Microstructure and Mechanical Properties of Geopolymer Ceramic Reinforced Sn-0.7Cu Solder

Z Nadiah Izzati¹, A Mustafa Al-Bakri¹, M A A Mohd Salleh¹, Romisuhani Ahmad¹, Nurul Aida Mohd Mortar¹ and Shamala Ramasamy¹

¹School of Materials Engineering, Universiti Malaysia Perlis (UniMAP), Kompleks Pusat Pengijian Jejawi 2, Taman Muhibbah, 02600, Jejawi, Arau, Perlis, Malaysia

E-mail: nadiahizzati.zulkifli@gmail.com

Abstract. This study investigates the effect of various geopolymer ceramic particles addition in matrix SnCu solder paste on its geopolymer ceramic’s microstructure and their mechanical properties after being reinforced in solder alloy. The composite solder was prepared by mechanically mixed 1.0 wt.% geopolymer ceramic powder into SnCu powder and compacted via powder metallurgy (PM) technique. Based on microhardness results, the presence of slag geopolymer shows an improvement after being reinforced with SnCu.

1. Introduction
Nowadays, the usage of lead-free solder has been widely used since the prohibition and restriction of toxic tin lead (SnPb) by the European Union (EU) Waste of Electrical, Electronic Equipment (WEEE) and RoHS (Restriction of Hazardous Substances) since it become hazardous and harmful to human health and environment [1-2]. Continuous development of lead-free solder on the electronic application such as Sn-0.7Cu solder were invented by the researchers to study the reliability concern to the solder joint properties. Sn-0.7Cu can be used as one of solder material’s alternative due to its excellence availability and was expected to have low cost production compare to traditionally monolithic SnPb [3-4].

Basically, plain lead-free solder matrix alloy was stated to have less properties compare to composite solder. Previous researcher agreed that some addition of alloying element such as rare earth element, transition metals and addition of micron or nanoparticles can improved the solderability of solder joint [3, 5]. Mostly, some researcher elucidates that addition of nanoparticles ceramic such as SiC, NiO, TiO₂ and many others obviously improved the properties of solder joint [6-8]. In current research, the addition of TiO₂ were able to suppress the interfacial intermetallic compound (IMC) formation by reducing the number of density and total area of Cu₆Sn₇ with multiple reflow cycles. Therefore, it can improve the mechanical strength such as hardness, shear strength, compressive strength as well as the decrement of wetting angle and IMC thickness [6, 7, 9-12].

The introducing of geopolymer ceramic as reinforcement particles was new to the solder industries. Geopolymer was discovered by Professor Joseph Davidots in early 1980s as one of concrete materials that gives higher mechanical and durability properties [13-19]. Geopolymer materials comes from polymerizations process between aluminosilicates materials such as alumina, silica, fly ash (FA), kaolin, slag, silica fume, red mud and etc. and alkaline activators such as NaOH and KOH based with a mixture of Na₂O and SiO₂ based (Na₂SiO₃) forming a paste or binder [20-26]. M Albitar et. al. [27] elucidates that geopolymer can withstand sulphate attack, corrosion and acid attack and therefore easy to get with
a low cost. The advantages of adding ceramic as reinforcement in metal matrix composite (MMCs) compared to unreinforced materials can improved the strength, reduced the density, controlled thermal expansion coefficient and improved stiffness [28].

