commonly called carbonation. High levels of carbonation near the surface of concrete samples have been widely studied since they may lower the pH of the concrete and alter the characteristics of a concrete structure if they migrate deeper into the structure. However, little information is available about the levels of CO₂ sequestration in interior portions of concrete pavements. This study focuses on the spatial distribution of carbonation levels across the thickness of concrete sidewalk samples that are many decades old. For 120 mm thick sidewalk samples, the average carbonation percentage was found to be 80% for specimens located near the surfaces, and 25% for specimens located in the inner part of the samples (20–80 mm from the top surface). For a 160 mm thick sidewalk sample, the average carbonation was 28% for all interior specimens at least 20 mm from surfaces. These interior levels for various concrete pavements at various ages can be used together with surface carbonation information to estimate CO₂ sequestration in concrete pavements.

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1. Introduction

One of the expected contributors to climate variability is the “greenhouse effect” produced by a group of gases referred to as “greenhouse gases” including carbon dioxide (CO₂). About 5% of the annual anthropogenic global carbon dioxide emissions are produced by the cement industry and a substantial portion is from the chemical reaction called calcination [1]. Cement is an important component in concrete which is used throughout the world for pavements, buildings and other infrastructure items. However, these structures made with cement can reabsorb some of those emissions over time, reversing a portion of the calcination reaction in a process referred to as carbonation. In order to understand the

overall carbon footprint of a structure made with concrete, it is important to determine how much carbon the different types of concrete can absorb, based on many parameters.

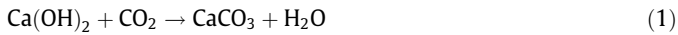
There are various chemical reactions that occur during the calcination and carbonation processes and these can also vary with different chemical compositions in cement and concrete. In Ordinary Portland Cement (OPC), which is used in most concrete applications, calcium oxide (CaO) is a major component. It has been formed by the high temperature dissociation of calcium carbonate (CaCO₃), as found in rock, to CaO and CO₂. In order to produce concrete, cement is mixed with water, aggregates, usually additional supplementary cementitious materials (SCMs) and various small amounts of chemical admixtures. The hydrated cement is what binds the aggregates together. When the cement is hydrated, calcium hydroxide Ca(OH)₂ is one of the compounds formed. Hydrated cement also has a complex calcium–silicate–hydrate (C–S–H) phase, in addition to tri-sulfates such as ettringite (AFt), calcium aluminate monosulfates (AFm) and other minor phases [2].

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The carbonation reaction occurs when CO₂ is sequestered into the concrete along with some water and the carbonation reaction occurs to form calcium carbonate. Some of this might occur from carbon dioxide mixed into the concrete initially, but this process also occurs slowly over time as carbon dioxide diffuses into the porous concrete matrix. Both the calcium hydroxide and the C–S–H phase can carbonate. A simplified summary of the carbonation process in the calcium hydroxide phase can be represented by Eq. (1) [3].



There are also some other residual components in concrete mortar such as aluminum (Al³⁺) and iron(III) (Fe³⁺) that are not significantly involved in the carbonation process due to not commonly complexing with carbonates [4], or magnesium (Mg²⁺) and iron(II) (Fe²⁺) that are not considered as significant due to their relatively low concentrations [3].

Many parameters can influence the amount of carbonation or carbon sequestration in concrete such as the cement's composition, the environment, the type of concrete, and time. As the carbonation process continues over time, calcium carbonate is formed which is a larger molecule than calcium hydroxide and the available pore microstructure decreases for diffusion of the carbon dioxide into the sample as time progresses [5]. The water available is one of the most important parameters. Most researchers assume that the carbon dioxide from the air must first dissolve into water before reacting with the calcium hydroxide to form the carbonate. Nevertheless, if the humidity is too high, the porous system can be blocked off by condensed water so the carbonation could be obstructed [1,3,5]. A higher CO₂ concentration in the environment that the concrete is exposed to might accelerate the carbonation process [6]. The water/cement ratio (w/c) is the initial mass ratio of water to cement in the concrete. Typically lower w/c's have lower rates of carbonation since a concrete with a lower w/c ratio may be a denser concrete since less extra water in the mix means there is less water to evaporate originally and thus fewer micropores.

Most of the research on the carbonation of concrete focuses on the front or depth from the surface where the amount of carbonation is progressively sufficient to cause a color change in a pH indicator test [1,3,5]. This phenomenon is a result of the reaction in Eq. (1) where the highly basic calcium hydroxide is converted to the less basic calcium carbonate. This carbonation front has been shown to slowly migrate from the surface of a concrete sample exposed to carbon dioxide and water (such as the humidity in the air) and is estimated to reach levels of about 75% carbonation based on the calcium available. Crushing concrete after a structure's service life increases the surface that is available to allow CO₂ to diffuse into the concrete, therefore, increasing the carbonation level [1].

