Effect of Hybrid Particles of Nano-Micron Sizes on Electrical Transport of Silver-Epoxy Composites

Muhammad Zulkarnain, M.Sobron Lubis

Abstract: Electrically Conductive Adhesive's (ECAs) mainly consist of a polymer as matrix and binder to filler as conductor. The performance of electrical properties in ECAs strongly depends on the concentration (volume fraction) of the metal filler, morphology of the filler, filler distribution within the polymer matrix, the interaction between filler surface, and the contact resistance between adjacent particles. In this work, the characteristics of electrical transport of ECA's systems on Ag-Epoxy composites are containing either micro or nano fillers are studied by the electric static network theory using FEM. The hybrid system will be formed for considering influence electrical transport properties. The spherical Silver (Ag) used 80 nm and 2 µm in diameter in this study. Two particles chain forming electrically transport with some number of particles and reach a conductivity value of ECA's. The result indicated that nano size silver usually lower its conductivity than micron size. During simulation, it has been shown the correlation between nano and micron size demonstrated a increasing in the electrical transport when the hybrid system performed.

Keywords: Conductivity, Electric Field, Current Density, Spherical Particle.

I. INTRODUCTION

Electrically Conductive Adhesive (ECA) has become a trend toward using high performance conductive adhesives to replace alloy solder because of its good electrical conductivity, low cost, extendibility to fine pitch interconnect and environmental friendly [1-5]. ECAs mainly consist of polymer as a matrix and conductive filler. The performance of electrical properties in ECAs strongly depends on the concentration (volume fraction) of the metal filler, morphology of the filler, filler distribution within the polymer matrix, the interaction between filler surface, and the contact resistance between adjacent particles [6]. The most important filler characteristics are particle size, size distribution, specific surface area and particle shape, while the main matrix property is stiffness [7].

One of the most important properties of electrical conductivity of ECA's is their morphology correlate to electrical properties which is especially interesting near the percolation threshold.

The morphological analysis is to describe spatial particles distribution in the ECA's system. New technology on nano materials has attracted many researches to investigate the properties and application of silver nano particles in ECA. However, due to high surface energy of nano particles, the particles tend to agglomerate and hence create non-homogenize particle distribution, which results in no cluster particle construction in composite to electrical transport [5,6]. Traditionally, micron size conductive adhesive extendibility interconnecting is greater achieved than nano size [7].

As implied above, as the filling of ECA with hybrid can make high performance of the interfacial interactions of particles, there is a need among the research communities in ECA to know the electrical transport. Hybrid system has shown by the electric network percolation theory, that the isotropic conductive adhesive (ICA) using a bimodal distribution of metal fillers is expected to have an increase in electrical properties [9]. Fu, (1999) has demonstrated computer modelling that was decrease of 10% in the percolation volume percentage when the volume percentage of the nano particles in the ICA increases from 2.76 to 13.8%. In nano-micro mixtures, nano particles occupy interstitial positions to improve particle-particle contact for conductivity. Hsien-Hsuen Lee, (2005) [10] developed substituting nano-sized silver particles for micro-sized particles. One conceivable advantage of using nano-particles is to avoid the settling problem encountered in some micro-sized Ag particle systems. When the composite consisting of micro-sized silver flake and PVAc is near the percolation threshold, the addition of a small amount of nano-sized silver colloids would help to establish the conductive path and hence lower the resistivity. In the hybrid system is still less information which is focus on the development of the electron transport system and many studies still need to be explored.

Therefore in this study 3-D computer simulation was carried out to predict the electrical conductivity behavior of 80 nm size and 2 µm of silver nano particle in polymer matrix. The present work was focused on electrical network transport of nano, micron and hybrid system at particle level with particular distance. The existing electrical conductivity model in 3-D was used to predict the electrical conductivity. The model was developed based on the spherical shape of Ag filler which produces electrical density typical on particles.

Revised Manuscript Received on December 05, 2019.
Muhammad Zulkarnain, Universiti Kuala Lumpur Malaysian Institute of Marine Engineering Technology (UniKL MIMET), 32200 Lumut, Perak, Malaysia.
M.Sobron Lubis, Department of Mechanical Engineering, Engineering Faculty, University of Tarumanagara, Jakarta, Indonesia

II. ELECTRICAL THEORETICAL

Electrical Conductivity

The schematic of apparatus used to measure electrical resistivity is illustrated in Fig. 1. The current flows (I) through a conductor when the potential difference between its ends is (V) and resistance (R) can be defined.

