Effect of Geopolymer filler in Glass Reinforced Epoxy (GRE) Pipe for Piping Application: Mechanical Properties

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Abstract. The present work is aimed to carry out the effect of geopolymer material which is fly ash as filler in the glass reinforced epoxy pipe on the microstructure of fly ash geopolymer, compression properties, and bulk density using the filament winding method. Conventional glass reinforced epoxy pipes has its own disadvantages such as high corrosion resistance at acidic environment and low strength which can be replaced by the composite pipes. Geopolymer is a type of amorphous alumino-silicate and can be synthesized by geopolymerization process. A series of glass reinforced epoxy pipe and glass reinforced epoxy pipe filled with 10 - 40 weight percentage geopolymer filler which is fly ash with 4 Molarity were prepared. Morphology of the raw material fly ash and fly ash based-geopolymer surface was characterized using scanning electron microscopy. It was found that the additions of fly ash at the beginning with 10 wt\% are showing higher compressive strength than glass reinforced epoxy pipe without fly ash geopolymer filler. The compressive test of these series of samples was determined using Instron Universal Testing under compression mode. It was found that compressive strength for samples fly ash based-geopolymer filler are higher as compared to glass reinforced epoxy pipe without geopolymer filler. However, the compressive strength of glass reinforced epoxy pipe with fly ash geopolymer filler continues to decline when added to 20 wt\% - 40 wt\% of geopolymer filler loading. The results showed that the mixing of geopolymer materials in epoxy system can be obtained in this study.
1. Introduction

Glass fiber reinforced epoxy (GRE) composite tubes have a great strength, lighter in weight, and lower risk of bursting which making them commonly preferred in the fields of aviation, structural engineering, and in oil and gas industries [1]. The increased usage of composite structures demands reliable testing methods to ensure the safety and predict their long-term performance. However, previous researches reported that GRE pipes have disadvantages such as lower strength compare to metal pipes, cannot stand at high temperature due to epoxy properties and have low corrosion resistance which can suffer strain corrosion at acidic environments [2-4]. This paper concerns these entire disadvantage includes the application in the offshore oil and gas industry, particularly composite pipelines for aqueous liquids and to produce higher strength of GRE pipe. GRE pipes are normally designed to withstand high pressure, lightweight, relatively thin-walled structure provides ease of handling and transportation, which results in reduced installation costs [5, 6]. Geopolymer filler which is fly ash is use in this research to overcome this problem.

Geopolymer is a type of amorphous alumino-silicate cementitious material and can be synthesized by polycondensation reaction of geopolymeric precursor and alkali polysilicates known as geopolymerization process [7]. Through a period of time, substantial research struggles have been focused towards the development of inorganic geopolymers, due to the wide range of potential applications for these materials. Several reports can be found in the literature on the synthesis, properties and applications of geopolymers [8]. Geopolymerization is an innovative technology that can transform several aluminosilicate materials into useful products called geopolymers or inorganic polymers. The remaining achievements in inorganic chemistry made through geopolymerization include mineral polymers which termed as polysiatate or geopolymers, making a great possibility to produce composite materials not only with excellent mechanical properties such as lightweight and high strength but also with ideal fire resistant, non-toxic fumes and smokes, and resisting all organic solvents [9-11]. For several years, researchers have been enthralled by the studies on geopolymers which offered low energy consumption and the absence of CO₂ emissions in the preparation process [12-14]. Composites based on clay have been more widely studied since clay materials are naturally available and contribute to excellent mechanical properties of either thermoplastic or thermosets matrices [15-17].

