Review: Potential Strength of Fly Ash-Based Geopolymer Paste with Substitution of Local Waste Materials with High-Temperature Effect

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Abstract. This research provided an overview of the potential fly ash based geopolymer paste for application in building construction. Geopolymer paste with various variations of fly ash substitution with local waste material and high-temperature influence exploited with the fresh and hardened condition. The local waste material which utilized for this study were sandblasting waste, carbide waste, shell powder, bagasse ash, rice husk and bottom ash. The findings of this study indicated that fly-based geopolymer paste with local waste material substitution which had high-temperature influence ash showed a similar nature of OPC binders potentially used in civil engineering applications.

Keywords: Geopolymer Paste, Fly Ash, Local Waste Material, High Temperature

1. Introduction

The growth of technology and current economic activity leads us to live in complex human activities and affect the growing demand for infrastructure business. Infrastructure and construction materials have a close connection; a component of building materials that become the primary requirement was cement. The demand for cement would continue to increase and was predicted to experience an imbalance compared to its production numbers until 2030, and this would be a global problem. On the other hand, the researchers' awareness towards this situation had been growing to comprehend and conceive more about the environmental sustainability system. The facts showed that cement industry had contributed to the accumulation of carbon dioxide (CO2) in the air close to 8% of other industrial waste contribution. Thus, it is suggested to conduct research in the field of building materials and construction focused on designing alternatives to cement usage. Therefore, this is the right time to promote alternative concrete with 100% non-cement hydration content, which is geopolymer concrete. Also, one of the advantages of geopolymer concrete is that it has an excellent resistance both in acid and salt environment [1] and shows a strength escalation as the curing time and temperature increase. Another factor that determines the strength of this concrete is the fineness of the material since it enhances the activation while the polymerization process held. But those aren't everything, some other

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parameters, such as the molarity of alkaline, the ratio of alkaline activator composers, the addition of superplasticizer, and other stuff, play major rule for geopolymer concrete's performances. The mechanism of the geopolymer binder was the synergy of the alumina-silicate material and the activator material. One of the most widely used activators of sodium silicate Na₂SiO₃ that served to speed up the polymerization reaction. Also, sodium hydroxide (NaOH) is used in alkaline solution to help bonding process between particles [2]. After those processes, usually, researchers cure the sample to harden the samples [3]. Fly Ash (FA), one of the alumina-silicate materials, is a waste from coal combustion process. The development of the FA-based geopolymer research in Indonesia was based on the physical and chemical characteristics of FA from each burning site that had highly variable characters [4-7]. It was due to the process and resources used in producing a different FA that became a separate issue to determine the overall FA based geopolymer quality[8]. Besides the ratio of alkaline activator composition, the molarity of alkaline and the usage of admixture will affect the compressive strength and workability of fresh geopolymer concrete. The utilized local waste materials were chosen to have the character of pozzolan or alumina-silicate to reduce the dominance of fly ash in geopolymer. This study reviews the possible usage of waste from local materials in geopolymer resistance of high temperatures.

2. Geopolymer Development with Local Waste Materials
Geopolymer is a new material which still developing the standard design procedure of its constituent material mixing up until this moment. However, trial and error method still used. Nevertheless, there are other approaches, for example, Taguchi method. One of the main reasons why there was no standard for mixing procedures is because fly ash's raw materials are quite varied based on the process of picking, transporting and burning coal. Below are the various compositions of local waste materials, with the objection to substituting FA with six local materials in East Java found by researchers.

2.1 Sand-blasting
The mixture of geopolymer paste, used by [13], with the composition of FA (50%) and Sand-blasting waste (50%) resulted in: time setting, compressive strength, porosity, UPV, and permeability. This result was better than Fly Ash composition (100%). Sand-blasting had the potential to increase the setting time of geopolymer. By using 100% sand-blasting composition to geopolymer, the setting time could be reached at about one week. Duan et al. [15]. Has varied the form of silica fume used in his study with 10%, 20%, and 30% respectively of its total binder (they compared with a reference sample of FA 100%). The curing method for the samples were made by heating exposures up to 200°C, 400°C, and 800°C after 28 days. Furthermore, the addition of silica fume could increase the compressive strength rapidly. The addition of silica fume resulted into a denser and more homogeneous sample, which supports the strong performance. The maximum silica used in the study was 30% of total binder. This lead into the creation of most impenetrable and homogeneous microstructure. In another word, it enhances the microstructure and improves its thermal resistance.

