The Influence of Sintering Method on Kaolin-Based Geopolymer Ceramics with Addition of Ultra High Molecular Weight Polyethylene as Binder.

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Abstract. The influence of sintering method on kaolin-based geopolymer ceramics with addition of Ultra High Molecular Weight Polyethylene as binder were studied. Geopolymer were formed at room temperature from kaolin and sodium silicate in a highly alkaline medium, followed by curing and drying at 80 °C. 12 M of sodium hydroxide solution were mixed with sodium silicate at a ratio of 0.24 to form alkaline activator. Powder metallurgy technique were used in order to produce kaolin geopolymer ceramics with addition of Ultra High Molecular Weight Polyethylene. The samples were heated at temperature of 1200 °C with two different sintering method which are conventional method and two-step sintering method. The strength and density were tested.

1. Introduction
Inorganic aluminosilicates produced at low temperatures, also known as geopolymers, have potential applications as matrix in cements, coatings, and ceramics [1]–[3]. Synthetic alkali aluminosilicate material called geopolymer are produced through a reaction between aluminosilicate materials with alkali or alkali silicates under highly alkaline conditions [4][5][6]. Chemically, geopolymers contain of cross-linked units of AlO4 and SiO4 tetrahedra, where charge-balancing cations are provided by alkali metal cations such as Li+, Na+, K+, and Cs+ [4].

The characteristics of geopolymer includes high strength, fast or low setting, low shrinkage, acid resistance, low thermal conductivity and fire resistance. For the reason, geopolymers are considered to be suitable for a broad range of engineering materials applications [7][8][9]. It can be used as sealants, concretes, fire and heat resistant fibre composites, ceramics, etc., depending on the chemical composition of the source material and the activators [10]. Furthermore, geopolymer technology provides a new route to produce advanced ceramics with beneficially short fabrication time with the proper processing procedure and low cost. The geopolymer with amorphous to semi-crystalline behavior will transform into crystalline ceramic phases on heating [11][12][13]. Geopolymer also can

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be directly transformed into final structural ceramic part of interest and also convenient to design the chemical composition of the final product [14].

Porous ceramics are significantly outstanding permeability and have a large surface area. Their greater properties can be extensively used in diverse fields such as ceramic filters, bioceramics, fuel-cell electrodes and membrane reactors [15]. In the spite of widen the applications, these ceramic should not have only large interconnections and high porosity but also high in strength and dense networks. Therefore, the addition of binder in production of geopolymer ceramics can be one of the solution to achieve all those excellent properties. The binder were used purposely to deform the plasticity between ceramic particles hence transmit an adequate strength to the green compact [16]. The sintering method will allow densification to occur without stimulating the grain growth which is suitable to microstructural refinement lead to produce dense ceramics. A great deal of different methodologies has been developed with a view to improve the mechanical properties through powder consolidation. Other than conventional method that were used by many researchers on their study of ceramic fabrication, the two-step sintering is also among the favourable method found to effectively control the undesirable grain coarsening during densification[17]. Furthermore, the removal of binder usually take place in oxidising atmosphere at moderately elevated temperatures, and generally become complete at around 600 °C to 700 °C for polymers [16]. Thus, sintering method plays an important role in order to produce ceramic materials with the desired properties.

This paper presents a study on the effect of sintering method of the kaolin-based geopolymer ceramics with addition of UHMWPE as binder. The addition of polymer (UHMWPE) into geopolymer ceramic materials prepared by using powder metallurgy method were sintered at 1200 °C. The effect of two different methods which are conventional method and two-step sintering method on flexural strength and density were studied.

2. Experimental

2.1 Materials

Kaolin was provided by Associated Kaolin Industries Malaysia as Si-Al sources materials. The summarization of chemical composition of kaolin has been analysed by X-ray fluorescence (XRF) (Table 1). Malvern particle size analyser were used to analyse the particle size distribution of aluminosilicate materials. Sodium hydroxide (NaOH) powder used was of caustic soda micropearls, 99% purity with brand name of Formosoda-P made in Taiwan. Sodium silicate (Na₂SiO₃) chemical composition are 9.4% of Na₂O, 30.1% of SiO₂, and 60.5% of H₂O (modulus, SiO₂/Na₂O = 3.2), specific gravity at 20 °C = 1.4 g/cm³ and viscosity at 20 °C = 0.4 Pa s. The UHMWPE in powder form, with a molecular weight of 5 X 10⁶ g/mol and density of 0.94 g/ml was used as binder in kaolin-based geopolymer ceramics and supplied from Ticona Engineering Polymer, China.

| Table 1. Chemical composition of kaolin. |
|-----------------------------------------|--------|--------|--------|--------|--------|--------|--------|
| Composition | SiO₂ | Al₂O₃ | Fe₂O₃ | K₂O | TiO₂ | MnO₂ | ZrO₂ | LOI |
| Percentage (%) | 54.0 | 31.7 | 4.89 | 6.05 | 1.41 | 0.11 | 0.10 | 1.74 |