2. Sample Preparation
The raw material used for the geopolymerization are FA, kaolin, and slag. Each raw material was produced with different mix design as shown in table 1 based on their optimum properties from previous research. The alkaline activator was prepared by adding NaOH solution to the $Na_2SiO_3$ solution and stirred for 5 minutes. Each raw material was weighted for 300g and mixed with alkaline activator homogenously for another 15 minutes. The geopolymer was poured in a 50 mm x 50 mm x 50 mm mould and wrapped before curing it for 3 days to allow geopolymerization. The solid geopolymer was then crushed before sintered it at 900°C and crushed again after sintering for a 38μm particles size.

| Table 1. Mix design of different raw material [29-32] |
|-----------------------------------------------|
| Molarity (M) | $Na_2SiO_3/NaOH$ | S/L | Curing temp (°C) |
| FAC          | 10               | 1.0 | 2.5 | 27         |
| Kaolin       | 8                | 0.32 | 1.0 | 80         |
| Slag         | 10               | 2.5 | 3.0 | 27         |

The geopolymer-reinforced composite solder was prepared by mechanically mixed 1.0 wt.% geopolymer particles into Sn-0.7Cu powder and compressed via powder metallurgy technique with 4 tonne pressure. The compacted green body were then sintered in a microwave oven at sintering temperature of 185°C for 3 minutes. The samples were mounted in a mixture of epoxy resin and hardener with a ratio of 1:1, grinded and polished until mirror surface. To produce a solder joint, about 0.1g of solder paste composite was then deposited on the cupper substrate and let it solder reflowed in a reflow solder oven up to 250°C.

3. Results and Discussions

3.1. Geopolymer ceramic powder

3.1.1. Microstructural
There are several criteria that must be satisfied for the selection of ceramic as reinforcement in manufacturing metal matrix composites (MMCs). Such criteria include size and shape of the reinforcement, density, melting temperature, thermal strength, and compatibility of the reinforcement with the matrix material [33]. The surface morphology of each geopolymer were identified using scanning electron microscope (SEM) image. Based on Mustafa et. al, the microstructure of raw fly ash appeared to be in the shape of spherical, hollow, and glassy which also known as cenospheres [34]. Kaolin is reported to have plate-like structure with some needle-like shape [35]. On the other hand, the morphological of slag when observed under SEM tend to have coarse and edgy shape compare to FA and kaolin [36-37].

Based on figure 1 (a), the micrographs show that the fly ash geopolymer has undergoes geopolymerization process since the reduction of cenospheres-shaped. However, only a few was not completely reacted with alkaline activator solution since there is some unreacted fly ash detected. During geopolymerization, there are 3 steps occur which are the dissolution of soluble species such as fly ash in the alkaline phase, followed by the reorganisation and diffusion of dissolved ions with formation of coagulated structure. The next step is the growth and condensation of these structures to form hydrated products [38-40].
3.2. Geopolymer reinforced SnCu solder

3.2.1. Hardness

The micro hardness test was operated using Vicker’s hardness test machine according to ASTM B933-09 standard. The hardness value of the solder matrix after the addition of geopolymer ceramic were tabulated in figure 2. Based on the result, SnCu/slag geopolymer shows the highest hardness up to 7.84 Hv compare to other composite solder. On the other hand, SnCu/kaolin geopolymer appeared as the lowest hardness which is 4.22 Hv which is nearly half than the hardness of SnCu/slag geopolymer. The controlled solder material which is the pure SnCu solder gave constant result (6.48 Hv) after being tested numerously under Vicker’s hardness which is slightly higher to the SnCu/fly ash geopolymer (6.42 Hv). This result was parallel to the compressive strength of the geopolymer ceramic of shows in table 2 below.

The higher the values of hardness, the lower the solder sample to deform and crack. Microstructural observations revealed that the distribution of reinforcing geopolymer ceramic particles along the grain boundaries of monolithic SnCu can be the main reason in providing high microhardness values [6]. Some researchers agreed that the formation of IMC and wettability influence the hardness of solder material [8]. According to the dispersion strengthening mechanism, homogenous distribution of reinforcing particle may lead to improvement of mechanical properties of the solder joint since the refinement of IMC formation. During indentation, the diamond indentation depth will either decrease or increase. Decreasing in indentation depth shows the sample’s strength, thus increasing hardness value due to the indenter was difficult to displace further depth. S. Chellvarajoo and M. Abdullah stated that when the addition of NiO nanoparticles increase, the grain size of the solder matrix decreased due to high viscosity and therefore increase the microhardness reading [8].
4. Conclusions
The addition of slag geopolymer to SnCu solder paste has a significant effect on the microhardness values. Results show that, slag geopolymer particles increased the hardness up to 7.84 Hv compare to monolithic SnCu. In addition, the hardness of geopolymer reinforced SnCu results was corresponding to the compressive strength of geopolymer itself where slag geopolymer tend to produce high strength compare to fly ash geopolymer and kaolin geopolymer. Therefore, future study need to be continued to investigate the criteria of reinforcer efficacy in MMCs.

