THE EFFECT OF POROSITY ON THERMAL CONDUCTIVITY OF SILICON CARBIDE FILLED POLYPROPYLENE COMPOSITE

Azlin Fazlina Osman, Banjuraizah Johar, Syed Nuzul Fadzli Syed Adam,
Sri Asliza Md Amin, & Amirro Osman
School of Materials Engineering, Universiti Malaysia Perlis,
Academics Complex -Jejawi (2), Taman Muhibah, Jejawi 02600 Arau, Perlis.

Abstract

Silicon carbide filled Polypropylene (PP) composites are fabricated, where the compounding is carried out by using Z-blade mixer with filler loadings of 0wt%, 10wt%, 20wt% and 30wt%. Composite sheets produced from hot press molding process are prepared for testing samples. The effect of filler loadings and incorporation of coupling agent on density, porosity and thermal conductivity of SiC filled PP composites are studied. Thermal conductivity test is performed by using Digi Sense Scanning Thermometer apparatus. The results reported demonstrate that, the density and thermal conductivity of SiC filled PP composites increased with increasing filler loadings while the porosity decreased with increasing filler loadings. When treated with 5% titanate coupling agent (Lica-12), the increase of density for 10%SiC-90%PP and the increase of porosity for 30%SiC-90%PP composite can be observed. On the other hand, the increase of thermal conductivity can be observed for the composite treated with Lica-12 at 10% filler loading. However this treated SiC filled PP composites showed a decrease in thermal conductivity as filler loading increased.

Introduction

Thermally conducting but electrically insulating polymer-matrix composites is increasingly important for electronic packaging because the heat dissipation ability limits the reliability, performance and miniaturization of electronics [3]. Ceramic fillers such as aluminum nitride, silicon carbide and alumina are used. That explained why ceramic powder reinforced polymer materials have been used extensively as electronic packaging materials [1, 2]. This work is focused on SiC as filler in PP matrix due to its combination of high thermal conductivity and low cost. It is known that the transport of heat in nonmetals occurs by phonons or lattice vibration. The thermal resistance is caused by various types of phonon scattering processes include pore scattering phenomenon [4]. Therefore, we investigate the effect of porosity in thermal conductivity seems the phonon-pore scattering phenomenon can affect the thermal transport. The research also studied the effect of titanate coupling agent (Lica-12) addition on the porosity and thermal conductivity of PP-SiC composite since this type of coupling agent are believed can enhance the properties of composites combining a wide range of fillers and polymers [7].

Experimental Methodology

Composite polypropylene filled with silicon carbide (SiC-PP) was prepared by using melt mix method. SiC-PP composite was prepared by using Z-blade mixer with the rotation speed of 35 rpm at 190°C and then compressed by using hot press molding with the pressure of 165 kg/cm² at 190°C. Composites with different weight percentage of filler contents varying from 0% to 30% were prepared. In order to improve properties of composites consisting of PP filled with SiC particles, the particles surface was treated with a titanate coupling agent. The effect of titanate coupling agent on density, porosity and thermal conductivity of SiC-PP composite were measured as a function of filler loading and coupling agent content.

Porosity Test

Porosity is determined using soaking process or water absorption test. The soaking process is done for 30-45 minutes. Absorption occurs when water liquid enter the pores existed in the sample solid. Porosity is the ratio between pores and total sample volume. While bulk density value is mass per total volume includes all the pores and filled portion of the pores. In this experiment, ethanol is used instead of water and the density value for ethanol is 0.7893 g/cm3.

$$\% \text{ Porosity} = \frac{W3 - W1}{W3 - W2} \text{ X } 100\%$$

Where;
W1 = sample weight in air = solid weight + open pores weight
W2 = sample weight in water
W3 = sample weight in air after soaking = W1 + water weight with closed pores

