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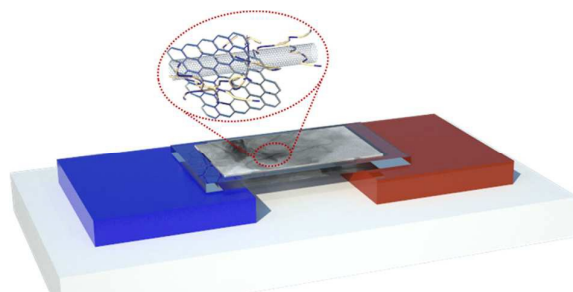
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Synergic effects of conducting polymer, carbon nanotubes, and graphene result in the enhanced thermoelectric properties of nanocomposites.



# Effects of One- and Two-Dimensional Carbon Hybridization of PEDOT:PSS on the Power Factor of Polymer Thermoelectric Energy Conversion Devices

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We investigated the thermoelectric properties of polymer composites based on a conducting polymer and carbon materials with various dimensionalities. PEDOT:PSS as a conducting polymer matrix was successfully hybridized with graphene sheets and multi-walled carbon nanotubes through in-situ polymerization of 3,4-ethylenedioxythiophene monomers in an aqueous solution with a presence of the carbon materials dispersed by using a polymeric dispersant. The hybrid structures of PEDOT:PSS, graphene, and carbon nanotubes in the composite showed an electrical conductivity, Seebeck coefficient, and power factor of 689 S/cm, 23.2  $\mu\text{V/K}$ , and 37.08  $\mu\text{W/mK}^2$ , respectively, values that are much higher than those of pristine PEDOT:PSS, PEDOT:PSS/graphene, or PEDOT:PSS/carbon-nanotube composites. The thermoelectric figure of merit increased from 0.017 in the pristine PEDOT:PSS to 0.031 in the composite, corresponding to an 80 % enhancement. We believe that the enhanced thermoelectric performance comes from the synergic effects of multi-component systems with excellent electrical bridging and electronic coupling between PEDOT and carbon materials.

## Introduction

Affordable and efficient thermoelectric devices that generate electricity from the temperature gradient have the potential to play a key role as a power source for portable electronics and as a means to recycle waste-heat.<sup>1-3</sup> Much of attention has been focused on inorganic semiconductors or alloy materials such as SiGe,<sup>4</sup> PbTe,<sup>5</sup> Bi<sub>2</sub>Te<sub>3</sub>,<sup>6</sup> and clathrates<sup>7</sup> for efficient thermoelectric materials. Although these materials have shown impressive thermoelectric performance, some of them are only efficient at temperature above 400 K and the others possess challenges such as a lack of abundance, limited processability, high cost, and the toxicity of constituent elements. To get around some or all of these limitations, alternative thermoelectric materials like organic and polymeric materials,<sup>8-11</sup> organic/inorganic hybrids,<sup>12-14</sup> inorganic/carbon composites,<sup>15,16</sup> and organic/carbon composites<sup>17-24</sup> have been investigated.

Intrinsically conducting polymers have been considered promising as thermoelectric materials because of low-cost manufacturing, easy processability, and ability to blend with other materials, as well as high electrical conductivity, high Seebeck coefficient, and low thermal conductivity. Among other conducting polymers, poly(3,4-

ethylenedioxythiophene):poly(4-styrenesulfonate) (PEDOT:PSS), which is a composite of a polythiophene derivative and a polymeric acid as a conducting component and a dispersant, respectively, is so far the most promising candidate, having a high electrical conductivity in ambient conditions, water-based processability, and easy control of oxidation levels.<sup>8,10,25,26</sup> Researchers have also applied hybridization by incorporating inorganic and carbon materials into PEDOT:PSS to further enhance the thermoelectric properties at ambient temperature.<sup>13,14,17</sup>

Low-dimensional carbon materials such as carbon nanotubes and graphene platelets have been very effective additives for PEDOT:PSS, showing stable hybridization and outstanding thermoelectric performance.<sup>17,22-24</sup> For example, Yu et al. obtained a thermopower of approximately 160  $\mu\text{W/mK}^2$  in the presence of single-walled carbon nanotubes (SWCNTs) with PEDOT:PSS.<sup>21</sup> Xu et al. acquired a power factor of 5.2  $\mu\text{W/mK}^2$  in PEDOT-reduced graphene oxide nanocomposites.<sup>22</sup> Woo et al. reported a thermoelectric power factor of 11.09  $\mu\text{W/mK}^2$  by using direct hybridization with PEDOT:PSS and 2 wt.% graphene bundles.<sup>23</sup> Yu et al. recorded a thermopower of approximately 25  $\mu\text{W/mK}^2$  from composites of PEDOT:PSS and 35 wt.% SWCNTs.<sup>24</sup> In these reports, carbon materials are

expected to affect the thermoelectric properties of PEDOT:PSS by electronic and electrical interaction between carbon and the polymers through electrostatic stacking. In our previous work, we studied the thermoelectric properties of PEDOT:PSS hybridized with graphene platelets via in-situ synthesis of the conducting polymer.<sup>17</sup> It was demonstrated that the presence of graphene sheets in the in-situ polymerized composites promotes charge transport and thermoelectric potential build-up across the composite thin films at the optimized point of graphene contents through strong interactions along the  $\pi$ - $\pi$  contacts between the graphene and PEDOT. It should be noted that the above-mentioned reports have only dealt with a single type of carbon material, and there have been only few studies that have tried to incorporate two different allotropes of carbon with different dimensionalities into the PEDOT:PSS matrix at the same time, and investigation of the effects of multi-dimensional carbon materials on the thermoelectric properties of polymer composites has not been performed.

