Development of Geopolymer Ceramic as a Potential Reinforcing Material in Solder Alloy: Short review

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Abstract. Nowadays, the consumption of lead-free solder has been widely used around the world since the utilization of SnPb solder has been banned and restricted by European Union. Variety of studies have been conducted by the researchers to find an alternative to replace the usage of SnPb such as SnCu, SAC, SnAg and etc. However, the development of plain lead-free solder was declared to provide low mechanical, thermal, and electrical properties in terms of interfacial intermetallic compound and wettability towards its solder joint compared to the traditionally monolithic SnPb solder alloy. Mostly, previous studies stated that addition of some additives such as ceramic particles (Si₃N₄, TiO₂, SiC, NiO and etc) may improves the solder joint reliability. At the same time, no major studies were done using geopolymer ceramic as reinforcing agent in plain matrix alloy. Therefore, this paper reviews the fabrication process of multiple geopolymer-based ceramic such as fly ash, kaolin, and slag as reinforcement in solder alloy. The development process includes the processing method of geopolymer ceramic and the characterization of geopolymer ceramic as reinforcing material consist of; i) chemical composition, and ii) phase identification.

1. Introduction

Soldering process becomes necessary in electronic packaging industries as one part of mechanically support and joint the components on the circuit board to assembly together and thus, providing electrical support by transmitting electrical connection within the solder joint between solder and circuit [1]. As solder will remain as primary assembly and interconnection technology for several period of time, the soldering process has been a fundamental aspect in realisation of all electronic products for a functioning lifespan [2].

Formerly, the soldering industries used Tin lead (SnPb) eutectic alloy solder material in the production of solder material for the past electronic devices. In fact, the used of SnPb eutectic alloy in soldering industries has successfully produced exclusive combination of chemical, physical, mechanical and electrical properties in producing good reliable solder alloy with reasonable price for many years [2], [3]. Unfortunately, the used of SnPb was reported to release toxicity and might be harmful to human’s health and environment if it is used for long-term usage [4]. As a result, the
legislation on restriction of the toxic SnPb by European Union (EU). Waste of Electrical, Electronic Equipment (WEEE) and RoHS (Restriction of Hazardous Substances) have strictly banned the usage due to health and environmentally concern since it might give negative impact to the human’s health and environment surrounding [3], [5].

Therefore, researchers find other alternatives in order to replace traditionally SnPb eutectic alloy with lead-free solder alloy. In soldering process, it is difficult to control the robustness of solder joint in electronic device. Moreover, the rapid modernity in electronic application is a great challenge faced by the electronic packaging industries as they need to keep in track with the emerging advances while struggling to stay within manufacturing cost limit [6]. addition of other reinforcement such as from ceramic based are widely acknowledged by the researchers as it can increase the mechanical and thermal properties of the solder joint. Recently, many investigation about Sn-0.7/Cu/Si$_3$N$_4$, Sn-0.7Cu-0.05Ni/TiO$_2$, Sn-3.0Ag-0.5Cu/SiC, Sn-3.0Ag-0.5Cu/ NiO and other related to this study has been observe to have various properties for the matrix alloy solder [7], [8]. Ceramic materials demonstrate enhanced properties in terms of microstructure, thermal properties, tensile strength, liquidous temperature and others under demanding conditions became one of the factors in choosing ceramic as reinforcement as it is reported to decompose β-Sn microstructure [9].

2. Processing of geopolymer for solder reinforcement

The history of alkaline activator was implemented since 1940s. In 1950s, Krivenko and Glukhovsky had invented the use of alkaline activator in cement system that comprised of alumino-silicate phases and calcium silicate hydrated (CSH) [10], [11]. Later on, geopolymer was introduced by J. Davidots in 1978 after 30 years of studies where the process occurs the combination of inorganic material such as silica and alumina with an alkaline activator [12]. The terms geopolymer came from an analogy of inorganic materials and organic polymers consist a group of mineral binders with some chemical composition similar to zeolite but in an amorphous structure. The mechanism of geopolymerization consist of dissolution, transportation or orientation, polycondensation and going through an exothermic process [13], [14]. The selection for solid raw material geopolymer-based could be natural minerals rich in silicone (Si) and aluminium (Al) such as clays, kaolinite, or also called waste materials like red mud, slags, fly ash etc. While the liquid for the alkaline activator usually consist of NaOH or KOH with a soluble alkali material such as Na$_2$SiO$_3$ [15]. Different fabrication methods in producing geopolymer ceramic gives different mechanical properties and relative densities. In processing geopolymer as reinforcement, this paper will focus more on mix design that provided optimum properties toward the geopolymer performance previously.

Nimiri et. al found that a maximum strength of metakaolin-based geopolymer produced when using NaOH solution as alkaline activator compare to KOH solution during thermal treatment at 700°C [12]. For fly ash geopolymer, a few researchers agreed that 12 M of NaOH solution as an alkaline activator gave the best performance in terms of mechanical properties [16]–[18]. Other than that, a concentration that is high than 12 M is difficult to be handle since it might be harmful when in contact to direct skin and a little bit costly due to the high usage of NaOH pallets [19]. In term of solid to liquid (S/L) and Na$_2$SiO$_3$/NaOH ratio, mostly researchers also approved that S/L ratio of 2.0 and 2.5 was the best mixture workability in providing high flexural strength [20], [21]. Basically, fly ash geopolymer and slag geopolymer can be cured at an ambient temperature. But due to the slow reactivity of kaolin geopolymer, a high cured temperature at 60°C was proposed by Heah et. al since it helps the geopolymerization to speed up [22], [23].

