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Potential Use of Fly Ash Base-Geopolymer as Aggregate Substitution in Asphalt Concrete Mixtures

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Abstract-Infrastructure industry is still dominant in developing countries. These activities requires a large number of aggregates. To make cement concrete and asphalt concrete, it requires 60% to 75% of aggregate in total volume of the mixture. This high volume of aggregates may cause reduction on availability of natural aggregate. Another problem is that not all area in Indonesia has the ability to provide adequate aggregates so that it can support the infrastructure development, especially in remote areas. The aggregate mobilization may face disruption. This research attempts to present an idea of creating an artificial aggregate. The artificial aggregate is made of power plant waste that is mixed with alkali silica, named as fly ash geopolymer. Previous study indicates that the use of fly ash geopolymer as filler replacement in asphalt concrete mixture, is able to double the stability of Marshall test. This experiment serves a role to design an artificial aggregate.

Keywords -artificial aggregate, fly ash geopolymer, asphalt mix materials

I. INTRODUCTION

Increasing infrastructure development in most parts of the world, especially in developing countries, has resulted in reduced availability of raw materials. The use of natural resources to produce raw materials on an ongoing basis can threaten environmental conditions. Aggregates are one of the raw materials produced from rock exploration in nature. The aggregate is made of stone-crusher installation. The provision of aggregates to meet the natural aggregate needs of the infrastructure industry leads to depletion of natural resources [1][2]. This is understandable considering to make concrete cement and asphalt concrete, it requires 60% to 75% of aggregate of the total mixed volume [3]. The need for raw materials in the form of aggregate to support this infrastructure development cannot be fully met in every Indonesia area, especially in remote areas. There can be various obstacles in aggregate mobilization because all of Indonesian areas are not provided by adequate transportation facilities and infrastructures. Thus, it is necessary for sustainable efforts to develop alternative materials such as construction and dismantling waste and industrial waste to be used as replacements of natural aggregates [3]. There have been many studies using waste as artificial aggregates. A research using dismantling of buildings or debris caused by earthquake, for example brick, has been done to find new alternative material in the form of coarse aggregate of waste material. The use of brickle brick can reduce the problem and simultaneously help the conservation of natural resources [1]. There is also an artificial lightweight aggregate production using bottom ash from urban solid waste combustion. This is applied by stabilizing the bottom ash by adding cement, lime as a binder and made of using rotary granulator pan [4]. Another artificial aggregate research is conducted by recycling stone sludge and waste silt. Stone sludge is taken from stone slab processing and waste silt is taken from aggregate wash site. The analyzes include compressive strength, absorption, and artificial aggregate resistance to abrasion under vibration. As a result, water absorption depends on packing density, applied compaction vibration, filling the cavity between the aggregate sludge and the stone waste sludge. The results present denser structure, thus it can reduce the amount of using compacting agent, decrease the water cement ratio, and ultimately increase the compressive strength [5]. Production of light aggregate from fly ash geopolymer has been done by agglomeration technique, including: procurement of row material, composition, mixing, agglomeration to form pellet. In agglomeration process, there is agitation or pressure provision. The agitation manner depends only on the strength of fall with addition of bonding agents such as water glass, kanji solution, and so on. The compaction manner is using a piston suppressor so that it can reduce pores, then it can produce very dense briquettes. Furthermore, the hardening process is by sintering, autoclaving and cold bonding, then there will be aggregatedestruction. To form the aggregate, it is sieved to obtain appropriate size of the classification [6]. The above description creates an idea to use waste as an artificial

aggregate material. Waste to be used is fly ash. Fly ash is the result of coal use as a waste from power plants; while there is currently less than 50% of the fly ash utilization. The issue of industrial waste disposal has become an environmental issue, due to pollution caused primarily in big cities where there are limited landfills [1][2]. This research is conducted by an idea to make artificial aggregate from fly ash base-geopolymer.

The artificial aggregates are made of fly ash geopolymer paste. This paper discusses the results of a compressive strength test of fly ash geopolymer paste using a cylindrical sample. The resulting compressive strength is compared with other research results. The sample used is cylindrical with 5 cm-diameter and 10 cm-height. Further issue discusses the possibility of the fly ash utilization as an aggregate material based on the results of previous research.

II. METHOD

A. Experimental program

The geopolymer paste is made in 3 mixtures design based on Na_2SiO_3 ratio with NaOH, namely 1.5, 2.0, and 2.5. The composition of fly ash and alkali is 75%: 25%. Each design mixture is used in 6 samples, therefore it requires total of 18 samples. The mixture design can be seen in Table 1 below.

