Analysis of Thermal Conductivity in Composite Material

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ABSTRACT

The world population continuously increasing and because of these our requirements is steadily increase, and resources (material) is reducing. This situation has led to the development of alternative materials, the various synthetic materials that have been explored and advocated, polymer composites claim a major participation as materials. There has been a growing interest in utilizing natural fibers as reinforcement in polymer composite for making low cost construction materials in recent years. Natural fibers are prospective reinforcing materials and their use until now has been more traditional than technical. One of the most important physical factors related to the thermal conductivity of composite material, the most popular expressions for predicting the effective thermal conductivity of composite materials using the properties and volume fractions of constituent phases. In this connection, an investigation has been carried out to make use of coir: a natural fiber.

Key words: fiber, polymer, composites, thermal conductivity

1. INTRODUCTION

In our everyday life timber plays a significant role. However timber resources are getting depleted continuously while the demand for the material is ever increasing. According to the literature, by the beginning of the next century the wood will be scarce for the whole world. This situation has led to the development of alternative material. Among the various synthetic materials that have been explored and advocated, plastics claim a major share as wood substitutes. Plastics are used for almost everything from the articles of daily use to the components of complicated engineering structures and heavy industrial applications. Plastics find an extensive application in buildings as flooring material because they are resistant to abrasion have a low heat conductivity and low water absorption, sufficient hardness and strength. They fail to swell when moistened, readily take on varnishes and paints. Hardware items like door and window frames, flushing cisterns, overhead water storage tanks and water fittings are commercially available and are finding acceptance in the building industry. Plastics are used to manufacture various sanitary wares, which include wash basins, bathtubs, sinks, shower cabins, washing racks and others. Plastic pipes are widely used in the installation of various industrial purposes, water supply etc.

The knowledge of the effective thermal conductivity of composites is becoming increasingly important in many engineering application and in technological developments. Numerous theoretical and empirical models have been proposed to predict the effective thermal conductivity [1-2]. Numerous numerical studies of thermal conductivity of filled polymer were conducted in the past. Deissler's [3] works were extended by Wakao and Kato [4] for a cubic or orthorhombic array of uniform spheres in contact. Shonnard and Whitaker [5] have investigated the influence of contacts on two dimensional models. They have developed a global equation with an integral method for heat transfer in the medium. Auriault and Ene [6] have investigated the influence of the interfacial thermal barrier on the effective conductivity and on the structure of the macroscopic heat transfer equations. Using the finite elements method, Veyret at al. [7] studied the heat conductive transfer in the periodic distribution of the filler in the composite materials. In their study, calculation was carried out on two and threedimensional geometric spaces. The same
method was used by Ramani and Vaidyanathan [8] that have incorporated the effect of microstructural characteristics such as filler aspect ratio, interfacial thermal resistance, volume fraction, and filler dispersion to determine the effective thermal conductivity of a composite with spherical and parallelepiped fillers. The thermal conductivity has increased from 0.3211 W m\(^{-1}\) k\(^{-1}\) for pure PA6 to 2.09 11. W m\(^{-1}\) k\(^{-1}\) for spherical copper powder filler with a 50% volume fraction. Recently, ANSYS software was used by Liang [9], to perform the numerical simulation of the heat-transfer process in hollow-glass-bead (HGB)-filled polymer composites. The effects of the content and size of the HGB on the effective thermal conductivity was identified. The effective thermal conductivity of the polypropylene (PP)/HGB composites was estimated at temperatures varying from 25 to 30 °C. Lattice Monte Carlo (LMC) and finite element analyses were used on the ETC of sintered metallic hollow spheres structures.

