
A Numerical Study on the Determination of Mechanical and Thermal Properties of Hybrid Polymeric Composites

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Abstract

Filled Polymeric composites due to its high strength, less weight and ease of fabrication are profoundly used in variety of applications. Depending on the type of filler added with polymeric resin, the enhanced property is achieved thereby making it as a tailorable material. Especially glass microsphere filled epoxy resin is used as a potting compound in electronic and aviation industries. Therefore, there is a need to characterise the material which is best suitable as a potting compound. In this work, the effective properties of three different glass microsphere filled epoxy system is evaluated numerically using ANSYS. A single Representative Volume Element (RVE) model is used to evaluate both the mechanical and thermal properties at various loading fractions. The numerically predicted values are then compared with the analytical models and with the literature experimental data. The significance of the analytical model on the determination of properties is reported. Further the suitability of the hybrid polymeric composite system comprising Solid and Hollow glass microspheres as potting compound is investigated numerically using random RVE. The effect of interface on the properties is also investigated and reported.

Keywords: Filled polymeric composites; Glass microsphere; Interface; Loading fractions; Potting compound

1. Introduction

Particles filled polymeric composite materials have been intensively developed since 1960 and have shown a great promise in the industrial and structural applications for the past decades. These leading classes of engineering materials are widely used because of its outstanding properties such as low weight, high stiffness and strength, thermal stability, high chemical resistance and ease of fabrication. In the development of these light weight composite structure for aerospace and automotive industries, mechanical and thermal property plays a crucial role under unusual operating condition such as extreme environmental condition and operating temperature. Many researchers have claimed that the mechanical and thermal properties improve by the addition of filler in micrometer to nanometer sizes [1-6]. Some of the filler which have shown dramatic improvements in the properties are silicon dioxide, glass, aluminum oxide, copper, carbon nanotubes, and carbon nanofibre and so on. Among these fillers, the glass microsphere is commonly used for improvement in high modulus, relatively high strength, low coefficient of expansion and low cost [5-7]. These are the demandable qualities of an electronic potting compound. Therefore, the glass bead filled epoxy system is best suited for electronic applications [8, 9]. Thus, it is desirable to calculate the effective properties from the knowledge of the structure of the composite material.

The determination of the effective mechanical and thermal properties of a composite material with complex microstructures is a challenging task to engineers and scientists, because of its fundamental and technological importance in almost every area of material science. Some of the literature available to predict the effective thermal conductivity and the effective young's modulus of glass bead filled epoxy system are reported here. The thermal conductivity of hollow glass bead filled polymeric resin such as epoxy and polypropylene have been investigated experimentally and numerically [10-12]. The authors [13] have reported the estimation of effective thermal conductivity of a solid glass bead filled epoxy both analytically and numerically using ANSYS. Similarly, many researchers have experimentally determined the mechanical properties of filled polymeric composites [14 -17].

Numerous researches are going on in this field to have an exact model for the evaluation of the effective thermal conductivity and effective modulus of a particle filled composite systems. However, the presence of the interphase between particle and matrix alters the properties of the composite. Several studies have been made to accurately model the experimental results for evaluating the stiffness of nano composites [14, 15] by using the interphase concept. Among several researchers Vo, et al. [16] is the first to investigate the effects of the interphase in the numerical modeling of particle filled composites, by accurately fitting various closed form models with the experimental data for composites composed of micron-sized Al₂O₃ particles in a silver matrix. The author Brown et al. [17] have discussed the effect of interphase thickness in polymer nano composites by using molecular dynamics. It is noted that reduced particle size can lead to an increasing influence of the interphase on overall composite behavior for a fixed particle volume fraction. The thickness of the interphase is usually determined indirectly from a composite property, but the results depend very much on the method of determination.

Most of the studies that have been reported are for single filler composites and only very few papers have reported the synergistic effects of two different kinds of fillers on the thermal, mechanical and dielectric properties. In this work the effective thermal conductivity and the effective mechanical properties such as modulus and tensile strength for glass microsphere filled epoxy composite and a hybrid system is evaluated by using finite element software Ansys in 2D. Further, the effect of the interface between the matrix and the filler is studied by modeling the interface region.