TABLE 1. The Design of A Fly Ash Geopolymer Paste Mixture

No.	Code Samples	Fly ash (%)	Alkali (%)	Molarity NaOH (mol / liter)	$\text{Na}_2\text{SiO}_3 / \text{NaOH}$
1	1.5-1	75	25	8	1.5
2	1.5-2	75	25	8	1.5
3	1.5-3	75	25	8	1.5
4	1.5-4	75	25	8	1.5
5	1.5-5	75	25	8	1.5
6	1.5-6	75	25	8	1.5
7	2.0-1	75	25	8	2.0
8	2.0-2	75	25	8	2.0
9	2.0-3	75	25	8	2.0
10	2.0-4	75	25	8	2.0
11	2.0-5	75	25	8	2.0
12	2.0-6	75	25	8	2.0
13	2.5-1	75	25	8	2.5
14	2.5-2	75	25	8	2.5
15	2.5-3	75	25	8	2.5
16	2.5-4	75	25	8	2.5
17	2.5-5	75	25	8	2.5
18	2.5-6	75	25	8	2.5

B. Materials

(1) Fly ash

Fly ash used is the remaining coal combustion from Paiton Steam Power Plant unit 5 and 6, Probolinggo Regency, East Java Province. The chemical element content testing is done at PT. Sucofindo Surabaya as shown in Table 2. Visual fly ash can be seen in Fig. 1. Based on their chemical content shown in Table 2, fly ash Paiton is categorized in class F. This indicates by the amount of Al_2O_3 , SiO_2 and Fe_2O_3 content by 74.39%, greater than 70% (ASTM C 618-96). According to the results obtained, generally coal is grouped in anthracite or bituminous coal types. Anthracite or bituminous coal is a type of coal having high carbon content, low moisture content, and slow speed in combustion.

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 TABLE 2. Chemical content of fly ash Paiton power plant

No	Chemical composition	Mass (%)	No	Chemical composition	Mass (%)
1	SiO ₂	36.50	10	SrO	0.17
2	CaO	19.65	11	CuO	0.04
3	Fe ₂ O ₃	19.27	12	ZrO ₂	0.03
4	Al ₂ O ₃	19.00	13	ZnO	0.02
5	SO ₃	3.45	14	HgO	0.02
6	K ₂ O	0.90	15	V ₂ O ₅	0.02
7	TiO ₂	0.71	16	Rb ₂ O	0.01
8	BaO	0.31	17	Cr ₂ O ₃	0.01
9	MnO	0.20	Jumlah		100.00



Fig. 1. Fly ash PLTU Paiton units 5 and 6

(2) Alkali Activator

The alkaline activator to the fly ash geopolymer mixture is a mixture of Sodium Hydroxide (NaOH) and Sodium Silicate (Na₂SiO₃). More detail description of the alkali activator used can be seen as follows:

(a) Sodium Hidroksida (NaOH)

It is used Sodium Hydroxide in the form of flake with a purity rate of 98%. It is also used Sodium Hydroxide in the form of flake as a solution that is dissolved by aquades. The concentration of Sodium Hydroxide used in this study is 8M. Fig. 2 (a and b) shows the flake and Sodium Hydroxide (NaOH) solutions.

(b) Sodium Silicate (Na₂SiO₃)

It is used Sodium Silicate in the form of viscous liquid (gel) with ready-made state. The liquid of Sodium Silicate (Na₂SiO₃) is shown in Fig. 2 (c).



a. NaOH flake b. NaOH solution

c. Na₂SiO₃ solution

Fig. 2. Alkali activator Sodium Hydroxide and Sodium Silicate

C. Mixing, casting, curing and testing

(1) Preparation of NaOH solution

Preparation of NaOH solution is done with molarity of 8 mol / l (8M) solution. The formula used to make a NaOH solution is:

$$n = V M \dots \dots \dots (1)$$

which:

n = number of solute moles (mole)

M = molarity of solution (mol / liter)

V = volume of solution (liter)

$$\text{Mass NaOH} = n M_r \dots \dots \dots (2)$$

Which:

n = number of solute moles (mol)

M_r = relative atomic mass (gram / mol)

