

Multi-walled carbon nanotube/polyimide composite film fabricated through electrophoretic deposition

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ABSTRACT

Multi-walled carbon nanotube (MWCNT)/polyimide composite films were fabricated through electrophoretic deposition (EPD) of MWCNT-polyamic acid colloidal suspension which was derived from carboxylated-MWCNTs and poly(pyromellitic dianhydride-co-4,4'-oxydianiline) (PMDA-ODA). Under electric field, both negatively charged MWCNTs and PMDA-ODA colloid particles migrate onto a positively charged anode simultaneously, and are converted to a coherent MWCNT/polyimide composite film in the ensuing imidization reaction. Uniform dispersion of MWCNTs in the composite film was observed using transmission electron microscopy. The thickness of the prepared composite film can be tuned by varying processing conditions such as deposition time and anode conductivity. The electrical conductivity of the composite film increased with increasing the concentration of MWCNTs in EPD suspension. The mechanical reinforcement of polyimide using MWCNTs was evaluated by tensile testing and nanoindentation testing.

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1. Introduction

The high electrical conductivity and excellent mechanical property of carbon nanotubes (CNTs) have prompted the development of electrically conductive CNT/polymer composites which could be used as conductive structure materials, electromagnetic interference shielding materials and static dissipating materials [1–3]. The electrical conductivity of CNT/polymer composites depends strongly on the dispersion of CNT in polymer matrix and, as a result, strategy to enhance CNT-polymer connectivity and CNT dispersion in polymer matrix is of great technological importance in the development of CNT/polymer composites [4–14]. So far, several techniques have been developed to incorporate CNTs into polymer matrix. The frequently used method is to disperse CNTs into polymer matrix through direct melt blending or shear-intensive mechanical stirring [5–12]. Conductive CNT/epoxy composites have been prepared through mixing CNTs into epoxy resin using a high speed mixer [5,6], and conductive CNTs/poly(ethylene terephthalate) has been obtained via twin-screw extrusion (blending) at 260 °C [7]. Even though the direct-mixing approach is convenient for industrial production, the choice of suitable polymer

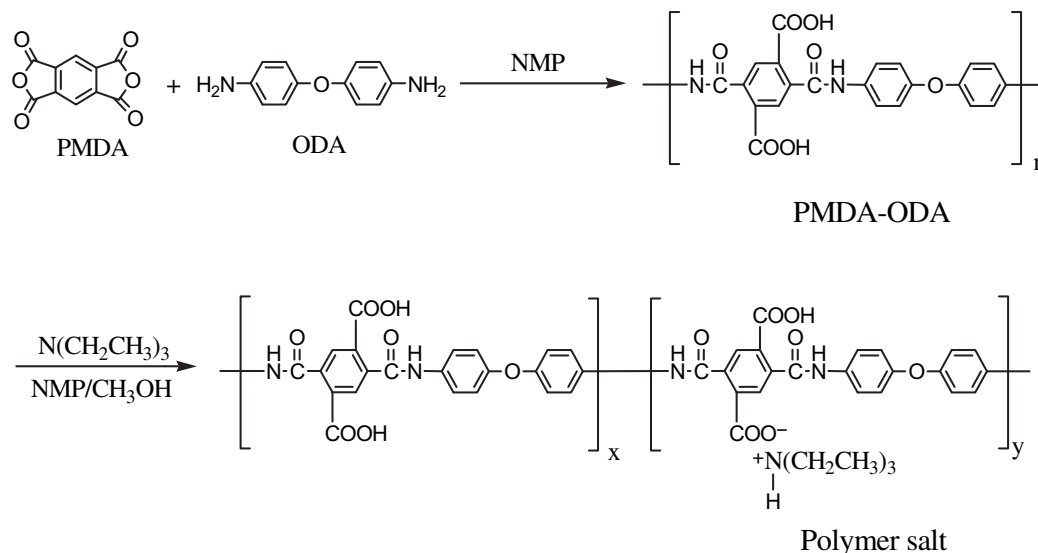
matrix is limited to thermoplastics and liquid polymer materials, and the dispersion of CNTs in the polymer matrix is not always satisfactory. An alternate method is the so-called solution-mixing method, in which an organic solvent is employed to dissolve polymer and disperse CNTs [13,14]. The problem with this method is that CNTs tend to agglomerate during solvent evaporation and the subsequent curing process, leading to non-uniform CNT dispersion in the polymer matrix. Moreover, solution-mixing always involves toxic and inflammable solvents.

