# Densification Behavior and Mechanical Performance of Nepheline Geopolymer Ceramics: Preliminary Study

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**Abstract.** Nepheline geopolymer ceramics have emerged as a promising sustainable alternative to traditional cementitious materials in various applications. As the sintering mechanism plays a crucial role in the densification and mechanical performance of ceramics, therefore, in this paper, a preliminary study was conducted to examine the effects of densification towards mechanical properties of geopolymer-based nepheline ceramics upon sintering. The said innovative geopolymer technology can convert raw materials of aluminosilicate activating with alkaline activator into ceramic-like materials requiring low temperatures. The experimental procedure includes the synthesis of nepheline geopolymer ceramics through the geopolymerization method, then sintered at different temperatures to explore the sintering behavior and its impact on the materials' microstructure and mechanical performance. The densification behavior of nepheline geopolymer ceramics during sintering was analyzed by evaluating the changes in density, shrinkage, and porosity.The microstructural evolution and are determined by using SEM. The relationships between sintering conditions, microstructure, and mechanical performance were investigated to understand the underlying mechanisms affecting the material's strength and durability. The geopolymer exhibited its highest flexural strength of 54.93 MPa when sintered at 1200 °C, while the lowest strength of 6.07 MPa was observed at a sintering temperature of 200 °C. The findings demonstrate a positive correlation between the sintering temperature and the flexural strength of the geopolymer ceramics, indicating that higher temperatures lead to increased strength. Ultimately, this knowledge can facilitate the broader utilization of nepheline geopolymer ceramics as sustainable materials in various engineering and construction applications.

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## 1 Introduction

In the production of high-strength ceramics, the conventional method often involves high processing temperatures exceeding 1600 °C. However, this high temperature can have adverse effects on the resultant materials, particularly on pore size and distribution, which are critical factors influencing mechanical properties. Methods like Hot Isostatic Pressing (HIP) and Spark Plasma Sintering (SPS) have been utilized to produce high-density ceramics. However, these methods often pose challenges in terms of cost and complexity, making them inaccessible to many industries and research institutions that lack the necessary equipment and expertise to carry out the intricate sintering process and control requirements [1]. Conventionally, sintering of glass-ceramics typically involved two main steps. Firstly, the raw materials are heated to high temperatures, reaching up to 1500 °C, to achieve vitrification, followed by the processes of nucleation and crystal growth [2]. Besides, the requirement for high sintering temperature, the resulting ceramics also suffer from various complications such as agglomeration, abnormal grain growth, and furnace contamination. Therefore, the novel method of using geopolymer in producing ceramic materials is introduced.

In the 1970s, a researcher Davidovits introduced geopolymer firstly as an alternative to developing inorganic polymer materials. Geopolymers, classified as ceramic-like inorganic polymers, are synthesised at temperatures below 100 °C. These materials consist of interconnected mineral molecules bonded by covalent links. The composition of geopolymers typically involves aluminosilicates, including rock-forming minerals, amorphous silica, and aluminosilicate-rich industrial by-products like coal fly ash and blast furnace slag. These raw materials offer ample sources of aluminosilicates for geopolymer production. By mixing the aluminosilicate sources into a strong alkali activator such as sodium hydroxide (NaOH), geopolymers were synthesised by the dissolution and polycondensation of aluminosilicate materials in alkali-activated solutions [3, 4]. Geopolymers offer numerous desirable characteristics, including low weight, impressive thermal and mechanical properties, strong resistance to chemicals, and good permeability. These advantageous traits make geopolymers suitable for various applications, such as high-temperature-resistant ceramics, heat-resistant coatings and adhesives, containment of radioactive waste, and as cementitious components in construction materials [5]. Moreover, geopolymers are gaining recognition as suitable matrices for reinforced composites, various refractory applications, corrosion-resistant coatings, and as precursors for the formation of ceramics [6].

