Comparison Study on Microstructure Properties of Kaolin Based Geopolymer Ceramics with Addition of UHMWPE under Different Sintering Condition

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Abstract. To better understand the structure-mechanical properties relation of additively fabrication of kaolin geopolymer ceramics with sintering method, a comparison study was performed. Kaolin based geopolymer ceramics were synthesized starting from the powders of kaolin based geopolymer, using powder metallurgy method. Typically, the sintering method used are one step (1200 °C – 5 min) and novel two step sintering processes (600 °C-5 min and 1200 °C – 5 min) for both kaolin based geopolymer ceramics with and without ultra high molecular weight polyethylene were applied and compared. The outcome revealed that there is no phase changes on the both sintering method pattern and the two step sintering method giving a smooth surface owing to the densification process during the preheat treatment.

1. Introduction
The combination of compacting and sintering process in the fabrication of ceramic materials has become increasingly important in modern industry due to superior mechanical and physical properties. A crystalline or partly semi-crystalline structure of ceramic are fabricated by a great number of small crystals, or grains separated from one another by grain boundaries [1]. Green properties of ceramic components are depending on the powder packing while the additives such as binder, surfactants, plasticizers, dispersants and lubricants also plays a major role, irrespective of the various processing technique [2]. The binder were used to plastically distort the ceramic particles in order to convey sufficient strength to the green compact, subsequently sustaining the integrity of the green part throughout ejection and presintering process. Although, the present of the binder also can affect the relative [3]. Thus, the usage of ultra high molecular weight polyethylene (UHMWPE) perhaps can develop the strength of the green bodies by binding the geopolymer ceramic particles due to their characteristic features includes the dimensional sheets covalently bond of network, the functional group types, and the length of molecular weight, chain and distribution [4].
Inorganic polymers called geopolymers harden at ambient temperature, having great potential applications as construction materials, matrix in ceramics other composite materials and coatings [5]. Geopolymer are synthesized by the polycondensation of silica-alumina structure. The reaction between solid aluminosilicate with high concentrated of aqueous alkali hydroxide or silicate solution can produces a synthetic alkali aluminosilicate material [6]. Geopolymer that having amorphous to semi-crystalline phases which contain random tetrahedral network of silicon and aluminium atom will transform into crystalline phase upon heating [7]. Among the unique feature of geopolymer is the high temperature in ceramic materials fabrication is no longer needed. With the help of geopolymerization, the homogeneous of the geopolymer system will influence the structural arrangement during the phase changes thus promoting the nucleation and densification process.

In general, the engineering properties of ceramic materials are measured by the microstructure such as grain size and density. In order to enhance this microstructural, the sintering mechanism is one of the factors that should be discovered due to the material densification will be promoted during this process [8]. Different methodologies have been made with a view to develop the mechanical properties through powder consolidation that appear to offer major property benefits. The influence on the sintering method in the microstructure of ultrafine alumina were studied by Chinelatto et al., (2012) [9]. By heating the samples to 1050 °C, smaller final grain were observed due to the removal of finest particles and narrow particles size distribution. Vogiatzis et al., (2015) found that the two step sintering can avoid any thermal shock that lead to cracking and deteriorating the structural of the samples [10]. Thus, this study was to investigate the effect of sintering method on the microstructural properties of kaolin based geopolymers ceramics with and without addition of UHMWPE as binder.

2. Experimental Work
2.1. Materials
Kaolin is a clay mineral composed of Al$_2$Si$_2$O$_5$(OH)$_4$ chemical composition (Table 1) with good characteristics such as natural whiteness, fine particle size, non-abrasive stability and chemical stability. Kaolin that a layer of silicate mineral with one sheet of tetrahedral silica (SiO$_4$), linked through oxygen atoms to one sheet of octahedral alumina (AlO$_6$) octahedral was bought from the Kaolin Industries Associated Sdn. Bhd. Malaysia in powder form.

| Composition | SiO$_2$ | Al$_2$O$_3$ | Fe$_2$O$_3$ | K$_2$O | TiO$_2$ | MnO$_2$ | ZrO$_2$ | LOI |
|-------------|--------|------------|------------|-------|--------|---------|--------|-----|
| Percentage (%) | 54.0   | 31.7       | 4.89       | 6.05  | 1.41   | 0.11    | 0.10   | 1.74|

Sodium silicate (Na$_2$SiO$_3$) solutions were supplied by South Pacific Chemicals Industries Sdn. Bhd. Malaysia. The chemical compositions of of the solution given by the supplier are 30.1% SiO$_2$, 9.4% Na$_2$O and 60.5% H$_2$O. While for the sodium hydroxide (NaOH) is supplied by Formosa Plastic Corporation, Taiwan. with a brand name of Formosoda-P were used in this study.