Recently, a latex-based process has been developed to prepare electrically conductive CNT/polymer composite with relatively low percolation threshold [15–17]. A stable colloidal suspension is prepared by mixing polymer latex and CNT suspension directly. When water is removed, the colloidal particles assume a close-packed configuration. This leads to the formation of a CNT network that occupies the interstitial space among colloidal particles. Such CNT network is locked in the coherent polymer matrix formed through the coalescence of colloidal particles in the subsequent curing process at high temperature. The percolation threshold of thus prepared conductive composite is exceptionally low because the free volume available for CNTs to form a conductive network is significantly reduced due to the excluded volume created by the colloidal particles [15]. In contrast to the solution-mixing method, this latex-based process avoids using toxic and inflammable solvents. But, water removal through freeze-drying or evaporation is a time consuming process and renders this process unsuitable for mass production.

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Scheme 1. Syntheses of PMDA-ODA and its ionization.

Although the methods mentioned above are generally satisfactory for preparation of respective CNT/polymer composite systems, they have limitation in preparing CNT/polymer composite film with controlled thickness for mass production. Therefore, it is desirable to develop a convenient solution to address these problems. Electrophoretic deposition (EPD) is a convenient and widely used process to deposit electrically charged colloidal particles onto conductive substrate to produce films [18–20]. It is well known for short processing time, simple apparatus and suitability for mass production. Moreover, EPD can be applied to produce film not only on flat surface but also on variously-shaped surfaces with controlled thickness. Recently, EPD was employed to prepare CNT thin film that shows good electron field emission stability under both continuous and pulsed operations [21,22]. In this study, EPD was employed to fabricate multi-walled carbon nanotube (MWCNT)/polyimide composite film from MWCNT-polyamic acid colloidal suspension. Both the conductivity and thickness of the composite film can be controlled easily by fine-tuning the processing condition. Transmission electron microscopy (TEM) study demonstrated that the dispersion of MWCNT in MWCNT/polyimide composite film is uniform. The mechanical reinforcement of polyimide film using MWCNTs was evaluated through tensile testing as well as nanoindentation testing. Unlike conventional latex-based method, solvent removal is not required in the EPD process and the lace solvent remained in the deposited film will be evaporated off in the followed imidization, which is suitable for mass production.

2. Experimental section

2.1. Materials

MWCNTs were purchased from Cheap Tubes, Inc. and used as received. Pyromellitic dianhydride (PMDA) and 4,4'-Oxydianiline (ODA) were purchased from Alfa Aesar and ChrisKev, respectively, and they were purified through sublimation before use. *N*-methylpyrrolidone (NMP) and methanol were purchased from TEDIA and used as received.

2.2. Preparation of MWCNT suspension

First, MWCNTs were treated using nitric acid. As a typical example, 600 mg of pristine MWCNTs was reacted with 240 ml of

nitric acid (2.6 M) under stirring for 1 h using mechanical stirrer at a speed of 400 rpm, followed by ultrasonication for 2 h using water bath ultrasonicator. The mixture was then vacuum-filtered and washed with distilled water to neutral pH. The acid-treated MWCNTs (MWCNTs-COOH) were dried through vacuum drying at 45 °C for 48 h. Then, 4.2 mg of dried acid-treated MWCNTs was suspended in 5.0 g of NMP through overnight stirring. Finally, after adding 15.0 g of methanol, the MWCNT suspension was prepared by ultrasonication the above mixture for 5 min using high power probe ultrasonicator (Sonics VCX 750 Ultrasonic Processor). MWCNT suspensions of different concentrations have been prepared according to the same procedure.

2.3. Preparation of PMDA-ODA colloidal suspension

In a 250 ml three-neck round-bottom-flask, 10.9 g of PMDA (0.05 mol) was added into 140 ml of NMP that dissolved 10.0 g (0.05 mol) of ODA. After stirring for 24 h at room temperature, poly (pyromellitic dianhydride-co-4,4'-oxydianiline) (PMDA-ODA) solution in NMP with concentration of 12.65 wt.% was produced. The synthesis of PMDA-ODA is depicted in Scheme 1.

