Civil Engineering and Architecture 11(1): 13-21, 2023 http://www.hrpub.org
DOI: 10.13189/cea.2023.110102

Manufacture of Geopolymer Artificial Aggregates by Pelletization and Crushing Processes

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Received August 22, 2022; Revised November 3, 2022; Accepted November 15, 2022

Cite This Paper in the Following Citation Styles

(a): *[1] Bimo Brata Adhitya, Anis Saggaff, Saloma, Hanafiah , "Manufacture of Geopolymer Artificial Aggregates by Pelletization and Crushing Processes," Civil Engineering and Architecture, Vol. 11, No. 1, pp. 13 - 21, 2023. DOI: 10.13189/cea.2023.110102.*

(b): *Bimo Brata Adhitya, Anis Saggaff, Saloma, Hanafiah (2023). Manufacture of Geopolymer Artificial Aggregates by Pelletization and Crushing Processes. Civil Engineering and Architecture, 11(1), 13 - 21. DOI: 10.13189/cea.2023.110102.*

Abstract Aggregates are an important ingredient of concrete. They are of two types: coarse aggregates and fine aggregates. The supply of natural aggregates on Earth is declining with technological advancement, hence, alternatives to natural aggregates are needed. Artificial aggregates have been manufactured using a coal burning waste, i.e. fly ash. On mixing fly ash with an alkaline activator, the mixture reacts and hardens. Aggregates are manufactured either by mixing materials using a granulator pan or by crushing materials using a stone crusher. The optimal manufacturing method was determined by comparing physical properties, such as bulk specific gravity, water absorption, and aggregate hardness, of the aggregates manufactured using these two methods with those of natural coarse aggregates. The average bulk specific gravity was 1.776 and 1.857 for the aggregates produced by the pelletization and crushing processes, respectively, and 2.957 for the natural aggregates. The average water absorption values were 11.62% and 8.37% for the aggregates produced by the pelletization and crushing processes, respectively, and 4.17% for the natural aggregates. The average aggregate hardness values, determined using the Los Angeles abrasion test, were 27.33% and 25.98% for aggregates produced by the pelletization and crushing processes, respectively, and 24.05% for the natural aggregates.

Keywords Artificial Aggregate, Fly Ash, Pelletization, Crushing, Geopolymer

1. Introduction

Aggregate in the form of gravel, crushed stone, and sand becomes the basic material in construction, such as concrete and asphalt, where the aggregate is mixed with a binder [1]. Aggregates can be classified into coarse aggregates (e.g. gravel) and fine aggregates (e.g. sand). Coarse aggregates are classified further into two types: natural aggregates and artificial aggregates. With the continuous development of technology, the stock of natural aggregates on Earth is declining, hence, an alternative is required to replace natural aggregates [2,3]. Geopolymer Artificial aggregates have been developed from waste materials such as fly ash from coal combustion [4]. Geopolymer is an eco-friendly alternative to replace Portland cement. They are synthesized by a chemical reaction (polymerization) between an aluminosilicate material (an industrial by-product, such as blast furnace slag or fly ash) and an alkaline activator [5]. Fly ash is considered a waste material formed from combustion [6]. Artificial aggregates can be formed by mixing fly ash with alkaline activator [6]. NaOH and Na_2SiO_3 have been used as alkaline activators of fly ash in mixtures with fly ash and alkaline activator ratio of 25:75 for making artificial aggregates [7-12]. The manufacture of artificial aggregates requires a further process, which includes determining the FA/AA mixture ratio, Na_2SiO_3/NaOH ratio, and NaOH concentration [14,15]. After determining the optimal ratio of the mixture, the manufacturing process proceeds with mixing the ingredients. There are two methods for making artificial aggregates: the pelletization method using a granulator pan and the crushing method using a crusher [16,17].

The manufacture of artificial aggregates using the pelletization method depends on various factors, including the slope of the granulator, rotational speed, stirring time, and water content, which are varied to obtain different artificial aggregates. The alkaline activator is sprayed during the mixing process in the granulator. Then, the curing process is carried out using various curing methods.

Shivaprasad and Das [18] used type F fly ash mixed with NaOH and Na_2SiO_3, with Na_2SiO_3/NaOH ratio of 2.5 and NaOH concentration of 10 M [19]. The aggregate was formed using a granulator pan of depth 100 mm and diameter 450 mm. The angle of the granulator was 45° [20-22], and the duration of rotation was 15 min, the speed was maintained at 10 radians per minute. The fly ash was mixed with the activator by rotating the fly ash on the granulator pan and gradually spraying the activator. The aggregates produced were uniformly round in shape, and the pellet size depended on the amount of activator used.