2. EXPERIMENTAL

2.1. Materials

Epoxy resin used in this work was diglycidyl ether of bisphenol-A (DGEBA), Curing agent, 2-ethyl-4-methylimidazole (EMI-2,4), Amino silane coupling agent, "-aminopropyl-triethoxysilane (A1100). Other agents utilized were analytically pure grade. SiC particles with average particle size of 0.75 µm and irregularly polyhedral shape. Micrographs of the micro-SiCs and the micro-SiC/epoxy composite can be seen in our earlier works [10-11], respectively. The material supplier’s specification, COOH-MWCNTs were synthesized by catalytic chemical vapor deposition with 50– 80 nm diameter, 10–20 µm length and 125–400 aspect ratio. The purity and the specific surface area were greater than 95% and 40 m\(^2\)/g, respectively. Before stored in desiccators, COOH-MWCNTs were dried at 110°C for 24 h in vacuum to eliminate the agglomeration caused by hygroscopic absorption, as well as removing planar water, which would hinder the interaction between coupling agent and COOH-MWCNTs. Micrographs of the rod-shaped COOH-MWCNTs and the COOH-MWCNT/epoxy composite can be seen in our previous work [12]. The starting material for GNPs was graphite intercalation compound (GIC) particles (500 µm), i.e. sulfuric acid-intercalated graphite; the content of intercalants was about 15 wt% and the carbon content was higher than 99%. GNPs were prepared according to the method reported in the literature [13]. Briefly, (a) as-received GIC particles were subjected to a thermal shock on rapid exposure to 1000°C for 20 s in a muffle furnace, causing the GIC particles underwent a significant expansion (~440 times) along the thickness direction since the entrapped intercalants within GIC layers decomposed or vapored instantly, (b) the obtained expanded structures were then dipped in absolute ethanol and then broken down to GNPs using a 30 min high speed shear mixing (2400 rpm) followed by a 12 h ultrasonic irradiation (100 W, 80 kHz), (c) the GNP-absolute ethanol dispersion was filtered and the obtained GNPs were dried at 110°C to remove residue solvents. The as prepared GNPs were then kept in a dry desiccator for testing and further use.

![Figure 1. FESEM micrographs of GNPs, 5k x (a) and 60k x (b)](image-url)
Field emission scanning electron microscope (FESEM) micrographs in Figure 1 show that the as-prepared GNPs have flat and smooth morphology besides sharp corners. Image analysis calculation results based on 100 GNPs showed that the as-prepared GNPs have average diameter of 21 µm with the range of 3–30 µm and average thickness of 47 nm with the range of 20–80 nm. Then the aspect ratio can be calculated to be ~447. Fourier-transform infrared (FTIR) spectrum of as-prepared GNPs, shown in Figure 2, illustrates that there are a lot of hydroxyl groups (–OH, intense band at 3650–3050 cm⁻¹) and carboxyl functional groups (C=O, band at 1600–1200 cm⁻¹) confirmed to exist on the surface of as-prepared GNPs, indicating that the treatment of natural graphite with sulfuric acid and thermal shock of GICs resulted in some carbon double bonds oxidized, leading to the presence of oxygen-containing functional groups on the GNPs, which will facilitate their interactions with coupling agent or micro-SiCs.

2.2. Surface modification of fillers Oxidation of micro-SiCs involved (a) dipping micro-SiCs in a 10 vol% hydrofluoric acid solution for 20 min, (b) rinsing with acetone by filtration to remove the passivating thin native oxide film on the surface of micro-SiCs, (c) oxidizing micro-SiCs at 900°C for 20 h in a preheated furnace. The oxidized micro-SiCs were left in the furnace, cooling to room temperature, and then stored in desiccators. Silane treatment of fillers using A1100 involved (a) making a silane-absolute ethanol solution at 0.001 g/mL concentration, and the amounts of A1100 used were 3% by weight of the micro-SiCs and 10% by weight of the nanofillers i.e. GNPs or COOH-MWCNTs, (b) adding filler particles to the solution and stirring with a high speed magnetic stirrer (1500 rpm) at 60°C for 30 min, in addition, dispersing the solution added with nanofiller particles by high intensity ultra-sonication for 1 h, rinsing with absolute ethanol by filtration, and drying at 110°C for 1 h in vacuum. Then the silane treated fillers were stored in desiccators.

2.3. Composite preparation The composites were prepared by solution blending and casting method, which involved (a) stirring DGEBA-absolute ethanol solution at 80°C with a high speed magnetic stirrer for 20 min, (b) adding appropriate amount of as-prepared or pretreated filler particles to the solution and continuing stir for 30 min, in addition, ultrasonicating the solution added with nanofiller particles for 2 h to ensure good homogeneity, (c) cooling to 60°C, (d) adding EMI-2,4, which is 4% by weight of DGEBA, to the mixture and continuing stir for 10 min, (e) casting the mixture in mould and repeatedly degassing the mixture in vacuum drying oven at 60°C until no air bubble appears on the surface of mixture, (f) curing the mixture at 65°C for 1 h, 120°C for 1.5 h, and 160°C for 1.5 h, (g) cooling to room temperature, then de-moulding.

