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A review of geopolymer ceramic as a potential reinforcement material in solder alloys.

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Abstract. Nowadays, lead-free solder has been currently used in electronic packaging technology as part of soldering material. Since SnPb was detected to produce toxicity and might harm the consumers, the usage of Pb solder has been banned by WEE and RoHS. Therefore, various studies have been developed as alternatives to replace the usage of SnPb. Since lead-free solder might not perform as well as their traditional SnPb, researchers suggested to add some elemental reinforcement particles in matrix alloy. Previously, addition of ceramic reinforcement has been widely known in enhancing the properties of solder-substrate. This paper reviews about the presence studies of ceramic as solder reinforcement, the characteristics of geopolymer ceramic as potential solder reinforcement, and their properties in providing a superior solder joint. In this review, the characterization is divided into two stages; 1) characterization of geopolymers in terms of microstructural and crystallographic; 2) characterization of solder properties in terms of intermetallic layer growth (IMC), wettability, and its mechanical properties

1 Introduction

Soldering has been widely known in electronic packaging industries for electronic assembly where the solder materials serves as a conductive materials that bond together with the substrate through reflow soldering process at controlled temperature forming one or more electrical interconnection within the circuits [1], [2]. Few years ago, lead (Pb) solder was typically known as the most superior solder material in electronic packaging industries as it provide good soldering properties, reliability, and manufacturability with an affordable price [3]. In early 2000, Needleman found out that the usage of Pb solder might lead to health and environmental issues for a long period of time. The toxicity of Pb solder may cause harm if in direct contact with skin, mouth, nose, and eyes if it is not properly controlled [4]. Therefore, European Union's Waste in Electrical and Electronic Equipment (WEE) and Restriction of Hazardous Substances (RoHS) has banned and restricted the used of Pb solder in electronic industries [1], [3]. Since the usage of Pb has been restricted, National Electronics Manufacturing Initiatives (NEMI) has recommend to find other alternatives in replacing lead solder by inventing new lead-free solder material [5]. As a result, the elimination of Pb solder

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has lead to a great response by electronic consumers industries in EU markets and later from around the world such as China, Japan, South Korea, Turkey, and United States [3], [4].

According to Xu et. al. lead-free solder must be designed to form electrical and mechanical connection to maintain integrity through manufacturing process and service conditions [1]. Various types of lead-free solder has been fabricated to replace SnPb solder such as tin silver (SnAg), tin bismuth (SnBi), tin copper (SnCu), tin zinc (SnZn), tin rhenium (SnRe), tin zinc aluminium (SnZnAl), tin silver copper indium (SnAgCuIn), tin silver copper (SAC) and many others but it still beneath the wide range availability of SnPb [2, 6-10]. In manufacturing the new lead-free solder, its future properties must be comparable with it traditional SnPb solder material. Since Pb is widely known as the least expensive elements on earth, the overriding concerns is one of its replacement material will have an increasing intrinsic cost and whether the new lead-free solder can have equally or higher performance level to their traditional lead solder [11].

2 Solder reinforcement

Numerous researchs have been studied for the replacement of harmful SnPb that might have better potential than the previous one [12-15]. Researchers discovered that the performance of solder material becomes higher when some small amounts of particles element such as transition metal or rare earth elements were added in matrix alloy as reinforcement compared to plain matrix alloy [16-17].

Previously, most conducted research illuminated that addition of ceramic reinforcement produced high performance in terms of mechanical and electrical properties compare to plain matrix lead-free alloy [16-17]. Kotadia et. al. stated that the new elemental addition towards solder alloys should improves the wettability by reducing the surface tension of pure Sn, improves IMC thickness by suppressing intermetallic layer growth, and boost their mechanical and electrical properties such as hardness, shear strength, fatigue, creep, and thermal ageing [11]. 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 [18].

Ceramics are divided into several types which are nitride, oxide, silicate, and oxide ceramic were widely used in most studies. Addition of amorphous nanosilica (SiO_2) in Sn-0.7Cu could effectively provide optimum mechanical properties in terms of yield and tensile strength, and elastic modulus as the weight percentage of SiO_2 increased up to 1.5 wt.%. The following performances can be due to ceramic properties that usually is harder than the matrix alloy [19]. The wettability and the thickness of IMC layer has been reported to improve when 0.75 wt.% SiC was added in monolithic Sn-0.7Cu [20]. The wettability shows reduction in contact angle while the thickness of IMC layer changed from scallop shape to combination of planar and scallop shape. Chellvarajoo and Abdullah studied that the addition of NiO nanoparticles increased the melting point of SAC305 solder paste and gradually decrease the IMC formation after several addition [21].