The amount of flake NaOH required to prepare an 8M NaOH solution is using the formula (1) as follows:

$$n = VM = 1 \text{ liter} \times 8 \text{ mol / liter} = 8 \text{ mol}$$

$M_r \text{ NaOH} = 40,$

Which: $\text{Na} = 23, \text{O} = 16, \text{H} = 1$

Mass NaOH = $n \text{ mol } M_r$

$$= 8 \text{ mol} \times 40 \text{ grams / mol}$$

$$= 320 \text{ grams}$$

So, to make 1 liter of 8M NaOH solution, it requires 320 grams of flake NaOH. The following weight of flake NaOH is 320 grams. The flake NaOH is put into a measuring cup with a capacity of 1000 ml. Then, it adds aquades to the measuring cup filled with NaOH until the volume is 1000 ml. Then it is stirred and waited until it is cool. Once it is cool, the solution is stored in a container and ready for use.

(2) Mixing and casting procedures

Before mixing the fly ash, firstly it is mixing Na_2SiO_3 and NaOH with comparison based on the design. It is necessary to determine the amount of mixture. Then, it is to weigh the fly ash as required in the mixture, namely a gram. If the ratio of fly ash with alkali is 75%: 25%, then the amount of alkali is 25%: (75% .a) gram. After all of the mixture ingredients are available according to the composition, then the mixing is done. In the mixing process, it is preferable to use mixer to create possible mixture homogeneous. After the mixture is evenly mixed, it is done the casting. Compaction at the time of casting should be perfect so that there are no large pores in the sample. After the mold is fully loaded then it is flattened, then it is clamped with the mold. It is a 20 mm-diameter and 40 mm-height cylinder mold. After setting, the sample can be removed from the mold.

For example, to make a geopolymer mixture of 200 grams, it requires 75% of fly ash and 25% of alkali activator. Where the ratio of Na_2SiO_3 with NaOH in alkali is 2, then it is prepared the materials as follow:

$$\text{Fly ash} = 75\% \times 200 \text{ grams} = 150 \text{ grams}$$

$$\text{Alkaline activator} = 25\% \times 200 \text{ grams} = 50 \text{ grams,}$$

then:

$$\text{NaOH} = 1/3 \times 50 \text{ grams} = 16,67 \text{ grams}$$

$$\text{Na}_2\text{SiO}_3 = 2/3 \times 50 \text{ grams} = 33,33 \text{ grams}$$

Mixing process:

Weigh 16,67 gram of NaOH solution and Na_2SiO_3 33,33 grams. Then mix the sodium hydroxide solution and the sodium silicate that has been weighed and stirred until fused. Then weigh fly ash 140 grams. Put the fly ash into the vessel and then pour the alkaline activator. Stir until smooth using a spoon and mixer. Pour the paste into the sample mold while damaging to avoid exposure to the pores in the paste sample. Flatten the surface then clamp. Afterwards, dry the sample mold. The geopolymer sample is stored in a container. Other samples are conducted in the same manner according to the mixture composition. Fig. 3 is the process of mixing fly ash geopolymer paste.

a. Treatment (Curing)

Samples removed from the mold are further wrapped with a damp cloth to keep it moist. This is important so there is no crack in the sample. The treatment is performed until the 28-day of tested sample.

b. Testing

Testing of pasta samples is conducted at 28 days. The sample surface of the paste cylinder should be ascertained in a flat state before testing. It is also important to pay attention to the test object flatness because it greatly influences on the value of compressive strength. Then, weigh the specimen to get its density value. To know the density value, it is the weight of the specimen divided by the cylindrical specimen volume. The compressive strength test may be performed after the specimen complies with the requirements.

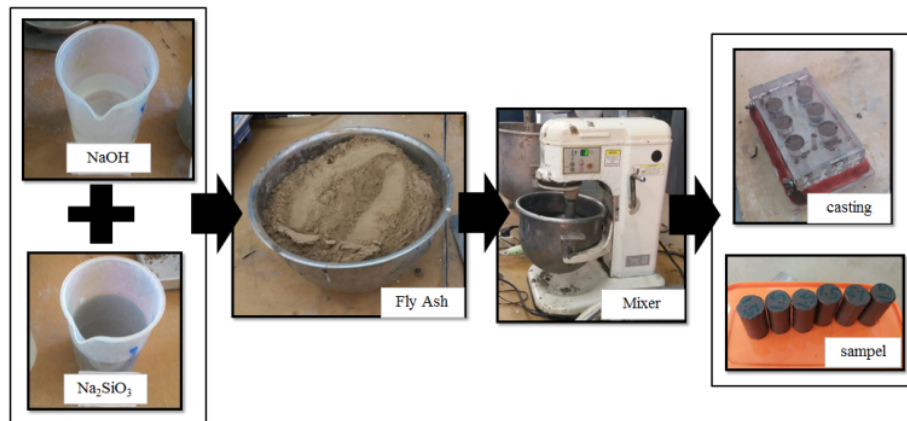


Fig.3. The process of mixing fly ash geopolymer paste

III. Results and Discussion

A. Result

The results of the compressive strength test as well as the complete density of the geopolymer paste samples in various compositions are shown in Table 3 below.