2.2 Shell Powder (SP)
The CaO's content on shell powder showed the interaction with FA. According to Freizna’s study, it was noted that the composition of the geopolymer paste were FA (50%) and SLP (50%). This composition resulted in a better compressive strength than FA full replacement of 100%.
Olivia et al. [16] found that the partially substituted-OPC with seashell could decrease the compressive performance but supporting the tensile and flexural strength. Based on research by Lertwattanaruk et al. [17], the calcium content of seashell is less than the OPC's therefore it caused a slow disruption. They have studied that the more shell powder used to replace some part of the OPC the faster its setting time, but the particle remains unreacted and acts as a filler as its particle is smaller than the OPC's. Although, the unreactive act could cause a diminution of strength compared to the reference sample, the compressive strength still met the standard and requirement for plastering. Furthermore, its thermal insulation showed a slightly better value compared to 100% OPC's.
Restu et al. [12] concluded that higher calcium content could disrupt the polymerization process and changes the microstructure, so they have a worse compressive strength compared with the reference's sample (100% FA).

2.3 Sugar Cane Ash (SCA)

The SiO$_2$ content on SCA was above 70% making SCA a potential material for geopolymer paste mixture. Alfi proved that SCA and FA composition of 20%: 80% with additional of Na$_2$SiO$_3$ and NaOH within 1.5 ratio were resulted into a similar compressive strength with FA composition 100%. This result proved that FA dominance reduced by the same result as FA 100%. Castaldelli et al. [18] characterized the ash by its forming compound, as it composed by 46.3% of Silica Dioxides and 23.9% Calcium Oxide, both are from the inorganic fraction (which is 67.8% of the total composition). Beside, this ash composed by high percentage of crystallized matter (quartz mostly, calcite, etc.), which was reactive in a high alkaline environment [19]. The organic fraction which found inside the Sugar Cane Bagasse Ash could affect the setting time above heating temperature of 650°C during calcination process. The determination of mortar’s mechanical properties samples for both compression and flexural test depend on the alkali activators used. Compressive mechanical strength was reported on the range of 30-55 Mpa. Bahurudeen et al. [20] did a deeper research of Raw Bagasse Ash preparation with 600, 700, and 800°C burning temperatures variations and used various fineness number of the ash from 210-45μm. They found that the optimum value of burning was at 700°C. A temperature higher than 700°C could produce trydimite and cristobalite crystallization which led into the degradation of pozzolanic activity[21]. Naturally, this material has less pozzolanic activity than the minimum requirement as per standard. Besides the burning process, we could enhance the rise of pozzolanic activity by grinding process. Sieving trough 300μm were suggested to achieve a maximum pozzolanic activity, lower the loss on ignition, and minimize the energy used in the course mixing. Finer particle gave a better chance of a denser sample. In this circumstances, the mix could fill the air and water cavity to enhance a better performance[3].

The existence of Sugar Cane Bagasse Ash triggered the reactivity of Blast Furnace Slag mixture at early ages [22]. The combination of mixed alkali activator solution and curing temperature of 20°C was resulted in a better compressive strength (55-65 Mpa). The different composition of BFS/SCBA mixture were 100/0, 85/15, 75/25, and 60/40. Pereira, et al.[19] Studied the same mix of BFS and SCBA. He replaced 25% of BFS with SCBA and got a similar/higher compressive strength than the reference (100% BFS) for a longer curing period. After seven hours of 65°C thermal activation, the samples cured for 3-90 days. Based on the results, found that the influence of alkali activator could determine its mechanical compressive strength. The usage of Natrium Hydroxide resulted in a high-early strength. However, when KOH were used as alkali activator, the final strength were two times greater than using NaOH.

2.4 Rice Husk Ash (RHA)

The use of rice husk ash in a geopolymer binder could decrease the compressive strength; it reinforced by the SEM RHA results described that RHA particles had "water absorbed" sponge surfaces and were slow to react. Therefore, the water which used for the polymerization process seemed to reduce due to the ASP absorption and resulted in longer binding time. This result was good for ASP combination with FA. This combination was proposed improvements in future research. Because particle refining with grinding and filtering could optimized the combustion temperatures and firing time. Hwang et al. [23] discussed the effect of substituting part of fly ash with RHA and found out that the optimum substitution proportion was 35% of total weight. Those two pozzolanic materials could be activated using Sodium Hydroxide and Sodium Silicate solution, with the concentration of 8; 10; 12; and 14 M of Sodium Hydroxide and the ratio of Silicate to Sodium Oxide 0.93; 0.81; 0.73; and 0.66. The research proved that the value of 10M Sodium Hydroxide concentrations in 35% RHA mixture at 56 days curing time was the optimum proportion to achieve an ideal compressive strength needed.
The samples were de-molded then cured at 35°C by placing it inside an automated temperature system with 50% relative humidity.

