

Enhancing the addition of fly ash from thermal power plants in activated high belite sulfoaluminate cement

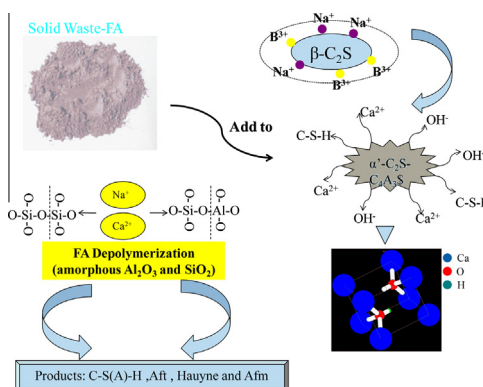
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HIGHLIGHTS

- Borax was introduced as an activator to change the polymorph of belite from β to α' .
- Hydration heat of activated HBSC was 382 J/g, while that of inactive HBSC was 311 J/g.
- Fly ash in cement was increased from 5 % (β -C₂S-C₄A₃\$) to 12 % (α' -C₂S-C₄A₃\$).

GRAPHICAL ABSTRACT



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ABSTRACT

This study focuses on the activation of belite to promote the addition of solid industrial waste, such as fly ash (FA), as the supplementary component in high belite–sulfoaluminate cement (HBSC). The results indicate that the polymorph of belite, which transforms from β to α' , principally depends on the concentration of sodium tetraborate. The incorporation of FA into inactivated HBSC is very slow because the inactivated belite is unable to participate in the early hydration process to release calcium hydroxide (CH) and break the glass structure of FA. Compared with the inactivated belite, considerable hydration heat is released during the early hydration process in activated HBSC, in which a large amount of CH is present to promote the pozzolanic reaction of FA. In addition, $(\text{Na,Ca})_8(\text{Si,Al})_{12}\text{O}_{24}(\text{SO}_4)_2$ is formed in the presence of sodium, which is a tectosilicate mineral with a bulk modulus that contributes to the compressive strength of the later period. The highest total content of FA in the activated HBSC, compared to inactive HBSC, improved from 5% to 12%. Thus, FA promotes the use of solid industrial waste and reduces the consumption of HBSC. Furthermore, the long term strength development with a large amount of FA can meet the requirements for engineering projects.

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1. Introduction

Energy conservation and the reduction of emissions have currently become focus for the environmental protection of the modern world. Calcium sulfoaluminate (CSA) cement, which was

manufactured in China 30 years ago, has been proposed as an alternative binder to reduce CO₂ emissions during the production of cement clinker [1–5]. The following cement nomenclature will be used throughout the manuscript: C = CaO, A = Al₂O₃, F = Fe₂O₃, H = H₂O, S = SiO₂ and \$ = SO₃. High belite–sulfoaluminate cement, which is based on sulfoaluminate cement, is produced by adjusting the proportions of belite (C₂S) and Ye'elimite (C₄A₃\$), and has nominal mineralogical compositions in the range of C₂S (40–70%),

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C_4A_3S (25–50%) and a minor aluminates phase. In high belite–sulfoaluminate cement (HBSC) clinker, belite is an important mineral phase in addition to calcium sulfoaluminate. Compared to ordinary Portland cement (OPC), HBSC has several advantages. (i) It requires less calcium oxide than OPC, which reduces the CO_2 produced per ton of HBSC in the sintering process by 0.19 ton. (ii) The sintering temperature of calcium sulfoaluminate is 100–150 °C lower than that of the calcium silicate in OPC. In addition, due to the high porosity of the sulfoaluminate clinker, it is easy to grind [6]. These temperature and porosity characteristics result in reductions of 0.04 and 0.02 tons of CO_2 per ton of CSA produced, respectively [7]. (iii) Furthermore, material resources are available to produce clinker because the solid industrial by-products, such as alumina-rich FA, anodization mud, red mud and desulfurization gypsum (FGD), can be used to replace bauxite and gypsum [8–12].

Many types of calcium sulfoaluminate-based cements have been developed in the past few decades for a wide range of potential applications [13–15]. However, the reactivity of belite is not sufficient for sustaining early strength development. Belite is a stable phase from room temperature to its melting point at 2130 °C, and thermal and X-ray analyses have revealed that five polymorphs of C_2S exist at ordinary pressures; it undergoes several polymorphic phase transitions in this temperature range, via $\alpha \xrightleftharpoons{1425} \alpha'_H \xrightleftharpoons{1160} \alpha'_L \xrightleftharpoons{630-680} \beta \xrightleftharpoons{500} \gamma$ [16]. Typically, the main polymorph of C_2S in industrial clinkers is the β phase.

FA, blast-furnace slag and steel slag are the typical solid industrial wastes released from the coal combustion process in thermal power plants or from ironworks sludge, respectively [17]. The chemical composition of FA primarily includes SiO_2 and Al_2O_3 , which are able to participate in the pozzolanic reaction during clinker hydration. The amorphous phase in FA reacts with the CH, which is released during the hydration of calcium silicates, to form calcium silicate hydrate and calcium aluminate hydrate, among other compounds. There is no CH as the absence of C_3S in HBSC. As a result, the progress of the pozzolanic reaction during the early hydration rarely occurred. Thus, the absence of alite limits the incorporation of materials such as FA and slag in HBSC [18].

