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Bloating of fly ash-based lightweight aggregates

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KEYWORDS

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ABSTRACT

The present work characterized two types of fly ash, one from pulverized coal combustion and the other from fluidized bed combustion. The effects of fly ash origins, raw mix proportions, and sintering temperatures on the bloating of lightweight aggregates were studied. The raw mixtures were calculated based on Riley's SiO2 – Al2O3 – fluxing composition diagram. The exothermic combustion reaction of a large amount of unburnt coal causes overheating, which results in boiling and abnormal bloating behavior. Besides, raw mixture compositions outside Riley's composition margin perform efficient bloating with most pore sizes from 0.5 to 1 mm. Hence, besides the fluxing content, one should consider the amount of unburnt coal in the fly ash in synthesizing lightweight aggregates.

1. Introduction

In recent years, environmental concerns and mining restrictions have promoted the use of industrial waste, of which coal-fired power plant fly ash is the most significant due to its large emission and suitability as a raw material for a wide range of building materials. Non-firing recycling solutions, including concrete admixtures [1], fly ash bricks [2], geopolymers [3], and landfill materials[4], are most commonly applied in fly ash treatment. Since each application has specific quality requirements for the raw materials, fly ash that does not meet these requirements remains in the storage yard and becomes an environmental concern. Firing-associated recycling solutions are more expensive but can help deal with the remaining fly ash while adding value to this material. Examples of this approach include but are not limited to ceramic tiles [5], glass and glass-ceramics [6], and concrete lightweight aggregates (LWA) [7].

Originally, LWA was produced by firing bloating clays that meet two requirements: (1) there are one or more reactions that liberate a gas at elevated temperature, and (2) at that time, the material produces a glassy phase with a viscosity high enough to prevent the gas from escaping. Riley has established the limits of bloating on a SiO_2 – Al_2O_3 – fluxing composition diagram, in which the flux is the total weight percent of CaO, MgO, FeO, Fe_2O_3, and $(K, Na)_2O$ [8].

Several studies are devoted to preparing sintered lightweight aggregates (LWA) using fly ash as a primary raw material. Since the mineralogical and physical properties of fly ash are much different from those of the bloating clay, a binder and one or more fluxes must be added to the raw mix before shaping, followed by drying and quick-firing [9-12].

Although the combustion conditions have a significant influence on the mineralogical properties of fly ash, to the authors' knowledge, no studies have been conducted to elucidate the effects of the coal combustion technique on the sintering behavior of fly ash in LWA preparation. Therefore, the present study compares the sintering behavior of LWA raw mixes using fly ash from two plants that apply two different combustion techniques, pulverized coal combustion (PCC) and fluidized bed combustion (FBC).

2. Methodology

2.1. Fly ash characterization

Fly ash was taken from Ninh Binh and Mao Khe power plants. The former applies the PCC technique, while the latter employs the FBC technique. To understand the influence of origin on the properties of fly ash and subsequently on the properties of lightweight aggregates, the fly ash was characterized by scanning electron microscopy on a JEOL JSM 6360LV microscope, X-ray diffraction on a Bruker D2 Phaser diffractometer, laser particle size analysis on a HORIBA LA-350 Laser Scattering Particle Size Distribution Analyzer, and thermal analysis on a Linseis STA PT1600 instrument.

2.2. Lightweight aggregates preparation and characterization

The raw mixtures are prepared from fly ash, clay, limestone, and soda ash, with the chemical composition shown in **Table 1**. In addition to providing network-building and fluxing oxides, the clay also acts as a temporary binder. Limestone and soda provide fluxing oxides and generate carbon dioxide for bloating. The raw mixtures were calculated based on Riley's diagram (**Figure 1**) and are presented in **Table 2**. The clay and soda ash contents were 40% and 5%, respectively. The fly ash content varied from 35% to 55%, while limestone content varied from 20% to 0%. From the top down in each series in Figure 1, the content of fly ash decreases from 55 to 50, 45, 40, and 35%. After drying and grinding, the raw materials were mixed in a ball mill for an hour, then added water to a moisture content of 15%, just enough for shaping in a stainless steel mold. Pellets of D10 ×

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H10 mm size were dried overnight at 115 °C, then fired in an electric furnace. As gas generating reactions are expected during firing, the heating rate was kept at 33 °C/min from room temperature to 400 °C, 17 °C/min from 400 to 900 °C, and 33 °C/min from 900 °C to the highest firing temperature (1100 – 1200 °C). The samples were maintained at the highest temperature for 5 minutes before being taken out of the furnace.