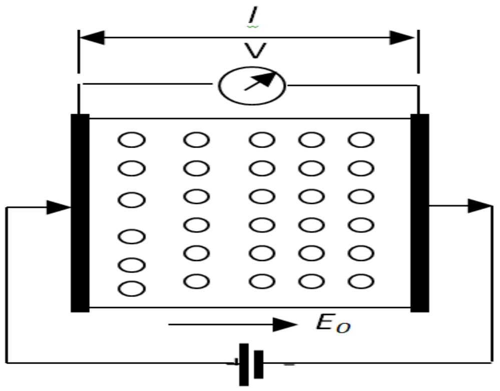

Fig. 1 Schematic of apparatus used to measure electrical resistivity

Current density flow of composite in x-axis direction is described in Fig. 2.

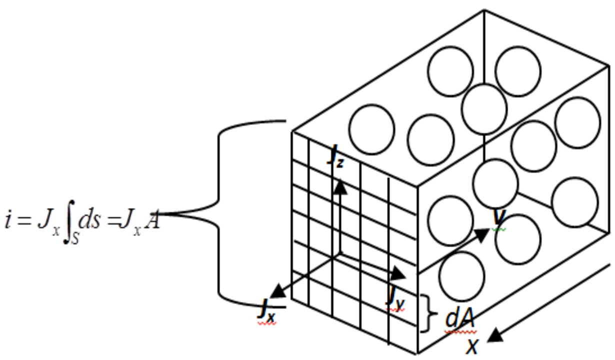

Fig. 2 The current flow of composite in x-axis direction

The current flow through a surface composite identified as current density (J). The differential current, di flowing through a surface element ds at which the current density (J) exists, is di = **J**.ds amperes. A constant current through any cross-section is

$$i = \int_S (a_x J_x)\cdot(a_x ds) = J_x \int_S ds = \sum_{n=1}^{m} J_x A \qquad (1)$$

Where A is cross section of composite. The resistivity ρ value of composite can be found by resistance of the volume composite through which the current is passing.

$$\sum_n^m R_i = \frac{\sum_{n=1}^{m} \Phi_i}{\sum_{n=1}^{m} J_{ix} A} \qquad (2)$$

Where Φ_x is voltage by loading input. Then resistivity ρ can be expressed

$$\bar{\rho}_x = \frac{\sum_n^m R_i A}{\sum_n^m l} \qquad (3)$$

Where l is distance between the two points at which the voltage is measured. Electrical conductivity (σ) of the composite can be applied by equation (4).

$$\bar{\sigma}_x = \frac{1}{\bar{\rho}_x} \qquad (4)$$

Finally, electrical conductivity (σ) for each volume fraction composition can be determined. This model can take into account any matrix and filler conductivity.

Conductivity calculation based on FEM Model

A constitutive FEA model to describe the potential distribution of ECA's model is given by equation (5)

$$[\Phi] = [E]\{\hat{r}\} \qquad (5)$$

The total electric field (E) due to a quantity of point charges, m_q, is simply the superposition of the contribution of each individual point charge, $\sum_{n=1}^{mq} E_i$ correspond to each distance $\sum_{n=1}^{mq} \hat{r}_i$. The potential total at field point P is the sum distance (8)

$$\Phi = \begin{bmatrix} E_{xx} & E_{xy} & E_{xz} \\ E_{yx} & E_{yy} & E_{yz} \\ E_{zx} & E_{zx} & E_{zz} \end{bmatrix} \begin{Bmatrix} \hat{r}_{xx} \\ \hat{r}_{yy} \\ \hat{r}_{zz} \end{Bmatrix}$$

Finite Element Method Model

C.1 Generate the structure model

In order to understand the electrical conductive adhesive behaviour, the finite element analysis were done by developing three dimensional ECA's composite system model and the performed current density (J) responses were analyzed by using the finite element software, ANSYS™. The simulation data is obtained from the previous work [Tee *et.al.* 2007] and the ECA's composite system was constructed by the components of particles metal conductor and a cubical matrix box as insulator. The model composition was arranged in the form particle and matrix by part partitioning method. Three models of particle were selected to capture electrical transport of composite which are nano size, micron size and hybrid system model. The complete FEM model consists are following steps:

1. Generate the structure ECA's which is metal/dielectric composite with known dimension parameters.
2. Defined material properties with electrical properties considering.