In contemporary times, there has been a burgeoning interest in innovative ceramic materials based on geopolymers, owing to their remarkable mechanical characteristics and ecological compatibility. These materials have garnered significant attention and are poised to exhibit substantial applicability in the times ahead. Geopolymer exposed to high temperatures by the sintering process produces ceramic materials. This is due to the effect of the sintering mechanism when heat is applied to the geopolymer body. For example, geopolymer was used to produce high-flexural strength ceramics and one-part-mixing GP [7]. Nevertheless, the utilization of high-temperature sintering alone can induce undesirable shrinkage and cracking, consequently compromising the overall strength of the end products. The utilization of low-temperature geopolymer-based ceramics offers convenience in the production of diverse and intricately shaped products. Moreover, these ceramics exhibit commendable fire resistance, environmental friendliness, and exceptional thermal properties [8]. Furthermore, the exploration of porous geopolymer-based ceramic materials has gained significant attention in current research due to their notable stability, high surface area, and permeability. These materials hold promise for various applications in lightweight structural components, filters, and other relevant areas.

The characteristics of the pores, such as their shape, volume, and distribution, in porous geopolymer-based ceramics are influenced significantly by the manufacturing technique and the composition of the composites. Research has demonstrated that by altering the chemical composition of the geopolymer-based ceramics, it is possible to control the water retention and adjust the porosity accordingly [9]. The progression of phase formation subsequent to the sintering process is contingent upon multiple variables, including the composition of the raw materials, the preparation of the ceramic mixtures, and the specific optimum temperature at which the ceramic structure is subjected to the sintering procedure [10]. However, there is a lack of studies investigating how the sintering mechanism affects the densification and crystallisation of geopolymer-based ceramics as it correlates with the mechanical properties of the end product. Therefore, in this paper, the sintering mechanism of geopolymer-based ceramics is studied. Effects on the densification of ceramics products and the mechanical performance are analysed when heat is applied during the sintering process.

## 2 Experimental Method

### 2.1 Geopolymer-Based Ceramic Sample Preparation

The aluminosilicate used in this study was kaolin supplied by Associated Kaolin Industries Sdn. Bhd, Petaling Jaya, Selangor, Malaysia. Kaolin has a chemical formula of $Al_2Si_2O_5(OH)_4$, where $SiO_2$ and $Al_2O_3$ contribute a large amount to its composition. The alkali activator was prepared by mixing liquid sodium silicate ($Na_2SiO_3$) and sodium hydroxide (NaOH) solution. The NaOH solution was prepared by mixing distilled water with caustic soda pellets from Formosoda-P, Taiwan. The liquid sodium silicate was supplied by South Pacific Chemicals Industries Sdn. Bhd. (SPCI), Pahang, Malaysia. It is composed of 30.1% $SiO_2$, 9.4% $Na_2O$, and 60.5% $H_2O$.

A fully dissolved 12 M NaOH solution was mixed with $Na_2SiO_3$ with a molar ratio of 0.24. Geopolymer-based nepheline ceramic is prepared by mixing kaolin raw materials with an alkali activator, producing a slurry geopolymer paste. The paste then being cured at 80 °C for 24 h before the geopolymer sample was milled and sieved using a 150 μm siever to obtain a fine geopolymer powder. The fine powder was compressed at 4.5 tons for 2 min. The geopolymer was sintered at six different temperatures of 200 °C, 400 °C, 600 °C, 800 °C, 1000 °C, and 1200 °C, soaking time of 180 min and 5 °C/min heating rate. Later, the nepheline geopolymer-based ceramics produced were characterized to study the density behaviour and its mechanical properties upon the increment of sintering temperature.

### 2.2 Characterization of Geopolymer Ceramic

The flexural strength of the geopolymer-based ceramics was evaluated as a measure of their mechanical properties. The testing was conducted using a three-point bending fixture with dimensions of 7 mm x 5 mm x 52 mm, following the ASTM C-1163b standard. To determine the flexural strength of kaolin geopolymer ceramic samples, an Instron-500 tester is utilized. The specimens are subjected to a three-point bending test using a fixture with a span length of 30 mm, and the crosshead speed is set at 0.3 mm/min. The density of geopolymer-based ceramics was determined by applying Archimedes' Rule. The percentage of shrinkage of the geopolymer-based ceramics after sintering at high temperatures was calculated by knowing the dimension before and after the sintering process. The JSM-6460LA model Scanning Electron Microscope (JEOL, Peabody, MS, USA) was utilized to examine the microstructural changes in the nepheline geopolymer-based ceramic. Prior to imaging, the samples were coated with a layer of gold using the JEOL JFC 1600 Auto Fine Coater (Peabody, MS, USA).

