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Thermoelectric and mechanical performances of ionic liquid-modulated PEDOT:PSS/SWCNT composites at high temperatures

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Abstract

Significant progress has been achieved for flexible polymer thermoelectric (TE) composites in the last decade due to their potential application in wearable devices and sensors. In sharp contrast to the exceptional increase in TE studies at room temperature, the mechanical performance of polymer TE composites has received relatively less attention despite the significance of the application of TE composites in high-temperature environments. The TE and mechanical performances of flexible poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/single-walled carbon nanotube (PEDOT:PSS/SWCNT) composite films with an ionic liquid (IL) (referred to as "PEDOT:PSS/SWCNT-IL") at high temperatures are studied in the present work. The fabricated composite films show increasing TE performance with increasing temperature and SWCNT content. The maximum value of the power factor reaches 301.35 μW m^{-1} K^{-2} at 470 K for the PEDOT:PSS/SWCNT-IL composite. Furthermore, the addition of the IL improves the elongation at break of the composites compared to the IL-free composites. This work promotes the advancement of flexible polymer TE composites and widens their potential applications at different temperature ranges.

Keywords: Thermoelectrics, composite films, ionic liquids, mechanical performance

INTRODUCTION

Thermoelectric (TE) materials have been studied extensively as a result of their unique ability to realize energy conversion between heat and electricity and their excellent potential for a range of versatile applications in complex environments to harvest waste heat[1-7]. TE conversion efficiency is reliant on TE performance, which is usually assessed by a dimensionless parameter known as the TE figure of merit ($ZT = \sigma S^2 T/\kappa$, where σ, S, κ and T are the electrical conductivity, Seebeck coefficient, thermal conductivity and absolute temperature, respectively). Furthermore, the power factor ($PF = S^2\sigma$) is also adopted to evaluate the TE performance of polymers and polymer composites due to their low or similar thermal conductivity[8].

In recent decades, inorganic TE materials (e.g., Bi_2Te_3, Sb_2Te_3 and their alloys) have been studied intensively due to their high TE performance[9-11]. However, their applications are impeded by their inevitable drawbacks that include intrinsic rigidity, toxicity, high cost and scarcity. Compared to their inorganic counterparts, conducting polymer (CP)-based flexible TE materials exhibit advantages such as intrinsic flexibility, low thermal conductivity and low toxicity. Poly(3,4-ethylenedioxythiophene) (PEDOT)[12,13], polypyrrole[14,15] and polyaniline[16] are the main CPs for TE applications[17,18]. Of these, PEDOT is one of the most successful CPs as a result of its satisfactory TE performance and commercial availability. The main issue for improving the TE performance of CPs is their low electrical conductivity. CP/carbon nanotube (CNT) TE composites represent an effective alternative for improved TE performance by utilizing the effective interfacial π-π conjugated interaction between CPs and CNTs[19,20]. Indeed, significant progress in the improvement of TE performance has been achieved for CP/CNT composites at room temperature[21-23]. However, their high-temperature TE performance has received significantly less attention than their room-temperature properties. In addition, it has been demonstrated that there is a trade-off between TE and mechanical performances[24-26]. The issue of whether this trade-off is valid or not has scarcely been discussed for CP/CNT composites at high temperatures. Although this trade-off is vital for the potential application of TE composites at high temperatures, the related properties of the composites remain unknown.

In our previous work, we reported the preparation of flexible PEDOT:poly(styrene sulfonate) (PSS)/single-walled (SW)CNT films with ionic liquid (IL) additives (referred to as "PEDOT:PSS/SWCNT-IL")[27]. The composite films showed an enhanced TE performance after the addition of IL into the system. In the present work, the temperature dependence of the TE performance of PEDOT:PSS/SWCNT-IL films is studied relative to IL-free PEDOT:PSS/SWCNT films. The carrier transport mechanism is discussed using the variable range hopping (VRH) model. The mechanical properties of PEDOT:PSS/SWCNT-IL and IL-free PEDOT:PSS/SWCNT are also evaluated at different high temperatures.

