



Thermal conductivity of polymer composites with oriented boron nitride[☆]



Hong Jun Ahn^a, Young Jun Eoh^a, Sung Dae Park^b, Eung Soo Kim^{a,*}

^a Department of Materials Engineering, Kyonggi University, Suwon, South Korea

^b Electronic Materials & Device Research Center, Korea Electronics Technology Institute, Seongnam, South Korea

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ABSTRACT

Thermal conductivity of boron nitride (BN) with polyvinyl alcohol (PVA) and/or polyvinyl butyral (PVB) was investigated as a function of the degree of BN orientation, the numbers of hydroxyl groups in the polymer matrices and the amphiphilic agents used. The composites with in-plane orientation of BN showed a higher thermal conductivity than the composites with out-of-plane orientation of BN due to the increase of thermal pathway. For a given BN content, the composites with in-plane orientation of BN/PVA showed higher thermal conductivity than the composites with in-plane orientation of BN/PVB. This result could be attributed to the improved degree of orientation of BN, caused by a larger number of hydroxyl groups being present. Those treated with C₁₄H₆O₈ amphiphilic agent demonstrated a higher thermal conductivity than those treated by C₂₇H₂₇N₃O₂. The measured thermal conductivity of the composites was compared with that predicted by the several theoretical models.

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1. Introduction

With a high integration of various components in electronic devices, heat dissipation becomes critical not only to maintain the life span of the device, but also to its performance and reliability. Ceramic/polymer composites have therefore been in particularly high from the electronics industry, in order to develop composites with a high degree of thermal conductivity. Numerous investigations have previously been conducted into improving the thermal conductivity of ceramic/polymer composites, including the following: the addition of ceramics with a high thermal conductivity, varying the orientation of anisotropic ceramics and changing the ceramic/polymer interface [1–3]. Furthermore, several types of numerical models have also been suggested to predict the thermal conductivity of composites, and to discover potential new composites more effectively [4–7].

Although the hexagonal boron nitride (h-BN) is widely used in ceramic/polymer composites due to its higher thermal conductivity, the thermal conductivity of the composite strongly depends on the anisotropy of platelet BN which changes its thermal

conductivity. Therefore, the thermal conductivity of the BN/polymer composite should be studied as a function of the orientation of BN to the heat flows.

On the other hand, in order to develop ceramic/polymer composites with a high thermal conductivity that are suitable for a variety of applications, the interfacial behaviour between ceramics and polymers needs to be controlled based on a fundamental understanding of the factors affecting it. The difference in surface properties between hydrophobic ceramics and hydrophilic polymers is particularly important, and it is expected that these properties could be controlled by the use of amphiphilic agents. Generally, an amphiphilic agent is composed of two groups; one is hydrophobic groups worked for hydrophobic ceramics, the other is hydrophilic groups worked for hydrophilic polymer. The interfacial behaviour between ceramics and polymers could be improved due to these two groups of amphiphilic agents.

From a preliminary study, the thermal conductivity of a boron nitride (BN) composite treated with the amphiphilic agent C₂₇H₂₇N₃O₂ was found to be higher than that of pristine BN. This result agrees with a report on BN nanotubes treated by catechin, which showed an improved thermal conductivity over pure BN nanotubes [8]. However, it is still unclear whether the thermal conductivity of BN/polymer composites was affected by the number of hydroxyl groups of amphiphilic agent or not.

In this study, the thermal conductivities of BN/polymer composites were investigated as a function of the degree of

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* Corresponding author.

E-mail address: eskim@kyonggi.ac.kr (E.S. Kim).

orientation of BN, and the types of amphiphilic agents with different number of hydroxyl groups. Two polyvinyl matrix materials were selected with polyvinyl alcohol (PVA) having a larger number of hydroxyl groups than polyvinyl butyral (PVB). The amphiphilic treatment agents $C_{27}H_{27}N_3O$ and $C_{14}H_6O_8$ have the same number of aromatic rings, yet the number of hydroxyl groups in $C_{14}H_6O_8$ is four times greater than in $C_{27}H_{27}N_3O_2$. It is thought that the aromatic rings of amphiphilic agent promote strong π - π interactions with the BN surface [8]. Also, hydroxyl groups of amphiphilic agent promote hydrogen bonding with polymer matrix. Fig. 1 illustrates the mechanism scheme of interaction between BN treated amphiphilic agent and polymer matrix. The several different thermal conductivity models were examined for predicting the thermal conductivity of a polymer composite with oriented BN was also discussed.

2. Experimental procedure

Starting materials were 8- μ m-sized boron nitride (BN, DENKA, Japan) particles as ceramic filler, and polyvinyl butyral (PVB, BM2L380, CS Technology, Korea) or polyvinyl alcohol (PVA, Kuraray Poval, Kuraray Co., Ltd., Japan) as the polymer matrices; 2-(4,6-diphenyl-1,3,5-triazin-2-yl)-5-[(hexyl)oxy]-phenol ($C_{27}H_{27}N_3O_2$, Sigma-Aldrich, USA) and ellagic acid hydrate ($C_{14}H_6O_8 \cdot xH_2O$, Sigma-Aldrich, UK) were used for the amphiphilic agents. To prepare the amphiphilic-agent-treated BN, the volume fraction of the $C_{27}H_{27}N_3O_2$ and $C_{14}H_6O_8$ to the BN was determined to 0.16 and 0.10 V_f , respectively due to the difference of densities with amphiphilic agent type. $C_{27}H_{27}N_3O_2$ and $C_{14}H_6O_8 \cdot xH_2O$ were stirred with 8- μ m-sized BN powders in toluene and dimethyl sulfoxide (DMSO), respectively, for 6 h at 150 °C in concentration of 20 wt.%. The treated BN powders were filtered through a suction flask by three times ethanol washing, with the remaining powder then dried at 80 °C for 24 h.

