Synthesis of Kaolin Geopolymer as Ceramic Reinforcement in Lead-Free Solder

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Abstract. This paper elucidates the fabrication of kaolin geopolymer as the ceramic material. The kaolin geopolymer ceramic (KGC) was used as a new potential reinforcement in the lead-free solder. The fabrication of KGC was started through combination of alkaline solution of NaOH and Na2SiO3 reaction with kaolin material. The mixture of kaolin geopolymer were prepared and the homogenised mixture were curing for 24 hours. The kaolin geopolymer is sintered at 1200°C and then crushed to produces a fine kaolin geopolymer. The KGC were then mixed with lead-free solder through powder metallurgy technique. The elemental distribution in the KGC was investigated by using Synchrotron Micro-XRF. Meanwhile, the phase analysis involved in KGC and composite solder with addition of KGC were investigated as well.

1 INTRODUCTION

Geopolymers is a termed referring to inorganic polymer which having a polymeric framework of Si-O-Al, formed through geopolymerization process [1]. During the geopolymerization process, the dissolution of high aluminosilicate sources raw materials (for instance kaolin, fly ash and slag) with alkali activated solution (commonly sodium hydroxide or mixture between sodium hydroxide and sodium silicate) took placed [2]. Geopolymers posses excellent early and long term mechanical properties, low density and good resistance to fire. Moreover, geopolymer consist of amorphous to semi-crystalline phases with random tetrahedral network of Si and Al atoms. These phases can be transformed to crystalline phases upon sintering process at elevated temperature. Moreover, sintering the geopolymers will produce a sintered body that possess improved properties [3]. Kaolin is a natural clay with high content of Si and Al and potentially to be utilized as raw materials in the production of kaolin geopolymer. This kaolin geopolymer will be sintered to high temperature and yielding crystalline phases. As stated by Rovnanik et. al [4], the crystalline phases that will be formed with sodium-based geopolymers were nepheline and albite. The

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formation of nepheline and albite crystalline phases in KGC was important to the strengthening of the ceramic [5]. Meanwhile, for the potassium-based geopolymer, the crystalline that may formed prior to sintering process were leucite and kalsilite [5]. Therefore, with the ability of geopolymerization reactions in converting the amorphous and semi-crystalline phases of geopolymer to crystalline phases during the sintering process providing an alternative way to produce a ceramic. Conventionally, ceramic was formed using high sintering temperature up to 1600 °C [6]. However, the advantageous posses through geopolymerization reactions could reduce the sintering temperature in order to form ceramic materials.

In the solder system, ceramic particles was added to the solder matrix, forming composite solder. The main intention in forming composite solder was to alter and improve the properties of existing solder alloy. Some example of ceramics that had been used in the formation of composite solder are titanium dioxide [7-9], silicon carbide [10-12], zirconium dioxide [13] and silicon nitride[14]. However, there is limited study in investigates the reaction geopolymer ceramic in the solder alloy. Therefore, this paper focusing to produce KGC and the reactions that might occur by adding KGCs as reinforcement particles in solder alloy.

2 Experimental procedure

For this experiment, the experimental procedure was divided into two phases. The first phase was the fabrication of kaolin geopolymer ceramic (KGC). Then second phase involved the fabrication of composite solder with KGC as reinforcement particles.

The fabrication of KGC was preceded with formation of kaolin geopolymer through geopolymerization process. The investigated kaolin geopolymer samples were fabricated by using kaolin as raw materials and alkaline activator solution at a solid to liquid ratio of 1.0 was mixed with kaolin materials. The mixture of kaolin and alkaline activator solution were mixed homogeneously using a mechanical mixer, producing kaolin geopolymer paste. The kaolin geopolymer paste were then put in a HDPE mould and underwent curing process using an oven with curing temperature of 80 °C for one day. Then, the as-cured geopolymer paste was crushed with a mechanical crusher and sieved with a 150 micron siever, producing kaolin geopolymer powder. The kaolin geopolymer powder produced was used to fabricate KGC by using powder metallurgy technique. The compaction of kaolin geopolymer powder process was carried out using a stainless steel mold with a load of 4.5 tons. Then, the pellets of kaolin geopolymer which had been compacted, were sintered in a furnace at sintering temperature of 1200 °C and using heating rate of 5°C/min, producing pellets of KGC. Then, the pellets of KGC were pulverized using a mechanical crusher. The KGC powder underwent milling process using a planetary ball mill for 10 hours with speed of 450 rpm as to obtain smaller particle size of KGC powder. The ball to powder ratio (BPR) 10 :1 was used and producing powder with average particle size of ~18 μm.

