

Evaluation of Pelletized Artificial Geopolymer Aggregate Manufactured From Volcano Ash

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Abstract: This paper introduced the new pelletized artificial geopolymer aggregate made from volcano ash. This is one of the effort to diminish the high quantity of volcano mud from East Java, Indonesia, that become a major issue since May 29, 2006 when it surfaced from the bowel of the earth and impacted an area of almost 770 hectare to a depth of 20 m, and thirty thousand people have been displaced which cost Indonesia \$3.7 billion in damages and damage control. The characterization of volcano ash was first examined. The major constituent was SiO₂ and Al₂O₃ for this material. The microstructure properties showed the volcano ash has a plate like structure and the shell covered outer surface of this aggregate appear naturally. The geopolymer artificial aggregate was examined with various curing temperature of 500 °C, 600 °C and 800 °C. The results show that the specific gravity of geopolymer artificial aggregate was in the range of 1.7-2.0 g/cm³. The lowest specific gravity was observed at sintering temperature of 800°C. The water absorption can be modulated by controlling the sintering temperature.

Key words: geopolymer, artificial geopolymer aggregate, XRF, XRD, density, water absorption.

INTRODUCTION

The high demand for construction materials and building products including aggregates need to find the new alternatives to fulfill the industry demand while the natural aggregate resource is depleting day by day. Some effort has been done on utilizing the new fly ash lightweight aggregate to be used in lightweight concrete (Monica *et al.*, 2005; Ramamurthy and Harikrishnan, 2006; Byung-wan *et al.*, 2007; Kayali, 2005; Sivakumar and Gomathi, 2012; Rafiza *et al.*, 2013). Introducing the new artificial geopolymer aggregate from volcano ash, this material is a novel geopolymer technology with low sintering temperature used. The idea to utilize the volcano ash as artificial geopolymer aggregate was based on the high quantity of volcano mud which erupted at Sidoarjo, East Java, Indonesia on May 29, 2006, two days after Yogyakarta earthquake of May 27, 2006 with a magnitude of 6.3 (Ryuta and Kiyoshi, 2009). Some expert stated that this eruption was caused by drilling of the gas exploration well in the Porong area, Sidoarjo, East Java (Davies *et al.*, 2007; Mazzini *et al.*, 2007; Davies *et al.*, 2008), then impacted an area of almost 3 square miles to a depth of 65 feet and thirty thousand people have been displaced which cost Indonesia \$3.7 billion in damages and damage control (Geoffrey *et al.*, 2008; Cryanoski, 2007). It has completely immersed many villages around the mining area.

It is also well known that geopolymers possess excellent mechanical properties, as well as fire and acid resistance (Guo *et al.*, 2010; Jimenez *et al.*, 2008; MMA Abdullah *et al.*, 2011). Davidovits (Davidovits, 2005) stated that geopolymers have applications including replacing cement in building materials, and as binders for concretes. In this research, the evaluation of pelletized artificial geopolymer aggregate from volcano ash using geopolymerization method has been studied as an effort to diminish the high quantity of volcano mud in Surabaya, Indonesia.

Experimental Method:

Preparation of Material:

The volcano mud was collected from the eruption site and transferred into sealed container. The original mud was medium gray in colour. Volcano mud was then dried in the oven at 105 °C for 48 hours. After drying, this material was then crushed and blended to form the volcano ash with particle size less than 300 µm.

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Materials Characterization:

Volcano ash samples were analyzed for different parameters. X-ray fluorescence (XRF) and X-ray diffraction (XRD) were performed to determine the raw materials characterization as geopolymer aggregate.

Mixing Process:

The materials used are volcano ash to be reacting with the geopolymer binder alkaline activator consisting of sodium silicate (Na_2SiO_3) and sodium hydroxide (NaOH) solutions. The sodium silicate has a chemical composition of 30.1% SiO_2 , 9.4% Na_2O and 60.5% H_2O (modulus $\text{SiO}_2/\text{Na}_2\text{O}=3.2$), specific gravity at $20^\circ\text{C} = 1.4 \text{ g/cm}^3$ and viscosity at $20^\circ\text{C} = 0.4 \text{ Pa s}$. The sodium hydroxide powder used was of 99% purity and fixed at 8M (Liew *et al.*, 2012) in this study.

