PAPER • **OPEN ACCESS**

Characterization of geopolymer ceramic reinforced Sn-0.7Cu composite solder: Effect of milling time and speed.

To cite this article: Marliza Mostapha *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **701** 012016

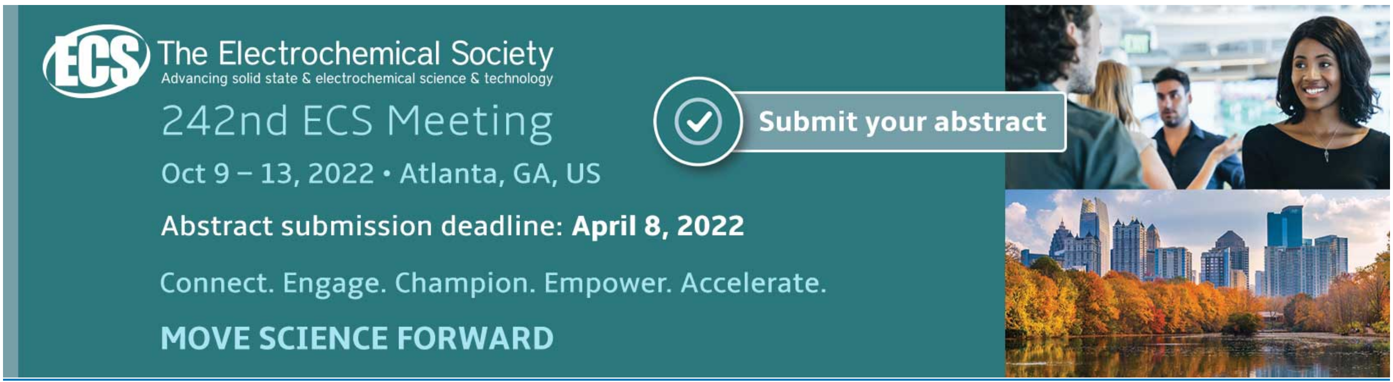
View the article online for updates and enhancements.

You may also like

- Fabrication, sinterabilty and characterization of non-colored and colored geopolymers with improved properties
S E Abo Sawan, M F Zawrah, R M Khattab et al.

- Study on the effect of emulsifiers on the pore structures of geopolymer prepared by emulsion templating
Zhuangzhuang Wang and Duyou Lu

- Development of Geopolymer Ceramic as a Potential Reinforcing Material in Solder Alloy: Short review
Nadiah 'Izzati Zulkifli, Mohd Mustafa Al Bakri Abdullah, Mohd Arif Anuar Mohd Salleh et al.



This content was downloaded from IP address 103.86.131.118 on 15/03/2022 at 04:49

IOP Conf. Series: Materials Science and Engineering **701** (2019) 012016 doi:10.1088/1757-899X/701/1/012016

Characterization of geopolymer ceramic reinforced Sn-0.7Cu composite solder: Effect of milling time and speed.

Marliza Mostapha[1,2*], *Mohd Arif Anuar* Mohd Salleh[1,2], *Mohd Mustafa Al Bakri* Abdullah[1,2], *Maiteli* Thiaghu[2], *Nur Syahirah* Mohamad Zaimi[1,2] and *Romisuhani* Ahmad[1,3]

[1]Centre of Excellence Geopolymer and Green Technology, School of Materials Engineering, Universiti Malaysia Perlis (UniMAP), Taman Muhibbah 02600, Jejawi, Arau, Perlis, Malaysia
[2]School of Materials Engineering, Universiti Malaysia Perlis (UniMAP), 01000 Jejawi, Perlis, Malaysia
[3]Faculty of Engineering Technology, Uniciti Alam Campus, Universiti Malaysia Perlis (UniMAP), Perlis, Malaysia

