Effects of water to solid ratio on thermal conductivity of fly ash-based geopolymer paste

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Abstract. Thermal conductivity is one of the most important properties of material when applied as an insulating material. It referred to how much heat can be transferred inside the material. Usually, a low thermal conductivity is needed as a good insulating material. This study focuses on the effect of various water to solid ratio on the geopolymers’ thermal conductivity. Six various waters to solid ratio ranging from 0.16 to 0.31 were studied. Mix design of geopolymers paste using fly ash as aluminosilicate was obtained from Paiton power station. Alkaline activator was combined with NaOH concentration of 10 M and constant Na₂SiO₃/NaOH ratio of 2.0. A maximum of 7 days compressive strength obtained was up to 34.12 MPa when water to solid ratio at 0.22. The thermal conductivity was varied with a temperature range from 50°C to 200°C. The average thermal conductivity of geopolymers paste ranges from 0.75 to 1.54 W/m·°C. The lowest thermal conductivity occurred when water to solid ratio was at 0.22. Based on this finding, fly ash-based geopolymer paste can be applied as an insulating material.

Keyword: water to solid ratio, thermal conductivity, geopolymer, fly ash.

1. Introduction

Geopolymers are durable to high temperature exposure because of its amorphous to semi-crystalline silico aluminate materials [1,2]. Alkaline solution is needed to produce a geopolymer by activating the aluminosilicate source like metakaolin, fly ash and slag. To activate aluminosilicates for geopolymer synthesis, Khale and Chaudhay [3] suggested the use of alkaline solution with pH value higher than 13. Vickers et al [4] described that many factors can affect geopolymer synthesis and properties of geopolymer. There are molar ratio of silicon to aluminium amorphous from raw material, type and concentration of alkali solution, water content, and curing treatment.

Water content is known to result in a good property of geopolymer. It consists of total mass of water including alkaline solution and additional free water. The ratio of water, to the sum of aluminosilicate source and solid form of alkaline solution described as water to solid geopolymer ratio (W/GPS). This ratio affects geopolymerisation rate. Temuuvin et al [5] reported that the higher W/GPS ratio, the lower the mixture rate to conduct geopolymerisation.
Geopolymer material is widely applied in engineering materials, especially as a protection from fire exposure because its amorphous inorganic structure makes a better thermal resistance. As an insulation material, a low thermal conductivity is needed as low as possible to minimize the heat transfer inside the materials [4]. There have been many researchers investigating the thermal conductivity of geopolymer. Sivasakthi et al. [6] reported thermal conductivity as a time function. Increasing the temperature will decrease the thermal conductivity. In a room temperature, geopolymer mortar results in a thermal conductivity value range from 0.7 -1.1 W/m.K. Sakkas et al [7] developed a sodium-based geopolymer for passive fire protection with a thermal conductivity of 0.27 W/m K at 300 °K.

Hence, as initial approach, this study aims to investigate the effect of water content to geopolymer solid (W/GPS) ratio on thermal conductivity of geopolymers. It is important to determine the optimal W/GPS ratio to improve the composition of geopolymers with other additives to produce a good insulating material.

2. Experimental

2.1. Materials
Fly ash (FA) from the Paiton power station in East Java, Indonesia was chosen as a source of aluminosilicate materials. True density of fly ash was 2.46 gr/cm³. Table 1 shows the chemical composition of FA as determined by X-ray fluorescence (XRF). Fly ash was classified as class C according to ASTM C 618-03 [8] because its has CaO contents higher than 10%. The activators used in this paper was combination between sodium silicate (SS) and sodium hydroxide (SH). Sodium silicate consist of 72.25% H₂O, 7.89% Na₂O and 19.86% SiO₂. Sodium hydroxide with 99 wt% of purity was supplied by Bratacem Co., Yogyakarta, Indonesia. Sodium hydroxide pellets and distilled water were mixed to obtain sodium hydroxide solution 10 M.

| Comp. | SiO₂ | Al₂O₃ | Fe₂O₃ | CaO | MgO | Na₂O | K₂O | TiO | MnO | Cr₂O₃ | SO₃ | LOI* |
|-------|------|-------|-------|-----|-----|------|-----|-----|-----|-------|-----|------|
| wt.%  | 38.90| 18.21 | 15.20 | 14.61 | 7.18 | 1.76 | 1.23 | 0.95 | 0.22 | 0.01 | 0.59 | 0.69 |

*Loss on ignition at 1000°C.

2.2. Preparation and synthesis of specimens
Geopolymer were synthesized with varied W/GPS ratio from 0.16 to 0.31. For clarity, all prepared samples are presented in Table 2. AA/FA varied from 25%-50% at a step of 5% while SS/SH was kept at 2.0. NaOH concentration used in this study was 10 M. The synthesis of geopolymer included three steps: (a) the preparation of activation solution, (b) the mixing of materials and (c) the molding and curing of specimen. FA and AA were mixed using Hobart mixer with the speed of 140 rpm for 5 minutes to form a homogenous slurry. The mixture was cast in a steel mold of 50 mm x 50 mm x 50 mm for compressive strength test. Bulk density used cylindrical acrylic mold of 10 mm height and 30 mm diameter. Thermal conductivity test used cylindrical acrylic mold of 2 mm and 4 mm height and 40 mm diameter. All test consist of three sample except for thermal conductivity test with one sample. The specimens were left for 24 hours at a room temperature and then it was demolded. All the samples were sealed with polyethylene film and cured under ambient conditions. Sample of fly ash-based geopolymers is represented in Figure 1.

