Composites: Part B 56 (2014) 431–434

Contents lists available at ScienceDirect

Composites: Part B

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

Estimation of thermal conductivity for polypropylene/hollow glass bead composites

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A R T I C L E I N F O

Article history:
Received 10 March 2013
Received in revised form 7 July 2013
Accepted 19 August 2013
Available online 29 August 2013

Keywords:
A. Polymer–matrix composites
B. Thermal properties
C. Analytical modeling
D. Thermal analysis

A B S T R A C T

The thermal conductivity of hollow glass bead (HGB)-filled polypropylene (PP) composites was estimated using the thermal conductivity equation of inorganic hollow microsphere-filled polymer composites published in the previous paper. The estimations were compared with the measured data of the PP composites filled with two kinds of HGB with different size (the mean diameter was respectively 35 μm and 70 μm). The results showed that the predictions of the thermal conductivity were in good agreement with the measured data except to individual data points. Furthermore, both the estimated and measured thermal conductivity decreased roughly linearly with increasing the HGB volume fraction when the HGB volume fraction was less than 20%; the influence of the particle diameter on the thermal conductivity was insignificant.

1. Introduction

Thermal conductive polymeric composites have been paid more and more attention in the past two decades [1–4]. Rigid hollow microspheres (e.g. hollow glass beads, hollow ceramic beads, rigid hollow plastic beads, etc.) contain inert gas, and have some advantages, such as low thermal conductivity coefficient and density. In addition, these micro-particles do not generate important stress concentration in the interface between the fillers and the matrix owing to their smooth spherical surface [5]. They are, therefore, usually used to fill and modify resins in polymeric industry and coating industry. Generally, polymer/hollow micro-sphere composites have good thermal and sound insulation, low density and good mechanical and rheological properties [6–13]. This type of composite is applied in building materials, space-flight and the aviation industry.

The heat transfer process in porous materials is very complicated, especially for polymer composites. It is quite important, therefore, to understand the heat transfer mechanisms of heat transfer in polymer composites. Thermal conductivity is an important characteristic of heat transfer properties of materials [14]. For porous materials, several researchers [15,16] derived some thermal conductivity equations based on the Maxwell expression, or established a more accurate formula for calculating the effective thermal conductivity of porous materials [17]. Relatively, the models proposed respectively by Nielsen [18] and Cheng–Vachon [19] may better estimate the thermal conductivity of filled composite materials, while the Agari–Nagai equation can predict the thermal conductivity of the composites with high-loading [20]. Liang [21] analyzed the thermal conductivity of a porous material with closed spherical and cylindrical holes. Suvorov et al. [22] studied the thermal conductivity of hollow emery filled composites. Recently, Hill and Supancic [23] proposed an indirect method to determine this interfacial boundary resistance by preparing large-scale "macro-model" simulations of the polymer–ceramic interface. They also investigated the effects of similar size and shape of platelet-shaped particles on the thermal conductivity of polymer/ceramic composite materials [24]. Yu et al. [25] measured the thermal conductivity of polystyrene–aluminum nitride composite, and found that the thermal conductivity of composites was higher for a polystyrene particle size of 2 mm than that for a particle size of 0.15 mm. The thermal conductivity of the composite was five times that of pure polystyrene at about 20% volume fraction of aluminum nitride (AIN) for the composite containing 2 mm polystyrene particles.

Hollow glass bead (HGB) contains inertia gas, and have some advantages, such as low thermal conductivity coefficient and light. In addition, these hollow micro-particles do not generate important stress concentration in the interface between the inclusions and the matrix owing to their smooth spherical surface. In the previous work, Liang and Li [6] measured the thermal conductivity (k_{eff}) of HGB-filled polypropylene (PP) composites by means of a thermal conductivity instrument, and simulated the heat-transfer process in PP/HGB composites by using finite element method (FEM) with ANSYS software [26]. More recently, Liang and Li [27] analyzed the heat transfer mechanisms in polymer/hollow

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http://dx.doi.org/10.1016/j.compositesb.2013.08.072

microspheres composites, and established a mathematical model for predicting the thermal conductivity. The objectives in this paper are to predict the thermal conductivity of the PP/HGB composites applying this mathematical model and to compare the predictions with the experimental data.

2. Modeling

2.1. Heat transfer mechanisms in polymer/hollow micro-sphere composites

In general, the thermal conductivity in thermal insulation materials is the combined effect of heat conduction, convection and radiation. According to the second law of thermodynamics, heat always transfer spontaneously from high temperature body to low temperature one. Namely, heat transfer will conduct in where there is difference in temperature. Generally, insulation materials only reduce the strength of heat exchange, and have a property of blocking heat transfer. Polymer/hollow micro-spheres composite is a kind of ternary composites, it includes three phases, namely resin, gas and spherical shell. During heat transfer in polymer/hollow micro-spheres composites, when heat quantity is close to a hollow microsphere, only a small part of heat quantity will conduct by it, while greater part of heat quantity will move around it due to its low conductivity, as shown in Fig. 1. Because of low thermal conductive coefficient of the hollow micro-spheres and longer heat transfer route and complication in the filled systems, the thermal conductivity of these composites will be reduced.