2. Theoretical models

For a binary or ternary composite system, the effective properties of a composite material is a complex function and rely on their geometry of the particle, the material property of the matrix and reinforcement phases, distribution of particle within the matrix medium and interface between the particles. Although it can be measured by experimental methods, analytical methods and equations are often essential to predict the properties of composite materials in order to have a better understanding of the micromechanics of the composite materials. This section reviews the important theoretical/empirical models especially for spherical inclusions reported in the literature for calculating the effective young's modulus and strength of particle filled composites for comparison. The Voigt and Reuss models [18] are the two most basic models used to characterize the mechanical behavior of a two-phase composite system consisting of a spherical filler inclusion embedded in a continuous matrix material. The Voigt and Reuss model, assumes that the particulate inclusions and matrix material are subjected to constant strain and constant stress respectively. These models predict the upper bound and lower bounds of modulus and the effective modulus of the composites lie within these bounds. In addition to the Voigt and Reuss model other theoretical models to predict the effective Youngs modulus of the composite system is proposed by Neilson and Einstein [19]. The Einstein equation considers the filler as rigid spheres and assumes perfect bonding between the filler and matrix. This equation is valid for low particle loading fraction. It is impossible to specify the conditions of constant stress or strain across the filler-matrix boundaries, hence more rigorous models have been proposed using bounding relationships or self-consistent field theories [20] to increase the predictive accuracy of the Voigt and the Reuss models. The modulus properties of the solid glass filled composites are modeled using the S-Combining Rule [18]. The C-Combining Rule [18] is more appropriate for the matrix filled with a more compliant filler, such as epoxy matrix loaded with hollow glass spheres.

3. Numerical Analysis

In the present work, using the finite element software ANSYS the numerical analysis of Solid Glass Microsphere (SGM), Hollow Glass Microsphere (HGM) and a combination of SGM/HGM filled epoxy composite is investigated. The size of hollow and solid microspheres is 20 μm and 100 μm respectively. The material properties of the filler and the matrix considered in this study are shown in the Table 1. In order to perform this analysis, a single representative volume element (RVE) is considered for SGM, HGM and

random RVE for hybrid SGM/HGM epoxy composite. The random position of SGM and HGM is modelled and analysed.

Table 1 Material parameters

Material	Property					
	Density g/cm ³	Young's Modulus GPa	Poisson's ratio	Rigidity modulus GPa	Bulk Modulus GPa	Thermal conductivity W/(mK)
Epoxy	1.19	2.45	0.33	0.92	2.47	0.363
SGM	2.54	76	0.23	30.9	46.9	0.00363
HGM	0.37	1.99	0.21	0.82	1.15	0.137

3.1 Structural Analysis of Glass Microsphere filled epoxy composite

The symmetric nature of single RVE model under study is taken as an advantage and a quarter model of the composite system is modeled using Plane 182 element. The loading and boundary condition for SGM and HGM loaded epoxy composite is shown in Fig.1 (a) & (b). The structural analysis of SGM, HGM and hybrid SGM/HGM loaded epoxy is performed and the average stresses and strain induced in three different composite system is computed using the trapezoidal rule. From the numerically obtained stresses and the strain, the effective Young's modulus is predicted.

Further, the effect of interface is studied by modeling the interfaces as three phase continuum represented by a matrix, homogenous spherical particle and the interface. The interface region of the filled composites is modeled using the linear correlation between the matrix and filler material.

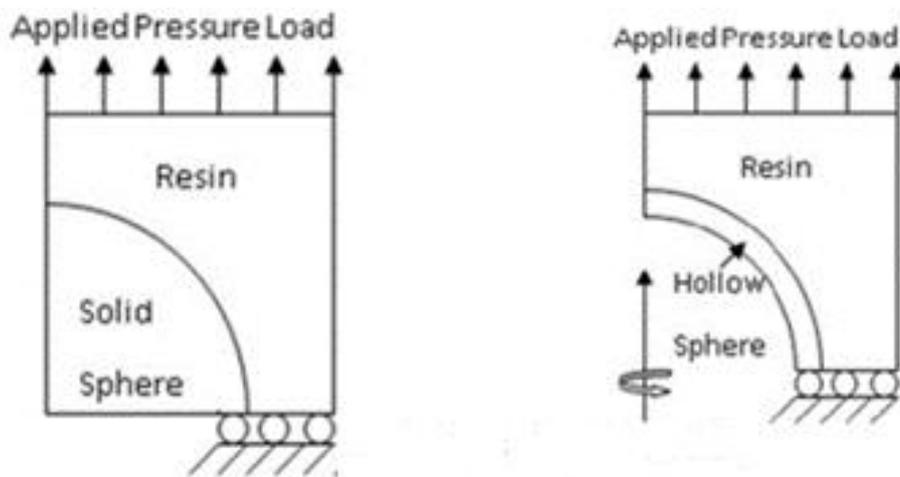


Figure 1 (a) & (b) Quarter Model showing loading and boundary Condition for SGM and HGM respectively