Commonly, kaolin refers to a family of kaolinitic clays that consist of 1:1-type clay mineral with one tetrahedral sheet and one octahedral sheet [11], [24]. The most important parameters that need to be emphasize in processing kaolin geopolymer is their low workability. Several researchers found out that S/L ratio of 1.0 with NaOH molarity of 8M gives the best result where it does not crack and were stable when exposed to the water for several days [22], [25]–[29]. A higher S/L ratio could lead to low workability and make it difficult during mixing process. Heah et. al concluded that a ratio of 0.32 $\text{Na}_2\text{SiO}_3$/NaOH could give an increment in strength [10], [14].
Slag is a non-toxic material which can be inserted as a raw material in geopolymer production for making high performance geopolymer products due to their high content of CaO as shown in Table 1 below [30]. Processing slag geopolymer might be tough since the CaO content is normally high causing low workability and fast setting during processing. Previously, slag was used as an additive to increase the reactivity or speed up the geopolymerization rate of base material such as fly ash class f and kaolin. Based on Aziz et al., 10 M concentration of NaOH with $Na_2SiO_3/NaOH$ ratio of 2.5 provided maximum compressive strength than low concentration of NaOH and alkaline activator. S/L ratio was the main contributor in order to produce good geopolymer properties. According to their studies, S/L ratio of 3.0 promotes higher compressive strength compared to S/L = 1.0 [31].

**Table 1.** Chemical compound based on XRF result of raw material for geopolymer-based [32]–[34].

| Compound | FAC Mass (%) | Kaolin Mass (%) | Slag Mass (%) |
|----------|--------------|-----------------|---------------|
| SiO2     | 21.23        | 54.5            | 33.56         |
| Al2O3    | 25.16        | 32.4            | 8.44          |
| Fe2O3    | 4.86         | 4.32            | -             |
| CaO      | 32.86        | -               | 47.49         |
| MgO      | 4.95         | -               | 4.74          |
| SO3      | 9.28         | -               | 4.14          |
| Na2O     | 0.43         | -               | 0.37          |
| K2O      | -            | 5.58            | -             |
| MnO3     | -            | 0.09            | 0.37          |
| TiO2     | 0.95         | 1.33            | 0.9           |
| ZrO2     | -            | 0.08            | -             |

3. Geopolymer Ceramic Powder

Geopolymer ceramic produced when the geopolymer was introduced to high temperature as high as 500°C and above depends on its material’s melting point. The removal of carbon in fly ash class f are also known as carbon burn-out (CBO) system. At temperature of 500°C, active carbon in fly ash class f was removed with a little change in inorganic portion. All carbon content was said to remove at thermal treatment range of 700°C. At this state, the crystalline structure already slightly appeared proved that ceramic-based already achieved [35]–[37] Nevertheless, only a few crystalline background on the XRD pattern produced at this state [38]. According to K. Zulkifli et al. and Heah et al., the crystalline phase started to increase when fly ash geopolymer and metakaolin geopolymer was subjected to 800°C. However, at temperature 1000°C, the amorphous phase dropped from 66% to 43% due to the crystallinity appearance of nepheline ($NaAlSiO_4$) and anorthoclase ($NaAlSi_3O_8$) [23], [39]. John Fox elucidated that fly ash class f started to crystalline starting at 700°C to 800°C while fly ash class c shows its appearance of crystalline phase after 800°C and becomes almost fully crystalline at 1000°. Due to the high percentage of amorphous phase in class f fly ash, John Fox concluded that higher amount of CaO content in class c fly ash contribute to high crystallization degree after being heated to 1000°C and above [38].
A study by Y. Liew found that the water absorption of kaolin geopolymer ceramic (KGC) reduced, produced maximum flexural strength when subjected to 1200°C temperature due to transformation of crystalline phases that help to decline water absorption [40], [41]. Sintering to a high temperature up to 1200°C resulted the presence of high nepheline phase which help the enhancement of mechanical strength due to the high amount of silicon-rich and polymerized species [42]. Previous study of an alkali activated shale-slag binder indicated that the amorphous content was gradually disappearing and transform into crystalline state starting from 800°C and above [43]. By sintering at high temperature, densification of structure will occur without stimulating the grain growth and therefore allow the refinement of microstructure into a dense ceramics [44].

4. Conclusion
This review paper summarized that it is possible to process numerous geopolymer powder as reinforcement in solder alloy. Although there aren’t many studies focused on this, it is possible to fabricate geopolymer reinforced solder alloy to characterize the difference in chemical composition either it might alter the solder joint performance or neither. Besides, the different geopolymer-based material also led to different phase diagram and chemical composition when subjected to a certain temperature. Depending on the raw material selection, geopolymer exhibit a wide variety of properties such in terms of compressive strength, low thermal conductivity, fire resistance, acid resistance and many other more. Geopolymer are considered as fine engineering materials with a broad application. Therefore, different characteristics and properties of geopolymer towards solder alloy can be explores further.

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