In this research, GRE pipe which is diglycidyl ether of bisphenol A (DGEBA) epoxy were mixed with fly ash geopolymer filler with 4 Molarity concentration of sodium hydroxide (NaOH) and ratio solid to liquid is 1. To produce a higher strength of geopolymer, the optimum sodium silicate to sodium hydroxide ratio should be in range of 0.67 to 1.00 [18]. The formulation design of geopolymer was referred to the composite system as stated from the previous researcher on composite system which is use the loading of flax fibers varied from 0 to 60 wt%. [19]. Epoxy resin DGEBA was mixed with geopolymer filler which one of the greatest current interests in thermoset-based clay composites since it offers low cost, ease of processing, fine adhesion to many substrates, and good chemical resistance for a wide range of applications [20]. Fly ash contains of finely divided ashes produced by pulverized coal in power stations. The spherical shape of fly ash improves the consolidation of pipe, which also reduces permeability [21]. Therefore, these materials (fly ash, kaolin, and white clay) has wide potential to be used as a source material to react with liquid alkaline activator within sodium silicate solution (Na₂SiO₃) and sodium hydroxide (NaOH) mixture [22].
2. Material and Experimental Details

2.1 Materials selection

The epoxy resin, Diglycidyl Ether of Bisphenol-A (DGEBA) supplied by Euro Pharma Sdn Bhd and hardener Isophorondiamine (IPDA) was obtained from Dr Rahmatullah Holdings was used in this study. The raw materials geopolymer filler was produced by Saudi local based materials which is Fly Ash was used in preparation of geopolymer. Geopolymer paste is formed by alkaline activator to induce the silicon and aluminum atoms in the material [23]. Alkaline activator liquid used in this research is combination of sodium hydroxide (NaOH) with 4 Molarity and sodium silicate (Na$_2$SiO$_3$).

Alkaline activator was prepared by mixing NaOH and sodium silicate before mixed with raw materials to increase the reactivity of solution with concentration 4 M of NaOH to determine the optimum concentration. The ratio of raw materials to alkaline activator and sodium silicate to NaOH is 1. The geopolymer pastes then were cured in oven for 24 hours to 48 hours at 60 $^\circ$C – 80 $^\circ$C and then were taken out to be crushed by using ring mill. Crushed geopolymer filler were sieved using sieve size of 150 µm.

2.2 Experimental procedure

The Epoxy Geopolymer was prepared according to the formulation in the table 1 by mechanical mixer using blade stirrer. The mixing epoxy geopolymer were cured with cycloaliphatic amine curing agent IPDA. Epoxy and geopolymer materials were mixed for about 2 hours to make it completely homogeneous, followed by curing agent/hardener for about 5 minutes. Then the resin was poured into the tank after mixing with epoxy hardener.

Continuous glass fibers (E Type) were impregnated (“wet-out”) with geopolymeric resin by means of homemade “impregnation machine” as filament winding technique. The velocity of the fiber during the impregnation process was chosen based on the best penetration on geopolymer resin into the fibers. The winding speed was controlled to generate the desired winding angle patterns. The feeding velocity of the fiber into the resin tank will depends on the mandrel rotational speed. It was selected based on the best impregnation of geopolymer resin into the fibers.

After the appropriate number of layers has been applied according to fully wounded, filament winding samples were allowed to be cured at the mandrel in the room temperature for 24 hours. After the curing process completed, the sample is ready for testing. In order to determine the mechanical properties, several tests are performed on composite structures which are compressive test and elasticity modulus.

| Geopolymer Material (4 M) | Epoxy + Hardener (%) | Geopolymer (%) |
|---------------------------|----------------------|----------------|
| Fly Ash                   | 100                  | 0              |
|                           | 90                   | 10             |
|                           | 80                   | 20             |
|                           | 70                   | 30             |
3. **Result and discussion**

3.1 **Surface morphology of raw material fly ash**

Figure 1 shows the surface morphology of the raw material fly ash. It can be seen that the microstructure of fly ash appeared to be a glassy, hollow and clearly shows the shape of fly ash as reported from previous researcher contains of spherical particles with smooth outer surfaces [24]. The smooth aluminosilicate spherical particles, also known as cenospheres are formed as a result of mineral particles during coal combustion process, where the minerals melt to form small droplets, which upon rapid cooling and action of surface tension forces adopt the spherical shape [25-27]. The raw material of fly ash shows that the spherical morphology of the raw fly ash was fully destroyed which mean there have some agglomeration of the fine grained particles due to mechanical milling processes.