The aim of the current study is to identify how hybridization of one-dimensional multi-walled carbon nanotubes (MWCNT) and two-dimensional graphene platelets with PEDOT:PSS influences the thermoelectric properties of the composites. We performed in-situ polymerization of 3,4-ethylenedioxythiophene (EDOT) monomers in water in the presence of MWCNTs and graphenes, dispersed by using a polymeric dispersant, PSS. The MWCNT bundles have high aspect ratios (>1000), providing a network of additional long-range bridges between conducting polymer domains. Graphene platelets, with their unique electronic properties, have strong electronic interactions with PEDOT:PSS. We found that the hybrid structures of graphenes and MWCNTs in the PEDOT:PSS/graphene/MWCNT (P/G/M) composites exhibited synergic effects. The thermoelectric figure-of-merit (ZT) of the P/G/M composites was higher than that of the pristine PEDOT:PSS by a factor of 1.8. The performance of the P/G/M samples was also better than that of the composites of PEDOT:PSS/graphene (P/G) or PEDOT:PSS/MWCNT (P/M) with the same weight ratios of carbon materials. Various analytical techniques were applied in order to understand the effects of the hybridization of carbon materials with different dimensionalities on the chemical and morphological features and electrical properties of the PEDOT:PSS composites.

## Experimental section

### Materials

Iron(III) sulfate [ $\text{Fe}_2(\text{SO}_4)_3$ , 97%], sodium persulfate ( $\text{Na}_2\text{S}_2\text{O}_8$ ,  $\geq 99.0\%$ ), 3,4-ethylenedioxythiophene (EDOT), cation exchange resin, anion exchange resin, and dimethyl sulfoxide (DMSO), and poly(sodium 4-styrenesulfonate) (PSS) were purchased from Aldrich Chemicals. Graphene platelets powder (N002-PDR, Angstrom Materials) and multi-walled carbon nanotube (MWCNT; TMC220-10, Nano Solution) bundles were obtained. All solvents and chemical reagents were used as received without any further purification.

### Preparation of PSS/carbon material dispersions

PSS was used as a dispersant for the graphenes and MWCNTs, similar to our previous report.<sup>17</sup> PSS was treated with a mixture of cation and anion exchange resins to eliminate extra ions before we utilized it as a dispersant. The desired amount of carbon materials (0.12 g or 0.20 g) and PSS (4.0 g) were directly added into deionized water (580 g). The aqueous mixture of PSS/carbon materials was stirred for 30 min by ultrasonication with a tip sonicator in order to obtain a well-dispersed system of PSS/carbon materials. The samples were prepared with graphene (denoted as 'G'), MWCNT ('M'), or a 1:1 weight ratio of graphene/MWCNT mixture ('G/M'). The carbon materials were 3 wt% or 5 wt% of PSS solid contents. The total composition of carbon materials was limited to 5 wt% in our experiment, because nanocomposites with more carbon materials had a large amount of agglomerates in thin films.

### In-situ polymerization of PEDOT with a presence of PSS and carbon materials.

The aqueous dispersion of PSS/carbon materials was bubbled with inert argon gas (99.999 %) for 60 min at a rate of 3 L/min in order to remove the dissolved oxygen from the aqueous medium and minimize over-oxidation of EDOT units during the polymerization. The EDOT monomer (5.01 g) was added and mixed with a mechanical stirrer for 30 min in the dispersion. The oxidizing reagents of iron (III) sulfate (0.20 g;  $5.01 \times 10^{-4}$  mol) and sodium persulfate (8.02 g;  $3.45 \times 10^{-2}$  mol) were dissolved in 50 mL of deionized water. The solutions were then added to the EDOT and PSS/carbon material dispersion to facilitate oxidative polymerization of EDOT with a presence of PSS and the carbon materials. The polymerization was carried out for 48 h at 5 °C with bubbling inert argon gas. After the polymerization, the composite product was blended with 300 mL of a mixture of bead-type cation and anion exchange resins for 1 h, followed by filtering through a 30- $\mu\text{m}$  mesh. As a control sample, pristine PEDOT:PSS was also prepared without the addition of any carbon materials.

