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Effect of shale addition on properties of sintered coal fly ash

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ABSTRACT

Shale can be used as a substitute for clay in sintered fly ash, due to the similar physical and chemical property to clay. In this paper, these characteristics of sintered fly ash with or without shale (binder) were investigated by physical property, X-ray diffraction and scanning electron microscopy. The results show that shale addition ranging from 30% to 50% (in weight) can be beneficial for properties of sintered products at temperature ranging from 950 to 1050 °C. However, a higher amount of shale easily caused significant bloating at 1100 °C. Considering energy saving and best performance, the sintering mix for shale and fly ash (50% in weight respectively, the same as below) fired at 1000 °C for 2 h was an optimal option. XRD results of the sintering mix show that intensity of hercynite increases with the increasing temperature, but intensity of quartz decreases on sintering. SEM confirms that addition of shale can improve microstructure and sintering of fly ash.

1. Introduction

Fly ash is a major solid waste collected from the flue gas of coal fired power plants. According to a report of the State Electricity Regulatory Commission (SERC), 1300 million tons of coal were consumed by fired power plants in China, which produced approximately 350 million tons of fly ash in 2006 [1]. Fly ash particles are mostly spherical in shape and range from less than 1 μm to 100 μm, with a low apparent density (typically between 530 and 1260 kg/m^3). The chemical composition of fly ash is similar to clay, with a total amount of SiO_2 and Al_2O_3 reaching 70–90% [2]. In addition, there are some unburnt coal particles in fly ash. Based on these properties, we can utilize fly ash to prepare sintered products.

In previous research [3–14], fly ash was adopted to prepare sialon ceramic, aggregate and brick at high temperature. For sialon ceramic, Gilbert and Mosset [3] used high-C fly ash to synthesize a nearly pure β-phase sialon ceramic by heating the raw materials, at 1500 °C for 1 h. Jansen et al. [4] reported that mechanical properties of the sialon ceramics synthesized from fly ash are comparable to the properties of sialon ceramics made from clay and pure oxides. Besides, the sialon phases and carbothermal temperature depend on properties of fly ash. For aggregate, Kayali [5] prepared lightweight aggregates by sintering fly ash, and used these aggregates to produce concrete which is around 22% lighter and at the same time 20% stronger than normal weight aggregate concrete. Huang et al. [6] used mining fly ash, mining residues and sludge to prepare lightweight aggregate. Mangialardi [7] obtained the best performance of concrete aggregate with washed fly ash at the sintering temperature of 1140 °C for 60 min.

For brick, Furlani et al. [9] studied the sintering of powders obtained from coal fly ash and paper mill sludge. Aineto et al. [10] studied the fly ash as an additive to clays for fabrication of building ceramic. Clay is added to fly ash in order to improve plasticity of pressed specimens and properties of sintered fly ash, without negative effects on shrinkage, color alteration or efflorescence. Xu et al. [11] studied the effect of fly ash at high replacement ratio of clay on firing parameters and properties of bricks. The results indicate that the plasticity index of mixtures of fly ash and clay decrease dramatically with increase of replacement ratio of fly ash. The fired bricks with high volume ratio of fly ash are of high compressive strength, low water absorption, no cracking due to lime, no frost and high resistance to frost-melting. Biernacki et al. [12] investigated the sinterability of a class F fly ash as a function of processing conditions including sintering temperature and sintering time. The results show that samples treated at 1050 °C exhibited poorly sintered structure, resulting in poor properties. Ilic et al. [13] reported the effect of high temperature on the density, water accessible porosity, mineralogy and microstructure of sintered samples. SEM results indicate that pyroplastic effects caused pore formation and bloating at 1190 °C.

Collectively these studies focus on the effects of fly ash particle size distribution, process parameters (such as compaction pressure, sintering temperature, and sintering time), and mix

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proportions (clay as binder), on the properties of sintered products [8–14]. Although it is popular to use clay as a cheap and convenient binder to adjust the plasticity in ceramic or brick industry, clay is one kind of key and decreasing resources, which has close relation with farming especially in developing country. Actually, Chinese government has already enacted a law to prohibit using clay to fabricate sintered solid clay brick. Therefore, finding a good solution to substitute clay with the other resources is an urgent and serious problem. According to the industrial experiences of ceramic or brick, shale can be a better binder reacted with fly ash to produce qualified sintered product and reduce the amount of solid waste at the same time. However few researches focus on the effect of shale addition on properties of sintered fly ash. In this paper, these characteristics of sintered fly ash with or without shale were investigated by physical properties, X-ray diffraction and scanning electron microscopy.

2. Experimental procedures

2.1. Materials

Fly ash was obtained from the Wuhan Yangluo power plant in China. This fly ash is categorized as second grade ash according to the Chinese national standard GB/T 1596–2005. The 45 μm square sieve residue is 20%. Major mineral phases of this fly ash are quartz and mullite. Red shale, used as a binder, was supplied from the Qing Huangdao region in China. The 45 μm square sieve residue of this shale is 18%. Major mineral phases of the shale are quartz, gismondine and muscovite.

The chemical composition of the fly ash and shale are shown in Table 1.

2.2. Sample preparation

Three mixtures were selected for experiments, 100% fly ash (control sample), a mixture of 70% fly ash and 30% shale, and a mixture of 50% fly ash and 50% shale. If "A" is used to stand for fly ash, and "a" is for shale, the mixtures mentioned above can be referred to as A100, A70a30 and A50a50.

