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Optimization of Ground Sediment and Calcium Carbide Residue as Raw Materials for Geopolymer

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Abstract

This paper aims to study the synthesized geopolymers by using Ground Sediment (GS) and Calcium Carbide Residue (CCR) as aluminosilicate sources. The efficiency of the source materials in producing geopolymer products was optimized by using Taguchi method via five parameters including Sodium Hydroxide (NaOH) concentration, Solid to Liquid ratio, Sodium Silicate to Sodium Hydroxide (Na_2SiO_3/NaOH) ratio, oven curing time and oven curing temperature. Each of the parameter was tested at four levels with L16 array. The performance of the specimens was assessed by density test and compression test. Taguchi analysis of the experimental results suggested a specimen with NaOH concentration of 8M, Solid to Liquid Ratio equals to 1, Na_2SiO_3/NaOH of 2, oven curing time for 36 hours and oven curing temperature at 60 °C to acquire the highest strength after 14 days of curing at room temperature.

1. INTRODUCTION

Cement is broadly utilized in construction development all over the world in spite of the fact that it requires a high amount of energy and produces a major quantity of greenhouse gases. The production of Ordinary Portland Cement (OPC) releases approximately 7% of the total greenhouse gases and mainly consists of carbon dioxide (CO_2) [1]. On top of that, it was reported that concerning one ton of CO_2 is formed for each one ton of OPC produced [2]. A research reported that in 2013, Australia cement production contributes to the emission of 36 billion tonnes of CO_2 in a year [3].

A huge amount of energy is needed when producing one ton of cement which is equivalent to about pounds of coal. It is estimated around 4.7 billion metric tonnes of cement will be utilized in 2020 considering an annual growth of 4% [3]. Therefore, researchers have started to investigate geopolymer concrete and alkali-activated construction materials such as geopolymer concrete, geopolymer mortars or geopolymer bricks for the last few years as alternatives options to replace OPC. According to M. N. S. Hadi et. al [3], since geopolymer does not accommodate any OPC, it is thought of as a green concrete and proven for its excellent mechanical properties, low shrinkage, fire resistance, and low energy consumption.

Geopolymer or also well known as an alkali-activated aluminosilicate material formed by the polycondensation of an individual $[SiO4]^{4-}$ And $[AlO4]^{5-}$ tetrahedral [4]. Geopolymerization is a reassuring advance technology supported by the principle which is a reaction between amorphous silica and alumina rich solids with a high alkaline solution to form amorphous to semi-crystalline aluminosilicate inorganic polymers [4].

In order to promote alternative binders by utilizing an abundance of alumina-silicate (pozzolanic) wastes from the industrial sector, several studies on different aluminosilicate materials have been done by researchers from all around the world. For example, Chindaprasirt et al. [4] has studied on fly ash based geopolymer, Hardjito and Fung [5] studied on geopolymer utilizing bottom ash, meanwhile, Nuruddin et al. [6] in 2011 conducted a study on geopolymer using Silica fume and rice husk ash and N.S Hadi et. al. [3] reported on ground granulated blast furnace slag (GGBFS) [3]. There are also some researches on CCR [2], [5], [7] and sediments [6]–[9] as the source materials for geopolymer. Most of the research studies have also revealed that alumina-silicate materials can be used as prime materials to synthesize a cementitious binder by activating with an alkaline solution. The main intention of this research is to study the synthesis of a geopolymer utilizing GS and CCR in terms of compressive strength and its density.

As mention before, the main factors affecting the geopolymer properties rely upon the use of the aluminosilicate sources. From previous research done, the most significant factor in producing geopolymer are aluminosilicate source type and size, oven curing temperature, oven curing time, alkali activator type, concentration and alkali activator to aluminosilicate weight ratio [4]. One may consider the factors affecting the properties of geopolymer by using a suitable design method.

In this research, the main materials used are GS and CCR together with NaOH and Na_2SiO_3 as alkali activator. GS was used as the primary material since it is considered as abundant material. The high amount of Calcium Hydroxide ($Ca(OH)_2$) in CCR promotes the material as a sustainable cementing material. Taguchi method was used to develop the design of experiments. A total of 16 experiments were conducted according to L16 array proposed by the Taguchi method. The efficiency of the geopolymer was determined by investing the compressive strength to obtain the optimum mixture. Analysis on density was also conducted.

2. EXPERIMENTAL PROGRAM

A. Materials

One of the primary materials used in this research is GS which was taken from Cameron Highland reservoir sediment landfill area at Sungai Jasik. The GS were sieved passing 75 μm sieved. The chemical composition of GS proved that it can be used as the primary material for geopolymer synthesis due to its similarity to aluminosilicate materials. The density of the GS is 2500 kg/m^3.