EXPERIMENTAL

Chemicals and materials

SWCNTs (purity of > 90 wt.% and diameter of 1-2 nm) were purchased from Shenzhen Nanotech Port Co., Ltd., China. A PEDOT:PSS aqueous dispersion with a solid content of 1.1 wt.% (Clevios PH 1000) was obtained from Heraeus Materials Technology Shanghai Ltd., China. The molecular formula of PEDOT:PSS is shown in Figure 1. 1-Butyl-3-(4-sulfobutyl)imidazolium trifluoromethanesulfonate IL 95% (HPLC grade) was provided by Shanghai Mackin Biochemical Co., Ltd., China and its molecular formula is shown in Figure 1. The selection of IL additives was based on their multiple roles in enhancing the TE performance of the PEDOT:PSS/SWCNT system[27].

Preparation of PEDOT/SWCNT films with IL

Figure 1 shows the preparation procedure of the flexible PEDOT/SWCNT films. Here, the fabrication procedure of a composite film of PEDOT:PSS/SWCNT (70/30 wt./wt.) with 50 wt.% IL is described as an

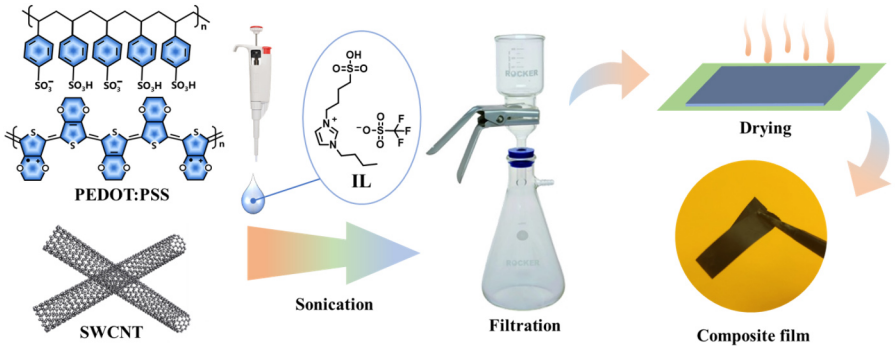

Figure 1. Schematic illustration of the preparation procedure for the PEDOT:PSS/SWCNT composite films with IL. PEDOT:PSS/SWCNT: Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/single-walled carbon nanotube; IL: ionic liquid.

example. In short, the SWCNTs (9.64 mg) and IL (8 μL) were successively added to an ethanol solution and ultrasonically dispersed to form a homogeneous SWCNT dispersion containing the IL, followed by the addition of a PEDOT:PSS aqueous solution (2 mL) with 20 min of sonication to form a homogeneously dispersed PEDOT:PSS/SWCNT solution with the IL. The final composite film was obtained by vacuum filtration, repeated rinsing and vacuum drying [Figure 1]. The detailed preparation procedures can be found in our previous work[27]. For brevity, the PEDOT:PSS/SWCNT composites without IL and the PEDOT:PSS/SWCNT composites with an IL content of 50 wt.% are referred to as IL-free PEDOT:PSS/SWCNT and PEDOT:PSS/SWCNT-IL, respectively. The IL content was chosen at 50 wt.% because it is the optimal content for TE performance at room temperature[27]. As emphasized in our previous work[27], the IL content is relative to the solid contents of PEDOT:PSS and SWCNTs, with the repeated rinsing procedure washing out most of the IL. Furthermore, for comparison, a pure SWCNT film (32.14 mg) and SWCNTs (32.14 mg) with an IL content of 50 wt.% were prepared without the addition of PEDOT:PSS.

TE and mechanical performance measurements

The temperature dependencies of the electrical conductivities (σ) and the Seebeck coefficients (S) in a temperature range from 298 to 470 K were measured by a commercially available Thin-Film Change Temperature Thermoelectric Parameter Test System (MRS-3 CT, Wuhan Joule Yacht Science & Technology Co., Ltd., China), with a quasi-steady-state mode was applied. The TE performance was evaluated by the power factor ($PF = \sigma S^2$) in the present work. The room-temperature tensile stress-strain measurements were performed using an EUT4103 testing instrument with a speed of 3 mm min^{-1} at 298 and 470 K. The high-temperature tensile stress-strain measurements were performed with the same instrument with a temperature-controlled chamber at the target temperature.

