DEVELOPMENT OF VOLCANIC MUD-BASED GEOPOLYMER ARTIFICIAL AGGREGATE

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ABSTRACT

Several attempts have been made to utilize volcanic mud from Sidoarjo, East Java, Indonesia, which has been erupting since May 2006. More than 600 hectares of productive land, housing and industrial areas, as well as infrastructures have been submerged. To date, after more than six years, there is no sign that the eruption will end in the near future. The contents of mud are predominantly SiO_2, Al_2O_3 and Fe_2O_3 in crystalline form. This paper reports an attempt to utilize the volcanic mud as artificial aggregate through geopolymerisation process. After pre-treatment in the form of calcinations and grinding, the volcanic mud is found to be reactive. Geopolymerisation is performed by mixing the more-amorphous form of the volcanic mud with the highly alkaline solution, followed by curing in an elevated temperature. The artificial aggregates resulted from this process is found to remain stable after 24 hours immersing in the water. The specific gravity of the artificial aggregates varies widely in between 1470-1800 kg/m^3, depending on the mixture composition, which can be attributed as lightweight aggregate. This study reveals the potential of volcanic mud from Sidoarjo, East Java, Indonesia, to be used as artificial aggregates, although further improvement in its characteristics needs to be carried out.

INTRODUCTION

Since May 2006, the eruption of volcanic mud in Sidoarjo, Indonesia, has been continuing; submerging more than 600 hectares of productive land, housing and industrial areas, schools and other infrastructure facilities. The eruption is predicted to last for decades (Rudolph, Karlstrom et al. 2011). To date, the height of the embankment dam encircled the submerged area is approximately 12 metres, and unlikely it can be increased anymore due to the safety reason, although the discharge is continuing (Agustawijaya and Sukandi 2012). To alleviate the pressure to the embankment dam, the mud is channelled to a nearby river, causing a newly formed island on its mouth.

The mud is rich in SiO_2, Al_2O_3 and Fe_2O_3 in crystalline form; and thus, in most cases, its use requires pre-activation in the form of calcinations and grinding (Nuruddin, Bayuaji et al. 2010; Hardjito, Antoni et al. 2012; Hardjito, Antoni et al. 2013). It has been reported that the mud possesses pozzolanic properties, enable its use to partially replace the use of cement in making mortar; i.e. about 10% cement replacement (Nuruddin, Bayuaji et al. 2010), or even in a semi high volume up to 40% cement replacement (Hardjito, Antoni et al. 2012). The authors previous study revealed that the calcined mud is also a suitable precursor for geopolymer, especially those with particle size less than 150εm (Mustafa Al Bakri, Rafiza et al. 2012; Rafiza, Bakri et al. 2012; Hardjito, Antoni et al. 2013).

Fansuri et al. (2012) reported the successful use of fly ash as raw material for manufacturing artificial aggregate using geopolymerisation technique. This study focuses on the possibility to utilize the Sidoarjo volcanic mud (also called LUSI, a short form of **LU**mpur **SI**doarjo, mud), as a base material for making geopolymeric artificial aggregate. This study is part of an effort to develop light-weight artificial aggregate utilizing Sidoarjo volcanic mud through geopolymerisation approach.

MATERIALS AND METHODS

The fresh volcanic mud was collected directly from the eruption site in Sidoarjo, East Java, Indonesia. The fresh mud was molded to be brick-like shape and dried in the oven at 100^0C for 24 hours to remove the moisture. It was then sent to a local tile manufacturer for calcinations at 945^oC for about 5 hours. Following the calcinations, the mud was ground in a rod-mill machine for about three hours, and then sieved to obtain fraction with particle size less than 63εm. Characterizations of mud were performed using the XRF analysis. The main objectives of calcining and grinding are to enable the mud particles to be more reactive, by changing its microstructure from crystalline into more amorphous form and to increase its specific surface area.

Tab. 1. The chemical composition of volcanic mud as measured by XRF (% by mass)

SiO_2	Al_2O_3	Fe_2O_3	MgO	Na_2O	CaO	K_2O	SO_3	TiO_2	MnO_2	Cr_2O_3	LOI
56.75	23.31	7.37	2.95	2.70	2.13	1.04	0.96	0.38	0.14	0.01	1.20

Table 1 shows the chemical composition of the calcined mud as measured by XRF analysis. The mud is rich in SiO_2, Al_2O_3 and Fe_2O_3, whereby the total content of these three components is more than 85%, indicating its potential to be a pozzolanic material and precursor for geopolymer. From the XRD analysis carried out in the previous study, it was revealed that the calcined volcanic mud is in a more amorphous form (Antoni, Hardjito et al. 2012). Using Scanning Electron Microscope (SEM), it was shown that the particles of the mud is plate-like shape, with particle size predominantly about 20 εm.

A combination of sodium silicate solution and sodium hydroxide solution was chosen to be the alkaline activator for geopolymerisation. Sodium silicate solution with Na_2O content of 17.14%, SiO_2 content of 36.71% was used throughout the study, while sodium hydroxide in flake form with 98% purity was chosen. Tap water was used to mix the alkaline activator.

Two stages of study were carried out, with mixture proportions were prepared based on the work by Fansuri et al. (2012). In first stage, only sodium silicate solution was used as the alkaline activator, while in the second one a combination of sodium silicate and sodium hydroxide solution was utilized. In first stage, the content of sodium silicate was taken as the parameter. Water was added until the required workability was achieved; i.e. the required workability for pelletizing the fresh geopolymeric paste, i.e. with the flow diameter in the range of 110-125mm; measured by using the flow table apparatus. The more the content of the sodium silicate used, the less the amount of added water needed to achieve the required workability. In this study, pelletizing was performed manually by using hands. The details of mixture proportion for the first stage are shown in Table 2. Each mixture composition was prepared enough to make three cubes of 50x50x50mm.

