Geopolymer Ceramic as Piezoelectric Materials: A Review

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Abstract. Diverse application for geopolymer so called inorganic polymer have been expanded as potential to continue growing at a realistic rate where the properties, processing tolerance and economical are comparable with the existing materials. An aluminosilicate inorganic polymer can be produced at low temperature under highly alkali conditions from a solid aluminosilicate and an alkali silicate solution. The conversion of amorphous to semi-crystalline behaviour of geopolymer into crystalline phases upon heating make the method be an alternate way in producing ceramic materials. For another application related to high temperature packaging and enclosure of electronic devices, piezoelectric behavior turn out to be important properties to the geopolymer ceramic materials. This paper summarize the review on the important research findings on the basic geopolymer systems, current knowledge of geopolymer ceramic, and outline potential piezoelectric effect on ceramic materials.

1. Introduction

The superior character of ceramic materials grow to a huge application in this modern industry. Recently, in the ceramics industry, much of the material becomes waste, and it is not recycled. Ceramics defined as an inorganic nonmetallic material comprises of metallic and nonmetallic elements bonded together with ionic and/or covalent bonds, are formed by a heating process. Typically, ceramics were labeled as refractory due to the high melting. Different type of ceramics originate with different properties, in general they are also high compression strength, high in modulus, low thermal conductivity, high hardness and chemically inert [1]. Besides, several factors were identified as important parameters that affect the properties and performance of ceramic materials such as material used, fabrication method and sintering profile. Thus, the use of geopolymer method in producing ceramic materials owning to the excellent mechanical properties with low production cost, short fabrication time, and increasing treat to the environment with the aim of emerging the application of the geopolymer technology.
Hardening at ambient temperatures, geopolymer that having amorphous to semi-crystalline phase contain a random tetrahedral network of silicon and aluminium atom [1]. In theory, any alkali and alkali earth cations can be used as the alkali component in the geopolymerization process. High alkali solutes, such as sodium silicate (Na$_2$SiO$_3$) and sodium hydroxide (NaOH) are integrated into source materials with high content of SiO$_2$ and Al$_2$O$_3$, yielding Si-O-Al-O bonds. The bonding between aluminosilicate and alkali solution expanded the application of geopolymer in many areas such as coatings, cements, composites and ceramic materials [2-4]. A unique feature of geopolymers is that sintering at high temperature transforms them into a crystalline ceramic phase. Iwahiro et al. reported that high temperature technique is no longer necessary by using geopolymerization process in order to develop materials of ceramic-like structure and properties [6]. Geopolymer give the potential possibilities to prepare inorganic bonds and building materials from the waste such as slag, fly ash and kaolinitic substances [7]. Ceramic wastes obtained from the industry in Malaysia has to be utilize to produce great application [8].

A new development of ceramic materials using geopolymer were produced by alkali activation of thermally activated aluminosilicate materials such as kaolin and metakaolin [4, 5]. Bell et al. reported that pollucite ceramic from the natural metakaolin-based geopolymer in the formation of pollucite (Cs$_2$O·Al$_2$O$_3$·4SiO$_2$) were possible to produce [6]. The pollucite crystallization gradually arises during sintering at temperatures in the range of 900°C to 1250°C [10]. While leucite ceramic derived from geopolymer were produced by Xie et al., (2010) providing a simple and low-cost approach with relatively high-strength and leucite-based glass compared with the conventional methods [50]. Despite the privilege on producing ceramic materials using geopolymer technology, being an aluminosilicates also could expected to be displayed in both thermal and electrical properties [9, 43]. Besides, piezoelectric effect received a great attention among the electrical properties due to their ability to produce electric current in response to mechanical stress. This work provides a review on the basic knowledge on geopolymer, geopolymer ceramic and remarks on the future trends and challenges for piezoelectric effect in the field of geopolymer system. Geopolymer ceramic also expected to show a promising performance on the piezoelectric effect.

2. Overview of Geopolymer System

The reaction between aluminosilicate sources with a highly concentrated alkali solution produces synthetic aluminosilicate materials, generally called geopolymer, also known as alkali activated aluminosilicates, hydroceramics, alkali-activated cement and alkali-bounded ceramics. In spite of those varieties of nomenclature, a more applicable term that describes the produced materials is ‘inorganic polymers’ [2,35-42]. According to Davidovits, geopolymer possesses amorphous to semi-crystalline phases, consist of three-dimensional tetrahedral silicate and aluminate bonded by oxygen atoms together with ion exchange properties. Porous structure are commonly found in geopolymer system, and water molecules along with the positive ion are trapped within the three-dimensional cross linked Si-O-Al structure [13,44-49]. The complex geopolymer structure involves of chains, three-dimensional networks and sheet-like made up of several Q unit types of connected SiO$_4$ and AlO$_4$ tetrahedra [14].

