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Long term permeation properties of different fly ash geopolymer concretes

Chamila Gunasekara *, David W. Law, Sujeeva Setunge

Civil and Infrastructure Engineering, School of Engineering, RMIT University, 124, La Trobe Street, Melbourne, Victoria 3000, Australia

HIGHLIGHTS

- Durability properties of geopolymer concretes improve from 28 to 365 days.
- Chloride diffusion coefficient of geopolymer concretes reduce from 28 to 365 days.
- Continuing gel production densifies microstructure and pore-structure over time.
- A high quantity of meso-pores in the gel paste increases water absorption.
- A high quantity of macro-pores leads to an increase in water and air permeability.

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ABSTRACT

Geopolymer is a sustainable construction material produced by the activation of fly ash using a high concentration alkali to initiate a polymerisation reaction. A key parameter in determining the potential adoption of geopolymer concrete in the construction industry is the long term durability of the material. To determine the durability characteristics a detailed investigation of the permeation properties of four different fly ash geopolymer concretes was carried out up to one year of age. An improvement in the durability properties is observed for all geopolymer concretes with time. This is attributed to an on-going geopolymerization which results in continuing gel formation leading to a more densely packed microstructure, with an associated reduction in meso-pores and macro-pores. The packing density coupled, with the pore size distribution, were observed to determine the permeation and diffusion characteristics of the concrete. The increased in meso-pores represents the increase in the gel of the matrix and in turn this affect the increase of water absorption. On the other hand, a high quantity of macro-pores leads to an increase in the water and air permeability of geopolymer concrete. A large quantity of coarse particles in fly ash results in an uneven gel distribution which reduces pore-filling ability, while the presence of a high quantity of CaO was observed to contribute to a densely packed microstructure. Notably the initial chloride diffusion coefficients are analogous to those observed in Portland and blended cement concretes and also decrease with the age in a similar manner.

1. Introduction

Portland Cement (PC) production consumes high quantities of fuel and raw materials using processes that are energy intensive and emit large amounts of greenhouse gases. This cement production alone contributes about 5–7% of anthropogenic CO_2 emissions worldwide [1,2], with the 0.6–0.8 kg of CO_2 emitted for every kilogram of cement manufactured [3,4]. Thus, the impact of the cement industry on the global environment is a significant problem. This on-going issue has inspired research into environmentally friendly green concrete utilizing alkali-activated cement, widely known as geopolymer, using materials containing alumina and silicates [5]. The reduction of CO_2 emission due to the replacement of PC with geopolymer is estimated to be between 26% and 45%, with no economic impact [6,7]. Low calcium fly ash has been identified as a possible source material for geopolymer concrete.

Fly ash based geopolymer concretes have been shown to be able to achieve comparable strengths to PC and blended cement concretes [8–10]. However, the durability characteristics of geopolymer concrete are as important as material strength since the failures of concrete structures are not only caused by excessive load, but also due to the deterioration of structural components.

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* Corresponding author.
E-mail addresses: chamila.gunasekara@rmit.edu.au (C. Gunasekara), david.law@rmit.edu.au (D.W. Law), sujeeva.setunge@rmit.edu.au (S. Setunge).

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The durability resistance of fly ash geopolymer concretes against elevated temperature [11–13], sulphate and acids [14–16], carbonation [17,18] and chloride penetration [19,20] has been widely studied. While these studies have shown that fly ash geopolymer concretes have a satisfactory performance, many of them were conducted by using a single type of fly ash using a mixing process unique to that study and often at an early age. However, durability properties, such as, water penetration, pore structure characteristics, chloride ingress and gas transport in concrete are long term issues, with these properties potentially changing with time. To date limited attention has been given to evaluate variations in long term permeation properties of geopolymer concrete with time. The most important parameter that influences water and chloride ion transport is the pore structure, particularly the pore volume, pore size distribution, connectivity and shape of the pores. In this study a series of geopolymer concrete specimens were prepared from four different fly ashes and the water absorption, water permeability, air permeability and chloride diffusivity were determined up to one year of age to identify any changes with time. In addition, the pore structure and microstructure changes of the different fly ash geopolymer concretes up to one year have been examined by using mercury intrusion porosimetry (MIP) and scanning electron microscopic (SEM) analysis to identify any correlation in changes in durability properties with microstructural changes.

