



Experimental study on effects of CO₂ concentrations on concrete carbonation and diffusion mechanisms



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HIGHLIGHTS

- Five different CO₂ concentration levels were used for the concrete accelerated carbonation study.
- Dense carbonated outermost layer reduces the rate of CO₂ diffusion in concrete.
- Diffusion mechanisms of CO₂ into concrete under high and low CO₂ concentration levels are different.
- An concrete accelerated carbonation test with 20% CO₂ concentration to predict natural carbonation is not appropriate.

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ABSTRACT

In this paper, concretes of different grades were exposed to five different CO₂ concentration levels (2, 10, 20, 50 and 100% by volume) using accelerated carbonation test methods. The relationship between the depth of concrete carbonation and CO₂ concentration was investigated. The diffusion mechanisms of CO₂ gas in concrete were analysed through the scanning electron microscopy (SEM) observations. Experimental results showed that, under the same carbonation age, the increase of carbonation depth was found to be high when concrete samples were exposed to low CO₂ concentrations (i.e. 2–20%); and low when the samples were exposed to high CO₂ concentrations (i.e. 50–100%). The reason for this is mainly due to the microstructure change caused by concrete carbonation, which makes the surface layer denser thus lowering the rate of CO₂ diffusion into concrete. SEM results showed that carbonation decreased the porosity of concrete by clogging up the pores and reducing the pore sizes. It is believed that the diffusion mechanisms of CO₂ into concrete under high and low CO₂ concentration levels would be different.

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1. Introduction

Carbonation occurs in concrete because the free hydroxides (mainly calcium hydroxide (Ca(OH)₂) in concrete react with carbon dioxide (CO₂) from air or water to form calcium carbonate [1,2]. Owing to the consumption of hydroxides the pH value of concrete pore solution reduces to below 9.0. Normal carbonation results in a decrease of porosity making the carbonated paste stronger. Carbonation is therefore an advantage in non-reinforced concrete. However, it is a disadvantage in reinforced concrete (RC) because it decreases the pH in concrete pore solution. It is known that rebar in concrete is protected against corrosion by a passive film that is stable due to the high alkalinity of the pore solution. Corrosion may

start once the protective passive film on the rebar surface breaks down (depassivation) due to the carbonation processes [3,4]. In general, cement paste contains 25–50 wt.% calcium hydroxide, which means that the pH of fresh cement paste is usually at least 12.5. The pH of a fully carbonated paste however is about 7. When the pH value of concrete pore solution drops to about 8.5–9.0, the passive film will be completely damaged and the protection to the rebar will be destroyed and the durability of RC structure will be significantly affected [5,6].

Carbon dioxide concentration is increasing due to fossil energy consumption and the damage of RC structures due to carbonation is reported to increase due to increasing CO₂ concentration. Concrete carbonation should be carefully considered in the durability design of RC structures especially those in metropolitan cities where the rise in CO₂ concentration is accelerating. The straight cause of carbonation is the presence of CO₂ in the surrounding

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environment of concrete. Carbonation starts with the diffusion of dissolved CO₂ molecules in pore water of concrete and then the formation of calcium carbonate with calcium hydroxide and calcium silicate hydrates (C–H–S) [7,8]. The diffusion of CO₂ in concrete depends on a number of factors, such as CO₂ concentration, porosity and moisture content of the concrete. The carbonation process starts at the surface and penetrates slowly to the interior of the concrete and the depth of carbonation is considered to be as one of the most important factors in assessing the durability of concrete structures.

Up to now, the only reliable data related to carbonation properties of concrete are those obtained in natural environment where concrete samples are exposed to real natural conditions over a long term. This method, however, is very time consuming, normally taking a few years. Therefore, various accelerated carbonation test methods have been developed, using which carbonated samples can be obtained in a short time and can be served as the basis for predicting the carbonation behaviour of concrete under real atmospheric environments. Note that, the CO₂ concentrations used in these test methods are significantly varied among countries. For example, the CO₂ concentration of 2% value is specified in the report of DURACRETE [9], whereas it is 20% as prescribed in GB/T 50082-2009 [10]. The European standard EN 13295 [11] claims that under certain testing conditions (i.e. 1% CO₂, 21 ± 2 °C, relative humidity 60 ± 10%), the carbonation products formed would be similar to those observed in naturally carbonated specimens. Bernal et al. [12] stated that the CO₂ concentration levels used in an accelerated method should be in the range of 1.0–3.0% by volume so that the carbonation process under natural environment can be revealed. When concrete is exposed to high CO₂ concentration levels, the resulting carbonated concrete may be different when compared to that formed under the natural environment.

