



Enhancing the heat and load transfer efficiency by optimizing the interface of hexagonal boron nitride/elastomer nanocomposites for thermal management applications



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ARTICLE INFO

Article history:

Received 5 February 2018

Received in revised form

29 March 2018

Accepted 30 March 2018

Available online 2 April 2018

Keywords:

Thermal conductivity

Boron nitride

Interface

Functionalization

Thermo-physical properties

ABSTRACT

Amine-terminated butadiene-acrylonitrile (ATBN) was covalently grafted on a hexagonal boron nitride (BN) surface to enhance the interface affinity between BN and a carboxylated styrene-butadiene rubber (XSBR) matrix for preparing thermal interface materials with robust thermo-physical properties. The strong attraction between the amino groups on the ATBN-functionalized BN (ABN) and the carboxyl groups on the XSBR chains can further enhance the interfacial adhesion between the fillers and polymer matrix. The mechanical properties, thermal stability, thermal conductivity, thermo-physical properties, and interfacial filler–matrix network of the prepared XSBR/ABN nanocomposites were studied. The XSBR/ABN nanocomposites exhibited better mechanical and thermo-physical behavior compared to the XSBR/BN nanocomposites over the entire range of filler concentration, representing an enhanced interfacial interaction owing to functionalization. Moreover, the XSBR/ABN nanocomposites exhibited superior thermal conductivity as compared to the XSBR/BN nanocomposites, with the former exhibiting approximately 490% higher thermal conductivity than neat XSBR, whereas the latter exhibited 395% higher thermal conductivity than neat XSBR at a filler loading of 90 phr.

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1. Introduction

Recently, with the advances in technology, the miniaturization of electronic devices is confronted with the issue of heat dissipation, which has urgently highlighted the demand for superior thermal interface materials for modern chip packaging [1]. The lifespan of an electronic component is profoundly dependent on the operating temperature; therefore, the enhancement of the heat diffusion rate for maintaining the required device operating temperature is attracting increasing interest [2,3]. Consequently, polymer composites incorporated with high thermal-conductivity fillers are emerging as highly efficient alternatives for addressing thermal management problems [4,5].

Thermal conductivity is the total quantity of heat that diffuses through a plate of a particular area and thickness per unit time until its opposite faces differ by 1° in temperature [6]. In the past few decades, high thermal-conductivity fillers, such as carbon nanotubes [7,8], graphene [9,10], (expanded) graphite [11,12,13], and

boron nitride (BN) [14,15,16], have been the most widely employed fillers for enhancing the thermal conductivity of polymer composites. Among them, carbon-based materials have different degrees of electrical conductivity, which limits their practical usage in electric devices. Consequently, insulated BN became one of the most favorable candidates for enhancing the heat diffusion of polymer composites. Li et al. [17] developed a new thermally conductive polyimide (PI)/BN composite film using 3-mercaptopropionic acid as the surfactant to modify the BN surface for the dispersion of BN in the polymer, and the thermal conductivity of the PI composite containing 30 wt.% of micro- and nano-sized BN filler could reach 1.2 W/mK. In the study by Huang et al. [18], a dielectric thermally conductive epoxy nanocomposite was fabricated using polyhedral oligosilsesquioxane-functionalized BN nanotubes (BNNTs) as fillers. Compared to the pristine epoxy resin, the prepared nanocomposites presented a significant thermal conductivity enhancement of 1360% when the BNNT loading fraction was 30 wt.%. Terao et al. fabricated BNNT/polyvinyl alcohol composite fibers via electrospinning to maintain the alignment of BNNTs in the fiber casting direction and the highest values (0.54 W/mK) were obtained along the long axes of aligned BNNTs.

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In addition to incorporating a high thermal-conductivity filler, improving the interfacial affinity between the filler and polymer matrix is a non-ignorable factor for enhancing the thermal conductivity of polymer composites [19]. A high interfacial affinity can significantly reduce the interfacial thermal resistance, thus hastening the heat diffusion. Yu et al. [20] prepared BN nanoplatelets using a sonication-centrifugation technique and investigated the effects of non-covalent and covalent functionalization of BN nanoplatelets on the thermal properties of epoxy composites. They determined that the composites with a stronger interface incorporated with covalently functionalized BN exhibited significantly enhanced thermal conductivity and dynamic thermal mechanical modulus, whereas the composites with non-covalently functionalized BN representing an intermediate interface interaction exhibited a moderate improvement in thermal conductivity.

In this paper, a novel idea of grafting amino-terminated macromolecular chains (amine-terminated butadiene-acrylonitrile (ATBN)) onto BN surfaces to enhance the interface affinity between the BN and rubber (carboxylated styrene-butadiene rubber (XSBR)) matrix is proposed, and simultaneously, the terminal amino groups react with the terminal carboxyl groups on the rubber chains to further enhance the interfacial interaction between the inorganic filler and organic polymer matrix. Physically robust rubber-based nanocomposites were prepared and their thermal conductivity and thermo-physical properties were comprehensively compared and studied.