Abstract. The effect on the addition of optimally ball milled kaolin and fly ash geopolymer ceramic as reinforcement on the morphology and electrical properties of Sn-0.7Cu composite solder were explored. Geopolymer ceramics from fly ash class F and kaolin were prepared using geopolymer technology, milled at various speed and time. 1.0 wt. % of each sample were used to form composite solder via microwave sintered through powder metallurgy method. Structural characterization via SEM reveals that kaolin geopolymer ceramics has nano-sized subangular powder particles with larger amount of open porosity compared to fly ash geopolymer ceramics when milled at optimum speed and time. Four Point Probe test results showed a decreasing trend of electrical resistivity for kaolin geopolymer ceramics as the milling speeds and times increased. Overall, the results compared to electrical resistivity of other composite solder with various typical ceramic reinforcement additions, proves that kaolin geopolymer ceramics reinforcement to be the best option so far in term of morphology, electrical properties and its sustainable manufacturability.

1 Introduction

Geopolymers is an inorganic polymer that formed at low temperature, normally under 100 °C [1,2] and contain amorphous to semi-crystalline crystal structure which can be converted to crystalline ceramic phase through sintering [3]. Compared to conventional ceramic production, via sol gel or hydrothermal techniques, this method of producing ceramic through geopolymer starting material has improvised the material strength [4]. In geopolymer process, there is no release of bounded carbon dioxide since this process does not involve any calcination of calcium carbonate and the process does not need for facilities

* Corresponding author: marliza@unimap.edu.my

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

such as extreme high temperature kilns, with large outlay on fuel, plants and equipment. The mechanical properties of this geopolymer ceramic are similar to those of industrial ceramics. The availability of suitable raw materials from appropriate geological resources also utilization from industrial waste product as raw material can be considered as an advantage of this new ceramic production technology [5,6].

Fly ash and kaolin geopolymer besides its plus point of vast availability, its good processing workability, and improved mechanical properties in final product is the cause why is being studied for its potential application as ceramic precursor [4]. Nano-geopolymer on the other hand portrays quality of forming thermally-stable high-strength and close to final shape structures at room temperature. Also, nanometres sized crystallite novel amorphous and crystalline materials are produced. High-energy ball milling becomes handy for particle size reduction (comminution) down to the nanometres scale which corresponds to increased reactivity of nanostructured powders promoted by higher surface area [7]. Among high-energy ball mills; the planetary ball mill is a mechanically simple, low cost, versatile device for efficient grinding and suitable for any class of materials, and a larger quantity of powder can be produced easily [8]. According to several reference papers, particle size reduction prior to milling not just increases the reactivity, but also proved to contribute increases in compressive strength of the geopolymers [9–13]. Therefore, the feasibility of forming nanostructured ceramics from geopolymer and its effect on mechanical properties has been widely demonstrated, hitherto there are limited literatures available on incorporation of nano-sized kaolin and fly ash geopolymer ceramics into composite solder [14-17]. Little work has been conducted to investigate the comparative effect of addition of these ceramic reinforcements to the electrical properties of solder.

In this paper, two types of geopolymer ceramics which from fly ash and kaolin were milled for optimal time and speed. Both of this geopolymer ceramics were characterized in aspect of particle size and surface morphologies. The electrical resistivity of composite solder incorporated with two different types of geopolymer ceramics has been investigated by mechanical mixing of 1wt% of respective ceramic reinforcement with Sn-0.7Cu solder, compacted via powder metallurgy technique, microwave sintered and tested via four-point probe method. The SEM analysis is used to correlate the particles morphology, shape, size and porosity presence with the electrical resistivity of composite solder.

2 Experimental procedure

In this research, fly ash and kaolin were used as raw materials to form geopolymer ceramic. Fly ash supplied by Manjung Power Plant, Perak and kaolin supplied by Associated Kaolin Industries Sdn Bhd, Malaysia. The element composition of as-received fly ash and kaolin were given in Table 1. Sodium hydroxide (NaOH) pellet with 99% purity supplied by Formsoda Plastic Corporation, Taiwan and Sodium Silicate (Na_2SiO_3) solution supplied by South Pacific Chemical Industries Sdn Bhd, Malaysia. Distilled water used throughout the research. Sn0.7Cu were used as base lead-free solder supplied by Nihon Superior Co. Ltd., Japan.