To produce BN/PVB composites, liquid PVB and amphiphilic-agent-treated BN powder were mixed by magnetic stirring for 4 h at room temperature. For BN/PVA composites, amphiphilic-agent-treated BN powder was dispersed in a 10 wt.% PVA aqueous solution, which was then mixed by magnetic stirring at 80 °C for 5 h, and then dried. The mixed BN/polymer slurries were de-aired in a vacuum chamber, and the viscosity of slurry was adjusted to 2000 mPa.s. To obtain composites filled with oriented ceramic

particles, the de-aired slurry was poured into a tape-casting machine and then laminated to a height of 5 mm at 65 °C under the pressure of 5 MPa. The significant orientation is expected in tape casting, where a strong field of shear stress is presented particularly at the boundary of the blade of the carrier sheet and vicinity. This laminated sheet was hot-pressed at 90 °C under 10 MPa. For the preparation of composite with a specific orientation, the laminated bulks, $5 \times 5 \times 5 \text{ mm}^3$, were first produced by cutting, and then the specimens with $5 \times 5 \times 1 \text{ mm}^3$ were cut from these laminated bulks. The laminated bulks of BN platelets which were perpendicular (out-of-plane orientation) to the thermal path, were used to measure the thermal diffusivity of the BN/polymer composites with out-of-plane orientation. For the thermal diffusivity of the composites with in-plane orientation, the laminate bulks were rotated 90° and finally BN platelet were aligned parallel (in-plane orientation) to the thermal path.

X-ray diffraction analysis (XRD, Model D/Max-3C, Rigaku, Japan) was used to determine the degree of orientation (D) for BN ceramics in polymer composites by comparing the relative intensity of the (100) peak, to the sum of the (100) and (002) peaks. ($D = I_{(100)} / (I_{(100)} + I_{(002)}) \times 100\%$) Orientation of the platelet BN in composites was also observed using a scanning electron microscope (SEM, Model JSM-7500F, Jeol, Japan).

The density of the specimens was measured by a gas pycnometer (GP, Model AccuPyc1330, Micromeritics, USA).

The volume fraction (V_f) was calculated using Eq. (1):

$$V_f = \frac{(W_f / \rho_f)}{(W_f / \rho_f + W_m / \rho_m)} \quad (1)$$

where, V_f , W_f , W_m , ρ_f , and ρ_m are the volume fraction of the filler, and the weight of the filler, and the matrix, and the density of the filler, and the matrix, respectively.

The BN composites treated by amphiphilic agents were investigated by Fourier-transform infrared spectroscopy (FT-IR), using a Bruker IFS-66/S spectrometer (Bruker UK Ltd., Conventry, UK) in the 4000–400 cm^{-1} range. Thermogravimetric analysis was performed using a NETZSCH STA 409 PC/PG (STA 409 PC/PG, Netzsch, Germany) instrument from room temperature to 1000 °C at a heating rate of 10 °C/min under air atmosphere.

The thermal diffusivity of composites with orientated BN was measured by a non-contact laser flash method (Model LFA-447, Netzsch, Germany) [9], with the laser pulse directed through the

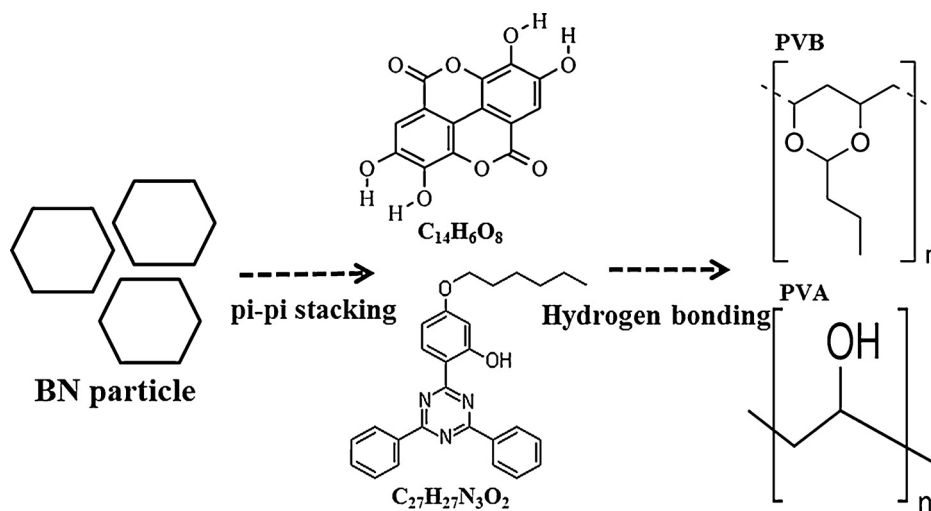


Fig. 1. Scheme of interaction between BN treated amphiphilic agent and polymer matrices.

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