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Enhanced thermal conductivity of poly(vinylidene fluoride)/boron nitride nanosheet composites at low filler content



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ABSTRACT

Due to the growing needs of thermal management in modern electronics, high thermal conductive polymer composites are increasingly demanded. Boron nitride nanosheet (BNNS) was prepared through molten hydroxide assisted liquid exfoliation of hexagonal boron nitride (h-BN) powder and used as thermally conductive filler. The poly(vinylidene fluoride) (PVDF)/BNNS films were obtained through solution blend and hot pressing. With only 4 wt% BNNS, the in-plane thermal conductivity of PVDF/BNNS composite achieved 4.69 W/mK, with a thermal conductivity enhancement of 2297% compared to neat PVDF. However, the through-plane thermal conductivity of the composites is only 0.23 W/mK, which shows a high thermal conductive anisotropy over 20. The thermal conductive anisotropy and the high in-plane thermal conductivity can be attributed to the formation of thermally conductive network in PVDF matrix. Thus, the BNNS reinforced PVDF films are promising for use as an efficient heat spreader for electronic cooling applications.

1. Introduction

Efficient removing the heat generated by high power device and highly integrated electronic components could greatly improve the work stability and lifetime of the devices [1]. However, current polymeric thermal conductive composites produced by simple blending can hardly get high thermal conductivity with a low filler content. The reasons can be attributed to the difficulty of thermal conduction path formation in polymer matrix at low fillers loading. Two dimensional (2D) nanomaterials, like graphene, boron nitride nanosheet (BNNS), and MXene [2-4] have been widely used as high thermal conductive fillers. Among them, BNNS, also called "white graphene" [5], is one of the best choice due to its high thermal conductivity and dielectric insulation [6,7]. BNNS possess similar structure with graphene, whose calculated thermal conductivity reaches 1700-2000 W/mK [8,9]. Poly (vinylidene fluoride) (PVDF) is currently used in high power electronics due to its high dielectric constant and high melting temperature. However, the low thermal conductivity of PVDF greatly limits its application. In order to obtain higher thermal conductivity, specific fillers were added in PVDF based composites. Xiao et al. [10] demonstrated that the thermal conductivity of BN/PVDF only reached the value of 0.97 W/mK even with 20 wt% BN particles content. The unsatisfactory result could be ascribed to bad distribution of BN in PVDF matrix. Generally, a high loading of fillers is essential for achieving a high thermally conductivity. However, the large amount of fillers commonly results in the deterioration of mechanical property and high cost, which will make the composites difficult to meet the demands of industrial applications [11].

Up to now, few-layered BNNS were mostly prepared via solid-phase mechanical method or liquid-phase exfoliation [12–14]. The solid phase mechanical exfoliation is generally high cost and time consuming. Liquid exfoliation normally exhibits higher efficiency and eco-friendliness [15–17]. The exfoliation process which occurs in a liquid environment, is easier, faster, and safer than solid phase mechanical exfoliation, and does not cause dust pollution. Here, we demonstrate a facile liquid exfoliation method of BN and introduced the obtained BNNS into polymer matrix. Our method of liquid exfoliation enables the production of BNNS with a yield of 19% [18], which is higher than most of the liquid-phase exfoliation methods reported so far. The inplane thermal conductivity of PVDF/BNNS composites is equal to 4.69 W/mK, while the through-plane one is equal to 0.23 W/mK with only 4 wt% BNNS loading. The PVDF/BNNS composites show great

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D. Microstructurar analysis

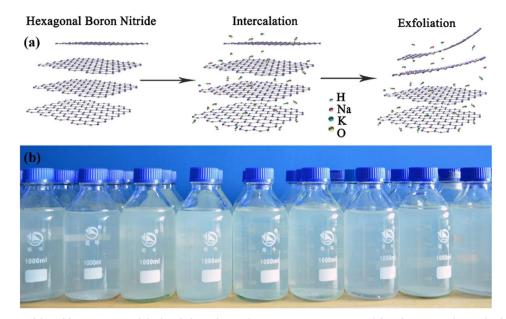


Fig. 1. (a) Schematic diagram of the exfoliation process and (b) digital photo of BNNS dispersions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

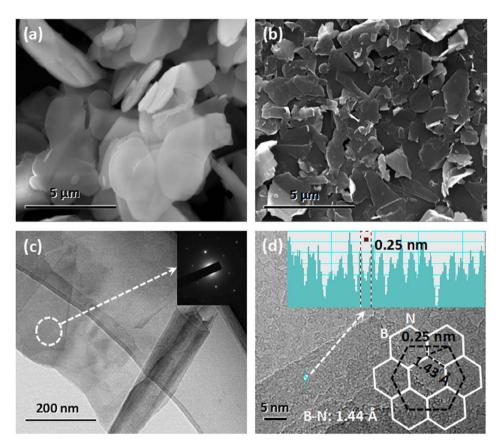


Fig. 2. Scanning electron microscopy (SEM) image of (a) pristine h-BN powder and (b) BNNS exfoliated from h-BN powder. (c) Transmission electron microscopy (TEM) image of BNNS. The inset is corresponding selected area electron diffraction (SAED) pattern. (d) The high-resolution TEM (HRTEM) image of BNNS. There are two insets of d, one shows TEM contrast intensity profile documented along blue line as a mark, the other one below it is the structure model of BNNS. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

potential used as a dielectric insulated heat spreader for electronic packaging application.

2. Experimental

2.1. Materials

Hexagonal boron nitride (h-BN, with lateral size of 7μ m) powder was purchased from ESK Ceramics GmbH & Co (Germany). Sodium

hydroxide (NaOH, \geq 96%) and potassium hydroxide (KOH, \geq 85%) and N,N-dimethylformamide (DMF, \geq 99.5%) were purchased from Sinopharm Chemical Reagent Co. Ltd, Shanghai (China) and used without further purification. Poly(vinylidene fluoride) (PVDF) powder was purchased from 3F Co. Ltd, Shanghai (China).

2.2. Preparation of BNNS

The molten alkali-assisted exfoliation of h-BN was following by two

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