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Regulating the chemical foaming reaction to control the porosity of geopolymer foams

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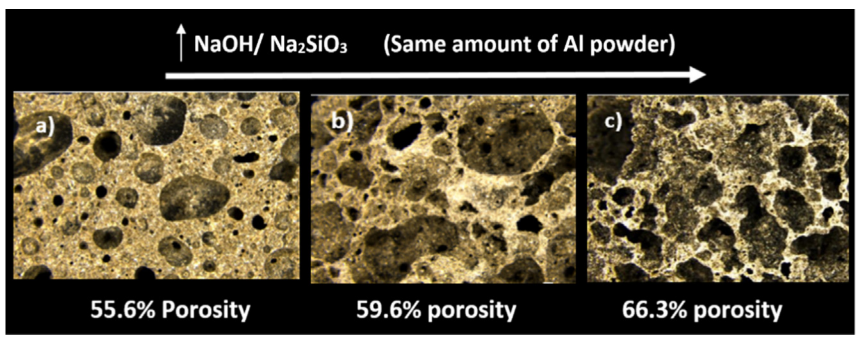
HIGHLIGHTS

- In-situ FTIR is used to study the kinetics of aluminium foaming agent reaction in different alkali environments.
- Aluminium reaction has been regulated to increase the porosity of geopolymer foams without adding more aluminium powder.
- Altering aluminium foaming reaction rate results in different morphology and size distribution of the pores.
- The impact of controlling the foaming reaction on geopolymerization kinetics is investigated.

GRAPHICAL ABSTRACT



High percentage of aluminium foaming agent is not desirable in geopolymer foams made with aluminium powder. Therefore, the kinetics of aluminium reaction is regulated to increase the porosity of geopolymer foams without increasing the extent of aluminium usage. In geopolymer foams made by Al powder, the geopolymerization reaction and aluminium foaming reaction occur simultaneously. By adjusting the ratio of alkali activators, the high degree of aluminium reaction has been coupled with faster setting of geopolymers, and therefore, the extent of porosity and the pore size distribution has been increased in geopolymer foams without the need to add more Aluminium powder. Aluminium powder has high embodied energy, and this study helps to minimise the negative environmental footprint of geopolymer foams which are aerated by aluminium powder.

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ABSTRACT

In lightweight geopolymers foamed with aluminium powder, it is desirable to increase the porosity without increasing the amount of aluminium usage. In this study, the kinetics of aluminium reaction is manipulated to increase the porosity of geopolymers without adding extra foaming agent, and the impact on porosity development and the characteristics of binding skeleton is investigated. It is shown that adjusting the ratio of alkali activators regulates the oxidation rate of aluminium powder and impacts the extent of foaming. Samples with higher aluminium oxidation rate developed higher porosities and reached lower densities without adding more foaming agent. Size distribution of the voids becomes wider, and their circularity is reduced in higher chemical foaming reaction rates. Also, by increasing the foaming rate, lower silica to alumina ratios contribute in geopolymer gel network, and geopolymer binder loses its compactness.

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1. Introduction

Geopolymer concretes are frequently referred to as an "environmentally-friendly" or "green" construction materials compared to the cement-based concretes [1,2]. Geopolymer production process is not as resource and energy intensive as cement manufacturing process, and the life-cycle analysis has shown that if the synthesis method and condition is controlled, geopolymers can significantly reduce the carbon dioxide emissions as cement replacement materials [3].

Cellular concrete (or foam concrete) is a lightweight concrete with air voids entrapped in its matrix which make it lighter and more sustainable by decreasing its embodied energy (i.e. energy required to produce, transport and install) [4,5]. Cellular concrete brings some advantages to the construction industry such as improving housing affordability, improving the thermal insulating properties and providing acoustic insulation [6,7]. Foam concretes have been widely used for various non-structural construction applications such as precast or cast-in-situ wall elements, insulation screeds, facades, slabs, or as a core of lightweight sandwich panels that are becoming increasingly popular in construction [8]. Geopolymer and alkali-activated binders have also been used to make foam concretes with the aim of producing more environmentally friendly construction materials compared to the Portland cement-based cellular concretes [9,10].

There are two usual methods of introducing air voids into the concrete matrix, i.e. mechanical foaming and chemical foaming. In mechanical foaming method, surfactants are added to the mixture to generate bubbles during the mixing process, or premade foams are produced and blended with concrete paste [11,12]. In chemical foaming method, chemicals are added to the mixture and gas voids are appeared within the concrete paste as a result of their reaction in alkaline environment of concrete. During the setting and hardening stages of concrete, the gas voids are confined within the binding skeleton and drop the density of matrix [13–18]. Mechanical foaming method can potentially result in a consistent distribution of fine voids [9], but the density of geopolymer concrete cannot always be extensively reduced by mechanical foaming. The chemical foaming seems to be more effective in dropping density, but it can result in generation of disorderly large voids which negatively affect the thermal and mechanical properties of concrete. The combination of both methods has also been used to bring their benefits together [15,19].