Table 1. Raw materials chemical compositions

Materials	Chemical composition (wt.%)								
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	LOI
Clay	63.20	19.10	5.56	0.14	0.80	2.52	0.22	0.97	6.46
Limestone	0.11	0.31	0.01	52.50	0.43	0	0	0.71	41.20
Soda ash	0.05	0.05	0	0.08	0	0	56.50	0	36.30
Mao Khe (FBC) fly ash	47.98	25.33	6.31	3.35	1.25	3.60	0.64	0.71	7.97
Ninh Binh (PCC) fly ash	45.20	19.99	6.86	1.52	1.29	3.13	0.32	0.57	20.15

Table 2. Raw mix composition

Rax mix label	Raw mix composition (wt.%)					LOI	$SiO_2 – Al_2O_3 –$ Fluxes equivalent (wt.%)		
	Clay	Limestone	Soda ash	Mao Khe fly ash (FBC)	Ninh Binh fly ash (PCC)		SiO_2	Al_2O_3	Flux
FBC35	40	20	5	35	0	15.43	49.78	19.59	30.63
FBC40	40	15	5	40	0	13.77	51.59	20.67	27.74
FBC45	40	10	5	45	0	12.11	53.34	21.70	24.96
FBC50	40	5	5	50	0	10.44	55.02	22.69	22.28
FBC55	40	0	5	55	0	8.78	56.65	23.65	19.70
PCC35	40	20	5	0	35	19.69	51.21	18.31	30.49
PCC40	40	15	5	0	40	18.64	53.32	19.28	27.41
PCC45	40	10	5	0	45	17.59	55.37	20.23	24.40
PCC50	40	5	5	0	50	16.53	57.37	21.15	21.48
PCC55	40	0	5	0	55	15.48	59.33	22.05	18.62

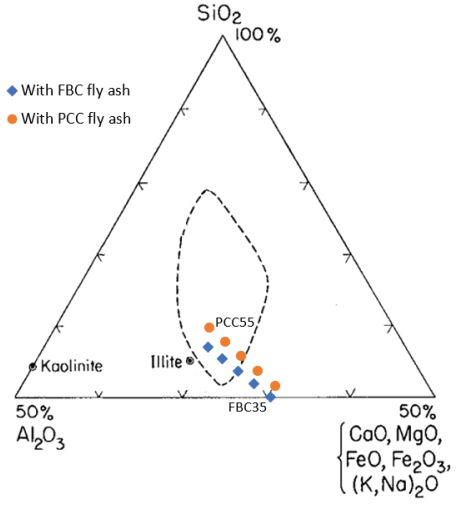


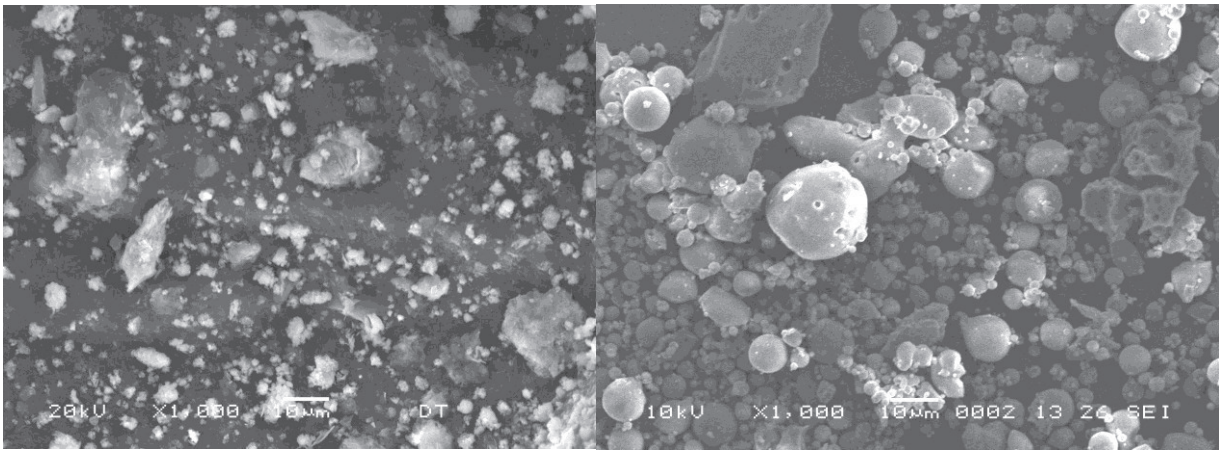
Figure 1. Raw mix chemistry on the Riley's diagram.