TABLE 3. Test results of fly ash geopolymer samples

Mixed Number	Code Sample	Weight (gr)	Density (gr/cm ³)	Compressive strength (MPa)	Mixed Number	Code Sample	Weight (gr)	Density (gr/cm ³)	Compressive strength (MPa)
1	1.5-1	29,50	2,35	36,35	10	2.0-4	29,80	2,37	25,91
2	1.5-2	29,40	2,34	23,27	11	2.0-5	28,90	2,30	30,01
3	1.5-3	30,10	2,39	53,33	12	2.0-6	29,40	2,34	78,99
4	1.5-4	30,00	2,39	49,83	13	2.5-1	28,80	2,29	19,72
5	1.5-5	29,30	2,33	78,07	14	2.5-2	28,80	2,29	27,73
6	1.5-6	29,10	2,31	47,71	15	2.5-3	28,80	2,29	40,66
7	2.0-1	30,20	2,40	48,77	16	2.5-4	29,00	2,31	27,07
8	2.0-2	31,60	2,51	36,50	17	2.5-5	29,30	2,33	30,47
9	2.0-3	29,50	2,35	23,93	18	2.5-6	28,60	2,28	37,06

Based on Table 3 above, the calculation of average density and compressive strength of the fly ash geopolymer paste is used to determine the inter-variablerelationship. The average density of fly ash geopolymer for the ratios of Na₂SiO₃ and NaOH by 1.5, 2.0 and 2.5 are 2.35, 2.38, and 2.30. While the average compressive strength of fly ash geopolymer paste for the ratios of Na₂SiO₃ and NaOH by 1.5, 2.0 and 2, 5 are 48.09, 40.68 and 30.45. Fig. 4 shows the compressive strength relationship with density. Fig. 5 shows the compressive strength relationship with Na₂SiO₃ with NaOH ratios.

B. Discussion

An artificial aggregate of fly ash geopolymer is made of fly ash based on geopolymer paste. Fly ash is a material having characteristics and as a source of aluminosilicate. The geopolymer characteristics of fly ash have been studied extensively in the last decade [7]. A number of fly ash geopolymer with alkali peractive utilization is done by various methods. The cost-effective method is by using low energy. One of the energy-saving methods used in the manufacture of fly ash geopolymer paste is cold bonding method[8]. The findings suggest that the fly ash is suitable for making geopolymers, and the resulting geopolymer indicates the mechanical properties and high durability[7]. This type of alkali activator plays an important role in the geopolymerization process and has a significant influence on the geopolymer mechanical strength. Currently, the most commonly used basic activation is the combination of Sodium Silicate (Na₂SiO₃) solution and Sodium Hydroxide (NaOH) solution with different Na₂SiO₃ and NaOH mass ratio [9].

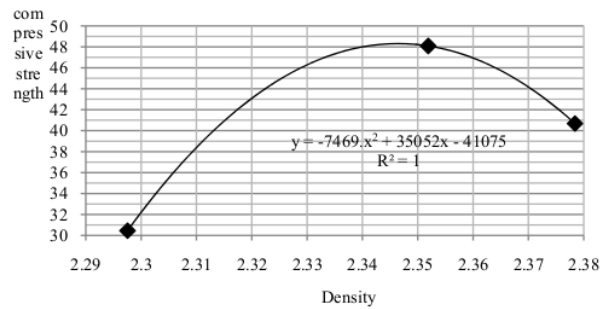


Fig. 4. Relation of compressive strength (y) with density (x)