![Figure 1: SEM micrographs of the raw fly ash at 2000x and 5000x magnification](image)

3.2 **Bulk Density Analysis**

Bulk density is one of the physical properties that can be measured by simple procedure and calculated manually with mass per unit volume of sample. From Figure 2, it can be seen that the fly ash geopolymer filler with 40 wt% of filler loading has the highest bulk density which is 1.585 g/cm³ as compared to other samples. GRE pipe with fly ash based-geopolymer with 10 wt% has the lowest bulk density which is 1.414 g/cm³ due to low filler loading. As a result, the density of the sample will decrease or less dense as the porosity increase.
3.3 Compression Analysis

Figure 3 shows the typical graph from the tests of compression strength tests for the glass reinforced epoxy (GRE) pipe without filler and glass reinforced epoxy (GRE) pipe filled with 10 wt% - 40 wt% of fly ash geopolymer filler. Compression test was performed according to the ASTM D3410 using Instron Universal Testing Machine [28]. The compression test consists of deforming a cylindrical hollow specimen to produce a thinner cylinder hollow of large diameter. It is a convenient method for determining stress strain response.

From Figure 3, it is clearly indicated that GRE pipe with 10 wt% of fly ash geopolymer filler shows the highest compressive strength compare to others composition. However, compressive strength of GRE pipe filled with geopolymer filler of fly ash are slightly continue to drop with the increase of the percentage of geopolymer filler loading and it was supported by the rule of mixture theorem which the addition of micro particles offer greater stiffness than epoxy matrix [29]. This may be due to the smaller agglomerated and disordered nature of clay and also the uses of lower molarity of NaOH. Even the compressive strength of GRE pipe with fly ash geopolymer filler drop when added with higher filler loading, the compressive strength is much higher than GRE pipe without geopolymer filler. Table 2 shows the values of compressive strength, compressive strain and compressive modulus of elasticity for the 0 wt% – 40 wt% fly ash geopolymer filler loading.

Figure 2: Comparison graph of bulk density between samples
Figure 3: Compressive strength of GRE pipe with 0 wt% - 40 wt% geopolymer filler loading

The high compressive strength revealed a strong interaction between polymer chains which is epoxy hardener with the geopolymer filler itself. However, the decreasing of compressive strength in higher contain of fly ash which 20 wt% – 40 wt% may be controlling the capability and capacity of relocating load and plastic deformation between particles and matrix interface.

Table 2. Compression Properties of Epoxy Hardener and Epoxy Geopolymer

|                | Compressive Strength (MPa) | Compressive Strain (%) | Modulus Elasticity (MPa) |
|----------------|---------------------------|------------------------|-------------------------|
| Epoxy          | 53.36                     | 0.04                   | 1681.28                 |
| Fly Ash        | 75.49                     | 63.75                  | 2295.85                 |

3.4 Surface morphology of fly ash based-geopolymer

Figure 4 shows the fly ash based-geopolymer that undergoes 24 hours of curing time in the oven within 70 °C and has been crushed into small particles using a milling machine (ring mill). The changes on the surface morphological of raw material of fly ash and fly ash based-geopolymer observed show changes due to the reactive dissolution of SiO2 and Al2O3 (contained in the cenospheres) in alkaline solution. This will leads to the formation of alumino silicate gel, which then acts as a precursor to geopolymer formation [30].
4. Conclusion

From this study, it can be concluded that glass reinforced epoxy (GRE) pipe filled with fly ash based-geopolymer was developed on epoxy resin with different weight percentage of the geopolymer material. The experimental result shows the performance of the product through the compression test, morphology, and bulk density test. Based on the results of the compression test, the product from GRE pipe with fly ash based-geopolymer shows the higher strength compared to the GRE pipe without geopolymer filler. GRE pipe with 10 wt% of fly ash based-geopolymer filler shows the highest compressive strength. Due to the good properties of the waste materials based geopolymer, it has a bigger potential to be a matrix filler of composite with glass fiber in filament winding technique. Geopolymer composite can be as a filler in the piping system application through filament winding technique that not only environmentally but also reducing the production cost of the product.

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