



Alkali-activation potential of biomass-coal co-fired fly ash



Christopher R. Shearer^{a,*}, John L. Provis^b, Susan A. Bernal^c, Kimberly E. Kurtis^a

^a School of Civil and Environmental Engineering, Georgia Institute of Technology, 790 Atlantic Dr., Atlanta, GA 30332-0355, USA

^b Department of Materials Science and Engineering, The University of Sheffield, Sir Robert Hadfield Building, Mappin Street, Sheffield, S1 3JD, United Kingdom

^c Department of Civil and Structural Engineering, The University of Sheffield, Sir Frederick Mappin Building, Mappin Street, Sheffield, S1 3JD, United Kingdom

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ABSTRACT

Co-fired fly ash, derived from the co-combustion of coal and biomass, is examined as a potential precursor for geopolymers. Compared to a coal fly ash, two co-fired fly ashes have a lower vitreous content and higher carbon content, primarily due to differing combustion processing variables. As a result, binders produced with these co-fired fly ashes have reduced reaction potential. Nevertheless, compressive strengths are generally highest for all ashes activated with solutions with a molar ratio of $\text{SiO}_2/(\text{Na}_2\text{O} + \text{K}_2\text{O}) = 1$, and these mixes reach the highest extent of reaction among those studied. Activation with sodium hydroxide solution forms zeolitic phases for all ashes. The thermal and dilatometric behavior of the coal and co-fired fly ash geopolymers is similar between equivalent mix designs. These results indicate that co-fired fly ashes can be viably used to form alkali-activated geopolymers, which is a new beneficial end-use for these emerging waste materials.

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1. Introduction

The practice of co-firing coal with biomass has increased worldwide as a result of stricter emissions regulations and legislation, because biomass combustion can be considered a CO₂-neutral process when the amount of CO₂ released into the atmosphere is less than or equal to the amount of CO₂ taken up by the plants during their lifetime [1]. In the U.S., biomass electricity generation is expected to increase by an average of 3.1% each year through 2030, primarily by co-firing biomass at existing coal power plants [2]. As more coal power plants convert to co-firing, it is critical for industries that beneficially reuse coal fly ash to recognize and understand the potential implications of these changing combustion practices on the quality of fly ash. The concrete industry is currently the primary user of coal fly ash as a supplementary cementitious material, but co-fired fly ash—which is the main by-product of the co-firing process—is not currently

addressed in the U.S. standard for fly ash use in concrete, ASTM C618-15 [3]. However, co-fired fly ash is permitted for use in concrete with additional restrictions under the European standard (EN 450-1) [4]. Research to find other pathways to utilize this emerging waste stream is undoubtedly warranted, given its projected increase in production in place of pure coal fly ash in many countries [5].

One potential reuse application is to generate alkali-activated geopolymers, which are sustainable binders that can be used as a cost-effective and durable alternative to portland cement concrete [6]. Since coal fly ash is one of the main precursors used for alkali-activated geopolymer production, this research examines the use of co-fired fly ash—which can have comparable chemical compositions and physical properties to coal fly ash—for geopolymerization. The influence of co-firing on geopolymer gel properties is relatively unknown. One study examined the zeolitic formation in two alkali-activated co-fired fly ashes (produced by firing coal with sunflower hulls and an unknown biomass) [7], while another study geopolymerized co-fired fly ash derived from the combustion of a blend of coal, wood and palm pits, with addition of blast furnace slag, to produce high strength mixes [8]. Pure biomass ash from the combustion of eucalyptus forest waste was also successfully geopolymerized, but the strengths of the resulting geopolymer products were relatively low [9].

* Corresponding author. Present address: Department of Civil and Environmental Engineering, South Dakota School of Mines and Technology, 501 E. Saint Joseph St., Rapid City, SD 57701-3901, USA.

E-mail addresses: chris.shearer@sdsmt.edu (C.R. Shearer), j.provis@sheffield.ac.uk (J.L. Provis), s.bernal@sheffield.ac.uk (S.A. Bernal), kimberly.kurtis@ce.gatech.edu (K.E. Kurtis).

Co-firing can yield fly ashes with reduced glassy phase contents and higher phosphate, alkali and magnesium levels compared to coal fly ash [10], depending on the biomass source and combustion conditions. Biomass ash compositions are also more variable than coal fly ashes (i.e., biomass ashes have a wider distribution of chemical and physical characteristics) [11]. This is important because biomass ash constitutes part of the co-fired fly ash, and its variability may present challenges when developing viable geopolymer mix designs. Moreover, many of the coal power plants initially being converted to co-firing facilities in the U.S. are smaller and less efficient, which could result in ashes with a higher unburned carbon content ultimately affecting their suitability for alkali-activation. However, this may not be an issue when more efficient processes are used during co-firing. Since the composition of the precursor material has a significant impact on the reaction potential and formation of geopolymeric products [6], more research is needed to understand the implications of co-firing on the properties of geopolymer binders.

In this study, three ashes—one commercial-quality coal fly ash and two co-fired fly ashes produced by burning coal with wood chips—were alkali-activated and examined using strength testing, infrared spectroscopy, X-ray diffraction (XRD), thermogravimetric analysis (TGA), dilatometry and scanning electron microscopy (SEM). Co-fired fly ash properties are linked to binder properties to better understand how potential changes in their composition may impact the fundamental characteristics of geopolymers. This research is necessary before co-fired fly ash can be utilized in future low-carbon infrastructure, and is important for understanding the future of geopolymer technology as the U.S., Europe and other regions move increasingly to biomass co-firing as a fuel source for electricity generation and as the availability of coal fly ash decreases.

