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Ordered multilayer film of (graphene oxide/polymer and boron nitride/polymer) nanocomposites: An ideal EMI shielding material with excellent electrical insulation and high thermal conductivity

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A R T I C L E I N F O

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ABSTRACT

To simultaneously resolve an undesirable electromagnetic wave and heat emissions that were caused by an electronic device, an electromagnetic interference (EMI) shielding material with excellent electrical insulation and high thermal conductivity was urgently required. However, keeping the EMI shielding material on electrical insulation still remained a challenge due to the superior electrical conductive network in it. We fabricated an ordered multilayer film through casting layer (graphene oxide/poly(-hydroxybenzate-*co*-DOPO-benzenediol dihydrodiphenyl ether terephthalate)) by layer (boron nitride/ (maleated styrene-ethylene/butylene-styrene)), and the special architecture of the film not only simultaneously built the superior electrical conductive path through-plane direction. Moreover, the ordered multilayer film (11 layers) exhibited a good EMI shielding effectiveness (37.92 dB), excellent electrical insulation (breakdown strength was 1.52 MV/m) and high thermal conductivity (12.62 W/mK) in-plane direction, indicating that the ordered multilayer film was a novel promising candidate for an ideal EMI shielding material with excellent electrical insulation and high thermal conductivity in today's electronic devices.

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1. Introduction

To effectively conserve space and conveniently employ electronic devices, the electronic components tended to integrate and be miniaturization. Generally, a high speed processor as the main electronic component would cause undesirable electromagnetic wave and bring about significant heat emissions, which would not only seriously interfere with the function of other nearby electronic components, but also deleteriously shorten the lifetime of the electronic devices. To effectively shield undesirable electromagnetic wave and dissipate heat, the effective electromagnetic interference (EMI) shielding material with excellent electrical insulation and high thermal conductivity (TC) were urgently required [1,2].

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Generally, a high electromagnetic interference shielding effectiveness (EMI SE) for the EMI shielding material required an excellent electrical conductivity (at least 1 S m^{-1}), which can be reached through controlling an electrical filler concentration far above the electrical percolation threshold so as to construct the superior conductive network in the EMI shielding composite [3,4]. As a novel two-dimensional (2D) carbon material, graphene had high electrical conductivity (~600000 S m^{-1}) and remarkable thermal conductivity (~5000 W/mK) [5], and was extremely expected to become a promising candidate for the excellent EMI shielding and high TC materials. For example, the EMI SE of the graphene/polystyrene (PS) composite reached to 29.3 dB when the graphene content was 30 wt% [6]. The EMI SE of the graphene or reduced graphene oxide (rGO) based on epoxy were also investigated in pervious literature [7]. However, to obtain high EMI SE, these composites usually required abundant graphene or reduced graphene oxide (rGO) because it had the homogenous dispersion







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structure. The multi-facet segregated architecture of the EMI shielding composites, in which the rGO selectively located on the surface of the PS particles, was designed, and the EMI SE reached to 45.1 dB with only 3.47 vol% rGO [8]. Nevertheless, only a low thermal conductivity (~2 W/mK) was achieved when the 10 wt% graphene was introduced into the polymer matrix [9,10]. Higher thermal conductivity (~5–10 W/mK) had also been reported through further increasing the graphene content (>20 wt%). [16,17] To obtain high EMI SE of the EMI shielding material with excellent TC, the flexible graphene oxide (GO) films were fabricated through direct evaporation of GO suspension, and the EMI SE and thermal conductivity in-plane reached to 20 dB and 1100 W/mK, respectively.

Although the EMI shielding composites based on graphene or graphene oxide (GO) can effectively shield undesirable electromagnetic wave and dissipate heat, keeping a high EMI SE of the EMI shielding composites on excellent electrical insulation still remained a challenge due to the superior electrical conductive network in the EMI shielding composites [11]. To resolve the dissipating heat of the electronic component, the hexagonal boron nitride (h-BN)/polymer composites were also utilized because the h-BN has high thermal conductivity (~120 W/mK) and excellent electrical insulation [12,13]. Similarly, to enhance the thermal conductivity, the superior thermal conductive network was also needed to be built in the polymer composites [14,15]. However, the thermal conductive mechanism and the electrical conductive mechanism were dramatically different: the electrons, photons and phonons were the hot carriers: meanwhile, the charge carriers were only electrons and holes (such as p-type semiconductor) [16]. Gui et al. reported that the h-BN and graphene hybrid filler had a synergistic effect to enhance thermal conductivity of the polymer composite [17]. Although the synergistic effect of the thermal and electrical conductive network was an effective way to enhance the thermal conductivity of the polymer composite, it was still difficult to make the thermal conductive composite simultaneously have excellent electrical insulation and high EMI SE.