Table 1. Elemental composition of as-received fly ash and kaolin.

Element (%)	Si	Al	Fe	Ca	K	Ti	Loss on ignition
Fly ash	25.8	14.6	5.3	2.9	1.4	1.3	0.6
Kaolin	26.23	14.71	4.60	-	5.03	1.36	0.99

NaOH solution with 12 M were used to prepare alkaline activator with Na_2SiO_3 /NaOH ratio of 0.24:1. The mixture of kaolin or fly ash, and alkaline activator with ratio of 1:1 were mixed together for a few minutes and cured at 80 ˚C for 24 hours. Kaolin and fly ash geopolymer were crushed and sieved into 150 µm and compacted at 5 tonnes for 5 minutes. The sample were heated at 1200 ˚C for 3 hours soaking time. Then, sample were crushed and sieved into 43 µm before ball milling process according to the parameter set up showed in Table 2 using stainless steel balls at 10:1 ball-to-powder ratio in dry environment. Monolithic Sn0.7Cu solder and its composites were prepared by powder metallurgy method through microwave sintered by mixed 1 wt.% of milled geopolymer ceramics with Sn0.7Cu solder in airtight container via planetary mill for 1 hour at 200 rpm. The sample were compacted in 12 mm diameter mould at 4.5 tonnes for 5 minutes and sintered through microwave sintering at 185 ˚C under ambient condition in 800W, 50Hz Panasonic microwave oven. The particle size of fly ash and kaolin geopolymer ceramic before and after milling were obtained using a Malvern particle size analyser. Scanning electron microscope (SEM) were used to study the changes on surface morphology before and after milled and four-point probe were used to analysed the electrical resistivity of monolithic Sn 0.7Cu and its composites.

Table 2. The ball milling parameter.

No.	Raw materials	Milling time (hours)	Milling speed (rpm)
1	Fly-ash geopolymer ceramic (FAGC)	5	0, 100, 300, 500
2	Kaolin geopolymer ceramic (KGC)		
3	Fly-ash geopolymer ceramic (FAGC)	0, 5, 7, 10	*optimum speed
4	Kaolin geopolymer ceramic (KGC)		

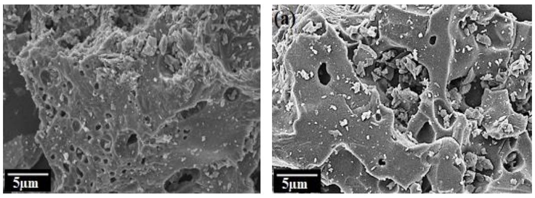
* Depend on the optimum speed that produced the smallest particle size.

3 Results and Discussions

3.1 Particle size and surface morphologies

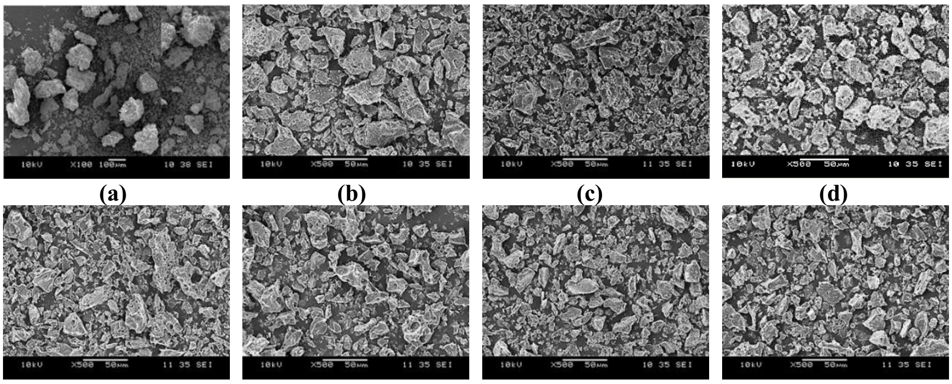
The surface morphologies of fly-ash geopolymer ceramic (FGC) before and after milling are shown in Fig. 1 (a) and Fig. 2 (a, b, c, d), and kaolin geopolymer ceramics (KGC) before and after milling are shown in Fig. 1 (b) and Fig. 2 (e, f, g, h), respectively. It can be seen that FGC surface contain large amount of open micropores, irregular and nearly rounded shape whereas KGC surface contain small amount of open micropores that actually larger in size compare to FGC, irregular and angular in shape. The presence of many large particles in FGC powder before milling are shown in Fig. 2 (a). Once the milling speed increases, the reduction of FGC particle sizes also happens from average size of 43 µm down to 7.5 µm. The irregular and nearly rounded shape of FGC starts to become more angular, smaller and nearly in the same size.

