

The Influence of Si:Al and Na:Al On The Physical and Microstructure Characters of Geopolymers Based on Metakaolin

SUBAER^{1,a*}, ABDUL Haris^{1,b}, NURHAYATI^{1,c}, ANDI Irhamsyah^{1,d},
JANUARTI Jaya Ekaputri^{2,e}

¹Laboratorium Fisika Material, Jurusan Fisika, Universitas Negeri Makassar, Indonesia
Jl. Daeng Tata Raya, Makassar, Indonesia

²Department of Civil Engineering, Institut Teknologi Sepuluh Nopember, Sukolilo, Surabaya, 60111, Indonesia

^ajzubayir@yahoo.com, ^bAbdulharis@yahoo.co.id, ^cNurhayati_unm@yahoo.com
^dirhamsyah.physics@gmail.com, ^ejanuarti_je@yahoo.com

Keywords: geopolymers, metakaolin, physical properties, microstructure characters

Abstract. A research has been conducted to investigate the physico-mechanical and microstructure properties of geopolymers synthesised from metakaolin activated with sodium silicate solution. A wide range of physical and mechanical properties of geopolymers were studied such as bulk density, porosity, Vickers hardness, compressive strength, thermal expansion and thermal conductivity. It was found that these properties were directly related to geopolymers process variables such as Si:Al, Na:Al, Na₂O:H₂O, time and curing temperature. The structure of the resulting geopolymers was studied by using X-Ray diffraction (XRD) and the microstructure of geopolymers paste and the interfacial transition zone (ITZ) between the aggregate and the matrix of geopolymer were studied by using Transmission Electron Microscope (TEM) and Scanning Electron Microscope (SEM). The results gave a new insight into the composition-microstructure-property relationship of geopolymers and paving the way to the production of geopolymers with improved performance in a variety of applications.

Introduction

Over the last thirty years, *geopolymers*, have received much attention as a promising new form of inorganic polymer material that could substantially substitute for conventional or ordinary Portland cement, plastics, ceramics-composites and many mineral-based products. Geopolymers are a subset of the broader class of alkali-activated binders [1]. The defining characteristic of a geopolymer is that the binding phase comprises an alkali aluminosilicate gel, with aluminium and silicon linked in a three-dimensional tetrahedral gel framework that is relatively resistant to dissolution in water [2,3].

Research has shown that geopolymers may be readily synthesised through alkali-activation of inexpensive and pure starting materials such as kaolinitic clays [4-10,26], as well as waste products such as fly ash and furnace slag [11-16].

Geopolymers have a potential for a wide variety of applications, whether used pure, with fillers or reinforced. In general, these applications can be divided into two categories; (1) Structural products such as reinforced for the manufacture of moulds, tooling, cement and concrete replacements in various environments, (2) Immobilisation technology for toxic chemical and radioactive waste containment. It is expected that in the near future these applications will also be found in automobile and aerospace industries, non-ferrous foundries and metallurgy, civil engineering and plastic industries [18-21]. Understanding of geopolymers is built to the point where binder properties can be tailored a priori by rational mix design, and the understanding of the binder structure is sufficient to explain why these properties can be expected to last for a sufficient period of time to render the material fit for purpose in an engineering sense [22].

Experimental

Geopolymers produced in this study were prepared by the alkali-activation (sodium silicate solution) of metakaolin. Metakaolin was obtained by dehydroxylation of kaolinite at 750°C for 6 hours. Chemical compositions of the activation solution in terms of the molar oxide ratio used in this study were divided into 3 groups. In each group, the nominal Si:Al ranged from 1 to 2 while varying Na:Al from 0.6 for group A, 0.8 for group B and 1.0 for group C. The oxide molar ratio of H₂O:Na₂O was kept at a value of 10. The resulting geopolymer resin was poured into the moulds, compacted and vibrated few minutes and allowed to mature at room temperature for 30 - 60 minutes. Curing was undertaken at 70 °C for 2 hours followed by drying at room temperature before demoulding. Natural sand (α -quartz) was used as aggregate and poured directly into the geopolymer resin. The grain size and concentration of sand aggregate used were 40 μ m, 125 μ m, 212 μ m, 500 μ m and 750 μ m, and 10 wt %, 20 wt %, 30wt % and 50 wt %, respectively. Another type of aggregate used was granite (known as blue metal), which was crushed and sieved to attain a grain size of 300 μ m and 600 μ m.