In their study, Hilda et al [23] used an FA/AA ratio of 25:75, and an Na_2SiO_3/NaOH ratio of 2.5:1 [24-26]. A granulator of diameter 120 mm and weight 20–50 kg was rotated at a speed of 26 rpm [27]. The size of the resulting aggregates ranged from 2.36 mm to 25 mm. Three slope angles were used 45°, 50°, and 55°. The best slope angle was determined to be 50° [28].

Using a granulator of depth 100 mm and diameter 450 mm at an angle of 45° for 15 min at a speed of 10 rpm [18]. They used three types of curing: ambient curing, heat curing, and solution curing. The highest compressive strength of 3.9 MPa was achieved by heat curing with water content of 21% and Na_2O content of 3.5%.

Artificial aggregates are produced using agglomeration techniques [14]. A granulator pan of diameter 500 mm and depth 95 mm was rotated at a speed of 60 rpm, and a slope angle of 48° was employed for 20 min. This method is carried out because it is proven to provide sufficient water absorption and strength as well as increase the durability of the mortar and concrete produced [31-33]. The curing method used was air drying at 20 °C.

Shivaprasad et al [34] reported the production of factory-made aggregates using a granulator of diameter 500 mm and depth 125 mm. The disk rotation speed was varied from 1 to 55 rpm, and the tilt angle was varied from 0 to 70°. The aggregate manufacturing process included the following steps: preparing the required material, placing uniform fly ash into the granulator, and rotating the disk while spraying an alkaline activator solution for 2–3 min. After the pelletization process, a heat curing method was applied for 24 h at a temperature of 80°C, then the aggregate was stored at room temperature for up to 28 days before further testing.

The manufacture of artificial aggregates using the crushing method comprised of several stages, including mixing, casting, crushing, and curing [17]. The first step in the manufacture of geopolymer aggregates was mixing. Fly ash (FA) and alkali activator (AA) were mixed after determining a suitable FA/AA ratio and alkaline activator ratio (Na_2SiO_3NaOH) [17,35,36]. These materials were mixed in a special mixer to ensure that they are mixed well.

A stone crusher was used to form artificial aggregates in. The hardness as determined by the Los Angeles abrasion test was 24.03% and the water absorption was 5.84%. These artificial aggregates had a composition of 75% fly ash and 25% alkali activator [19].

In the next step, the geopolymer mixture is molded into a cubic mold. After vibration and disassembled after 24 hours at laboratory temperature (25°C), all cubes are covered with plastic sheets [11,17]. The cubic specimen mixture is prepared to produce sufficient aggregates for the manufacture of concrete.

After demolding, the cubes were crushed using a stone crusher. Then the crushed cube fragments were collected and further crushed with a hammer until all the aggregate passed the 20 mm sieve, and the aggregate with a diameter of less than 5 mm was discarded [17,35].

The aggregates with diameters between 5 and 20 mm were collected and placed in an oven at 105 °C [17]. As shown in past examinations, premature heat preservation is very successful in lessening the shrinkage of geopolymer concrete, and higher temperatures will generally better diminish shrinkage issues. After 24 h, the aggregates were removed and stored in fixed plastic packs for another 26 days at 20 °C. The properties tested after 28 days included apparent density, bulk density, water absorption, and compressive strength.

2. Materials and Methods

2.1. Fly Ash

The fly ash used in this study was obtained from PT. Pupuk Sriwidjaja Palembang and analyzed using XRF (X-Ray Fluorescence), XRD (X-Ray Diffraction), and SEM (scanning Electron Microscopes) [6,11,36,37]. The chemical composition of the fly ash is given in Table 1, and the results of SEM and XRD are shown in Figure 1 and 2, respectively.

Most fly ash granules are spherical shown in Fig. 1, which is a SEM photo of fly ash at 10,000× magnification. The spherical shape of fly ash allows it to react rapidly with other particles when making geopolymer mixtures.

The fly ash can be classified as type C according to the ASTM 618 standard [37], because it contains 50% $\leq SiO_2 + Al_2O_3 + Fe_2O_3 \leq$ 70%.

