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Degradation of fly ash concrete under the coupled effect of carbonation and chloride aerosol ingress

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1. Introduction

Reinforced concrete structures face the problems of performance deterioration during their designed service life, due to the penetration of harmful chemical substances from the surrounding environments. Many existing studies have investigated the deterioration mechanism of concrete materials caused by a single item such as chloride attack or carbonation and its corresponding influence on the durability performance of concretes [1–6]. However, most of reinforced concrete structures are subjected to combined attacks of multiple items in the field where the structures are exposed. For instance, structures built in coastal regions are often subjected to carbonation and chloride arracks. The corrosion of reinforcing steel due to the combined attack of carbonation and chlorides is different from that due to carbonation or chloride attack alone. Concrete carbonation is a neutralization reaction between the carbon dioxide (CO₂) penetrated into concrete from surrounding atmosphere and the alkaline hydration products (e.g. calcium hydroxide) in concrete [7–9]. Concrete carbonation can cause a local fall of alkalinity of pore solution due to the consumption of calcium hydroxide [10–12]. The reduction of the concrete alkalinity can destroy the passive film initially-formed around the steel rein-

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ABSTRACT

This paper presents an experimental investigation regarding the coupled effect of carbonation and chloride aerosol ingress on the durability performance of fly ash concrete. Test results demonstrate that carbonation significantly affects the chloride ingress profile, reduces the chloride binding capacity, and accelerates the rate of chloride ion diffusion. On the other hand, the carbonation rate of fly ash concrete is reduced by the presence of chlorides aerosol. The interaction nature between concrete carbonation and chloride aerosol ingress is also demonstrated by the microscopic analysis results obtained from scanning electron microscope and mercury intrusion porosimetry.

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forcement and thus lead to the corrosion of reinforcing steel when both moisture and oxygen are present there. In view of the increasing amount of CO₂ emission around the world because of the traffic pollution and the increasing use of cement products, the deterioration effect of carbonation on reinforced concrete structures becomes even more serious. In addition to carbonation, chloride attack is a different durability problem, which occurs mainly in offshore and coastal concrete structures [13–16]. It is known that when the concentration of chloride ions in the vicinity of steel bars exceeds to a certain threshold value, the protective film around the steel bars becomes unstable and its protection will be broken down, leading to a state of active steel corrosion [17–20]. The chlorideinduced corrosion of reinforcing steel results in the reduction of reinforcement cross-sectional area, the onset of concrete cracks, and even spalling of concrete cover due to the swelling pressure of the corrosion products [20,21]. In many conventional studies dealing with chloride ingress, the concrete structures considered are either immersed into seawater or in tidal zones. In addition, most publications on this subject only described the laboratory studies in which the concrete samples are submerged into a NaCl solution [22-24]. In fact, when the reinforced concretes are located in a coast close to the sea or in other salt aerated conditions, the atmospheric chlorides (e.g. chloride aerosol) can accumulate on the surface of concrete and gradually ingress into concrete if the exposure duration is long enough, and thus cause the corrosion of reinforcing steel. The chloride-contained aerosol is mainly generated over the

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sea and is carried inland by wind [25]. Despite the large number of studies on chloride ingress in saturated concrete, there are not many research works focusing on the mechanism of chloride aerosol ingress. Moreover, the performance of concretes under the coupled effect of carbonation and chloride aerosol ingress is even more complicated. The presence of carbonation can tremendously affect the behaviour of chloride aerosol ingress, and vice versa. Due to the significance of this problem, it is crucially important to investigate this interacted mechanism for better assessment of the existing concrete structures in real conditions.

During the past few decades, the pozzolan materials have been used as cement replacement in concrete, which has gained increasing attention due to the energy conservation, economic and environmental consideration [26]. It is acknowledged that the pozzolanic additions have profound influence on the permeability of concretes, particularly to chloride ion transportation [27,28]. Fly ash is one of the most common pozzolan materials, which is a byproduct of coal combustion from power plants and metallurgical industries. As demonstrated by a number of studies found in the literatures, the utilization of fly ash in hardened cement and concrete materials causes high porosity at early ages, which results in the less resistance to the penetration of chlorides and CO_2 [29–31]. Therefore, the concretes incorporating fly ash may have less resistance to chloride ingress. However, fly ash material has micro-filler effect and pozzolanic reaction in the late age of curing, which is beneficial in improving the resistance of concrete against the ingress of harmful substances. In order to gain a better understanding of the influence of pozzolan materials on the coupled environmental actions in the real structures, there is a need to investigate the resistance performance of fly ash concretes against the coupled effect of carbonation and chloride aerosol ingress.

In the present work, an experimental investigation is carried out to understand the durability behaviour of fly ash concretes against the coupled effect of carbonation and chloride aerosol ingress. The effect of carbonation on chloride ingress profile, chloride binding capacity and chloride ion diffusion coefficient is examined. In addition, the influence of chloride aerosol ingress on carbonation depth and alkalinity change in concrete is investigated. Microscopic properties such as morphology, porosity, and pore size distribution for fly ash concretes are also explored by using scanning electron microscope (SEM) and mercury intrusion porosimetry (MIP) techniques.

