

Enhance the figure of merit for flexible thermoelectric materials by reducing the screening effect

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Abstract A flexible thermoelectric materials (FTEM) films based on Polydimethylsiloxane as the polymer matrix and (Zinc Oxide, Polyaniline and multi wall Carbon nanotube) as the fillers were prepared. The fillers were prepared using modified sol–gel method to enhance the distribution of the nanoparticles inside the polymer matrix and good performance. The Seebeck coefficient, electrical conductivity, thermal conductivity and dielectric measurements were carried out for all of the four films. The Seebeck coefficient results for the investigated films were $-40, 220, -120, 280 \mu\text{V}/\text{k}^\circ$ at 200°C for FTEM 1, 2, 3, 4 alternatively. The sign and the value for the Seebeck measurements show a great dependent on the filler type add to the film. The electrical conductivity results for the films was varied from 2×10^{-8} to 0.14 S/m and strongly dependent on the fillers electrical conductivity. The less dependent on the thermal conductivity of the fillers and a slight change in the thermal conductivity value of the films were obtained. The figure of merit for the films was ranged from 1.14×10^{-16} to 3.134×10^{-8} . The dielectric results ranged from 4 to 3500. Reducing the screening effect will increase the electrical conductivity and Seebeck coefficient values.

Keywords Flexible thermoelectric nanogenerator · PDMS · Seebeck coefficient · Thermal conductivity

Introduction

Energy harvesting converts different types of energy into electric energy. These energies can be found everywhere in nature such as mechanical, light and thermal energies all of these energies can be converted to electricity via different mechanisms (Hicks et al. 1996; Candolfi et al. 2011; Vaqueiro and Powell 2010).

Thermal energy is one of the most popular energy sources for generating electricity and can be considered as the most used method for generating electricity. Different method used to convert thermal energy to electricity and they can be divided into two methods. The first one is the conversion of thermal energy into mechanical energy then mechanical energy convert to electrical energy for example, burning fuel in electric power station and nuclear reactor. The second one is the direct generating of electricity from thermal energy via Seebeck effect; this method can be used to directly convert thermal energy in our bodies, in different layer of the earth (geothermal) and in some radioisotope materials into direct electricity. The conversion efficiency of thermal to electrical energy in Seebeck effect depends on the figure of merit (ZT). Figure of merit depend on three parameters (electrical conductivity, thermal conductivity and Seebeck coefficient). The problem in developing a new material that has high figure of merit (i.e., high conversion efficiency) is the direct dependence of these parameters on each other. Last decades scientist found a suitable way to enhance one or two of these parameters independently on the other parameters using quantum effect that been formed in different shapes of nano materials (Hsu et al. 2004; Sales 2007; Pichanusakorn and Bandaru 2010; Gibson 2010; Liang et al. 2011).

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The screening effect reduces the electrostatic field and Coulomb potential of an ion in semiconductor or metal material by electron clouds. The formation of the built-in electric field near the p–n junction boundary will effectively deplete excess electrons in n-type material and excess holes in p-type material; this will result in reducing the screening effect and increase the electric field. Different researches have been done to enhance the output power of piezoelectric generator by utilizing the reduction of screening effect between p-type and n-type materials (Shin et al. 2013; Lei et al. 2015). Other research in enhance piezoelectric nanogenerator output power was done by incorporating high electrical conductivity material like MWCNT (Sun et al. 2013) and PANI as an alternative to MWCNT (Sultana et al. 2015) to make electrical bridge in the polymer matrix. On the other hand, the modification of thermoelectric output power was focused on making thermoelectric paste material consist of p-type material and polymer matrix (Andrei et al. 2014) or adding conductive filler to the thermoelectric paste to enhance the figure of merit by increasing the electrical conductivity (Andrei et al. 2016; Luo et al. 2016). The MWCNT filler was investigated in many researches. The thermoelectric performance of functionalized and non-functionalized MWCNT polymer composite shows little difference (Liebscher et al. 2014). Other research investigated the thermoelectric performance of PANI with MWCNT which show a great enhancement in figure of merit (Chatterjee et al. 2016).

In this work, a new method was investigated to enhance the figure of merit by increasing both Seebeck coefficient and electrical conductivity through the reduction of the screening effect. The polymer matrix shows an important role in maintaining low thermal conductivity for the films and also gives the films a flexible feature.

