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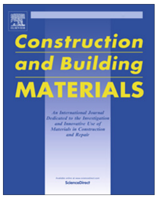

The influence of air cooled blast furnace slag (ACBFS) aggregate on the concentration of sulfates in concrete's pore solution

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HIGHLIGHTS

- Air-cooled blast furnace slag (ACBFS) aggregate contribute sulfate to pore solution.
- Smaller particles of ACBFS release the sulfate faster than the coarse particles.
- Rate of release is higher during early stages of contact with pore solution (APS).
- Chemical composition of APS had no effect on rate and amount of sulfate release.

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ABSTRACT

This study evaluates changes in chemical composition of artificial pore solutions (APSs), representing the actual pore solutions of plain, binary, and ternary cementitious systems, resulting from contact of APSs with air cooled blast furnace slag (ACBFS) aggregate. The maximum increase in the sulfate concentration of APSs was 12.23 and 16.87 mmol/L for, respectively, the coarse and crushed ACBFS aggregate. More than 40% of the total sulfate released by ACBFS aggregate occurred within the first 3 days of contact with APSs. After 28 days of contact with ACBFS aggregates, the pore solution of plain cement paste had the highest sulfate concentration.

1. Introduction

Recycled materials are being increasingly used in concrete to reduce the cost and the environmental impact associated with construction activities and solid waste disposal. The U.S. geological survey reported that approximately 1.5 million metric tons of air cooled blast furnace slag (ACBFS) aggregate was used in manufacturing of concrete in the United States in 2006 [1]. Although, when compared to the total amount of aggregates consumed in production of concrete, the percentage of ACBFS used as aggregate is low, this product is still commonly utilized in concrete in regions close to iron/steel production plants.

Several researchers [2,3] showed that fresh and hardened properties of concrete incorporating ACBFS aggregates are comparable to properties of concrete manufactured using naturally mined aggregates. However, it has been reported that concrete containing ACBFS aggregates showed more frequent instances of infilling of air voids by ettringite compared to concretes with naturally mined aggregates [4,5]. The infilling process reduces the space available for water to expand on freezing, and therefore renders the microstructure of concrete more susceptible to freeze-thaw damage. Previous studies have suggested that ACBFS aggregate itself can be the potential internal source of sulfates being released into the pore solution, and that these sulfates contribute to the observed infilling of the air voids in concrete [6,7].

The ACBFS, a by-product of iron and steel industry, is generated during smelting of iron ore, the process which utilizes limestone (or dolomite) as fluxing agents. Though the composition of ACBFS

Abbreviations: ACBFS, air-cooled blast furnace slag; APS, artificial pore solution; APSs, artificial pore solutions; IC, ion chromatography; ICP-OES, inductively coupled plasma optical emission spectroscopy.

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depends on the chemistry of the raw materials used in the iron production process, this by-product is mainly composed of four major oxides: CaO, SiO_2, Al_2O_3, and MgO [7]. Generally, the sulfur content of ACBFS varies between 1% and 2% (by mass). Amongst several forms of sulfur that can be found in the ACBFS, the crystalline calcium sulfide (CaS – also known as oldhamite) plays potentially important role with regard to the performance of ACBFS as aggregate in cementitious systems [7]. Specifically, in his 1999 study Hammerling [6] showed that oldhamite is highly soluble in a high pH environment. Therefore, when used in concrete, ACBFS aggregates can potentially contribute sulfates to the pore solution. As previously mentioned, the presence of excessive amount of sulfates in the pore solution can result in the infilling of air voids due to the precipitation of ettringite [6,7]. However, clear evidence of the extent of release of sulfur from ACBFS aggregates into the pore solution of concrete is still lacking. Therefore, to ascertain the durability of concrete structure incorporating ACBFS aggregate, it is essential to determine the amount and the rate of release of sulfur from ACBFS aggregate into the pore solution of commonly used cement paste matrices.

Modern day concretes often utilize fly ash (FA) and/or slag cement (SC) as a partial replacement for the ordinary portland cement (OPC). The chemistry of the pore solution of the resulting cement paste matrix is therefore a function of the chemistry, mineralogy, and dosage of the supplementary cementitious materials used [8,9]. Partial replacement of OPC with SC has been reported to reduce the alkali concentration of the pore solution because of the dilution effect [10] and increased binding of alkalis by the hydration products [11]. Similarly, Wang showed that the chemistry of the pore solution is dependent on the type and dosage of FA used [12]. Considering these facts, the question arises as to whether the leaching (or dissolution) of sulfur from ACBFS in any given cementitious system will also be influenced by the composition of the pore solution present in this system.