2. Experimental

2.1. Materials, mixture proportions and specimen details

The cement used in this research work was type I ordinary Portland cement (OPC) made in Starfish Onoda Cement Limited Company of Shenzhen, China. Type I fly ash material was obtained from a power station in Ma Bay of Shenzhen, China. The chemical composition and the physical properties of the cement and fly ash used are presented in Table 1. Crush stone with a density of 2700 kg/m³ was used as course aggregate, which was obtained from a quarry of AnTuo Mountain, Shenzhen, China. The maximum and minimum aggregate sizes of the coarse aggregate were 20 mm and 5 mm, respectively. River sand with a fineness modulus of 2.61 and a density of 2632 kg/m^3 was employed as fine aggregate. Distilled water was used in the mixtures of concrete. The water-to-binder ratio for all tested concrete specimens was 0.47. The fly ash was used as partial replacement of cement at 0%, 15%, and 30% by weight of binder. The volume ratio of the fine aggregate to total aggregate (fine aggregate + coarse aggregate) used in all concrete specimens was kept as 0.4. The detailed mix proportions of the concrete specimens are listed in Table 2.

Table 1

Chemical composition and physical characteristics of ordinary Portland cement and fly ash.

| | Cement | Fly Ash |
|--|--------|---------|
| Composition (mass% as oxide) | | |
| Calcium oxide (CaO) | 64.67 | 4.74 |
| Silica (SiO ₂) | 18.59 | 62.32 |
| Alumina (Al ₂ O ₃) | 4.62 | 23.95 |
| Iron Oxide (Fe ₂ O ₃) | 4.17 | 1.33 |
| Magnesium oxide (MgO) | 2.35 | 2.04 |
| Sulfur trioxide (SO ₃) | 3.32 | 1.25 |
| Potassium oxide (K ₂ O) | 0.92 | 0.76 |
| Physical characteristics | | |
| Loss on ignition (LOI) | 1.03 | 3.12 |
| Specific surface area (m ² /kg) | 345 | 391 |
| 80 μm sieving fineness (%) | 4.15 | 8.30 |

All mixtures were cast into a mould with dimensions of $100 \text{ mm} \times 100 \text{ mm} \times 100 \text{ mm}$ and then demoulded after 24 h, followed by the moist curing at $20 \,^{\circ}$ C room temperature and 95% relative humidity for 28 days. More specifically, merely the top surface was exposed to carbon dioxide or chloride aerosol while the other surfaces were coated with epoxy resin, in order to simulate a one-dimensional diffusion process. After the surface treatment, the concrete specimens were separated into four groups, representing different exposure conditions, namely, group A (chloride aerosol ingress), group B (carbonation-chloride aerosol ingress), group C (carbonation), and group D (chloride aerosol ingress-carbonation). More details of the experimental scheme for these four groups of specimens are described in Table 3.

2.2. Accelerated carbonation condition, pH value and carbonation depth measurement

The accelerated carbonation test was carried out in a carbonation chamber for a period of 28 days according to the Chinese National Standard GB/T 50082-2009 "Standard for test methods of long-term performance and durability of ordinary concrete" [32]. The concentration of CO₂ in the carbonation chamber was 20%, the temperature was 35 ± 2 °C, and the relative humidity was 70 ± 2 %, respectively. Although the 20% concentration of CO₂ was much higher than the normal atmospheric condition and the carbonated hydration products under such a high CO₂ concentration might differ from that under the natural carbonation condition, the use of the high concentration of CO₂ could achieve more obvious carbonation results in given short periods of 28 days in this research work.

In order to evaluate the effect of carbonation on the change in alkalinity, pH value of the pore solution in the carbonated zone was measured. During the experimental work, the pH measurements were carried out within 0-16 mm depth interval in the specimens. The method of pH measurements was based on mixing powdered samples with distilled water as a solvent and measuring the pH of the suspension, which was proposed and validated from the literature [33]. The concrete specimen was sliced using a diamond saw at an interval of 1 mm for the first 10 mm starting from the exposed surface and at an interval of 2 mm in the range from 10 mm to 16 mm. These slices were further pulverized into fine powers by a grinding machine. Thereafter, 1 g powder and 50 g distilled water were then weighted, and mixed together for 24 h prior to titration process. A device of pH metre (version: PHS-3C, with accuracy of \pm 0.01 pH) was employed in this study to determine the pH value of the pore solution in the carbonated concrete.

In addition, the carbonation depth was measured by using a phenolphthalein test. The carbonated concretes were split into two blocks. The fresh surface was cleaned and sprayed by a phenolphthalein pH indicator. The carbonation depth was recorded by a

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