2. Experimental program

2.1. Materials used

Four types of low calcium, Class F fly ash conforming to AS 3582.1 standard [21], obtained from Gladstone, Pt.Augusta, Collie and Tarong power plants in Australia were used to manufacture the geopolymer concrete. The chemical composition, amorphous and crystalline content, and physical properties of each fly ash, determined by X-ray fluorescence, X-ray diffraction (XRD) and Malvern particle size analyser (Mastersizer X), respectively are shown in Tables 1 and 2. The XRD data were obtained using a Bruker AXS D4 Endeavor wide angle X-ray diffractometer with copper anode at 40 kV and 35 mA. After 28 days samples were cut into thin slices, then ground into fine particles and filtered using 75 μm sieve to obtain the required powder samples for testing. The semi-quantitative analysis was conducted using BRUKER DIFFRAC.EVA-4 software in order to identify the crystalline phases. Brunauer Emmett Teller (BET) method by N_2 absorption was used to determine the fly ash surface area. The sulphur trioxide (SO_3) is less than 1%, which is expected to ensure high volume stability, which is desirable for good durability.

The alkaline liquid used in the geopolymer production consisted of a mixture of commercially available sodium silicate solution (specific gravity = 1.53, Na_2O = 14.7% and SiO_2 = 29.4% by mass), and sodium hydroxide solution (15 M). The sodium hydroxide solution was prepared by dissolving sodium oxide pellets of 99% purity in deionised water one day prior to usage. Both coarse and fine aggregate were prepared in accordance with AS 1141.5 standard [22]. The moisture condition of the aggregate was in a saturated surface dry condition. The fine aggregate was river sand in uncrushed form with a specific gravity of 2.5 and a fineness modulus of 3.0. The coarse aggregate was crushed basalt aggregate of two-grain sizes: 7 mm (2.58 specific gravity and 1.60% water absorption) and 10 mm (2.62% specific gravity and 0.74% water absorption). Demineralized water was used throughout the experiment.

2.2. Mix designs

The optimized mix design (activator modulus) for each fly ash based geopolymer mortar [23] was initially applied to the respective fly ash in the manufacturing of geopolymer concrete. The AM was then varied by 0.125 intervals ranging from 1.0 to 1.75 until the optimum compressive strength for the each fly ash geopolymer concrete at 28 days was determined. The activator modulus (AM) is characterized by the blended sodium silicate and sodium hydroxide solutions as the mass ratio of SiO_2 to Na_2O in alkaline activator. In all cases, the Na_2O dosage (i.e. mass ratio of Na_2O content in alkaline activator to fly ash) is fixed at 15% while the total aggregate in the concrete is kept to 64% of the entire mixture by volume for all mixes. The ratio of ingredients (fly ash, chemical activator, aggregate, and water) was calculated based on the absolute volume method [24], as a result, the total weight of binder and water was varied to keep the volume of material and water/solid ratio (0.37) constant. The mass of water in the mix was taken as the sum of mass of water contained in the sodium silicate, sodium hydroxide and added water. The mass of solid is taken as the sum of fly ash, the solids in the sodium silicate solution and the sodium oxide pellets. Table 3 summarizes the optimized mix details.

2.3. Mixing, casting and curing

The mixing of geopolymer concrete was carried out using a 90 L concrete mixer. The dry materials (fly ash, fine and coarse aggregates) were mixed first for 4 min. Then activator and water were added to the dry mix and then mixed continuously for another 8 min until the mixture was glossy and well combined. The mixture was then poured into 100 mm × 100 mm × 100 mm cubic Teflon moulds for compressive strength and chloride diffusion

Table 2
Physical and mineralogical properties of fly ash.

Properties investigated		Gladstone	Pt.Augusta	Collie	Tarong
BET Surface Area (m^2/kg)		2363	1228	1095	1876
Specific gravity		2.26	2.23	2.42	2.08
Percentage passing at X micron sieve	X = 5	24.8	30.1	26.1	22.7
	X = 10	43.1	46.7	40.9	43.0
	X = 20	61.9	62.1	54.6	63.0
	X = 45	82.7	80.2	70.0	81.8
Amorphous content (%)		71.8	59.5	72.5	66.3
Crystalline (%)	Quartz	6.8	29.2	18.2	14.8
	Mullite	17.9	7.5	8.7	18.9

Table 1
Chemical composition of fly ash.

Fly ash	Component (wt.%)											
	SiO_2	Al_2O_3	Fe_2O_3	CaO	P_2O_5	TiO_2	MgO	K_2O	SO_3	MnO	Na_2O	LOI[a]
Gladstone	47.87	28.0	14.09	3.81	1.81	1.99	0.93	0.62	0.27	0.21	0.41	0.43
Pt.Augusta	49.37	31.25	4.47	4.80	1.65	2.94	1.28	2.21	0.24	0.04	1.30	0.51
Collie	53.82	29.95	9.24	1.03	1.28	2.19	0.58	0.79	0.34	0.04	0.75	0.63
Tarong	75.66	19.0	1.38	0.30	1.0	1.83	0.0	0.63	0.03	0.02	0.15	1.16

[a] Loss on ignition (unburnt carbon content).