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Controlled direction of electrical and mechanical properties in nickel tethered graphene polyimide nanocomposites using magnetic field





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ABSTRACT

Oriented hybrid nickel tethered graphene polyimide resin nanocomposites with different degrees of orientation were prepared by in-situ magnetic field solvent casting method. Magnetization of the hybrid Ni-graphene polyimide nanocomposites exhibited a maximum in the magnetic field direction and a minimum perpendicular to the magnetic field direction indicating the orientation of the superparamagnetic nickel nanoparticles. In-plane dc electrical conductivity of the 1.3 vol.% Ni-graphene was 2.5 times higher when cast in a high magnetic field compared to films cast without an applied magnetic field. The through-plane dc conductivity of the 1.3 vol.% oriented Ni-graphene polyimide nanocomposites decreased with increasing magnetic field strength and reached insulation (10^{-12} S/cm) for the films cast in high magnetic field. The in-plane tensile modulus of the polyimide exhibited a 35% increase when 0.16 vol.% Ni-graphene was added to the polyimide and cast in a low-strength magnetic field. Further addition of Ni-graphene, up to 1.3 vol.%, to the polyimide resulted in nearly constant tensile moduli. Tensile strength of nickel graphene nanocomposites showed up to 2-fold increase compared to the neat polyimide. Scanning electron microscopy (SEM) revealed that the Ni-graphene nanosheets were oriented in the magnetic field direction.

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1. Introduction

Graphene is a high aspect ratio two-dimensional nanoparticle with extraordinary in-plane properties of high Young modulus (1 TPa), high intrinsic stiffness (130 GPa), theoretical charge carrier mobility, and excellent thermal conductivity (4000 W/ m K) made from hexagonal carbon rings [1–4]. Graphene's micron-sized lateral and monoatomic thin dimensions consist of continuously connected hexagonal carbon ring with π electron cloud and a theoretical surface area of $2600 \text{ m}^2/\text{g}$ [1–4]. Graphene's applications include use in transistors, super capacitors, photonics, and polymer nanocomposites [5–11]. Addition of graphene to polymers results in mechanical property enhancements [10], electrical property improvements [12], increases in thermal conductivity [13], barrier property enhancements [14], and light-induced actuation [15]. The dispersion of graphene in the polymer host matrix is generally isotropic with some alignment tendency of the

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two dimensional nanoparticles. The property enhancement of the graphene polymer nanocomposites is a function of the degree of graphene's interfacial interactions with the polymer, available interface dictated by the dispersion or aggregation, and three-dimensional orientation of the graphene nanoparticles.

Aromatic polyimides are high temperature engineering polymers with a range of important properties including; a wide thermal stability range (-65 to 357 °C), high stiffness and mechanical properties, excellent radiation resistance, and low dielectric constant [16]. The excellent properties of aromatic polyimides make them suitable for advanced technologies such as aerospace components, circuit board printing and packaging, and engine components. Graphene is a two-dimensional nanoparticle with exceptional in-plane properties. Controlling the planar orientation of graphene leads to maximum property enhancements in the targeted direction as indicated by simulations and modeling [17]. Alignment of graphene in polymethylmetacrylate (PMMA) and polystyrene (PS) using multilayer coextrusion resulted in a 118% increase in reinforcement of single layer [18]. This increase is significantly higher than previous reports of randomly oriented graphene which is attributed to the planar orientation of graphene in the polymer layer [18]. Random and oriented graphene oxide polyurethane nanocomposites were prepared and exhibited a 21-fold increase in tensile modulus and excellent improvement in barrier properties [19]. Alignment of graphene in Nafion[®] was achieved by in-situ reduction of graphene oxide with hydrazine in aqueous solution [20]. The oxygen and water barrier properties of aligned graphene oxide polyvinyl alcohol (PVA) exhibited 98% and 68% decreases at 0.72 vol.% graphene content [21]. Orientation of graphene in PVA/graphene (1 wt.%) by uniaxial stretching of oxide films resulted in 42% suppression of swelling, barrier property improvement, increases of modulus and strength, and exceptionally high toughness (260% increase) [22].