Nazari et al. [3] have varied the material based on its fineness mixture proportion. They have compared the fine fly ash (3μm) and coarse fly ash (75μm) with RHBA fine particle (7μm) and coarse particle (90μm). They used sodium hydroxide and sodium silicate solution with the molarity of 4; 8; or 12 M as alkali activator. The samples obtained were pre-cured at room temperature for 24 hours and soaked in water. There were three ways of curing method. First, the samples were placed in a 25°C ambient temperature and second, the samples were placed inside the oven for 36 hours at the elevated temperatures of 50-90°C; and the last method, samples were heated at 80°C for 36 hours. All samples were tested after 7 and 28 days after it was de-molded. Those processes have shown us that the highest compressive strength were obtained by using 12 M of sodium hydroxide with ratio of 2.99 silica to aluminum oxide and a proportion of 70:30 fine fly ash and fine rice husk ash which obtained 58.9 Mpa of compressive strength.

Sturm et al. [24] concluded some factors which affect the high compressive strength samples. Those factors including:

(a) No swelling found on the hardened paste,
(b) Better workability
(c) A good homogeneity for both for fresh and hardened geopolymer paste.

During geopolymerization, the formation of crystalline by product like Zeolite would enhance the bad strength, which fortunately did not find in the geopolymerization process of RHA one-part geopolymer based. Another advantage of RHA usage is its high reactivity towards the alkali activator. This circumstances will lead into a high SiO2/Al2O3 ratio, which affected the microstructure (ranged start from 3.5 mole/mole) density and homogeneity. RHA has a higher specific surface area compared to other silica-rich material, therefore, it has high reactivity.

2.5 Carbide Waste (CW)

The CaO compound in carbide waste was quite high. It can be seen from Phetchuay et al. research [25]. About 70% of Calcium Carbide chemical composition residue is CaO. It is an indication that this material is reactive and a potential cementitious material [26]. Some studies have done to observe its performances including strength, durability, and microstructure for pavement or subgrade material. Peng Du et al. [27] investigated two type of silicon carbide (silicon carbide particles and whiskers) into their metakaolin geopolymerization. The microstructure analysis concluded that a denser and a less porous geopolymer composite could be achieved by using spherical material particles and whisker's shape in reverse. A spherical carbide particle could induce a better compressive property. Although, both particles have a good interfacial bonding between the filler and geopolymer, its silica reactivity of silica is questioned.

Hanjutsiwan et al. [28] presented that the usage of bottom ash based geopolymer was varied by CCR. He figured out that it has high content calcium hydroxide (which is about the same as the OPC’s). The Bottom Ash which induced with calcium rich-material is a good alternative for the geopolymer binder instead of using Fly Ash.

Previous research has provided a source that a large CaO would affect the acceleration of setting time. Putri's research showed that a good combination of CW and FA-based geopolymer paste at 1: 1 ratio was good portion to achieved compressive strength. While, NaOH 8M, Na2SiO3 ratio also NaOH 0.5 was still better than FA 100% composition.

2.6 Fly Ash and Bottom Ash

Apha Sathonsaowaphaka [29] utilized the bottom ash from power generation waste to develop geopolymer mortar. The conclusion of this study was NaOH addition could slightly increase geopolymer mortar’s resilience while maintaining its strength.

A small ratio of silica to alumina compound, could reduce the compressive strength of bottom ash based geopolymer as reported by Hanjitsuwan et al. [28]. Geopolymer performance will increase after the
addition of SiO2/Al2O3 and calcium or silica promoter like rice husk ash, glass, sugar cane ash, carbide waste, and other materials. Through a microscopically scanning figured, the paste which made from bottom ash contains some unreacted particle or partially reacted particle in the matrix (less dense form compared to the mixture which involving a promoter. According to Lee W et.al [30], aggregate-binder interfaces were stronger in geopolymer than it was in the OPC's, [8] conclude that this particular character could enhance the performance of geopolymer concrete. The use of bottom ash can't substitute the fly ash for improving geopolymer's strength. In other words, the existence of bottom ash could worsen the compressive strength's results instead of improve it [31]. In the same report, the researcher proved that the usage of gypsum gas desulfurization flue (FGDG) could increase the fly ash compressive strength up to 10%. According to SEM observation, a simple fly ash mixture of geopolymer was more homogenous and denser compared to a simple bottom ash mixture. In a high content of fly ash, the usage of FGDG could disrupt the matrices' density. It was in contrary with fly ash.