The second material used in this project is CCR which was collected from Magnalium Sdn. Bhd., Perak, Malaysia. CCR was sieved passing sieve at 75 μm. The density of CCR is 2300 kg/m$^{3.}$

Na_2SiO_3 or also known as waterglass or liquid glass was used as alkali activator for geopolymerization. The chemical composition of Na_2SiO_3 consists of 35.40% Silicon dioxide (SiO_2), 17.93% of Sodium oxide (Na_2O) and 46.67% of water.

NaOH was in pearl form estimated 2 to 3 mm size with 98%. The alkaline solution was prepared by dissolving the pearl with distilled water. The molecular weight of NaOH is 40g. In order to ensure that the NaOH is fully dissolved before proceeding with experimental works, the solutions were prepared and left overnight.

B. Sample Preparation and Testing

L16 array was developed by Taguchi method. Parameters used in the design are NaOH concentration, solid to liquid ratio, Na_2SiO_3/NaOH Ratio, curing time and curing temperature. Each parameter was conducted at four levels. The design of experiments is shown in Table 1.

Firstly, NaOH pallets and Na_2SiO_3 solution were used as alkali activator in the mixture. NaOH was diluted for at least 24 hours prior to the experiment by distilled water to the designed concentration. NaOH pellets were dissolved in the distilled water and stirred continuously until the solution become clear which indicates that the NaOH has fully dissolved. The solution was then transferred to the volumetric flask and left for 24 hours at room temperature until the excess heat had completely vanished in order to avoid speed-up the geopolymeric specimens setting. After a day, the NaOH solution was poured into a beaker together with Na_2SiO_3 solution into the beaker and mixed for approximately 2 minutes. As soon as both solutions were mixed and stirred gradually, a reaction known as an exothermic reaction occur and an extreme amount of heat is evolved.

The mixing procedure starts with mixing the GS and CCR using a stirrer. Later, the alkali activator was poured into the mixture and mix for a few minutes until a homogenous solution is obtained. The mixture was cast in the 50mm $\times$ 50 mm $\times$ 50 mm steel moulds. The moulds were half-filled and then compacted for a few minutes to avoid any voids. The paste was filled up to the top level of the mould and finish the top surface by trowel o the last layer. The samples were placed into the oven for assigned temperature and curing time. After completing the elevated curing, the geopolymer specimens were evacuated from the oven and demoulded after 24 hours. The specimen was placed at room temperature for 14 days until testing ages.

The density of samples was calculated by measuring the weight and applied the $rho = \frac{m}{V}$ formula. The compression test was conducted in accordance with ASTM C39 standard. The compressive strength of the cube was calculated by using formula $\sigma = \frac{F}{A}$. where σ = the compressive strength, N/mm^2, F = the maximum load, N and A = the cross-section area in which load is applied, mm^2.

3. RESULTS AND DISCUSSION

A. Density Result

Table 2 shows the density for each sample. Generally, the density for geopolymer utilizing GS and CCR ranged between 1235 kg/m^3 to 1616 kg/m^3. Therefore, the geopolymer has a tendency to become a light-weight material. The density of the samples are lesser compared to OPC paste which density is more than 1800 kg/m^3 [1]. Besides, the density recorded was also slightly lower if compared to fly ash-based geopolymers that ranged between 1500 kg/m^3 to 1800 kg/m^3[1].

Figure 1 and Table 3 illustrate the significance of the optimum level of each factor by using Taguchi Analysis. Although the optimum condition of mixture would produce the lowest density, compressive strength of the sample is the essential mechanical properties to be considered.

B. Compressive Strength

The compressive strength for all mixes was illustrated in Figure 2. G11 produced the highest compressive strength which is 5.29 MPa with NaOH concentration of 10M and solid to liquid ratio of 1 which was oven cured for 36 hours at 80°C and then cured at room temperature for 14 days. It was proved that geopolymer production produced a high strength when the oven curing temperature was increased due to the increasing in geopolymerization reactions in higher temperatures [4].