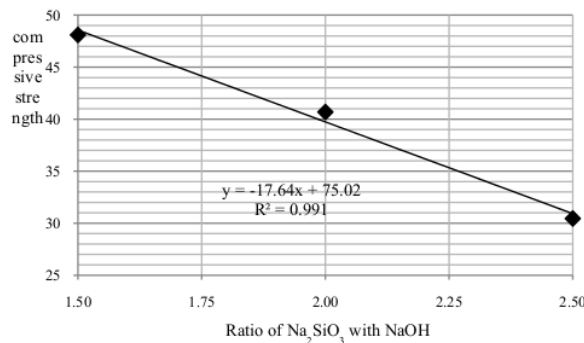


Fig. 5. The compressive strength (y) relationship with the alkali activator ratio of Na₂SiO₃ with NaOH (x)

Likewise in this study, in Fig. 5 it can be seen that the geopolymer compressive strength is influenced by the Na₂SiO₃ and NaOH ratios. Lower Na₂SiO₃ and NaOH ratios lead to higher paste compressive strength. The highest average compressive strength of 48.09 MPa can be obtained at ratio of Na₂SiO₃ and NaOH by 1.5. The lowest average compressive strength is 30.45 MPa in Na₂SiO₃ and NaOH ratio of 2.5. According to Shuaibu [10] in addition to the influence of Na₂SiO₃ and NaOH ratios, the optimal strength of geopolymers is also influenced by the molarity of NaOH, fly ash ratio and alkali activator and temperature preservation [9]. The statement is supported by a study which states that the increase in molarity of NaOH from 8M to 14M with the same 2.5 Na₂SiO₃ and NaOH ratios increases the compressive strength by 10 MPa, from 57 MPa to 67 MPa [10]. With the same molarity of 8 M NaOH and the variation in the ratio of Na₂SiO₃ to NaOH, 0.5, 1.0, 1.5, 2.0, and 2.5, Risdanareni et al. found that there is an increase of the paste compressive strength from a ratio of 0.5 to 2.0, then it is decreased by 2.5. However, there are different results when using molarity of 10M NaOH, where the compressive strength increase is based on the increase of Na₂SiO₃ to NaOH ratios. The highest compressive strength can be achieved in ratio of 2.5 by 30.97 MPa [11]. This means that the geopolymer compressive strength can be increased by modifying the mixture namely by changing Na₂SiO₃ to NaOH ratio as well as the molarity of NaOH. So that, natural aggregate compressive strength of 33.0 MPa [11] can be achieved by fly ash geopolymer paste. Even fly ash geopolymer pastes is greatly possibly to have higher compressive strength. The activator ratio and the concentration of NaOH solution have an important role to the compressive strength value of the geopolymer paste. Changes in compressive strength are generally linear with changes in NaOH concentration as well as Na₂SiO₃ and NaOH ratios [12]. The compressive strength of fly ash geopolymer can be increased by changing Na₂SiO₃ and NaOH ratios, resulting in compressive strength up to 103 MPa [13]. However, in the study by Tajunnisa et al. (2016), it was found that the compressive strength was not only influenced by the ratio of Na₂SiO₃ to NaOH, but also by the type of fly ash used. This research used the fly ash taken from Japan and Indonesia. The ratios of Na₂SiO₃ to NaOH used were 1.5, 2.0 and 2.5. For the geopolymer made of Indonesian fly ash, it is obtained the highest compressive strength at a ratio of 1.5, of 57.5 MPa. As for the geopolymer made of Japan fly ash, the highest is 47.7 MPa, at a ratio of 2.5. The geopolymer made of Indonesian fly ash has a higher compressive strength than the Japanese fly ash. This is because the chemical content of Indonesian fly ash is rich in CaO and Fe₂O₃. Fly ash with high CaO levels causes a reciprocal reaction of polymerization and hydration [14].

Several studies have been conducted by making the relationship between the compressive strength of the fly ash base-geopolymer paste with the artificial aggregate properties. There is a research of making artificial aggregate from fly ash geopolymer with compressive strength of 22,8 Mpa; it results in aggregate with water absorption value by <13,01%[15].Srinivasan et al.[16], found out that the quality of artificial aggregate is lower than the natural one. This research, it is used natural aggregate and the artificial one for concrete mixture. The results indicates that in the age of 28 days, compressive strength of concrete with natural aggregate is 25.86 N/mm² meanwhile, those with artificial aggregate is 18.07 N/mm² [15]. However, the research studying on the quality of artificial aggregate is continuously performed out.The aggregate properties are: crushing and impact values, abrasion and water absorption. For crushing values, artificial aggregates obtain a value by 31.8% that is lower than the natural aggregates, but it has 26.4% higher impact values. The abrasion value is too low so that it is necessary for upgrading to be used as a road material. Another result is the percentage of aggregate water absorption which is 9 times higher than the natural aggregate, but it can be eliminated by various repair methods such as by using sodium silicate and other materials. Other studies result in specific gravity aggregates of 1.7, bulk densities of 1050 kg / m³, water absorption of 18%, abrasion values <40% and aggregate resistance <12%[17].

