Composites Communications 5 (2017) 36–39

Contents lists available at ScienceDirect

Composites Communications

journal homepage: www.elsevier.com/locate/coco

Mechanical, thermal and dielectric behavior of hybrid filler polypropylene composites

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ARTICLE INFO

Keywords:
Polypropylene
Aluminium nitride
Solid glass microsphere
Microelectronics application

ABSTRACT

In this study, thermal and dielectric behaviour of particle reinforced polymer composites are investigated experimentally. The main aim is to explore composite materials based on polymer matrix for microelectronics application. The continuous development of electronic devices requires optimum solutions for heat dissipation. This prompts the needs to the development of unique polymer composites that possess high thermal conductivity and low dielectric constant. In present work, hybrid composites were fabricated with micro-sized aluminum nitride and solid glass microspheres as filler material in polypropylene matrix in order to achieve the required property values. Experimental results are discussed in light and concluded that this new class of polymer composite can finds its potential application in future electronic packaging materials.

1. Introduction

For most of the semi-conductor industry's history, engineering effort has been focused on higher level of integration, improved performance and speed. The ever increasing demand for such high performance electronic devices necessitates new materials to be used in microelectronics application that possess superior properties when matched to the conventionally used ones. These materials should satisfy certain diverse requirements such as low relative permittivity to reduce the signal propagation delay, low dielectric loss for better device performance, high thermal conductivity to dissipate the heat generated, low coefficient of thermal expansion (CTE) to maintain desired dimensional stability, moisture absorption resistance and mechanical flexibility [1]. The polymeric materials in the components are increasingly important as thermal paths for the removal of excess heat that builds up and may cause failure. Unfortunately, polymeric materials are inherently poor thermal conductors, and they must be modified to assist in heat removal from electronics [2].

It has been seen that single filler cannot improve the thermal conductivity and decrease the dielectric constant of the composite simultaneously [3–5]. So the incorporation of hybrid fillers in terms of a combination of high thermal conductive with low dielectric constant filler to the polymer matrix is suggested. Among the various fillers, an inorganic crystalline filler i.e. aluminum nitride (AlN) which are combined individually with an inorganic amorphous filler i.e. solid glass micro-spheres (SGM) have been considered superlative candidates for hybrid filler polymer composite for the present investigation owing to high thermal conductivity of Al_2O_3 and low dielectric constant of SGM. Moreover, both the fillers are non-toxic and are having low CTE. In view of this, the present work has been undertaken to study the effect of adding hybrid filler on various mechanical, thermal and dielectric properties for such composite. Also, the measured effective thermal conductivity (k_{eff}) is compared with calculated values obtained from the mathematical model developed by the authors in their previous work [6].

2. Models for effective thermal conductivity for composites

Predicting effective thermal conductivity analytically is an important step to obtain thermal conductivity through constitutional design and decrease the development cost of new composites. To predict the k_{eff} of composite materials, several theoretical models have been proposed in the past [6–9]. All the models discussed above are applicable for composite with single filler only and not much work has been reported regarding mathematical model for hybrid filler. However this aspect has been considered in a recently proposed model by the authors [6]. The expression for effective thermal conductivity as per proposed model is given by:

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http://dx.doi.org/10.1016/j.coco.2017.06.005
Received 8 December 2016; Received in revised form 16 June 2017; Accepted 26 June 2017
Available online 11 July 2017

$$k_{eff} = 2 \times \left[\left(\frac{1}{k_p} - \frac{1}{k_p}\left(\frac{12\phi_a}{\pi}\right)^{\frac{1}{3}} + \frac{2}{\left(k_p\left(\frac{2\pi}{3\phi_a}\right)^{\frac{1}{3}} + \left(\frac{4\phi_a}{9\pi}\right)^{\frac{1}{3}}\pi(k_a - k_p)\right)} \right) + \left(\frac{1}{k_p} - \frac{1}{k_p}\left(\frac{12\phi_b}{\pi}\right)^{\frac{1}{3}} + \frac{2}{\left(k_p\left(\frac{2\pi}{3\phi_b}\right)^{\frac{1}{3}} + \left(\frac{4\phi_b}{9\pi}\right)^{\frac{1}{3}}\pi(k_b - k_p)\right)} \right) \right]^{-1} \quad (1)$$

where Φ and k represents the volume fraction and thermal conductivity respectively and suffix a, b and p are for filler A, filler B and matrix material respectively.

3. Experimental details

3.1. Material considered

Homopolymer polypropylene, resin belonging to the thermoplastic group is used as a matrix material. It possesses very low density of 0.92 g/cm^3. Polypropylene (PP) used in present work have tensile strength of 45 MPa, compressive strength of 83 MPa and its hardness is 0.059 GPa. CTE of PP is quite high (111.5 × 10^{-6}/°C) but have very low dielectric constant (2.25 at 1 MHz) and is procured from Ciba Geigy Limited, India. Aluminium nitride is used as primary filler materials and has been procured from M/s Alfa Aesar Limited-Beijing, China. The average size of AlN used is about 60–70 µm. AlN powder possesses unique combination of high thermal conductivity (160 W/m-K), good dielectric properties (8.9 at 1 MHz) and low CTE (4.5 × 10^{-6}/°C). Solid glass microspheres (SGMs) with an average size of 90–100 µm procured from NICE Limited located at Bangalore, India is used as the second filler material. They have lower CTE (7.5 × 10^{-6}/°C), low dielectric constant (2.25 at 1 MHz), high compressive strength and high surface hardness with improved surface smoothness.