Table 1. Design of experiments by Taguchi method (L16 array)

Mix	NaOH concentration (M)	Solid to liquid ratio	Na_2SiO_3/NaOH ratio	Oven curing time (h)	Oven curing temperature (°C)
G1	5	0.3	0	24	24
G2	5	0.8	0.16	36	50
G3	5	1	2	48	60
G4	5	1.25	3	72	80
G5	8	0.3	0.16	48	80
G6	8	0.8	0	72	60
G7	8	1	3	24	50
G8	8	1.25	2	36	24
G9	10	0.3	2	72	50
G10	10	0.8	3	48	24
G11	10	1	0	36	80
G12	10	1.25	0.16	24	60
G13	12	0.3	3	36	60
G14	12	0.8	2	24	80
G15	12	1	0.16	72	24
G16	12	1.25	0	48	50

Table 2. Density for each mix design

Mix	**Average density (kg/m^3)**
G2	1325.33
G3	1274.67
G4	1234.67
G6	1562.67
G8	1378.67
G10	1344.00
G11	1568.00
G12	1608.00
G13	1509.33
G14	1301.33
G15	1616.00
G16	1456.00

Figure 1. The significance of the main effects of the considered parameters on the density

Meanwhile, there are four specimens, G1, G5, G7 and G9 where no data recorded due to its low strength and undetectable by the machine. The four samples show a common physical appearance, where a layer of crystalline white substances was found on the surface of the geopolymer samples which is known as 'efflorescence'. Efflorescence can be described as a fine, white and powdery deposit left on the surface of the geopolymer which occurs spontaneously and involved the relocation of soluble alkalis [6]. Technically, the alkalis diffused on top of the geopolymer through the pores and react with CO_2 right after being dissolved by water. This process forms white carbonate products on the geopolymer surface. [6]. One of the factors to durability problems for alkali-activated metakaolin geopolymeric materials is efflorescence [8] and according to S. Hanjitsuwan et. al [9] research on Fly ash mortar with CCR, the presence of efflorescence demolished the mortars as it leads to low strength which in line with the current compressive strength results for GS and CCR based geopolymer.

Although G11 shows the highest strength, G8 with the second-highest strength recommends other parameters may also contribute to strengthening mechanism such as Na_2SiO_3/NaOH ratio. The similarities between G11 and G8 are the curing time which is 36 hours. Other parameters such as NaOH concentration, solid to liquid ratio, Na_2SiO_3/NaOH and curing temperature are different.

Most of the failed specimens have the lowest solid to liquid ratio of 0.3 proving that the ratio is inefficient in order to achieve a high strengths geopolymer. No failed specimens were found for specimens with NaOH concentration of 12M. This is in line with research done by Ali Nazari et. al [10] on utilizing fly ash and rice husk-bark ash, which stated that the highest strength of geopolymer specimens was achieved when using 12M of NaOH concentration. However, Chindaprasirt et al. [4] reported that 10-20M of NaOH concentration has a small effect on enhancing the strength of the specimens. To conclude, NaOH concentration depends on the aluminosilicate source for optimum strength. In this study, NaOH concentration of 10M shows the highest average strength although one of the specimens with concentration 10M has no data due to unsuitable solid to liquid ratio.

Table 4 and Figure 3 shows the optimum level for each parameter on the compressive strength obtained by Taguchi Analysis. Based on Table 4, the most influential parameter

Table 3. Parameters ranking and optimum level based on Taguchi analysis for density

Level	NaOH concentration (M)	Solid to liquid ratio	Na_2SiO_3/NaOH ratio	Oven curing time (h)	Oven curing temperature (°C)
1	1278	1509	1529	1455	1446
2	1471	1383	1516	1445	1391
3	1507	1486	1318	1358	1489
4	1471	1419	1363	1471	1368
Delta	228	126	211	113	121
Rank	1	3	2	5	4
Optimum Level	5	0.8	2.00	48	80