The structural can be designated depending on the Si/Al ratios, namely poly-sialate (-Si-O-Al-O-) (Si:Al = 1), poly-sialate-siloxo (-Si-O-Al-O-Si-O-) (Si:Al = 2), poly-sialate-disiloxo (-Si-O-Al-O-Si-O-Si-O-) (Si:Al = 3), and sialate links (Si:Al > 3) [10, 51-56]. De Silva et al., (2007) described that geopolymers that have poly (sialate-siloxo) and poly (sialate-disiloxo) structures are more rigid, more stable, and stronger than poly (sialate) structures. The geopolymer have the following empirical formula as describes by equation 2.1:

$$M_n[-(\text{Si-O}_2)_{z} - \text{Al-O}]_n \cdot w\text{H}_2\text{O}$$

(1)

Where m is the alkaline element, the symbol – indicates the presence of a bond, z is 1, 2, or 3, and n is the degree of polymerization.
The source material and alkali activator play a crucial role in the formation of geopolymer. The source material provides silica (Si) and alumina (Al) for reaction by an alkali activator solution. Wide variety of materials is presently being used in the geopolymerization process. Other than industrial by-product materials such as fly ash, Ground Granulated Blast Furnace Slag (GGBFS), red mud and rice husk ash, natural minerals such as kaolin and metakaolin could also be used [17,18]. Past research described that the strength of geopolymers is dependent on the nature of the source materials. The use of calcined source materials demonstrate a higher compressive strength compared to those formed from non-calcined materials [19-21]. Meanwhile, alkali activator is used to activate the aluminosilicate materials besides prompt the crystallization and precipitation of the silica and alumina species which present in the solution. The hydroxyl radical (OH-) in the mixture of alkali hydroxide will act as a reaction catalyst while the alkali metal (Na+) as a structural forming component and balance the negative framework carried by the tetrahedral aluminium [20]. The presence of alkali silicate solution in alkali reactant solution is essential, leads higher rate of reaction occur in which offering a better microstructure and strength properties.

![Figure 1. Process occurred during geopolymerization process [18].](image)

An exothermic reaction called geopolymerization is a chemical process that rapidly transforms partially or totally amorphous aluminosilicate sources into three-dimensional polymeric structure. The process is carried out through oligomers (dimer, trimer) which are the fundamental unit structures for three dimensional macromolecular structures. Generally, the process involves a number of simultaneous processes including dissolution, reorientation, and solidification as shown in figure 1 [18]. The dissolution of the solid aluminosilicate source releases small silicate and aluminate species into surrounding solution undergoes a rapid exchange and oligomerization reactions. The oligomeric were later undergoing a gelation process and delay the diffusive transport of the dissolved species. The gel structure remains to evolve and harden thus become a fully coordinated aluminosilicate network [11]. The extent of generation of both Si and Al were depending on few aspects: concentration of the alkaline solution, alkali metal cation in alkali solution, design of mixing and basic properties (e.g.,
structure and composition) of aluminosilicate raw materials [24]. The concentration of alkali solutions and the basic properties of the aluminosilicate raw materials are dominant.

Geopolymer received extensive attention in research and development due to outstanding properties such as excellent physical and mechanical properties, low energy consumption, and reducing greenhouse emissions during manufacturing process [25-27]. With all the advantages of geopolymer properties, the viability of forming ceramic materials using geopolymer technology has been verified by few researchers as geopolymer are a class of amorphous, aluminosilicate materials that harden hydrothermally at ambient temperatures and can be transformed into crystalline ceramics during heating [28, 29].

3. Geopolymer Ceramics

The unique composition of the geopolymer system with the help of geopolymerization reaction will improve the crystallization process as well as reducing the sintering temperature required, and with an appropriate processing method, geopolymer can be directly transformed into the final structural structural leucite (K2O·Al2O3·4SiO2), pollucite (Cs2O·Al2O3·4SiO2) or nepheline (Na2O·Al2O3·4SiO2), ceramic part of interest and practically design the chemical compositions of the final product [19]. Furthermore, the homogenous of the geopolymer system will influence the structural rearrangement during the change of amorphous to crystalline phase hence promote the nucleation and densification of the geopolymer. However, the difficulty of fabrication method of geopolymer ceramics for various applications requires the properties and performance required can be obtained in a reliable and cost-effective way. Both goals of appropriate reliability and are cost dependent on the impact of components composition as well as shape, size and surface finish requirement on fabrication routes and the parameters of the processing technique within this routes [20].

Previous work has generally concentrated on the microstructural and physical evolution on heating of as-cast geopolymers. The monolithic bodies typically induce cracking due to large capillary forces acting on capillary channels during drying by heating of as-cast, [31]. In the study by Peigang et al., (2013), a leucite ceramic disk was developed by sintering the geopolymer without first grinding it into a powder. The geopolymer was obtained by mixing metakaolin with potassium silicate solution. The geopolymer were cured at 70 °C for 48 hours, then sintered at different temperatures with 120 minutes of soaking time. In result, substantial amount of leucite was formed at a temperature of 800 °C. At heating rate of 2 °C/m minutes, the bullet-shaped together with the plated-shaped of leucite ceramic were observed without cracking by directly heating the geopolymer precursors [32, 59]. In another study by Peigang and Dechang, (2013), leucite ceramic directly derived from the geopolymer has developed. The crystallization, sintering and thermal expansion behaviors of a synthetic-metakaolin cesium-based geopolymer were studied. Based on crystal nucleation and growth mechanism, the amorphous Cs-geopolymer completely crystallized into pollucite on heating at 1200 °C for 2 hours and the ceramic products produced showed a low mechanical strength.

