The Effects of Na2SiO3/NaOH Ratios on the Volumetric Properties of Fly Ash Geopolymer Artificial Aggregates

by I Dewa Made Alit Karyawan

The Effects of Na_2SiO_3/NaOH Ratios on the Volumetric Properties of Fly Ash Geopolymer Artificial Aggregates

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Abstract This study was conducted as a part of a research to assess the influence of the volumetric properties of geopolymer artificial aggregates made of fly ash to the performance of asphalt mixture to be used in pavement layers. This paper presents the parameters adopted during the production of geopolymer artificial aggregates which may affect the volumetric properties and the structure of the aggregates. The investigated volumetric properties included specific gravity and water absorption. In the experiment laboratory, two variables were utilized for producing the artificial aggregates, which might affect the considered volumetric properties. Those variables are pan-granulator slopes and alkaline ratios (the ratios of Sodium Silicate to Sodium Hydroxide). The pan-granulator slopes were set at 3 different angles, i.e. 45°, 50°, and 55°. The selected alkaline ratios were 1.5, 2.0, 2.5, and 3.0. The test results indicated that the best volumetric properties were obtained at a slope of 50^0 and alkaline ratio of 2.5. The bulk specific gravity values at the best volumetric properties were found to be: 1) oven dry at 1.9 grams/cm^3; 2) saturated surface dry (SSD) at 2.0 grams/cm^3, and 3) apparent at 2.1 grams/cm^3 with 6% water absorption. In addition, the interfacial transition zone and microstructure aggregates were examined by using Scanning Electron Microscope (SEM). In this study, it was found that there were effects of Na_2SiO_3/ NaOH ratios and the granulation method on the volumetric properties.

Introduction

The increasing of infrastructure development has caused an increase in aggregate demand. Aggregates are the main ingredients for concretes, both portland cement concrete and asphalt concrete. Aggregates are produced by crushing natural rocks into smaller sizes, according to the required sizes, in gradations. Therefore, natural rock explorations are carried out to supply the required aggregates in the infrastructure industry which causes the resource to decrease [1]. As an illustration, to make a mixture of asphalt concrete, 90%-95% of the aggregates from the mixture weight is used which is equal to 75%-85% of the mixture volume [2]. The efforts to fulfill this aggregate needs include creating artificial aggregates. One of them is by using fly ash geopolymer [3][4]. Fly ash geopolymers have higher mechanical strength, temperature resistance, and durability in comparison with cement. Due to this nature, fly ash geopolymers gain more attention and interest from researchers. An investigation on the fly ash geopolymer durability with metakaolin at high temperatures and acids conclude that geopolymers have better durability compared to Portland cement under the same conditions [5].

*corresponding author

Artificial aggregates, used for construction, must have several properties to comply with the requirements. Therefore, to be used as an asphalt concrete mixture, these properties must be identified through testing. These properties include specific gravity and absorption [6]. Geopolymer aggregates based on fly ash holds the potential to be utilized as artificial aggregate materials because their quality can be improved in the mixing process [1]. The process of making aggregates with fly ash materials includes making pellets through a sintering process by heating the materials above 1,000^{o}C [7] and through pellet making by cold bonding methods [3][7]. Geopolymerization is highly dependent on the fly ash's physicochemical properties and on the availability of silicate solutions and aluminate. In this case, an additional concentration of sodium hydroxide, capable of increasing the amount of silicate and aluminate, can be dissolved in the mixture. Therefore, the ratio of alkali activators plays a crucial role in the geopolymerization process [4].

This paper is a part of a study on the manufacture of geopolymer aggregates made from fly ash and alkali activators. This paper only explained specific gravity properties and aggregate absorption. The emphasis was on the effects of pan granulator slopes and the alkali activator ratios on the volumetric properties of artificial aggregates. This resulted in a part of the research to get aggregate with the properties that complies the requirements to be utilized as a mixture of asphalt concrete.

Experiment

The mixture material was fly ash taken from Suralaya Steam Power Plant Unit 1-4, Indonesia. The chemical content of fly ash based on the XRF test is indicated in Table 1. The main difference between class F fly ash and class C fly ash is that the latter contains a higher amount of calcium [8]. Fly ash is classified as class C when the CaO content is more than 10% and the amount of SiO_2, Al_2O_3, and Fe_2O_3 is more than 50%. The proportion of fly ash in geopolymer paste was 75%, while the alkali activator amounted to 25%. The basic activator contained 8M of Sodium Silicate (Na_2SiO_3) and Sodium Hydroxide (NaOH). The aggregates were made through cold mixing process with granulation system using a granulator. The materials for making artificial aggregate are presented in Figure 1.