3 High Temperature Effect on Geopolymer Concrete

Geopolymer concrete has characteristics like ceramics. It shared the same strength characteristics and has a high temperature resistant. Daniel [32] undertook an experimental in geopolymer study under high temperatures that focused on metakaolin-based geopolymer. To compare the performance of metakaolin-based geopolymer on paste, mortar, and concrete, he used the FA-based geopolymer comparators. The highest temperature which used during the study was 800°C. Aggregate dimension factor was the most important parameter to show the performance of geopolymer concrete at high temperature, and the minimum aggregate dimension factor was 10. The comparison of geopolymer mortar strength and OPC showed in Dao's research [33]. This study utilized F class FA from Phalai Power Plant and 14M NaOH activator with K2SiO3 modulus of 3. Complete result of compressive strength ratio with high-temperature influence on mortar presented in Table 1.

| Temperature (°C) | Geopolymer Mortar (GM) (Strength) | OPC Mortar (OM) (Strength) |
|-----------------|-------------------------------|---------------------------|
| 25              | 41.98                         | 40.71                     |
| 200             | 71.01                         | 38.05                     |
| 400             | 68.53                         | 35.21                     |
| 600             | 47.42                         | 29.35                     |
| 800             | 31.77                         | 12.79                     |
| 1000            | 26.36                         | 5.91                      |
Table 1 explained that GM has excellent temperature resistance compared to OM at room temperature. The GM compressive strength sample was about 42 MPa, almost the same as the OM sample. As the temperature increased to 600°C, the compressive strength of OM gradually decreased from 40.71 MPa to 38, 35 and 29 MPa (7, 14 and 18%, respectively, compared to the value at room temperature). On the one hand, when the temperature rose to 600°C, the compressive strength of GM showed an increase of 70, 63 and 13%, compared to the value at room temperature. When the temperature was increased up to 1000°C, OM compressive strength dramatically reduced to 6 MPa, equivalent to only 14% of the strength at room temperature. Similarly, GM's sample also experienced a decrease in strength; however, its strength remained above 26 MPa, equivalent to 63% of the power achieved at room temperature.

Geopolymer was produced using environmentally friendly alkaline activators (based on Rice Husk Ash and potassium hydroxide). The aluminosilicate particles, carbon fibers, and ceramics were used as reinforcing materials. The effect of reinforcement materials on bending strength, linear shrinkage, thermo-physical properties and micro-geopolymer structures in the room and high temperature (1200 °C) were studied. The result showed that the composite toughness increased by 110.4% for geopolymer which reinforced with ceramic fiber (G-AF) at room temperature. The presence of particles improved by 265% on bending behavior for geopolymer which reinforced by carbon fibers after exposure to 1200 °C. Linear-shrinkage occurred in geopolymer which reinforced by ceramic. Its particle grains and G-AF geopolymer compared with reference samples (without fibers and particles) increased by 27.88% and 7.88% respectively at 900 °C. The geopolymer material which developed in this work was a porous material with low thermal conductivity and excellent mechanical properties with potential thermal insulation applications for building applications.

Based on an observation done by [34] the usage of hot-stage TEM could be used to observe the geopolymer microstructure. The researcher explained that four hour geopolymer heat exposure at 1000°C could stabilize the Nano sized microstructure. This researcher tried to compare three different factor of curing, pressure less, warm, and highly Pressured sample in an autoclave with the purpose to find out the effects on the diminution of entrapped air, gain of reactivity, improvement of compound dissolution, and the degree of workability.

A review of [35] suggested that the standard curing temperature was at the range of 60°C to 90°C for 24 hours. [36] agreed to that statement and conducted the research at temperature of 65°C for 24 hours inside oven. A slightly different method of curing performed by [31]. The researcher placed the samples in an electric oven at 40°C for 48 hours. This curing treatment will increase the strength up to 70% within three to four hours.

4 Conclusion
Local material waste could be potentially used to substitute fly ash in geopolymer binders. It could improve geopolymer concrete setting time and compressive strength. Another factor that determines the compressive strength was the fineness of the material since it enhances the activation during the polymerization process. Yet, some other parameters such as alkaline molarity, activator ratio, alkaline composites, superplasticizer addition and any other way play important role for geopolymer concrete's performances. The addition of silica fume will also increase the compressive strength rapidly. In addition, the addition of silica fume can also create a denser and more homogeneous sample, which supports the strong performance. The usage of the seashell in OPC substitution could disrupt the composition of calcium which could potentially change the microstructure and distract the polymerization process. Yet, seashell substitution in OPC could increase the flexural and tensile strength. Bottom ash cannot be used to partly or wholly substitute fly ash due to its bad homogeneity and density. Therefore, the incorporation of other material such as FGDG or silica-rich material is highly recommended. Geopolymer concrete has a better ability than conventional concrete with high-temperature exposure.
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