3. Create the meshing discrete on the structure model.
4. Define voltage loading between two ends of a path to generate electrical potential.
5. Analysing and testing of the electron transport through the ECA's model.
6. Determinate relation particular distance and electrical transport analysis.

C.1.1 Nano and Micron Size model

Two particle conductive conditions was modeled in this part; the first one is the particle conducting path where contact performed physically touching each other to exhibit continuous network through ECA's composite for both nano and micron size. The second was the conductive particles were in minimum distance and electron can jump the gap between the particles and creating current flow. These electrical conductivity mechanism models structure as shown in Fig. 4(a) and 4(b). Permittivity for each material denoted by ε_1 and ε_2 are conductor and isolator, respectively. The spherical Silver (Ag) used 80 nm and 2 µm in diameter in this study.

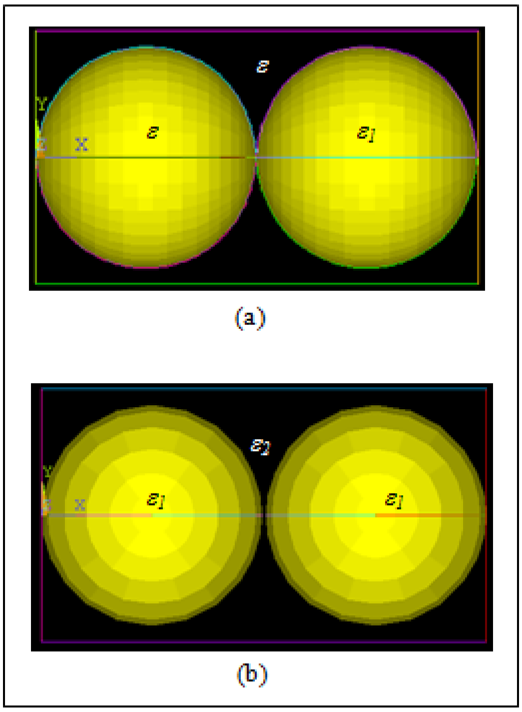

Fig. 4 Examples of two conductive Ag spherical composite structures prepared by (a) two particles contact model and (b) interstice of two spherical model

C.1.2 Hybrid System

The hybrid system is used when the composite consist the micro size and adding of small amount of nano size to improve the electrical performance. In this model are used 2 particles of micron-sized with a distance of 1 nm with the addition of some particles of nano size. The formation of a conduction path of hybrid system was modeled by ANSYS^{TM} and illustrated in Fig. 5.

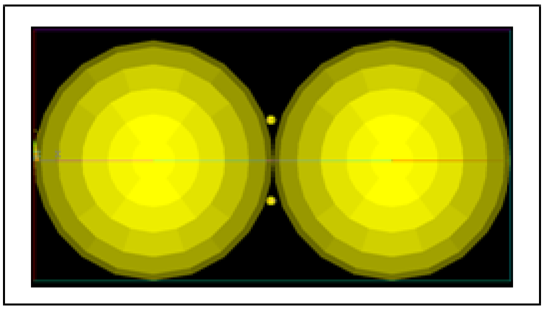

Fig. 5 Examples of hybrid composite structures prepared by two particles of micron size with addition of 2 particles of nano size.

C.2 Meshing and boundary condition

All the models were discretizated by tetrahedral electric solid mesh element to create the finite element model. The elements used for the model is 10 nodes 3D linear tetrahedral elements (solid232) for particles filler and matrix in electrical conductivity 3D model. The element was considered as maximum node number and allowable in curve shape. The three dimensional finite element models are shown in Fig. 6a. The models were mapped meshed by using the size control technique with same size at edge side. A surface area voltage was applied to couple surfaces area on the model as shown in Fig. 6b by applying 2 difference level voltage in the end of path that is automatically generate electrical potential in axial direction along x-direction. For boundary condition, no displacement constraints were applied on the model as axial and transverse forces are not applicable to the model.