Synthesis and self assembly of magnetic nanoparticles and hybrid magnetic nanoparticles have a wide range of applications in aerospace [23,24], recording media [25], nanomedicine [26,27], and hyperthermia [28]. Magnetic nanoparticle properties depend on the nanoparticle chemical composition, shape, aspect ratio, and size [29]. The dipole moment of superparamagnetic nanoparticles is freely moving and will take the direction of an applied magnetic field. Therefore, superparamagnetic nanoparticles can be aligned in a magnetic field [30].

The significant barrier property improvement in graphene TPU nanocomposite has been partly attributed to the tendency of high aspect ratio sheetlike structure of graphene to orient in the film direction [14]. Spherical magnetic nanoparticles were incorporated in the polystyrene matrix and were aligned by moderate magnetic field which resulted in directionally oriented reinforcements [30]. The alignment of the nanoparticles was investigated by small angle X-ray scattering and transmission electron microscopy [30].

Tuning and controlling the direction of mechanical property, thermal and electrical charge transport increase is highly desirable in advanced technological applications. Thermal energy dissipation in portable electronic device and multimedia is only achievable using directionally controlled thermal transport with highly conductive carbon materials. Electrical charge dissipation in airplane components requires directional control of the electrical charge.

We previously reported the synthesis and characterization of hybrid nickel tethered graphene nanoparticles, and their magnetic conductive polyimide nanocomposites [31]. Alignment and property enhancement of graphene in rubbery and lower modulus polymers such as polyurethane, polyvinyl alcohol, and Nafion[®] have been investigated [19-22]. Recently, coextrusion of multilayer polystyrene and PMMA graphene has resulted in alignment and property enhancements [18]. However, these studies did not use an applied magnetic field to induce orientation. To our knowledge, this paper presents the first report of using a magnetic field to align nickel graphene nanoparticles in high-performance polyimide matrix. This study demonstrates that the use of a magnetic field is a simple and easy method by which magnetic nanoparticles tethered to graphene can be manipulated to enhance the properties of highly thermally stable, high modulus, high performance polyimide in a controlled manner.

Here, we report controlling and tuning the bulk electrical and mechanical properties of the nickel tethered graphene polyimide nanocomposites using external magnetic fields. The properties of the magnetic field oriented Ni-graphene polyimide nanocomposites are compared to the Ni-graphene polyimide nanocomposites prepared without magnetic field. The morphology of the Ni-graphene polyimide nanocomposites are examined by scanning electron microscopy and transmission electron microscopy. This work attempts to provide a correlation between the orientation and the examined properties.

2. Experimental

2.1. Materials

Bisphenol A dianhydride (BPADA) and 4,4'-Bis(4-aminophenoxy)biphenyl (BAPP) were purchased from Polyscience Inc. and Chriskev, respectively. Nickel acetyl acetonate, methanol, chloroform, N-methyl-2-pyrrolidone (NMP), and acetone were obtained from Sigma methanol, and J.T. Baker Chemical Co., respectively. Oxygenated graphene was generously donated by Vorbeck Materials Corp.

2.2. Sample preparation

Polyimide was synthesized based on a previously described method by the reaction of bisphenol A dianhydride (BPADA) and 4,4'-Bis(4-aminophenoxy)biphenyl (BAPP) in anhydrous NMP followed by thermal imidization [32,33]. Nickel graphene nanoparticles were prepared based on previously published procedure [31]. Briefly, graphene oxide NMP dispersion (0.0023 g/cc) was heated to 190 °C when a solution of nickel acetyl acetonate (0.0514 g/cc) introduced dropwise in 1 h under nitrogen. The dispersion was centrifuged, washed with acetone and, then dried in vacuum. Nickel tethered graphene polyimide nanocomposite films (1.3 vol.%) were prepared using solvent mixing with dispersion based on the previously reported procedure [31]. The solvent was evaporated while mixing until high-viscosity dispersion was obtained. The sam-

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