Table 1 Chemical composition of fly ash based on XRF test results

No	Compound	Content (%)	No	Compound	Content (%)
1	MgO	0.20	9	MnO	0.35
2	Al_2O_3	18.05	10	Fe_2O_3	18.45
3	SiO_2	45.00	11	CuO	0.04
4	K_2O	1.47	12	ZnO	0.05
5	CaO	10.85	13	MoO_3	3.29
6	TiO_2	1.70	14	BaO	0.27
7	V_2O_5	0.07	15	Re_2O_7	0.25
8	Cr_2O_3	0.03	16	Na_2O	-

Fig 1. Materials for making aggregates

The pan granulator slopes were altered by adjusting the screw on the back of the pan as seen in Figure 2. The properties of aggregates as a mixture of asphalt concrete include specific gravity, absorption, hardness, adhesion to asphalt, durability, and impact. Nevertheless, this paper only discussed the volumetric properties, i.e. specific gravity and absorption. Specific gravity consists of bulk, apparent, and effective specific gravity. Specific gravity depends on the element and the porosity of the aggregate particles which can be defined as follows: a) all of the pore spaces (bulk specific gravity); b) some of the pore spaces (effective specific gravity); c) none of the pore spaces (apparent specific gravity) [9]. The absorption was in comparison with the change in the aggregate mass because of the presence of water absorbed in the pore space in constituent particles with dry conditions [10].

Fig 2. Granulator and pan slope

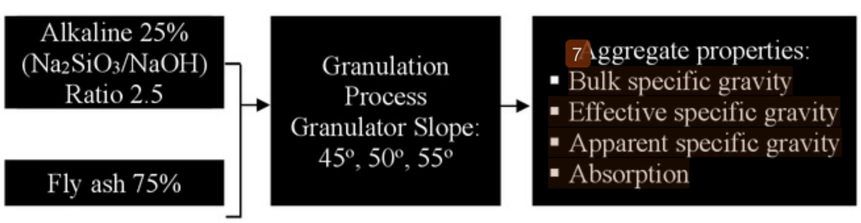


Fig 3. Experiment to find the best slope [11]

The influential parameters in the process of making pellets included (i) rotation speed of pelletizer discs; (ii) angle of pelletizer disc; (iii) content moisture; and (iv) pelletization duration [12]. To determine the effects of the slopes on volumetric properties, 3 slope degrees were trialed, i.e. 45°, 50°, and 55° with an alkali activator ratio of 2.5 [11]. The best properties produced from these 3 slopes were then selected. The slope that produced the best properties were utilized in an attempt to create aggregates with activator ratios of 1.5, 2.0, 2.5 and 3.0. From the experiment, the best angle and ratio were identified to obtain the best volumetric properties.

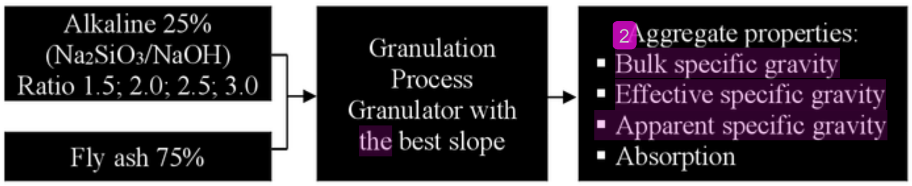


Fig 4. Experiment to find the best alkaline ratio

All test of artificial aggregates were carried out in the laboratory of Institut Teknologi Sepuluh Nopember, Surabaya. The aggregates were tested after 28 days [13]. The properties of volumetric aggregates, i.e. specific gravity and absorption, were tested at the Laboratory of Transportation and Road Materials of Institut Teknologi Sepuluh Nopember. The XRF testing was conducted to determine the chemical composition of fly ash [4]. Furthermore, SEM test was performed on the best artificial aggregates from the property test analysis results. The SEM test results were employed to determine the density of the matrix and fly ash reaction [4]. XRF and SEM tests were conducted at the Energy Laboratory of Institut Teknologi Sepuluh Nopember.

Results and Discussion

Aggregate properties based on pan granulator slope variations

The aggregates made with a ratio of Na_2SiO_3/NaOH of 2.5 and slopes of 45°, 50° and 55° are shown in Figure 5 as well as the test results of volumetric property value based on pan granulator slopes. The aggregates were made by setting the ratio of Na_2SiO_3/ NaOH at the value of 2.5. In the picture, it is evident that specific gravity did not indicate a linear correlation. On the other hand, in general, 50^0 slope had the lowest value, except for the apparent specific gravity. The Normal Bulk Specific Gravity of 2.4 to 2.9 was set for natural aggregates [14]. The results obtained from 3 slopes did not correlate with the natural aggregates.

