



Chloride-induced corrosion of reinforcement in low-calcium fly ash-based geopolymer concrete

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ABSTRACT

Geopolymer concrete (GPC) has significant potential as a more sustainable alternative for ordinary Portland cement concrete (PCC). However, as a rather new engineering material, there are some concerns over the durability aspects of geopolymer-based binders. In this study, the performance of chloride-contaminated reinforced GPC specimens manufactured using a blended low-calcium fly ash and slag cement is investigated by long-term monitoring of corrosion parameters such as open circuit corrosion potential, polarization resistance and Tafel slopes. The electrochemical results are validated by contrasting the electrochemical mass losses with the mass losses obtained from the gravimetric measurements. The investigated low-calcium fly ash-based GPC exhibit a comparable electrochemical performance to a similar strength PCC during the propagation phase of corrosion. Additionally, some of the conventional classifications which are commonly used to assess the severity of corrosion in Portland cement-based corroding systems might need some recalibration to be used for low-calcium fly ash-based corroding systems.

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1. Introduction

Ordinary Portland cement concrete (PCC) is the most commonly used material in the construction industry [1] and global demand for it is projected to increase steeply until 2050 reaching 5.5 Gt/year [2]. Concrete is indeed an attractive construction material regarding mechanical performance, durability, shape adaptability and availability of raw precursors. Nonetheless, production of PCC is associated with some significant environmental impacts such as massive consumption of natural resources and responsibility for as much as 6–7% of all the greenhouse gasses emitted worldwide [3–5].

PCC can be prone to degradation and premature failure and using Portland cement-based concretes can lead to serious problems with millions of dollars spent for maintenance, repair or replacement of damaged structures [6–8]. In this regard, developing alternative binders can be considered as an efficient way of overcoming the hurdles associated with the production and application of Portland cement binders. Among all the available alternative binders, geopolymer binders have a great development potential and are widely considered as a promising alternative for Portland cement binders [9]. Geopolymer binders are produced by the chemical reaction of alumina-silicate oxides with alkali polysilicates to produce polymeric Si—O—Al bonds [10]. Due to the low cost and wide availability, industrial waste materials such as fly

ash are commonly used as the source of aluminosilicate for the manufacture of geopolymer concrete (GPC).

While emitting up to nine times less carbon dioxide [11], GPCs exhibit many of the engineering characteristics of traditional concretes, despite their vastly different chemical composition and reaction mechanisms [12–14]. Moreover, geopolymer binders can display other benefits as well, such as higher stability when exposed to elevated temperature [15], higher resistance against chemical attacks [16–18] and better resistance to freeze-thaw cycles [19,20]. However, being a comparatively young engineering material, the quantity of available durability data (crucial for predicting the service life of reinforced GPC structures) such as chloride ingress properties or corrosion rate of embedded reinforcing steels is limited, and the long-term performance of GPC structures is yet to be determined [9,21].

Low-calcium fly ash-based (Class F) GPC has been reported by some researchers to have lower chloride diffusion coefficients, chloride content and porosity compared to high-calcium (Class C) fly ash-based GPC and PCC [22], while others have found that the exact raw materials and mixture design can result in varying performance [23]. Lloyd et al. [24] concluded that the presence of calcium is essential to lower the permeability of pore system and prevent alkalis from leaching and consequent pH drop which can lead to depassivation of embedded reinforcement. Previous studies reveal that due to the high alkalinity of the pore system, fly ash based GPC can passivate the reinforcement steel as efficiently as PCC [24–28]. Type and concentration of the alkaline solution have also been found to play a crucial role in the stability of passive film [1,24,28]. While performance of fly ash-based GPC is

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observed to be similar to PCC in passive state (i.e. in the absence of chloride ions) [25,28], an (at least) equivalent performance of low-calcium fly ash-based GPC compared with PCC in severe marine environments during the propagation phase of corrosion is emphasized [28–30].

Considering the limited literature available related to the chloride-induced corrosion of reinforcements in geopolymer concretes, this work aims to:

- Assess the performance of low-calcium fly ash-based GPC during the propagation phase of corrosion process through long-term monitoring of corrosion parameters such as open circuit corrosion potential, polarization resistance and Tafel coefficients. The results are compared with the reported values for PCC. Direct mass loss measurement is carried out as a way to validate results of the corrosion tests.
- find out if the existing electrochemical test methods calibrated and validated for reinforced Portland cement concrete are suitable for geopolymer concrete.

2. Experimental program

2.1. Geopolymer concrete mix design and precursors

An experimental investigation is conducted to assess the corrosion of reinforced GPC fabricated from a blend of Class F fly ash and ground granulated blast-furnace slag (GGBFS) stockpiles. In accordance with a previous study [31], three different sources of aluminosilicate precursors have been used: 1 - fly ash (FA) from Eraring Power Station (New South Wales, Australia), 2 - an ultra-fine FA branded as Kaolite high-performance ash (HPA), sourced from Callide Power Station (Queensland, Australia), and 3 - ground granulated blast-furnace slag (GGBFS) supplied by Blue Circle Southern Cement Australia. Both fly ashes are low-calcium class F fly ash (ASTM C 618 Class F). Chemical compositions of the aluminosilicate sources determined by X-ray fluorescence (XRF) analysis are listed in Table 1.