In this article, to make the polymer composite simultaneously have superior EMI SE, excellent electrical insulation and high thermal conductivity, the ordered-layer architecture, which was similar with "Sandwich" structure, was fabricated through casting layer (graphene oxide/polymer layer) by layer (boron nitride/ polymer layer). The graphene oxide/polymer layer was a core layer while the boron nitride/polymer layers were the skin layers. This special architecture not only simultaneously built the superior electrical and thermal conductive networks for the EMI composites in-plane direction, but also effectively blocked the electrical conductive path through-plane direction. Therefore, contradiction between electrical insulation and high EMI SE was expected to be resolved. Moreover, to further enhance EMI SE, the EMI shielding composites were designed as a multilaver structure, and the graphene oxide layer/polymer still remained as the core layers. The final results revealed that the ordered multilayer film of (graphene oxide and boron nitride)/polymer nanocomposites was indeed as a novel promising candidate for an ideal EMI shielding material with excellent electrical insulation and high thermal conductivity in today's electronic devices.

2. Experimental

2.1. Materials

A maleated styrene-ethylene/butylene-styrene (SEBS-g-MAH) was purchased from TSRC Co., Ltd. (China), and the poly (ethyleneco-1-butene) block and polystyrene block ratio was 71/29. A phosphorus-containing liquid crystalline copolyester (PHDDT, Fig. 1) was synthesized from *p*-acetoxy-benzoic acid, terephthalicacid, 2-(6-oxid-6H-dibenz <c, e> <1, 2> oxaphosphorin6-yl) 1, 4-benzenediol, and 4, 4'-diacetoxydiphenyl ether at the overall molar ratio of 10:5:4:1. The detailed synthetic route of PHDDT was described in our previous investigations [18,19]. The graphene oxide (GO) was prepared from expanded graphite through the modified Hummers method. The average particle size and thickness of the GO were 5.7 μ m and 1–2 nm, respectively. Boron nitride (BN) with a particle size of 10-15 µm was purchased from Shandong Pengcheng Special Ceramics Co., Ltd. (China). y-aminopropyltriethoxysilane (KH550) was supplied by Chengdu Xinlong Chemical Co., Ltd. (China).

2.2. Preparation of the functionalized BN and GO particles

100 mL aqueous solution of H_2O_2 (30 wt%, Chengdu kelong Chemical Reagents Co., Ltd) and BN (~5 g) were put into a threenecked flask (250 mL). Next, the mixture was refluxed at 105 °C for 4 h with vigorous stirring [20]. Then KH550 (2 g), 90% ethanol (200 mL) and the BN-OH particles (20 g) were mixed. After agitating at 60 °C for 4 h and washing with distilled water several times, the amino-functionalized BN (f-BN) particles were dried at 70 °C for 24 h. Through the same steps, the amino-functionalized GO (f-GO) particles were also obtained.

2.3. Preparation of the flexible multilayer film

The schematic of the fabrication of the ordered multilayer film was shown in Fig. 2. The SEBS-g-MAH was dissolved in xylene solvent (20 g/L). After that, the f-BN particles (80 g/L) were added into the solution. Meanwhile, the PHDDT was dissolved in phenol/ tetrachloromethane (volume ratio, 1:1) mixed solvent (30 g/L). Then, the f-GO particles (70 g/L) were added into the PHDDT solution. The SEBS-g-MAH/f-BN solution was cast onto a glass plate at room temperature for 24 h, and then it was dried in a vacuum oven for 12 h at 80 °C (*step I*as shown in Fig. 2(c)). The PHDDT/f-GO solution was cast onto the SEBS-g-MAH/f-BN film, and then the glass plate cast was dried in a vacuum oven for 12 h at 80 °C (*step II*as shown in Fig. 2(c)). The PHDDT/f-GI solution was cast onto the SEBS-g-MAH/f-BN film, and then the glass plate cast was dried in a vacuum oven for 12 h at 80 °C (*step II*as shown in Fig. 2(c)). Through repeating the above *step I*and *step II*, the multilayers film can be achieved.

2.4. Characterization

The atomic force microscopy image of the GO was obtained through the tapping mode on a Veeco Digital Instruments Multimode Scanning Probe Microscope with a Nanoscope III Controller. Scanning electron microscopy (SEM) observation was achieved by a Hitachi S-4800 field emission SEM at an accelerating voltage of



Fig. 1. Chemical structure of PHDDT, where X, Y, and 1-X-Y denoted the overall composition in molar ratio.

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