Structural and morphological characterization

The SWCNTs were observed with high-resolution scanning electronic microscopy (SEM, FEI APREO S). The statistical distribution of SWCNT bundles was analyzed using Image J software based on the SEM images. The carrier concentration and mobility were measured using an Ecopia HMS-3000 Hall effect measurement system. Thermogravimetric analysis (TGA) was performed on a TA TGA-Q50 thermal analysis system in a N_2 atmosphere from room temperature to 1073 K at a heating speed of 10 K min^{-1}.

RESULTS AND DISCUSSION

Morphologies of neat SWCNT and SWCNT-IL films

Typical SEM images of the SWCNT and SWCNT-IL films are compared in Figure 2. Basically, SWCNTs in

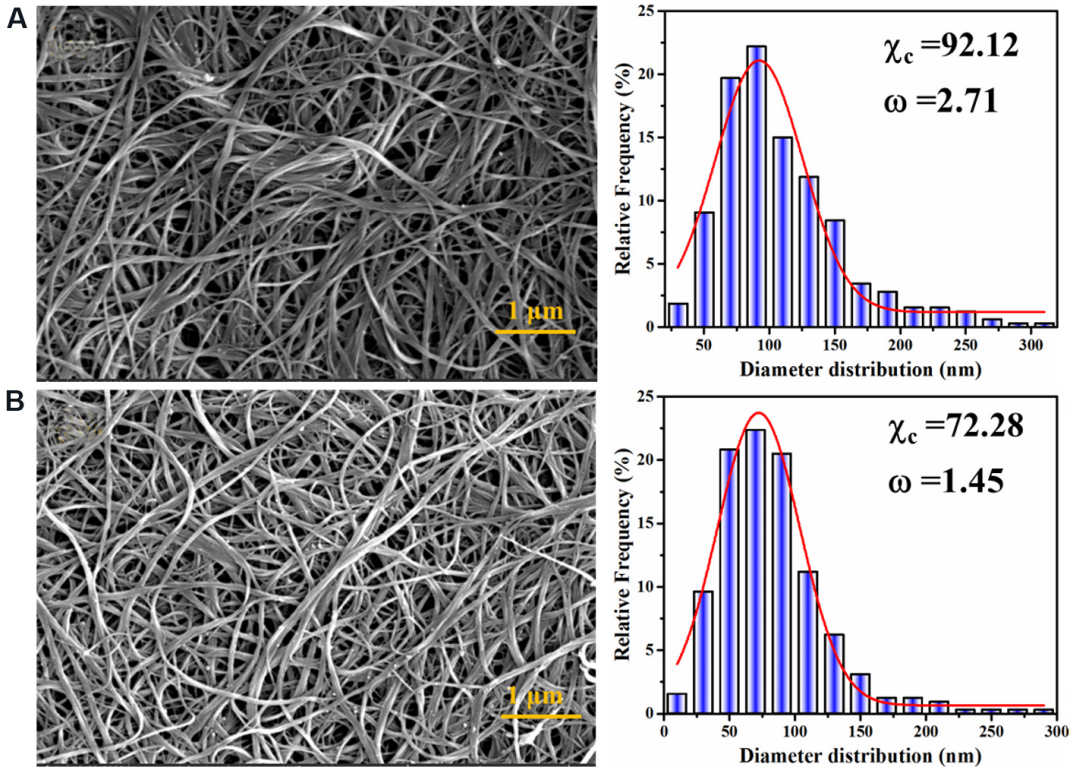

Figure 2. Scanning electronic microscopy images of (A) pure SWCNTs and (B) SWCNT-IL. Statistical distributions of the SWCNT bundle diameters are presented on the right. SWCNTs: Single-walled carbon nanotubes; IL: ionic liquid.