Experimental section

Materials used in the preparation of the FTEM films are: sol–gel synthesized ZnO, PANI from Panichem Company, MWCNT from cheap tube Company, tetrahydrofuran (THF) and PDMS Slygard 184 from Dow Corning Company.

Preparation of the ZnO nanoparticle

Materials used in the preparation of the ZnO are: Zinc nitrate hexahydrate, Cetrimonium bromide, Lithium hydroxide monohydrate, and ethanol.

First dissolve the Zinc Nitrate into 100 ml ethanol to get a molarity of (0.86 M) then add to the solution 0.08 g of

Cetrimonium bromide to help prevent the formation of agglomeration when add the lithium hydroxide solution. Then dissolve the lithium hydroxide into 100 ml ethanol at 0 °C to get (1.7 M) solution, and then dropwise add the lithium hydroxide solution into the Zinc Nitrate solution with continuous stirring at 60 °C for 1 h. After filtering white precipitate powder was obtained and dried in the furnace for about 4 h at 500 °C, after that washed, filtered and dried the powder to get the final ZnO powder.

Functionalization of the MWCNT

Materials used in the MWCNT functionalization process are: MWCNT, Sulfuric acid (H₂SO₄), Hydrochloric acid (HCl), Nitric acid (HNO₃), Sodium hydroxide (NaOH) and water.

First mix 25 ml of HNO₃ 68% concentration with 75 ml of H₂SO₄ 98% concentration then added 1 g of MWCNT and mix the solution for 1 h using ultrasonic device and then leave the mixture for 19 h then after that add 40 ml of concentrated HCl to the solution and mix using ultrasonic for 1 h then add 120 ml of (4 M) NaOH solution to the solution above to get a pH of about 7 then filter the solution using the vacuum pump and wash the powder with distilled water then dry the solution at 80 °C for 2 h.

Preparation of the FTEM films

FTEM 1 was prepared by dispersion of 10% wt of ZnO into 5 ml of THF and then mixed with the solution using ultrasonic device. A solution of 90% wt of PDMS and 5 ml of THF was mixed ultrasonically then add the PDMS solution to ZnO solution with mechanically stirring for 1 h at 70 °C to get a uniform solution then let the mixture cooled to room temperature and left it for 1 day to complete the evaporation of the THF. After evaporation of the THF, the PDMS composite was mixed in wt ratio 16:1 with the curing agent with continuous stirring. After that the composite was drop casted on glass substrate and set to 70 °C for 2 h to cure. Then the film was annealed at 140 °C for 4 h.

FTEM 2 was prepared by dispersion of 10% wt of ZnO and 20% wt of PANI into 5 ml of THF and then mixed the solution using ultrasonic device. A solution of 70% wt of PDMS and 5 ml of THF was mixed ultrasonically then add the PDMS solution to ZnO solution with mechanically stirring for 1 h at 70 °C to get a uniform solution then let the mixture cooled to room temperature and left it for 1 day to complete the evaporation of the THF. After evaporation of the THF, the PDMS composite was mixed in wt ratio 16:1 with the curing agent with continuous

stirring. After that the composite was drop casted on glass substrate and set to 70 °C for 2 h to cure. Then the film was annealed at 70 °C for 4 h.

FTEM 3 was prepared by dispersion of 10% wt of ZnO, 0.8% wt F-MWCNT and 0.8% wt MWCNT into 5 ml of THF and then mixed the solution using ultrasonic device. A solution of 88.4% wt of PDMS and 5 ml of THF was mixed ultrasonically then added the PDMS solution to ZnO solution with mechanically stirring for 1 h at 70 °C to get a uniform solution then let the mixture cooled to room temperature and left it for 1 day to complete the evaporation of the THF. After evaporation of the THF, the PDMS composite was mixed in wt ratio 16:1 with the curing agent with continuous stirring. After that the composite was drop casted on glass substrate and set to 70 °C for 2 h to cure. Then the film was annealed at 140 °C for 4 h.