TABLE 4.Properties of artificial aggregates and natural aggregates

No.	Aggregate Properties	Artificial aggregates					Natural aggregates
		Could bonded [15]	Sintered pellets[17]	Aardelite Aggregate[17]	Pelletizationprocess[18]	Sintered fly ash aggregate[19]	Crushed stone [19][17]
1	Compressive strength (MPa)	22.81	-	-	13.72	26,7	33.0
2	Water absorption (%)	<13.01	14%-18%	18%	-	15%-16%.	-
3	Specific Gravity	-	1.30-1.60	1.7	2,12	1,66	2.75-2.95
4	Shape	-	Round	Round	Round	-	Angular
5	Bulk Density (kg/m3)	-	750-900	1050	942.68	-	1450-1750
6	Particle size (mm)	-	5-15	-	-	-	5-40 and greater

Table 4 shows the comparative properties of artificial aggregates and natural aggregates. Previous research has shown that the artificial aggregate has lower compressive strength than the natural aggregate. Looking at other properties such as water absorption, specific gravity and bulk density, these also show lower values. These values are linear with the compressive strength value. The results of the study review obtain primarily by reviewing the compressive strength of cylinders, reinforcing the hypothesis that the artificial aggregates with fly ash geopolymer materials can potentially replace natural aggregates. The compressive strength is correlated with other aggregate properties, so that it can obtain aggregates with better properties than the natural aggregate. These results simultaneously show that the artificial aggregates of fly ash geopolymers qualify as asphalt mixtures. This reasoning is supported by higher compressive strength test results than the compressive strength test for aggregates in some studies. Furthermore, compressive strength the geopolymer may be further added by varying the mixture with modification of the ratios of Na₂SiO₃ and NaOH, NaOH molarity, fly ash ratio and alkaline activator and temperature preservation. This experiment can be improved by designing artificial aggregates. The quality of artificial aggregate of fly ash geopolymer is directly proportional to the compressive strength of the geopolymer paste used. There is a fact that the paste compressive strength can be increased, then it is possible to obtain a good quality of aggregate. Success will be enhanced by using a pelletization process as an effective way to produce artificial light aggregates [20]. The results show that the use of fly ash geopolymer is prospective for using an artificial aggregate material. Because, it can produce the fly ash geopolymer paste formula with high compressive strength. Since the artificial aggregates have properties such as natural aggregates, it can be used as a mixture of asphalt for roads and airports. Previous research has shown that the use of fly ash geopolymer as a filler on concrete asphalt mixture can lead to double increase of the stability of Marshall test [21].

The statement of Aodah, et al, supports the idea of successful use of artificial aggregates from fly ash. Which it is stated that the determinants of asphalt mixture performance are not only based on the properties, but rather by aggregate gradation. In this case, the parameter is the gradient ratio, which correlates with strength

parameters and mixed performance [22]. On the other hand, workability is one of the important parameters in designing asphalt mixtures [23]. This is more easily achieved with artificial aggregates, because the grain size can be planned to follow the desired gradation. Review of some papers by Vali and S, allows the use of lightweight aggregates as a modern structural element. The utilization of fly ash for artificial aggregates is a form of attention to the diminished natural aggregate resources. This success will reduce the use of natural aggregates, amid improvements in the infrastructure industry [24].

IV. CONCLUSION

The conclusions of this study based on Results and Discussion are as follows:

- 1) Fly ash-based geopolymer is potentially used as an artificial aggregate material for as a replacement aggregate in a concrete asphalt mixture.
- 2) The quality of artificial aggregate can be increased by increasing the compressive strength of the geopolymer paste.
- 3) The compressive strength of the fly ash base-geopolymer paste can be increased by modifying the alkali activator, ie the ratio of Na_2SiO_3 to NaOH, and the molarity of NaOH.
- 4) Higher compressive strength leads to better result of artificial aggregate properties, therefore, it is necessary for well plan of the fly ash based-geopolymer paste mixture.
- 5) The successful utilization of aggregate made from fly ash based-geopolymer, will reduce the use of natural aggregate, so that it can be considered as a form of attention to the depletion of natural resources.

V. ACKNOWLEDGEMENT

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