the form of bundles were observed in both samples. The diameters of the SWCNT bundles were measured at more than 300 diverse sites and the corresponding statistical histograms are shown at the right side of each SEM image. The mean values (x_c) and standard deviations (ω) of the SWCNT bundles are used to characterize the diameter distributions in the histograms, which are fitted by a Gaussian distribution[28]:

$$y = y_0 + Ae^{-(x-x_c)^2/2\omega^2} \quad (1)$$

where x is the SWCNT bundle diameter (nm) and the parameters y, y_0, x_c, ω and A represent the count, offset, center position of the distribution fitting peak, width of the distribution and amplitude of the distribution, respectively. Apparently, the SWCNT film without IL has a higher x_c and there are more SWCNT bundles with larger diameters [Figure 2A]. After the IL is added, x_c decreases [Figure 2B], suggesting that the SWCNT dispersion is enhanced with the IL[27].

TE performance of composites with IL at high temperature

Figure 3 shows the TGA curve of the PEDOT:PSS-IL film, which indicates that the film can maintain its thermal stability up to 548 K. There is some mass loss at ~350 K due to the evaporation of water molecules in the samples. After that, the sample shows a significant mass loss at ~548 K because the PSS components start to degrade[29]. When the temperature increases to 773 K, a further significant breakage of the PEDOT skeleton occurs, leading to another dramatic mass loss[30]. Thus, it is believed that PEDOT:PSS possesses excellent thermal stability under 470 K (the maximum testing temperature for TE and mechanical performances), below which the PEDOT:PSS will not experience any molecular degradation.

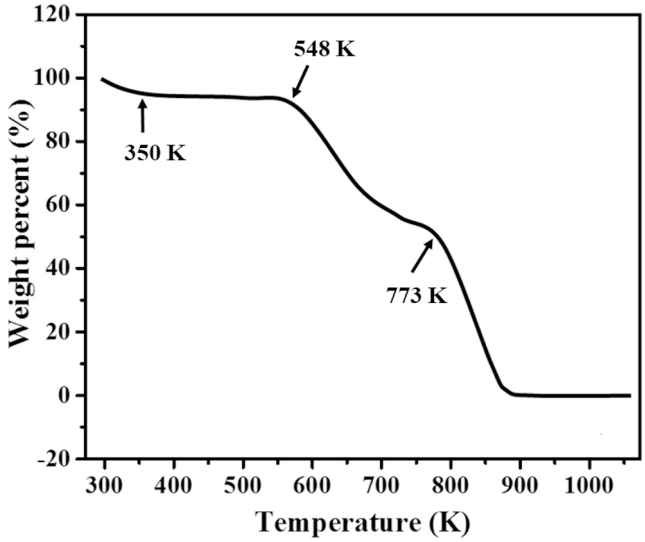

Figure 3. Thermogravimetric analysis curve of PEDOT:PSS-IL film. PEDOT:PSS: Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate); IL: ionic liquid.

Figure 4A displays the influence of temperature on the electrical conductivity for the composites with and without IL. It can be found that the electrical conductivity is dramatically enhanced at the same SWCNT content (30 or 60 wt.%) after the addition of IL additives. The IL additive interacts with PEDOT:PSS through an ion exchange effect, resulting in the transformation of the PEDOT conformation to a more conductive linear conformation and hence an improvement in the electrical conductivity[31-33]. Furthermore, the electrical conductivity decreases with increasing temperature from 293 to 470 K, exhibiting typical metal behavior[34]. In contrast, the Seebeck coefficients of the composites with and without IL increase distinctly with increasing temperature between 293 and 470 K [Figure 4B]. The addition of IL does not significantly affect the Seebeck coefficient if the experimental errors are considered. The variations of the electrical conductivity and the Seebeck coefficient are verified by the change in carrier concentration and mobility in Figure 4C and D. Compared to the IL-free composite (SWCNT content of 30 or 60 wt.%), both the carrier concentration and mobility are significantly improved for the composite with IL, which accounts for the dramatic increase in electrical conductivity after the addition of IL additives [Figure 4A]. In contrast, the Seebeck coefficient is negatively correlated with the carrier concentration and positively correlated with the carrier mobility[24,35]. The simultaneous carrier concentration and carrier mobility lead to a very slight variation in the Seebeck coefficient for the composites with and without IL [Figure 4B].