2.4. Characterization In this paper, morphological studies of GNPs and the fracture surfaces of composites were carried out using FESEM (Quanta 3D FEG, FEI Co., USA; LEO1550, LEO Electron Microscopy Ltd., UK). GNPs were pre-coated with a thin platinum layer. The aspect ratio of GNPs was calculated based on the measured dimensions of 100 GNPs from several FESEM images. Composites were fractured in liquid nitrogen and then the fracture surfaces were coated with a thin platinum layer before FESEM study. FTIR spectrum was recorded on a Thermo Nicolet IS-10 Smart iTR™ spectrophotometer (Thermo Fisher Scientific Co., Ltd., USA) with the smart iTR™ ATR (Attenuated Total Reflectance) accessory. Thermal diffusivity (mm²/s) at room temperature was measured on disk samples (12.7 mm diameter, 2 mm thickness) by laser flash method (nanoflash LFA 447 system, NETZSCH Instruments Co., Ltd., UK), specific heat (C, J/gK) at room temperature was measured on disk samples (6 mm diameter, 1 mm thickness) by DSC (DSC-7 system, PerkinElmer Co., Ltd., UK), and bulk density (ρ, g/cm³) of specimen was measured by water displacement. For each measurement, three samples were tested three times. After that, thermal conductivity (W/m·K). The viscosity of epoxy resin and epoxy composites at room temperature was measured using a stress rheometer (ARES9A, TA Instruments, USA) in a steady flow mode. A cone-and-plate geometry was used. A freshly prepared liquid uncured sample was dispensed on the plate before the run and the experiments were conducted under stepped shear rate from 0.01 to 100 s⁻¹.
3. RESULTS AND DISCUSSION

The filler content dependence of thermal conductivity of the oxidized and silane treated micro-SiC/epoxy composites. In this work, micro-SiCs were oxidized followed by silane treatment since it was demonstrated that the ability in improving the thermal conductivity of epoxy follows the sequence: oxidized and silane treated micro-SiC > silane treated micro-SiC > untreated micro-SiC. It can be seen that below the percolation threshold (52.1 wt%), the thermal conductivity of oxidized and silane treated micro-SiC/epoxy composites rises slowly with the increasing micro-SiC content because of a lack of continuous micro-SiC heat conductive chains, but above the percolation threshold, the thermal conductivity increases rapidly, and when 71.7 wt% oxidized and silane treated micro-SiCs were added, the thermal conductivity reached the maximum, 20.7 times that of epoxy.

Thermal conductivity of the epoxy composites containing GNPs or COOH-MWCNTs. The thermal conductivity approximately increases linearly with the increasing nanofiller content for all the investigated composites. More importantly, at identically low nanofiller contents (6 wt%) silane treated GNPs produced epoxy composites with higher thermal conductivities than silane treated MWCNTs despite the inherently lower thermal conductivity of individual GNP (250 W/(m·K)), the flat surface of 2-D GNPs dramatically enhances the GNP/epoxy or GNP/GNP contact area, moreover, the rigidity of 2-D GNPs allows for better preservation of their high aspect ratio in comparison with the more flexible 1-D MWCNTs, thus 2-D GNPs are more efficient in forming heat conductive networks in epoxy matrix as compared with 1-D MWCNTs, and ii) in sharp contrast to 1-D MWCNTs, the flat surface of 2-D GNPs minimizes the geometric contribution to the thermal interface resistance since the contribution of phonon acoustic mismatch to the interface contact resistance increases with the decreasing radius of nanoparticles. It is noticed that the use of inorganic nanofillers has proven to be effective in reducing the filler content required for relatively high thermal conductivity and thereby minimizing the problems associated with density and mechanical properties, and in this work silane treated GNPs did efficiently improve the thermal conductivity of epoxy with low loadings and thus were considered attractively good nanofiller candidates.

4. CONCLUSIONS

In this study, 2D-GNPs with high aspect ratio (447) were used to improve the thermal conductivity of epoxy. By adding 12 wt% GNPs or 71.7 wt% micro-SiCs to an epoxy resin the thermal conductivities of the composites reached maxima that were respectively 6.3 and 20.7 times that of the epoxy alone. To further improve the thermal conductivity a composite designing way, i.e. partial replacement method that partially replaces micro-SiCs with GNPs, was utilized to unite the respective advantage of microfiller and nanofiller in improving the thermal conductivity of epoxy. Epoxy composites with a mixture of GNPs and micro-SiCs were prepared, and a thermal conductivity, 26.1 times that of the epoxy, was obtained with 7 wt% GNPs + 53 wt% micro-SiCs, thus not only break the bottleneck of further improving the thermal conductivity of epoxy composites but also broaden the applications of GNPs.

REFERENCES