45° Slope 50° Slope 55° Slope

Fig 5. Artificial aggregates with $Na_2SiO_3/NaOH$ ratio of 2.5 and slope variations [11]

In Figure 6 on absorption values, it was indicated that the smallest value was obtained at 50° slope. The absorption requirement to be asphalt mixture was 3% at most. Because all absorption values obtained were less than 3%, the best value was obtained at the slope angle of 50°. The granulation process speed affected the specific gravity and absorption [12]. In Figure 6, it is evident that slopes did not correlate with the absorption level.

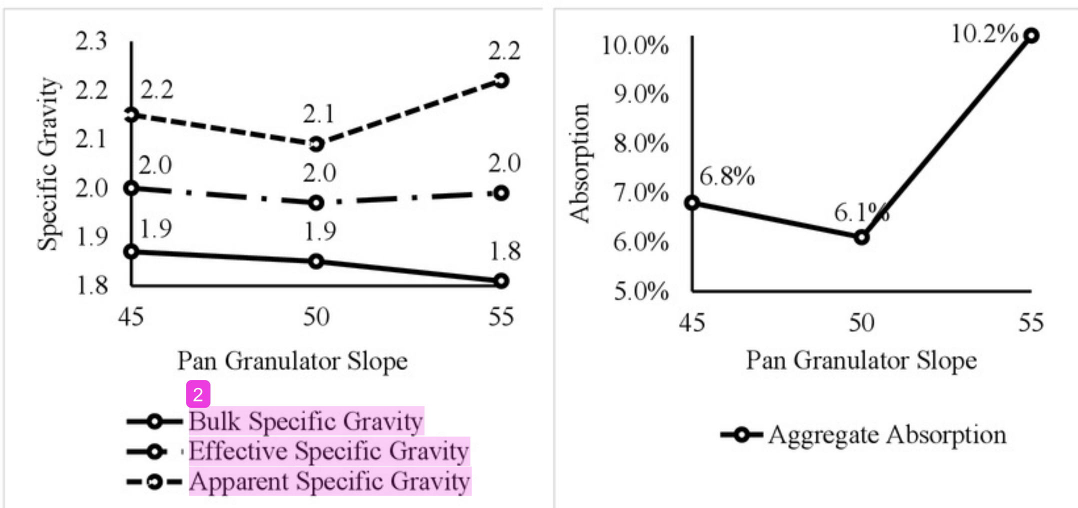


Fig 6. The correlation between pan granulator slopes and specific gravity and absorption [11]

Aggregate properties based on alkaline ratio variations

Based on the test results in Figure 6, to get the properties based on variations in the ratios of $Na_2SiO_3/NaOH$, the pan granulator slope was set at 50°. Figure 7 displayed the aggregates made with 50° slope and variations in the ratios of $Na_2SiO_3/NaOH$ of 1.5, 2.0, 2.5 and 3.0. The test results in Figure 8 suggested the value of volumetric properties based on pan granulator slopes. In the figure, it can be seen that specific gravity had the same trend on the effective specific gravity and apparent specific gravity. However, ratio 2.5 suggested a different tendency for bulk specific gravity. The results indicated that a high ratio did not necessarily produce a high value or vice versa. The normal bulk specific gravity were 2.4 to 2.9 for natural aggregates [14]. The results obtained from the 3 ratios indicated that the properties of artificial aggregates did not correlate with the natural aggregates.

There was an effect of alkaline ratios on the compressive strength of geopolymer paste [15][16]. In related research about the correlation of compressive strength and the ratio of Na_2SiO_3/NaOH, the ratios of 0.5, 1, 1.5, 2, and 2.5 were utilized. It was found that the optimum ratio of 2 and 2.5 resulted in the highest compressive strength of 2.86 MPa [17][18]. Therefore, to obtain good aggregate properties, the alkali activator needs to be modified. Such a modification can be made on the use of alkali activator/fly ash ratio, Na_2SiO_3/ NaOH ratio, and NaOH molarity. Therefore, a trial must first be conducted to obtain good properties. Higher compressive strength will produce better aggregate properties [1].

Fig 7. Artificial aggregates with a slope of 50º and variations in the ratios of Na_2SiO_3/NaOH

In Figure 8, at a ratio of 2.5, the smallest absorption values were obtained. To obtain the maximum absorption requirements of 3% to make asphalt mixture, the best value was obtained at an alkali ratio of 2.5 [11].

