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## Utilization of coal fly ash from a Chinese power plant for manufacturing highly insulating foam glass: Implications of physical, mechanical properties and environmental features

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### HIGHLIGHTS

- Large amounts of fly ash generated from power plants in Xinjiang.
- Low gaseous emissions from fly ashes.
- Manufacturing foam glass from fly ash/recycled glass mixture.
- Benign mechanical, thermal and acoustic insulation properties of foam glass.
- High application potential of foam glass with low environmental burden.

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### ABSTRACT

This research focuses on the potential use of fly ash (Waf) from a Chinese pulverized-coal combustion (PCC) power plant for manufacturing foam glass. The influences of fly ash properties, fly ash/recycled glass proportion in the raw materials, addition of fluxing and foaming agent, firing temperature, and residence time at the peak temperature on the final properties (apparent density, compressing strength and leachable potential of major and trace elements) of the foam specimens are investigated. The optimal foaming products were obtained consuming up to 33.3–43.3% fly ash, adding 9–11% Na<sub>2</sub>CO<sub>3</sub> fluxing agent and 0.5% SiC foaming agent, firing at 865–915 °C during 15 min residence time at the peak temperature. The obtained fly ash-based product revealed comparable compressive strength but slightly higher densities than commercial foam glass, with low CO<sub>2</sub> emissions and low leachable potential for environmentally relevant trace elements within the limits for inert/no-hazardous material established by the European Council Decision 2003/33/EC for land waste disposal. The large porosity of the foam glass indicates a high potential of thermal and acoustic insulation. The foaming behavior and the final properties the foam glass are strongly influenced by properties of fly ash (mainly fluxing temperature and organic C content), fly ash/recycled glass proportion in the raw materials, addition of Na<sub>2</sub>CO<sub>3</sub> fluxing agent and SiC foaming agent, firing temperature, and residence time at the peak temperature. The low C content and high refractory character of Waf give rise to poor foaming of the specimens when only fly ash and Na<sub>2</sub>CO<sub>3</sub> mixtures are used. The consumption of relatively high proportions of recycled glass and SiC is essential to produce foaming.

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### 1. Introduction

China is the world's largest coal market, producing and consuming the largest proportion of global coal supplies in the recent years. In 2016 the production and consumption of coal in China attained 16.86 Gt and 18.88 Gt oil equivalent, accounting for 46.1% and 50.6% of the global coal production and consumption,

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respectively [1]. The high coal consumption for energy and economic development, especially in pulverized-coal combustion (PCC) power plants is one of the most important emission sources of gaseous pollutants ( $\text{CO}_2$ ,  $\text{SO}_x$ ,  $\text{NO}_x$ ) and particulate matter. Large amounts of coal combustion products (CCPs), especially fly ash, are generated, and the majority are disposed in landfills, potentially resulting in environmental and economic burdens. On the other hand, fly ash has been successfully used for many years in a number of applications ranging from low added value to technological advanced applications, bringing substantial economic benefits [2–5]. Research on potential alternatives to develop high added value fly ash-based products is therefore of high environmental and economic relevance.

With the increasing energy demand in China, Xinjiang is expected to be the largest coal-producing and consuming province in China due to its enormous coal reserves and high coal quality, potentially producing large amounts of fly ash with low environmental concern [6,7]. The fly ash from Xinjiang power plants have been successfully used to synthesize zeolites, which can be used as slow-release fertilizer for plant growth in nutrient-limited soils or for soil reclamation [8]. These fly ashes have also been reported to be optimal as raw materials for manufacturing fire-resistant panels [9].

Given the high  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{CaO}$  contents in fly ash [10–13], special attention should be paid to potential used in foam glass preparation. Foam glass is a porous heat-insulating and soundproof material with true porosity of up to 90% vol. Compared to the polymeric foams currently employed, foam glass possesses significant advantages for thermal and acoustic insulation applications [14–16]. The excellent structural properties enable optimal performance as insulation in roofs, walls, and traffic areas such as flat roofs or floors. It is also used as industrial insulation for a number of minor uses such as sandwich panels, and it has been found to display optimal long term behavior in extreme environmental conditions due to its numerous advantages, such as, inert, incombustible, impermeable to moisture and steam, chemically stable, low density, long-time stable in physical characteristics, environmentally safe, and not subjected to degradation [14,17].

The production of foam glass may follow two distinct processes. The first method dating back to the 1930's consists of the direct introduction of gases ("blowing") into molten glass [18]. The second is the powder method, the essence of which consists in sintering a mixture of glass powders and special powder additives (foaming agents) facilitating the formation of a gaseous phase during the foam formation under firing [19]. When the temperature of the mixture exceeds the softening temperature, glass particles start sintering and form a continuous sintered body. Particles of the pore-forming agent become insulated by softening glass. Further heating prompts gas emission causing glass melt frothing and emerging pores throughout the sintered body where the particles of the pore-forming agent were blocked. Foaming agents are known to undergo two types of reactions i.e. decomposition or oxidation [11,15]. Decomposition reactions are associated with the presence of carbonate or sulfate foaming agents [15]. Intense gas release during their decomposition breaks the walls of individual pores favoring coalescence and creating a maze-like system of cavities in glass. On the other hand, oxidation reactions derive from the carbon-containing foaming agents, such as coke, anthracite, soot, graphite and silicon carbide (SiC), which react with oxygen present in the sintering furnace and produce gas emission from the glass melt. In general the powder method is the preferred approach for foam glass manufacturing given that (i) substantially lower temperatures are required for glass sintering in relation to those reported for the blowing method and (ii) the reuse of the crushed recycled glass brings environmental benefits. However an important issue associated with this application is the release

of  $\text{CO}$ ,  $\text{CO}_2$  or  $\text{SO}_2$  and therefore method optimization to minimize the emissions needs to be addressed.

