Characteristics of Australian brown coal fly ash blended geopolymers

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HIGHLIGHTS

- Usage of Australian brown coal fly ash as geopolymer source material.
- Blending brown coal fly ash with slag and class F fly ash enhances strength.
- Sulphate present in brown coal fly ash may adversely affect geopolymers.

ARTICLE INFO

ABSTRACT

The present study evaluates the potential re-use options for a suite of low rank brown coal fly ashes (ASTM class C fly ash) as feedstock for geopolymer binder synthesis. Ash suitability was evaluated using a number of conventional characterisation tools including XRF, calorimetric and microscopy to analyse solid phase distribution in relation to compositional differences derived from ternary diagram analogues. Blends of brown coal ash with selected black coal fly ash (class F) and blast furnace slag were developed for compressive strength and durability tests. The study show that brown coal fly ash with higher Al content improved the rate of reaction of blended binders and that observed high contents of sulphate and magnesium oxides available in some brown coal fly ashes induced accelerated decomposition in high moist environments. Overall, it was evident from both microscopy and mineralogical analysis of binder mixtures that brown coal fly ash participate in geopolymer network formation rather than act as filler material. Factors influencing brown coal ash reactivity and the potential for incorporating this class of ash in developing stable and usable geopolymer binder systems are discussed.

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1. Introduction

Approximately 90% of electricity in the state of Victoria, Australia is generated by burning of brown coal (65 million tonnes per year) deposits mined from the Latrobe Valley in Victoria [1]. As a result 1.13 million tonnes of ash by-products are discharged into landfills with minimal utilisation. Fly ash produced from Brown coal (typically derived from younger sub-bituminous coal sources) has different chemistry from conventional black coal fly ash (ASTM class F) due to the characteristics of coal. Therefore, current usage of this material as a supplementary cementitious material is restricted resulting in high amount of ash deposits with environment consequences [2].

Geopolymer is a novel binder that can be used to replace Portland cement binder [3]. Alkali-activated binder or geopolymer can generally be prepared by using a wide range of aluminate–silicate feed stock materials. Source material for geopolymer can be by-product obtained from industrial processes such as fly ash, slag, red mud and rice husk ash (RHA) or from geological resource like metakaolin [4,5]. More recently, researchers have started to focus on other non-traditional materials such as lower grade clays, have shown that the properties of final geopolymer are quite often difficult to control [6]. Traditionally, class F fly ashes have been identified as the preferred source material for geopolymer binders given their chemical composition, amorphous content and availability [4,5]. The primary difference of classes F and C fly ashes is that class C typically contains 20% higher CaO content compared to class F fly ash. Brown coal fly ash, generally classified as class C fly ash type although it differs somewhat from conventional class C ash characteristics owing to its genesis from lower rank coal types compared to lignite. Altogether, very limited work has been done on geopolymer binder synthesis using brown coal fly ash [7].

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Bankowski et al. [8] investigated the use of brown coal fly ash geopolymer binders as containment matrices to reduce the leaching of heavy metals in land fill applications. Metakaolin was incorporated in their geopolymer synthesis because the brown coal fly ash had limited amounts of reactive aluminium and silicon. This study however, did not provide data on engineering properties of brown coal fly ash based geopolymer binders investigated. The authors stated that utilisation of 100% Victoria, Australia brown coal fly ash in geopolymer systems remained unsatisfactory. More recently, Law et al. [9] prepared geopolymer mortars based on Australian brown coal dry precipitator fly ash. Their binders attained compressive strengths of 50 MPa in 7 days demonstrating potential for use of this material in geopolymer systems. However geopolymer binders in the latter study were produced using high alkaline systems (15 M) and high curing temperatures (90°C), representing processing conditions over and above general concreting practise. The researchers also did not analyse the phase evolution and reaction mechanisms of brown coal fly ash based geopolymers deemed essential to understanding key requirements for binder mix design and system properties.

In the present work, four types of brown coal fly ashes (class C fly ash) were evaluated in order to assess mineralogical and chemical suitability for geopolymer matrix formation. Various Geopolymer formulations were developed based on fly ash mineralogy to monitor binder compressive strength development as well as solid phase evolution.