Thermal Conductivity Test
The thermal conductivity is measured by using Digi Sense Scanning Thermometer apparatus (Figure 1). The sample which is in bar shape with dimension of 30mm x 3mm x 3mm is placed onto a holder and then screwed tight to position the sample firmly. The end of the bar is connected to the 300 Ohm, 3W resistor which is then connected to Phywe 12V Power supply so that heat can flow through the sample. A non-silicone heat transfer compound is put at the contact point between the capacitor and the sample to facilitate the heat flow from the capacitor to the sample. Two probes are used in this test.
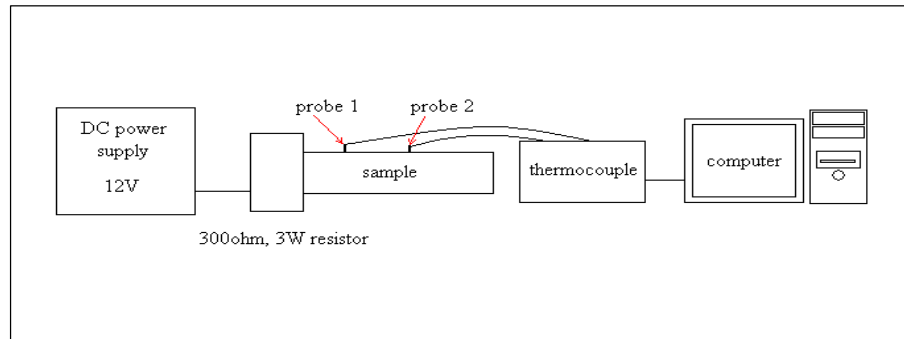

Figure 1: Apparatus setup for thermal conductivity testing

Thermal conductivity is calculated using equation:

$$k = \frac{Q\,L}{A \times \Delta T}$$

Q is the heat supplied to the bar (watt)
L distance between probe 1 to probe 2 (meter)
A is the cross-sectional area of the bar (m^2)
ΔT is the difference in temperature between two probes in the bar ($T_2 - T_1$),
k is the coefficient of thermal conductivity of the bar. (W / m K)

Results and Discussion
Figure 2a shows the bulk density of both treated and untreated composite with a function of filler loading. It can be seen that the density of untreated composite increased with increasing filler loading. However, the treated composite showed an increment in density at 10% filler content but exhibit a decrement in density at 30% filler content. Figure 2b shows the result for porosity test on the composite samples. According to the result, untreated 90%PP-10%AlN shows the highest porosity value at 5.7%. For overall untreated composite, porosity decreased as filler loading increased. But for the treated composite, the result is inconstant where 70%PP-30%AlN showed the highest porosity value. The result predicts that the addition of coupling agent caused a decrease of porosity for low filler loading composite but promotes an increase in porosity for the higher filler loading composite. Previous research [8] stated that an increase in porosity is intrinsically related to the processes that take place during the mixing and compression molding procedures.

Figure 3 demonstrates the thermal conductivity value for both treated and untreated composite, where the untreated composite showed the increasing value with increasing filler loading. Previous research [1] on polystyrene-AlN composites also showed the same trend of thermal conductivity with increasing of filler loading. The thermal conductivity of SiC is much larger than that of PP, so the addition of AlN filler to the PP matrix will result in an increased in the thermal conductivity of the composite. At higher particle content, the filler tends to form agglomerates and conductive chains resulting in a rapid increase in thermal conductivity [6]. The increase in thermal conductivity is also due to the decrease in the filler-matrix thermal contact resistance through the improvement of the interface between matrix and particle [3]. From morphology study (Figure 4), it can be seen that the filler distribution and AlN particle contact is improved by increasing the filler loading. Good filler distribution in the matrix improves the contact between filler and filler thus providing good channel for heat to transfer [10]. Figure 2b indicates that for untreated composite, the increase of filler loadings decreased the degree of porosity in the PP-SiC composite. Reduction of porosity increased the density of particle contacts that probably exceeded the thermal conductivity. It is believed that this condition provides a

261

better contact for the powder particles and therefore, facilitates thermal transport by improving contact in the powder chains [8].

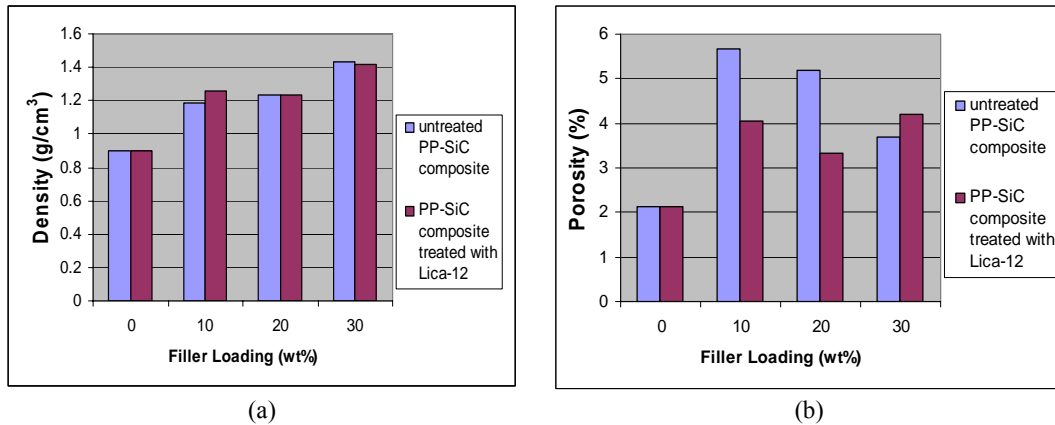(a)   (b)

Figure 2 a) Bulk density and b) Porosity value of PP-SiC composite in different filler loadings.

