



Mechanical properties and simulation of nanographene/polyvinylidene fluoride composite films

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ABSTRACT

Nanographene possesses unique mechanical and electrical properties. The mechanical properties of nanographene/polyvinylidene fluoride (PVDF) composite films are studied by experiment and simulation analysis in this paper. Different amounts of nanographene/PVDF samples were prepared to obtain general field emission scanning electron microscopy overviews and perform tensile experiments for obtaining the equivalent elastic moduli. Materials Studio was used to establish nanographene microscopic models to obtain linear elasticity data. The digital simulation models were established using ANSYS to conduct the finite element simulation for the equivalent elastic modulus of the composites in different graphene contents and assumptions.

Results of the experiments and simulations showed that the equivalent elastic modulus of the composites increases first with increasing graphene content and then decreases after the mass fraction of graphene reaches 10%. The change in the mechanical properties of nanographene/PVDF composites is due to graphene agglomeration. The agglomeration phenomenon increases with the increase in graphene.

1. Introduction

Graphene is a type of two-dimensional planar honeycomb-lattice-structured carbon material with excellent physical properties, such as superior thermal conductivity and light transmission [1], good conductivity (up to 6000 s cm^{-1}), and high Young's modulus (1100 GPa) [2]. Several kinds of experimental methods, such as micromechanical stripping, chemical vapor deposition, redox, and epitaxial growth [3–6], have been employed to prepare graphene. Graphene exhibits unique, interesting physical and chemical properties. Its derivatives include graphene oxide (GO) derivatives [7], graphene obtained by hydrogenation, and graphene derivatives with magnetic properties. Graphene capsule is also a widely used graphene derivative which can be mixed with metal to prepare composite material [8,9].

In recent years, considerable work has been conducted in the research and practical exploration of graphene. Graphene offers extensive application prospect in high-performance energy storage materials, computer chips, sensors, transparent electrodes, and liquid crystal display materials. As a new functional material in the high-tech field, graphene will significantly influence future technological revolution [10–14]. Moreover, graphene-related composite materials constitute a

popular research topic. The strong force between carbon atoms in the structure confers ultrahigh strength to graphene, which is currently known to exhibit the highest physical strength. As an additive, graphene is widely used in the preparation of new high-strength composite materials with increasing performance advantages in defense, industry, and other fields.

Graphene can be combined with a variety of other materials, including metals, metal compounds, and polymers. Among numerous existing polymers, polyvinylidene fluoride (PVDF) has been widely used because of its multiple advantageous properties. PVDF is a semi-crystalline polymer with good mechanical properties [15]. In the past decades, the polymer has been widely used as a functional or structural material due to its easy processability, excellent chemical resistance, thermal stability, and good mechanical properties [16–21].

Considerable exploratory research has been conducted on the preparation of graphene/PVDF composites. Dao-an Zha et al. [22] formed a hybrid graphene/PVDF gel by applying the diffusion of a nonsolvent (methanol or water) into the graphene/PVDF suspension in *N,N*-dimethylformamide (DMF). This drying method effectively prevents pore shrinkage because of the capillary force during the drying of gels, yielding porous materials with a high specific surface area and surface

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roughness. Md Ataur Rahman et al. [23] used a simple solution of casting GO/PVDF to prepare nanocomposite films and then produced PVDF/reduced GO films by in situ thermal reduction of GO/PVDF films.

The use of graphene/PVDF-related composites has been extensively explored. Z. Y. Jiang et al. [24] synthesized GO/PVDF nanocomposites and investigated their structural, ferroelectric, and pyroelectric properties. The group conducted a dielectric spectrum analysis and P–E loop tests, which indicated that the nanocomposites exhibit enhanced ferroelectric and pyroelectric properties relative to those of PVDF samples. The isothermal crystallization kinetics of GO/PVDF nanocomposites quantitatively determined using differential scanning calorimetry demonstrated that GOs facilitate the crystallization of PVDF. Jinhong Yu et al. [25] developed a new thermally conductive composite by combining PVDF with hybrid functionalized graphene sheets (GSs) and nanodiamond filler. The PVDF composite showed different thermal conductivities at different proportions of the hybrid filler and exhibited good thermal conductivity, thermal stability, and electrical conductivity. Numerous experiments were conducted to obtain the best composition of composite material based on various performance indicators. Jin Woo Jang et al. [26] manufactured PVDF-based nanocomposite films with different graphene contents of 0.1–10.0 wt% by ultrasonicated solution mixing and melt compression. The thermal stability of the nanocomposite films under oxygen gas atmosphere noticeably improved, and the electrical volume resistivity of nanocomposite films substantially decreased. The research showed that the mechanical storage modulus is markedly enhanced with increasing graphene content in the nanocomposite films. Varrla Eswaraiah et al. [27] prepared novel foam composites comprising functionalized graphene (f-G) and PVDF and investigated the electrical conductivity and electromagnetic interference shielding efficiency of the composites with different mass fractions of f-G.