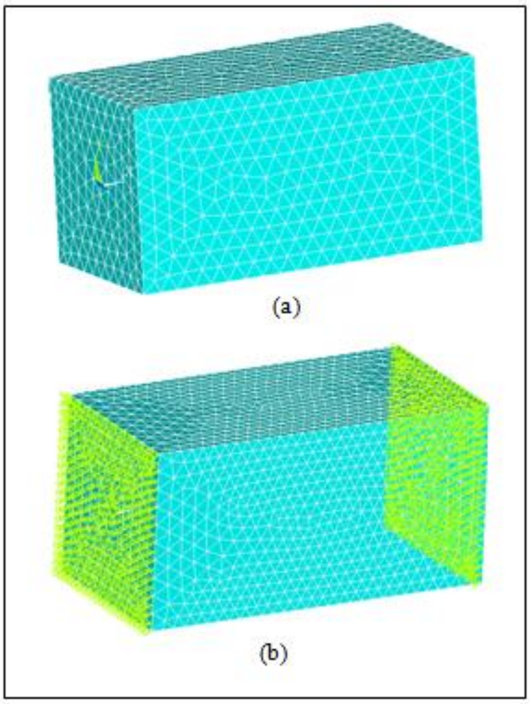

Fig. 6 Mesh detail of the model (a) Meshing of tetrahedral elements (b) Voltage coupled loading on 2 surfaces

C.3 Material properties

The material properties detail is referred from our previous study [Tee *et.al.* 2007]. The electrical conductivity of Ag and epoxy are 6.301 x 10^5 S/cm and 2.13 x 10^{-6} S/cm, respectively. Ag and Epoxy have electrical resistivity of 1.59 x 10^{-6} Ω-cm and 4.71x 10^5 Ω-cm, respectively. The data have been used in the FEM model.

III. RESULTS AND DISCUSSION

The information from finite element model enable fully capture important phenomenon due to conductivity effect on composite. The composite conductance is strongly influenced by the electrical transport by particle structure properties. These phenomenons can be described by common term used in percolation threshold.

Finite Element Method Model

A.1 Nano size phenomenon

The visualization model of the complete 3D structures will perform tunnel junction transport and contact particle transport of the composite structures. This section will carry out the transportation of the electron on contact and the some particular distance of particle. Figure 10 presences the current density vector distribution of nano particle on varying distance. Contact particle transport shown in Fig. 7 (a). It was found that total current density through the composite occurred at 14402.419 J/cm^2. The tunnel junction by separating two conductors with a thin distance insulator is showed in Fig. 7 (b) which is 1 nm of partial distance. The total current density through the composite is 6326.7794 J/cm^2. The much different clearly seen in electrical transport, when the particles are start fully contact and electrically interconnected and its effectively increasing electrical. This electrical transport behaviour correlates between the electrical conductivity with particle distribution for ECA applications.

A.2 Micron Size phenomenon

The model performed current density as shown in Figure 8. Two various particles transport are analysed to observed current density for the study electron transport in x-axis. Fig. 8a illustrates that high current density are observed around particle contact. It found that total current density through the composite occurred at 614.70196 J/cm^2. While Fig. 8(b) shown the total current density at 1 nm spatial distance is 383.627318 J/cm^2.

A.3 Hybrid System

Fig. 9 shows the current density vector distribution phenomenon of hybrid system. The structure was consisting bimodal distribution of silver fillers which is of 2 nano particles and 2 micron. Fig. 9(a) shows current density vector distribution which is 2 particles nano at outside of electrical path among micron particles. Particle nano distribution has shown a decrease in current density which is lower than micron performance its self. This phenomenon occurs because of current density was distributed divergently; causing the current flow through the composite was decrease. The total current density through the composite was 320.38486 J/cm^2. While Figure 9(b) illustrated the nano particles making interconnect network between nano and micron particles. Nano particles were arranged among micron position to generate electrical path on micron particle. The results of current density were observed that increase with addition nano particle as illustrated in Figure 9(b). The total current density through the composite shown at 395.32314 J/cm^2 that was higher than single micron particle.