A mixture of the sodium hydroxide (NaOH) solution and sodium silicate (Na_2SiO_3) solution was used [31,32]. The sodium hydroxide solution used was prepared by dissolving the technical grade NaOH pellets in water. The sodium hydroxide white pellets with a purity of at least 98% were supplied by Ajax Finechem under the commercial name of UNIVAR A-302. These pellets have a specific gravity of 2.1 g/cm^3 and a pH of approximately 14. The concentration of sodium hydroxide solution used is 12 M (12 M) which consisted of 480 g of NaOH pellets per liter of solution or 361 g of NaOH pellets per kg of solution. Grade D sodium silicate, which was supplied by PQ Australia under the commercial name of Vistrol D-A53, has a chemical composition of $\text{Na}_2\text{O} = 14.7\%$, $\text{SiO}_2 = 29.4\%$ and $\text{H}_2\text{O} = 55.9\%$ (by mass). The Na_2SiO_3 solution used is a thick adhesive liquid with a viscosity of 400 cps at 20°C , has a

specific gravity of 1.53 g/cm^3 and a pH of 12.9 (values provided by the supplier, PQ Australia). Also, the ratio of sodium silicate to sodium hydroxide solution used was 2.5:1 (by mass) to maximize the compressive strength, while having an acceptable workability and pH level [33–35]. The modulus (the molar ratio of $\text{SiO}_2/\text{Na}_2\text{O}$) of the solution is 1.17.

Mix proportioning of the raw material ingredients, as shown in Table 2, was carried out by mass. About 85% of the blend is composed of low-calcium class F fly ash. Sydney sand was used as fine aggregate, and the coarse aggregate was 10 mm nominal size crushed basalt. The aggregate's mass shown in Table 2 is in the saturated surface dry (SSD) condition.

2.2. Fabrication of specimens, curing and exposure condition

To prohibit any external corrosion, a specific method of fabrication has been adopted (Fig. 1), similar to the approaches employed in some previous studies to assess the steel corrosion in PCC [36,37]. The reinforcing bars used were normal ductility grade 12 mm deformed bars with 500 MPa yield strength. All embedded bars were 50 mm long and were machined at both ends. Rebars were gently wire-brushed to eliminate any pre-formed rusts which could affect the gravimetric mass loss calculations. All steel bars were then weighed and tagged for gravimetric mass loss measurements at the end of the testing period (after doing the destructive Tafel test on each sample). One acrylic tube was attached at each end. The internal diameter of the acrylic tubes was almost equal to the external diameter of the machined part of the steel bars, so the steel bar could easily fit in the tube, although a thin layer of silicone sealant was used as an adhesive and also to avoid any corrosion on the machined parts of the steel bars. Acrylic tubes were then filled with Silicone sealant to block the access for chloride ions to the steel bar (Fig. 1). A copper wire was welded on each steel bar to perform as a working electrode during the electrochemical measurements. Both top and bottom sides of samples were coated with an anti-chloride resin (supplied by the Australian supplier "Parchem" under the commercial name of Emer-Stop S100 N), to enforce peripheral penetration of chloride ions.

After casting, moulds were sealed with either a proper lid or a plastic sheet to prevent excessive loss of moisture and were stored in a chamber at 40°C for 24 h. The specimens were then demoulded, sealed again and placed in a hot water bath at 80°C for another 24 h. After that, samples were stored in a room with a fixed temperature of $23 \pm 2^\circ\text{C}$ and relative humidity (RH) of 50% until the testing dates. The average compressive strength (average of 3 samples) at 28th day was 54.5 MPa with a standard deviation of 1.10 MPa. Also, the average elastic modulus (average of 3 samples) at 28th day was 23.8 GPa with a standard deviation of 0.61 GPa.

Table 1
Chemical compositions of FA, kaolite HPA and GGBFS by X-ray fluorescence (XRF) analysis.

Oxide	FA [wt.%]	Kaolite HPA [wt.%]	GGBFS [wt.%]
Silicon dioxide (SiO_2)	66.56	45.14	31.52
Aluminium oxide (Al_2O_3)	22.47	33.32	12.22
Iron oxide (Fe_2O_3)	3.54	11.99	1.14
Calcium oxide (CaO)	1.64	4.13	44.53
Potassium oxide (K_2O)	1.75	0.13	0.33
Sodium oxide (Na_2O)	0.58	0.07	0.21
Magnesium oxide (MgO)	0.65	1.37	4.62
Manganese oxide (MnO)	0.06	0.23	0.36
Phosphorus oxide (P_2O_5)	0.11	0.56	0.02
Titanium oxide (TiO_2)	0.88	2.19	1.03
Sulphur trioxide (SO_3)	0.10	0.48	3.24
Loss of ignition (LOI)	1.66	0.41	0.79
Specific gravity	2.1	2.4	2.8

Table 2
GPC mix proportions.

Material	kg/m ³
Coarse aggregate	1138
Fine aggregate	730
FA	200
Kaolite HPA	55
GGBFS	45
12 M sodium hydroxide solution (NaOH)	45.7
Sodium silicate solution (Na_2SiO_3)	114.3
Free water	31
Coarse aggregate/fine aggregate	1.60
Total binder (FA, HPA, GGBFS)	300
Water/binder ¹	0.35
Modulus ($\text{SiO}_2/\text{Na}_2\text{O}$)	1.17
$\text{Na}_2\text{O}/\text{binder}$ (wt.%)	9.86

¹ Calculated considering the total water and the total solids (precursors + anhydrous activator).

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