Ultra high molecular weight polyethylene (UHMWPE) was used as binder with a molecular weight of 5 X 106 g/mol and density of 0.94 g/ml, in powder form was purchased from Ticona Engineering Polymer, China.

2.2. Fabrication of Kaolin Based Geopolymer Ceramics
Alkali activator was prepared by mixing 12M of NaOH solution with Na$_2$SiO$_3$ at a ratio of 0.24. The solutions were then allowed to mature for 24 h permitted to dissolve the silica homogeneously. The geopolymer paste was prepared by mixing the kaolin powder with alkali activator solution at solid-to-liquid ratio of 1.0 and was stirred by using mechanical mixer and cured at 80 °C for 24 h. The ratio was chosen based on our previous work since it allowable for optimal physical, mechanical and thermal properties.
The kaolin geopolymer were then crushed and sieved manually by using 150µ siever. The kaolin based geopolymer powder was then mixed with 4 wt.% of UHMWPE by using planetary mill in dry condition method for 4 minutes at 100 rpm with reverse direction. Then, the samples through compaction process by using a 12-mm-diameter cylindrical stainless steel die at 5 ton for 2 min and sintered at two different sintering condition which are one step and two steps method with a cooling and heating rate of 5 °C/ min. For the two steps method, the samples were heated at lower temperature of 600 °C with 60 min of soaking and proceed to second steps of heating up to 1200 °C with 120 mi of soaking.

2.3. Microstructure Testing

XRD 6000, SHIMADZU diffractometer were used to characterize the phase involved in the samples. Samples were prepared by classic preparation technique based on the pressing of powder samples into aluminium holders with the operating settings at 40 kV and 30 mA using Cu-Kα radiation. The peak were identified by using X’Pert High Score Plus. The microstructure of the sample was revealed by using JSM-6460LA model Scanning electron microscope (JEOL). The specimens were prepared and coated by using Auto Fine Coater; model JEOL JFC 1600.

3. Results and Discussion

3.1. Microstructure

The XRD patterns of kaolin based geopolymer ceramics with and without addition of UHMWPE at different methods of sintering are shown in Figure 1 and Figure 2. Both samples with and without addiotion of the binder shows the same pattern. Nepheline (NaAlSiO₃) phase appeared as major phase in both XRD patterns. Crystallization of nepheline (NaAlSiO₃) from kaolin geopolymer gradually occurred on heating [11] and its proof that there is no phase change occurred due to the same sintering temperature used. However, in the XRD pattern kaolin geopolymer ceramics with addition of various content of UHMWPE, appearance of extra peak of carbon (C) were observed at 2θ values of 17.7 °, 31.0 °, 35.9 °, 37.5 °, 41.7 °, 45.7 ° and 56.2 ° credited to the decomposition of UHMWPE while sintering at high temperature [12]. The samples sintered with two step method illustrate a sharper peak compared to samples sintered with conventional method. In addition, the intensities increased in the XRD pattern of samples that sintered using two step method in which consistence with the high value of strength [13, 14].

![Figure 1. XRD pattern for kaolin based geopolymer ceramics at different sintering method (N= Nephaline, Z= Zeolite).](image-url)
Figure 2. XRD pattern for kaolin based geopolymer ceramics with addition of UHMWPE at different sintering method (N= Nepheline, C= Carbon).

Figure 3. SEM micrograph of kaolin based geopolymer ceramics sintered using (a) one step method, (b) two step method and with addition of UHMWPE, (c) one step method, (d) two step method.
The SEM micrographs of the fractured surface of kaolin based geopolymer ceramics with and without addition of UHMWPE at different sintering method were compared in Figure 3. Both samples with and without the addition of UHMWPE that sintered using two step method shows a more uniform and homogeneous microstructure compared with the samples using one step sintering method. This is due to the preheating phase in two step method that induced densification process by eliminating the finest particles of the samples [9]. The fractured surface of both samples with and without addition of UHMWPE shows formation of pores for both method. However, the pore size become irregular and some of the pores confined within the grains during the faster densification of finest particles in two step sintering method [15-18]. In the study on the effect of the pre-sintering on microstructural evolution of alumina carried out by Lin et al. (2005), the average of pore size of a pre-sintered compact were found to increases after the preheat treatment and the pore-size distribution becomes narrower [13].

In general, the microstructure of the ceramic bodies showed a network of small dense zones interconnected by a porous phase. However a noticeable improvement is shown in the microstructure of the samples that heated with two sintered method in which would improve the physical and mechanical properties of the ceramic materials.

4. Conclusion
The use of two steps sintering methods shows a noticeable improvement in microstructural characteristics properties for both samples with and without the addition of UHMWPE. XRD results of all the samples show significant similarities in term of peaks appearance of nepheline phase which used to occur at the same sintering temperature. While the microstructure analysis demonstrated that the two step method allowing an improvement of the microstructure, thus can improves the mechanical properties of the samples.

5. References
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