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Preparation of sintered foamed ceramics derived entirely from coal fly ash

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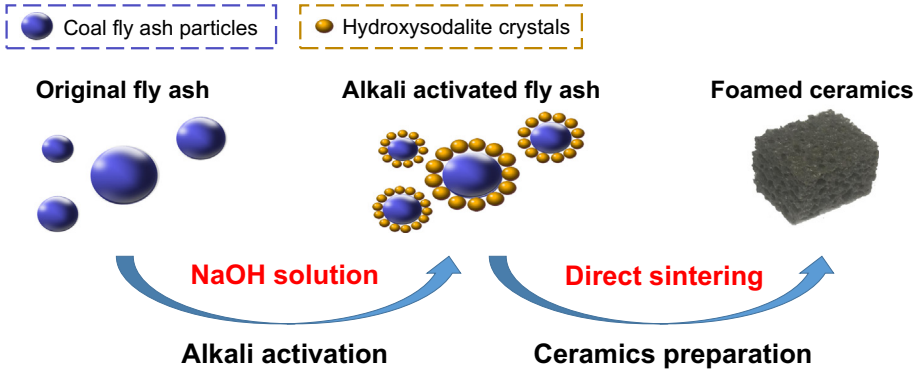
HIGHLIGHTS

- An alkali activation pretreatment of coal fly ash is proposed.
- Fly ash is evenly coated with a layer of hydroxysodalite during alkali activation.
- Alkali activated fly ash has a self-foaming performance during sintering.
- The obtained ceramics have better practical properties compared with reported ones.
- The vitrified encapsulation mechanism explains the reduction in leaching toxicity.

GRAPHICAL ABSTRACT



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ABSTRACT

A new foamed ceramic was successfully synthesized entirely from coal fly ash (CFA). Before sintering, the CFA was pretreated with an alkali activation that evenly coated CFA particles with a layer of hydroxysodalite crystals. Because of the pretreatment, the alkali activated CFA underwent a self-foaming reaction during sintering. When sintered at 1200 °C, the foamed ceramics exhibited optimal properties, including an apparent density of 0.41 g/cm^3, porosity of 83.60%, a compressive strength of 8.3 MPa, and thermal conductivity of 0.0983 W/m·K. The leaching toxicity tests revealed that the hazardous heavy metals were encapsulated within the glassy phases during sintering.

1. Introduction

Because of its vast coal natural resources, China's energy structure is dominated by coal [1]. With rapid economic development in China, more than four billion tons of coal are burned annually to meet increasing electricity demands, making coal fly ash the largest single solid waste in China [2]. The total production of coal fly ash (CFA) in China hit an all-time high of 620 million tons in 2015; however, the actual recycle utilization rate of CFA is less than 70% [3]. The air, soil, and water pollution that accompanies coal combustion is detrimental to the environment and human health. For example, potentially toxic substances such as heavy metals in coal fly ash leach into soil and groundwater, and then accumulate in the food chain [4,5]. Hence, there's an urgent need

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to find other beneficial uses for CFA in order to improve the amount recycled and reduce its pollution hazard.

Over the past few decades, CFA has mainly been used in concrete [6], cement [7], paper making [8], steam-cured bricks [9], ceramics, and other related industries. Because the chemical contents of CFA are close to those of ceramic raw materials, research into using fly ash in applications similar to other ceramic products has attracted much attention. He et al. prepared batches of fly ash-based α-cordierite glass-ceramics containing 68 and 64 wt% CFA [10]. Zhang et al. investigated the leaching toxicity characteristics of a glass-ceramic containing 90 wt% CFA, and found that the glass-ceramic was non-hazardous [11]. Erol et al. produced glass, glass–ceramic, and ceramic materials from CFA without any additives [12], and Li et al. synthesized high-purity mullite ceramics from bauxite and high-aluminum CFA, and reduced the sintering temperature by 50 °C by adding 5–10 mol% V_2O_5 [13]. These examples show that there is a strong interest in employing mature manufacturing technologies to produce basic ceramic products using CFA.

Recently, several researchers reported using conventional ceramic fabrication methods to prepare high value-added foamed ceramics from CFA. Fernandes et al. [14] produced glass foams using sheet glass cullet, CFA (20 wt%), and carbonate as a foaming agent to produce foams with apparent density values of 0.36–0.41 g/cm^3 and compressive strength values of 2.4–2.8 MPa. Chen et al. [15] combined 40–50 wt% red mud, 26.25–40 wt% CFA, 15–20 wt% sodium borate, and 5 wt% sodium silicate to synthesize foamed ceramics with 64.14–74.14% porosity, 4.0–10.6 MPa compressive strength, and a bulk density of 0.51–0.64 g/cm^3. Liu et al. [16] used 60 wt% CFA to prepare porous ceramics that exhibited a bulk density of 0.93 g/cm^3, porosity of 65.6%, and a flexural strength of 11.9 MPa (the compressive strength was not measured). These foamed ceramics prepared by the conventional ceramic fabrication methods using a foaming agent have very desirable values for porosity.