The sodium hydroxide and sodium silicate was first mixed and stir until homogeneous solution was achieved to form as alkaline activator. The ratio $\text{Na}_2\text{SiO}_3/\text{NaOH}$ used was 0.6. Geopolymer paste were prepared by mixing volcano ash with the alkaline activator. The ratio of volcano ash/alkaline activator was 1.7. The mixing material was mixed for five minutes to obtain a homogeneous paste mixture. The paste need to be palleted as shown in Fig. 1 then dry at the temperature 60°C for 30 minutes to get the shape of the aggregate with 14-20 mm sizes. Then the pelletized artificial geopolymer aggregate was sintered at temperature 500, 600 and 800°C for 1 hour. The grain size distribution must meet the ASTM C 33 requirement for the use as artificial aggregate.



Fig. 1. Pelletized artificial geopolymer aggregate.

Testing and Analyzing:

The optical microscope and scanning electron microscope (SEM) with JSM-6460L model scanning electron microscope (JEOL) was performed to reveal the microstructure of artificial geopolymer aggregate at different sintering temperature. The water absorption and specific gravity were tested according to the ASTM C 127 (ASTM C 127, 2007) to determine the stability of the artificial geopolymer aggregate produced.

RESULTS AND DISCUSSION

Characterization of volcano ash:**Chemical composition of volcano ash:**

The chemical composition of volcano ash is shown in Table 1. The major constituents are SiO_2 with 38.5%. The content of Al_2O_3 for volcano ash is 14.2%. The content of Fe_2O_3 is 23.8%. The total of $\text{SiO}_2+\text{Al}_2\text{O}_3+\text{Fe}_2\text{O}_3 > 70\%$ indicated that this volcano ash can be used as pozzolan materials (ASTM C618-92a, 1994) and suitable to be used as a raw material for geopolymer.

Table 1: Chemical composition of volcano ash [21].

Compound	Al_2O_3	SiO_2	K_2O	TiO_2	Fe_2O_3	CaO	MnO	SO_3	V_2O_5
Conc Unit (%)	14.2	38.5	4.31	1.76	23.76	5.62	0.35	0.78	0.067

X-Ray Diffraction (XRD):

An x-ray diffraction (XRD) analysis of volcano ash is shown in Fig. 2. This volcano ash is almost dominated by quartz phase and amorphous in nature. This material exhibit a highest peak at 2 theta where 2 theta = 26.4° due to higher intensity of quartz (Q) revealed the silicon oxide. This statement also proved by the XRF results which shows that the higher contents of SiO_2 represent with high peak of intensity in XRD result. Volcano ash showed other five intense diffraction peaks at 2 theta values of 21.0° , 36.4° , 39.5° , 42.5° and

50.2°, which are associated with quartz (Q) (Mustafa *et al.*, 2012). The 2 theta values of 19.7° and 24.0° showed unidentified element (X).

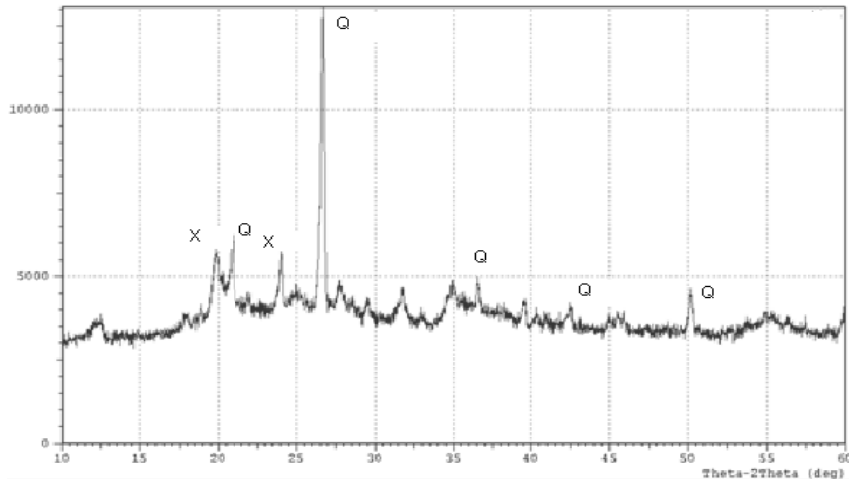


Fig. 2. XRD pattern of volcano ash

Microstructure Properties:

Stereo Microscope:

Fig. 3 examines the topography of the artificial geopolymer aggregate at various sintering temperature using an stereo microscope 200X and 350X magnification. It is clearly shows that the colour of artificial geopolymer aggregate was changed at each types of sintering temperature. Fig. 3(a) shows the artificial geopolymer aggregate was started to burned at 500 °C at the outer surface around an aggregate. Sintering temperature at 600 °C shows the aggregate burned at first stage constantly due to uniform colour as shown in Fig. 3(b). Fig. 3(c) shows this aggregate burned at second stage with higher temperature of 800 °C in the inner part. Artificial geopolymer aggregate at 500 °C shows grey in colour while light brown at 600 °C, then turned to dark brown at 800 °C sintering temperature for 1 hour. It can be seen that there is a thick shell with 934.2 µm and 255.0 µm at the outside surface of the aggregate at 500 and 800 °C sintering temperature, respectively.

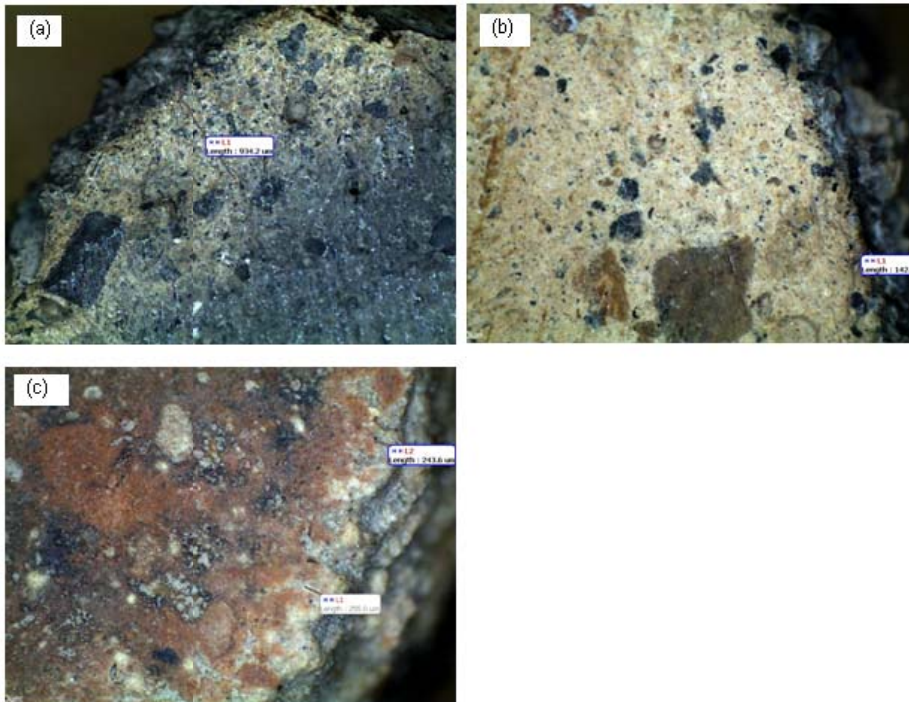


Fig. 3: Section of the artificial geopolymer aggregate sintering at (a) 500 °C; (b) 600 °C; and (c) 800 °C.

Scanning Electron Microscope (SEM):

Fig. 4 shows the SEM images of artificial geopolymer aggregate from volcano ash before and after sintered at 500 °C, 600 °C, and 800 °C at 2000X magnification. The particles observed as plate which forming a layer-like structure. The structure of artificial geopolymer aggregate before sintering shows layer stick together to form the structure due to the existence of water. The artificial geopolymer aggregate after sintered at 500 °C, 600 °C, and 800 °C showed the existence of pores marked with arrows as shown in Fig. 4(b), (c) and (d). More pores can be observed at sintering temperature of 500 °C. The smaller pores distributed uniformly in aggregate sintered at 600 °C indicate the better binding between volcano ash and binder but still results in higher water absorption. Some large voids can be observed at sintering temperature of 800 °C but this structure more dense with less smaller pores. The void sizes of artificial geopolymer aggregate were in the range of 115.0 - 319.2 μm. The plate-like structure contributed to smaller surface area for geopolymerization process compared to fly ash which has sphere microstructure.