3.2 Thermal Analysis of Glass Microsphere filled epoxy composite

The prescribed boundary conditions for the thermal analysis of particle filled composite and the direction of heat flow for the conduction problem is as shown in Fig. 2. The temperature at the nodes along the surface ABCD is $T_{high}=100^{\circ}\text{C}$ and the analysis of the problem is done at a room temperature of $T_{low} = 27^{\circ}\text{C}$.

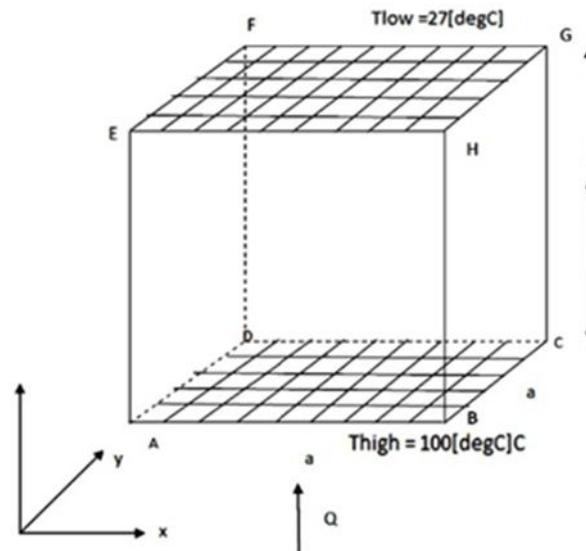


Figure2 Heat flow and boundary Conditions for Filled Polymer composites

The remaining surfaces parallel to the direction of the heat flow are assumed as insulated. The average thermal conductivity in three different composite system is computed using the trapezoidal rule. The numerical analysis results for SGM filled epoxies [21] with interface is validated with the literature experimental data and compared with analytical models. In this study, the effective thermal conductivity prediction is extended for HGM and hybrid filled epoxies, at different loading fractions loading concentrations and the results are compared.

4. Result and Discussion

4.1 Structural Analysis of Glass Microsphere filled epoxy composite

The structural analysis of solid, hollow and hybrid glass microspheres filled epoxy is carried out by single RVE model at different filler proportions. The different volume fractions ranging from 0.05 to 0.45 of the glass microsphere of 100 micrometer is considered in this study for SGM filled epoxies. Fig.3 shows the stress distribution of SGM filled epoxy at $V_f = 0.15$ for the given boundary conditions of a RVE model. The effect of interface between SGM and epoxy at $V_f=0.05$ is shown in Fig 4. With the stresses and strain values the effective young's modulus is computed.

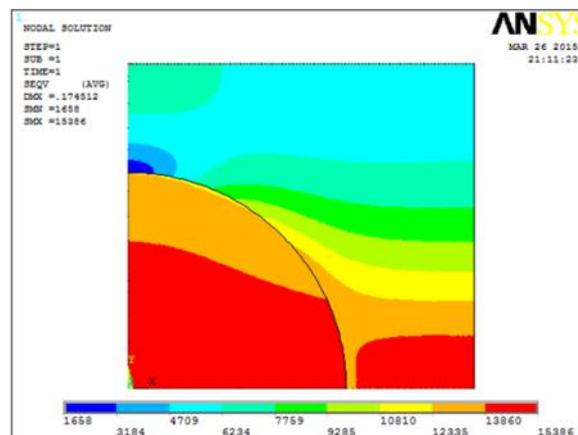


Figure 3 Stress distribution of SGM filled epoxies ($V_f = 0.15$)