### Characterization of PEDOT:PSS/carbon composites.

In order to analyse the electrical and thermoelectric properties of the composites, thin films of the composites were prepared by the bar-coating method on a glass substrate. Prior to the deposition, the dispersion of PEDOT:PSS/carbon material hybrid was mixed with DMSO and stirred for more than 1 h at ambient conditions to enhance the electrical conductivity.<sup>27</sup> The as-coated thin films were dried at 150 °C for 2 min in a convection oven. The gold electrodes for measurement of the conductivity and the Seebeck coefficient were deposited onto the surface of the hybrid film with a distance of 10 mm to minimize electrical contact resistance. The conductivity of the samples was obtained by the van der Pauw method using a home-built system. The Seebeck coefficient of the hybrid samples was measured by using two Peltier devices attached to an aluminum heat sink using a thermal paste to prevent thermal

disturbance and to maintain a controlled temperature gradient across the thin films.

The Fourier-transform infrared spectra (FT-IR) and X-ray diffraction (XRD) data were obtained from the composite thin films by using an FT-IR spectrophotometer (SENSOR 27, Bruker) and an X-ray diffractometer (Ultima IV, RIGAKU), respectively. The electron binding energy of the composites was measured by using X-ray photoelectron spectroscopy (XPS; K-alpha, Thermo U.K.) equipped with a monochromatic Al K $\alpha$  X-ray source (1486.6 eV). Raman spectra of the composite films were obtained by a Raman spectrometer (LabRam Aramis, Horriba Jovin Yvon) using a Nd:Yag laser source with a wavelength of 532 nm. Thickness of the composite films was measured with a surface profiler (Alpha Step IQ, KLA-Tencor). The morphology of the composites was imaged by using a transmission electron microscope (TEM; JEM-2010, JEOL Ltd.) operating at 200 kV. The thermal conductivity ( $\kappa$ ) of the composites was obtained by using a laser flash apparatus (Netzsch LFA 457, MicroFlash Instruments Inc.) with a xenon flash lamp. The thermal conductivity is a function of the specific heat ( $C_p$ ), thermal diffusivity ( $D$ ), and density ( $\rho$ ) of the material, according to the relationship:  $\kappa = C_p D \rho$ .

## Results and discussion

Fig. 1 shows a schematic illustration of the direct synthesis of PEDOT:PSS/carbon composites in an aqueous medium. The carbon materials were added in a deionized water medium with the PSS dispersant. Then the mixed solution containing the carbon materials and PSS was subsequently homogenized by using a probe-tip sonicator. Finally, EDOT monomers were dispersed and polymerized in the PSS/carbon dispersion. PEDOT:PSS/MWCNT (hereafter denoted as 'P/M') composites and PEDOT:PSS/graphene (denoted as 'P/G') composites were synthesized in the presence of one-dimensional MWCNT bundles and two-dimensional graphene platelets, respectively. PEDOT:PSS/graphene/MWCNT (denoted as 'P/G/M') composites were synthesized by polymerization with MWCNT and graphene. As a control sample, pristine PEDOT:PSS was also prepared by polymerizing EDOT monomers in an aqueous solution of PSS without any carbon materials. It is important to stabilize the dispersion for processing of polymer/carbon hybrid systems.<sup>17</sup> PSS worked as a polymeric surfactant in an aqueous medium in our system, providing nanometer-scale polymerization sites for EDOT and maintaining stable dispersion of the PEDOT/carbon hybrid nanoparticles in water. For this reason, aqueous dispersions of PEDOT:PSS and PEDOT:PSS/carbon material composites have long-term stability of more than a month.

We confirmed the chemical structures of the synthesized PEDOT:PSS and P/G/M (5 wt%) composite films by using FT-IR spectra obtained in the range from 2000  $\text{cm}^{-1}$  to 400  $\text{cm}^{-1}$  (Fig. 2). The IR bands of 1520, 1310, and 1201  $\text{cm}^{-1}$  are mainly from the C=C and C-C stretching of the quinoidal structure of the thiophene rings and the sulfonic acid groups of PSS. The

bands at 1143 and 1090  $\text{cm}^{-1}$  are attributed to the C-O-C bond stretching in the ethylene dioxide units. The C-S stretching mode has IR bands at 982, 840, and 691  $\text{cm}^{-1}$ .<sup>28</sup> The FT-IR spectra indicate that the polymerization of EDOT with the existence of graphenes and MWCNTs was successful in the aqueous medium.