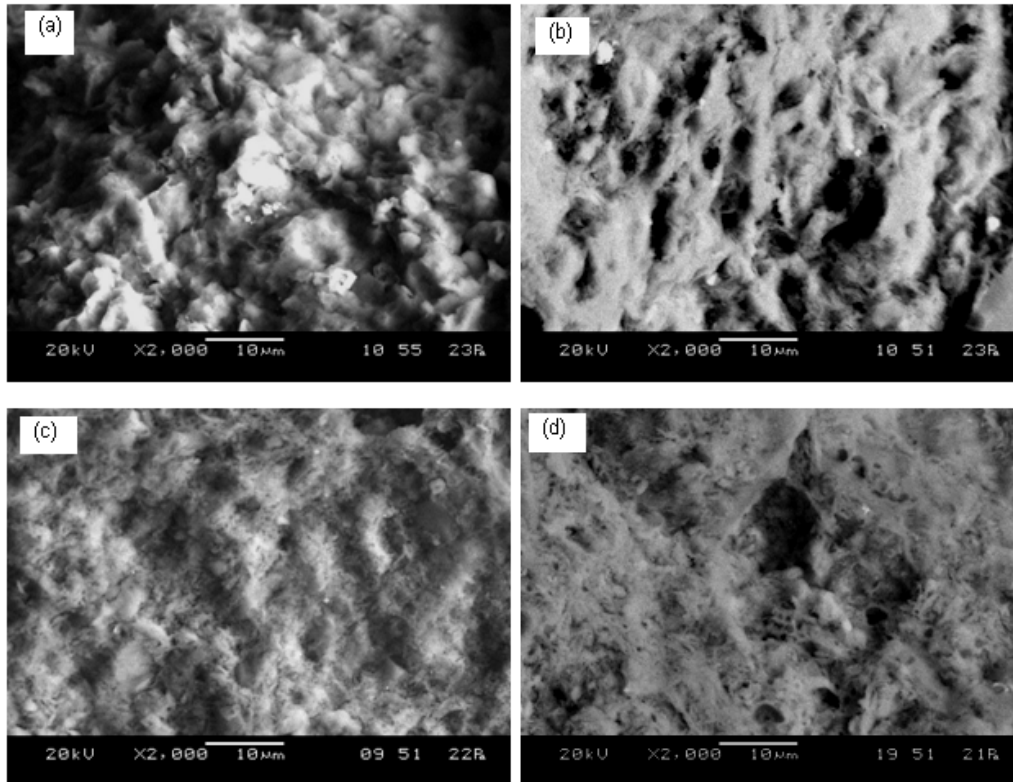


Fig. 4: SEM micrographs of artificial geopolymer aggregate at various sintering temperature (a) room temperature; (b) 500 °C; (c) 600 °C; and (d) 800 °C.

Mechanical Properties Of Artificial Geopolymer Aggregate:**Specific Gravity:**

The specific gravity of an aggregate is the mass of the aggregate in air divided by the mass of an equal volume of water (ACI Education Bulletin E1-07, 2007). This specific gravity is significant to control the density of concrete. Fig. 5 shows the specific gravity of artificial geopolymer aggregate produced at different sintering temperature. The higher sintering temperature will result in lower specific gravity value. The specific gravity of artificial geopolymer aggregate was in the range of 1.7-2.0 g/cm³. This value was comparable with the normal weight aggregate of 1.76 g/cm³ and lightweight aggregate determined by past researchers using fly ash (Monica *et al.*, 2005; Ramamurthy and Harikrishnan, 2006), but they need high temperature (more than 1000 °C) to get this value compared in this study (500-800 °C). The specific gravity below than 1.7 g/cm³ could be achieved by increasing the sintering temperature.