The *PF* values in Figure 4E show a positive temperature dependence for the composites with and without IL [Figure 4B]. The *PF* values of the composites with IL are always higher than the IL-free ones and the maximum *PF* value of 301.35 μW $m^{-1} K^{-2}$ is reached for PEDOT:PSS/SWCNT (60 wt.%)-IL composites at 470 K [Figure 4E]. The carrier transport mechanism is analyzed by the VRH model:

$$\sigma = \sigma_0 \exp\left[-\left(\frac{T_0}{T}\right)\alpha\right] \quad (2)$$

where σ_0 represents the electrical conductivity at an infinite temperature (0 K), T_0 is the characteristic temperature, T is the experimental temperature and the parameter α is related to dimensionality via the expression of $\alpha = 1/(1+D)$[36,37]. The carrier transport mode can be judged by the α values. Specifically, values of 1/2 and 1/4 correspond to one- and three-dimensional transport, respectively. The VRH model describes

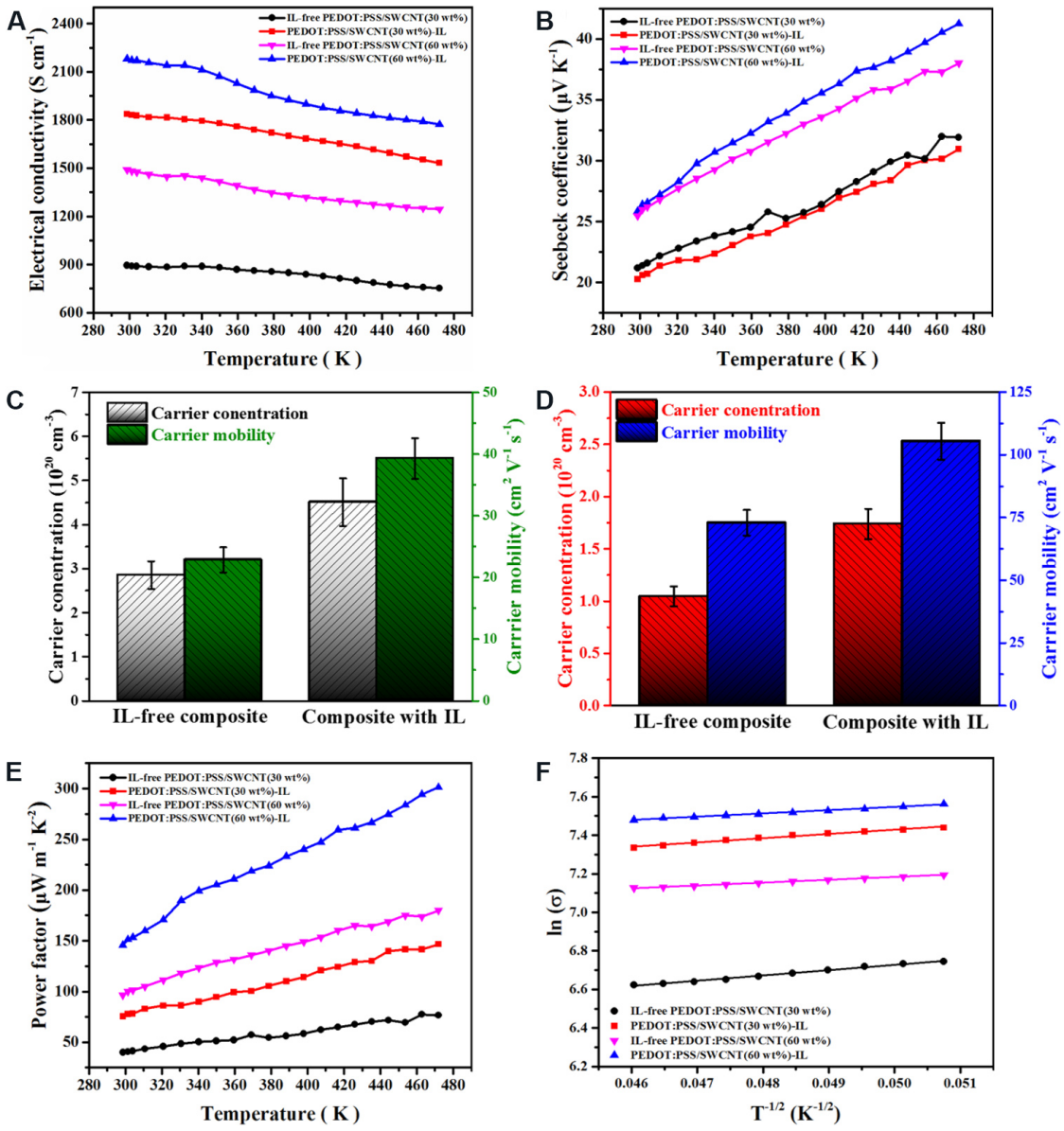

Figure 4. (A, B, E) TE parameters for the composites with and without IL containing SWCNT contents of 30 and 60 wt.%. Comparison of carrier concentration and mobility of composites with and without IL at SWCNT contents of (C) 30 and (D) 60 wt.%. (F) Dependence of lnσ on $T^{-1/2}$ for the composite with and without IL. TE: Thermoelectric; IL: ionic liquid; SWCNT: single-walled carbon nanotube.