Acknowledgements
This work was supported by Universiti Malaysia Perlis, Nihon Superior Co. Ltd and Institutional Links grant, ID 332397914, under the Newton-Ungku Omar Fund partnership. The grant is funded by the UK Department of Business, Energy and Industrial Strategy (BEIS) and Malaysia and delivered by the British Council.

References
[1] Amir M, Muhammad R, Ahmed N, Sadiq M and Waqas M 2017 J. Eng. Appl. Sci. 36 p 1–9
[2] Cheng S, Huang C M and Pecht M 2017 Microelectron. Reliab. 75 p 77–95
[3] Mohd Salleh M A A, Mustafa Al Bakri A M, Kamarudin H, Binhussain M, Zan Hazizi M h and Somidin F 2011 Phys. Procedia 22 p 299–304
[4] Zeng G, Xue S, Zhang L and Gao L 2011 J. Mater. Sci. Mater. Electron. 22 p 565–578
[5] Said R M, Mohd Salleh M A A, Ramli M I I, Saud N, Al Bakri Abdullah M M A and Sandu A V AIP Conf. Proc. 1835
[6] Ramli M I I, Saud N, Salleh M A A M, Derman M N and Said R M 2016 Microelectron. Reliab. 65 p 255–264
[7] Fathian Z, Maleki A and Niroumand B 2017 Ceram. Int. 43 p 5302–5310
[8] Chellvarajoo S and Abdullah M Z 2016 Jmade 90 p 499–507
[9] Mohd Salleh M A A, McDonald S D, Gourlay C M, Yasuda H and Nogita K 2016 Mater. Des. 108 p 418–428
[10] Kanlayasiri K and Kongchayasukawat R 2018 Trans. Nonferrous Met. Soc. China 28 p 1166–1175,
[11] Yakymovych A, Švec P, Orovcík L, Bajana O, Ipser H. 2018 J. Electron. Mater. 47 p 117-23.
[12] Nasir S M, Ismail K A, Shayfull Z and Shuaib N A 2013 Intl. Rev. Mech. Eng. 7 977-990
[13] Jaya N A, Al Bakri M M, Ghaazali C M, Binhussain M, Hussin K and Ahmad R 2016 Mater. Sci. Forum 857 p 405-411
[14] Fauzi A, Nuruddin M F, Malkawi A B and Abdullah M M 2016 Procedia Eng. 148 p 487-93
[15] Jaya N A, Abdullah M M, Ghazali C M, Hussain M, Hussin K and Ahmad R 2016 MATEC Web Conf. 78 p 01063
[16] Aziz I H, M M A B Abdullah, H C Yong, L Y Ming, K Hussin, A A Kadir and E A Azimi 2016 MATEC Web of Conferences EDP Sciences pp. 01023
[17] Aziz I H, M M A B Abdullah, M M Salleh, E A Azimi, J Chaiprapa and A V Sandu 2020 Constr. Build. Mater. 250 118720Shahedan N F, Al Bakri Abdullah M M, Mahmed N, Kusbiantoro A, Kamarudin H, Sandu A V and Naveed A 2019 Rev. Chim. (Bucharest) 70(8) 3027-3031
[18] W Wan Mastura, H Kamarudin, I Khairul Nizar, M M Al Bakri Abdullah and H Mohammed 2013 Adv. Mater. Res. Trans Tech Publ pp. 937-941
[19] Yahya Z, Abdullah M M, Hussin K, Ismail K N, Razak R A and Sandu A V 2015 Mail 8 p 2227-42