Water Absorption:

Water absorption is a measure of the total pore volume accessible to water and it can be calculated from a specific gravity determination (ACI Education Bulletin E1-07, 2007). The highest water absorption was found at sintering temperature of 600 °C with 16.0 %. The lowest water absorption can be found at sintering temperature

of 500 °C with 9.9 % as shown in Fig. 5. The range of water absorption for artificial geopolymer aggregate with volcano ash was 9-16 %. Other researcher (Ramamurthy and Harikrishnan, 2006) stated that the water absorption of sintered fly ash aggregate without binders was in the range of 21-22%. However, with the addition of 20% bentonite as binder, water absorption reduced significantly to 15-16% (Ramamurthy and Harikrishnan, 2006). The water absorption found by Byung-wan *et al.* (Byung-wan *et al.*, 2007) was 11.8% for alkali-activated fly ash lightweight aggregate. Monica *et al.* (Monica *et al.*, 2005) found lower water absorption of the fly ash lightweight aggregate when sintered at 975-1050°C. This shows that the water absorption can be modulated by controlling the expansion sintering temperature.

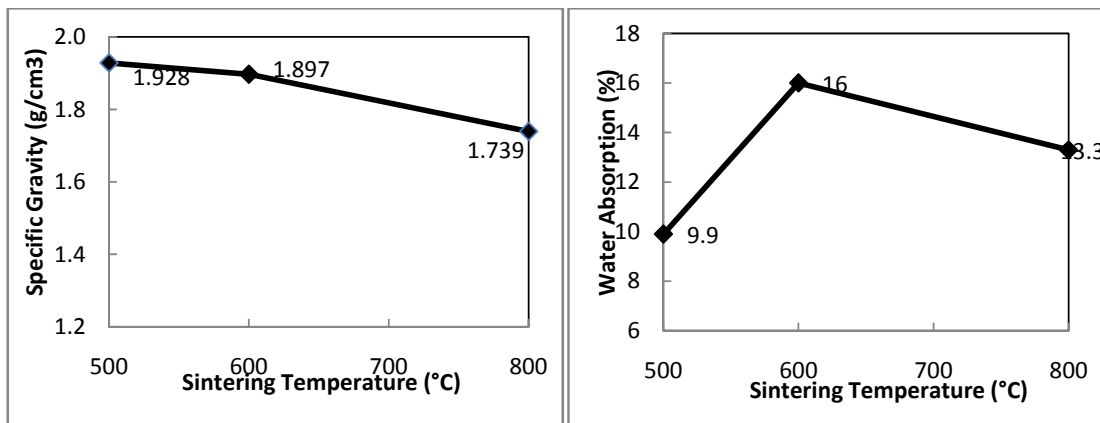


Fig. 5: The specific gravity and water absorption of artificial geopolymer aggregate of volcano ash at various sintering temperature.

Conclusion:

From the results produced, this volcano ash has a good potential to be commercialized as artificial geopolymer aggregate. The major constituents are SiO₂, Al₂O₃, and Fe₂O₃ with total 76.5 % > 70% indicated that this volcano ash is suitable to be used as a raw material for geopolymer. The volcano ash is almost dominated by quartz phase. There is a thick shell at the outside surface of the aggregate at 500 °C and 800 °C sintering temperature due to burning process and change the colour at different temperature. The particles observed as plate which forming a layer-like structure. Some large and small voids can be observed at all three types of sintering temperature. The specific gravity of artificial geopolymer aggregate was in the range of 1.7-2.0 g/cm³. The lower specific gravity could be achieved at higher temperature (>800 °C). The range of water absorption for artificial geopolymer aggregate with volcano ash was 9-16 %. The water absorption can be modulated by controlling the expansion sintering temperature. In the future works, the density and water absorption of this aggregate can be improved by finding the best design ratio during mixing.

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REFERENCES

- Mustafa Al Bakri, A.M., A.R. Rafiza, D. Hardjito, H. Kamarudin, and I. Khairul Nizar, 2012. Characterization of LUSI Mud Volcano as Geopolymer Raw Material. *Advanced Materials Research*, 548: 82-86.
- Rafiza, A.R., A.M. Mustafa Al Bakri, H. Kamarudin, I. Khairul Nizar, D. Hardjito, and Y. Zarina, 2013. Reviews on the Properties of Aggregates made with or without Geopolymerisation Method. *Advanced Materials Research*, 626: 892-895.
- ACI Education Bulletin E1-07, 2007. *Aggregates for Concrete*, in Developed by ACI Committee E-701, Materials for Concrete Construction, American Concrete Institute.
- ASTM C 127, 2007. Standard Test Method for Density, Relative Density (Specific Gravity), and Absorption of Coarse Aggregate.
- ASTM C618-92a, 1994. Standard specification for fly ash and raw or calcinated natural pozzoland for use as mineral admixture in Portland cement concrete. American Standard for Testing Materials. Annual book of ASTM Standards. Vol. 04.02, Pennsylvania.