Similarly, the same pattern was observed in KGC powder where the powder starts to decrease in size from average size of 43 µm down to 7 µm. The process of particle size reduction causes the morphological changes of FGC and KGC and this can be clearly observed in the SEM micrograph shown in Fig. 2. The plastic deformation started to increase as the milling speed increases and in turn increased the changes in particle morphologies. Further increase in milling speed lowered the ability of powder to withstand the deformation without fracturing [18,19]. Consequently, smaller particle size was formed as tabulated in Table 3. Thus, with the optimal speed of 500rpm that was observed and found to have the lowest particle size, the geopolymer ceramic powder further milled for 7 and 10 h at 500rpm to investigate the optimal milling time in particle size reduction.



(a) **(b)**

Fig. 1. Surface morphologies of **(a)** FGC before milling and **(b)** KGC before milling.



(a) **(b)** **(c)** **(d)**

(e) **(f)** **(g)** **(h)**

Fig. 2.Surface morphologies of geopolymer ceramic powders milled at 5 hours for different milling speed **(a)** FGC at 0 rpm, **(b)** FGC at 100 rpm, **(c)** FGC at 300 rpm, **(d)** FGC at 500 rpm, **(e)** KGC at 0 rpm, **(f)** KGC at 100 rpm, **(g)** KGC at 300 rpm and **(h)** KGC at 500 rpm.

Table 3. Particle size of KGC and FGC with different milling speed for 5 hours.

Speed of milling (rpm)	Particle size (KGC)	Particle size (FGC)
0	43 µm	43 µm
100	23 µm	29 µm
300	13 µm	16 µm
500	7 µm	7.5 µm

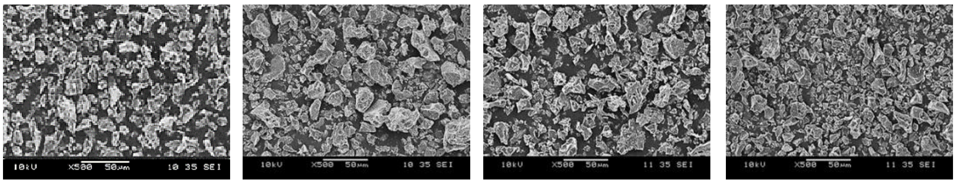
The effect of milling time on the particle size of FGC and KGC powder at 500 rpm was tabulated in Table 4. Increase of ball milling duration contributes to particle fineness indicating the extent of breakdown of structure [20]. When the milling time was increased up to 7 hours, particle size of FGC powder was drastically decreased while particle size of KGC was slightly decreased compared to FGC. This shows that FGC experienced major deformation and lost their strength to withstand the fracture effect that took place after 5 hours of milling. Fig. 5. (a) shows that the particle size become smaller and more angular for KGC powder while Fig. 5 (c) shows that there were small changes in size and shape of KGC powder compared to KGC powder milled for 5 hours. Further increased in milling time to 10 hours, particle size of FGC was increased significantly. This aligned with the increased in particle size shown in Fig. 5 (b). The FGC particles happen to be in cold-welding stage when milled for 10 hours.