However, there can be additional carbon sequestration beyond this front. Previous laboratory tests on cement samples exposed to the atmosphere have shown that there are significant amounts of calcium carbonate formed well beyond the carbonation front, and in many cases at a fairly constant level throughout the remainder of the sample [7,8]. For hydrated cement paste samples made with OPC and water, the average interior levels of carbon dioxide sequestration based on moles of CaO in the samples was found to be approximately 25% in just a few years unless otherwise inhibited by low water to cement ratios or additional water saturation [8]. This previous work proposed an interior model of a quasi-steady state equilibrium of sequestration levels applicable to specific mixes, environmental exposures and fairly constant throughout the interior of a sample over long periods of time such as years. These fairly constant levels could then be compiled into isopleths (graphs of similar conditions) of interior carbon dioxide sequestration for certain conditions and timeframes. This simplified interior

sequestration method, along with the results of carbonation front research, might together provide a more comprehensive estimate of the overall carbon dioxide sequestration level of concrete for various applications at certain ages [8]. Note that this previous laboratory work was on hydrated cement paste, which has been shown by other researchers to have a significantly faster rate of carbon dioxide diffusion than concrete [9]. It is therefore expected that the time frames associated with quasi-steady state equilibrium conditions of sequestration in concrete pavement will be longer than those for similar levels for the hydrated cement paste analyses. The hypothesis is that concrete pavement samples exposed to carbon dioxide and water will have interior carbon dioxide sequestration levels that may be estimated using the quasi-steady state equilibrium (isopleths) method for certain extended periods of time such as years or decades. At similar earlier time frames, these levels in the pavements may be less than the previous laboratory work on hydrated cement paste [8] due to the differences in carbon dioxide diffusion in cement versus mortar, but may still be significant and might increase to the higher levels with time [9]. This research extends the work on interior carbon dioxide sequestration from the laboratory work on hydrated cement paste samples to actual concrete pavement samples. It is predicted that concrete pavements in regular use re-absorb significant amounts of carbon dioxide with respect to the calcium content of the cement used in its production. The absorption is more prevalent near or at the pavement surfaces, but is still measurable throughout the pavement. Therefore evaluations of concrete pavement samples can be used to estimate total re-absorption during primary use, combining both the carbonation front research for near surface portions of a sample, with the quasi-steady state equilibrium method for the remaining interior portions of the sample.

There were two objectives of the research. The first was to examine the surface levels of carbon dioxide sequestration on various concrete pavement samples, both surfaces that had been exposed to ambient (air or soil) conditions in their primary use for many decades and also broken surfaces which had been exposed to the air for a few years after primary use. The second objective was to map the interior carbon dioxide sequestration levels in the concrete pavement samples to determine if they had reached a quasi-steady state equilibrium that was fairly consistent throughout the interior.

2. Methods

Carbonation front research is typically performed with a pH indicator method which provides depth information to the location of the front where the pH drop is noticeable [1,5]. Thermal gravimetric analysis (TGA) is another method used to estimate the actual amounts of CO₂ absorbed in a specimen. In the TGA process small mortar or cement specimens taken from a sample are heated and several physical and chemical processes occur. The most significant processes/steps which occur during the TGA heating process are depicted in Fig. 1 and can be summarized as follows: (1) free or loosely bound water is released from the concrete between 50 and 300 °C, (2) the calcium hydroxide is dehydrated forming water and calcium oxide during the dissociation reaction which typically occurs between 310 and 520 °C, and (3) at higher temperatures, calcium carbonate dissociates and carbon dioxide is released [10]. Note that different forms of calcium carbonate do not dissociate at the same temperature range: Calcite CaCO₃ dissociates between 650 and 950 °C, while other CaCO₃ species dissociate between 520 and 750 °C [11,12].

This process is referred to as TGA since as the specimen heats, the mass of the specimen decreases as compounds are released such as water and carbon dioxide, and the mass losses are measured and used to estimate the amounts of water and carbon dioxide released over the three main steps. TGA is the method used in this study as it can provide carbon dioxide sequestration information for specimens taken at various locations from a sample. The TGA instrument machine has a thermal analyzer which portrays the thermogravimetric curve, and the derived thermogravimetric curve. Each process/step is characterized by its own temperature range which is defined by the edges of the characteristic peaks of the derived thermogravimetric curve. The TGA instrument was set at a ramp rate of 20.00 °C/min up to 1000.00 °C in a nitrogen environment. In Fig. 1 the top curve is the mass loss, and the bottom curve is its derivative. The left peak in the derivative curve represents loosely bound water release, the small peaks in the center are water from

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