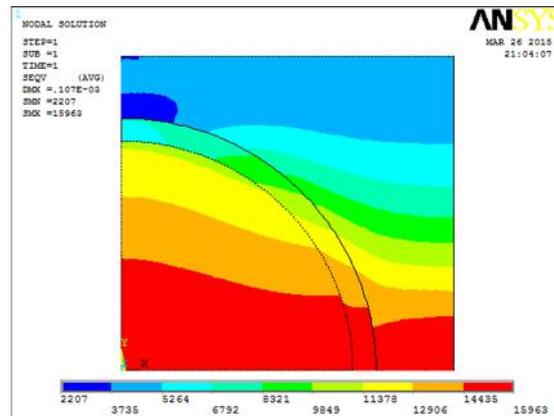


Figure 4 Stress distribution of SGM filled epoxies with interface ($V_f= 0.05$)

Further, the numerically obtained results are compared with the theoretical model described in section 2.2 and with literature experimental data. Table 2 summarise the effect of SGM filler content on the elastic modulus computed both numerically and theoretically. From the Table 2, it is clear that the effective modulus increases with the increase in filler content. The numerically obtained results are closer to the experimental values. Thus, emphasising the efficacy of the numerical tool. However, the deviation of numerical results from experimental value may be due to the assumption of perfect bonding between the particle and matrix. Of the various theoretical model discussed, the S-combining rule agrees with the experimental and numerical results. From the structural analysis results, the tensile strength of SGM filled epoxy is computed and is shown in Table 3. Table 3 shows the comparison of the results obtained with the literature experimental data [18] and with the analytical models. It is noticed that with addition of SGMs, tensile strength of the composite decreases and this decrement is a function of the SGM content.

Table 2: Effect of filler content in SGM filled the epoxies on Elastic Modulus

Filler	Volume fraction, V_f	Elastic modulus, GPa								
		Theoretical						Ansys		Literature experimental data [18]
		Neilson's model	Einstein's theory	Voigt model	Reuss model	Wong's model	S-combining Rule	WOI	WI	
Neat	-	2.45	2.45	2.45	2.45	2.45	2.45	-	-	2.45
	0.05	2.76	2.57	6.13	2.57	2.51	2.72	2.93	3.09	3.13
	0.1	3.06	2.69	9.8	2.71	2.57	3.04	3.46	3.76	---
	0.15	3.37	2.82	13.48	2.87	2.63	3.41	4.06	4.7	3.9
SGM	0.2	3.67	2.94	17.16	3.04	2.69	3.85	4.75	5.25	---
	0.25	3.98	3.06	20.83	3.23	2.75	4.36	5.6	7.4	4.99
	0.3	4.29	3.18	24.51	3.45	2.82	4.97	5.98	4.71	5.7
	0.35	4.59	3.31	28.19	3.7	2.88	5.71	7.22	5.27	---
	0.4	4.90	3.43	31.87	4.00	2.94	6.6	8.59	5.52	---
	0.45	5.20	3.55	35.55	4.34	3.00	7.69	11.82	---	10.1

WOI – Without interface; WI – With interface

Table: 3 Effect of filler content in SGM filled epoxies on Tensile Strength

Filler	Volume fraction,	Elastic modulus (GPa)			Theoretical modulus (GPa)					
		ANSYS		Experimental	Neilsons model	Einstein's theory	Voigt mod	Reuss mod	Wongs mode	C-Combination
		WOI	WI							
Neat	-	-	-	2.45	2.45	2.45	2.45	2.45	2.45	2.45
HGM	0.05	2.323	2.323	----	2.75	2.57	2.43	2.42	2.51	2.43
	0.1	2.318	2.319	----	3.06	2.69	2.4	2.39	2.57	2.41
	0.15	2.316	2.316	2.7	3.37	2.82	2.38	2.37	2.63	2.38
	0.2	2.313	2.313	----	3.67	2.94	2.36	2.34	2.69	2.36
	0.25	2.31	2.319	3.07	3.98	3.06	2.33	2.32	2.76	2.34
	0.3	2.306	2.306	----	4.29	3.18	2.31	2.29	2.82	2.32
	0.35	2.303	2.303	2.61	4.59	3.31	2.29	2.26	2.88	2.3
	0.4	2.299	2.299	----	4.9	3.43	2.26	2.24	2.94	2.28
	0.45	2.298	2.298	2.67	5.2	3.55	2.43	2.22	3.00	2.26