[20] Azimi E, M Abdullah, L Ming, H Yong, K Hussin and Aziz I H 2016 MATEC Web of Conferences EDP Sciences pp. 01090
[21] Aziz I H, M M A Al Bakri, H Cheng Yong, L Yun Ming, K Hussin and E A Azimi 2015 Key Engineering Materials Trans Tech Publ pp. 34-38Shahedan N F, Abdullah M M A B, Mahmed N, Kusbiantoro A and Bouissi A 2018 AIP Conf. Proc. 2030 020294
[22] Ibrahim W M W, Hussin K, Abdullah M M A B and Kadir A A 2017 Geopolymer lightweight bricks manufactured from fly ash and foaming agent In AIP Conference Proceedings 1835(1) 020048.Supriadi W, Bayuaji R, Burhan R Y and Fansuri H 2016 Mater. Sci. Forum 841 p 178-185
[23] Aziz I H, M M Al Bakri Abdullah, H C Yong, L Y Ming, K Hussin, A Surlevea and E A Azimi 2019 P. I. Mech. Eng. L-J Mat. 233 721-733
[24] Bayuaji R, Nuruddin M F, Francis S, Ekaputri J J, Junaedi S and Fansuri H 2015 Mater. Sci. Forum 803 p 49-57
[25] Albitar M, Ali M M, Visintin P and Drechsler M 2017 Constr. Build. Mater. 136 p 374-85
[26] Surappa M K 2003 Sadhana - Acad. Proc. Eng. Sci. 28 p 319–334
[27] Sofri L A, Abdullah M M, Hasan M R and Huang Y 2018 AIP Conf. Proc. 2030 020293
[28] Heah C Y, Kamarudin H, Al Bakri A M, Binhusain M, Luqman M, Nizar I K, Ruzaidi C M and Liew Y M 2011 Phys. Procedia 22 p 305-11
[29] Aziz I H, Abdullah M M, Yong H C, Ming L Y, Panias D and Sakkas K 2017 IOP Conf. Ser. Mater. Sci. Eng. 209 p 012040
[30] Al Bakri Abdullah M M 2013 Rev. Chim-Bucharest 64 p 382–387
[31] Cui C, Shen Y, Meng F and Kang S 2000 J. Mater. Sci. Technol. 16 p 619–626
[32] Mustafa A M 2011 J. Eng. Technol. Res. 3 p 44–49
[33] Liew Y M, Heah C Y, Li L Y, Jaya N A, Abdullah M M, Tan S J and Hussin K 2017 Constr. Build. Mater. 156 p 9-18
[34] Yazdi M A, Liebscher M, Hempel S, Yang J and Mechtcherine V 2018 Constr. Build. Mater. 191 p 330-41
[35] Aziz I H, M M A B Abdullah, C-Y Heah and Y-M Liew 2019 Adv. Cem. Res. 1-11
[36] Alonso S and Palomo A 2001 Cem. Concr. Res. 31 p 25-30
[37] Azimi E, M M Al Bakri Abdullah, L Y Ming, H C Yong, K Hussin and Aziz I H 2016 Rev. Adv. Mater. Sci. 44 warid
[38] Shahedan N F, Abdullah M M A B, Ghazali C M R, Binhusain M, Al Husaini M, Hussin K and Ramasamy S 2016 MATEC Web Conf. IConGDM 78 01069
[39] Ibrahim W, K Hussin, M Abdullah, A Kadir and L Deraman 2017 AIP Conference Proceedings AIP Publishing LLC pp. 020011
[40] Gain A K, Chan Y C and Yung W K 2011 Microelectron. Reliab. 51 p 975-84