This paper discusses the common β -modification and α' - C_2S , which is more reactive than β - C_2S but still reasonably easy to stabilize [19]. The purpose of this study is to enhance the reactivity of belite, which is essential for promoting the pozzolanic reaction so that an increased number of solid industrial waste materials can be incorporated into HBSC.

2. Experimental

2.1. Raw materials

Alumina-rich FA and desulfurized gypsum were obtained from the Zhejiang Tianda plant, limestone was obtained from the Hebei Taihang plant, and sodium tetraborate ($Na_2B_4O_7 \cdot 10H_2O$, AR, 99.996%) was provided by Bayer (Shanghai, China). The primary oxide compositions of the raw materials, which were determined by chemical analysis, are shown in Table 1.

2.2. Mixture of raw materials preparation

Six mixtures were prepared by combining the raw materials in different proportions (for C1: the proportions of limestone, FA and FGD are 68.1%, 24.5%, 7.4% by mass, respectively) and homogenizing in a laboratory ball mill to be able to pass through an 80 μm sieve. Sodium tetraborate was added to the mixtures (0.5, 1, 1.5, 2, and 2.5 wt% of $Na_2B_4O_7$) to promote the formation of reactive belite polymorphs (the α and α' forms). Table 2 provides additional information regarding the mixtures (C1–C6) used to prepare these clinkers.

2.3. Synthesis of inactive and active belite sulfoaluminate cement

The mixture was mixed with water and pressed into a disk mold (φ 40 mm \times 25 mm) at a pressure of 15 MPa. The disks were dried in an oven at 100 °C for 1 h, calcined in a Si–Mo rod resistance furnace at 1280 °C for 30 min, then

Table 1

Chemical composition of raw materials (wt.%).

Material	limestone	FA	FGD
CaO	51.55	4.04	34.01
SiO_2	3.26	45.61	0.02
Al_2O_3	0.99	40.16	0.92
Fe_2O_3	0.57	3.73	0.19
SO_3	0.07	3.01	39.96
MgO	1.37	1.26	0.13
LOI ^a	40.68	1.67	24.94

^a LOI – loss of ignition at 950 °C.

Table 2

Proportions used to obtain the different clinkers, expressed as oxide in wt.%.

Label	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3	MgO	$Na_2B_4O_7$
C1	51.14	21.03	16.94	2.53	5.6	1.72	0.0
C2	50.88	20.92	16.86	2.52	5.57	1.71	0.5
C3	50.62	20.82	16.77	2.50	5.54	1.70	1.0
C4	50.37	20.71	16.69	2.49	5.52	1.69	1.5
C5	50.12	20.61	16.60	2.48	5.49	1.69	2.0
C6	49.86	20.50	15.52	2.47	5.46	1.68	2.5

removed from the furnace and rapidly cooled with forced air. All of the clinker samples were ground to pass through a 80 μm sieve. The HBSC was prepared by mixing the clinker and the gypsum in the following proportion: clinker/gypsum = 85/15 (Blaine surface area was 357 m²/kg). The clinkers were relatively soft and friable to OPC; consequently, the entire process involved considerable energy conservation.

2.4. Preparation of hydration samples

All the tests were conducted using inactive and active HBSC. The tests were conducted with two different matrices, with one matrix as a control to determine the effect of the presence of FA. The first matrix (C1) consisted solely of HBSC without activation. In the second matrix (from C2 to C6), varying amounts of FA were added, with the mass ratio from 5% to 12%. All of the pastes were mixed with water at a water/solid ratio (w/s) of 0.26. The hydration of the solid phase at different curing ages was stopped by submerging a part of the crushed pastes into ethanol for 2 days, and the stopped hydration samples were dried and placed into desiccator for further study. Herein, it is worth noting that the w/s ratios as well as the curing conditions, the type of binder, and the use of mineral and chemical admixtures have significant effects on the value of the total porosity and the pore size distribution [20].

2.5. LOI tests of hydration specimens

After stopping hydration, all samples were ground by agate mortar below 0.08 mm to determine the amounts of hydrates that were formed by the samples that were cured at different ages. LOI analysis was conducted in an atmosphere of air utilizing approximately 1 g of sample in a muffle instrument at 10 °C/min for a temperature up to 150 °C. Chemically bound water of ettringite was determined as the weight loss at 100–150 °C. The amount of ettringite in the hydrated pastes was determined as previous described [21], assuming that the weight loss between 50 °C and 120 °C corresponds to 32 molecules of crystal water per molecule of ettringite. Each LOI value represents the average of the twice-measured values of specimens.

2.6. Compressive strength tests

The composition of the standard mortars was as follows: HBSC/sand/water = 1/3/0.5. Prismatic samples (40 \times 40 \times 160 mm) were cured at 20 \pm 1 °C and 95% relative humidity (RH). Then, the samples were removed from the mould after 1 d and immersed in water at 20 °C until they were utilized for compressive strength testing. Before the compressive strength tests, the prismatic samples were first subjected to a flexural test. After the specimens were cured for 1, 3 and 28 days, the compressive strengths were tested using the unconfined compression machine with a maximum load of 200 kN. The loading rate of the machine is 5 mm/min. Each compressive strength value represents the average of the results from 6 specimens tested.

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