



Distribution of oxides in fly ash controls strength evolution of geopolymers



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HIGHLIGHTS

- Geopolymer synthesized from six different types of Australian fly ash.
- Uniform distribution of oxides in fly ash is the key role of synthesis of geopolymers.
- Geopolymers with same SiO₂ and Al₂O₃ distribution pattern have higher strengths.

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ABSTRACT

Compressive strength of six types of geopolymer pastes made from different fly ash sources were studied. Alkali activation was performed by a mixture of sodium silicate and sodium hydroxide solution (NaOH). Concentration of NaOH for all mixtures was 8 M. Specimens were cured at 60 °C and 100% RH for 24 h. The effect of different parameters on compressive strength was studied via contour analysis. Microstructural analysis of both fly ashes and geopolymers reveals that strength evolution mainly depends on the distribution of SiO₂ and Al₂O₃ in the starter fly ash rather than their ratios. Localized short order diffusion for geopolymerization necessitates both uniform distribution of SiO₂ and Al₂O₃ in the whole fly ash powder, and presence of these oxides in approximately same pattern.

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1. Introduction

Alkali activated binders (Geopolymers) are made from different silica rich raw materials. These substances which contain a moderate level of alumina are called aluminosilicate sources. Fly ash is the most attractive aluminosilicate source for production geopolymers and is widely used for construction technology as a possible replacement of ordinary Portland cement (OPC) [1–5]. However, as a result of different characteristics of fly ash sources, scattered results are achieved. Therefore, it is important to conduct a study where different types of fly ash sources are compared. In this study, six types of class F fly ashes widely produced in Australia are considered as a possible source of production geopolymers.

The most important parameters influencing compressive strength of geopolymers are particle size distribution of fly ash, weight ratio of constituent oxides such as Al₂O₃, SiO₂ and Na₂O, liquid to solid weight ratio, NaOH concentration, NaOH to sodium silicate weight ratio, CaO content, pore size distribution and age of

geopolymer. While the effect of some parameters on compressive strength is established well, other parameters have not a constant effect and vary due to different ways of geopolymerization.

Ryu et al. [6] studied the effect of different parameters on compressive strength of geopolymers. They showed that compressive strength increases when higher concentrations of NaOH are used. Furthermore, increasing of Al₂O₃/Na₂O and SiO₂/Na₂O weight ratios at a constant sodium silicate to NaOH weight ratio negatively impact on compressive strength. On the other hand, when the ratio of sodium silicate to NaOH is changed, Al₂O₃/Na₂O and SiO₂/Na₂O weight ratios have a different effect on compressive strength. Songpiriyakij et al. [7] studied properties of geopolymers made from a mixture of fly ash and rice husk bark ash. Their results were completely contradictory to those obtained by Ryu et al. [6]. Songpiriyakij et al. [7] found that increasing of Al₂O₃/Na₂O and SiO₂/Na₂O weight ratios positively affect on compressive strength evolution. Variable effects of Si/Al and content of Na in fly ash based geopolymers has been reported by Zheng et al. [8] as well. Álvarez-Ayuso et al. [9] have mentioned that the effect of NaOH concentration on compressive strength of geopolymers depends on the type of utilized fly ash. While in most cases increasing of

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NaOH concentration results compressive strength to rise, in some cases it may have an optimum condition. This has been mentioned in Chindaprasirt et al. [10] study as well for high calcium fly ash based geopolymers. Sathonsaowaphak et al. [11] have obtained an optimum for liquid to solid ratio to attain maximum compressive strength in bottom ash geopolymers. Their interesting result is the negative impact of bottom ash particle size on compressive strength which disagrees with the most findings for this factor.

Rickard et al. [12] studied possibility of production of geopolymers by five types of Australian fly ashes (four of them are similar to those used in this study). Several material science features of these geopolymers have been studied in that paper. However, compressive strength evolution and the reasons behind these changes in different types of fly ashes have not been studied in depth. In the present work, attempts have been made to evaluate the effect of each parameter on strength evolution of these materials.