Several researchers explored the mechanical properties of graphene/PVDF composites. Jinhong Yu et al. [28] prepared PVDF with GSs, which are novel fillers, through a solution method. The team studied the structure–property relationships of PVDF/GS nanocomposites and found that the nanocomposites exhibited significant increases in dynamic mechanical properties and thermal stability compared with neat PVDF. Chaozhi Yu et al. [29] adopted hot-pressing treatment to convert regular reduced GO (RGO) from chemical exfoliation to high-quality graphene (HQG), and the HQG/PVDF composite films were prepared by spin coating. Their experiment showed that the storage modulus of the HQG/PVDF composite was almost two and eight times higher than those of RGO/PVDF composite and pure PVDF, respectively. Rama K. Layek et al. [30] prepared poly(methyl methacrylate)-functionalized graphene/PVDF nanocomposites and observed significantly increased thermal stability of nanocomposites and large T_g increase (20 °C). Storage modulus showed an increase of 124%, stress at break 157%, and Young's modulus 321% for 5% MG. However, their research was mainly experimental and lacked simulation. The experimental parameters were limited and contingent, and the conclusion was not universal. The abovementioned scientific research explored the preparation, application, and the related properties of nanographene/PVDF composites. The physical and mechanical properties of the composites were not studied from an engineering application perspective.

Nanographene/PVDF composite films were studied in this paper. The different amounts of nanographene/PVDF composite films were prepared and then pressed into regular samples. The general field emission scanning electron microscopy (FESEM) overviews of the composites with different graphene mass fractions were observed to obtain microscopic information. Tensile test was conducted to evaluate the equivalent elastic modulus and tensile strength limit of the sample. Moreover, the microscopic models of nanographene were established using Materials Studio to obtain linear elasticity data. Digital simulation models were established using ANSYS platform to obtain the different graphene contents and the equivalent elastic modulus under

Table 1
Amount of graphene and PVDF.

Mass fraction of nanographene	Graphene (g)	PVDF (g)	Quantity (g)
0%	0	0.300	4
5%	0.015	0.285	4
10%	0.030	0.270	4
15%	0.045	0.255	4
20%	0.060	0.240	4

different assumptions combined with the material properties provided by the microscopic model.

2. Material and experiment

2.1. Preparation of composite films

The composites were prepared by mixing graphene powder with the polymer PVDF by using the blending and hot-molding procedure. The selected solvent in the experiment was DMF, which is an excellent solvent for dissolving PVDF [31]. The sources of reagents used in this experiment are as follows. PVDF was obtained from Beihang University Chemical Laboratory (China), and Graphene powder was supplied by Suzhou Hengqiu Technology CO.LTD (China). DMF was acquired from Beijing Chemical Reagents Company (China).

The composites were divided into five groups according to the amount of nanographene. The percentages of nanographene in the overall mass were 0%, 5%, 10%, 15%, and 20%. The amounts of nanographene and PVDF used in the experiment are shown in Table 1.

First, PVDF was added to DMF. After the PVDF was completely dissolved in DMF, the graphene powder was added to the mixed solution. The solution was stirred for more than 10 min and subjected to ultrasound to maximize the degree of dispersion. After being dried at a temperature of 100 °C for 120 min and cooled to room temperature, the solution was evaporated to film-state samples. Finally, the dried mixture was collapsed and compressed into wafers for 10 min at 220 °C under 5 MPa and cooled to room temperature under the same pressure.

2.2. Microstructure of composite films

Grain morphology and size were observed using FESEM on a JSM-6700F microscope. The general FESEM overviews of nanographene/PVDF composites with different graphene mass fractions are shown in Fig. 1a–d.

The FESEM images of the composite in Fig. 1 show that graphene aggregates into clusters. In the composite material with a graphene content of 5%, the graphene retained a good separation state. With the increase in graphene content, the aggregation became increasingly evident. In composites with a graphene mass fraction of 20%, graphene aggregated and presented minimal separation. The graphene agglomeration phenomenon seriously influences the mechanical properties of the composites.

2.3. Tensile experiments of composite films

Shimadzu AGS-X universal testing machine (Japan) was used in the mechanical tensile test. This test machine allows the maximum tensile force of 1000 N, a force sensor accuracy of 0.0001 N, and displacement sensor accuracy of 0.0001 mm. The appearance of the test machine and the composite film is shown in Fig. 2.

When the object was extended within the small deformation range, the length of the elongated object and the force of the relationship are expressed as follows [32]:

$$\Delta L = \frac{FL}{EA}, \quad (1)$$

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