3.2. Composite fabrication

For fabricating different set of composites, AlN and SGM are added in polypropylene resin. Composites are fabricated using compression molding technique. Rheomix 600 batch mixer is used to melt and mix PP and fillers. The mixing is done as per ASTM standard D-2538. As the mixing is over, the material is taken out from the chamber and it is kept in a hot air oven for about an hour. It is then taken for compression molding. Materials are kept in die which is used to make a composite sheet of thickness 3 mm and area 150 × 150 mm^2. Compression moulding is done as per ASTM D-256 standard. Composites of six different compositions with wide range of filler content are fabricated. The compositions of various samples prepared for present work is shown in Table 1.

Table 1
Polypropylene based composites filled with different inorganic fillers.

S. No.	Composition
1	PP + 5.0 vol% AlN + 5.0 vol% SGM
2	PP + 10.0 vol% AlN + 10.0 vol% SGM
3	PP + 15.0 vol% AlN + 15.0 vol% SGM
4	PP + 5.0 vol% AlN + 10.0 vol% SGM
5	PP + 15.0 vol% AlN + 10.0 vol% SGM
6	PP + 20.0 vol% AlN + 10.0 vol% SGM

3.3. Characterization

Computerized Instron 1195 universal testing machine is used to measure the strength of the composites. Micro-hardness measurement is done using a Leitz micro-hardness tester. Unitherm™ Model 2022 is used to measure thermal conductivity of the fabricated composite. The tests are done in accordance with ASTM E-1530 standard. Coefficient of thermal expansion is measured with the help of Perkin-Elmer thermal mechanical analyzer (TMA-7). Hioki 3532-50 Hi Tester LCR analyzer with an applied AC voltage of 500 mV is used to measure dielectric constant (ε_c) in the frequency range of 1 kHz–1 MHz.

4. Results and discussion

4.1. Mechanical properties

In the present work, micro-hardness, tensile strength and compressive strength values are measured and the test results for all the fabricated composites are presented in Table 2. It is evident from the tables that with addition of fillers, micro-hardness of the composites improved with increase in filler content. The mean micro-hardness values of the PP composites exhibit improved hardness with the increase in the content of AlN and also with increasing SGM content. With 15 vol% AlN and 15 vol% SGM reinforced in PP matrix, an improvement of about 440% in the value of micro-hardness is obtained. It is noticed that with addition of fillers, tensile strength of polymers decreases. However, the rates of decrease are quite marginal. Tensile strength value reduces to 39.9 MPa i.e. by 11.3% with 20 vol% AlN and 10 vol% SGM. This reduction is may be due to the fact that chemical bond strength between filler particles and the matrix body is not adequately strong to transfer the tensile load. The other reason for the decrement of tensile strength may be due to the increase in void percentage in the composites with increase in filler content. Compressive strengths of the fabricated specimens are evaluated and is noticed that with addition of fillers, compressive strength of polymers increases with filler content. There is an increment of 46% with 20 vol% AlN and 10 vol% SGM combination is observed. The compressive strength of PP which is 83 MPa increases to 121.2 MPa. It is mainly because of the high compressive strength of filler material. Further, in a compression test, any crack or flaw introduced by dispersion of the filler will get healed and made ineffective, contrary to the crack opening mechanism occurring in a tensile loading situation.

4.2. Thermal properties

The effective thermal conductivity values of all the fabricated samples are represented in the form of bar graph as shown in Fig. 1. The calculated thermal conductivity values using proposed theoretical model and the measured values are shown and compared. This comparison is done for a wide range of filler content and found that values obtained from the proposed model are in close approximation with the measured values. The various assumptions taken while deriving the correlation is the reason of marginal deviation observed between the calculated and measure values. Among the various sets of fabricated composites, a maximum k_{eff} value of 0.397 W/m K is obtained for combination of 20 vol% AlN and 10 vol% SGM. This improvement in k_{eff} amounts to about 261% compared to the neat PP. The coefficient of thermal expansion values of all the fabricated composites in present work are shown in Fig. 2. It can be seen from the figures that by increasing the content of either of the fillers, coefficient of thermal expansion of the respective composite system decreases appreciably. It can also be seen that with presently used hybrid fillers, a uniform decreasing trend is obtained unlike the variable trend in case of single filler reinforced PP composites [10]. This uniform trend is because of the presence of perfectly spherical shape SGM which is pre-mixed with Al_2O_3. SGM in present case helps in restricting the formation of any