The X-ray diffraction (XRD) patterns of the graphene platelets, MWCNT bundles, PEDOT:PSS films, and P/G/M 5 wt% composites were taken in order to characterize the solid-state morphology (Fig. 3). The pristine graphene platelets and MWCNT bundles have a broad peak at 21.5° and 26.2°, respectively. Diffraction of the PEDOT:PSS films and P/G/M composites shows patterns similar to each other, with two features at 17.5° and 26.5°. There is no detectable XRD feature from either graphenes or MWCNTs in the P/G/M composites, suggesting that the stacked structures of graphene platelets and MWCNT bundles disappeared and that the carbon materials were well dispersed in the PEDOT:PSS during the in-situ polymerization.<sup>29</sup>

To visualize the morphology of the samples, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images were obtained. Fig. 4 shows the images of pristine samples of MWCNT bundles, graphene platelets, PEDOT:PSS, and their composites. The MWCNT bundles were typically 10–15 nm wide and 10–20  $\mu\text{m}$  long [Fig. 4(a)]. The pristine graphene platelets had a diameter of 1–2  $\mu\text{m}$  and exhibited porous structures consisting of expanded multiple layers similar to a croissant [Fig. 4(b)]. The PEDOT:PSS films had nanometer-size dark PEDOT grains in the bright PSS matrix [Fig. 4(c)]. The images in Fig. 4(d–f) are P/M, P/G, and P/G/M composites, respectively, all with 5 wt% carbon materials in the PEDOT:PSS matrix. In the P/M composites, the conductive PEDOT grains were connected by long MWCNTs. The P/G composites had graphene sheets covered with conductive PEDOT grains as dark lumps. In the P/G/M composites, graphene sheets covered with conductive PEDOT were combined with long MWCNTs, forming electrical meshes.

X-ray photoelectron spectroscopy (XPS) analysis was conducted to study changes in the electronic environment in the carbon composites. Figure 5 shows the carbon-carbon binding peaks of PEDOT:PSS, P/M, P/G and P/G/M samples, deconvoluted from the C1s spectra shown in Figure S3. The electron binding energy of C 1s in a carbon-carbon bond is typically at 284–285 eV. The corresponding binding energy peaks in the PEDOT:PSS and P/M 5 wt% samples were 284.38 and 284.42 eV, respectively, suggesting a small interaction that can affect the electronic environment. In the case of P/G 5 wt%, the XPS peak shifted to 284.47 eV which implies a stronger interaction between PEDOT:PSS and graphene than between PEDOT:PSS and MWCNT. The larger shift in P/G than in P/M originates in part from the larger contact area of the planar graphene platelets than of the one-dimensional MWCNTs with a high aspect ratio (>1000) when in contact with PEDOT:PSS nanoparticles. We note that such a shift toward a higher binding energy has also been previously observed and attributed to the



sign of the partial charge exchange between PEDOT:PSS and graphene due to the  $\pi$ - $\pi$  interactions.<sup>23</sup> The P/G/M 5 wt% composite had an even larger degree of interactions as the binding energy reached 284.53 eV.

Raman spectroscopy confirmed the effects of electrical stacking and interaction between PEDOT and the carbon materials (Fig. 6). The spectrum of the pristine PEDOT:PSS has four main peaks indicating C-C inter-ring stretching ( $1261\text{ cm}^{-1}$ ), C-C single-bond stretching ( $1364\text{ cm}^{-1}$ ), C=C symmetrical stretching ( $1429\text{ cm}^{-1}$ ), and C=C anti-symmetrical stretching ( $1501\text{ cm}^{-1}$ ) modes, respectively.<sup>30</sup> Focusing on the Raman bands of the C=C symmetrical stretching mode in the samples, we can see that the band of  $1431\text{ cm}^{-1}$  in the P/M 5 wt% composite is very close to that of  $1429\text{ cm}^{-1}$  in the pristine PEDOT:PSS. This negligible shift shows that  $\pi$ - $\pi$  interactions between the PEDOT grains and the MWCNTs are not significant. On the other hand, the same C=C symmetrical stretching bands in P/G 5 wt% and P/G/M 5 wt% composites shifted to larger wavenumber at  $1441\text{ cm}^{-1}$  and  $1442\text{ cm}^{-1}$ , respectively. The strong shift of the Raman bands when graphene is incorporated confirms again the strong interactions between the constituent materials.<sup>22,31</sup>

With the hybridization of MWCNTs and graphenes in the P/G/M composites, we were able to achieve the synergic effects of multi-dimensional electrical bridges on the electrical conductivity and the thermoelectric properties (Fig. 7 and Table 1). Although the pristine conducting polymer, PEDOT:PSS, had an electrical conductivity of 548 S/cm and a Seebeck coefficient of  $18.0\text{ }\mu\text{V/K}$ , the polymer/carbon composite films showed higher electrical conductivity and Seebeck coefficient. The increment of the values also increases with the incorporation of more carbon materials in the composite. The P/M composites consisting of PEDOT:PSS and 5 wt% MWCNTs had a conductivity of 612 S/cm and a Seebeck coefficient of  $19.0\text{ }\mu\text{V/K}$ . A more positive effect on the electrical properties was obtained when graphene platelets were added, resulting in a conductivity of 638 S/cm and a Seebeck coefficient of  $20.7\text{ }\mu\text{V/K}$ . We believe that the main reason for the conductivity enhancement in the composites is better long-range electrical bridging between conductive domains through closer interaction. Although both graphene and MWCNT can have high conductivity, the effect is more significant for graphene as it has a two-dimensional shape and a stronger interaction with PEDOT:PSS, as revealed by XPS and Raman analyses. The high charge-carrier mobility of the individual graphene sheet and MWCNT might also be the reason for the enhanced Seebeck coefficient in the composites.<sup>23</sup> The effect on the Seebeck coefficient may be more significant in the graphene composites because of the stronger interaction. Interestingly, the P/G/M composites, which have PEDOT:PSS and both graphene platelets and MWCNTs, shows much better enhancement. The conductivity and Seebeck coefficient are 689 S/cm and  $23.2\text{ }\mu\text{V/K}$ , respectively, in the P/G/M composite films with 5 wt% carbon materials. With this synergic effect as a result of carbon material incorporation, the power factor, a product of the conductivity and the square of the Seebeck