3. Results and Discussion

3.1. Properties of the fly ash

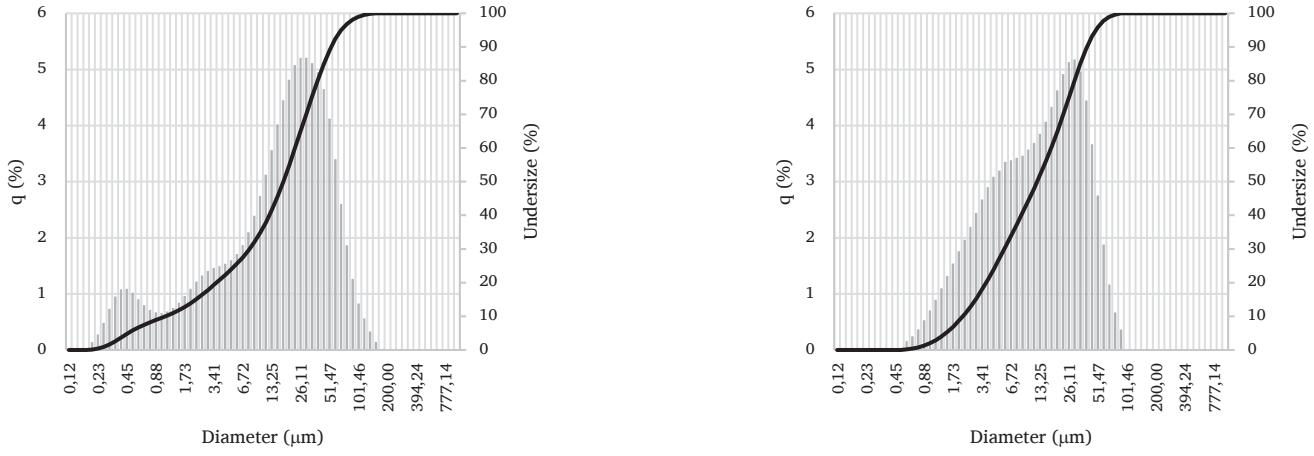
Mao Khe FBC fly ash particles are very irregular in shape, while Ninh Binh PCC fly ash particles are primarily spherical (Figure 2). This morphological difference is due to the combustion conditions. FBC boilers operate at typically around 900 °C, which is not high enough for melting the ash. On the other hand, PCC boilers work at over 1400 °C, which is high enough to cause partial melting of the ash particles followed by rounding under surface tension. Ninh Binh PCC fly ash also contains irregular-shaped particles attributed to unburnt coal that causes high loss-on-ignition (LOI).

Mao Khe FBC fly ash performs a broader particle size distribution range than Ninh Binh PCC fly ash. The particle sizes of Mao Khe FBC fly ash range from 0.2 to 174.6 μm, with 90% having a size below 58.9 μm (Figure 3-a). It performs a trimodal distribution with the maxima peaks at 0.4 μm, 3.4 μm, and 34.2 μm. The particle sizes of Ninh Binh PCC fly ash range from 0.6 to 88.6 μm, with 90% having a size below 40.0 μm (Figure 3-b). It performs a bimodal distribution with the maxima peaks at 6.7 μm and 29.9 μm.



(a) (b)

Figure 2. SEM images of (a) Mao Khe FBC fly ash and (b) Ninh Binh PCC fly ash.



(a) (b)

Figure 3. Particle size distribution of (a) Mao Khe FBC fly ash and (b) Ninh Binh PCC fly ash.

The combustion temperature also greatly impacts the mineralogical properties of the fly ash. As presented in Figure 4, Mao Khe FBC fly ash contains quartz, phengite, and hematite, while Ninh Binh PCC fly ash contains quartz and mullite. Mullite is the crystalline phase that forms at temperatures above 1000 degrees Celsius, and the formation of mullite is accelerated more rapidly in the presence of the liquid phase.