During the process of milling, the geopolymer ceramic powder is subjected to three important process that are severe plastic deformation that leads to change in the size of particles, fracturing effect that cause the breaks of particles into smaller size and cold-welding process where this process cause the re-joining of the particles and thus increases the size of the particles [19]. For independent ceramic powder, the average particle size tends to be increased upon attaining steady-state equilibrium when balance between welding rates is achieved after milling for certain time interval [8]. At this point, the smaller FGC particles can withstand the deformation without further fracture and re-welded into larger particles [9].

In contrast, particle size of KGC was continue decreased. Reduction in size of particles larger than average and propagation through smaller particle cluster of trashes smaller than average results in finer particle size of KGC at this stage in agreement with Rao et al.'s work [20]. After 10 hours of milling time at 500 rpm, the KGC powder became sub angularly shaped with coarse surface morphology as shown in Fig. 5 (d) .

Table 4. Particle size of KGC and FGC with different milling time at 500 rpm.

Milling time (hours)	Particle size (KGC)	Particle size (FGC)
0	43 µm	43 µm
5	7 µm	7.5 µm
7	5.5 µm	0.97 µm
10	0.88 µm	6 µm



(a) **(b)** **(c)** **(d)**

Fig. 3.Surface morphologies of geopolymer ceramic powders milled at 500 rpm for different milling time **(a)** FGC at 7 hours, **(b)** FGC at 10 hours, **(c)** KGC at 7 hours**, (d)** KGC at 10 hours.

3.2 Electrical resistivity of Sn0.7Cu solder composites

Good electrical conductivity is a key criterion for a solder which serves its function as electrical interconnection which allows current conduction through it. Thus, a low electrical resistivity of the material is expected for effective functioning of an electronic device. Presence, shape, volume, size and type of reinforcement and matrix are found to be a few factors that affect electrical resistivity of a composite material [21-26]. In this research, three of the factors; presence, type and size of reinforcement definitely affected the electrical resistivity of the composite solder.

Fig. 4 shows the effect on electrical resistivity of the composites solder when FGC and KGC prepared by milling at different time and speed were used as reinforcement material to form composite solder. Base on Fig. 4. (a), the lowest electrical resistivity was from composite solder incorporated with 1wt% of KGC milled at 500 rpm. A slightly similar low electrical resistivity value of 8.82 µΩ.cm obtained for composite solder incorporated with 1wt% of FGC powder milled at 500 rpm indicating 500 rpm as the optimal milling speed for the reinforcement material. Preceding the study in finding the optimal milling time, the result as shown in on Fig. 4. (b) suggests, 7 hours as the optimal milling time for FGC powder as the resistivity value tend to increase when the powder milled for period of time longer than 7 hours, whereas 10 hours is found to be the optimal milling time for KGC powder upon achieving the lowest electrical resistivity value of 2.78 µΩ.cm. These results do not just directly relate to the size of the reinforcement, but also the volume of porosity in the reinforcement material itself [22].

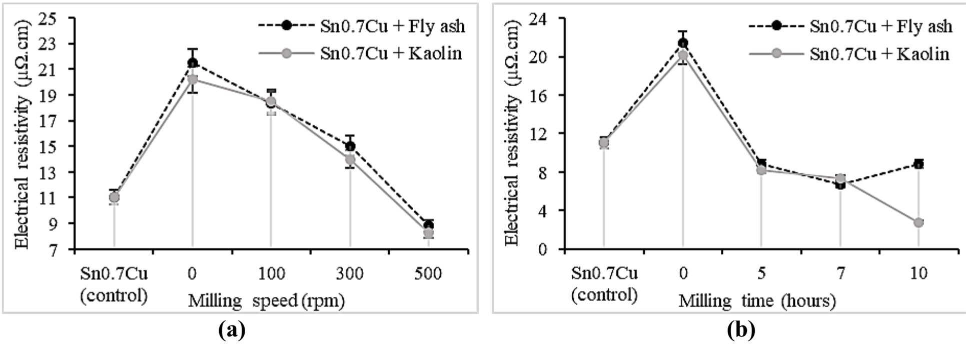
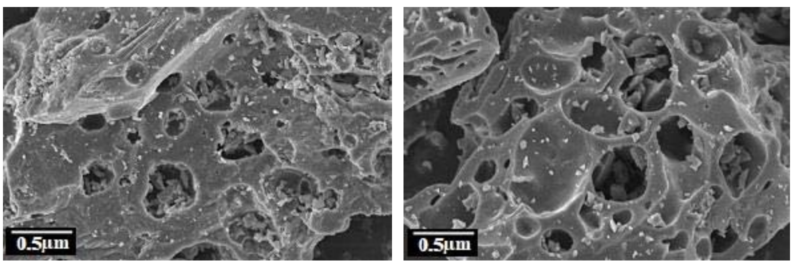