2. Experiment procedure

2.1. Materials

Six types of class F fly ashes obtained from six different Australian power stations including Gladstone, Collie, Mt Piper, Eraring, Tarong and Bayswater were used as main aluminosilicate source materials in this study. D-Grade sodium silicate solution (29.4% SiO₂ and 14.7% Na₂O by weight) from PQ Australia and analytical grade NaOH solid from Sigma Aldrich were used as alkaline activators.

2.2. Characterization of fly ashes

The overall chemistry and mineralogy of fly ashes were analyzed by XRD and XRF techniques. XRD analysis was carried out using a Bruker D8 Advance X-ray diffractometer. Scans were collected between 5° and 70° of 2θ with a step size of 0.02° and a scan rate of 5 s per step. An internal standard (10 wt.% corundum, Al₂O₃) 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 Diffrac^{plus} Topas software. The chemical compositions of the fly ashes are given in Table 1 while Table 2 shows percentage of mineralogical phases of fly ashes. Using the formula of the mineral, the amount of crystalline Al₂O₃ and SiO₂ present in the mineral phases can be calculated. By subtracting this value from the total values given in chemical composition, the amorphous content or reactive components of Al₂O₃ and SiO₂ present in each fly ash can be obtained. Particle size of the materials was obtained using Cilas laser diffraction particle analyser. Fig. 1 shows the particle size distribution variation for Australian class F fly ashes and Table 3 summarizes particle size distribution in percentage values. Fig. 2 shows elemental mapping analysis for all six types of fly ash types. Since mapping is conducted automatically, it could not illustrate specific elements which their X-ray radiations are weak. Therefore, in some cases, Mg and Ca are not mapped. For a certain element, the lighter the image indicates the larger amounts of the corresponding element. It is obvious that the lightest image for Si and Al belongs to Gladstone fly ash (Fig. 2a). This is while that in accordance to Table 1, this fly ash has the lowest amount of SiO₂. In other words, one can assume that only Gladstone has the best distribution of elements in random sampling of fly ash powders. Moreover, Gladstone, Collie, Eraring and Mt piper fly ashes have homogeneous distribution of Al and Si.

Table 1
Bulk chemical composition of the fly ashes.

	Gladstone	Collie	Mt Piper	Eraring	Tarong	Bayswater
SiO ₂	51.1	60.0	65.8	63.2	73.1	80.4
Al ₂ O ₃	25.6	24.6	26.7	25.2	23.2	14.0
CaO	4.30	0.15	0.05	0.07	0.07	0.04
MgO	1.45	0.99	0.25	0.57	0.14	0.31
Fe ₂ O ₃	12.5	8.56	1.32	3.36	0.89	3.57
Na ₂ O	0.77	0.36	0.32	0.72	0.06	0.10
P ₂ O ₅	0.89	0.22	0.14	0.25	0.05	0.09
Cr ₂ O ₃	-	0.04	-	-	-	-
K ₂ O ₅	0.70	0.36	2.69	1.81	0.52	0.85
MnO	0.15	0.04	0.02	0.07	0.02	0.04
SO ₃	0.24	0.18	0.11	0.18	0.06	0.08
LOI	0.57	1.49	1.44	1.31	0.75	0.54
TiO ₂	1.32	1.53	1.11	0.99	1.31	0.49

Table 2
Mineralogical phases of fly ashes.

	Gladstone	Collie	Mt Piper	Eraring	Tarong	Bayswater
Quartz	13.0	8.42	5.58	8.12	5.88	8.75
Mullite	14.6	7.76	9.15	13.1	7.11	8.56
Hematite	4.17	1.87	1.88	-	-	2.21
Magnetite	1.83	1.71	-	-	-	6.52
Amorphous (SiO ₂ + Al ₂ O ₃ + CaO + other oxides)	66.0 (32.3 + 16.8 + 4.30 + 12.6)	81.0 (48.5 + 19.9 + 0.15 + 12.5)	84.1 (56.6 + 21.2 + 0.05 + 6.27)	76.7 (49.8 + 17.4 + 0.07 + 9.41)	86.5 (64.6 + 18.9 + 0.07 + 3.16)	81.7 (68.2 + 8.9 + 0.04 + 4.58)

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