Meanwhile, the use of geopolymer precursors reported by Xie et al., (2010) offers a simple, low-cost approach to form relatively high-strength leucite based glass compared with the conventional methods. Leucite ceramic was fabricated by grinding the K2O·Al2O3·4SiO2·11H2O geopolymer into a powder, isotactic pressing, and sintering at 950 °C to 1200 °C. Samples heated at 1200 °C for 3 hours provide a maximum in density, fracture toughness, and biaxial flexure strength [60]. Kuenzel et al., (2013) studied the properties of metakaolin based geopolymer mortars that contained 50 % by weight of silica sand, exposed to temperature up to 1200 °C with 20 °C/minute of heating rate and 2 hours soaking time. The sodium aluminate crystalline phase was formed at above 800 °C and the amorphous phase of geopolymer was transformed to nepheline (NaAlSiO4) and carnegieite (NaAlSiO4) on heating at 900 °C. At 1000 °C, the mortar samples were transformed into polycrystalline nepheline/quartz ceramics [57, 58].

Common source material used for previous research in producing geopolymer ceramics is metakaolin which proved to have issues in consuming more energy for calcination thus producing a cost efficient final product. For this reason, our previous research groups were studied on the
development of kaolin based geopolymer ceramics. The optimum ratio of solid to liquid ratio [22], alkali activator ratio [61] and NaOH concentration [24] on the kaolin based geopolymer ceramic were studied. The production of lightweight properties also have been investigated by using Ultra High Molecular Weight Polyethylene (UHMWPE) [23]. Thus, the ability of using geopolymer technology in fabricating ceramic materials at low temperature create an innovative ceramic application due to the forming of various crystalline types dependent on the raw material used. Besides, the method selection and material used play important roles towards reducing energy consumption in fabrication of ceramics.

4. **Piezoelectric Effect on Ceramic Materials**

Piezoelectric phenomenon has been widely used in numerous applications such as vibration sensor, transducers, actuators, and frequency controller. Material with piezoelectric effect is crystalline material which develops an electrical charge subjected to mechanical stress were discovered by Curie brothers in 1880. The most common used piezoelectric materials are lead zirconate titanate, piezoelectric ceramic, or piezoceramic called PZT. Other than that, natural piezoelectric materials also exist include quartz, topaz, tourmaline, cane sugar and Rochelle salt [24]. Piezoelectric phenomenon were then realized to show a converse effect; a geometric strain/deformation proportional to an applied voltage. The illustration schematic diagram of direct and converse piezoelectric effects is shown in figure 2(a) and 2(b), respectively [30]. The electromechanical properties of piezoelectric ceramics are mainly influenced by the processing conditions. Among the important processes that effect the product characteristics and properties are preparation of powder, calcining and sintering.

![Figure 2. Schematic diagrams of direct and converse piezoelectric effect [29].](image)

The polarizing is the key technique in fabrication piezoelectric materials due to the randomness of the grains in as-prepared piezoelectric ceramics. Piezoelectric ceramics are normally polycrystalline that having a complex of randomly oriented tiny crystal and present as non-piezoelectric before poling [31]. With the absence of an electric field, the domains having a random orientation in which polarization aligned the domain in the direction of the electric field as shown in figure 3. The randomly oriented dipole will rotates from its original orientation towards a direction that minimizes the overall electrical and mechanical energy stored when subjected to mechanical stressed applied. The piezoelectric effect exhibits will be negligible as the rotation is not significantly change the
macroscopic net polarization of the materials [30]. Thus, electric polling is required for piezoelectric materials in which resulting in polarization to get oriented to the directions of poling field.

![Figure 3. Schematic diagram of poling process in piezoelectric process [32].](image)

A study on piezoelectric effect on geopolymer has conducted by previous researchers that quartz state could possibly contribute to electrical dipoles creation during mechanical stress, thus increase the piezoelectric activity. The piezoelectric effect generated by the charge imbalance and local dipoles under compressive stress by the migration within the network of interconnected pores of geopolymer. Another significant finding is the presence of sodium ion in charge balancing disposition that leads to piezoelectric effect [31]. During dehydration process of geopolymer, pores undergo a reduction of size while sodium ion and framework generate strong electrostatic interactions as a consequence of the reduced in dielectric screening of water, resulting inhibition of the cation migration. Indeed, geopolymers also do not need to be cut along particular directions like quartz crystals, neither to be polarized.

5. Conclusions
This study presents a compressive review on the development of geopolymer ceramics with the piezoelectric effect. The review helps in realizing the basics of geopolymer technology outcomes and scalability to model piezoelectric materials and later, with the improved understanding and matured technology, growing effort can be devote to develop geopolymer ceramic with the piezoelectric effect.

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