Aluminium metal powder is widely used in lightweight concrete industry as a chemical foaming agent [4,9,20], and recently cellular geopolymer concretes are also made utilising similar methods targeting lower densities with the addition of higher percentage of aluminium powder [13–16]. Aluminium can react with water and produce hydrogen gas. However, a passive oxide film that covers the surface of the metal Al in ambient condition inhibits the Al reaction in water [21]. An alkaline solution, e.g. sodium hydroxide solution can accelerate the corrosion of Al in water as a catalyst, and therefore; the alkaline environment of concrete paste is a suitable medium for aluminium reaction and hydrogen generation. The reactions of aluminium with aqueous solutions of sodium hydroxide have been previously studied [21–23]:

$$2Al + 6H_2O + 2NaOH \rightarrow 2NaAl(OH)_4 + 3H_2 \quad (1)$$

$$NaAl(OH)_4 \rightarrow NaOH + Al(OH)_3 \quad (2)$$

$$2Al + 6H_2O \rightarrow 2Al(OH)_3 + 3H_2 \quad (3)$$

Sodium hydroxide is initially consumed in the hydrogen generation reaction (1), but when the aluminate concentration exceeds the saturation limit, aluminate undergoes a decomposition reaction (2) that produces a precipitate of aluminium hydroxide with the regeneration of the alkali. The amount of Al, the particle size of the metal powder and the reaction temperature are known to be important factors affecting the kinetics of aluminium reaction [24].

Since geopolymer foam concretes are intended to be a more environmentally-friendly alternative of cement-based foam concretes, their synthesis condition and foaming method should be carefully controlled to preserve their environmental benefits. Aluminium powder has very high embodied energy and extensive usage of this material in geopolymer foams will diminish their environmental benefits [9]. In order to minimise the negative impact of the metallic powder on the environmental footprint of geopolymeric products, it is important to minimise the dosage of aluminium used as a foaming agent. In most of the studies conducted on geopolymer foams with aluminium foaming agent, the amount of aluminium powder is increased in order to increase the porosity. Although there are studies that used different formulation of geopolymers together with different amount of aluminium powder [25], the impact of mix design on porosity development was not studied and differentiated with the impact of different aluminium content. It is important to explore the reason how different geopolymer formulation can impact the extent of porosity to be able to custom design the geopolymer foams of low densities without extensive usage of aluminium powder.

Altering the quantity of aluminium powder or the temperature of the reaction are not suitable options for controlling the kinetics of foaming since higher temperature or higher amount of aluminium both increase the embodied energy of geopolymer concretes. The alternative method is regulating the reaction by using accelerators and inhibitors. As discussed earlier, alkaline solutions like sodium hydroxide act as a catalyst of aluminium reaction accelerating the corrosion in water. Sodium silicates have also been known to be an effective inhibitor of aluminium corrosion in alkaline systems [26]. The inhibition effect of the sodium silicates is related to the formation of amorphous aluminosilicate film on metal surface. Sodium hydroxide and sodium silicate are the commonly used activators in geopolymers, and they are usually used together to improve the performance of geopolymer concretes [2,27,28]. Varying the ratios of these two activators to control the foaming reaction will also change the silica modulus that impacts the geopolymerization reaction and the properties of binding skeleton [27–31]. Therefore, it is important to study both geopolymerization and foaming reactions and investigate the porosity development together with binder formation mechanism.

In this study, geopolymer foams with different porosities are synthesised with similar amount of aluminium powder as a foaming agent. Two commonly used alkaline activators in geopolymers (i.e. sodium hydroxide and sodium silicate) are also the well-known accelerator and inhibitor of the aluminium reaction. The ratio of these activators is manipulated to control the kinetics of aluminium reaction and the extent of hydrogen gas generation in geopolymer foams. Also, since the high strength development is not central in this study, no foam modifiers are added to the matrix to minimise the impact of other factors on pore size distribution and circularity of voids. The impact of varying alkali activators ratio is studied on aluminium reaction, characteristics of geopolymer foams and the properties of the binding skeleton.

2. Materials and methods

Fly ash used in this study (with the commercial name of Melbourne Ash) is supplied from Cement Australia. Table 1 shows the chemical composition of fly ash. Quantitative XRD analysis indicated that 75.5% of Melbourne Ash is amorphous. Anhydrous sodium metasilicate with the composition of 50.5 wt% Na_2O and 46.2 wt% SiO_2 is obtained from Redox, and sodium hydroxide pellets with the composition of 99.9 wt% NaOH is purchased from Sigma-Aldrich. Aluminium metal

Table 1
Oxide composition of Melbourne Ash as determined by XRF*a*.

SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	K_2O	P_2O_5	SO_3	Na_2O
42.09	1.44	25.13	13.16	0.18	1.27	13.56	0.41	1.10	0.41	0.81