Unfortunately, they also have undesirable pore size distributions and relatively low compressive strengths due to the poor dispersion of the foaming agents. Zhao et al. [17] and Hlaváček et al. [18] used a low-temperature direct foaming method to prepare foamed materials from CFA with bulk densities in the range of 0.4–0.8 g/cm^3, compressive strengths in the range of 4.5–6.8 MPa, and a closed pore network. The direct foaming method is simple and economical, but has some inherent risks in possible leaching of toxic heavy metals compared with conventional sintering methods. The thermal conductivities of foamed ceramics made with CFA have also been investigated by several researchers. Zhu et al. [19] prepared ceramic foams with a 40 wt% fly ash addition, and sintered foams had a bulk density of 0.46 g/cm^3, a compressive strength of 5 MPa, and a thermal conductivity of 0.36 W/m·K. Using software, they also evaluated the energy conservation effect of their foamed products. Li et al. [20] used 90% fly ash to fabricate porous anorthite ceramics that exhibited ultra-high open porosity of 94% and an ultra-low thermal conductivity of 0.042 W/m·K, but the compressive strength was not considered. Therefore, the development of a new kind of foamed ceramic incorporating a high volume fraction of CFA, a good balance of thermal insulation properties and mechanical properties, i.e. high porosity, uniform pore size distribution, high compressive strength and low thermal conductivity, is necessary to fully understand the potential of recycling CFA into foamed ceramics.

This study aspires to possibly address both environmental advantages due to the hazardous waste consumption and economic advantages due to the low cost of CFA. The objective of this work is to prepare sintered foamed ceramics completely derived from CFA with both good thermal insulation properties and optimal mechanical properties. Alkali activation pretreatment effects on the crystalline phase, microtopography, particle size, and thermal behavior of CFA were investigated in detail. A simple and common conventional ceramic preparation method was adopted in this study without a foaming agent. The evolution of ceramic phases and morphology with the increasing temperature was also characterized and discussed. Finally, the heavy metal leaching toxicity of samples was tested, and the corresponding results were compared with the Chinese identification standard for hazardous waste.

2. Experimental procedure

2.1. Materials

CFA was sampled from a thermoelectric power plant in Inner Mongolia, China. The chemical composition of the original CFA is listed in Table 1 in Section 3.1. The loss on ignition of CFA is about 2.83%, and it mainly represents the carbon residue.

Sodium hydroxide was reagent grade (Xilong Chemical Co., Ltd., China) and was used as-received without further purification.

2.2. Ceramic processing

The alkali-activation parameters were optimized to maximize activation effects and economic feasibilities. Alkali activation pretreatment of CFA was performed under stirring in a 5 L high-temperature reactor with external heating and internal cooling (Model GSHA-5, Xintai Chemical Equipment Corporation, China). The system was equipped with an automatic proportional-integral-derivative control system to regulate the heating rate (3 °C/min), agitation (300 r/min), and temperature. CFA and NaOH solution (150 g/L) were added to the reactor, and mixtures with a liquid-to-solid ratio of 5 mL/g were digested at 150 °C for 3 h. After the reaction, the slurry was filtered and washed with hot deionized water (∼95 °C) five times to minimize the amount of adsorbed sodium ions. The obtained samples were dried in an oven at 80 °C for 12 h.

Production of foamed ceramic materials from alkali activated CFA involves two stages: green body production and sintering. Prior to preparing green bodies, the dried alkali activated CFA was mixed with deionized water in a 10:2 mass ratio, and the mixtures were pressed into 40 × 40 × 20 mm bars with the help of a uniaxial tablet press (Model 769YP-60E, Tianjin Keqi High & New Technology Corporation, China) under 20 MPa. For the sintering process, the obtained green compacts were dried at 105 °C for 12 h and then sintered in air atmosphere at 1180, 1190, 1200, or 1210 °C in a laboratory electrical sintering furnace (Model SX-G13135, Tianjin Zhonghuan Lab Furnace Co., Ltd, China). All samples were heated from room temperature to the required sintering temperature at a rate of 5 °C/min, held at the maximum temperature for 30 min, and cooled to room temperature at the rate of 3 °C/min. The resulting ceramic samples were denoted as FCRE 1180, FCRE 1190, FCRE 1200, and FCRE 1210 depending on the sintering temperature used.

2.3. Characterization techniques

The sample chemical compositions and the leaching toxicity test solutions were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS, Model iCAP Qc, Thermo Scientific) under the following conditions: the power was set at 1548.6 W, the nebulizer gas flow was 1.0689 L/min, and the peristaltic pump flow was 1 mL/min. Crystalline phases were identified by X-ray diffraction (XRD, Model X'Pert Pro MPD, PANalytical B.V.) performed at 40 kV and 30 mA using Cu K_α radiation. The microstructures of the samples were observed by scanning electron microscopy (SEM, Model MLA Quant 2500, FEI Company) at an acceleration