coefficient ( $\text{PF}=\sigma\times S^2$ ), also increased in the composites. The P/G/M 5 wt% composite sample showed a power factor of  $37.08\text{ }\mu\text{W/mK}^2$  as the topmost value in this study.

One of the most important factors that govern the thermoelectric properties is the thermal conductivity ( $\kappa$ ) of the material. Therefore, we also characterized the thermal conductivity of the pristine PEDOT:PSS, P/M, P/G, and P/G/M composites. As shown in Fig. 7(e), the pristine PEDOT:PSS had a thermal conductivity of  $0.313\text{ W/mK}$ , which is similar to the value reported in the literature.<sup>32</sup> The P/M, P/G, and P/G/M composites containing the hybridizing carbon materials exhibited thermal conductivities of 0.345, 0.338, and  $0.360\text{ W/mK}$ , respectively. These values in the composites are higher than that in the pristine PEDOT:PSS. The increase in the thermal conductivity is likely from the fact that the pristine states of the carbon materials (MWCNTs and graphenes) without hybridization have higher thermal conductivities. The thermal conductivities of MWCNT and graphene have been reported to be 200 and  $500\text{ W/mK}$ , respectively.<sup>33,34</sup>

We evaluated the thermoelectric figure of merit (ZT) from the electrical conductivity, Seebeck coefficient, and thermal conductivity. The ZT values of pristine PEDOT:PSS, P/M, P/G, and P/G/M composites are summarized in Table 1 and plotted in Fig. 7(f). All the hybrid composites have higher ZTs than the pristine PEDOT:PSS. The P/G/M composite in particular revealed the highest ZT value of 0.031 at ambient temperature, corresponding to an 80 % enhancement as compared with the pristine PEDOT:PSS. These results demonstrate that hybridization of conducting polymers and various carbon materials in different dimensionalities has a synergic effect on the properties of polymer/carbon composites. The electrical conductivity and thermoelectric properties of the composite films can be significantly enhanced by taking advantages of the properties of the carbon materials.

## Conclusions

In summary, we have demonstrated that the electrical conductivity and Seebeck coefficient of PEDOT:PSS composites could be tuned by controlling the hybrid composition of carbon materials during in-situ polymerization of PEDOT:PSS. A thorough analysis on the polymer/carbon composites was performed by using SEM, TEM, FT-IR, Raman spectroscopy, and XPS, as well as electrical and thermoelectric measurements. We found that the conducting polymers and carbon materials with a low-dimensionality have strong electronic interactions that result in better electrical bridging between conductive domains. We also found that the synergic effects between PEDOT:PSS and carbon materials improve significantly when carbons with different dimensionalities, namely multi-walled carbon nanotubes and graphene platelets, are simultaneously incorporated, as the advantageous properties of both materials can be utilized. As a result, the hybrid composites of PEDOT:PSS, graphenes, and MWCNTs with 5 wt% carbon content showed higher electrical conductivity, Seebeck coefficient, power factor, and thermoelectric figure of

merit as compared to the pristine PEDOT:PSS and composites of PEDOT:PSS and single-type carbon material. These results emphasize the potential of in-situ polymerization and hybridization of conducting polymers with multiple low-dimensional materials for development of polymer-based energy devices such as thermoelectric modules, organic photovoltaic cells, and supercapacitors.

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Table 1. Electrical and thermoelectrical properties of PEDOT:PSS/carbon composites with various carbon types and compositions at room temperature.

Sample	Electrical conductivity (S/cm)	Seebeck coefficient ( $\mu\text{V/K}$ )	Power factor ( $\mu\text{W/mK}^2$ )	Thermal conductivity (W/mK)	Figure-of-merit, ZT
PEDOT:PSS	548	18.0	17.75	0.313	0.017
Pristine graphene platelets (G)	5.32	0.89	0.00042		
Pristine MWCNT bundles (M)	0.07	9.25	0.00059		
PEDOT:PSS / MWCNT 3 wt.% (P/M 3)	582	18.3	19.49		
PEDOT:PSS / MWCNT 5 wt.% (P/M 5)	612	19.0	22.09	0.345	0.019
PEDOT:PSS / graphene 3 wt.% (P/G 3)	591	19.1	21.56		
PEDOT:PSS / graphene 5 wt.% (P/G 5)	638	20.7	27.34	0.338	0.024
PEDOT:PSS / graphene / MWCNT 3 wt.% (P/G/M 3) <sup>a</sup>	621	19.8	24.35		
PEDOT:PSS / graphene / MWCNT 5 wt.% (P/G/M 5) <sup>a</sup>	689	23.2	37.08	0.360	0.031

<sup>a</sup> 1:1 weight ratio of graphene and MWCNT in the carbon mixture.



