

# Influence of slag incorporation on electrochemical behavior of carbonated cement



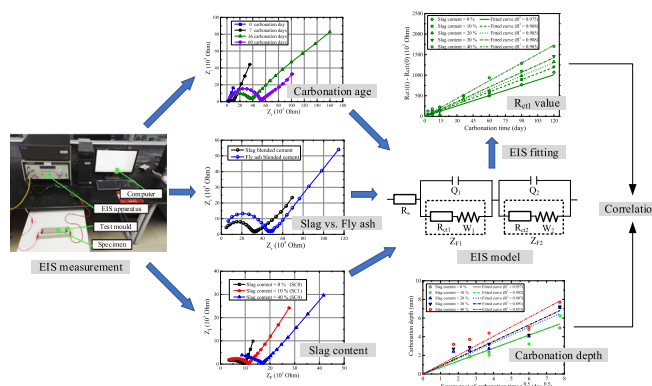
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## HIGHLIGHTS

- Slag effect on electrochemical behavior of carbonated cement is studied.
- Slag cement exhibits larger semi-circles of Nyquist curve than plain cement paste.
- An increase of slag content can hinder the ion transport process in cement system.
- Slag cement has less resistance of ion transport than fly ash cement.

## GRAPHICAL ABSTRACT



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## ABSTRACT

A research work is carried out to understand the influence of slag incorporation on the electrochemical behavior of carbonated cement systems. Measurement of carbonation depth and electrochemical impedance spectroscopy is conducted. A suitable equivalent circuit model is applied to fit the measured electrochemical impedance data (Nyquist curve) and quantitatively obtain the electrochemical property for carbonated slag blended cement. Test results demonstrate that increasing the slag replacement ratio in cement can increase the carbonation depth, enlarge the semi-circle of the Nyquist curve, and improve the resistance of ion transfer at the “solid-liquid phase” interface. We also compare the slag incorporation effect on the electrochemical system of carbonated cement with another common mineral admixture (i.e. fly ash). It is found that slag blended cement exhibits smaller semi-circle of Nyquist curve and larger carbonation depth than fly ash blended cement. Based on the electrochemical modification, we also predict the carbonation depth of blended cements with different slag contents.

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## 1. Introduction

As an essential component of concrete, ordinary Portland cement is one of the most widely used building materials around the world. In China, the cement production has grown rapidly

alongside the national economy since the 1980s [1]. However, a large amount of carbon dioxide (CO<sub>2</sub>) is emitted from the fossil fuel combustion and the calcination process in cement industry [2]. Generally, one ton cement production can release one and a half tons CO<sub>2</sub> to the atmosphere, and it is estimated that CO<sub>2</sub> emission will alarmingly increase to 38.8 × 10<sup>9</sup> tons by 2025 [3]. Such high CO<sub>2</sub> emission would lead to escalate the global greenhouse effect. Moreover, cement production consumes a large amount of energy

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and accounts for a large portion of the financial expenditure in the construction market [4–8]. To remedy these problems, a promising solution is using the mineral admixtures which partially replace the cement constituent. With the development of construction technology, slag has been widely used as a mineral admixture for concrete materials. It is obtained as a by-product or waste material from the ferrous and nonferrous metal industries [9–16]. Slag has numerous advantages which favor it as a constituent of cement, for example, the low initial capital cost of the raw material and the associated energy savings [17,18].

Despite the above beneficial aspects, aggressive chemical substances from the outside environment can penetrate into the blended cement through the capillary pores [5,19–23]. When CO<sub>2</sub> penetrates the cementitious materials in this way, carbonation occurs. Carbonation is a neutralization reaction that can lead to reduce the alkalinity of pore solution in cement and concrete materials owing to the consumption of hydrate cement compounds [24–27], which is an unwanted chemical phenomenon for steel-reinforced concrete structures [28,29]. Based on the alkalinity change, conventional testing methods (e.g. phenolphthalein indicator [30,31], pH meter [32]) have been applied for carbonation characterization. However, these methods require the damage of specimen, and the measurement accuracy depends on the test duration and the experience of inspectors. Repairing the damage after measurement by using phenolphthalein indicator also requires much time and heavy workload. In addition to the chemical modification, previous studies have indicated that carbonation can tremendously affect the electrochemical behavior (e.g. resistance of ion transfer and diffusion) of various cement systems [33–36]. This indicates that the carbonation assessment can be achieved by obtaining electrochemical characteristics of carbonated cement systems with the use of electrochemical measurement techniques like electrochemical impedance spectroscopy (EIS). EIS method is non-destructive, steady-state, sensitive, highly reproducible, and relatively easy to be used in construction field. The equipment and devices used in EIS measurement are easily available from electrochemical community, which are not so expensive especially for the portable EIS instruments. The data analysis and post-treatment can be performed with computer program and the results can be visualized automatically. From the engineering point of view, investigating the factors influencing the electrochemical behavior of carbonated cementitious materials is an essential step to develop a reliable electrochemical-based approach for analysis of carbonation of different cement-based materials. From the review of past research work, effects of water-to-cement ratio [36], river sand aggregate [37], and fly ash incorporation [38] on the electrochemical performance of carbonated cementitious material have been reported. Nevertheless, the role of slag on the electrochemical behavior of cement under carbonation has not yet been well clarified thus far. As compared to fly ash, slag material is self-cementing and does not require calcium hydroxide to form cementitious products, resulting in a distinct pore structure in cement system [39–42]. As a result, a special carbonation behavior as well as electrochemical behavior can be formed for slag blended cement. Understanding the impact of slag on the electrochemical property of carbonated cement would be of great importance for durability design of blended cement lining with an excellent carbonation resistance.