10 g of PMDA-ODA solution prepared above was diluted with 6.5 ml of NMP, and the carboxylic acid group in PMDA-ODA was ionized by adding 169 μL of triethylamine with ionization degree of 20 mol%. Then, a negatively charged PMDA-ODA colloidal suspension was prepared by dropping 58.8 ml of methanol into the ionized PMDA-ODA solution under stirring. The weight ratio of methanol to NMP is 3:1 and the final content of PMDA-ODA is 2 wt.%. The effective diameter and the average zeta potential of the prepared PMDA-ODA colloidal particles are around 600 nm and -20.5 mV, respectively, as measured by Zetaplus Particle Sizing equipment.

2.4. Fabrication of MWCNT/polyimide composite film and neat polyimide film

The PMDA-ODA colloidal suspension prepared above was mixed with equal volume of MWCNT suspension under water bath ultrasonication for 20 min. The obtained MWCNT-filled PMDA-ODA colloidal suspension was then transferred to a beaker and kept at 25 °C by putting the beaker into a water bath. A chromium (Cr)-coated silica wafer and a workpiece (Cr-coated

silica wafer or copper plate) were used as a cathode and an anode, respectively, and immersed into the colloidal suspension. The cathode and anode were separated by a spacer with length of 1.5 cm. Then, the electrophoretic deposition (EPD) was carried out under a constant electric current generated by LAMBDA InvenSys Genesys™, a direct current (DC) supplier. Different composite films were fabricated by varying the deposition time and MWCNT concentration in the colloidal suspension. The deposited film was then washed with methanol and imidized in an air oven with the following heating steps: 100 °C for 30 min, 200 °C for 30 min, and 280 °C for 60 min.

To fabricate a neat polyimide thin film as a control, the PMDA-ODA colloidal suspension prepared above was diluted to 1 wt.% by mixing with equal volume of methanol/NMP mixed solvent with weight ratio of 3:1 in a beaker. Then, EPD and imidization were carried out under the same processing condition as that for fabrication of MWCNT/polyimide composite thin film.

2.5. Mechanical property testing

The tensile strength measurements were carried out using an Instron 5543 microforce tester with deformation rate of 40 mm min⁻¹ and free length of 10 mm. The composite films were cut to rectangular shape with dimensions of 20 mm × 6 mm × ~40 μm. The average of five measured values was taken for each sample, and the measurements were reported as mean ± standard deviation.

Nanoindentation tests were performed by an MTS Nano Indenter XP (MTS Cooperation, Nano Instruments Innovation Center, TN) using a continuous stiffness measurement (CSM) technique. A three-sided pyramid (Berkovich) diamond indenter was employed for the indentation experiments. MWCNT/polyimide composite film with thickness of approximately 40 μm was mounted on a flat aluminum stub using superglue. In the indentation test, the indenter was pressed into the composite film with a constant strain rate, i.e., 0.05 s⁻¹, from the sample surface to 600 nm deep into the sample. Here, a constant strain rate was chosen to load on the samples in order to avoid a strain-hardening effect on the measurements. At least 10 indentations were performed on each sample, and the distance between each two indentations was 100 μm to avoid interaction.

2.6. Characterization

Fourier transform infrared (FTIR) spectra of MWCNTs and MWCNTs-COOH ground in KBr pallet were recorded on a Bio-Rad 165 FTIR spectrophotometer; 64 scans were signal-averaged with a resolution of 2 cm⁻¹ at room temperature. Thermogravimetric analyses (TGA) were carried out using a TA Instruments SDT 2960. Samples were heated at 10 °C min⁻¹ from room temperature to 800 °C in a dynamic nitrogen atmosphere (flow rate = 70 ml min⁻¹).

TEM observation of thin sections of the composite film was performed with a JEOL 2100 TEM under an acceleration voltage of 200 kV. Thin sections with thickness of about 70 nm of the composite film were cut from the prepared composite film embedded in epoxy resin under cryogenic conditions using a Leica ultramicrotome equipped with a diamond knife. To prepare TEM samples of pristine MWCNTs and MWCNTs-COOH, pristine MWCNTs and MWCNTs-COOH were dispersed into DMF and cast onto holey copper TEM grid. Surface resistance of composite film was measured using a Fluke 187 multimeter, and surface resistivity was calculated by multiplying the measured resistance with the width of the surface across which the current flows divided by the path length of the current along the surface. Film thickness was measured using a digimatic caliper.