WOI – Without interface; WI – With interface

Table: 4 Effect of filler content in HGM filled the epoxies on Elastic modulus

Filler	Volume fraction, Vf	Max. stress (MPa)			Theoretical stress (MPa)	
		Ansys		Experimental [23]	Tavman Model	Wong Model
		WOI	WI			
Neat	-			58	58	58
SGM	0.05	57.73	57.82	57.34	48.47	54.06
	0.1	57.81	56.25	57.14	42.88	51.75
	0.15	57.23	55.9	56.92	38.19	49.81
	0.2	56.03	55.07	56.59	34	48.08
	0.25	55.02	53.5	56.18	30.15	46.49
	0.3	54.58	53.14	55.65	26.55	45
	0.35	52.37	40.49	55.06	23.14	43.59
	0.4	36.19	32.03	---	19.9	42.26
	0.45	24.33	0	---	16.79	40.97

WOI – Without interface; WI – With interface

The study is extended to predict the stress distribution for HGM and SGM/HGM filled epoxies for different particle loading fraction both numerically and theoretically. The effect of filler content on the elastic modulus of HGM and SGM/HGM filled epoxies is shown in the Table 4 and Table 5. Fig. 5 shows that, the elastic modulus

increases with increase in filler content and the hybrid polymeric composite has high modulus when compared with SGM and HGM.

Table: 5 Effect of filler content in SGM/HGM filled epoxies on Elastic modulus

Filler	Volume fraction, Vf	Elastic modulus (GPa)	Theoretical modulus (GPa)		
		Ansys	Neilson's model	Einstein's	Wongs model
Neat	-	-	2.45	2.45	
Solid/Hollow	0.05	3	2.75	2.51	2.51
	0.1	3.63	3.06	2.57	2.57
	0.15	4.498	3.37	2.63	2.63

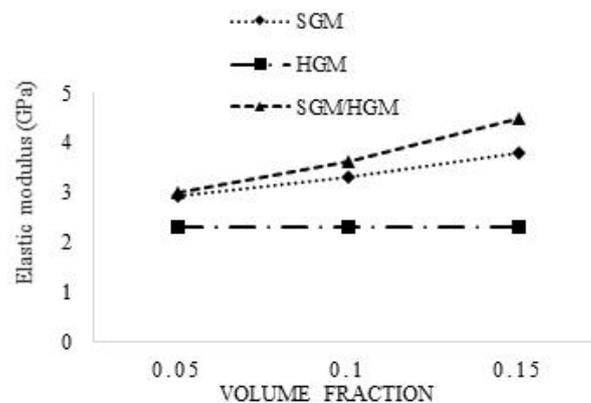


Figure 5 Effective elastic modulus comparison of SGM, HGM and SGM/HGM filled epoxies

4.2 Thermal Analysis

The thermal analysis of solid, hollow and hybrid glass microspheres filled epoxies is carried out by single RVE model and random RVE model at different filler proportions. The different volume fractions ranging from 0.01 to 0.18 of the glass microsphere is considered in this study for filled epoxies. Fig. 6 and Fig. 7 shows the heat flux and temperature distribution of HGM filled epoxy at Vf = 0.15 for the given boundary conditions of a RVE model. From the heat flux value obtained numerically, the average heat flux is calculated using the trapezoidal rule thereby the effective thermal conductivity is computed. The thermal conductivity value decreases with the addition of SGM, HGM and hybrid glass microsphere. This fact is clearly shown in Fig. 8. Also for hybrid i.e. SGM and HGM filled epoxy composite is having higher thermal conductivity and is suitable as an electronic potting compound