The resulting materials were subjected to various physico-mechanical characterisations, including bulk density, apparent porosity, vickers microhardness, compressive strength, aggregate-geopolymers interfacial bond, thermal conductivity, and thermal expansion. The structure of geopolymers was studied by using X-Ray diffraction (XRD). The microstructure of geopolymers paste and the interfacial transition zone (ITZ) between the aggregate and the matrix were studied by using Transmission Electron Microscope (TEM) and Scanning Electron Microscope (SEM).

Results and Discussion

Fig. 1 depicted the XRD of metakaolin indicates all the kaolinite reflections have been eliminated leaving an amorphous pattern with ancillary peaks due to α -quartz (SiO₂) and anatase (TiO₂) although the morphology of the original kaolinite is maintained as depicted in fig. 1.

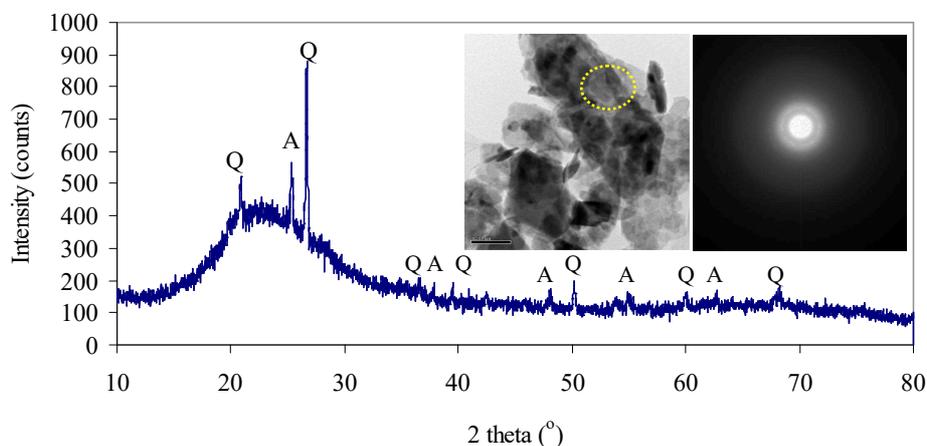


Figure 1. XRD pattern of metakaolin, A = anatase (TiO₂) and Q = α -quartz (SiO₂), decorated with its TEM image and Selected Area of Electron Diffraction (SAED).

Geopolymers prepared in this study were divided into three groups based on the nominal ratio of atomic Si:Al and Na:Al. Geopolymers with Si:Al = 1.04 and 1.25 are designated as *sodium-poly(sialate)* (Na-PS) type, and geopolymers with Si:Al = 1.50, 1.75 and 2.00 are designated as *sodium-poly(sialate-siloxo)* (Na-PSS) type. The bulk density and apparent porosity of all specimens were measured by using Archimedes method. It was found that magnitude of the bulk density range from 1.40 to 1.70 g/cm³ and the apparent porosity from range from 33 to 24%. Both parameters depend on the ratio of Si:Al and Na:Al.

The XRD results showed that all Na-PS type geopolymers enclose the formation of zeolite-A and zeolite-X (Fig. 2a). These material revealed low bulk density, high porosity, and low compressive

strength. The formation of zeolite-A and zeolite-X from the geopolymer processing route was also observed by Rowles & O'Connor [23]. They also reported that the formation of zeolite from the geopolymer route occurs with either low Si:Al or high Na:Al ratios. Similarly, Grutzeck, Kwan & DiCola [25] reported the formation of zeolite in alkali-activated cement based on fly-ash. Na-PSS geopolymers were formed by increasing the Si:Al atomic ratio from 1.50, 1.75 and 2.00 for Na:Al atomic ratios of 0.6, 0.8 and 1.0. For these compositions the XRD patterns indicated that the structure of the resulting geopolymers were essentially amorphous. Fig. 2 (b) shows the XRD patterns of a series of Na-PSS geopolymers formed with a Na:Al atomic ratio = 0.6.