A large amount of unburnt coal in Ninh Binh PCC fly ash is not only reflected via a high loss-on-ignition in chemical analysis (Table 1) and SEM imaging (Figure 2) but also in thermal analysis (Figure 5). Up to 900ºC, the total weight loss of Ninh Binh PCC fly ash is 22.3%, while that of Mao Khe FBC fly ash is just 9.4 %. Those weight losses include about 2% of the initial moisture content. In DTA analysis, Ninh Binh PCC fly ash performs a higher and broader exothermic peak than that of Mao Khe FBC fly ash. This difference is due to the combustion reaction of a higher content of unburnt coal upon heating in the air. Because the volatiles may already be evaporated in the boiler, the unburnt coal becomes more difficult to burn, extending the exothermic peak to 900 ºC.

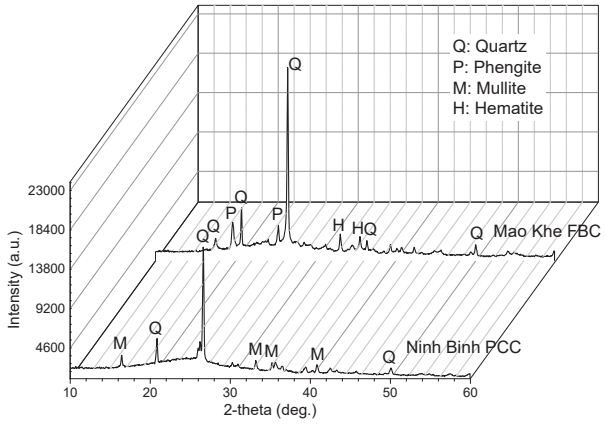


Figure 4. XRD of (a) Mao Khe fly ash and (b) Ninh Binh fly ash.

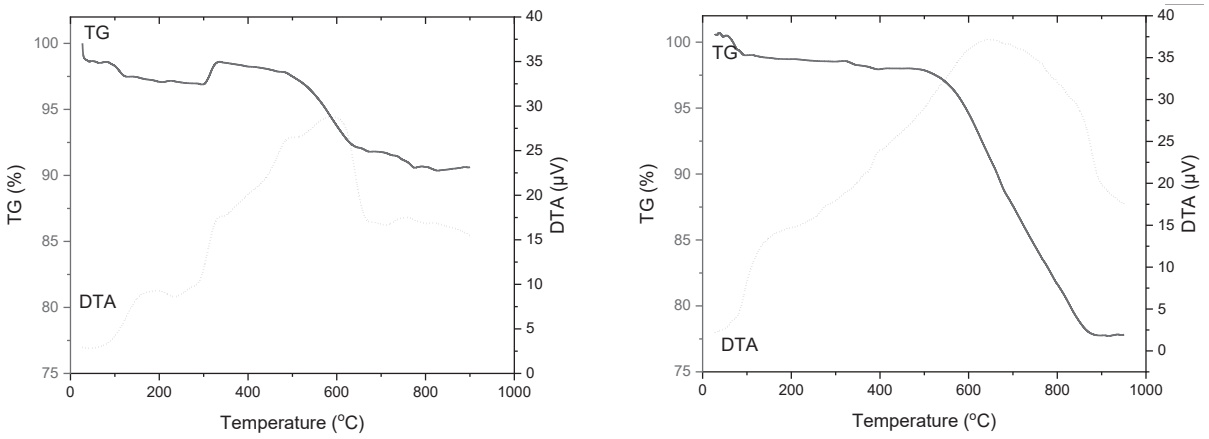


Figure 5. Thermal analysis of (a) FBC fly ash and (b) PCC fly ash.

3.2. Bloating of the lightweight aggregates

Except for PCC55, all the samples shrank after firing at 1100 °C (Figure 7). Samples in the FBC series get less glossy while those in the PCC series get more glossy as the fly ash content increases from 35% to 55%, equivalent to decreasing the flux content from ~ 30% to ~ 19%. With less than 50% fly ash content, samples in the PCC series shrink more than samples in the FBC series. With 50% and 55% fly ash content, samples in the PCC series shrink less, even expanded, than those in the FBC series. This opposite trend is attributed to unburnt coal combustion that raises the temperature inside the samples. As a result, liquid phase formation is promoted in the PCC series, and slight bloating occurs. In addition, samples fired at 1100 °C get darker as the fly ash content increases (Figure 6). This phenomenon is induced by fly ash unburnt coal, facilitating iron-containing compounds' redox reactions. The formation of iron (II) compounds changes the samples' color and lowers the liquid phase formation temperature and viscosity [13]. Therefore, unburnt coal significantly impacts bloating when present at a considerable amount.

Figure 6. Bloating of aggregates fired at 1000 °C and 1200 °C.