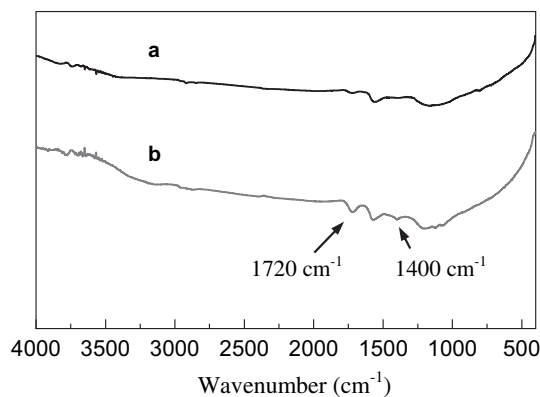


Fig. 1. FTIR spectra of pristine MWCNTs and MWCNTs-COOH: (a) pristine MWCNTs; (b) MWCNTs-COOH.

3. Results and discussion

In order to deposit both MWCNTs and PMDA-ODA colloidal particles onto the same electrode simultaneously, it is vital to prepare uniform and stable MWCNT-filled PMDA-ODA colloidal suspension with both MWCNTs and PMDA-ODA colloidal particles of the same charge. Here, negatively charged MWCNT suspension and PMDA-ODA colloids were prepared. PMDA-ODA was synthesized from condensation reaction of PMDA with equimolar ODA in NMP at room temperature. Then the synthesized PMDA-ODA was ionized by adding triethylamine to produce 20 mol% ionized carboxylic groups (COO⁻). Finally, negatively charged PMDA-ODA colloid was prepared by dropping methanol slowly into the prepared PMDA-ODA solution in NMP. The weight ratio of methanol to NMP was 3:1. The effective diameter and the average zeta potential of the prepared PMDA-ODA colloidal particles are around 600 nm and -20.5 mV, respectively, as measured by Zetaplus Particle Sizing equipment. The schematic illustration of the synthesis of PMDA-ODA and its ionization are shown in Scheme 1.

Carboxylated-MWCNTs (MWCNTs-COOH) were synthesized from pristine MWCNTs through acid-oxidation using dilute nitric acid (2.6 M). Attachment of COOH onto MWCNTs was verified by the observation of a characteristic band at 1720 cm⁻¹ corresponding to C=O stretching of COOH and the other at 1400 cm⁻¹ corresponding to O-H bending of COOH in the FTIR spectrum of MWCNTs-COOH (Fig. 1b), while there is no feature absorption peaks observed with the FTIR spectrum of pristine MWCNTs as shown in Fig. 1a.

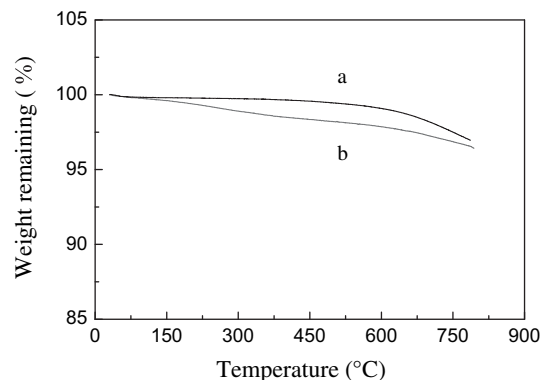


Fig. 2. TGA curves of pristine MWCNTs and MWCNTs-COOH: (a) pristine MWCNTs; (b) MWCNTs-COOH.

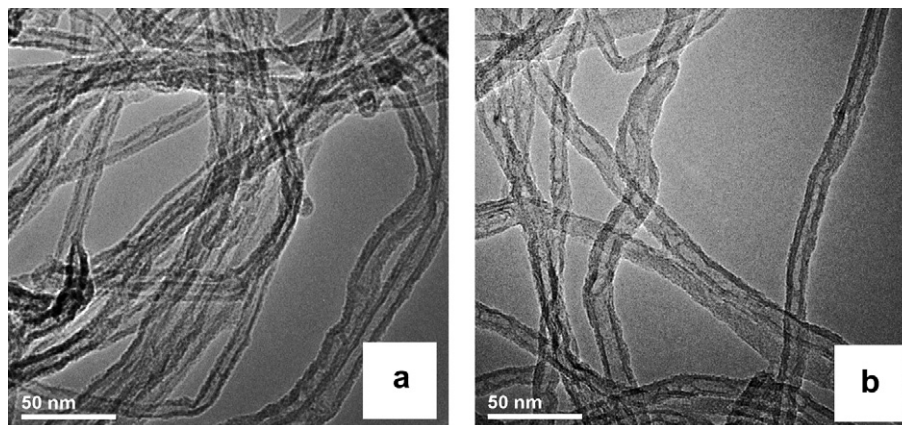


Fig. 3. TEM images of pristine MWCNTs and MWCNTs-COOH: (a) pristine MWCNTs; (b) MWCNTs-COOH.