Table 1. Chemical composition of the fly ash used in this study

Components	% in mass
MgO	0.2866
Al_2O_3	18.6427
SiO_2	36.3552
P_2O_5	0.2055
SO_3	0.2849
K_2O	0.7973
CaO	2.9143
TiO_2	0.9807
MnO	0.0932
Fe_2O_3	5.7413
CuO	0.0116
ZnO	0.0246
SrO	0.0931
Y_2O_3	0.0095
ZrO_2	0.0555
Balance	33.5039

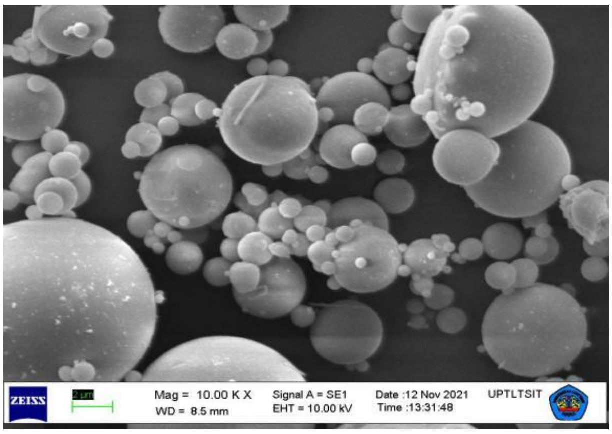

Figure 1. SEM image of the fly ash used in this study

The diffractogram obtained by conducting XRD analysis of the fly ash is shown in Fig. 2. It is observed that the crystalline peak of fly ash is very small, i.e., it occurs at a diffraction angle of 26.7° with an intensity of 1433.333 cps. This shows that the fly ash used in this study has an amorphous structure.

