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Sound absorption of electrospun polyvinylidene fluoride/graphene membranes

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ABSTRACT

In this study, a novel sound-absorbing material was developed using electrospun piezoelectric polyvinylidene fluoride (PVDF) membranes. The effects of graphene (Gp) and electrospinning on the crystal structure and piezoelectric properties of PVDF/Gp nanofibrous membranes were examined. The results showed that electrospinning effectively induced the beta phase and increased the piezoelectricity. Adding graphene further improved the piezoelectric properties through interfacial polarization. Electrospun nanofibrous membranes exhibited an increase in surface area, and, consequently, their contact with sound waves was increased, which enhanced the sound-energy absorption in the middle-frequency region through the friction and vibration of the internal nanofibers. The piezoelectric electrospun PVDF samples were crucial for converting sound energy into electric potential and absorbing sound waves in the low-frequency region, and the sound absorption performance of the electrospun PVDF/Gp membranes with the highest piezoelectricity was shifted further to a lower frequency region. Thus, the electrospun PVDF/acoustic nonwoven presented herein is potentially a practical and efficient sound absorber because of its favorable absorption performance, particularly in low-frequency regions.

1. Introduction

Polyvinylidene fluoride (PVDF)-based piezoelectric polymers are of interest for applications in tactile sensor arrays, strain gauges, and audio transducers. PVDF has been widely investigated because its conformable properties facilitate its application to complex structural surfaces. PVDF polymers are light, strong, and conformable, and they can be cut into various shapes and sizes. Furthermore, the polymeric nature and chemical compatibility of PVDF allow precise tailoring of the material by using other nanomaterial species. PVDF exhibits five crystalline phases featuring these three conformations [1]: (1) the all-trans (TTTT) planar zigzag conformation in the case of the β phase, (2) TGTG′ for the α and δ phases, and (3) T3GT3G′ for the γ and ε phases. Theα phase is nonpolar and is the phase most commonly found in commercial PVDF films, whereas the β phase is polar, and in this phase, dipole moments are oriented in the same direction; thus, the β phase determines the piezoelectric properties of PVDF polymers [2]. The β phase can be obtained through various posttreatments, such as mechanical stretching and high-electric-field poling [3], and two recent additions aimed at obtaining the β phase are the use of electrospinning (ES) and the blending of PVDF with organic or inorganic additives, nanoparticles, or carbon materials [4–7].

Graphene (Gp) has attracted considerable attention because it exhibits unique characteristics such as highly favorable mechanical properties, electrical conductivity, mobility of charge carriers, specific surface area, and transport phenomena

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[8]. Graphene is not only optimal as a nanofiller for improving the mechanical, electrical, and thermal properties of polymers [9–11]; it can also serve as the nucleation agent for generating the polar phase in PVDF [12]; this is because the large π bond in graphene specifically interacts with the highly electron-dense fluorine atoms in PVDF, which results in β-phase formation in PVDF/Gp composites. Furthermore, because graphene is conductive, the dielectric constant of PVDF/Gp composites is higher than that of neat PVDF [13]. A drawback here is that dispersing graphene sheets uniformly in a polymer matrix is challenging because the sheets can readily reagglomerate or restack into graphite because of the strong van der Waals interactions between them. However, promoting only the dispersion of graphene in a polymer matrix can strengthen graphene–polymer interactions, and then facilitate the formation of the β phase in PVDF and improve the properties of PVDF/Gp. Therefore, various methods (e.g., modification of graphene and addition of dispersing agents into composites) have been employed for enhancing graphene dispersion in a PVDF matrix [14,15].

