



High thermal conductive polyvinyl alcohol composites with hexagonal boron nitride microplatelets as fillers



Bin-Huan Xie, Xiao Huang*, Guo-Jun Zhang*

State Key Laboratory of High Performance Ceramics and Superfine Microstructures, Shanghai Institute of Ceramics, Shanghai 200050, China

ARTICLE INFO

Article history:

Received 19 March 2013

Received in revised form 14 June 2013

Accepted 17 June 2013

Available online 24 June 2013

Keywords:

A. Polymer–matrix composites (PMCs)

B. Thermal properties

C. Anisotropy

E. Casting

ABSTRACT

Organic polymers are generally thermal insulators. There is a huge demand especially from electronic industries to develop highly thermal conductive polymers. The most common way to increase the thermal conductivity of a polymer is to introduce highly thermal conductive inorganic fillers into the polymer matrix. But up to today, high loading of the fillers is required to achieve high thermal conductivity which sacrifices the lightweight, excellent processability and low cost of polymeric materials. In this work, hexagonal boron nitride (h-BN) microplatelets are introduced as thermal conductive fillers into polyvinyl alcohol (PVA). Due to the unique shape, h-BN microplatelets orient during processing, forming textured structure within the polymer matrix, which helps the formation of thermal conductive pathways. Thus, high thermal conductivities can be achieved at very low filler loadings. The thermal conductivity of the h-BN/PVA composite reaches 1.45 W/m K at only 1 wt% (0.6 vol%) loading, while that number rises up to 3.92 W/m K at 10 wt% (5.9 vol%) loading. Our parallel experiments show that the shape of h-BN microplatelet plays an important role. The surface modification on h-BN fillers improves the compatibility between inorganic fillers and organic matrix, leading to better dispersibility and higher degree of orientation, thus higher thermal conductivity.

© 2013 Elsevier Ltd. All rights reserved.

1. Introduction

The past decades witness the uncannily accuracy of Moore's law that the packaging density of components in an electrical device doubles approximately every two years [1]. Such an increase of the packaging density leads to an unwanted yet unavoidable side effect – the generation of a greater amount of heat in a smaller space. Most electrical components can only function normally at relative low temperatures. Thus, to ensure proper device operation, the unwanted heat must be dissipated [2,3].

Organic polymers have been widely used in modern electronic industries such as printed circuit boards (PCBs), thermal interface materials (TIMs), and device bases/holders [3,4]. But polymers are usually considered as thermal insulators. Most polymers' thermal conductivities are far less than 1 W/m K [5]. This is not a big issue in earlier days when the components packing density was not this high. But today, it becomes one of the bottle-neck problems for the continuous development in electronic industries.

The most effective and promising way to improve polymers' thermal conductivities is by introducing inorganic fillers with high

thermal conductivities into the polymer matrix. A lot of inorganic materials such as metals [6,7], carbon materials [8,9], metal oxides [10,11], metal nitrides [12,13] and clay [14] etc. have all been applied. But polymers with satisfying thermal conductivities are mostly achieved at the filler loading greater than 20 wt% (see Table 1), which sacrifices the lightweight, low cost and excellent processability of the polymeric materials.

Among all kinds of fillers, during the past decade, MWCNTs and SWCNTs have been expected to be capable of improving the thermal conductivities of polymeric materials effectively at relatively low loadings. But up to today, the results are rather frustrating. The poor performances of one-dimensional thermal conductive fillers are due to several possible reasons: the high interfacial thermal resistance between nanotubes and matrix, high tube–tube contact resistance, etc. [9,15,16].

The issues brought by one-dimensional fillers may be avoided by two-dimensional fillers. Due to its extra large in-plane thermal conductivity and inspiration from global research interest, graphene has been received special attention and given a lot of expectations as thermal conductive fillers. Recent works on two-dimensional fillers, including graphene and graphite nanoplatelets, have shown some promising results [17–19]. However, carbon-based fillers due to their electric conductivity are not applicable in applications where both good thermal conductivity and electronic insulation are required [3,4,20].

* Corresponding authors. Tel.: +86 21 52411080, +86 21 52414318; fax: +86 21 52413122.

E-mail addresses: xiaohuang@mail.sic.ac.cn (X. Huang), gjzhang@mail.sic.ac.cn (G.-J. Zhang).

Table 1
Thermal conductivities of polymer composites with different fillers.