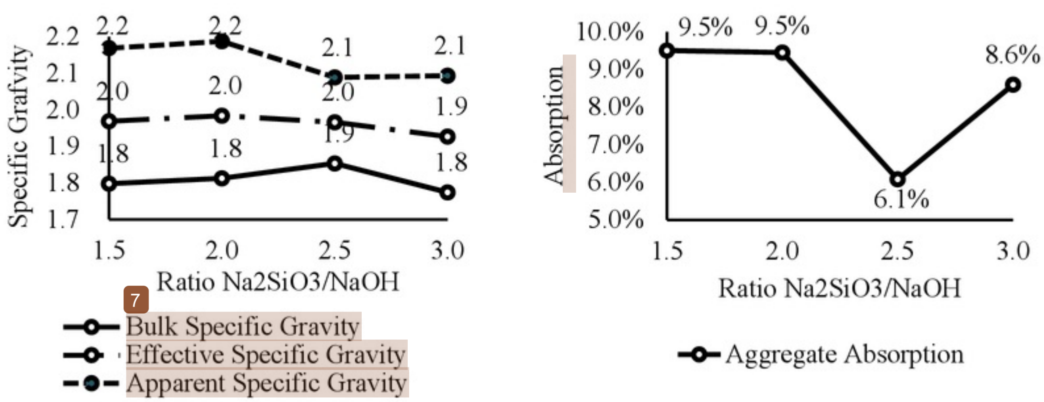
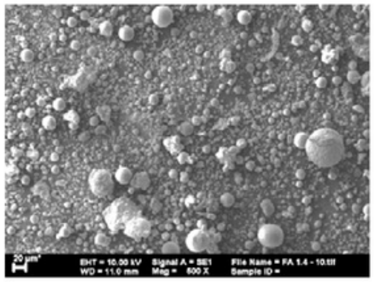


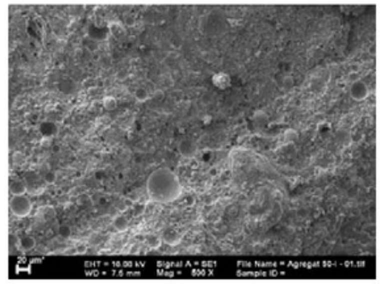
Figure 8. The correlation between specific gravity and absorption with Na_2SiO_3 and NaOH ratios

Based on the obtained results, artificial aggregates did not meet the requirements as asphalt mixture. However, from these results, the best aggregates were obtained at 2.5 ratio of Na_2SiO_3/ NaOH with 8M NaOH and 50º slope [11]

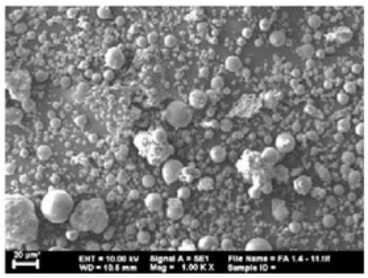
In Figure 9, the fly ash morphology characterization analysis with SEM indicated fly ash microstructure with various features of size, shapes, and surface. The particles were mostly spherical called as cenospheres[19]. On the other hand, the artificial aggregates suggested that in this proportion, good solidification on the fly ash granules with alkaline activators took place. In addition, there was a geopolymerization reaction between fly ash and alkali. This was evident where most of the fly ash particles were dissolved by the alkaline activator. Only a few reacted irregularly or partially [13].



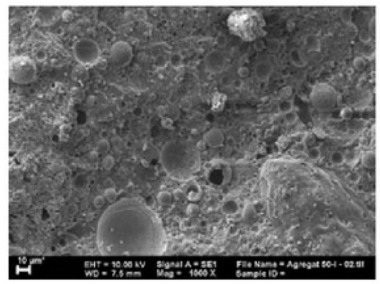
Fly ash (500X)



Artificial Aggregates (500X)



Fly ash (1,000X)



Artificial Aggregates (1,000X)

Fig 9. Morphology of fly ash and artificial aggregates with 50° slope and 2.5 ratio

Summary

- The aggregate properties varied for each difference in the alkaline ratio ($Na_2SiO_3/NaOH$). The difference in the ratio of $Na_2SiO_3/NaOH$ was not directly proportional to the specific gravity of the aggregates. The same phenomenon occurred for absorption where it had no correlation with the increase in the alkaline ratio. The smallest absorption took place at the ratio of $Na_2SiO_3/NaOH$ of 2.5.
- Two variables were employed in this study, namely the granulator slopes and the alkali ratios. The results indicated that both variables caused effects on aggregate volumetric properties. However, these effects had no linear correlations, meaning that a high tilt angle did not necessarily increase the volumetric properties or vice versa. The same correlation also applied for the alkaline ratios.

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