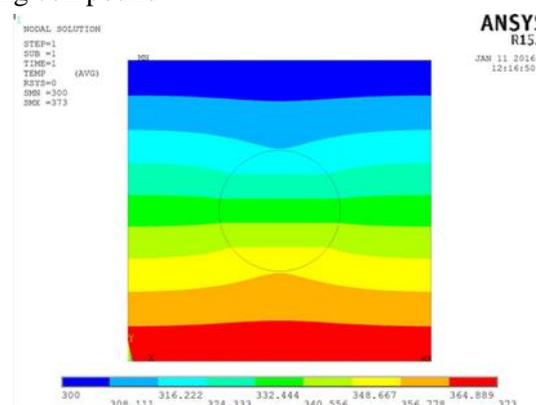


Figure 6 Temperature distribution of HGM filled epoxies

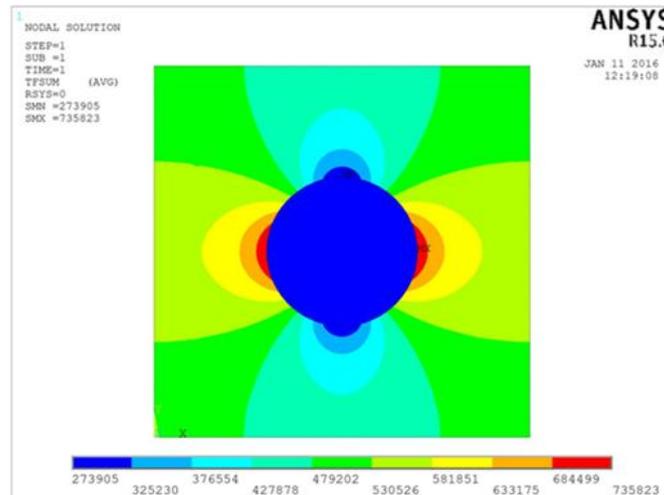


Figure 7 Heat flux distribution of HGM filled epoxies

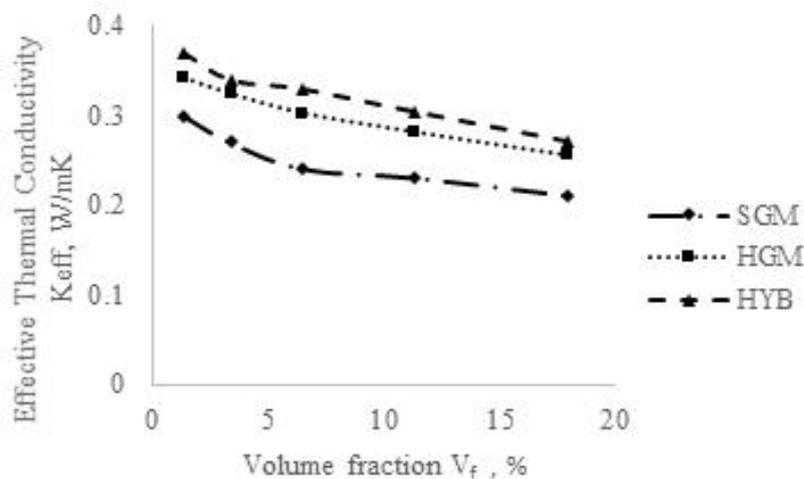


Figure8 Effective Thermal conductivity comparison of SGM, HGM and SGM/HGM filled epoxies

5. Conclusion

In this work, the thermal and mechanical properties of glass microsphere filled epoxy composite material to be used as electronic potting compounds are evaluated numerically. Through this material characterisation, effective thermal conductivity and the key mechanical properties such as effective modulus, Poisson's ratio and tensile strength are determined. The observations obtained from this work are summarised below.

-) Effective elastic modulus increases with increase of SGM and HGM content in epoxy.
-) In SGM/HGM ternary composite system also, the effective elastic modulus increases as a function of volume fraction. The SGM/HGM ternary composite system has a higher modulus with least density compared with SGM and HGM.
-) The effect of interface as a function of particle diameter has shown a significant effect on composite modulus.
-) The numerical results for a model with interface matches well with the literature experimental data and several mathematical models.
-) Thermal conductivity of glass bead filled epoxy system decreases with increase in glass bead content. However the conductivity values is higher in SGM/HGM ternary composite system
-) From the work, it is clearly understood that using SGM/HGM ternary composite system, greater strength is obtained at low density i.e. high strength to weight ratio. This characteristic is highly desirable for electronic potting compounds.

