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Carbonation characteristics of γ-dicalcium silicate for low-carbon building material

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HIGHLIGHTS

- γ-C_2S blocks exhibits rapid strength gain with high CO_2-absorbing capability.
- Solid phase volume increase densifies the matrix and leads to high strength.
- Ca^{2+} ion dissociation is accelerated by CO_2.
- Ca- and Si-rich products tend to grow in clusters separately.

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ABSTRACT

γ-Dicalcium silicate (γ-C_2S) is characterized by its high carbonation reactivity and has the potential to be utilized as a construction material with the added benefit of CO_2 sequestration. The present work addresses the characteristics of pressure-molded γ-C_2S samples subjected to accelerated carbonation curing, including the developments in microstructure, carbonation degree and mechanical strength. The results indicate that the mechanical strength and surface density of carbonated γ-C_2S block correlate well with its carbonation degree, which is attributed to the generation of more voluminous carbonation products. This strengthens the structure and densifies the matrix. The carbonation products include calcite as the primary morphology and aragonite as the minor phase, and amorphous Ca-modified silica gel formed by coordinated SiO_4 tetrahedrons. Conductivity and ion concentration measurement elucidates the accelerated Ca^{2+} ion dissolution in the presence of CO_2 is the reason for the high carbonation reactivity of γ-C_2S. A conceptual model of carbonation process is proposed based on the distribution of Ca-modified silica gel and calcium carbonates. Additionally, the ecological evaluation demonstrates that γ-C_2S would hopefully reduce 40% of the CO_2 emissions compared to ordinary cement, which opens up a new area of a novel low-carbon construction material.

1. Introduction

Ever since its invention in 1824, Portland cement has become the most widely used construction material all over the world for its convenience and good performance [1,2]. However, cement industry contributes significantly to the greenhouse gas emissions [3]. This causes heavy obligations to cut carbon dioxide (CO_2) emissions in countries with considerable cement production [4]. Thus, for the prospect of developing sustainable building materials, great efforts have been made, among which CO_2 curing for construction materials is one of the most efficient ways [5], which will improve their properties, and meanwhile benefits the long-term storage of CO_2 [6]. In this context, γ-dicalcium silicate (γ-C_2S) is put forward as a potential eco-friendly construction material.

γ-C_2S is the most stable crystal form among five polymorphs of dicalcium silicates with negligible hydraulic activity at ambient temperature [7]. γ-C_2S is now found to be an ideal low-carbon building material due to strong CO_2-absorbing capacity and potential energy conservation associated with a self-pulverization phenomenon during its manufacturing. [8,9]. Taking advantage of its high carbonation reactivity, several new eco-type γ-C_2S based cementitious systems have been explored. Higuchi et al. [10–12] invented an environmentally friendly concrete, CO_2-SUICOM, made mainly of γ-C_2S and fly ash and cured in CO_2 atmosphere with negative CO_2 emissions. A novel concrete composed of ordin-

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ary Portland cement (OPC), α-quartz and γ-C_2S was developed by Saito et al. [13,14], in which γ-C_2S sequestered CO_2 during the curing process to form a densified surface layer, thus protecting the concrete from erosion.

These explorations of γ-C_2S have triggered increasing interests in its application in construction materials attributed to the environmental and performance-enhancing merits. However, the explorations focus mainly on the application results, and there is still a lack of research into the carbonation characteristics of γ-C_2S, such as the carbonation products and the physic-chemical changes in the carbonation process, which are significant for the further development of low-carbon γ-C_2S building material. In this work, the evolution and relation of carbonation degree, mechanical strength and surface density of compression molded γ-C_2S material in carbonation process are discussed. The types and distribution of carbonation products of γ-C_2S are characterized by an array of experimental techniques such as X-ray diffraction (XRD), thermogravimetry coupled with mass spectrometry (TG-MS), ^{29}Si nuclear magnetic resonance (NMR) and scanning electron microscope coupled to an energy dispersive spectrometer (SEM-EDS). The carbonation mechanism is investigated by means of conductivity and ion concentration test. And a conceptual model for the carbonation mechanism of γ-C_2S is also proposed. Finally, the reduction of CO_2 emission is estimated with the association of γ-C_2S as a building material. The results will hopefully shed more light on the development of low-carbon sustainable construction materials.

2. Experimental programs

2.1. γ-C_2S synthesis

2.1.1. Raw materials

Analytically pure $Ca(OH)_2$ and SiO_2 produced by Sinopharm Chemical Reagent Co., Ltd were used with the chemical compositions listed in Table 1. The two materials at a 2:1 M ratio were homogenized in an agate jar mill for 3 h, blended with 10 wt% water, pressed into tablets and fully dried at 105 °C. The tablets were then heated at a rate of 10 °C/min to 1100, 1200, 1300 and 1400 °C respectively for 3 h, and cooled naturally inside the furnace.