## 3 Results and Discussion

### 3.1 Physical and Mechanical Properties of Geopolymer-Based Ceramic

Figure 1 presents the changes in the density of unsintered and sintered geopolymer-based ceramics. It is observed that the sintering mechanism for geopolymer sintered at 1200 °C has the highest density (2.254 g/cm$^3$) compared to geopolymer sintered at 200 °C (1.396 g/cm$^3$) and unsintered geopolymer (1.349 g/cm$^3$). As the sintering temperature increase, the bulk density shows an increment. The sintering mechanism facilitates the increase in viscous flow as a result of the presence of a dissolved glassy phase. The highest density, indicating the completion of the densification process, was achieved at a temperature of 1200 °C. Upon sintering to 1200 °C, the water vapour liberates from the pores, of the geopolymer structure [11], lead to the pore formation. This outcome is due to the thermal influence, which facilitates the expansion of the geopolymer matrix [12]. As the sintering process takes place, the application of higher temperatures encourages the densification of kaolin geopolymer-based ceramics. This process involves the migration, rearrangement, and shrinkage of particles, which leads to changes in the bulk density of the ceramics. When the temperature is increased, the diffusion of grains near each other is enhanced, causing a decrease in the distance between alumina grains and promoting grain growth. Consequently, the ceramics become more compact and denser in structure [13, 14].

The sintering temperature plays a significant role in determining the flexural strength of the kaolin-based geopolymer. Figure 2 shows the trend of flexural strength for kaolin-based geopolymer when the sintering temperature increased. The flexural strength of geopolymer-based ceramics sintered at 1200 °C recorded the highest reading (54.932 MPa), while the unsintered geopolymer had the lowest flexural strength

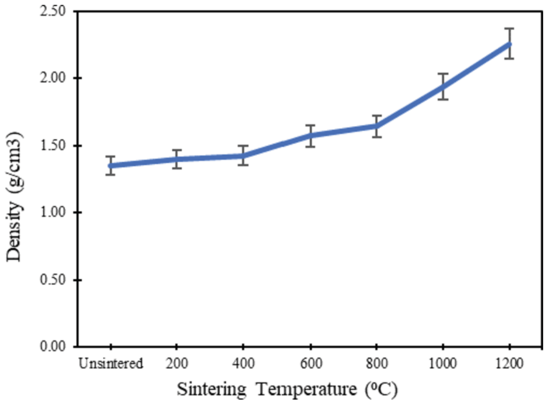

**Fig. 1.** Density of unsintered and sintered geopolymer-based nepheline ceramics at various temperatures.

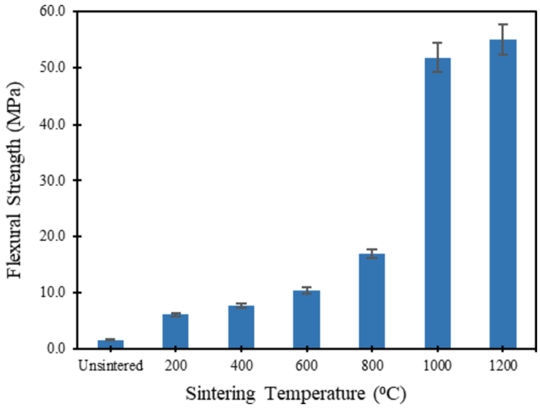

**Fig. 2.** Flexural strength of unsintered and sintered geopolymer-based nepheline ceramics at various temperatures.

(1.588 MPa). The geopolymer-based ceramics happen to increase a drastic strength when sintered at 1000 °C as the geopolymer matrices start to densify.

Overall, the flexural strength of geopolymer-based ceramics exhibited an upward trend as the exposure temperature increased. The observed enhancement in strength can be attributed to exothermic reactions that facilitated the continued geopolymerization of unreacted precursor materials. This led to the generation of additional reaction products, contributing to the overall strength improvement. Additionally, the densification of the geopolymer structure, facilitated by the solidifying melt and the development of refractory phases within thin geopolymers, contributed to enhanced flexural strength [18]. The elevated temperature played a crucial role in promoting the densification process within the geopolymer matrix, allowing it to fill voids and cracks, ultimately leading to improved strength. The formation of a more compact structure played a significant role in enhancing the overall strength performance [19].