the transport behavior of carriers in disordered systems in which the carrier hops from one localized site to another rather than the whole region[38,39]. In the present work, lnσ shows an obvious linear relationship with $T^{-1/2}$ for both composites at high temperatures in Figure 4F, suggesting that the carrier transport route of the PEDOT:PSS/SWCNT films is one-dimensional.

Morphology and flexibility of composites with IL

Figure 5 compares the SEM images of the PEDOT:PSS/SWCNT-IL composites at room temperature and after the 470 K test. It can be seen that the SWCNT bundles are coated with PEDOT:PSS at room temperature [Figure 5A and C]. After the TE test at 470 K, some PEDOT:PSS components partially melted

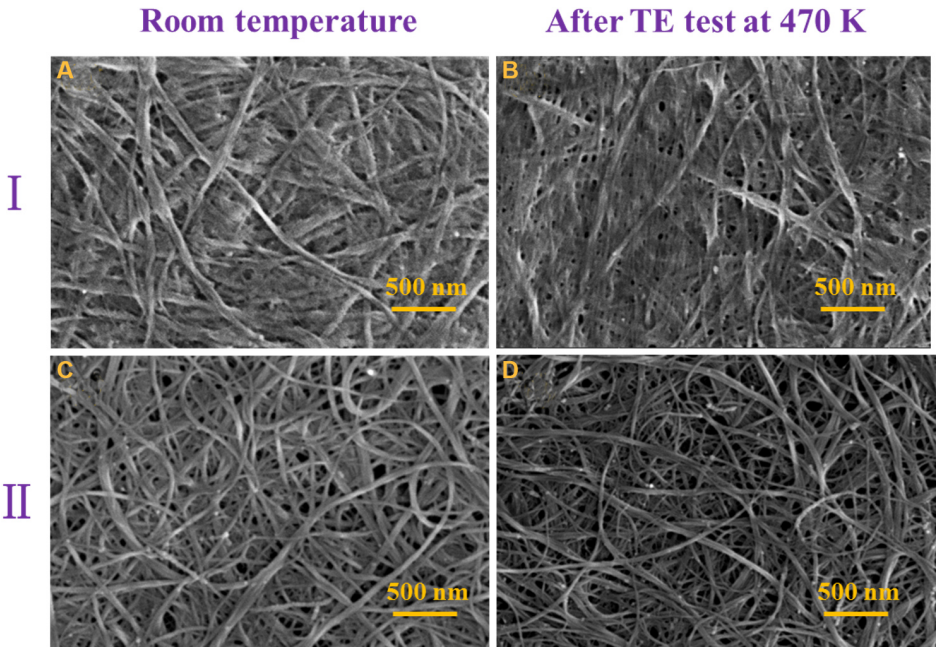

Figure 5. Typical scanning electronic microscopy images of PEDOT:PSS/SWCNT-IL composites with a SWCNT content of 30 (Row I) or 60 wt.% (Row II) after TE test at (A, C) room temperature and (B, D) after TE test at 470 K. PEDOT:PSS/SWCNT: Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/single-walled carbon nanotube; IL: ionic liquid.

and covered the surfaces of the SWCNT bundles, leading to a denser conductive network [Figure 5B and D]. Such a structure may be beneficial to the enhancement of the TE performance at high temperature.