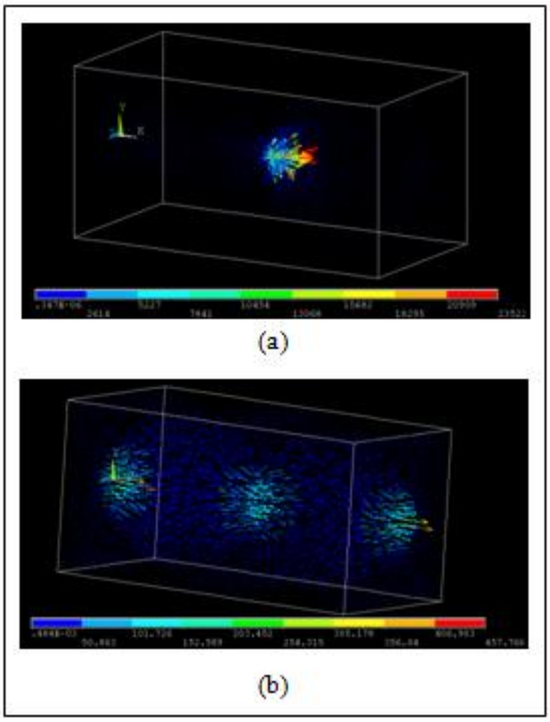

Fig. 7 Current density vector distribution of nano size (a). Contact particle (b). Partial distance of 1 nm

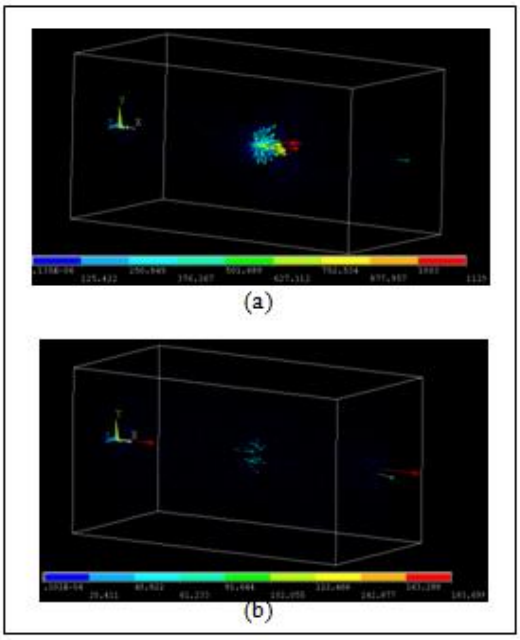

Fig. 8 Current density vector distribution of micron size (a) Contact particle (b). Partial distance of 1 nm

Retrieval Number: B9027129219/2019©BEIESP
DOI: 10.35940/ijitee.B9027.129219

Published By:
Blue Eyes Intelligence Engineering
& Sciences Publication

ISSN: 2278-3075, Volume-9 Issue-2, December 2019

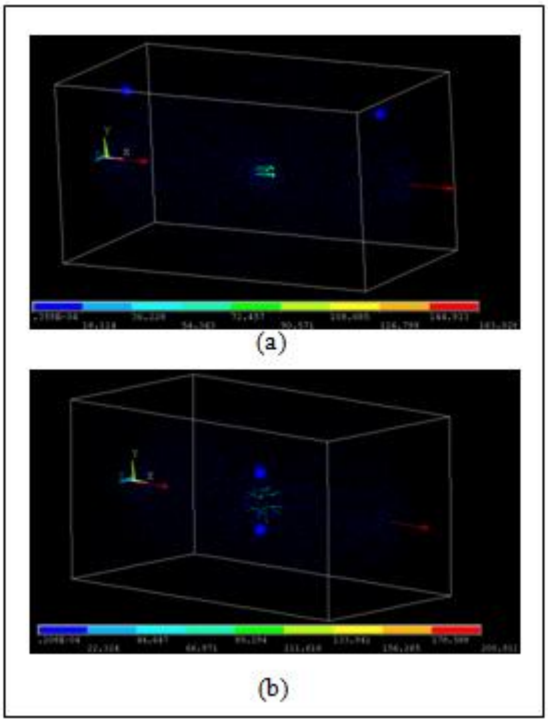

Fig. 9 Current density vector distribution of hybrid system; (a) two nano particles outside of electrical path among micron particles, (b) interconnect network between nano and micron particles.