Significant research progress has been made since 1980 on concrete carbonation and its impact on the corrosion of reinforcing steel. However, there is little research on the relationship between the CO₂ concentration level and the mechanism of carbonation in concrete. Rozière et al. [13] studied the resulting concrete samples exposed to a high CO₂ concentration (50% by volume) using the accelerated method and compared the results with the samples exposed to natural environment. Their results indicated that there was no significant difference in carbonation depths between the two sets of samples and the relationship between the high CO₂ concentration and mechanism of carbonation has not been discussed.

According to Fick's first law of diffusion, the larger the CO₂ concentration gradient, the quicker the CO₂ gets into the concrete and the faster the carbonation reaction would be. In this paper, different CO₂ concentration levels on the mechanism of concrete carbonation and the depth of concrete carbonation were investigated experimentally. The purpose of the present study is to characterise the relationship between the carbonation depth and CO₂ concentration and the microstructure change of the carbonated concrete and its effect on the diffusion of CO₂ in carbonated concrete.

2. Experimental

2.1. Materials

Ordinary Portland cement conforms to BS 12 was used (28 day compressive strength is 54.2 MPa). The specific gravity and specific surface area of cement were 3.4 g/cm³ and 3950 cm²/g, respectively. The chemical composition of cement is given in Table 1. The fine aggregate used was natural river sand having a fineness modulus of 2.4. The coarse aggregate used was graded gravel with a maximum size of 30 mm. Three concrete mixes with different

Table 1
Chemical composition of cement expressed as oxides (wt.%).

Oxides (wt.%)	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	K ₂ O	MgO	TiO ₂	Si/Ca
OPC	64.6	21.10	5.90	3.10	-	1.00	-	0.327

water-to-cement ratios were investigated. Table 2 shows the concrete mix details. The concrete was mixed first, then cast, and demoulded, Afterwards it was cured for 28 days in a controlled environment before the accelerated carbonation test.

2.2. Specimen preparation and testing

For each concrete mix, 85 cubic specimens of side length 100 mm were cast. Five of them were used for the determination of compressive strength according to the GB/T50081-2002 [14]. The remaining 75 samples were sub-divided into five groups and exposed to different CO₂ concentration levels (2, 10, 20, 50 and 100% by volume) using the accelerated carbonation method (20 °C and 70% RH) according to GB/T50082-2009. The depths of concrete carbonation at 7, 14, 28, and 56 exposure days were measured by spraying 1% phenolphthalein indicator.

Mercury intrusion porosimetry (MIP) tests were conducted to obtain the porosity of samples of carbonated and uncarbonated concretes. In this study, we used an AutoPore IV 9500 (Micrometrics) porosimeter capable of producing up to 33 × 10³ psi. The pores were modelled as cylindrical channels and the test pressure was linked to their radius by the Washburn equation.

The microstructure of carbonated concrete after exposed to different CO₂ concentrations were examined using JSM 5910LV Scanning Electron Microscope (SEM) with an acceleration voltage of 15–20 keV and working distance of 5–9 mm. Small strips (25 mm × 25 mm) were first cut out from the surface of carbonated concrete cubes using a diamond saw. Then the samples were cut into thin sections using a thin-bladed low-rpm precision circular saw. To obtain a high quality smooth surface, a rotary disc equipped with silicon carbide papers (#1200 and #2000) was used to polish the samples. The samples were then immersed in acetone to stop further hydration in the surface zone and cleaned using an ultrasonic vibrator. Finally, the specimens were vacuum dried and coated with a thin layer of gold. Each sample was photographed at a magnification of 1000, 5000, 10,000 and 20,000 times accordingly.

3. Results and discussion

3.1. Carbonation depth

Figs. 1–3 show the results of carbonation depth of G30, G40 and G50 with different CO₂ concentration levels at different test ages, respectively. Based on CO₂ concentrations used in this study, four CO₂ concentration change zones can be identified (see Fig. 1); they are zone ① for 2–10% CO₂, zone ② for 10–20% CO₂, zone ③ for 20–50% CO₂, and zone ④ for 50–100% CO₂.

From Fig. 1, it can be observed that the concrete carbonation depth increased with increased CO₂ concentration but the significance is reduced when the CO₂ concentration is higher than 20%. The increase rates in different zones are also different. In Fig. 1, the arrow lines display the slopes of 56d carbonation depth lines for different CO₂ concentration zones. The slope of zone ① (2–10%) is the biggest. It indicates that when CO₂ concentration increased from 2% to 10%, increase of concrete carbonation depth is the most obvious. The slope of zone ② (2–10%) is smaller than that of zone ①. The slope of zone ③ reduces greatly, and the slope

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