ES is a manufacturing technique that can be used to extract continuous nanofibers presenting several favorable characteristics such as a low diameter, high specific surface area, flexibility in surface functionality, and high porosity [16–18]. Thus, such nanofibers can be used as filter materials, biomedical elements, tissue scaffolds, biosensors, photoelectric components, and reinforced composite materials [19–21]. Andrew electrospun PVDF nanofibers and determined that the β-phase fraction increased with the working voltage and viscosity [22]. The electrospinning procedure, which induces a confinement of the polymer in a fiber of small diameter, further represents a favoring element for β-phase formation. Such a role may magnify by the presence of nanofiller such as graphene, carbon nanotube and clay, because in this case the polymer is subject to the spinning process which forces the chains to elongate in a fiber of small diameter, at the same time having to overcome the resistance to flow produced by the nanofiller [12,23]. Nanofiller play a double role in influencing the polymer crystallization process. It works as a nucleating agent, and also it is a physical obstacle to the motion of polymer chains. Kalinová demonstrated that electrospun nanofibers are porous fibrous materials that are highly efficient sound absorbers [24]. Electrospun nanofibers possess high surface area, porosity, and flexibility [25], which increase their sound-absorbing rate to beyond those of conventional materials such as nonwoven and acoustic foams [26]. Moreover, the frequency of the nanofibers decreases with increasing areal density of the nanofibrous membranes [27], which enhances the high-frequency absorption. However, to improve low- and middle-frequency absorption, other mechanisms for sound absorption are required.

When a piezoelectric material such as PVDF is subjected to stress or force, it generates an electrical potential or voltage proportional to the magnitude of the force; this property makes the material optimal for converting mechanical energy into electric potential. For example, a piezoelectric audio transducer can convert sound pressure into electric voltage and voltage into sound pressure. Thus, piezoelectric materials can be used to reduce vibrations and noise, and the materials can be employed in energy acquisition and novel sound-absorbing applications.

In the present study, piezoelectric PVDF nanofibrous membranes were prepared through ES. Graphene were added and coelectrospun with PVDF to improve the piezoelectric properties of the membranes. The polymorphism, piezoelectricity, and sound absorption of the PVDF/Gp nanofibrous membranes were investigated.

2. Experimental

2.1. Materials

PVDF films (Solef® 101 0, Solvay Co., USA), PVDF pellets (MW = 263,000; Kynar®720, Arkema Group, France), and few-layer graphene (GRE-PF00001, Allightec Group, Taiwan) [28] were used in this study. Poly(diallyldimethylammonium chloride) solution (PDDA; Echo Chemical Co, Taiwan) was used as a dispersing agent. Indium tin oxide-coated polyethylene terephthalate (ITO/PET; Sigma-Aldrich, Taiwan) featuring a sheet resistivity of 200 Ω/□ and thickness of 0.13 mm was used as the substrate for ES. Automotive interior used acoustic nonwoven (Feyrer Co., Taiwan), composed of polypropylene (PP) and polyethylene terephthalate (PET) and featuring a thickness of 20 mm and areal density of 2586 g/m^2, was used as the acoustic substrate.

2.2. Preparing electrospun pvdf/gp nanofibrous membranes

PVDF solution was prepared by adding 2.2 g of PVDF pellets to 6 mL of DMF solution and stirring gently at approximately 100 °C for 2 h. The Gp/DMF solution (30,000 ppm) was sonicated for 30 min by using ultrasonic cleaner, following which 1 mL of PDDA solution was added to 9 mL of the Gp/DMF solution; the suspended Gp/DMF solution was then crushed for 20 min at 40 kHz by using an ultrasonic cell disruptor. Subsequently, 0.75 mL of the suspended Gp/DMF solution was added to the as-prepared PVDF solution at 100 °C with stirring, and then the solution was cooled to room temperature. The PVDF/Gp solution (18 wt%) used in ES was prepared by adding 4 mL of acetone to the as-prepared PVDF/Gp solution in order to enhance evaporation.

An in-house-designed ES device consisting of an injection spinneret was powered by a syringe pump (KDS 101 Series, KD Scientific, USA), which was connected to a Teflon tube attached to a stainless steel needle (diameter, 0.23 mm). An electrostatic controller (SM 4030-24NIR, You-Shang Technical Corp., Taiwan), connected to the spinneret and collector, was