A.4 Electrical conductivity phenomenon

Fig. 10 shown relationships between conductivity and ratio distance to radius of particle (*d*/*r*) of nano and micron size particles. By considering various values of electrical conductivity it shown that consistent with the theoretical. Correlation electric field to their distributions on surrounding conductance particle is showed by uniformly charged *E* graph with variations concerning distance *r*. In this case, the conductance amount is restricted by the presence of the insulator polymer among particle. The particle phenomenon when embedded in dielectric medium showed that electric field inside the sphere is uniform in x-direction and it is effectively reduced by surrounding dielectric medium. Two interstice spherical charge problem, the electric field obtained are declined with increasing distance *r*. This situation indicates similar trend to the micrographs of discontinuous metal prepared on dielectric matrix with the potential charge distribution theory [9]. When charged spherical uniformly, electric field distribution by variation with *r* is depicted by $E_r = \frac{\rho_v r}{3\epsilon_0}$. Where ρ_v is integrate volume, it found that *E* is proportional with *r* as illustrated in Figure 10, conductivity of micron size higher than nano size. It can be seen that the higher conductivity is achieved by micron size which is 3.07 x 10^{-2} S/cm at particle contact. While the maximum conductivity value of nano size particle is obtained at 2.89 x 10^{-2} S/cm. The electrical conductivity decreases with increasing ratio *d*/*r* due to insulator matrix of epoxy.

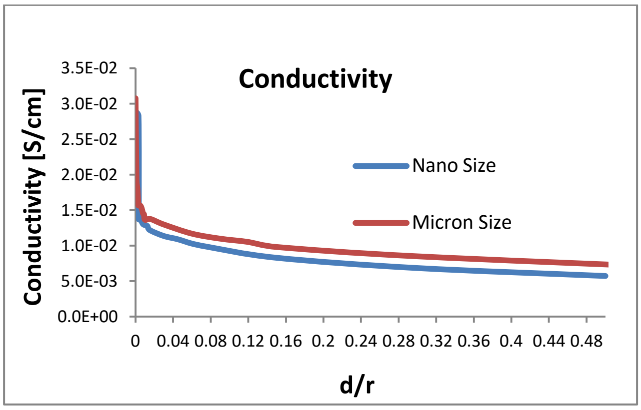

Fig. 10 Comparison of electrical conductivity on the nano and micron size correspond to variations in the partial distance

In hybrid stage system (b), the composite structure was constructed by 1 nm of partial distance both nano and micron size with interconnect network between nano and micron particles. The comparison of electrical conductivity on the nano, micron size and hybrid system as shown in Fig. 11 . The conductivity value was found to be higher as compared to single composition both nano and micron size at 1 nm of partial distance. This difference value might be due to nano size help to establish the conductive path among micron particle and hence lower the resistivity. The hybrid results of the electrical conductivity obtained at 1.98 x 10^{-2} S/cm.

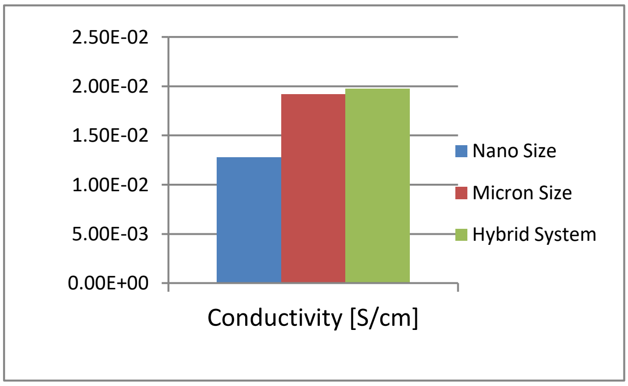

Fig. 11 Comparison of electrical conductivity on the nano, micron size and hybrid system

IV. CONCLUSIONS

It has been founded that the nano particles produced the lower electrical conductivity due to smaller number of electric field (*E*) value. While electric field (*E*) of micron size obtained higher; causing the current flow through the composite was increase. The main feature of the systems studied is the fact that, the conductivity value of the composite will increase. In hybrid system, the results observed shows that electrical conductivity has close connection between electrical transport and morphology of composite. When the composite consisting of micro size, the addition of a number of nano size at interconnecting between nano and micron particle would increasing performance of conduction path among particle.

The reason is that the network paths among microns particle are satisfied by nano particle.