FTEM 4 was prepared by dispersion of 10% wt of ZnO, 20% wt of PANI, 0.8% wt F-MWCNT and 0.8% wt MWCNT into 5 ml of THF and then mixed the solution using ultrasonic device. A solution of 68.4% wt of PDMS and 5 ml of THF was mixed ultrasonically then added the PDMS solution to ZnO solution with mechanically stirring for 1 h at 70 °C to get a uniform solution then let the mixture cooled to room temperature and left it for 1 day to complete the evaporation of the THF. After evaporation of the THF, the PDMS composite was mixed in wt ratio 16:1 with the curing agent with continuous stirring. After that the composite was drop casted on glass substrate and set to 70 °C for 2 h to cure. Then the film was annealed at 70 °C for 4 h. The digital photos for four prepared films are shown in Fig. 1.

Characterization

The crystal structure for the ZnO film was measured by XRD device (D/Max-RB Model). The electrical conductivity for the films was carried out with two-point electrical



Fig. 1 The digital photo of FTEM films

resistance method using digital multimeter (twintex TM-8155) as the resistance measurement unit and heat source, the measurements temperature was from room temperature 26 to 200 °C. The Seebeck coefficient measurement was carried out by homemade setup as shown in Fig. 2; which consists of digital multimeter (twintex TM-8155) as the voltage measurement unit, digital thermometer, AC dimmer circuit and 500 W heating element. The films dimension is (6 × 2.5) cm² and the copper tape electrode was attached to both end sides. The distance between the hot and cold side was set to 1.5 cm. The thermo voltage was recorded every 5 °C temperature difference between the hot and cold side and the temperature range was from room temperature to 200 °C. The Seebeck coefficient was determined from the formula below:

$$S = -\left(\frac{\Delta V}{\Delta T}\right) \quad (1)$$

where S is the Seebeck coefficient, ΔV is the thermo voltage and ΔT is the temperature difference between the two ends of a film. The highest values recorded for Seebeck coefficient and electrical conductivity was taken at 200 °C for all films, for this reason the values of other temperature are not recorded.

The thermal conductivity measurements for FTEM films was carried out using Lee's disk method, the films were put between two brass plate (plate a, plate b). The heat source for the experiment was the constant voltage and current of 6 V 0.25 A, respectively. The thermal conductivity equation is shown below:

$$\begin{aligned} H &= I \cdot V \\ &= \pi r^2 e (T_A + T_B) \\ &\quad + 2\pi r e \left(d_A T_A + d_S \frac{1}{2} (T_A + T_B) + d_B T_B + d_C T_C \right) \end{aligned} \quad (2)$$

where H is the average time of the applied energy to heat the coil, (T_A , T_B , T_C) is the (A , B , C) disk's temperature, respectively, (d_A , d_B , d_C) is the (A , B , C) disk's thickness, respectively, d_S is the Sample S thickness, r is the disk diameter, I is the current flow, V is the supply voltage

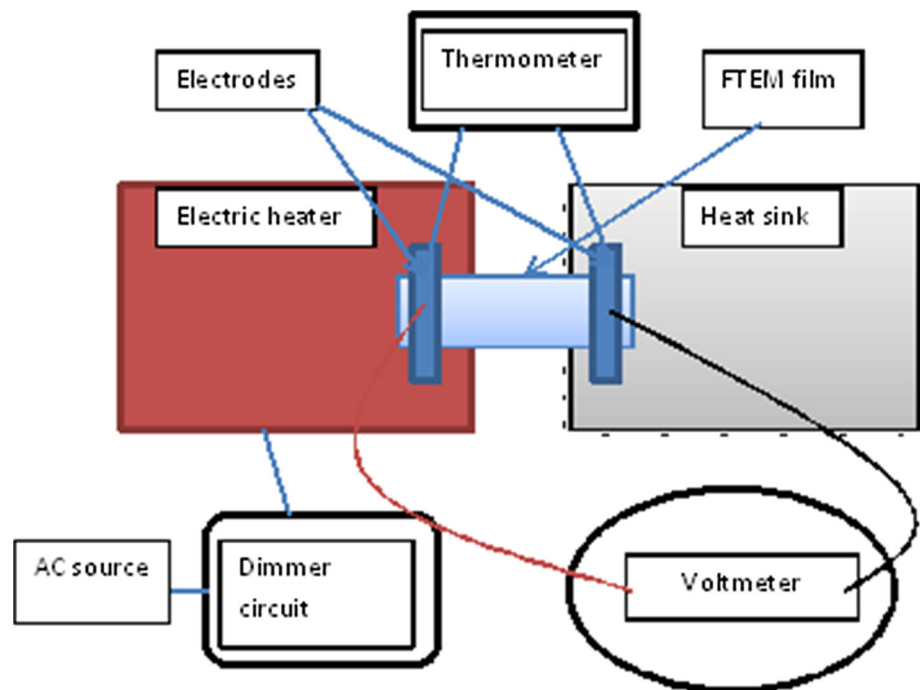
$$K \left(\frac{T_B - T_A}{d_S} \right) = e \left(T_A + \frac{2}{r} \left(d_A + \frac{d_S}{4} \right) T_A + \frac{1}{2r} d_S T_B \right) \quad (3)$$