To determine COOH content in MWCNTs-COOH, TGA was performed with both MWCNTs and MWCNTs-COOH under a nitrogen atmosphere as reported previously [23,24], and Fig. 2 shows the TGA scan results. As shown in Fig. 2a, pristine MWCNTs are thermally stable up to 600 °C and show negligible weight change. In contrast, MWCNTs-COOH shows a two-step thermal degradation, with the first step occurring between 150 °C and 550 °C and the second step starting from 600 °C at which the pristine MWCNTs starts to decompose. In comparison with the TGA curve of pristine MWCNTs, the first weight loss can be attributed to the decomposition of COOH in MWCNTs-COOH and the second weight loss to the decomposition of MWCNTs. Therefore, the COOH content in MWCNTs-COOH could be calculated from the two-step degradation profile, and it is 2.0 wt.%.

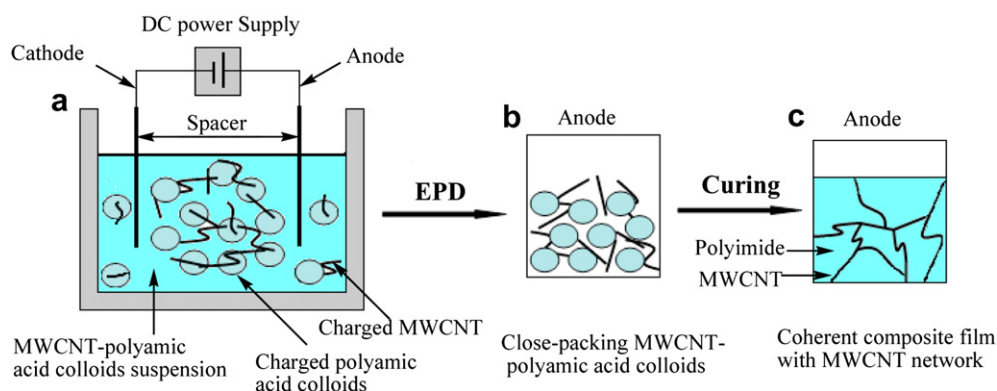
In order to study the influence of acid-oxidization on the integrity of MWCNTs, the morphology of pristine MWCNTs and MWCNTs-COOH were observed using TEM. As shown in Fig. 3, there is no apparent change on the diameter and wall thickness, demonstrating that acid-oxidization did not result in apparent detrimental effect on the MWCNTs structure.

MWCNT suspension was then prepared by suspending the obtained MWCNTs-COOH into methanol/NMP mixed solvent with identical ratio as that used in the preparation of PMDA-ODA colloids under ultrasonication for 5 min using high power probe ultrasonicator. The MWCNTs-COOH was further deprotonated to give negatively charged MWCNTs with COO^- groups through partial neutralization with triethylamine.

Uniform and stable MWCNT-filled PMDA-ODA colloidal suspension was prepared by mixing MWCNTs-COOH suspension

and PMDA-ODA colloid under water bath ultrasonication for 20 min. The concentration of PMDA-ODA was maintained at 1 wt.% and the content of MWCNTs-COOH to PMDA-ODA varied from 0.2 to 2 wt.%. The obtained MWCNT-filled PMDA-ODA colloidal suspension was quite stable even after one week.

The fabrication of MWCNT/polyimide composite films through electrophoretic deposition (EPD) is depicted in Scheme 2. Scheme 2a shows the schematic structure of an EPD apparatus used in our study. In our EPD apparatus, the anode functioned as the working-piece and it can be made up of Cr-coated silica wafer or any other conductive substrate intended for coating. The cathode at the other hand was a piece of Cr-coated silica wafer. Both cathode and anode were immersed into the MWCNT-filled PMDA-ODA colloidal suspension and separated from each other at a constant gap distance of 1.5 cm, as shown in Scheme 2a. The EPD was carried out over different deposition time under a constant electric current generated by a DC source supplied by LAMBDA InvenSys Genesys™. In the electric field, both negatively charged MWCNTs and PMDA-ODA colloidal particles migrate towards positively charged anode simultaneously, leading to close-packing of MWCNTs and PMDA-ODA colloidal particles on the working-piece, as reflected in Scheme 2b. Due to the excluded volume created by the PMDA-ODA colloidal particles, the MWCNTs are pushed into the interstitial space among PMDA-ODA colloidal particles to form MWCNT network. During curing, PMDA-ODA colloidal particles coalesce to form a coherent film and, through imidization at high temperature, the film was converted to polyimide film with MWCNT network impregnated within. Thus, a smooth MWCNT/polyimide composite film is produced on the



Scheme 2. Schematic description of the fabrication of MWCNT/polyimide composite films through EPD.