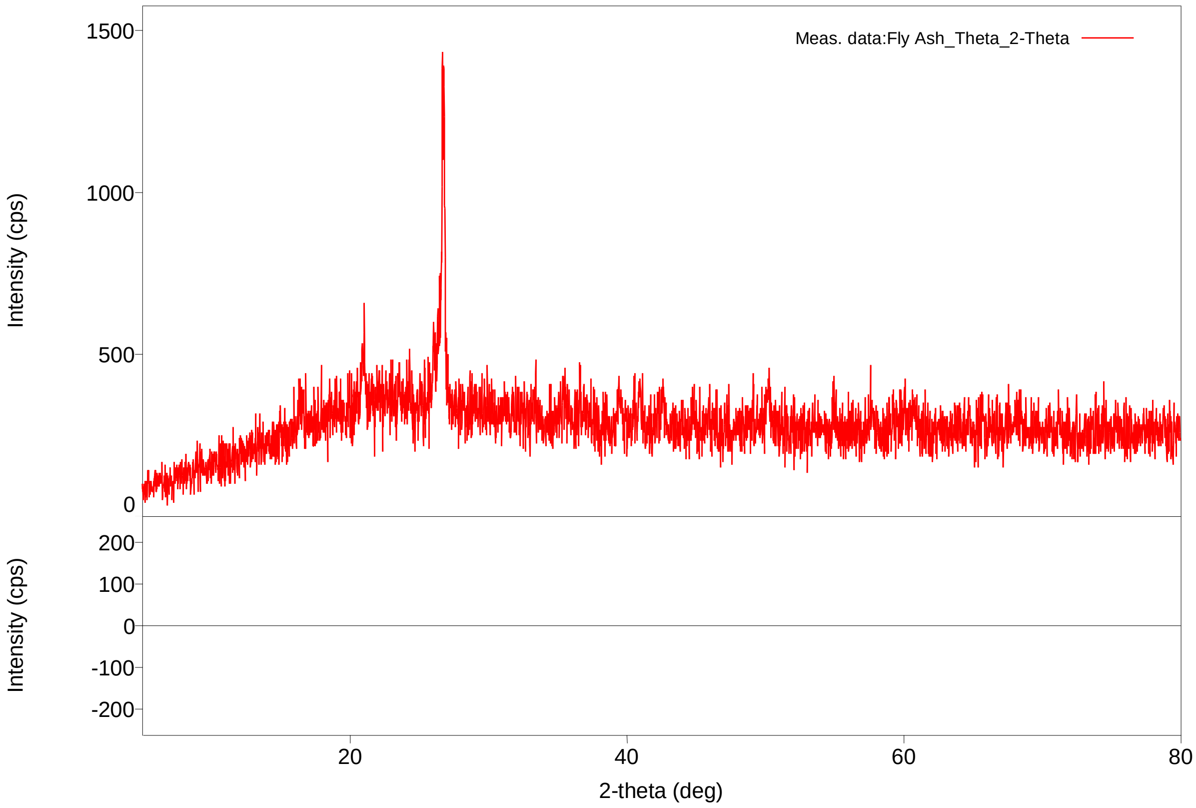

Figure 2. XRD of the fly ash used in this study

2.2. Alkali Activator

Sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3) were used as alkali activators [38-40]. Activators are substances or elements that cause other elements to react [5,41]. The activator used contains NaOH and silica, which is a strong acid and hence will react with a strong base. Na_2SiO_3 has the function of accelerating the polymerization reaction. Meanwhile, NaOH reacts with the Si and Al in the fly ash to produce strong polymer bonds. To prepare the alkaline activator [42], an NaOH solution was mixed with a Na_2SiO_3 solution, and the mixture as allowed to stand for 1 day to achieve equilibrium before being used. In this study, the FA/AA ratio was 3:1, the Na_2SiO_3/NaOH ratio was 1.5:1 and the concentration of NaOH was 16 M [41].

3. Methods

The process of manufacturing artificial aggregates first involves making the alkaline activator solution and then inserting the fly ash into the granulator pan. For the pelletization process, a granulator pan of diameter 60 cm and depth 20 cm was used, with a tilt angle of 55° and a speed of 30 rpm. The pan was rotated while the alkaline activator was allowed to drip for 15 min. The granulator pan is shown in Figure 3. Artificial aggregates were also manufactured with a stone crusher using the same mixture. The material was mixed and then placed into a mold, after it hardened, it was placed into the stone crusher (Figure 4).

Various curing methods are used in the aggregate manufacturing process, including curing at room temperature (20-25 °C) [43, 44], heating in an oven (80-100 °C), and the sintering method, where the temperature exceeds 1000 °C [45,46]. In this study, the curing process of heating in an oven at 80 °C was used for 24 h, after which the materials were allowed to cool at room temperature.

Figure 3. Granulator pan used in this study

Figure 4. Stone crusher used in this study

4. Results and Discussion

Discussion of the results of this study is as follows: visual assessment of artificial aggregates and results from laboratory tests of aggregate physical properties.

4.1. Visual Assessment of Artificial Aggregates

The shape of artificial aggregates produced by pelletization was generally round because they were rotated using a granulator pan. The aggregate texture was coarsened not slippery. The size of the resulting aggregates ranged from 2.36 mm to 12.5 mm, on average, most aggregates were produced with a size of 6.3 mm−9.5 mm. The size and shape of the artificial and natural aggregates are shown in Fig. 5. The size of crushed aggregates was easier to adjust compared to that of pelletized aggregates and not slippery. The size of the resulting aggregates ranged from 2.36 mm to 12.5 mm, on average, most aggregates were produced with a size of 6.3 mm−9.5 mm. The size and shape of the artificial and natural aggregates are shown in Fig. 5. The size of crushed aggregates was easier to adjust compared to that of pelletized aggregates.

4.2.1. Bulk Specific Gravity

Specific gravity testing was carried out according to [48]. The normal bulk specific gravity of natural aggregates is 2.4−2.9 [13]. The bulk specific gravity of artificial coarse aggregates produced by the crushing and pelletization methods in this study were far less than that of natural coarse aggregates. The bulk specific gravity value of each aggregate can be seen in Table 2. The average specific gravity was determined to be 1.776 for aggregates produced by the pelletizing process, 1.857 for aggregates produced by the crushing process, and 2.957 for natural aggregates.

4.2.2. Water Absorption

Aggregates generally have the ability to absorb water, although high water absorption is usually avoided. The absorption value is determined as the ratio of the weight of water absorbed by the pores of the aggregate to the weight in dry condition. Water absorption testing was carried out according to ASTM C 127 [47].

4.2. Physical Properties of Artificial Aggregates

Bulk specific gravity, water absorption, and aggregate hardness were analyzed in this study. The aggregates analyzed were the geopolymer aggregates produced by the pelletization method, geopolymer aggregates produced by the crushing method, and natural coarse aggregates from Merak.

The water absorption value of each aggregate can be seen in Table 3. The average water absorption was determined to be 11.62% for aggregates produced by the pelletization process, 8.37% for aggregates produced by the crushing process, and 4.17% for natural aggregates. The artificial aggregates produced by the pelletization and crushing methods have higher absorption values than the natural aggregates used in this test. A lower percentage of water absorption means that the aggregate has a higher structural density [17].