Fig. 4. Effect on the electrical resistivity of Sn0.7Cu composites solder with different **(a)** milling speed and **(b)** milling time.

Fig. 5 shows the comparison between presence of porosity in KGC particles and FGC particles. This micro porosity present in respective geopolymer ceramic particles largely influence the homogeneity of matrix and reinforcement as the porosity allows better bonding between reinforcing particles and solder thus enhancing the overall uniform distribution [27].

Based on Fig. 5, the obvious effect of porosity to electrical resistivity can be understood. Although practically in most cases, porosity is found to have very little effect on conductivity, but theoretically presence of open porosity found to increase the conductivity. This is possible if the electronically conducting species are adsorbed onto the surface of pores, proving additional conduction paths also said to act as active site provider [28]. Also, as per Hall Petch relationship, grain size increase causes removal of high resistance grain boundaries. This is the factor which contributes to electronic conduction of ceramics. KGC surface morphology as in Fig. 5. (b) shows a large number of open pores compared to FGC where this in turn became a path for above phenomenon to happened and consequently decreased the electrical resistivity of the composites solder. This finding is aligned with the trend that can be observed in Fig. 4. (b).

Due to above mentioned factor, many nano-sized porous ceramics have been used as reinforcement material in composite solder. Even so, addition of kaolin and fly ash geopolymer ceramic is found to be most effective not just in term of electrical resistivity, but also in term of sustainability. Production of geopolymer ceramic is a simple, low cost process that requires very low temperature sintering for ceramic formation compared to production of typical ceramics.



(a) (b)

Fig. 5. **(a)** FGC at 7 hours and **(b)** KGC milled at 10 hours.

4 Conclusion

In conclusions, the milling speed and the milling times are two predominant parameters that affects the size of particles and morphological changes in geopolymer ceramics. The optimal milling speed to produce nano-sized powder particles for both KGC and FGC is found to be 500 rpm, with different optimal milling time of 10 hours for KGC and 7 hours for FGC at the optimal milling speed. KGC is found to be composed of sub angular structured, irregularly shaped particles with larger amount of open porosity initiating from its plate like structure compared to FGC that have lesser particle size reduction, shape irregularity and open porosity which is the factors that support the resulting larger electrical resistivity of FGC compared to KGC in composite solder. Overall, comparing and considering the performance of KGC and FGC also other typical ceramics in aspect of morphology, electrical properties and sustainability, KGC is found be the best ceramic reinforcement for composite solder.

Acknowledgements

This work was supported by an 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. The authors acknowledge the financial support through the Ministry of Education (MoE), Malaysia, under the Fundamental Research Grant Scheme (9003-00612) (FRGS/1/2017/TK10/UNIMAP/03/2).