Table 1

Thickness of MWCNT/polyimide composite film fabricated on Cr-coated silicon wafer with various deposition time.

| Deposition time (Min) | 1 | 2 | 3 | 4 |
|----------------------------------|----|----|----|----|
| Film thickness (μm) | 16 | 31 | 41 | 55 |

DC current: 15 mA; MWCNT feeding content: 1 wt.% of PMDA-ODA.

anode (Scheme 2c). Unlike the conventional latex-based process, the close-packing of MWCNTs and PMDA-ODA colloidal particles in EPD is driven by electrical static force and not through solvent removal. Hence, this EPD approach for preparation of MWCNT/polyimide composite film can be employed for continuous fabrication in mass production using simple apparatus. Most importantly, it can be utilized to fabricate composite film with tunable thickness and electrical conductivity as well as onto variously-shaped surface.

The thickness of the prepared composite film can be easily tuned by varying the deposition time. Table 1 shows the thickness of MWCNT/polyimide composite film deposited on Cr-coated silica wafer over various deposition time. The DC current was 15 mA and MWCNT feeding content was 1 wt.% of PMDA-ODA. It can be seen that the film thickness increases from 16 to 55 μm with increasing deposition time from 1 to 4 min. This is because more MWCNTs and PMDA-ODA colloidal particles migrated onto the anode electrode with prolonged deposition time. The film thickness is also affected by the conductivity of the working-piece. For example, under identical DC current and colloidal suspension concentration, film fabricated on copper sheet had thickness of 73 μm , while that fabricated on Cr-coated silicon wafer was 55 μm in thickness. Copper sheet with higher electrical conductivity might facilitate the deposition of both MWCNTs and PMDA-ODA colloidal particles.

Fig. 4 shows the TEM image of MWCNT/polyimide composite film fabricated over 3 min with current of 15 mA and MWCNT feeding content of 0.8 wt.% of PMDA-ODA. The patchy distribution of MWCNTs verifies the assumption that MWCNTs were pushed into the interstitial space among PMDA-ODA colloidal particles to form MWCNT network during EPD. The TEM image also demonstrates the uniform distribution of MWCNTs within its network. Both qualities are essential for the fabrication of electrically conductive MWCNT/polyimide composite with low MWCNT content.

While neat polyimide film is electrically insulating, the fabricated MWCNT/polyimide composite film is electrically conductive when MWCNT content is high enough to form a conductive three dimensional MWCNT network in the film. Table 2 shows the surface resistivity of MWCNT/polyimide composite films fabricated on Cr-coated silicon wafer at various MWCNT feeding content with DC current of 15 mA and deposition time of 3 min. It shows that the

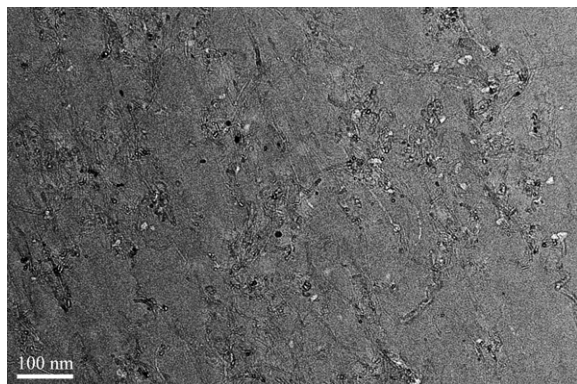


Fig. 4. TEM image of an MWCNT/polyimide composite film fabricated through EPD with MWCNT feeding content of 0.8 wt.% of PMDA-ODA and deposition time of 3 min.