The mixtures were homogenized in a blender, and then aged with addition of water (18%, dry basis) at 20 °C for 72 h, all samples were compacted at 25 MPa for 30 s. Cylindrical "green" samples, 50 mm in diameter and 60 mm high, were dried at 105 °C for 10 h to drive off the moisture prior to sintering at high temperature. Then samples were sintered from 950 to 1100 °C at around 2 or 3 °C/min for a dwell time at the maximum temperature of 2 h before being allowed to cool to room temperature within the furnace.

2.3. Characterization of sintered specimens

Compressive strength was conducted on five samples of each mixture at 0.6 kN/s load rate, averages and coefficient of variation were computed to provide statistics and error estimates.

The apparent density and apparent porosity were measured using Archimedes' method. The apparent porosity was measured from the increase in weight after the sample was immersed in distilled water for 24 h.

Crystalline phases of the fly ash, shale and sintered samples were determined using X-ray diffraction (XRD, Rigaku D/Max–RB) at 50 mA, 40 kV Cu target. The microstructures of sintered samples were investigated by scanning electron microscopy (SEM, JEOL JSM–5610LV).

3. Results and discussion

3.1. Physical characterization

The compressive strength for sintered products is shown in Fig. 1. It is notable that the compressive strength was improved significantly with the increase of sintering temperature. When the temperature increased from 950 to 1050 °C, the compressive strengths of the A100, A70a30 and A50a50 increased from 4.5 to 11.2 MPa, from 7.7 to 28.3 MPa, and from 13.3 to 27.3 MPa, respectively. At the same temperature, the compressive strengths of samples with shale (A70a30 and A50a50) were significantly higher than that of pure fly ash samples (A100). In addition, the A50a50 was 5.6 MPa higher in compressive strength than the A70a30 at 950 °C. However, the strengths of sintered products differed little at temperature between 1000 °C and 1050 °C. All samples fired at 1100 °C resulted in unreliable strength due to bloating or expanding, therefore we cannot list them in this paper.

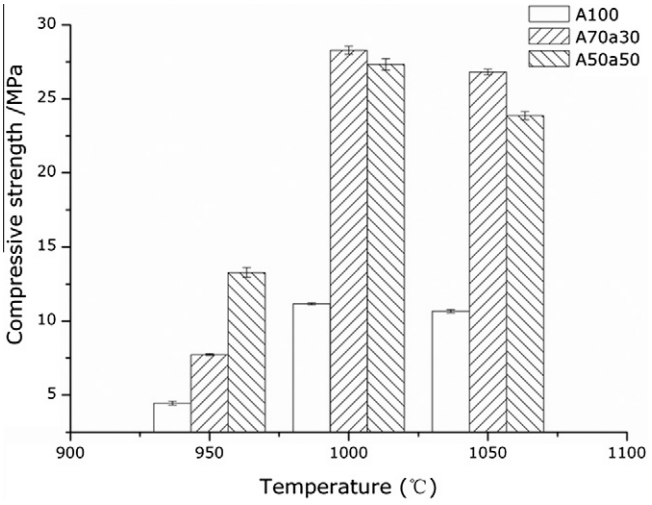

Fig. 1. Compressive strength of sintered product.

Fig. 2 indicates that there was a negative correlation between apparent porosity and sintering temperature. As the temperature increased from 950 to 1100 °C, apparent porosity of the A100 reduced by 19.2%; apparent porosity of the A70a30 and A50a50 reduced by 11.5%, and 10.8% respectively. Hence, apparent porosity of the A100 decreases significantly with a further increase in sintering temperature. It is pointed out that there was an increasing trend in the apparent porosity of the A50a50 from 1000 °C to 1100 °C, this results should be related with bloating or expanding of sintering samples.

Fig. 3 shows that the effect of sintering temperature on the apparent density of sintered fly ash. The apparent density gradually increased as the temperature was increased from 950 to

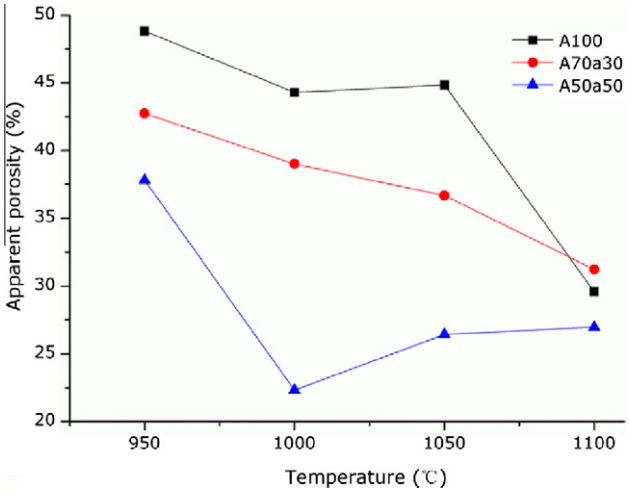

Fig. 2. Apparent porosity of sintered product.

Table 1
Chemical compositions for raw materials (W%).

Sample	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	Loss
Shale	59.04	19.29	8.83	0.22	2.59	5.10	0.25	0.02	4.65
Fly ash	54.76	32.74	4.56	4.27	0.65	1.57	0.30	0.92	0.23