2.1.2. Sintering temperature

The XRD patterns of the blended powders sintered at four different temperatures were shown in Fig. 1. At 1100 °C, lots of CaO and SiO_2 remain in the calcined sample, and no γ-C_2S but β-C_2S generated. As the sintering temperature increases to 1200 °C, the peak height of β-C_2S declines, and the peak of γ-C_2S appears. After sintered at 1300 °C, γ-C_2S becomes the major product with a minor presence of lime. Pure γ-C_2S is produced in the sample sintered at 1400 °C. The free-lime content of this sample conducted by alcohol-glycerol method [15] is 0.06 wt%, which means the calcareous and siliceous materials are well combined, indicating a sintering temperature at 1400 °C is required for producing pure γ-C_2S.

2.1.3. Chemical and physical properties of synthesized γ-C_2S

The synthesized γ-C_2S product is white powder with an autogenous pulverization characteristic owing to the volume expansion when transforming from β to γ-form [16] (Fig. 2a). The SEM image of γ-C_2S particles (Fig. 2b) shows the presence of cracks and complex folds on the surface of γ-C_2S due to the pulverization phenomenon. The particle size of γ-C_2S obeys the Gaussian distribution with its D_{10} = 3.283 μm, D_{50} = 16.401 μm and D_{90} = 45.223 μm (Fig. 3), which are the intercepts for 10%, 50% and 90% of the cumulative mass, respectively. The density is measured to be 2.91 × 10^3 kg/m^3 by Le Chatelier Flask method, and the specific surface area tested by Blaine method is 161 m^2/kg.

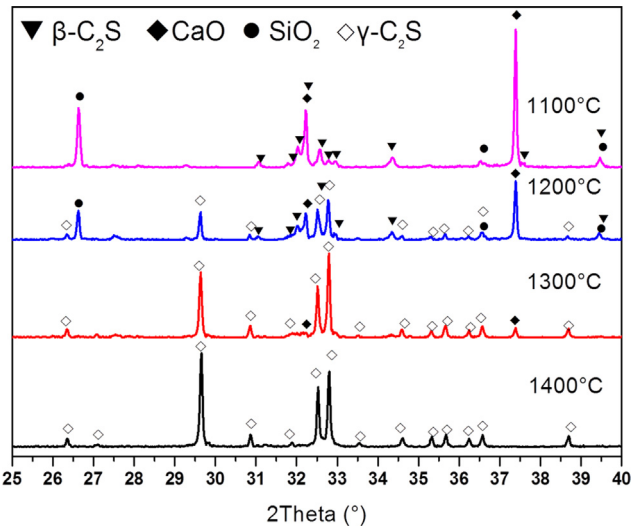

Fig. 1. XRD patterns of γ-C_2S synthesized at different sintering temperature.

2.2. Preparation of test specimens

2.2.1. Pretreatment of synthesized γ-C_2S powders

Before making samples, synthesized γ-C_2S powders were ground in a ball mill for 10 min, after which they would all pass through 200-mesh sieves with a specific surface area of 280 ± 20 m^2/kg which is similar to ordinary Portland cement.

2.2.2. Preparation of precast samples

Compaction is a routine procedure prior to carbonation that helps to get stronger mechanical properties [17,18]. To characterize the carbonation process of γ-C_2S, compressed cylindrical blocks with its basal diameter of 20 mm and a height of about 20 mm were made. Each of the blocks made of 10 g γ-C_2S and 1.5 g water was compacted under a pressure of 30 MPa for 1 min. The water-to-solid (w/s) ratio and compacting pressure are determined from the compressive strength results and the technical requirement for specimen compaction. The prepared blocks were covered with a plastic film to avoid moisture loss during the pressing process.

2.2.3. Specimens for volume change test

Every batch of 60 g of the synthesized γ-C_2S were fully mixed with 9 g deionized water, and the homogenized mixture was piled in a polystyrene disk (15 cm in diameter and 3 cm in height). A steel plate (20 × 20 × 0.5 cm) was then placed on top of the pow-

Table 1
Oxide composition of raw materials.

	CaO	SiO_2	MgO	Al_2O_3	P_2O_5	SO_3	SrO	K_2O	Fe_2O_3	LOI
SiO_2	0.00	99.12	0.03	0.59	0.01	0.01	0.00	0.09	0.05	0.09
$Ca(OH)_2$	74.58	0.16	0.65	0.06	0.00	0.05	0.04	0.00	0.00	24.45