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Review article

Effect of particle size and alkali activation on coal fly ash and their role in sintered ceramic tiles

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ABSTRACT

We focus on fly ashes of different sizes and their alkali-activation on ceramic products. Backscattered electron imaging-energy dispersive X-ray spectroscopy was used to classify coal fly ash particles according to particle size and to study the pre-activation of particles with different sizes. Secondary electron imaging-energy dispersive X-ray spectroscopy was used to study the role of coal fly ash particles of different sizes in ceramic bodies before and after alkali-activation. Ash particles can be divided into three classes based on size: clay-, quartz- and feldspar-like particles, which act as clay, quartz and feldspar, respectively, in ceramic bodies. The pre-activation process contributes to the plasticity of ash particles, the crystal skeleton role of clay-like particles and the fluxing agent role of feldspar-like particles, so pre-processing can improve the performance of ash-based ceramic tiles significantly. This research provides a new pretreatment method for coal fly ash in ceramic fields.

Contents

1. Introduction

Coal fly ash (CFA) is a type of solid waste that is produced from coal-fired power plants. CFA emissions have increased annually with developments in the power industry [1]. China's CFA emissions exceed 580 million tons per year [2]. Despite 70% of China's CFA being reused, large amounts of CFA are stored in many locations, which has caused serious environmental pollution problems [3]. The increasing additional use of CFA implies a need for guidance on scientific theory for its disposal [4].

Urbanization has led to the recent rapid development of China's building ceramic industry in recent years, and ceramic tile production has ranked first in recent years [5]. Expanding ceramic industries have resulted in an increased deficiency in traditional

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ceramic raw materials, such as clay and feldspar, and this lack of raw materials has restricted the sustainable development of this industry [6]. CFA, as a type of aluminum-silicate system material, is very similar to ceramic materials in terms of their chemical and physical compositions [7]. Consequently, applying CFA as a substitute for traditional ceramic raw materials in ceramic production could be of great significance [8].

Much work has been conducted to use CFA as ceramic-production raw material. Lin et al. [9] and Ji et al. [10] found that ceramic tiles that were fabricated by using CFA as the main raw material exhibit a better performance compared with traditional ceramics. Lin et al. believe that mullite in CFA provides strength for ceramic-body sintering, and the glass phase promotes sintering by its lower melting point. Ji et al. believe that CFA plays the role of feldspar in sintering. There is no scientific consensus on the role of CFA in ceramic bodies. The differences result from two main reasons: the complex chemical composition of CFA, and vast differences in its particle size and morphology. [11]

To date, most research has considered the macroscopic properties of CFA. In recent years, studies of microscopic particle properties of CFA have increased gradually. Dai et al. [12] sieved high-alumina CFA into six size fractions (<120, 120–160, 160–300, 300–360, 360–500 and >500 mesh) and separated these into magnetic, mullite + corundum + quartz and a glass phase for mineralogical and chemical analysis. They found that the glass abundance in fly ash decreases with decreasing particle size. In contrast, corundum increases with decreasing particle size. CFA particles of different size have rarely been studied systematically or in-depth in ceramic applications because CFA has a broad particle-distribution range. Therefore, it is very necessary to study the basic properties of different-sized CFA and their roles in ceramic bodies.

The hydrothermal alkaline reaction is used commonly to treat CFA in alumina extraction, cement manufacture and zeolite synthesis. For example, Ding et al. [13], Mobili et al. [14] and Shoumkova et al. [15] studied alumina extraction from high-alumina CFA using a high alkaline solution system, the possibility of manufacturing CFA-based mortars that were activated by an alkaline solution under room conditions and zeolite formation by hydrothermal alkali-activation of CFA, respectively. Because CFA is generated at high temperature during sintering, it has a low reactivity. Pre-activation of ash by an alkaline medium is often used to improve the ash reactivity. However, alkali-activation pretreatment is seldom used in ash-based ceramics, hence research on the activation characteristics of different CFA particles and the different activation effects on the properties of ceramic products would be of great use.