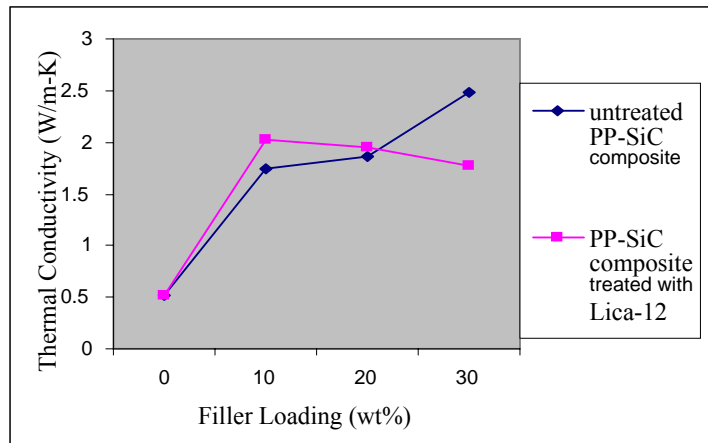Figure 3: Effect of filler loadings and Lica-12 content on thermal conductivity of PP-SiC composites

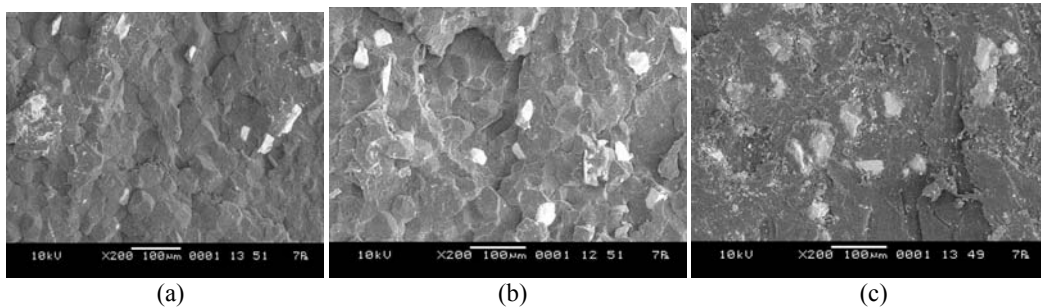(a)   (b)   (c)

Figure 4: SEM micrograph of a) 90%PP-10%SiC b) 80%PP-20%SiC c) 70%PP-30%SiC at 200x magnification

However, the unsteady result performed by the treated composite with an increase in thermal conductivity at 10% filler content followed by a slight decrease in thermal conductivity beginning at 20% filler content. The increased of thermal conductivity of 90%PP-10%SiC composite can be related with the effect of titanate coupling agent (Lica-12). The addition of coupling agent enables the matrix to form stronger bonding with the filler forming a 'molecular bridge' between the two, preventing gap where air can trapped at the interface and enhanced heat transfer between the matrix and the filler at the interface

262

[5]. Coupling agent can enhance the compatibility between the resin and the filler thus enhance the dispersion of the fillers, improved wet-out between resin matrix and filler which in turn improve the thermal conductivity ability of the polymer composites [9]. Comparison between morphology of treated and untreated PP-AlN composite is shown in Figure 5. Figure 5a shows the poor dispersion of filler due to weak filler-matrix interaction while Figure 5b shows better filler dispersion in matrix and better wetting by matrix caused by the addition of Lica-12. Good wetting by matrix can reduce porosity and enhance thermal conductivity and these statement supported by the result obtained in Figure 2b. Overall, treated 70%PP-30%SiC composite shows the lowest thermal conductivity. As referred from the previous result, treated composite also exhibit the highest degree of porosity at 30% filler loading. This shows that the increase in porosity in the composite system decreased the thermal conductivity. The lowering of thermal conductivity proved that the porosity produces either a small effect on the heat transfer properties or it forms a barrier for thermal transport in between particles [8]. Parrot and Stuckes [4] also stated that composite in high porosity present a decrease in thermal conductivity values in proportion to an increase in porosity degree. The lower value of thermal conductivity supports this argument. Figure 6a and 6b show the morphology of 90%PP-10%SiC and 70%PP-30%SiC respectively, where more pores exist in the 70%PP-30%SiC compared to 90%PP-10%SiC sample.
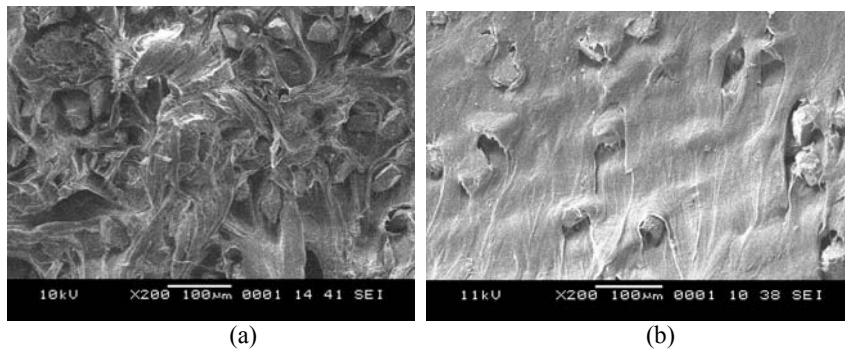