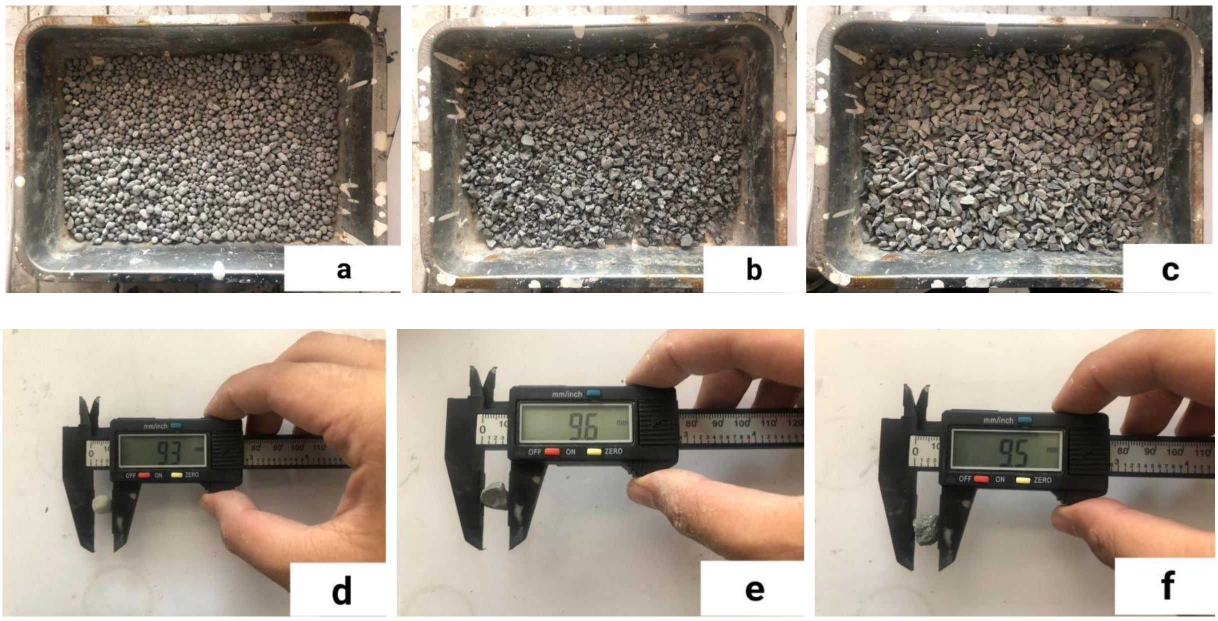

Figure 5. a) Shape of pelletized aggregates, b)Shape of crushed aggregates, c) Shape of natural aggregates, d) Size of pelletized aggregates, e) Size of crushed aggregates, and f) Size of natural aggregates

Table 2. Bulk Specific Gravity values for different types of aggregates

Sample	Bulk Specific Gravity		
	Pelletization	**Crusher**	**Natural**
Sample 1	1.651	1.852	2.850
Sample 2	1.879	1.830	3.057
Sample 3	1.738	1.907	3.048
Sample 4	1.850	1.786	2.927
Sample 5	1.760	1.912	2.896
Average	1.776	1.857	2.957
Standard Deviation	**0.091**	**0.053**	**0.093**

Table 3. Water absorption values for different types of aggregates.

Sample	Water Absorption (%)		
	Pelletization	Crusher	Natural
Sample 1	11.85	8.22	4.30
Sample 2	10.98	8.75	4.12
Sample 3	11.47	8.47	3.92
Sample 4	11.66	8.09	4.28
Sample 5	12.14	8.32	4.25
Average	11.62	8.37	4.17
Standard Deviation	**0.43**	**0.25**	**0.16**

Table 4. Los Angeles test results.

Sample	Los Angeles test results (%)		
	Pelletization	Crusher	Natural
Sample 1	27,86	25,69	25,68
Sample 2	26,71	26,71	22,16
Sample 3	27,40	25,54	24,26
Sample 4	26,88	26,46	24,85
Sample 5	27,82	25,51	23,32
Average	27,33	25,98	24,05
Standard Deviation	**0,53**	**0,56**	**1,36**

Figure 6 presents data on the specific gravity and water absorption parameters of the aggregates. Based on Fig 6. it shows that the higher the specific gravity value of the aggregate used, the lower the water absorption value.