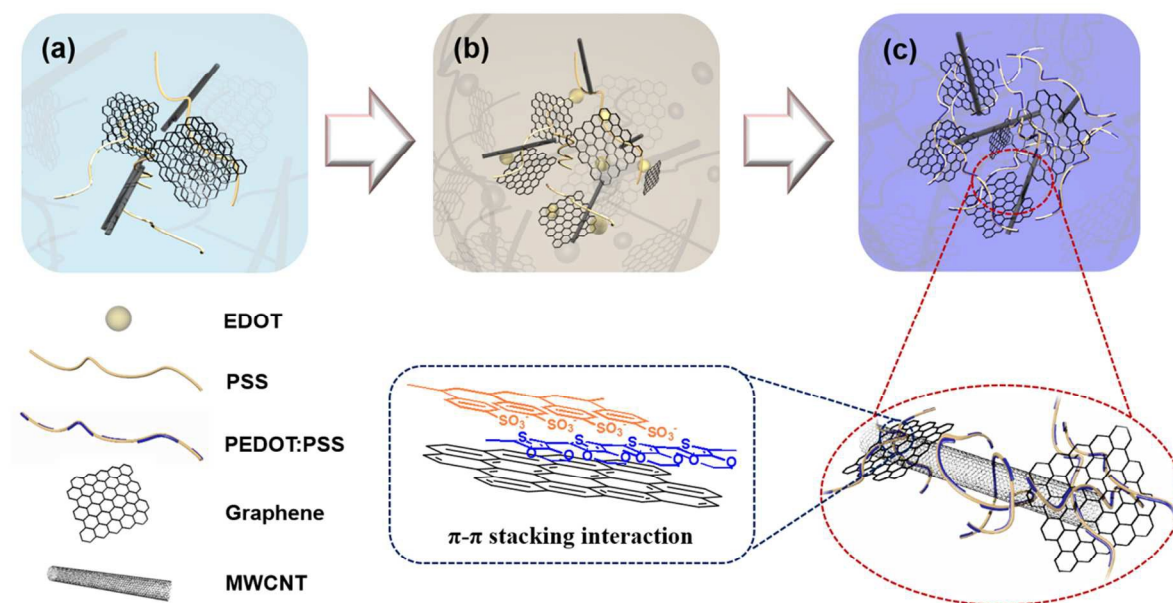


Fig. 1. Synthetic scheme of direct in-situ synthesis of PEDOT:PSS/graphene/MWCNT (P/G/M) composites.

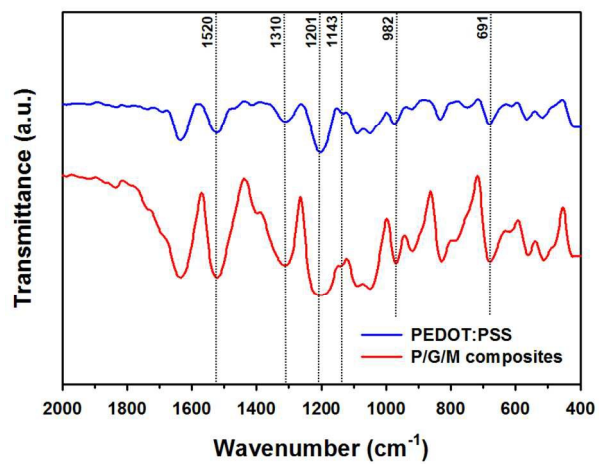


Fig. 2. FT-IR spectra of PEDOT:PSS and PEDOT:PSS/graphene/MWCNT (P/G/M) composites.

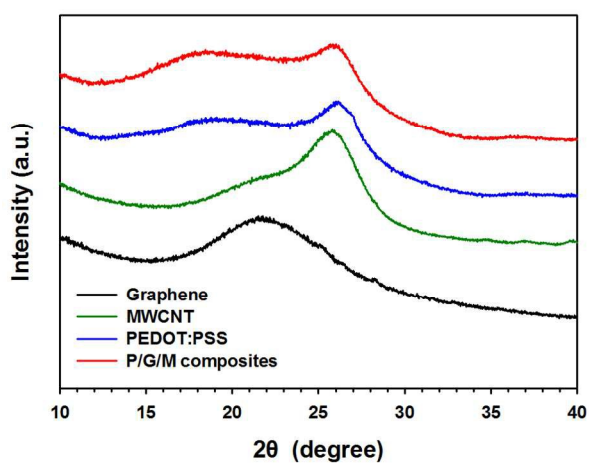


Fig. 3. XRD patterns of thin films of graphene, MWCNT, PEDOT:PSS, and PEDOT:PSS/graphene/MWCNT (P/G/M) composites.