The second phase involving fabricaton of composite solder with reinforcement particles of KGC. Powder metallurgy technique was used in fabricating the composite solder. Sn-3.0Ag-0.5 Cu (SAC305) lead free solder powder were used as base matrix material. Meanwhile, 1 wt. % of KGC was mixed with SAC305 lead free solder using a planetary mill for one hour at a speed of 200 rpm. Then the mixture was uniaxial compacted at a load of 4.5 tons. The sintering process was performed by using a technique of hybrid microwave sintering in a 50 Hz panasonic oven. A silicon carbide which functioned as susceptor material was used in sintered the samples.
The phase and crystallinity of KGC was characterized by using XRD 6000, Shidmazu diffractometer. X-Ray diffraction analysis was performed by using Xray tube Cu radiation operated at 40 kV and 30 mA. The data of XRD was collected at 2θ values in the range of 10° to 80° and scan rate of 2°/min with step size of 0.02°. Then, the results obtained were analysed using X-pert Highscore software. The elemental mapping analysis of KGC was carried by using synchrotron μ-XRF. Synchrotron μ-XRF was performed by using BL6b beamline at Synchrotron Light Research Institute (SLRI), Thailand. The utilization of synchrotron radiation and beam size of 30 x 30 μm² resulting in the mapping of elemental distribution for KGC. The data obtained from synchrotron μ-XRF was analyzed by using Pymca software. Meanwhile, to determine the existence of crystalline phase of KGC in the composite solder, X-ray diffraction analysis was used as well. The data of XRD was collected at 2θ values in the range of 20° to 80° and analyzed using X-pert Highscore software.

3 Results and discussions

3.1 Phase and elemental distributions

Fig.1 shows the XRD spectrum of kaolin geopolymer ceramics. According to Fig.1, there were appearance of nepheline peaks in the sample of KGC. The appearance of nepheline peaks was due to the sintering of kaolin geopolymer at high temperature which is 1200°C. The sintering process lead to the transformation of amorphous phase to fully crystalline phase [5]. The finding was consistent with studied done by Kuenzel et al. (2013) where crystalline phase of nepheline was observed as the geopolymer was heated above 900 °C.

![Fig. 1. XRD spectrum of kaolin geopolymer ceramics](image)

In order to obtain further investigations on the éléments presents in KGC, the elemental mapping using synchrotron μ-XRF was carried out. Fig.2 shows the elemental mapping results of kaolin geopolymer ceramic (KGC) obtained from synchrotron μ-XRF. The utilization of synchrotron μ-XRF enable to locate the distribution of major as well as light éléments in the KGC sample. In the elemental mapping as in Fig.2, blue,green and red colour indicates the low, medium and high intensity of Al, Si, K, Ti, Mn, Fe and Zr éléments distributed in the sample, respectively. According to Fig.2, Al and Si elements exhibit higher
distributions that are well distributed across the KGC sample. The elements of Al and Si were important during the geopolymerization process since it may contribute to the strengthening of the sample [15].

![Image of elemental mapping determined by Synchrotron Micro-XRF of kaolin geopolymer ceramic.](image)

**Fig. 2.** Image of elemental mapping determined by Synchrotron Micro-XRF of kaolin geopolymer ceramic.

### 3.2 Phase analysis in Sn-3.0Ag-0.5Cu with addition of kaolin geopolymer ceramic particles

Fig.3 depicts the XRD spectrum of SAC305 lead free solder and SAC305 with the addition of KGC. As shown in Fig.3 (a), the sample of SAC305 consists of three phases which are β-Sn, Cu₆Sn₅, and Ag₃Sn phases. Meanwhile in Fig.3(b), additional peaks were appeared at 2θ of 37.4°, 55.16° and 64.4°. The appearance of additional peaks in this sample was correlate with the nepheline phases originated from KGC added in the solder. This can be concludes that, SAC305 solder was successfully mixed with the KGC particles during the powder metallurgy method. Moreover, there is no new phase formed in the sample of SAC305 with addition of KGC. This can be inferred that, the reinforcement particles added are chemically inert with the matrix of SAC305 solder which then lead to no interaction during the solder melting. In the study done by Tsao et al. (2010) [16] also reported that there is no new phase formed in the sample with addition of titanium dioxide in Sn-3.5Ag-0.25Cu.
Fig. 3. XRD spectrum of (a) SAC305 and (b) SAC305 with addition of kaolin geopolymer ceramic.

4 Conclusions

As a conclusion, the results obtained proved that, the addition of kaolin geopolymer ceramic particles in the Sn-3.0Ag-0.5Cu (SAC305) lead free solder will not formed new phases in the solder system. Therefore, the used of kaolin geopolymer ceramic in the solder alloy could be a potential reinforcement particles in the formation of composite solder. In a future, the further characterization and analysis based on mechanical properties and thermal properties will be carried out.

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