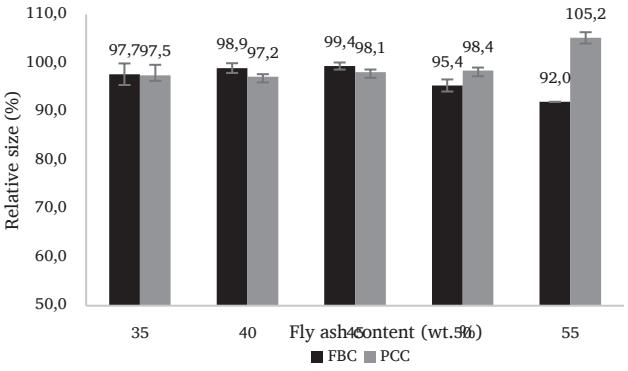


Figure 7. The relative size of the pellets fired at 1100 °C.

When calcined at 1200 °C, all samples in the FBC series bloat except for those containing 45% fly ash. FBC35 and FBC40 are rounded and bloat with a smooth surface, while FBC50 and FBC55 bloat but remain their cylindrical shape. Significantly, the surface of FBC55 samples shows traces of deflated boiling bubbles that originate from overheating due to the combustion of unburnt coal. At the same firing temperature, samples in the PCC series show a flatter deformation and darker color. Traces of boiling bubbles are demonstrated by large open pores, indicating that the sample temperature is higher than the furnace temperature due to the exothermic combustion reaction of the unburnt coal in the fly ash.

Of particular interest is FBC35 fired at 1200 °C because it has a spherical shape, smooth surface and its raw mixture lays furthest from the composition margin suggested by Riley. The fracture surface of this sample exhibits a dense presence of closed pores, most of them about 0.5 to 1 mm in diameter, some up to 2.5 mm in diameter (Figure 8). The X-ray diffraction pattern of the sintered aggregate shows the predominant amorphous phase (Figure 9). The diffraction peaks demonstrate that the only crystalline phase appearing on this diagram is anorthite (Al2 Ca O8 Si2 #PDF 96-100-0035).

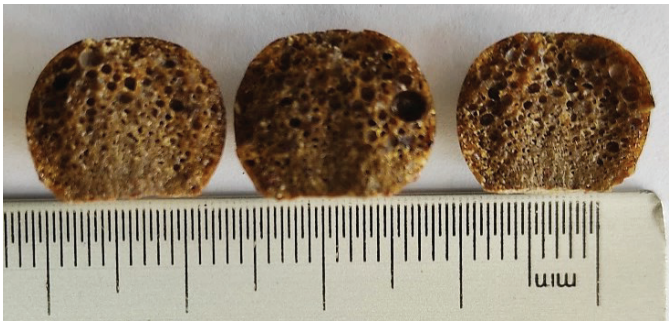


Figure 8. Fracture surface of FCB35 fired at 1200 ºC.

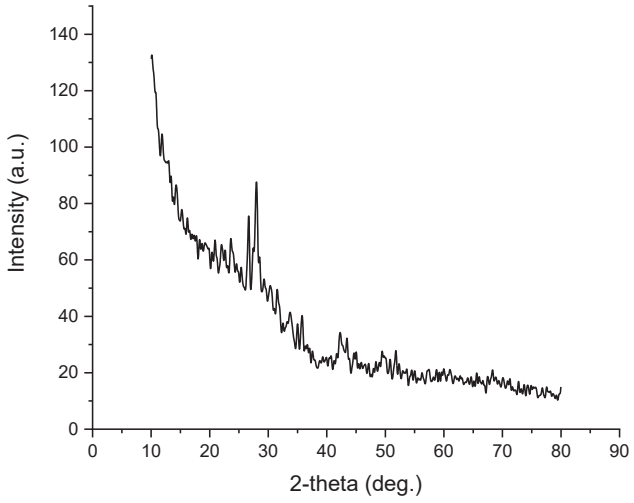


Figure 9. XRD of the FCB35 fired at 1200 ºC.

4. Conclusion

The PCC fly ash used in the present study has a high unburnt coal content, reflected by a high loss-on-ignition, a large exothermic peak in thermal analysis, and large irregular-shaped particles in SEM imaging. This high unburnt coal content has a great impact on the bloating behavior of the aggregates as the combustion of this unburnt coal raises the temperature of the samples, promoting liquid phase formation and lowering the liquid phase viscosity. Therefore, raw mixture calculation for fly ash-based LWA should consider both the fluxing oxide content and the unburnt coal content.

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