The enhanced strength observed at 1200 °C can be attributed to the effective diffusion and coalescence of particles, forming large sintered area and a uniform geopolymer matrix. The exposure to elevated temperatures facilitated the sintering process, leading to increased strength by promoting stronger interparticle bonding and effectively mitigating thermal damage at high temperatures. The synergy of polymerization and sintering mechanisms likely contributes to the observed enhancement in geopolymer strength. By subjecting the sample to high temperatures, not only did it solidify, but it also underwent crystallization, leading to enhanced mechanical properties. The flexural strength of the materials improves as the sintering temperature increases, primarily due to the promotion of densification and the suppression of significant grain growth [20].

## 3.2 Microstructural Evolution of Geopolymer-Based Ceramic

Figure 3 illustrates the results of morphological analyses conducted on kaolin geopolymer ceramics subjected to high-temperature sintering.

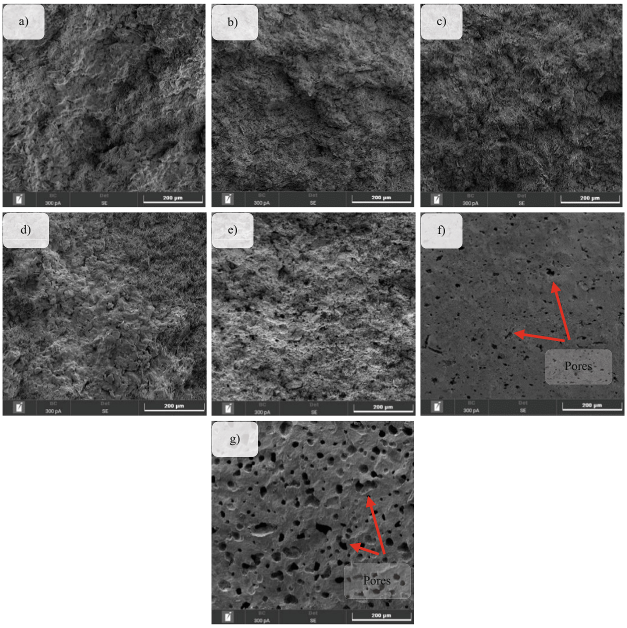

**Fig. 3.** SEM images of unsintered and sintered geopolymer-based nepheline ceramics at various temperatures.

SEM images of kaolin geopolymer ceramics sintered at temperatures between 200 °C and 400 °C, display the scanning image that closely resembles the unsintered samples.

These images depict a network of amorphous geopolymers with visible cracks, which may be associated with the mechanical strength tests or the evaporation of water during sintering.

Distinct morphological variations are observed when comparing the sintered samples at 800 °C, 1000 °C and 1200 °C with the unsintered geopolymers. These differences signify the crystallization of geopolymers beyond 800 °C, resulting in the elimination of surface irregularities and the formation of a crystalline structure. The transformation from an amorphous network to a crystalline in kaolin geopolymers thereby enhancing their resistance to high temperatures. Although the porosity remains relatively steady above 1000 °C, the open pores decrease due to the partial melting of the geopolymer matrix and the release of gases [21].

The presence of small pores indicates the occurrence of a phase transition from amorphous to crystalline. At higher temperatures, there is a notable expansion of pores as liquid water undergoes a transformation into vapour. This transformation applies pressure on the walls of the pores and creates connections between neighbouring pores, leading to the elimination of smaller pores.

The elevated sintering temperature contributes to the formation of a uniform and smooth microstructure. Densification occurs as fine particles are eliminated, leading to the entrapment of certain pores within the grains and the development of irregular pore sizes [22]. These phenomena arise from thermal expansion due to the release of residual water content, oxidation, and crystallization processes occurring within the geopolymer. Consequently, pore formation and the initiation of cracks are observed.

## 4 Conclusion

The preliminary results indicate that the sintering mechanism significantly influences the densification behavior and mechanical properties of nepheline geopolymer-based ceramics. Higher sintering temperatures led to enhanced densification, resulting in increased density and reduced porosity. The microstructural analysis revealed that sintering promoted grain growth, leading to the formation of denser and more interconnected microstructures. The presence of well-developed interparticle bonding contributed to the improved mechanical properties of the sintered ceramics.

The findings from this study contribute to the fundamental understanding of sintering processes in nepheline geopolymer ceramics, aiding in the development of optimized manufacturing techniques and improved mechanical properties. Ultimately, this knowledge can facilitate the broader utilization of nepheline geopolymer ceramics as sustainable materials in various engineering and construction applications.

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