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Graphene foam/carbon nanotube/poly(dimethyl siloxane) composites as excellent sound absorber

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ABSTRACT

The sound damping capabilities of graphene foam (GF)/carbon nanotube (CNT)/poly(dimethyl siloxane) (PDMS) composites to low-frequency sound waves are studied. To our best knowledge, this work reports for the first time the development of three dimensional graphene-based composites for sound absorption achieving a commercially viable absorption coefficient higher than 0.3 over a wide frequency range of 100–1000 Hz. Cellular GF is fabricated by template-directed chemical vapor deposition and the composites are prepared by infiltrating solvent-diluted PDMS with and without multi-walled CNTs (MWCNTs) into the porous structure of GF. The GF/PDMS composites with a porosity of 51.5% and a thickness of 1.6 mm are capable of shielding a maximum 70% of the incident sound waves at a low frequency between 100 and 200 Hz. This value is much better than the damping performance of commercial sound absorbing materials with much larger thicknesses. The incorporation of MWCNTs into the PDMS matrix allows the frequency band to be expanded to a wider range from 100 to 1000 Hz while achieving a uniform absorption of more than 30% of the incident sound waves. The outstanding sound damping capabilities of GF/PDMS and GF/CNT/PDMS composites make them excellent candidates for low-frequency noise shielding in many premises.

1. Introduction

Sound damping capability is a desired characteristic of a material because of the serious noise pollution problems everywhere. Porous structures, such as foams, lotus type, meso-perforated and porous fibrous materials, have been demonstrated to be particularly effective for acoustic damping applications [1,2]. Characteristics of pores – porosity, pore morphology, pore size and distribution of pores – influence both the sound absorption coefficient values and the absorption peak positions [3,4]. Open-, semi-open- and close-celled metallic foams have been extensively studied [3,5,6]. The first two types of foams are effective sound attenuators: the sound waves enter into the foam structure through the surface opening, reverberate and eventually dissipate or become absorbed [3,5]. However, the close-celled foams are poor absorbers [3]. Various wood-based absorbers have been developed as alternatives to high-density metallic materials [7–9].

The ability of materials, especially polymers, to damp vibration and absorb sound is a function of their viscoelastic nature where four major mechanisms of sound attenuation can occur [10–12]. They include: (i) scattering by inhomogeneities, (ii) mode conversion at boundaries, (iii) redirection, and (iv) intrinsic absorption by conversion to heat, especially with viscoelastic materials [13]. Judging from these mechanisms, sound absorption by a solid viscoelastic material is inefficient. Inclusions, such as microcells or large voids, may also help attenuate the sound energy by converting the incidental, longitudinal waves to shear waves or by scattering them to other modes of wave propagation. Although most commercial sound-absorbing foams perform well at high frequencies, the attenuation of low-frequency sounds, especially those at below 1000 Hz, has always been a challenging task because the dissipation of materials is inherently weak in this region [14]. Micro-perforated panel (MPP) absorbers have been widely studied due to their ability to absorb low-frequency sounds, but they have a critical disadvantage of narrow sound absorption bandwidths around their respective resonance frequencies. Thus, a wide-band

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sound absorber was designed with holes drilled in multiple sizes instead of a uniform size [15].

Porous polymer composite foams reinforced with nano-carbon materials, such as nanoclay, carbon nanotubes (CNTs) and graphene, are excellent sound dampers because they are light and possess excellent sound absorption properties [2,16]. The exceptionally large specific surface areas of nanoscale carbon fillers contribute to improved friction between the polymer matrix and the fillers, leading to effectively enhanced acoustic dissipation capabilities [17]. Among many different polymers, polyurethane (PU) foams have been extensively studied for sound absorption because of their light weight, easy processing and excellent viscoelasticity. The incorporation of carbon fillers before the foaming process effectively changed the morphology of cellular structure of PU foams and induced interfacial interactions by friction between the fillers and matrix [18,19], resulting in enhanced acoustic absorption properties. However, the absorption capabilities of currently cellular foams at low frequencies are insufficient, unable to fulfill the requirement for many practical applications. Therefore, it is necessary to develop novel materials for the absorption of low-frequency acoustic waves.

In this paper, we report the highly porous, cellular graphene foam (GF)/poly(dimethyl siloxane) (PDMS) composites of 1.6 mm in thickness for the absorption of low-frequency sound waves. A maximum 70% absorption of the incident sound waves at frequencies ranging from 100 to 200 Hz is achieved by the thin composites, which is much better than commonly used carbon foams and MPPs. The incorporation of multi-walled carbon nanotubes (MWCNTs) in the PDMS matrix gives rise to a uniform absorption of more than 30% of the sound waves in an expanded frequency band up to 1000 Hz. The effect of porosity on sound absorption effectiveness and the mechanisms behind the excellent sound absorption capabilities are discussed.