| (a) | (b) |

Figure 5: SEM micrograph of 90%PP-10%SiC (a) untreated (b) treated with 5%Lica-12 at 200x magnification showed better dispersion of filler and improved wet-out between PP (matrix) and SiC (filler)
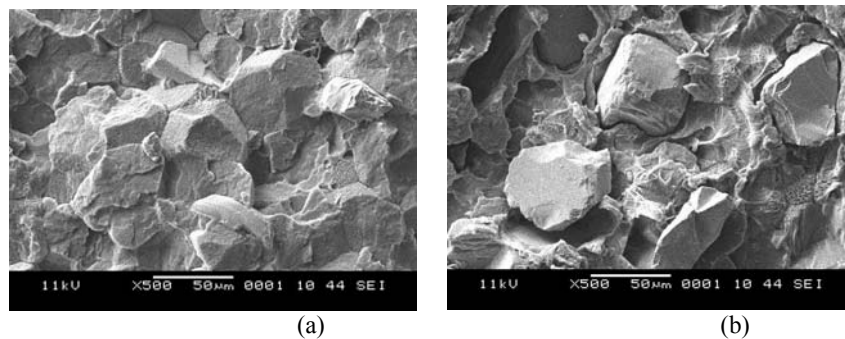

| (a) | (b) |

Figure 6: SEM micrograph of (a) treated 90%PP-10% SiC with less pores existence (b) treated 70%PP-30%SiC with more pores caused by de-wetting of filler by matrix; at magnification 500x.

Conclusion

The thermal conductivity increases with increasing filler volume fraction. The addition of 5%Lica-12 promotes an increase in porosity for PP-SiC composite with 30% filler content and therefore, decrease the thermal conductivity of the composite. Porosity may affect the permeability and transferring of heat, and therefore it drives the thermal conductivity values.

References

[1] Suzhu Yu, Peter Hing and Xiao Hu. (2002). Thermal Conductivity of Polystyrene-Aluminum Nitride Composite, *Composites: Part A,* 33, pp. 289-292.
[2] Reichmanis R. (1995). Microelectronics Technology: Polymers for Advanced Imaging and Packaging. ACS Symposium Series, American Chemical Society.

[3] Yunsheng Xu, D.D.L. Chung and Cathleen Mroz, (2001). Thermally Conducting Aluminum Nitride Polymer-Matrix Composites, *Composites Part A,* 32, pp. 1749-1757

[4] Parrott JE and Stuckes A.D (1975). Thermal Conductivity of Solids, New York:Methuen.

[5] Murphy J.(1996), *The Additives for Plastic Handbook,* Elsevier Science, p.202

[6] Sofian N.M., Rusu M., (2001). Metal Powder-Filled Polyethylene Composites. Thermal Properties. J*ournal of Thermoplastic Composite Materials,* 14, pp. 20-33.

[7] Monte S.J, Belgian Plastic & S.J, Belgian Plastic & Rubber Institute, Spring Conference 2003, titanates & zirconates.

[8] Lima W.M, Biondo V, Weinand W.R, Baesso M.L and Bento A.C. (2005) The effect of porosity on thermal properties, Journal of Physics :Condensed Matter, 17, pp 1239-1249.

[9] Seymore R.B and Carraher C.E (1984). *Structure–property relationship in polymers,* 11, Plenum Press, New York.

[10] Delmonte, J. (1978). *Handbook of Fillers and Reinforcement for Plastics*. Van Nostrand Reinhold, New York.