The objective and originality of study is to investigate the electrochemical behavior for carbonated cement systems with slag incorporation. The impedance spectra of slag blended cement, fly ash blended cement and plain cement is compared. Factor of slag content on the carbonation depth and the electrochemical system is evaluated. An equivalent circuit model is applied to quantitatively analyze the EIS data. Furthermore, the electrochem-

ical property of slag blended cement is correlated with the carbonation depth. Finally, the prediction of carbonation depth is performed.

## 2. Materials and methods

### 2.1. Specimen details

Type I ordinary Portland cement was used in accordance with GB175-2007 [43], which was obtained from the Onoda Cement Limited Company in Shenzhen, China. The percentages of calcium silicate (C<sub>3</sub>S), dicalcium silicate (C<sub>2</sub>S), tri-calcium aluminate (C<sub>3</sub>A), and tetra-calcium aluminoferrite (C<sub>4</sub>AF) in cement were 65.35%, 5.06%, 10.23%, and 10.40%, respectively. Class I slag and fly ash was obtained from the Mawan power plant in Shenzhen, China, according to the standard GB/T 18046-2008 [44]. The chemical components of cement, slag and fly ash are shown in Table 1. Cement without incorporation of slag was fabricated as a control specimen. In order to figure out the different effects between slag and fly ash, the fly ash blended cement was also fabricated. Four slag contents were considered in the slag blended cement. The mix proportions of all the cements are given in Table 2.

The dimensions (width × height × length) of all specimens were 40 mm × 40 mm × 160 mm. The cement raw material, mineral admixture and distilled water were mixed in a treater for approximately 90 s. The mixture was then cast in a steel mold and compacted by using a vibrating table. The steel mold was removed after 24 h of the matrix hardening under ambient conditions. Thereafter, the hardened specimens were stored in a water-curing tank for 28 days of curing. Finally, the specimens were transferred into a drying chamber at a temperature of 40 °C. The total number of specimens for each mixture type was 24. For each measurement time, three identical specimens for each mixture type were taken for carbonation depth measurement. Before carbonation depth measurement, one of the above three specimens was selected as the representative for EIS measurement.

### 2.2. Accelerated carbonation condition and carbonation depth measurement

Before exposure to carbon dioxide in the laboratory, the side faces of the specimens were sealed with wax while the two remaining end surfaces were left without wax, as shown in Fig. 1(a). This was to prevent CO<sub>2</sub> from penetrating into the cement specimens from the side faces, only allowing the carbonation to proceed inwards from the end surfaces in one-dimensional manner. Under the 28 days underwater curing age, the drying condition and the above surface treatment, it is assumed that the hydration activity of plain cement and blended cement was almost completed and had little impact on the carbonation process and the EIS measurement results.

Specimens were placed in an accelerated carbonation chamber where the temperature was 30 °C (±1 °C), the humidity was 65–70% and the concentration of CO<sub>2</sub> was 20%. After carbonation condition, the carbonation depth was measured according to the suggestions from these literatures [45–47] and Chinese GBJ820-85 Standard for test methods of long-term performance and durability of ordinary concrete [48]. The carbonation depth measurement was conducted at the ages of 0, 3, 7, 14, 36, 60, 90 and 120 carbonation days. 1% phenolphthalein solution was sprayed on the split cross section to determine the carbonation depth. In order to alleviate reading errors, each specimen was tested at seven equidistant points on each split face, where the carbonation depth was defined as the perpendicular distance from the carbonation front to the exposed surface. The average value was calculated and presented as the carbonation depth, with the precision of 0.1 mm.

### 2.3. EIS measurement and modelling

The EIS measurements were conducted at different carbonation ages (0, 3, 7, 14, 36, 60, 90, 120 carbonation days). During EIS measurement, the specimens were placed between two parallel electrodes (made of stainless steel) mounted in a test mold (Chinese Patent No. ZL 201120473976.2), as shown in Fig. 1(b). In order to

**Table 1**  
Chemical composition and physical properties of the cement and fly ash.

Composition (Mass% as Oxide)	Cement	Slag	Fly ash
Calcium oxide (CaO)	64.67	37.73	4.74
Silica (SiO <sub>2</sub> )	18.59	34.62	62.32
Alumina (Al <sub>2</sub> O <sub>3</sub> )	4.62	11.81	23.95
Iron Oxide (Fe <sub>2</sub> O <sub>3</sub> )	4.17	2.73	1.33
Magnesium oxide (MgO)	2.35	9.43	2.04
Sulfur trioxide (SO <sub>3</sub> )	3.32	1.42	1.25
Potassium oxide (K <sub>2</sub> O)	0.92	0.65	0.76
Loss on ignition (LOI)	1.03	1.2	3.12
<i>Physical Properties</i>			
Specific surface area (m <sup>2</sup> /kg)	345	443	391
80 μm sieving fineness (%)	4.15	–	8.30

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