This paper presents the results of an experimental study on leaching of sulfates from ACBFS aggregate into pore solutions of different types of cementitious matrices. The study involved immersing two types of ACBFS aggregate (coarse and fine) in different types of artificial pore solutions (APSs) that represented the following cementitious systems: a) plain system (100% Type I OPC), b) two types of binary systems (80% OPC + 20% Class C FA and 75% OPC + 25% SC), and c) one type of ternary system (60% OPC + 23% SC + 17% FA). The changes in the chemistry of APSs in contact with the ACBFS aggregates were monitored over time and the results were used to quantify the role of the type of cementitious system in controlling the amount of sulfates released to the solution. Though these simplified laboratory tests did not exactly replicate the real concrete system, comparing and contrasting changes in the sulfate concentration of APSs (representing different cement paste compositions) in contact with ACBFS aggregates should help to identify the susceptibility of a given cement paste systems to potential internal sulfate attack. Please note that throughout this paper the acronym "APS" will be used when referring to a particular (i.e. single) pore solution whereas the acronym "APSs" will be used when referring to multiple pore solutions.

2. Materials

Table 1 summarizes the chemical composition of all materials used in this study. The cement used had a chemical composition representative of the ASTM C150-17 [13] Type I OPC. Chemical compositions of FA and SC used in this study are, respectively, representative of a typical Class C fly ash and ASTM C989-16 [14] grade 120 slag cement. The coarse ACBFS aggregate used in this study was in compliance with the Indiana Department of Transportation (INDOT) specification for No. 8 coarse aggregate [maximum diameter (D_{max}) = 25 mm or 1 in.] [15]. Although, in general, the ACBFS is typically used to replace the coarse aggregate, it is also reasonable to assume that some of the coarse ACBFS particles will abrade during mixing and thus produce smaller sized aggregates. The higher surface area of such smaller particles might accelerate the rate of sulfur leaching from ACBFS aggregates into the pore solution. In order to determine the effects of the size of the aggregate on the change in chemical composition of the pore solution, this study utilized both, the coarse ACBFS particles and the fine ACBFS particles (which were obtained by crushing the coarse ACBFS aggregate). The maximum diameter of the fine aggregate was 10 mm (0.4 in.).

3. Experimental methods

Fig. 1 summarizes the methodology used to quantify changes in pore solution chemistry due to presence of ACBFS aggregates. The first step involved extraction of pore solution from plain, binary, and ternary binder system paste samples. The chemical composition of the pore solution extracted from the paste samples was determined by both, the ion chromatography (IC) and inductively coupled plasma optical emission spectroscopy (ICP-OES) techniques. The resulting composition information was used to prepare the APSs which were used as a soak solution for the ACBFS aggregates. The changes in the chemical compositions of the APSs over time were evaluated using both, the IC and the ICP-OES techniques.

3.1. Paste composition and curing conditions

Table 2 shows compositions (based on volumetric design) of four different cement paste mixtures which were used in the course of this study. All pastes shown in Table 2 were prepared using a Hobart mixer following the procedure described in ASTM C305-14 [16]. The size of each of the individual batches was 1 L. The prepared pastes were cast in cylindrical plastic containers (5 cm in diameter and 10 cm tall). Once the casting was completed, the containers were sealed and placed in an environmental chamber (maintained at 23 °C and 100% relative humidity (RH)) where they remained for specified period of curing time of either 3, 7 or 28 days.

3.2. Pore solution expression and analysis

Once the paste specimens reached the prescribed curing age of 3, 7, or 28 days, two test cylinders from each paste mix were demolded and immediately placed in a pore solution extraction dye (previously described by Barneyback and Diamond [17]). The pressure on each paste sample was increased from 0 to 435 MPa (63,000 psi) in cyclic steps until 5–10 ml of pore fluid was recovered through the fluid drain. The pore solution was then filtered and transferred into a sealed vial for analysis.

Pore solutions obtained from paste samples were diluted 50 times using deionized water. The diluted solution was analyzed using Dionex ICS 900 ion chromatograph (IC) and Perkin Elmer OPTIMA 8300 inductively coupled plasma optical emission spectrometer (ICP-OES) to determine the anions, namely, chlorides and sulfates (Cl^- and SO_4^{2-}), and elements of sodium (Na), potassium (K), calcium (Ca), magnesium (Mg), aluminum (Al), and iron (Fe), respectively. The following wavelengths were used to determine the concentration of different elements using ICP-OES: 589.592 nm for Na, 766.490 nm for K, 317.933 nm for Ca, 285.213 nm for Mg, 396.153 nm for Al, and 238.204 nm for Fe. Prior to analysis in the ICP-OES, the solutions were acidified by adding the HNO_3 (5% by volume of the solution).