2.2 Samples preparation

The 12 M of NaOH solution were mixed with Na₂SiO₃ at a ratio of 0.24 to form an alkaline activator solution. In order to dissolve the silica completely, the solution was then allowed to mature for 24 h. Kaolin powder were mixed with alkaline activator with the solid-to-liquid ratio of 1.0. A mechanical
mixture was used to stir the mixture for few minutes to obtain a homogeneous mixture. The details of the mixtures involved were shown in Table 2. The ratio was selected based on our previous work, as it allowed an optimal physical and mechanical properties. The kaolin-based geopolymer samples were crushed using mechanical crusher after the samples cured in the oven at 80 °C for 24 h. Then the samples were sieved by using 150 µ siever to obtained fine powder. The UHMWPE (4 wt. %) were added into kaolin-based geopolymer powder by using planetary mill in dry condition method. The mixture was mixed for 4 minutes at 100 rpm with reverse direction until achieve their homogeneity. After that, the mixture was compacted using a 12-mm-diameter cylindrical stainless steel die at 5 tons for 2 min. The final green body was sintered in a high temperature Carbolite furnace 1200 °C with two different sintering method which are conventional and two-step sintering method as shown in Figure 1 with a cooling and heating rate of 5 °C/ min.

| S/ L | NaOH molarity | Na₂SiO₃/ NaOH | Total molar ratio | Na₂O/ SiO₂ | SiO₂/ Al₂O₃ | H₂O/ Na₂O |
|------|---------------|--------------|-------------------|------------|------------|-----------|
| 1    | 12            | 0.24         | 0.3950            | 3.2022     | 11.5730    |

Figure 1. Sintering method used in fabrication of kaolin-based geopolymer ceramic with addition of UHMWPE (a) conventional method and (b) two-step sintering method.

2.3 Testing
2.3.1 Flexural Strength
Flexural strength test was conducted on specimens (7 mm x 5 mm x 52 mm) by using a three-point-bending fixture on an instron-500 tester. With a span length of 30mm at a crosshead speed of 0.5 mm/min, five samples for each group were subjected to flexural strength testing in accordance to ASTM C1161.

2.3.2 Density
Pycnometry (AccuPyc II 1340 He pycnometer, Micromeritics, GA, USA) were used to analyzed density of the geopolymer ceramic. A cylinder shaped sample with diameter of 12 mm and length of 4 mm were used in this experiment.
3. Results and Discussion

3.1 Flexural Strength

The effect of sintering method on the kaolin-based geopolymer ceramics heated at 1200 °C is shown in Figure 2. The flexural strength of kaolin-based geopolymer ceramics sintered using two-steps sintering method is higher compared to the kaolin-based geopolymer ceramics sintered using conventional method. The pre-treatment heating in two-steps sintering method increased the fracture toughness of the grain boundaries, reduced the subcritical growth rate of sintering flaws hence resulting in increasing of the strength [18]. The addition of 4 wt. % of UHMWPE to the kaolin-based geopolymer ceramics shows the highest strength of 94.32 MPa. The effect of binder on kaolin-based geopolymer ceramics is obvious with an increasing of strength greater than the kaolin-based geopolymer ceramics without addition of UHMWPE. This contribution to the strength were due to the advantageous of the carbonaceous residue for the resistance against crystallization thus produces a complex microstructure for carbon-rich of geopolymer ceramics [19].

![Figure 2. Flexural Strength of Kaolin-Based Geopolymer Ceramics with and without UHMWPE at different sintering method](image)

3.2 Density Analysis

Figure 3 shows the density of the kaolin-based geopolymer ceramics with and without addition of UHMWPE at different sintering method. Kaolin-based geopolymer ceramics with and without addition of UHMWPE that sintered with two-steps sintering method shows a lower density compared to the kaolin geopolymer ceramics sintered with conventional method. The kaolin-based geopolymer ceramics with addition of 4 wt. % of UHMWPE sintered with two-steps method achieved the lowest density of 2.095 g/cm³. It was clear that by having pre-treatment heating at lower temperature consequently reduce the densification rate. According to the Maglia et al (2013), the neck formation among particles during pre-heating leads to the more compaction, produce a slighter distribution in pore size and promoting the eliminating of fine particle thus becoming a factor in decreasing the density [20]. The decreasing of density in terms of addition of UHMWPE as a binder were contributed...
by the thermal decomposition of UHMWPE that resulting in formation of pores hence affected the density of the green part [21].

![Figure 3](image)

**Figure 3.** Density of Kaolin-Based Geopolymer Ceramics with and without UHMWPE with different sintering method

### 4. Conclusion

This paper is purposely to study the influence of sintering method on kaolin-based geopolymer ceramics with addition of Ultra High Molecular Weight Polyethylene (UHMWPE) as a binder. Results determined that the sintering method used affected the strength of the kaolin-based geopolymer ceramics. The flexural strength of kaolin-based geopolymer ceramics sintered by using two-steps sintering method is higher compared with using conventional method. Kaolin-based geopolymer ceramics with addition of 4 wt. % UHMWPE give the highest strength which is 94.32 Mpa. The density of the kaolin-based geopolymer ceramics with addition of UHMWPE using two-steps sintering method give a lowest value (2.095 g/cm³) and decreasing in density were observed for the addition of UHMWPE for both method of sintering.

### 5. Acknowledgments

The authors of the present work wish to dedicate their great thanks to Centre of Excellence Geopolymer System Research @UniMAP.

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