Table 2 Mixture composition of fly ash-based geopolymers

| Mix No. | Composition of mixture (kg/m³) | W/GPS* | Si/Al (mol) |
|---------|-------------------------------|--------|-------------|
|         | FA | SS | SH |      |      |      |
| 1       | 1766.32 | 224.39 | 147.19 | 0.16 | 1.96 |
| 2       | 1672.02 | 334.40 | 167.20 | 0.19 | 1.99 |
| 3       | 1587.28 | 370.37 | 185.18 | 0.22 | 2.02 |
| 4       | 1510.71 | 402.86 | 201.43 | 0.25 | 2.05 |
*Water to geopolymers solid by mass ratio. Water including H₂O in AA. Solid including FA and solid in AA.

|    | 5     | 6     | 7     | 8     | 9     |
|----|-------|-------|-------|-------|-------|
| 1  | 1441.20 | 1377.80 | 432.36 | 459.27 | 216.18 |
| 2  | 0.28   | 0.31   | 2.09   | 2.12   | 0.31   |

2.3. Methods of Analysis

The 7-day compressive strength of geopolymers cube was measured. A Compressive testing machine from Avery Dennison was used in accordance with ASTM C109/109 M [9]. Three specimens were made for each variable. Thermal conductivity was recorded by thermal conductivity measuring apparatus (HVS-40-200 SE from Tokyo, Japan) at varied temperature (50°C, 100°C, 150°C and 200°C) in accordance with ASTM C 177 [10]. Fig.2 shows the testing machine used in this study. The bulk density value was calculated by dividing the mass by the volume of the samples.

3. Results and Discussion

3.1. Thermal conductivity

Fig 3 illustrates the influence of the W/GPS ratio on the geopolymers’ thermal conductivity. The geopolymers thermal conductivity was observed at temperature range from 50°C to 200°C. It was observed that the thermal conductivity of geopolymer varied with the increase of the temperature. Commonly, low thermal conductivity occurred when it was exposed to higher temperature. Six varied water to geopolymer ratio were reported to have a thermal conductivity in the range of 0.75 to 1.54 W/m°C. It has a similar value with the common building brick (0.69 W/m°C), and cement mortar (1.16 W/m°C), but it is higher if its compared with gypsum board (0.16 W/m°C) and particle board

(a)

(b)

Figure 1. Sample for compressive test (a) and thermal conductivity test (b)

Figure 2. Testing machine for (a) Compressive strength and (b) thermal conductivity test.

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The lowest thermal conductivity occurred when W/GPS was at 0.22. Overall, specimen shows the highest thermal conductivity at temperature of 150°C. W/GPS ratio at 0.22 conducted a low thermal conductivity below 0.80 W/m°C. This value was near to the thermal conductivities of various insulating material. Some typical values were of 0.038 W/m°C for glass wool and 0.78 W/m°C for window glass [11].

Thermal conductivity is influenced primarily by porosity [12]. Vickers et al [4] reported that porosity had the main effect on reducing thermal conductivity value. The smaller the size of pores, the lower the value of thermal conductivity. In addition, cracks and coarse pore of higher than 5 mm should be prevented. Sivasakthi et al. [6] found that bulk density is the main indicator to change the thermal conductivity, especially on higher temperatures. The amorphous structure of new minerals was formed partially when it was exposed to high temperature that decreased the density.

**Figure 3.** Thermal conductivity at different temperature of geopolymers paste

### 3.2. Bulk Density and Compressive Strength

Fig 4 shows the bulk density on different W/GPS ratio of fly ash-based geopolymer paste. It shows that bulk density of the sample increased firstly and decreased slightly with the increasing amount of water. The bulk density increased from 2113.93 kg/m³ to 2164.79 kg/m³ when the W/GPS ratio increased from 0.16 to 0.19. The bulk density decreased slowly to 1958.43 kg/m³ when the W/GPS 0.22 increased to 0.31. This may be due to the difference in density between fly ash and activating solution. The higher W/GPS ratio resulted in a slight amount of fly ash contained in the mixture that lowered the bulk density of the specimen. This finding is in agreement with those reported in the previous studies [13].

The result of compressive strength is as shown in Fig 4. It can be observed that compressive strength increased slowly from 27.12 MPa to 34.12 MPa before dropping drastically until 18.40 MPa. W/GPS ratio significantly affected compressive strength of fly ash-based geopolymers paste. Strength decreased as the ratio of W/GPS by mass increased. Khale and Chaudhay [3] found that this trend is similar to water-to-cement ratio in the compressive strength in OPC. Reducing the ratio of water to geopolymer solid obtained a higher compressive strength because it affected the volume of pores and porosity in the matrix.[14,15]. The result shows a similar trend with [16–18] for fly ash-based geopolymers or different alkali type[19].
Figure 4. Bulk density and compressive strength of various W/GPS ratio

4. Conclusion
The aim of this paper is to determine the optimum of water content to geopolymer solid (W/GPS) ratio on thermal conductivity of geopolymer paste. Six various mix design based on W/GPS ratio were studied. From the behaviour of geopolymer on thermal conductivity and compressive strength, it is revealed that it is clearly possible to use this material as an insulator. Thermal conductivity test has a similar result with other insulating material. It is proposed that the higher W/GPS ratio obtained lower compressive strength. Thermal conductivity act as the main indicator to determine the optimum W/GPS paste used at geopolymer synthesis in this study. Therefore, the suitable preparation of fly ash-based geopolymer paste with W/GPS ratio of 0.22 is recommended because its has lower thermal conductivity and the higher compressive strength.

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