Figure 6 presents optical photographs of the PEDOT:PSS/SWCNT-IL composite film (SWCNT content of 60%) at room temperature and after the high-temperature test. Evidently, the PEDOT:PSS/SWCNT-IL film can be easily bent and even wrapped around a rod with a radius of 1.5 mm without any obvious breakage [Figure 6A], indicating its high flexibility. This is consistent with our previous work[27]. After the high-temperature test (393 or 470 K), the PEDOT:PSS/SWCNT-IL composite still maintained its high flexibility [Figure 6B and C]. This characteristic is significant for the application of TE composites at high temperatures[40,41].

Mechanical properties of flexible composite films

Figure 7A-C shows the tensile-strain curves of the IL-free PEDOT:PSS/SWCNT and PEDOT:PSS/SWCNT-IL composites at room temperature, 393 K and 470 K. The samples at 393 and 470 K are represented by sample@393 K and sample@470 K, respectively. At room temperature [Figure 7A], the PEDOT:PSS/SWCNT-IL composite presents a steady enhancement in the elongation at break compared to IL-free PEDOT:PSS/SWCNT composite. Specifically, the elongation at break increases to 5.0% for the PEDOT:PSS/SWCNT-IL composites (30 wt.% SWCNTs) from 1.6% for the IL-free PEDOT:PSS/SWCNT composite (30 wt.% SWCNTs). A similar increase in the elongation at break is also obtained for the PEDOT:PSS/SWCNT-IL composites (60 wt.% SWCNTs). This is possibly due to the formation of a fibrillar PEDOT structure stemming from the modulation of the IL[42-44]. The role of the IL that can increase the elongation at break also can be demonstrated for the composites at 393 K [Figure 7B] and 470 K [Figure 7C]. Figure 7D compares the influence of temperature on the elongation at break of the composites with and without IL. These results suggest that increasing temperature can enhance the elongation at break

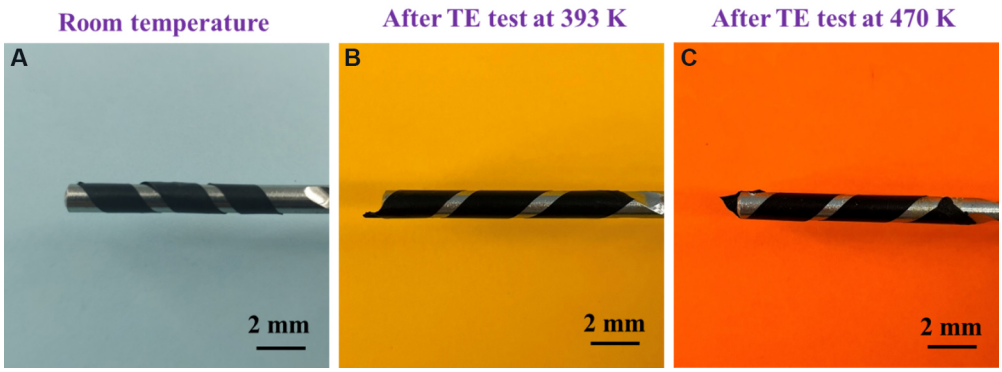

Figure 6. Optical photographs of PEDOT:PSS/SWCNT-IL composite film with 60 wt.% SWCNTs after TE test at (A) room temperature, (B) 393 K and (C) 470 K. PEDOT:PSS/SWCNT: Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/single-walled carbon nanotube; IL: ionic liquid; TE: thermoelectric.