References

- [1] S.S. Ray and M. Okamoto, Polymer/layered silicate nano composites: a review from preparation to processing, *Prog. PolymSci*, 28 (2003) 1539-1641.
- [2] B.B. Johnsen, A.J. Kinloch, R.D. Mohammed, A.C. Taylor and S. Sprenger, Toughening mechanisms of nanoparticle-modified epoxy polymers, *Polymer*, 48 (2007) 530-541.
- [3] M.H. Kim, C.I. Park, W.M. Choi, J.W. Lee, J.G. Lim and O.O. Park, Synthesis and material properties of syndiotactic polystyrene/ organophilic clay nanocomposites, *J ApplPolym Sci.*, 92 (2004) 2144-2150.
- [4] H.L. Tyan, Y.C. Liu and K.H. Wei, Thermally and mechanically enhanced clay/polyimide nanocomposite via reactive organoclay, *Chem. Mater.*, 11 (1999) 1942-1947
- [5] Y. Nakamura and M. Yamaguchi, Effect of particle size on the fracture toughness of epoxy resin filled with spherical silica. *Polymer*, 33 (1992) 3415-3426.
- [6] J. Ma, M.S. Mo, X-S. Du, P. Rosso, K. Friedrich and H-C Kuan, Effect of inorganic nanoparticles on mechanical property, fracture toughness and toughening mechanism of two epoxy systems, *Polymer*, 49 (2008) 3510-3523.
- [7] T.H. Hsieh, A.J. Kinloch, K. Masania, A.C. Taylor and S. Sprenger, The mechanisms and mechanics of the toughening of epoxy polymers modified with silica nanoparticles, *Polymer*, 51(2010) 6284-6294.
- [8] C.A. Harper, *Electronic Packaging and Interconnection Handbook*, 3rd Ed. McGraw-Hill Publishing Company, NewYork, USA (2000).
- [9] J.C.Smith, Experimental Values for the Elastic Constants of a Particulate-Filled Glassy Polymer, *Journal of Applied Physics and Chemistry*, 80A (1976) 45–49.
- [10] J.Z. Liang and F.H. Li, Measurement of thermal conductivity of hollow glass bead filled polypropylene composites, *Polym. Test*, 25 (4) (2006) 527-531.
- [11] J.Z. Liang and F.H. Li, Simulation of heat transfer in hollow glass-bead- filled polypropylene composites by finite element method, *Polym.Test*, 26 (3) (2007) 419-424.
- [12] J.Z. Liang and F.H. Li, Heat transfer in polymer composites filled with inorganic hollow micro-spheres: A theoretical model, *Polym. Test*, 26 (2007) 1025-1030.
- [13] Debasmita Mishra, Lucy Mohapatra, AlokSatapathy and Amar patnaik, *Determination of Thermal Conductivity of Polymer composites filled with Solid Glass Beads*, Proc. Int.Conference on Advancement in Polymeric Materials, CIPET, Chennai (2011).
- [14] V. Cannillo, F. Bondioli, L. Lusvarghi, M. Montorsi, M. Avella, M. Errico and M. Malinconico, Modeling of ceramic particles filled polymer–matrix nanocomposites, *Compos Sci Technol.*, 66 (2006) 1030-1037
- [15] H. Liu and L. Brinson, Mimicking mussel adhesion to improve interfacial properties in composites, *Compos Sci Technol.*, 68 (2008) 2042-2048.
- [16] H. Vo, M. Todd, G. Shi, A. Shapiro and M. Edwards, Towards model-based engineering of underfill materials: CTE modelling, *Microelectron J.*, 32 (2001) 331-338.
- [17] D. Brown, V. Marcadon, P. Mele, N. Alberola, Effect of particle size on the properties of model nanocomposites, *Macromolecules*, 41 (2008) 1499 – 1511.
- [18] M. J. Quesenberry, P. H. Madison, and R. E. Jensen, Characterization of Low Density Glass Filled Epoxies, Army Research Laboratory, Aberdeen Proving Ground (2003).
- [19] H. Ku, J. Epaarachchi, M. Trada, and P. Wong, Modelling of tensile properties glass powder/epoxy composites post-cured in an oven and in microwaves, *Journal of Reinforced Plastics and Composites*, 32 (2013) 10689-699
- [20] R. Hill, Continuum Micro-Mechanics of Elasto plastic Polycrystals, *Journal of the Mechanics and Physics of Solids*, 13 (1965) 89 –101.
- [21] S.Supriya, Dr.J.SelwinRajadurai and B.Sankar, A Numerical Study of Interface Effect on the effective thermal conductivity of Glass Microsphere Filled Polymer Composites, *Int. Journal of Appl. Engg. Research*, 10 (2015) 8967-8980
- [22] D. Mishra, Ph.D. Thesis, A Study on Thermal and Dielectric Characteristics of Solid Glass Microsphere Filled Epoxy Composites, National Institute of technology, Rourkela, India (2004)