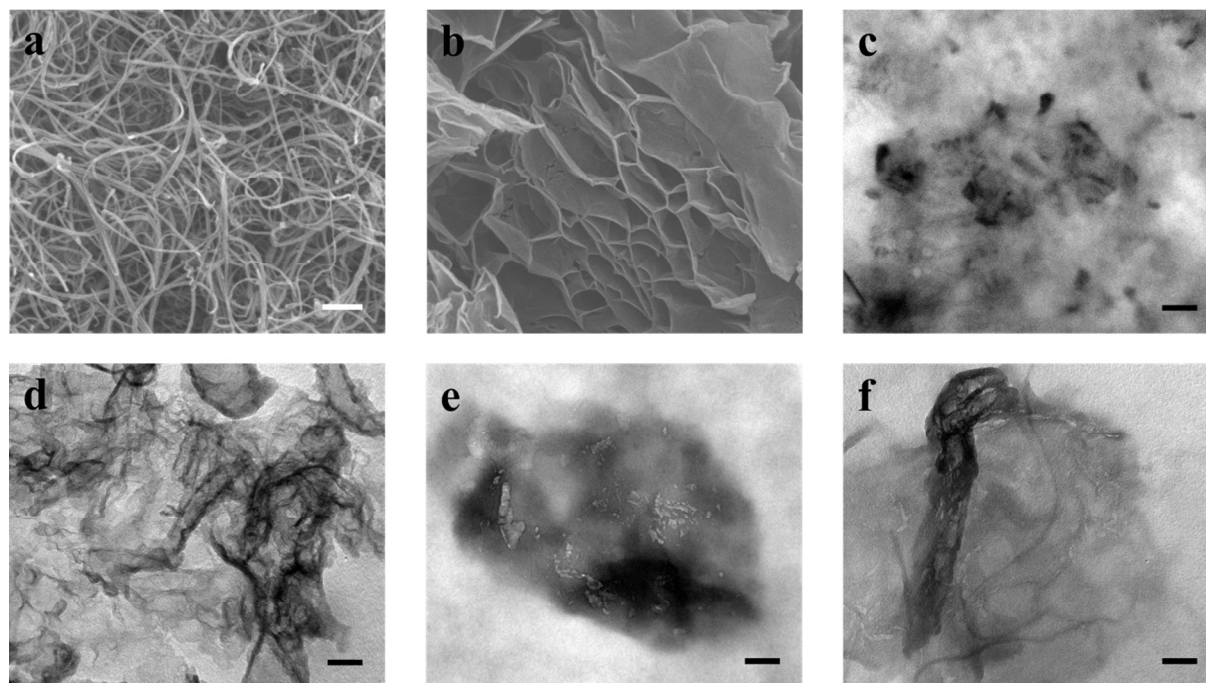


Fig. 4. Morphology of the polymer/carbon composites. SEM images of (a) MWCNT bundles and (b) graphene platelets. TEM images of (c) PEDOT:PSS, (d) PEDOT:PSS/MWCNT (P/M) composites, (e) PEDOT:PSS/graphene (P/G) composites, and (f) PEDOT:PSS/graphene/MWCNT (P/G/M) composites (scale bar : 100 nm).

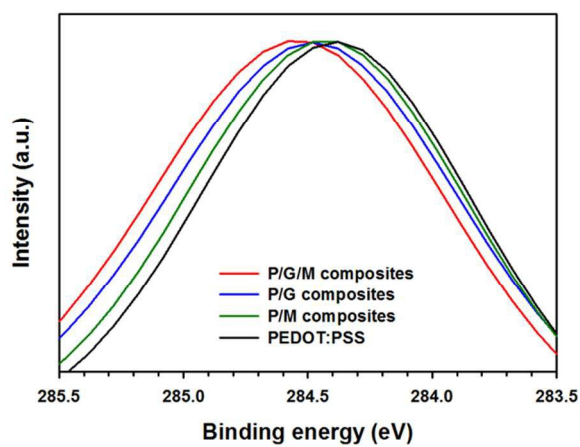


Fig. 5. Carbon-carbon binding peaks of deconvoluted C1s XPS spectra of PEDOT:PSS, PEDOT:PSS/MWCNT (P/M) composites, PEDOT:PSS/graphene (P/G) composites, and PEDOT:PSS/graphene/MWCNT (P/G/M) composites.