- Byung-wan, J., P. Seung-kook, and P. Jong-bin, 2007. Properties of concrete made with alkali-activated fly ash lightweight aggregate (AFLA). *Cement and Concrete Composites*, 29: 128-135.
- Cryanoski, D., 2007. Volcano gets choke chains to slow mud. *Nature*, 45: 470..
- Davidovits, J., 2005. Geopolymer, Green Chemistry and Sustainable Development Solutions, Institute Geopolymer.
- Davies, R., M. Brumm, M. Manga, R. Rubiandini, R. Swarbrick, and M. Tingay, 2008. East Java mud volcano (2006–present): an earthquake or drilling trigger? *Earth and Planetary Science Letters*, 272: 627.
- Davies, R., R. Swarbrick, R.J. Evans and M. Huuse, 2007. Birth of a mud volcano: East Java, 29 May 2006, *GSA Today*, 4-9.
- Geoffrey, S., T.J.C. Plumlee, Handoko T. Wibowo, Robert J. Rosenbauer, Craig A. Johnson, and George N. Breit, 2008. Preliminary Analytical Results for a Mud Sample Collected from the LUSI Mud Volcano, Sidoarjo, East Java, Indonesia. U.S. Geological Survey, Reston, Virginia. 1-26.
- Guo, X., H. Shi, and W.A. Dick, 2010. Compressive Strength and Microstructural Characteristics of Class C Fly Ash Geopolymer. *Cement and Concrete Composites*, 32: 142-147.
- Jimenez, F., M. Monzo, M. Vicent, A. Barba and A. Palomo, 2008. Microporous and Mesoporous Materials, 108: 41-49.
- Kayali, O., 2005. Flashag - New Lightweight Aggregate for High Strength and Durable Concrete, 2005 World of Coal Ash (WOCA), Kentucky, USA.
- Liew, Y.M., H., Kamarudin, A.M. Mustafa, Al Bakri, M., Luqman, I. Khairul Nizar, C.M. Ruzaidi, C.Y., Heah, 2012. Processing and characterization of calcined kaolin cement powder. *Construction and Building Materials*, 30: 794-802.
- Abdullah, M.M.A., H. Kamarudin, H. Mohammed, I. Khairul Nizar, A. R. Rafiza, Y. Zarina, 2011. The relationship of NaOH Molarity, Na₂SiO₃/NaOH Ratio, Fly Ash/Alkaline Activator Ratio, and Curing Temperature to the Strength of Fly Ash-Based Geopolymer. *Advanced Materials Research*, 328-330: 1475-1482.
- Mazzini, A., H. Svensen, G.G. Akhmanov, G. Aloisi, S. Planke, A. Malthe-Sorensen and B. Istadi, 2007. Triggering and dynamic evolution of the LUSI mud volcano, Indonesia. *Earth and Planetary Science Letters*, 261: 375-388.
- Monica, A., A. Anselmo, M.R. Jesus and R. Maximina, 2005. Production of Lightweight Aggregates from Coal Gasification Fly Ash and Slag, 2005 World of Coal Ash (WOCA), Kentucky, USA.
- Ramamurthy, K. and K.I. Harikrishnan, 2006. Influence of binders on properties of sintered fly ash aggregate. *Cement and Concrete Composites*, 28: 33-38.
- Ryuta, H. and S. Kiyoshi, 2009. Environmental Assessment of Natural Radioactivity in Soil Samples from the LUSI Mud Volcano, Indonesi. *Environment Asia*, 2: 45-49.
- Sivakumar, A. and P. Gomathi, 2012. Pelletized fly ash lightweight aggregate concrete: A promising material. *Journal of Civil Engineering and Construction Technology*, 3(2): 42-48.