The presence of diamond nanoparticles in crystalline phase in SAC305 was observed by Chellvarajoo et. al. as Fe_2NiO_4 was used as composite solder [22]. Before addition, the IMC shape was thick and formed as elongated scallop-shaped. After Fe_2NiO_4 nanoparticles were added, it reacts as catalyst to reduce Sn activities and therefore the nucleation and growths of both Cu_6Sn_5 and Cu_3Sn phase on Cu substrate were suppressed.

2.1 Geopolymer ceramic as reinforcement

Formerly, there were not much research conducted on geopolymer ceramic as reinforcement in monolithic solder alloy. Geopolymer was discovered by Professor Joseph Davidovits in early 1980s as one of concrete materials that gives higher mechanical and durability properties. Other studies found that geopolymer materials gives excellent resistance to chemical attack [23]. The word geopolymer came from geopolymerization process where geopolymerization occurs when chemical reaction between aluminosilicate sources and alkaline silicate solution was mixed together and cures or sinter at several temperature depends on its properties [23], [24]. Therefore, geopolymerisation happened when the raw materials rich in alumina and silica or pozzolanic materials dissolved in an alkaline activator solution [25-27]. Basically

In geopolymer industries, there are various mix design for each type of geopolymer. The alkaline activator solution that mostly used in geopolymer industries were the combination of Na_2SiO_3 , and NaOH or KOH but NaOH was mostly used due to their high properties. The rate of geopolymerization are depends on molarity of alkaline activator solution, ratio of $\text{Na}_2\text{SiO}_3/\text{NaOH}$, ratio of solid/liquid, and the time for curing or sintering per temperature [28-29]. Different types of raw materials geopolymer gives different optimum mix design to ensure their performances. In this paper, the structure of fly ash (FA), kaolin, and slag as raw material for geopolymer were reviewed.

Fly ash-based geopolymer was obtained by coal burning fire plants and have been widely used due to its good quality coal available and the low cost of producing this source [30]. Based on Mustafa et. al. the microstructure of fly ash appeared to be in the shape of spherical, hollow, and glassy which also known as cenospheres [31]. On the other hand, kaolin is reported to have plate-like structure with some needle-like shape [32]. The morphological of slag when observed under SEM as slag tend to have coarse and edgy shape compare to FA and kaolin just like Fig 1 below [33]. Slag can be classified into two group which is from ferrous and non-ferrous. Ferrous slag are such as blast furnace slag, iron steel slag, and ferroalloy slag while non-ferrous which is lead, nickel, cadmium, tin, copper, and zinc slag [34-35] With different microstructure of geopolymer ceramic reinforced monolithic matrix alloy, it might change the performance of solder joint.

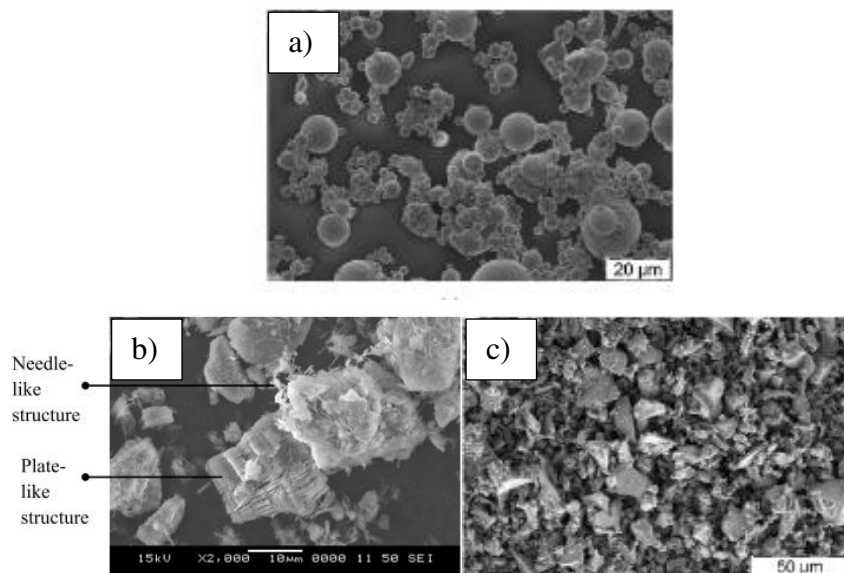


Fig. 1. SEM image of a) fly ash, b) kaolin, and c) slag [23], [24].

Geopolymer ceramic is produced by sintering the raw geopolymer at 600°C temperature as the crystalline phase already detected from the XRD result [36]. Based on Liew et al., the changed of geopolymer colour occurs for each sintering temperature. This is due to dehydration of moisture and phase transformation during sintering. The geopolymer ceramic reached their optimum strength when sintered it at 1200°C compare to 900°C, 1000°C, 1100°C and 1300°C. This is because the amorphous phase of geopolymer ceramic which tend to form brittle effect transformed into nepheline phases which is more stable [32]. In most research, the size of particles reinforcement needs to be smaller or the same size as the matrix alloy to ensure both materials pinned together during mixing process. The bigger size of particle reinforcement can lead to unhomogenous distribution mixing and causing agglomeration of specific material [37].