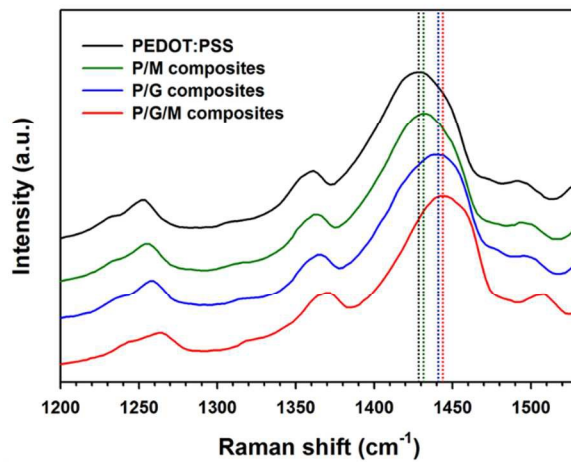


Fig. 6. Raman spectra of PEDOT:PSS, PEDOT:PSS/MWCNT (P/M) composites, PEDOT:PSS/graphene (P/G) composites, and PEDOT:PSS/graphene/MWCNT (P/G/M) composites.

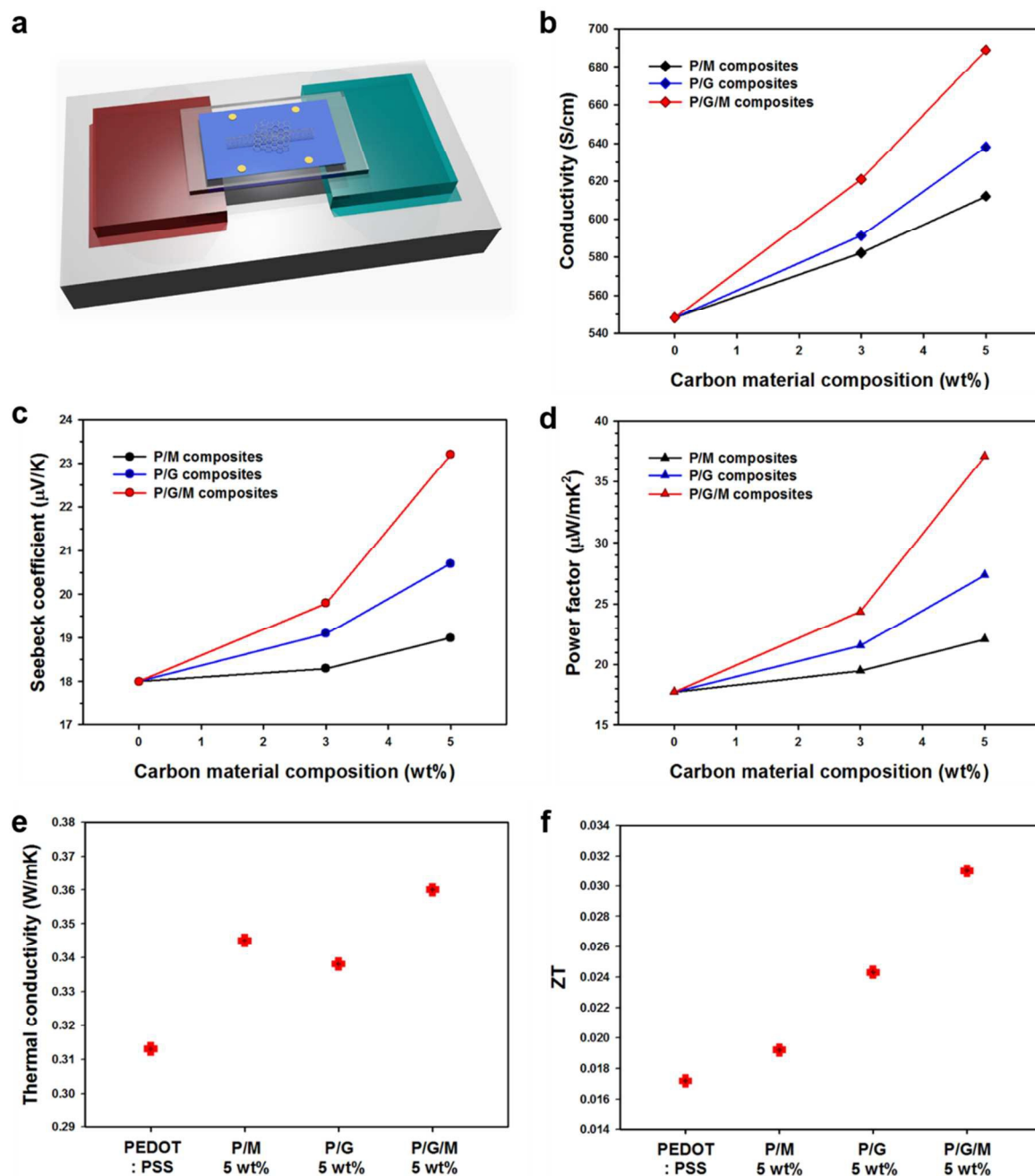


Fig. 7. (a) Schematic illustration of thermoelectric measurement setup. (b) Electrical conductivity, (c) Seebeck coefficient, (d) power factor, (e) thermal conductivity, and (f) thermoelectric figure of merit (ZT) of PEDOT:PSS, PEDOT:PSS/MWCNT (P/M) composite, PEDOT:PSS/graphene (P/G) composite, and PEDOT:PSS/graphene/MWCNT (P/G/M) composite films.