REFERENCES

1. Kandare E., Khatibi A.A., Yoo S., Wang R., Ma J., Olivier P., Gleizes N., Wang C.H. (2015), "Improving the through-thickness thermal and electrical conductivity of carbon fibre/epoxy laminates by exploiting synergy between graphene and silver nano-inclusions", Composites: Part A 69, 72–82.
2. Yao Y., Zeng X., Guo K., Sun R., Xu J.B. (2015) "The effect of interfacial state on the thermal conductivity of functionalized Al2O3 filled glass fibers reinforced polymer composites", Composites: Part A 69, 49–55.
3. Hongsheng Zhao, Tongxiang Liang, Bing Liu, Synthesis and Properties of Copper Conductive Adhesives Modified by SiO2 Nanoparticles, International Journal of Adhesion and Adhesives, p.p.429-433, 2006.
4. Tee D. I., Mariatti M., Azizan A., See C. H. & Chong K. F., Effect of Silane-based coupling agent on the properties of silver nanoparticles filled epoxy composites. Composites Science & Technology 67, 2584-2591, 2007.
5. Irfan Mir, D. Kumar, Recent advances in isotropic conductive adhesives for electronics packaging applications, International Journal of Adhesion & Adhesives 28, p.p. 362–371, 2008.
6. Li. Y, Wong. CP, Recent advance of conductive adhesives as a lead-free alternative in electronic packaging: Materials, Processing, reliability and applications, Materials Science and Engineering R 51, p.p. 1-35, 2006.
7. Ja´nos Mo´czo´, Be´la Puka´nszky, Polymer micro and nanocomposites: Structure, interactions, properties, Journal of Industrial and Engineering Chemistry 14 (2008) 535–563.
8. Schwarz J.A., Contescu C.I., Putyera K., Dekker encyclopedia of nanoscience and nanotechnology, vol. 5, Marcel Dekker Inc, New York, p.p. 3973-3979, 2004.
9. Fu Y, Liu J, Willander M, Conduction Modelling of a Conductive Adheshive with Bimodal Distribution of Conducting Element, *International Journal of Adhesion & Adhesives 19*, p.p.281-286, 1999.
10. Hsien-Hsuen Lee, Kan-Sen Chou, Zong-Whie Shih, Effect of nano-sized silver particles on the resistivity of polymeric conductive adhesives, International Journal of Adhesion & Adhesives 25, 437–441, 2005.
11. Carl T.A Johnk, Engineering Electromagnetic Fields and Waves, John Wiley & Sons, Canada, 1975.

ACKNOWLEDGMENT

The author acknowledges the financial support of the FRGS grant (203/P.Bahan/6071125) and post-graduate research fund (PGRU), as well as the facilities provided by the School of Materials & Mineral Resources Engineering and Post Graduate Research Grant Scheme (1001/PMEKANIK/8042005) of Mechanical Engineering, Universiti Sains, Malaysia.

AUTHORS PROFILE

Muhammad Zulkarnain PhD graduated with B.Eng (Hon) in Mechanical Engineering from Syiah Kuala University (UNSYIAH), Indonesia in 2002. He got his M.Eng and PhD degree in Mechanical engineering from Universiti Sains Malaysia (USM), Malaysia in 2007 and 2014 respectively. His Degree, Masters and PhD thesis was based on optimization and using Finite Element Analysis on Composite Material as a Predict Failure Analysis and Conductivity of Electrical. The main programming tool for his research was ANSYS and Matlab. He got expertise in Matlab and practicing it since 2011. He also had a great contribution in publishing articles and symposium papers in the field of Electrical Conductive Adhesives (ECAs). Currently he is the course leader of the courses like Fluid Mechanics and Fundamental Thermodynamics. He also excellent in writing high impact Q1 journals which is has been published through higher citation index measurement including SCOPUS, ISI and many of them placed on J Mater Sci: Mater Electron, Journal of Microsystem Technologies, Journal of Electronic Materials, Asia-Pac. J. Chem. Eng., Arab J Sci. Eng., International Journal of Technology and Journal of Electronic Materials for publications.

Muhammad Sobron Yamin Lubis, Ph.D graduated with B.Eng in Mechanical Engineering from Islamic University of North Sumatera (UISU), Indonesia in 1991, He got his M.Sc and Ph.D Degree in Mechanical Engineering from Universiti Sains Malaysia (USM), Malaysia in 2000 and 2008 respectively. In 1994-2011 he worked at Muhammadiyah University of North Sumatera as a lecturer in the Mechanical Engineering department and in 2011 until now he has worked as a lecturer and Head of Manufacturing Engineering Section at Tarumanagara University in Indonesia. He did a lot of research in the field of the manufacturing process and machining and development of the material of cutting tools.