The objective of this work was to study the particle-size and pre-activation effects on the properties of CFA particles and the roles of CFA particles in ceramic tiles, respectively. The aim of doing so is to understand the mechanism of utilization of CFA as a type of ceramic raw material and to guide the efficient utilization of CFA. Scanning electron microscopy (SEM)-backscattered electron imaging, which can identify different phases in a specimen [16], and SEM-secondary electron imaging, which can evaluate the specimen structure and morphology [17], were used in this study. Particle-size and pre-activation effects on ceramic particles are characterized mainly by backscattered electron imaging-energy dispersive X-ray (EDX) spectroscopy. The particle-size and pre-activation effects on the role of CFA particles in ceramic tiles were characterized mainly by secondary electron imaging-EDX.

2. Experimental procedure

2.1. Materials

CFA was sampled from a thermoelectric power plant in Inner Mongolia, China. Its X-ray diffraction (XRD) patterns, as shown in Fig. 1(a), show that the main phases comprise quartz, hematite and mullite and some amorphous material. The CFA particle size and morphology are shown in Fig. 1(b) and (c). The CFA particles are glossy microspheres with different particle sizes; the CFA particle size has a normal distribution with a peak at 85.32 μm and the d(0.5) at 75.32 μm, respectively. The chemical composition of the raw materials, including CFA, feldspar, kaolin and bentonite, as determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) is shown in Table 1.

Table 1
Chemical composition of raw materials (wt.%).

Raw material	Al_2O_3	SiO_2	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	K_2O	LOI
Coal fly ash	21.47	55.57	6.80	0.01	5.12	3.97	2.42	1.22	2.83
Feldspar	21.45	62.06	1.08	–	0.23	0.33	2.43	9.45	2.72
Kaolin	26.58	68.74	0.40	0.03	0.47	0.52	0.36	1.21	1.77
Bentonite	19.57	68.23	1.12	0.20	0.01	0.03	0.36	1.30	7.82

LOI: loss on ignition.

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

2.2. Experimental processing

CFA was sieved using several standard test sieves with different pore diameters on an electric mechanical sieve shaker (GS-86 type, Beijing Ever Light Medical Equipment Company Ltd., China) to obtain two size fractions of 50–150 μm and 0–5 μm, and denoted CFA 1# and CFA 2#, respectively.

CFA pre-activation was performed in a 1-L stirred high-temperature reactor with external heating and internal cooling. An automatic proportional–integral–derivative control system was used to control the heating rate, agitation and temperature in the reactor. CFAs of each particle size range and sodium hydroxide solution (100 g/L) were added to the reactor, and mixtures with a solid-to-liquid ratio of 4 mL/g were digested at 100 °C for 2 h. After the reaction, the slurry was filtered and washed with heated deionized water five times to reduce sodium-ion adsorption on the product structure. The samples were dried in an oven at 80 °C for 12 h and labeled CFA 3# and CFA 4# (corresponding to CFA 1# and CFA 2#, respectively).

The production of ceramic tiles from CFA consists of two stages: green-body production and sintering. Prior to making the green body, the dried original ashes or pre-activated ashes (~50 wt.%) were homogenized by milling with feldspar (~30 wt.%), kaolin (~15 wt.%) and bentonite (~5 wt.%). The homogeneous raw materials were mixed with deionized water in a 10:2 mass ratio. The mixtures were pressed into 100 mm × 100 mm × 4 mm cuboids by using a uniaxial tablet presser (Model WE-300B) at 20 MPa. During sintering, the green compacts were dried at 105 °C for 12 h and then were sintered in a laboratory-type electrical sintering furnace (Model HTF1400) at 1200 °C for 2 h. Samples were heated at 15 °C/min. The muffle was cooled at approximately 1.5 °C/min.

2.3. Characterization techniques

Samples were subjected XRD (X'Pert Pro MPD, Panalytical Company; 40 kV, 30 mA, 2θ = 5°–90°, Cu-Kα X-ray source) to identify the crystalline phases, and SEM (JSM 7100F) to determine the morphology. The sample chemical composition was analyzed by using ICP-OES (Optimal 5300DV, PerkinElmer Instruments; 1300W, carrier gas flow = 0.08 L/min, peristaltic pump flow = 1.5 L/min). The powder particle-size distribution was measured by laser particle-size analyzer (Beckman Coulter, LS 13320). A Nicolet Nexus FTIR Spectrometer (Nicolet Instrument Co., Madison, USA) equipped

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