Table 2

Surface resistivity of MWCNT/polyimide composite films fabricated on Cr-coated silicon wafer with various MWCNT feeding content.

| MWCNT content (wt.%) | 0.65 | 0.8 | 1.0 | 1.2 |
|--|-------------------|-------------------|-------------------|-------------------|
| Surface resistivity (Ω/\square) | 1.4×10^7 | 2.4×10^6 | 5.3×10^4 | 4.2×10^2 |

DC current: 15 mA; deposition time: 3 min.

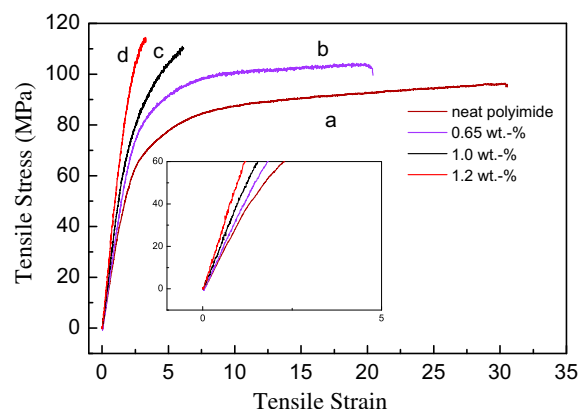


Fig. 5. Representative stress–strain curves for neat polyimide film and MWCNT/polyimide composite films with various MWCNT feeding content: (a) neat polyimide film; (b) 0.65 wt.% of PMDA-ODA; (c) 1.0 wt.% of PMDA-ODA and (d) 1.2 wt.% of PMDA-ODA. (DC current: 15 mA; Deposition time: 3 min; the inset shows the enlarged portion of stress–strain curve at the low strain regime).

surface resistivity of composite film decreases from 1.4×10^7 to $4.2 \times 10^2 \Omega/\square$ when MWCNT feeding content increased from 0.65 to 1.2 wt.%. This demonstrated that the electrical conductivity of obtained MWCNT/polyimide composite can be easily tuned through varying the MWCNT feeding content. At higher MWCNT feeding content, more MWCNTs will migrate to the anode to be incorporated into MWCNT/polyimide composite film. This leads to higher MWCNT content in the composite film showing higher electrical conductivity.

The mechanical reinforcement of polyimide using MWCNT was evaluated by tensile testing. Fig. 5 shows the representative stress–strain curves for neat polyimide film and MWCNT/polyimide composite films with various MWCNT feeding content fabricated at DC current of 15 mA and deposition time of 3 min, respectively, and the numerical values are summarized in Table 3. It can be seen that, in comparison with neat polyimide film, the MWCNT/polyimide composite films exhibit higher Young's modulus and tensile strength but smaller strain at break, and the strain at break decreases gradually with increasing MWCNT feeding content, while a significant increment in Young's modulus and a moderate increment in tensile strength are observed simultaneously. As listed in Table 3, the strain at break decreases from 32 ± 5 for neat polyimide film to

Table 3

Tensile properties of neat polyimide film and MWCNT/polyimide composite films fabricated with various MWCNT feeding content.

| Tensile properties | Neat polyimide | MWCNT/polyimide composite | | |
|------------------------|-----------------|----------------------------------|----------------------------------|-----------------------------------|
| | | 0.65 wt.% ^a | 1.0 wt.% ^a | 1.2 wt.% ^a |
| Young's modulus (GPa) | 3.6 ± 0.2 | 4.1 ± 0.6 (14) ^b | 4.8 ± 0.4 (33) ^b | 5.4 ± 0.1 (50) ^b |
| Tensile strength (MPa) | 101.5 ± 6.7 | 106.3 ± 2.3 (5) ^b | 110.7 ± 1.9 (9) ^b | 115.0 ± 3.6 (13) ^b |
| Strain at break (%) | 32 ± 5 | 21 ± 3 (34) ^b | 7 ± 1 (78) ^b | 3 ± 1 (91) ^b |

^a MWCNT feeding content of PMDA-ODA.

^b Value in parentheses represents percentage increase/decrease as compared to neat polyimide.