2. Experimental

2.1. Fabrication of GF/PDMS and GF/CNT/PDMS composites

GFs were produced based on chemical vapor deposition (CVD) of multilayer graphene using CH_4 carbon precursor on Ni foam templates [20–22]. The Ni foams with a thickness of 1.6 mm and an area density of 320 g/cm^2 (supplied by Heze Tianyu Technology Development) were cleaned by sonication in acetone for 1 h and rinsed using deionized water for several times followed by drying in an oven. They were cut into rectangular pieces before placing in the quartz tube of CVD chamber. The quartz tube was initially evacuated to a base pressure of 5 mTorr and 500 standard cubic centimeter per minute (sccm) Ar and 200 sccm H_2 were introduced to purge the tube. The temperature was raised to 1000 °C at a ramp rate of 20 °C/min and kept for 10 min to anneal the Ni template. A CH_4 concentration of 1.4 vol% (or 15 sccm) in the mixture of 500 sccm Ar and 200 sccm H_2 was used to grow uniform graphene onto the Ni template. The above growth condition was established after a series of trial and error experiments.

PDMS (Sylgard184, Dow Corning, base agent/curing agent = 10/1 by weight) solutions were dip-coated onto the graphene/Ni foam after dilution with ethyl acetate (Anaqua) solvent. PDMS solutions with different PDMS content were prepared by varying the solvent to base agent weight ratio from 0 to 10, so that GF/PDMS composites with different graphene contents and porosities were produced after evaporating the solvent. After the composites were cured at 80 °C for 2 h, the Ni foam template was etched using 3 M HCl at 60 °C for 12 h to obtain porous GF/PDMS composites.

MWCNTs (supplied by NanoKarbon Co. with an outer diameter of 60–80 nm and a length of ~20 μm) were treated by UV/O_3 on a UV/ozone cleaning system (Jelight 144AX-220) for 2 h to create oxygenated functional groups on the surface of CNTs [23]. 2 wt% treated CNTs were dispersed in ethyl acetate by sonication for 1 h before adding into the PDMS solution. The mixture was magnetically stirred and sonicated for 1 h for uniform dispersion of CNTs, which was then dip-coated onto the graphene/Ni foam followed by the same procedure as above to fabricate the GF/PDMS composites. The fabrication process is schematically shown in Fig. 1a. Neat PDMS foams were also prepared as a control group by dip-coating the pure PDMS solution directly onto the bare Ni foam followed by thermal curing and Ni etching.

2.2. Characterization of materials

The morphologies of freestanding GFs, GF/PDMS and GF/CNT/PDMS composites were characterized on a scanning electron microscope (SEM, JEOL JSM-6700F and 7100F) with a 15 kV accelerating voltage and a field emission transmission electron microscope (FETEM, JEOL 2010) at an acceleration voltage of 200 kV.

The porosities, ε, of the composites were determined by [24]:

$$\varepsilon = \left(1 - \frac{\rho_a}{\rho_t}\right) \times 100\% \quad (1)$$

where ρ_a is the apparent density calculated by the ratio of total mass to total volume of composites; and ρ_t is the true density of composites determined from the densities of graphene (2.2 g/cm^3) [25] and PDMS (0.965 g/cm^3) according to the following equation:

$$\rho_t = \frac{m_C}{m_G \rho_G^{-1} + m_{PDMS} \rho_{PDMS}^{-1}} \quad (2)$$

where m and ρ are the mass and density, respectively, and the subscripts C and G represent the composite and graphene, respectively. Because MWCNTs constituted less than 1 wt% of the total volume of the composites, an identical porosity was assumed for the composites with and without CNTs. The dynamic mechanical properties of rectangular composites of 15 mm long × 6.5 mm wide × 1.6 mm thick were evaluated on a dynamic mechanical analyzer (DMA, TA Instrument DMA Q800) at temperatures ranging from 0 to 100 °C at a heating rate of 3 °C/min. The measurement was performed at a vibration frequency of 1 Hz and an amplitude of 50 μm under a tensile mode.

2.3. Measurements of sound absorption coefficients

The sound absorption coefficients, $\boldsymbol{\alpha}$, were measured on a modified impedance tube apparatus comprising two Brüel & Kjær Type-4206 impedance tubes with the sample sandwiched in between, as shown in Fig. 1b. The front tube had a loud speaker at one end to generate a plane wave and two sensors to sense the incident and reflected waves, thereby obtaining both the reflection amplitude and phase. The third sensor in the back tube which was terminated with an anechoic sponge sensed the transmitted wave to obtain the transmission amplitude and phase. The absorption coefficient, $\boldsymbol{\alpha}$, is given by: [26]

$$\boldsymbol{\alpha} = 1 - |r|^2 + |t|^2 \quad (3)$$

where r and t are the measured reflection and transmission coefficients, respectively. The detailed derivations of r, t and $\boldsymbol{\alpha}$ can be found in the Supporting Information.

3. Results and discussion

3.1. Morphologies of freestanding GF, GF/PDMS and GF/CNT/PDMS composites

The cellular GF produced with CH_4 concentration of 1.4 vol% contained few-layer graphene, as shown in Fig. 2, consistent with