where e is the amount of the thermal energy flows over disk' cross section for every second and K is the thermal conductivity.

The dielectric constant measurement for the FTEM films was carried out by LCR meter using (Gw-insteck LCR-8105G) device at room temperature.

The dielectric constant for the FTNG films was evaluated from equation below:

Fig. 2 Seebeck coefficient measurement schematic



$$C = \frac{\epsilon_0 \epsilon_r}{d} \cdot A \quad (4)$$

where ϵ_0 is the dielectric constant of the free space, A is the area of the electrical conductor, d is the thickness of the dielectric layer, and ϵ_r is the dielectric constant.

Result and discussion

XRD

Figure 3 shows the ZnO nanoparticle have a polycrystalline structure and the characteristic peaks of the ZnO occur at $2\theta = (31.8596, 34.5275, 36.3581, 47.6471, 56.7023, 62.9798, 66.4780, 68.0460, 69.1904)$ which correspond to the miller indices (100, 002, 101, 102, 110, 103, 200, 112, 201), respectively. All the observed diffraction peaks are indicating the formation of hexagonal phase of

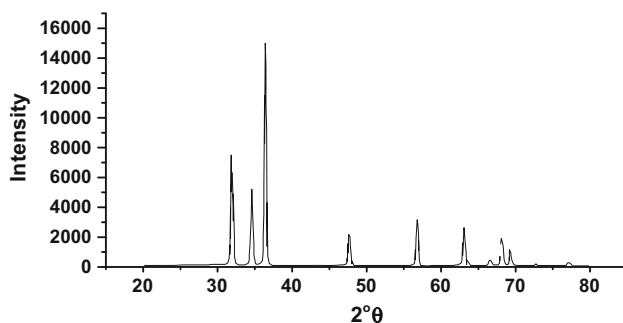


Fig. 3 XRD of the ZnO nanoparticle

the ZnO. There are no peaks of impurities observed, which means that the ZnO nanoparticle with high purity was obtained (Kolekar et al. 2011). The average crystallite size of the ZnO was calculated using Scherrer equation. The average crystallite size of the ZnO nanoparticle D was found to be 19.5 nm.

Electrical conductivity

Figure 4 shows the electrical conductivity of the four FTEM films at 200 °C. The result show large difference in electrical conductivity between the FTEM films; the reason could be related to the conductivity of the filler added to the film. The reason for increasing the electrical conductivity of the FTEM films can be assigned to hopping and tunneling effect (Khanam et al. 2015), also the high concentration of the fillers inside the insulating matrix could lead to physical interaction between the adjacent particles thus increase the electrical conductivity.

Thermal conductivity

Figure 5 shows the thermal conductivity of the four FTEM films. The thermal conductivity results show slight increases in value between the FTEM films; although the fillers (ZnO, MWCNT) have high thermal conductivity. This small influence of the high thermal conductivity filler on the overall thermal conductivity of the polymer Nano composites can be assigned to the fact that the thermal conductivity of the polymer nano composites is highly