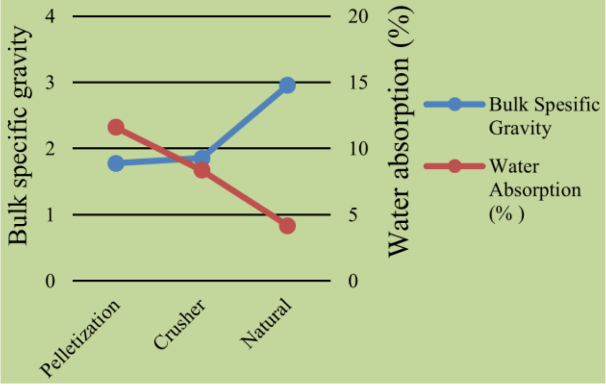

Figure 6. Bulk specific gravity and water absorption values

4.2.3. Aggregate Hardness

Artificial aggregates must have almost the same hardness as natural aggregates for use as replacements. Aggregate hardness tests were carried out using the Los Angeles test with standard procedure ASTM C 131 [48], where an aggregate hardness of < 30% is generally considered appropriate. The test data are given in Table 4.

The aggregate hardness value of each aggregate can be seen in Table 4. The aggregate hardness was determined using the Los Angeles tool type B gradation, resulting in average values of 27.33% for pelleted aggregates, 25.98% for crushed aggregates, and 24.05% for natural aggregates. All three types of coarse aggregates can be considered good aggregates as their hardness values are < 30%.

Figure 7 presents data on specific gravity and aggregate hardness parameters tested with the Los Angeles tool. Figure 7 shows that the higher bulk density value, the lower aggregate hardness value. The value of aggregate hardness indicates that the level of wear of the aggregate, the higher the wear value, the lower the ability of the aggregate to withstand friction.

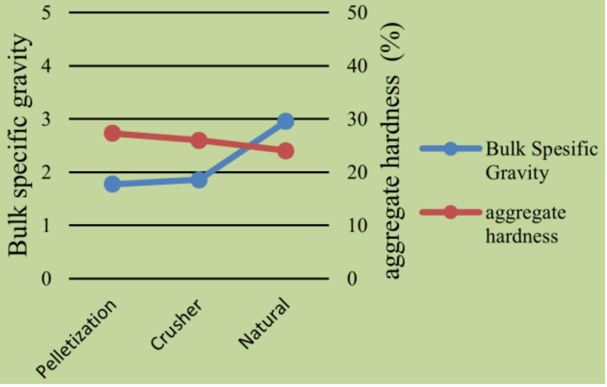

Figure 7. Bulk specific gravity and aggregate hardness values

4.3. Discussion

4.3.1. Artificial Aggregates For concrete

Artificial aggregate is an innovation of sustainable materials that can be used as an alternative to natural aggregates. Natural aggregates are replaced with artificial aggregates with concrete, which seems to be a fairly useful technique, as the artificial aggregates have nearly the same characteristics as natural aggregates. The use of artificial aggregates can also reduce waste material. The use of artificial aggregates for the manufacture of concrete was reported in [49].

5. Conclusions

The results of data analysis show that the aggregates produced in the present study have good characteristics, except for the water absorption properties. The artificial aggregates produced are quite suitable for use as a mixture in concrete. The technical details are as follows:

1. The size of the artificial aggregates produced by pelletization is from 2.36 mm to 12.5 mm. In contrast, the size of the aggregates produced using the stone crusher was easier to adjust.
2. The average specific gravity was determined to be 1.756 for aggregates produced by the pelletizing process, 1.863 for aggregates produced by the crushing process, and 2.985 for the natural aggregates.
3. Water absorption was > 3%, however, a high water absorption rate is acceptable if the aggregates pass the criteria of durability and resistance to weathering. The average water absorption values are determined to be 11.55% for aggregates produced by the pelletization process, 8.31% for aggregates produced by the crushing process, and 4.17% for natural aggregates.
4. The average hardness test values indicate that the produced aggregates were good, these values are 27.14% for aggregates produced by the pelletization process, 25.62% for aggregates produced by the stone crusher, and 24.03% for natural aggregates.
5. Geopolymer Artificial aggregates can be used as an alternative to natural aggregates in materials such as concrete.

Acknowledgments

Thanks to PT. Pupuk Sriwidjaja for providing fly ash and thanks to the University of Sriwijaya, where the testing and manufacture of the artificial aggregates was carried out.

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