2. Experimental methods

2.1. Materials

Three different ash samples were alkali-activated in this study, one derived from ordinary coal combustion and two derived from co-firing (i.e., produced from the co-combustion of coal and biomass during full-scale trials). The sample names in Table 1 use the general notation “X-YA-##”, where “X” is the plant at which the sample was produced, “YA” is the type of ash (i.e., FA is coal fly ash and CA is co-fired fly ash) and “##” indicates the replacement percentage (by weight) of coal with biomass in the wet feedstock. B-CA-4 and C-CA-15 were produced by co-firing bituminous coal with pine chips and hardwood chips, respectively. A-FA-0 is a commercially available coal fly ash produced by firing bituminous coal. The elemental compositions of these ashes are listed in Table 2, and were measured by X-ray fluorescence (XRF) on a Philips PW-2400 instrument. Loss on ignition (LOI) was determined gravimetrically using a LECO TGA 601. The ash median particle sizes and particle size distributions shown in Fig. 1 were obtained by dry-dispersion laser diffraction (Sympatec RODOS T4.1 Particle Size Analyzer). The Brunauer–Emmett–Teller specific surface area (BET

Table 1
Fly ash sample nomenclature.

Attribute	A-FA-0	B-CA-4	C-CA-15
Combustion plant	A	B	C
Ash type	Coal	Co-fired	Co-fired
Coal type	Bituminous	Bituminous	Bituminous
Biomass type	–	Pine chips	Hardwood chips
Biomass weight percentage (%)	0	4	15

Table 2
Fly ash properties.

Composition (%) or physical characteristic	A-FA-0	B-CA-4	C-CA-15
LOI	1.4	5.5	15.0
TOC	1.1	4.6	13.6
SiO ₂	55.3	42.8	47.1
Al ₂ O ₃	27.2	26.2	23.1
Fe ₂ O ₃	8.0	13.9	8.7
Σ(SiO ₂ + Al ₂ O ₃ + Fe ₂ O ₃)	90.5	82.9	78.8
CaO	1.3	4.0	0.8
MgO	1.2	1.2	0.8
SO ₃	0.1	0.4	0.1
Na ₂ O	0.5	0.5	0.3
K ₂ O	3.0	2.1	2.2
Na ₂ O _{eq}	2.5	1.9	1.7
P ₂ O ₅	0.2	0.7	0.5
TiO ₂	1.4	1.3	1.4
Mn ₂ O ₃	0.1	0.1	0.0
Cr ₂ O ₃	0.1	0.1	0.0
Sum total of XRF	99.6	98.8	99.9
Moisture content	0.2	0.3	0.5
Median particle size (μm)	16.2	26.9	11.4
BET SSA (m ² /g)	1.3	2.5	6.1
Rietveld quantification of XRD data (%)			
Quartz	14.1	10.5	10.6
Mullite	17.4	26.5	20.6
Iron oxide crystalline phases	1.1	6.1	2.5
Amorphous content (total)	67.5	56.9	66.3
Amorphous content (excluding carbon)	66.4	52.3	52.7

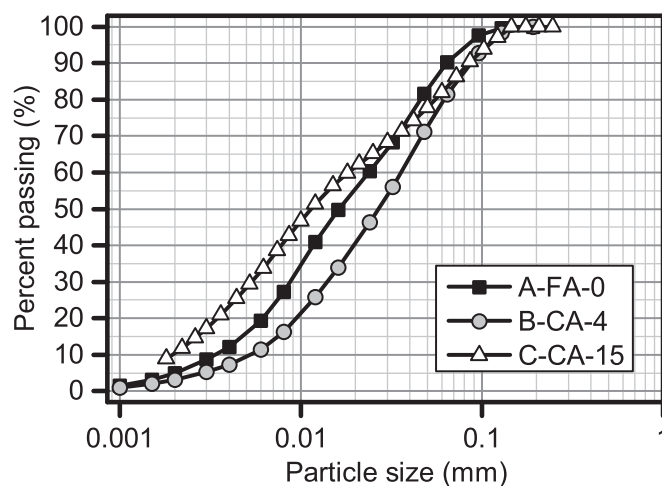


Fig. 1. Fly ash particle size distributions.

SSA) of each ash was measured by nitrogen adsorption at 77 K using a Micromeritics ASAP 2020 Physisorption Analyzer. XRD results measured with a Bruker D8 using an internal standard (rutile) were quantified by the Rietveld method [12]. Total organic carbon content was measured with a Shimadzu TOC analyzer. These properties are listed in Table 2.

All three ash samples are consistent with the category of Class F ashes under ASTM C618 on the basis of their primary oxide contents (i.e., the sum of SiO₂, Al₂O₃ and Fe₂O₃ is greater than 70%). B-CA-4 has 4% CaO, which is the highest among these ashes. A-FA-0 has the highest aluminosilicate and amorphous content, which indicates it is primarily composed of a glassy phase. B-CA-4 and C-CA-15 have high carbon contents, which for C-CA-15 is above the 6% limit stipulated in ASTM C618. B-CA-4 and C-CA-15 also have lower amorphous glassy contents compared to A-FA-0, after subtracting the amorphous phase associated with organic carbon in

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