2. Experimental

2.1. Materials

Latex of carboxylated styrene butadiene rubber (XSBR) KSL 203 was purchased from Korea Kum Ho Petrochemical Company. BN particles with purity of 98% and lateral size of 1 μm were purchased from Sigma–Aldrich. 4, 4'-diphenylmethane diisocyanate (MDI) and *N,N*-dimethylmethanamide (DMF) were purchased from Sigma–Aldrich. ATBN with an acrylonitrile content of 18% was purchased from Shenzhen Jiadida Chemical Co., China. Rubber additives such as sulfur, stearic acid, zinc oxide, dibenzothiazole disulfide (DD), and *n*-cyclohexyl-2-benzothiazole sulfonamide (CBS) are commercially available.

2.2. Synthesis of ATBN-terminated BN (ABN)

Fig. 1(a) shows the schematic of the surface functionalization of

BN. BN nanoplatelet was prepared via the exfoliation of BN particles by sonication for 8 h. Subsequently, the as-prepared BN nanosheets were treated in 5 M NaOH solutions 85 °C to introduce the hydroxide groups [14] and thereafter, the product was dispersed in DMF via ultrasonication for 40 min. Subsequently, 6 g MDI powder was dissolved in the solution and the mixture was subjected to water bath and stirred at 80 °C for 12 h and then washed with ethanol and de-ionized water to remove the residual MDI. The resultant is designated as BN-NCO where “-NCO” represents the isocyanate groups.

For preparing ABN, BN-NCO was dispersed in DMF and treated by sonication for 20 min. Subsequently, a solution of 10 wt% ATBN in DMF was added by stirring and the reaction was conducted at 80 °C for 36 h. The solid product was collected by washing and filtration.

2.3. Preparation of XSBR/ABN and XSBR/BN nanocomposites

Latex compounding, which can confirm homogeneous filler dispersion, was employed to prepare the nanocomposites. Quantitative XSBR latex and nanofiller aqueous suspension were directly mixed, followed by vigorous stirring for 20 min for each sample. The emulsion mixture was thereafter immediately co-coagulated using 0.5% wt. of a CaCl_2 aqueous solution, and subsequently dried in an oven at 70 °C for 48 h. Rubber vulcanizing ingredients and additives were compounded with the dried prefabricated samples on a conventional two-roll mill according to the standard vulcanization formulation, as presented in Table 1. The as-compounded nanocomposites were vulcanized by hot-pressing at 160 °C and a pressure of 10 MPa for the optimum cure time (t_{c90}).

2.4. Characterization

FT-IR spectra of the fillers was recorded using Perkin Elmer Spectrum 100 S. X-ray diffraction (D2 Phaser, Bruker) was carried out at ambient temperature from 5° to 80° (2 θ). N_2 adsorption–desorption isotherm was determined at 77 K by employing a gas adsorption analyzer (BEL–SORP; BEL Co., Japan). The thermal stabilities of the fillers and nanocomposites were evaluated using thermogravimetric analysis (TG 209 F3, NETZSCH, Germany) with a heating rate of 10 °C min^{-1} under the N_2 atmosphere. X-ray photoelectron spectroscopy (XPS, VG Scientific Co., ESCA LAB MK-II) analyses were carried out to confirm the surface chemistry of the functionalized BN-NCO and ABN. Transmission electron microscopy (TEM) was performed on a JEM-2100 F (JEOL,

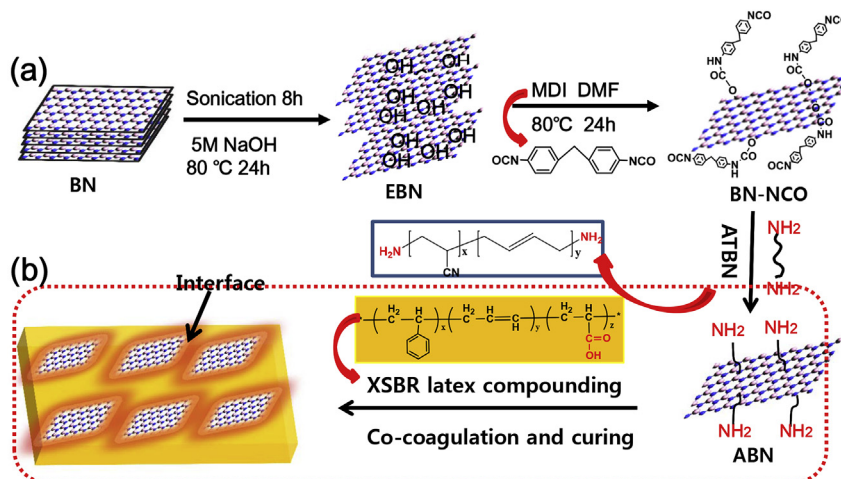


Fig. 1. (A) Schematic for the preparation of ABN and (b) schematic diagram for the preparation of the XSBR/ABN nanocomposites.

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