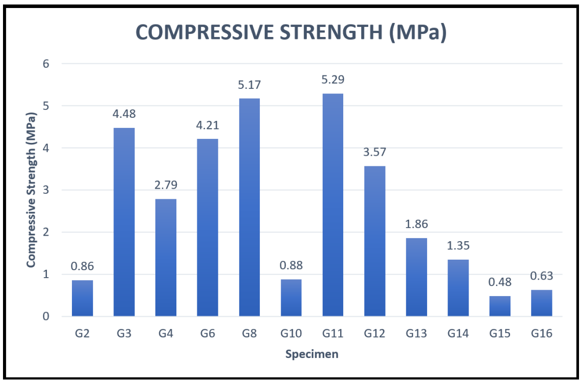

Figure 2. Compressive strength of the geopolymer specimens

is NaOH Concentration with delta 3.61. From Table 4, the optimum NaOH concentration is 8M. Ali Nazari et. al [10] reported that NaOH concentration more than 10M shows a negative impact on strength towards the geopolymer. Moreover, increasing the NaOH concentration more than 10M results in a reduction in the strength of the paste. When the additional concentration of NaOH could not be utilized by the primary materials, the strength will decrease because of incomplete geopolymerization reaction [7]. Chindaprasirt et. al [4] have proved that the NaOH concentration between 10 M and 20 M produces an inefficient effect on the strength. Besides, Somna et al.[10] indicated that NaOH concentration range between 12 M to 14 M create new crystalline sodium aluminosilicate products and the increase in concentration will only decrease the geopolymer paste strength due to early precipitation of aluminosilicate. Therefore, geopolymer with GS and CCR based with NaOH concentration of 8M has the most effective effect on increasing the strength and also the increase of NaOH concentration higher than the optimum level will only reduce the strength of the geopolymer.

Oven curing temperature was ranked as the second important parameter. The results show the effects of oven curing temperature on compressive strength of geopolymers is optimum at 60°C and compressive strength decreased when the oven curing temperature is at 80°C. This is in line with research conducted by Ali Nazari et. al [10] that increasing the curing temperature higher than 75°C for ash-based geopolymer proved to decrease the strength of the geopolymer. However, there is also a research stated that geopolymer specimens show a complete geopolymerization at 70°C and 90°C but the strength was incomparable with the geopolymer that was cured at 25°C and it is closely related with NaOH concentration [10]. To conclude, GS and CCR based geopolymer are able to produce high strength at an optimum oven curing temperature of 60°C with 8M of NaOH concentration.

The third in rank is Na_2SiO_3/NaOH ratio with optimum level equals to 2.00. M. J. A. Mijarsh et. Al [11] stated that the compressive strength for fuel ash-based geopolymer increase as Na_2SiO_3/NaOH ratio increases up to 2.5. The reaction rate increase as the ratio increase due to higher $[SiO_4]^{4-}$ concentrations [6]. During experimental work, the workability of geopolymer paste with Na_2SiO_3/NaOH ratio of 3.0 started to decrease because of the higher viscosity. The rate of influence parameters was evaluated by determining the formation of C-S-H and geopolymer (N-A-S-H) gels together with an increase in the amorphous silica content [6]. The polymerization process was enhanced by the addition of Na_2SiO_3/NaOH into the activating solutions which results to a higher compressive strength. However, decreased in silica concentration also leads to a less-polymerized distribution which eventually reduced the compressive strength [6].

The optimum solid to liquid ratio (S/L) in this study is 1.00. S/L more than 1.00 might decrease the compressive strength due to the increase in water content which may dilute the reaction [11]. In the previous study conducted by Kong et al. [12], it was found that solid to liquid ratio more than 0.8 produces low workability and deteriorates the properties of the paste produced for metakaolin-based geopolymer. C. Heah et. al [12] study on Kaolin geopolymers stated that S/L ratio of 1.00 is an applicable ratio in the matter of its strength and workability which is in-line with this current research.

4. CONCLUSION

To conclude, it is possible to produce a light-weight geopolymer materials the with the ranged between 1235 kg/m^3 to 1616 kg/m^3. Based on Taguchi Analysis, NaOH concentration of 5M with S/L ratio of 0.8 and Na_2SiO_3/NaOH of 2.00 at 80°C oven curing temperature for 48 hours produced the lowest density. As the density of the geopolymer specimens increases, the compressive strength increased.

Table 4. Parameters ranking and optimum level based on Taguchi Analysis for compressive strength

Level	NaOH concentration (M)	Solid to liquid ratio	Na_2SiO_3/NaOH ratio	Oven curing time (h)	Oven curing temperature (°C)
1	2.71	1.86	3.3767	246	2.1767
2	4.69	1.825	1.6367	3.295	0.745
3	3.2467	3.4167	3.6667	1.9967	353
4	1.08	3.04	1.8433	2.4933	3.1433
Delta	3.61	1.5917	2.03	1.2983	2.785
Rank	1	4	3	5	2
Optimum level	8	10	2.00	36	60

Figure 3. The significance of the main effects of the considered parameters on the compressive strength

Form this study, the optimum level of NaOH concentration was 8M to produce geopolymers with the highest strength. Solid to liquid ratio equals 1.00 and Na_2SiO_3/NaOH ratio of 2 at 60°C of oven curing temperature for 36 hours is the optimum level for geopolymer to reach its desire strength. Based on the optimum condition for both density and compressive strength, it was believed that GS and CCR based geopolymers are able to produce a low density geopolymer materials with high compressive strength. However, further research works need to be carried out to understand the mechanism of the geopolymer.

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