2. Experimental analysis

2.1. Materials

Brown coal fly ashes used in this work were sourced from, the La-Trobe Valley, Victoria Australia. Three types of brown coal fly ashes were collected directly from the precipitators in dry condition while Loy Yang lagoon fly ash was collected from the ash ponds in a wet state and class F fly ash was obtained from Gladstone power station Queensland, Australia. Ground granulated blast furnace slag was supplied by Independent Cement Australia Pty Ltd. Table 1 provides detailed chemical composition and source origin of each fly ash type. In the paper, the notations are used thereafter for the fly ashes and slag as shown in Table 1 (example: Loy Yang Lagoon ash-LYLG).

A commercial D grade sodium silicate solution (29.4% SiO_2 and 14.7% Na_2O by weight) from PQ Australia and 8 M sodium hydroxide solution was used throughout this study. The mass ratio between Na_2SiO_3 and NaOH was kept constant at unity. Feedstock blending was employed in order to achieve target SiO_2, Na_2O, CaO and Al_2O_3 oxide compositions and target SiO_2/Al_2O_3 (2.5–4.5 M

ratio) and Na_2O/Al_2O_3 (1.5–2.5 M ratio) oxide ratios. Thus, brown coal fly ash was mixed with class F fly ash and slag according to blending formulations given in Table 2. The water content was calculated considering the water in 8 M concentrated solution has 73.8% water and D-grade sodium silicate solution has 55.5% water. The blends were mixed with alkali activator solutions in a small mortar mixer. It should be noted that LYLG ash was dried in an oven prior to synthesis in order to remove moisture.

Alkali activated paste specimens prepared on equal flow basis were cast in 50 mm cube moulds. All specimens were cured for 24 h at 60°C (100% RH) and kept thereafter in air tight containers under ambient condition at 23°C temperature before compressive strength tests 7 days. The specimens which showed better compressive strength were prepared again and cured at 60°C for 8 h (100% RH). These latter set of specimens were used to measure compressive strength up to 120 days. Compressive strength test was conducted using an ELE International Universal Tester, at a loading rate of 1.0 kN/s for the 50 mm cubes.

2.2. Analytical techniques

Microstructural and chemical analyses were performed on raw materials or pastes samples after 1, 7 and 28 days of curing, through:

– The XRD analysis was carried out using a Bruker D8 Advance X-ray diffractometer. Scans were collected between 5° and 70° (2θ) with a step size of 0.02° and a scan rate of 5 s per step. An internal standard (10%wt corundum, Al_2O_3) was added to allow quantitative analysis using Rietveld refinement. Phase identification was completed using Materials Data, Inc., Jade 9.3 software and Quantitative Rietveld analysis using Bruker Diffracplus Topas software.
– Particle size of the materials was obtained using Cilas laser diffraction particle analyser. 1–10 g of fly ash was dispersed in water in order to get particle size.
– Isothermal calorimetry experiments were conducted using a TAM Air isothermal calorimeter, at a base temperature of 25 ± 0.02°C. Fresh paste was mixed externally, weighed into an ampoule, and immediately placed in the calorimeter, and the heat flow was recorded for the first 140 h of reaction. All values of heat release rate are normalised by total weight of paste.
– Environmental scanning electron microscopy (ESEM) with energy dispersive X-ray (EDX) analysis, at an accelerating voltage of 15 kV using an FEI Quanta ESEM instrument. Paste samples were sectioned, and polished up to 1 μm surface fineness using diamond paste.

Table 1
Bulk chemical composition of source materials.

Chemical composition	Loy Yang dry precipitator ash (%) LYDP	Loy Yang lagoon ash (%) LYLG	Hazelwood dry precipitator ash (%) HZDP	Yallourn dry precipitator ash (%) YLDP	Granulated blast furnace slag (%) SL	Class F fly ash (%) GFA
SiO_2	48.90	41.51	2.18	0.81	32.38	51.88
Al_2O_3	15.16	31.61	1.21	1.27	12.24	25.92
CaO	2.44	2.12	25.01	6.81	44.04	4.35
MgO	5.38	7.27	20.26	19.53	5.13	1.54
Fe_2O_3	9.68	5.87	13.30	39.35	0.49	12.66
MnO	0.05	0.04	0.34	0.45	0.37	0.15
K_2O	0.41	0.56	0.34	0.72	0.33	0.71
Na_2O	4.86	3.19	4.08	9.37	0.22	0.78
TiO_2	1.58	2.12	0.17	0.06	0.51	1.30
SO_3	7.48	0.46	13.37	19.82	4.21	0.24
LOI	4.04	5.05	19.76	1.81	0.08	0.55
Total	100	100	100	100	100	100