3 Properties of solder

3.1 Interfacial reactions between solder substrates

During soldering, intermetallic layer phase will grow due to interaction between solder alloy and solder substrate forming solder joint interface. However, rapid Cu dissolution between solder and Cu substrate caused high intermetallic layer thickness. Usually, high IMC thickness gives disadvantages to the solder joint as it is typically brittle and therefore contribute to failures joint [11]. In another words, the smaller the thickness of IMC layer growth, the higher performance provided since the initiation and propagation of cracks become less [38]. Figure 2 show the area of IMC layer produced during solder reflowed.

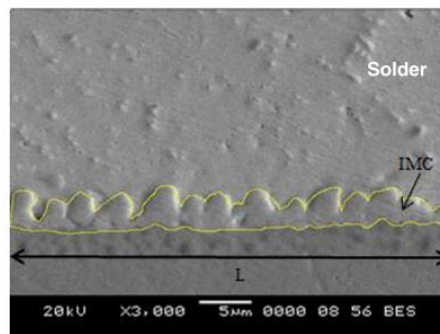


Fig. 2. Area of IMC phase at the solder joint interface [38].

A few studies clarified that longer period of time for reflow solder and dwell time gave rapid growth of IMC layer and reflect to the solderability performance of solder joint [21]. Mohd Salleh stated that the formation of intermetallic layer may leads to the formation of Kirkendall voids around the $\text{Cu}_6\text{Sn}_5/\text{Cu}$ substrate interface and therefore may deteriorate the reliability of the solder joint [39]. The presences of other reinforcing elements appear to limit the growth of neighbouring grains making the IMC layer more stable [6]. During reflow soldering, the mobility of reinforcing particles embed in matrix solder grain structure and substrate. Thus, the movement of Cu atoms will slowly reduced the formation of IMCs layer as it can act as an obstacle in the diffusion of Sn and Cu [16].

3.2 Wettability ability

In soldering, wettability refers to the ability of the molten solder to spread on the Cu substrate to create a solder joint layer when the solder is subjected to high temperature [18]. Wettability can be determined by the contact angle (θ°) between molten solder alloy and the Cu substrate such as Figure 4 below. The lower the contact angle, the greater wettability achieved by the solder on its application [40]. M. Ramli clarified that the contact angle of Sn-0.7Cu-0.05Ni decrease from 35.6° to a minimum 22.38° after added 1.0 wt.% of TiO_2 powder which indirectly improves the wettability of solder [38].

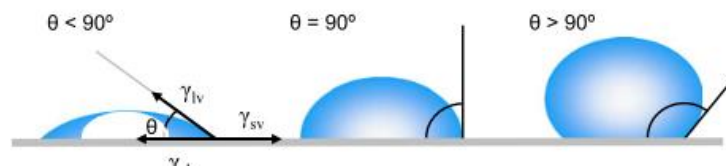


Fig. 3. Illustration of contact angle formed by molten solder alloy on a smooth solder-substrate [32].

Rate of wettability depends on the solder alloy proportion and surface properties of the substrates [41]. Dusek et. al. elucidate that there are differences in wettability between lead solder and lead-free solder where lead-free solder has a smaller wettability angle but is prone to obtain a tombstoning effect compared to lead solder. Tombstoning effects frequently take place in lead-free solder material when one side of the soldered component is not the same or rises during the remelting process [41].

3.3 Mechanical properties

Hardness and shear strength can be categorized as mechanical properties of solder alloys. Based on Salleh et. al., higher values of hardness (H_v) reduce deformation and cracks in the solder part. The microhardness values of Sn-Cu-Ni with TiO_2 became higher with an average of 16.64 H_v when sintered in a microwave oven compared to plain Sn-Cu-Ni [42]. This result shows that the addition of particle reinforcement might alter the hardness of the solder sample. Besides, the particle size of the reinforcement also plays an important role since the fine distributed size of particle reinforcement of solder influenced the hardness value due to the pinning effect [38, 42].

In terms of shear strength, high intermetallic layer growth contributes to low shear strength since the presence of a high intermetallic layer produces a brittle effect to the solder joint. Several researchers proved that the addition of In as reinforcement in Sn-Ag-Cu could modify their hardness, shear stress as well as their tensile strength [43].

4 Conclusion

Geopolymer ceramic reinforcement shows significant potential to be reinforced in a matrix alloy for the future. The different shape of geopolymer particles and chemical composition also might lead to different solder-joint performance. On the other hand, the high thermal might improve the solder joining. Therefore, further research is needed to investigate the suitable criteria for reinforcing particles in solder alloy.

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