Tab. 2. Mixture Proportion of Paste with only Sodium Silicate Solution as the Alkaline Reactor

Specimen	Volcanic Mud, by mass (gr)	Sodium Silicate solution, by mass (gr)	Added Water, by mass (gr)	Flow Diameter (mm)
SS00-1	600	90	225	112
SS00-2	600	120	220	126
SS00-3	600	180	155	121
SS00-4	600	240	100	127

In the second stage, a combination of sodium hydroxide and sodium silicate solution was chosen as the alkaline reactor, while the content of the sodium hydroxide was kept constant. Water was added in order to achieve the required workability for manual pelletizing. For this mixture, it was found that the flow diameter of the paste was in the range of 155-165mm. The details of the mixture composition are shown in Table 3.

After manual pelletizing to form circular artificial aggregate with diameter about 20mm, the fresh pellets were then cured in the oven at 100^{o}C for 24 hours duration. Properties of the artificial aggregates observed were the stability of the aggregates after soaking in the water for at least 24 hours. Compressive strength and unit weight of the artificial aggregates were measured on the cubes specimens of

50x50x50mm at the age of 7 days in accordance to the relevant ASTM standards. Each data presented in various tables is the mean value of the three test results.

Tab. 3. Mixture Proportion of Paste with Combination of Sodium Hydroxide and Sodium Silicate Solution as the Alkaline Reactor

Specimen	Volcanic Mud, by mass (gr)	Sodium Hydroxide, solid, by mass (gr)	Sodium Silicate solution, by mass (gr)	Added Water, by mass (gr)	Flow Diameter (mm)
SSSH-1	600	6.3	90	190	160
SSSH-2	600	6.3	120	165	162
SSSH-3	600	6.3	180	115	162
SSSH-4	600	6.3	240	90	156

RESULTS AND DISCUSSION

Properties of Artificial Aggregates with only Sodium Silicate Solution as the Alkaline Reactor

Table 4 shows the properties of the volcanic mud-based artificial geopolymeric aggregates. Artificial aggregates made from all four different sodium silicate solutions in this stage was found stable after soaking for 24 hours in the tap water. The unit weight of the artificial aggregates range from 1470 to 1760 kg/m^3, which satisfies the requirement for lightweight aggregates (Badan Standarisasi Nasional 2002).

Tab. 4. Properties of Artificial Aggregates with only Sodium Silicate Solution as the Alkaline Reactor

Specimen	Unit Weight (kg/m^3)	Stability after Soaking in the Water for 24hrs	Compressive Strength (MPa)
SS00-1	1470	OK	0.59
SS00-2	1550	OK	0.31
SS00-3	1608	OK	5.73
SS00-4	1706	OK	8.40

The unit weight and the compressive strength of the aggregate is increased with the increase of the sodium silicate content, whereby the artificial aggregate with low sodium silicate content (specimens SS00-1 and SS00-2) shows very small compressive strength. Initially, it was expected that with the increase in the sodium silicate content, the unit weight of the artificial aggregate is reduced, as the more sodium silicate content induces swelling of the aggregate (Fansuri, Prasetyoko et al. 2012). Increase in the compressive strength is understandable, as the more the sodium silicate content provides more alkali content to react with the Si and Al atoms in the mud.

Properties of Artificial Aggregates with Combination of Sodium Hydroxide and Sodium Silicate Solution as the Alkaline Reactor

Table 5 shows the properties of the volcanic mud-based artificial geopolymeric aggregates with a combination of sodium hydroxide and sodium silicate solution as the alkaline activator. All four specimens are stable after soaking 24 hours in the tap water. The unit weight of the aggregates were ranged in between 1610 to 1800 kg/m^3, which satisfies the requirement for lightweight aggregates, i.e. between 1000-1800 kg/m^3 (Badan Standarisasi Nasional 2002).

Tab. 5. Properties of Artificial Aggregates with Combination of Sodium Hydroxide and Sodium Silicate Solution as the Alkaline Reactor

Specimen	**Unit Weight (kg/m^3)**	**Stability after Soaking in the Water for 24hrs**	**Compressive Strength (MPa)**
SSSH-1	1610	OK	3.20
SSSH-2	1660	OK	2.53
SSSH-3	1780	OK	6.80
SSSH-4	1800	OK	8.67

Compared to aggregates prepared only by using sodium silicate as the alkaline reactor; aggregates prepared by using a combination of sodium hydroxide and sodium silicate solution resulted in a slightly higher unit weight as well as compressive strength. The higher the alkaline content, the higher the unit weight and the compressive strength. Compared to commonly available artificial aggregates, such as those of expanded clay with unit weight about 800 kg/m^3, the unit weight of artificial aggregates resulted in this study is still very big. Further improvement on its characteristics is imperative.

CONCLUSIONS

This study reveals the possible use of Sidoarjo volcanic mud as base material for making lightweight artificial aggregate through geopolymerisation. All the artificial aggregates produced in this study are stable after soaking in the tap water for 24 hours, with unit weight and compressive strength vary widely in between 1470-1800 kg/m^3 and 0.3-8.7 MPa, respectively, depending on the mixture composition. The higher the content of the alkaline reactor, the higher the unit weight and the compressive strength of the artificial aggregate. Further study needs to be carried out, especially in optimizing the mixture composition, to lower its unit weight.

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