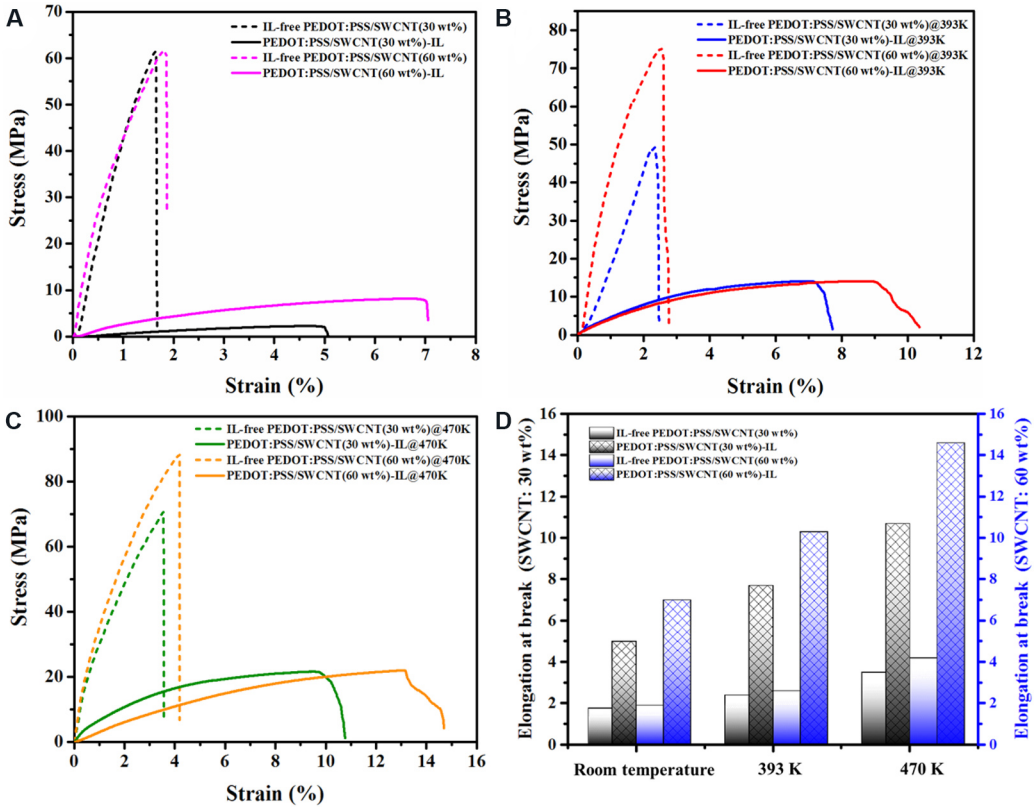

Figure 7. Stress-strain curves of PEDOT:PSS/SWCNT composite films with different SWCNT and IL contents at (A) room temperature, (B) 393 K and (C) 470 K. (D) Comparison of elongation at break of composites with and without IL. PEDOT:PSS/SWCNT: Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/single-walled carbon nanotube; IL: ionic liquid.

of the composites, which may result from the accelerating relaxation of polymer chains at high temperature[45]. Accordingly, the elongations at break can be improved by the addition of IL and the increasing temperature.

CONCLUSIONS

In summary, we have reported the TE and mechanical properties of flexible IL-free PEDOT:PSS/SWCNT and PEDOT:PSS/SWCNT-IL composite films at different SWCNT contents and temperatures. The PEDOT:PSS/SWCNT-IL composites show higher TE performance than that of their IL-free counterparts with the same SWCNT contents at room and high temperatures. Both composites show increasing trends of *PF* values with increasing temperature. The maximum *PF* value of 301.35 μW m^{-1} K^{-2} is obtained for the PEDOT:PSS/SWCNT-IL composite at 470 K. Furthermore, the addition of IL can improve the elongation at break of the composites compared to the IL-free composites. This work clarifies the influence of IL and temperature on the TE and mechanical properties of PEDOT:PSS-based composites and thus helps widen their applications at different temperature ranges.

DECLARATIONS

Authors' contributions

Experiments, data analysis, image editing, writing a draft: Deng W
Data discussion: Deng L
Data analysis: Hu Y
Reviewing and editing, supervision, funding acquisition: Zhang Y, Chen G
All authors have given approval to the final version of the manuscript.

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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