The present study focuses on investigating the promising utilization of the fly ash collected from a power plant in Xinjiang for the manufacture of foam glass. The gaseous emission during heating and the fusion behavior of fly ash were examined given that these are key properties in foam glass preparation. Optimization of the foaming conditions was undertaken by producing foam glass using a range of proportions of fly ash, recycled glass and  $\text{Na}_2\text{CO}_3$  and SiC addition. The physical, mechanical, and environmental properties of the foam glass products were determined to evaluate their practical suitability as insulating materials.

## 2. Materials and methodology

### 2.1. Materials

A fine fraction of fly ash (WAF,  $d_{50} = 4 \mu\text{m}$ ) was collected from the WEI PCC power plant, in Xinjiang (NW China) and used as the main raw material for making foam glass at a laboratory scale. Recycled glass (true density  $\rho = 2.52 \text{ g/cm}^3$ ), which is another solid waste from a float glass factory, was also used as raw material. Recycled glass can provide sufficient amorphous phase, which is necessary for good heating insulation performance [20]. The glass fragments were ground with a hammer mill until obtaining glass power  $< 500 \mu\text{m}$ . The chemical composition and the estimated foaming C contents of fly ash, recycled glass and  $\text{Na}_2\text{CO}_3$  are summarized in Table 1. Detailed information on characteristics of WAF has been reported elsewhere [7]. It is worth noting that the relatively high organic C content (3.0%) in WAF, which is desirable to enhance gas-forming and pore-forming thus improving heat-insulating and soundproof properties of foam glass [11].

Appropriate doses of powdered  $\text{Na}_2\text{CO}_3$  were used as the fluxing agent to reduce the economic cost by lowering the melting temperature [21]. It is worth noting that  $\text{Na}_2\text{CO}_3$  decomposition during the firing process will produce  $\text{CO}_2$ . Given that  $\text{Na}_2\text{CO}_3$  has a low decomposition temperature when it melts at around  $851 \text{ }^\circ\text{C}$  [22], it may also act as a gas-forming agent in some cases when the glass-fly ash mixture melt occurs before  $\text{CO}_2$  is released from  $\text{Na}_2\text{CO}_3$ .

Powdered SiC ( $d_{50} = 9.3 \mu\text{m}$ ) was selected as foaming agent to enhance porous foam structure by means of gas release due to the oxidation reaction during the firing process. The foaming C in the mixtures derives from the addition of SiC foaming agent as well as from the fly ash itself.

### 2.2. Analyses of gaseous emission during heating and fusion behavior of fly ash

#### 2.2.1. Gaseous emission of fly ash

The study of emission from WAF subjected to thermal treatment was conducted using the TG-DSC-QMS-FTIR analytical technique. This consists of thermogravimetric (TG) analysis equipment with differential scanning calorimetry (DSC) fitted with a Fourier transform infrared (FTIR) and a quadrupole mass spectrometer (QMS). This device enables detection of acid emissions (e.g.,  $\text{CO}_2$ ,  $\text{SO}_x$ ,  $\text{NO}_x$ ,  $\text{HCl}$ , or  $\text{HF}$ ...) in powdered materials, under different atmospheres up to the peak temperature of  $1200 \text{ }^\circ\text{C}$ .

#### 2.2.2. Firing behavior of fly ash

To examine the firing behavior of the WAF, a complete fusion assay was carried out using a heating microscope MISURA. Accordingly, the shrinkage-temperature curve and the following characteristic temperatures were obtained from the recorded images:

- Onset of shrinkage ( $T_{15}$ ), considered as such, when the area of the silhouette of the specimen is 99% of the initial area.
- End of shrinkage ( $T_{ES}$ ), when the cylinder stops contracting.
- Softening ( $T_{SO}$ ), when the edges of the specimen begin to round.
- Sphere ( $T_{SP}$ ), when the morphology of the specimen is similar to a sphere.
- $\frac{1}{2}$  Sphere ( $T_{1/2}$ ), when the morphology of the specimen is similar to a hemisphere.
- Fusion ( $T_F$ ), when the morphology of the specimen resembles a spherical cap, equivalent to  $\frac{1}{3}$  the volume of a sphere.

### 2.3. Foam glass preparation process

Optimization of foam glass products was undertaken by preparing a number of mixtures containing different proportions of fly ash, recycled glass and  $\text{Na}_2\text{CO}_3$  (FG1, FG2, FG3, FG4, FG5, and FG6 with the fly ash/recycled glass/fluxing agent ratio in%weight of 33.3/66.7/0, 16.7/83.3/0, 8.4/91.6/0, 33.3/57.7/9.0, 79.2/0/22.8, and 43.3/45.7/11.0, respectively, Table 2). Addition of  $\text{Na}_2\text{CO}_3$  as fluxing agent in different proportions in FG4, FG5 and FG6 was investigated with the aim of increasing the fly ash dosage that can be potentially used whilst reducing the overall economic

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