Fillers	Polymer matrix	Solids loading	Thermal conductivities (W/m K)	Refs.
Mercapto-functionalized AlN	Epoxy	65 vol%	~6	[44]
Graphene	Epoxy	25 vol% CNTs and 25 vol% GNPs	7.30	[45]
Graphene–multilayer graphene nanocomposite	Thermal grease	2 vol%	14	[17]
Noncovalently functionalized graphene flakes	Epoxy	10 wt%	1.53	[19]
Silica-coated multi-walled carbon nanotubes	Epoxy	1 wt%	0.240	[46]
Py-PGMA–graphene nanosheets	Epoxy	4 phr (parts per hundreds of resin)	1.91	[47]
Graphene	Epoxy	12 wt%	0.73	[48]
Micro-SiC	Epoxy	63 wt%	2.8	[49]
MWCNT		6 wt%	0.8	
MWCNT + micro-SiC		5 wt% MWCNTs + 55 wt% micro-SiC	~7	
Benzenetricarboxylic acid grafted MWCNT	Epoxy	5 vol%	0.96	[50]
BNNTs	PVA	10 wt%	0.54	[26]
Micro and nanosized BN	Polyimide	30 wt%	1.2	[25]
h-BN	Polyimide	30 vol%	~3	[2]
		60 vol%	7	
Graphite nanoplatelet	Epoxy	10 wt%	~2	[18]
AlN + h-BN	Polyimide	49 vol% AlN + 21 vol% h-BN	9.3	[13]
BNNTs	PMMA	10 wt%	0.51	[27]
		24 wt%	3.16	
BNNTs	PVF	10 wt%	0.45	[34]
	PVA	3 wt%	0.3	
Si ₃ N ₄	Polystyrene	40 vol%	3.0	[51]
DWCNT	Epoxy	0.6 vol%	0.252	[15]
BNNSs	Polysiloxane	5 vol%	0.13	[52]
h-BN microplatelets	PVA	0.6 vol% (1 wt%)	1.45 () ^a ; 0.15 (⊥) ^b	This work
		5.9 vol% (10 wt%)	3.92 () ^a ; 0.44 (⊥) ^b	

^a ||: In-plane direction.

^b ⊥: Cross-plane direction.

h-BN is a structural analog and isoelectronic counterpart of graphite. Like graphite, it possesses many exceptional in-plane properties including high thermal conductivity [21,22]. Meanwhile, h-BN is also an excellent electrical insulator and has the highest thermal conductivity among most electric insulators. Thus, h-BN is a very promising filler candidate for preparation of high thermal conductive polymeric materials with excellent electric insulation [23,24]. h-BN particles [2,23,25] and nanotubes (BNNTs) [26,27], have been investigated as thermal conductive fillers. Like other fillers, thermal conductivities greater than 1 W/m K can only be achieved at loadings over 20 wt%.

In this study, we use commercial available h-BN microplatelets as thermal conductive fillers. Due to the platelet-like morphology, the h-BN fillers can be easily oriented during processing, thus leading to anisotropic thermal conductivity of the composites. Because it is very easy to handle in the laboratory, polyvinyl alcohol (PVA) is chosen as the polymer matrix to simplify the research process [26,28,29]. We show that by using two-dimensional fillers and controlled orientation of the fillers, polymer composites with very high thermal conductivities can be prepared at very low filler loadings. The in-plane thermal conductivity of the PVA composite can reach 1.45 W/m K at only 0.6 vol% h-BN loading. And that value can further increase almost to 4 W/m K at 5.9 vol% loading.

2. Experimental

2.1. Materials

h-BN powder (purity >99.5%, ~0.6 μm, with perfect platelet morphology) was purchased from SICCAS, Shanghai, China. Their thicknesses are in the range of 45–116 nm, and the diameter–thickness ratios are in the range of 4.1–13.7. h-BN II (purity >99.5%, ~0.8 μm, with irregular shape) was purchased from Sanxing Co. Ltd., Gongyi, China.

PVA (MW: 1750 ± 50) and 30% hydrogen peroxide (AR grade) were obtained from Sinopharm Chemical Reagent Co. Ltd.,

Shanghai, China, used as received. Deionized water was prepared in-house by Thermo Scientific Easypure II system.

2.2. Surface modification on h-BN

h-BN powder was mixed with 30% hydrogen peroxide at 0.1 g/ml. The mixture was sealed in an autoclave and stirred for 24 h. And then, the mixture was heated up to 100 °C and kept at that temperature for 24 h [30]. After being cooled to room temperature, the mixture was filtered and washed by de-ionized water three times, and then dried at 50 °C under vacuum. In the text, the original h-BN is denoted as p-BN while surface-modified h-BN is denoted as m-BN.

2.3. Preparation of BN/PVA composites

To prepare the composites, weighed amount of p-BN or m-BN was first dispersed in an 8 wt% PVA aqueous solution. The mixture was magnetically stirred at 90 °C for 5 h. Then the BN/PVA composite films were prepared by two different techniques. One is referred to simple casting: the mixture was poured into a Teflon mold and then the mold was placed in an oven at 50 °C for 24 h. After cooling, a composite film with the thickness of about 200 μm was obtained. The other technique is referred to so-called tape-casting: the mixture was poured onto a glass panel and spread to a thin layer using a doctor blade with a tape casting height of 0.4 mm at a speed of 1 cm/s. After the tape casted layer was dried in air at room temperature for 24 h, a flexible film with a thickness of about 15 μm was obtained.

2.4. Instrumentation

SEM images were taken by a Hitachi S-4800 scanning electron microscope. XRD patterns were recorded via a Rigaku X-ray diffractometer (D/Max-2250 V) using Cu Kα radiation. The FT-IR spectra were recorded in a Thermo Nicolet NEXUS spectrometer between 400 and 4000 cm⁻¹. X-ray photoelectron spectroscopy

Download English Version:

<https://daneshyari.com/en/article/7215943>

Download Persian Version:

<https://daneshyari.com/article/7215943>

[Daneshyari.com](https://daneshyari.com)