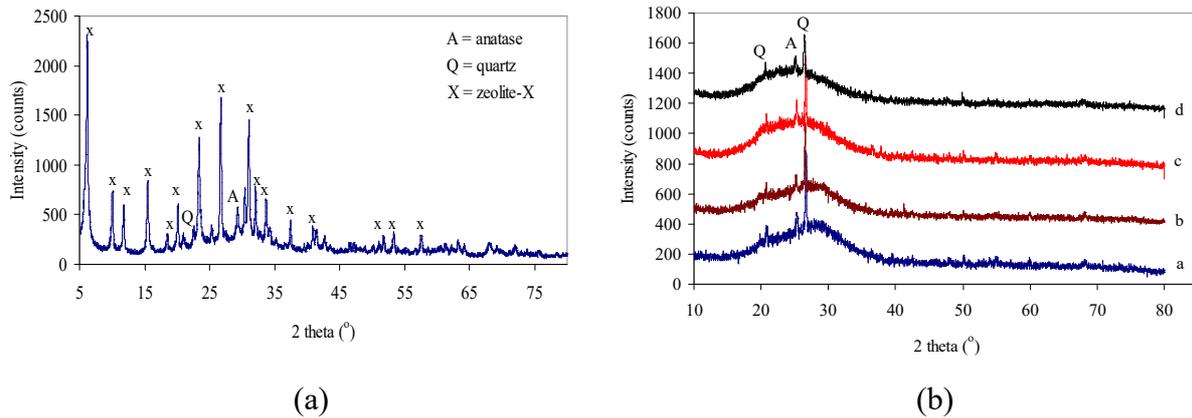


Figure 2. XRD patterns of geopolymers prepared with Na:Al molar ratio of 0.6. (a) Si:Al = 1.25, (b) Si:Al = 1.50, (c) Si:Al = 1.75 and (d) Si:Al = 2.0. The sharp diffraction peaks are anatase (A) and quartz (Q). Each pattern has been offset for clarity.

The diffraction patterns of these geopolymers, like the pattern of the original metakaolin, have a broad amorphous hump in the region $20^\circ - 38^\circ(2\theta)$. This suggests that the Na-PSS geopolymers consist of disordered frameworks with short-range order, with structures similar to those of feldspathic glasses [23,8].

Fig. 3(a) show SEM images of a sample prepared with Si:Al = 1.25 and Na:Al = 0.8. Based on the XRD results, a greater amount of zeolite-A was formed in this sample. Fig. 3(b) shows SEM image of a sample with Si:Al = 1.25, Na:Al = 1.0. The sample appears more dense than the sample shown in fig. 2 (a). Large voids on the surface of the sample are believed to be due to the grain pullout during polishing. The images indicates the influence of composition (Na:Al) to the microstructure characters of geopolymers.

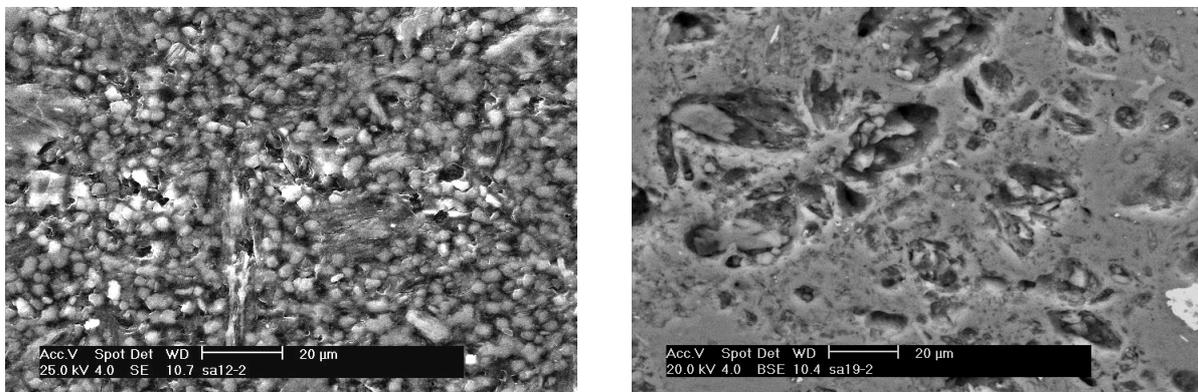


Figure 3. (a) SEM image of a geopolymer sample with Si:Al = 1.25, Na:Al = 0.8, (b) SEM image of geopolymer sample with Si:Al = 1.25, Na:Al = 1.0.

Fig. 4(a) shows that geopolymer prepared with Si:Al = 2.0, Na:Al = 1.0 has high homogeneity. This indicates that there was adequate sodium silicate solution to be able to dissolve most of the