References

1. Davidovits, J. J. Ceram. Sci. Technol. 8, 335–350 (2017).
2. Azimi. E.A, M. M. A. B. Abdullah, Y.M. Liew, C.Y. Heah., K. Hussin, I.H. Aziz, Matec Web of Conferences, 78, (2016)
3. Jaya, N. A. et al. Key Eng. Mater. 700, 3–11 (2016).
4. Jaya, N. A. et al. Appl. Mech. Mater. 754–755, 152–156 (2015).
5. Al Bakri, A. M. M. et al. Key Eng. Mater. 594–595, 575–580 (2013).
6. I. H. Aziz, M. M. A. B. Abdullah, C.Y. Heah, and Y.M. Liew, D.Panias, K.Sakkas. IOP Conference Series : Materials Science and Engineering, 209,(2017).
7. Abdullah, M. M. A. B., Ming, L. Y., Yong, H. C. & Tahir, M. F. M. Clay-Based Materials in Geopolymer Technology. in Cement Based Materials (InTech, 2018). doi:10.5772/intechopen.74438
8. Broseghini, M. et al. J. Eur. Ceram. Soc. 36, 2205–2212 (2016).
9. Sharma, P., Sharma, S. & Khanduja, D. Mater. Manuf. Process. 30, 1370–1376 (2015).
10. Assaedi, H., Shaikh, F. U. A. & Low, I. M. J. Asian Ceram. Soc. 4, 19–28 (2016).
11. Assi, L. N., Eddie Deaver, E. & Ziehl, P. Constr. Build. Mater. 167, 372–380 (2018).
12. Zhang, P. *et al*. *Appl. Sci.* **9**, (2019).
13. Zhang, L. & Tu, K. N. *Mater. Sci. Eng. R Reports* **82**, 1–32 (2014).
14. N.M. Nasir, N. Saud, M.N. Deraman, A.A.M. Salleh, M.I.I. Ramli, and R.M. Said, Materials Science Forum. **803** (2015)
15. M.I.I. Ramli, M.S.S. Yusof, M.A.A.M. Salleh, R.M. Said, and K. Nogita, Solid State Phenomena. **273** (2018)

16. F. Somidin, H. Maeno, M.M. Salleh, X.Q. Tran, S.D. McDonald, S. Matsumura, and K. Nogita, Materials Characterization. **138** (2018)
17. M.I.I. Ramli, M.A.A.M. Salleh, F.A.M. Sobri, P. Narayanan, K. Sweatman, and K. Nogita, Journal of Materials Science: Materials in Electronics. **30**(4) (2019)
18. Biyik, S. & Aydin, M. Acta Phys. Pol. A 127, 1255–1260 (2015).
19. Indexed, S., Vasamsetti, S., Dumpala, L. & Subbarao, V. V. 9, 1579–1589 (2018).
20. Rao, J., Narayanaswami, P. & Prasad, K. Int. J. Eng. Sci. Technol. 2, 284–299 (2010).
21. Allemang, R., De Clerck, J., Niezrecki, C. & Wicks, Conf. Proc. Soc. Exp. Mech. Ser. 45, 577–617 (2014).
22. M.A.A.M. Salleh, M.H. Hazizi, M.A.B.M. Abdullah, N. Noriman, R. Mayapan, and Z.A. Ahmad, Advanced Materials Research. **626** (2012)
23. K. Nogita, M.A.A.M. Salleh, S. Smith, Y. Q Wu, S.D. McDonald, A.G.A. Razak, T. Akaiwa, and T. Nishimura, 2017 International Conference on Electronics Packaging (ICEP). (2017)
24. K. Nogita, M.A.A.M. Salleh, E. Tanaka, G. Zeng, S.D. McDonald, and S. Matsumura, JOM. **68**(11) (2016)
25. M.A.A.M. Salleh, A.M.M.A. Bakri, F. Somidin, and H. Kamarudin, International Review of Mechanical Engineering. (2013)
26. M.A.A.M. Salleh, M.M.A.B. Abdullah, F. Somidin, A.V. Sandu, N. Saud, K. Hussin, S.D. McDonald, and K. Nogita, Revista de Chimie **64**(7) (2013)
27. Babaghorbani, P., Nai, S. M. L. & Gupta, M. J. Mater. Sci. Mater. Electron. 20, 571–576 (2009).
28. Dorey, R. A. (William Andrew, 2011).