It can be seen from Fig. 1 that the heat transport in inorganic hollow microsphere filled polymer composites has three kinds of ways: (1) thermal conduction by solid; (2) heat radiation on the surface between neighboring hollow particles; (3) the natural thermal convection of gas in the hollow particles. After finishing the experiments, Skochdopole [28] pointed out that the natural thermal convection of the gas in a micro-bubble would not occur when the bubble diameter was less than 4 mm. Because the diameter of the hollow micro-spheres as fillers is usually less than 0.1 mm, the natural thermal convection of the gas in it may be neglected. Furthermore, polymer composite works usually under lower temperature conditions where the proportion of the thermal radiation in the total heat transfer is very small, hence the thermal radiation may also be neglected.

Generally, the heat transfer process in inorganic hollow microspheres filled polymer composites is more complicated, because they are a type of material with three phases, namely resin, gas and spherical shell.

2.2. Effective thermal conductivity equation of polymer/hollow micro-sphere composites

On the basis of the law of minimal thermal resistance [29] and the equal law of the specific equivalent thermal conductivity, a mathematical model for describing the relationship between the effective thermal conductivity and other materials parameters of polymer/hollow micro-sphere composites is derived [27]:

$$k_{eff} = \left\{ \frac{1}{k_p}\left(1-\frac{6\phi_f}{\pi}\right)^{\frac{1}{3}} + 2\left[k_p\left(\frac{4\pi}{3\phi_f}\right)^{\frac{1}{3}} + \pi\left(\frac{2\phi_f}{9\pi}\right)^{\frac{1}{3}}\left(k_g\left(\frac{\rho_s-\rho_a}{\rho_g-\rho_a}\right) + k_a\left(\frac{\rho_g-\rho_s}{\rho_g-\rho_a}\right) - k_p\right)\right]^{-1}\right\}^{-1} \quad (1)$$

where ρ_g, ρ_a and ρ_s are the effective densities of the spherical shell, gas and micro-sphere respectively, ϕ_f is the volume fraction of the hollow micro-spheres, k_p, k_g and k_a are the thermal conductivities of polymer matrix phase, micro-spherical shell phase and gas phase, respectively.

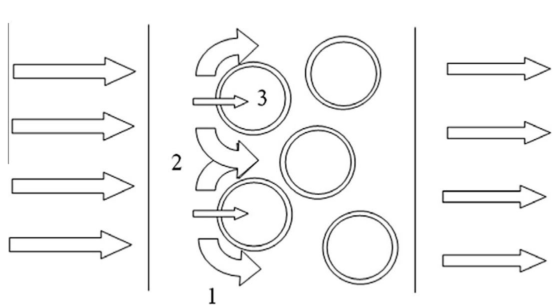

Fig. 1. Heat transfer model of polymer/hollow sphere composites.

3. Experimental

3.1. Raw materials

An injection grade of polypropylene (PP) with trade mark of CJS-700, supplied by Guangzhou petrochemical Co., Ltd. in China, was used as the matrix resin, the density and melt flow index (230 °C, 2.16 kg) of the resin were 0.91 g/cm^3 and 12 g/10 min, respectively.

Two kinds of hollow glass beads (HGB) supplied by Molüs Co., Ltd. in German, TK35 and TK70, with different size were used as the fillers in this work. The mean diameters of the fillers were 35 μm and 70 μm, and the density was 680 kg/m^3 and 210 kg/m^3, respectively. The surface of the particles was pretreated with silane coupling agent by the supplier. The particle size distribution of the fillers was measured by means of a laser size instrument (Model LS-C(I)) supplied by Omik Co., Ltd. in Zhuhai, China.

3.2. Sample preparation

After mixing simply, the PP resin and the HGB with different proportions were compounded in a twin-screw extruder [6]. The blending was conducted in a temperature range of 160–230 °C and screw speed of 25 r/min, and then the extrudate was granulated to produce the composites. The volume fractions of the HGB were 0%, 5%, 10%, 15% and 20%. The specimens for thermal conductivity measurement were molded by using an injection molding machine in temperature range of 160–240 °C after drying the composites. The specimens were the square plates; the length and thickness of the specimen were 50 mm and 6 mm, respectively.

3.3. Apparatus and methodology

The thermal conductivity of the composites was measured by means of a protecting heat plate method in this test, and the main apparatus was a protecting heat flow type of thermal conductivity instrument (model NF-7) supplied by South China University of Technology. Two thermocouples were set on the two cold faces of the test pieces, and two thermocouples were set on the two heated face of the test pieces. The temperatures and heating power were measured as soon as the heat transfer reached the steady state. All measured data were collected and recorded by a computer. The environmental temperature for test was 27 °C. The specimens was plates with length of 50 mm, width of 50 mm and thickness of 6 mm. 4 measuring points were set up equally on a plate, and the average was reported for each specimen [6]. The physical property parameters including density and thermal