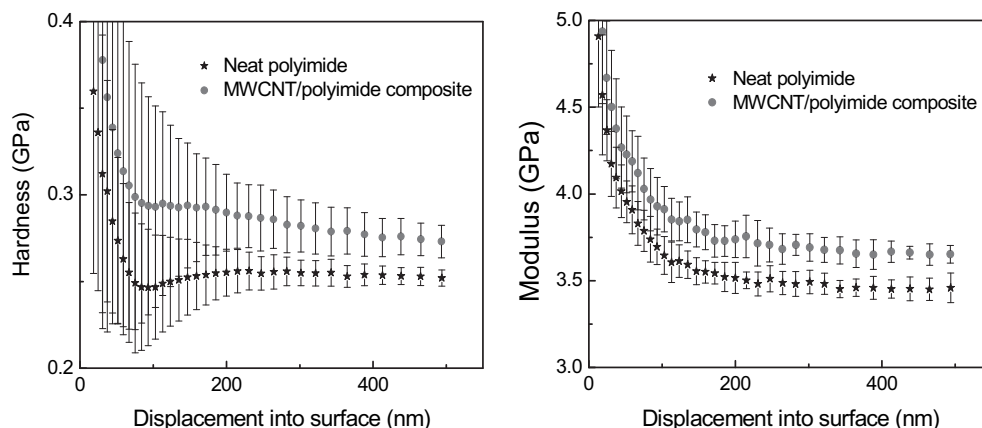


Fig. 6. E and H profiles with respect to displacement into sample surfaces for neat polyimide film and MWCNT/polyimide composite film with MWCNT feeding content of 1.0 wt.% of PMDA-ODA and deposition time of 3 min.

21 \pm 3, 7 \pm 3 and 3 \pm 1 for MWCNT/polyimide composite film with increasing the MWCNT feeding content from 0.65 to 1.0 and 1.2 wt.%, respectively. At the same time, the Young's modulus and tensile strength increase from 3.6 \pm 0.2 GPa and 101.5 \pm 6.7 MPa for neat polyimide film to 4.1 \pm 0.6 GPa and 106.3 \pm 2.3 MPa, 4.8 \pm 0.4 GPa and 110.7 \pm 1.9 MPa and 5.4 \pm 0.1 GPa and 115.0 \pm 3.6 MPa for MWCNT/polyimide composite film with increasing the MWCNT feeding content from 0.65 to 1.0 and 1.2 wt.%, respectively. The enlarged portion of stress–strain curves at the low strain regime as shown in the inset of Fig. 5 clearly demonstrate the modulus difference between the polyimide film and MWCNT/polyimide composite films fabricated with various MWCNT feeding content. Enhancement on Young's modulus and tensile strength with compromised strain at break is also commonly observed with other CNT reinforced polymer system. This may be due to the relatively weaker interfacial interaction between the MWCNT and matrix polyimide in comparison to those in other systems because of the lower COOH content in MWCNT-COOH (2.0 wt.% as measured using TGA).

The mechanical properties of neat polyimide film and MWCNT/polyimide composite film were further characterized by nanoindentation testing. Nanoindenter is a reliable equipment to study both elastic modulus (E) and hardness (H) of thin film fabricated on a substrate. Fig. 6 shows the E and H profiles with respect to indentation depth, respectively, for neat polyimide film and MWCNT/polyimide composite film with MWCNT feeding content of 1.0 wt.% of PMDA-ODA and deposition time of 3 min. It can be seen that both E and H of MWCNT/polyimide composite film are enhanced by about 10% compared to those of neat polyimide film. These increments are significant in view of the inherent high E and H of the neat polyimide.

During imidization at high temperature, COOH from MWCNTs is expected to react with NH from PMDA-ODA to form amide group that links MWCNTs and polyimide together, which in turn improves the MWCNT-polyimide interfacial interaction. Thus, polyimide is reinforced effectively.

4. Conclusion

A convenient approach to fabricate MWCNT/polyimide composite films with tunable thickness and electrical conductivity has been developed successfully. By utilizing EPD, MWCNTs have been dispersed into polyimide matrix uniformly to produce MWCNT/polyimide composite film. The patchy distribution of MWCNT network and uniform dispersion of MWCNTs in polyimide

matrix were verified by TEM study. In comparison to those of neat polyimide film, the results of tensile testing and nanoindentation testing with the prepared MWCNT/polyimide composite films demonstrated the mechanical reinforcement of polyimide film using MWCNTs. Unlike conventional latex-based method, solvent removal is not required in EPD and this is in favor of mass production. With tunable electrical conductivity, the prepared MWCNT/polyimide films have potential applications as electrostatic dissipating materials or electromagnetic interference shielding materials for device packaging and printed circuit boards. Moreover, we believe that the novel MWCNT/polymer composite film fabrication technique can efficiently be employed to fabricate MWCNT/polymer composite films onto irregular substrate.

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