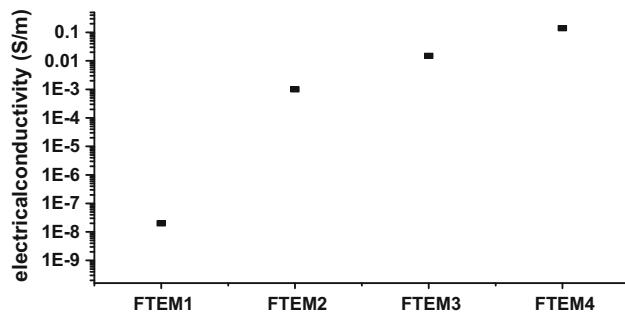


Fig. 4 The electrical conductivity of the FTEM films at 200 °C

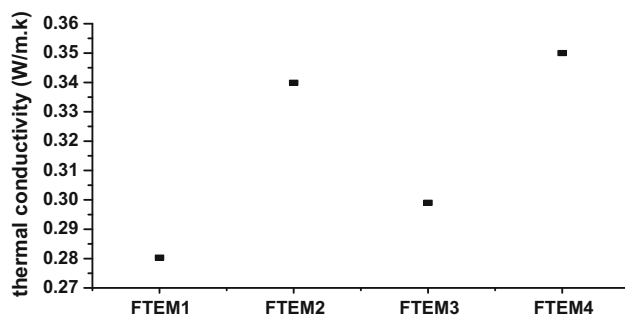


Fig. 5 Thermal conductivity of the FTEM films

affected by phonon scattering with polymer chains (Choi et al. 2011). The agglomeration of MWCNT filler in the FTEM films due to the fact that CNT are self-agglomerated this could lead to dissipation of heat by randomly formed phonon trajectories (Hong et al. 2010). Lastly for the FTEM films that contain the PANI filler the thermal conductivity slight change due to low thermal conductivity PANI filler.

Seebeck coefficient

Table 1 shows the Seebeck coefficient for the four FTEM films at 200 °C. FTEM 1 and 3 films Seebeck coefficient sign show negative sign; this may be due to the dominance of electrons carrier result from higher concentration of ZnO filler inside the polymer matrix. The FTEM 2 and 4 films Seebeck coefficient sign show positive sign; this may be due to the dominance of holes carrier result from higher concentration of the PANI filler inside the polymer matrix. FTEM 1 film shows lower Seebeck coefficient compare to the ZnO bulk or nanoparticle; The reason for this can be assigned to the restricted movement of the charge carrier inside the insulating polymer matrix that increase the losses of the thermally energized charge carrier. The FTEM 2 film shows a large increase in the Seebeck coefficient; this can be assigned to the decrease of the electrical resistivity of the FTEM films by the high conductivity of the PANI filler (Luo et al. 2016). Another reason for increasing Seebeck

Table 1 The value of Seebeck coefficient for the FTEM films at 200 °C

Film	Seebeck coefficient S ($\mu\text{V}/\text{k}^\circ$)
FTEM 1	−40
FTEM 2	220
FTEM 3	−120
FTEM 4	280

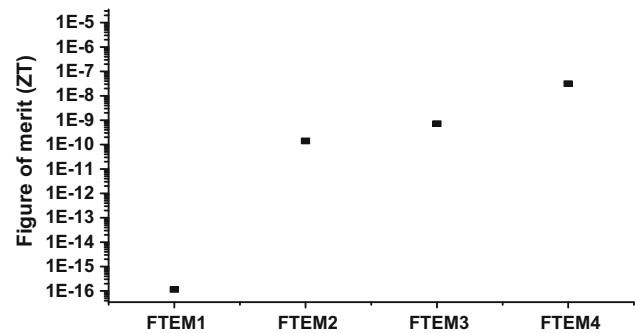


Fig. 6 Figure of merit for the FTEM films

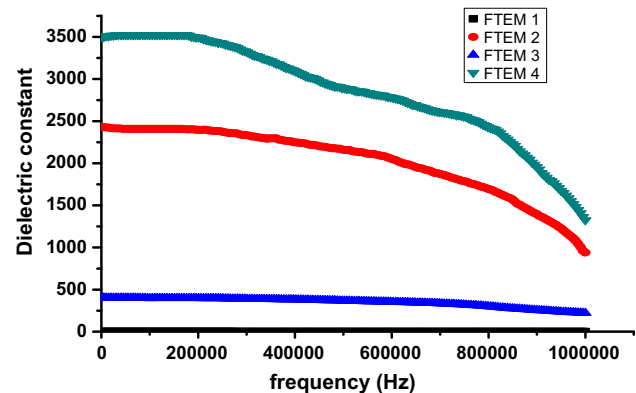


Fig. 7 Dielectric constant of the FTEM films

coefficient of FTEM 2 film can be subjected to reduce the screening effect for PANI filler (Lei et al. 2015). The FTEM 3 film shows a moderate enhancement in the Seebeck coefficient; this can be assigned to the high electrical conductivity of MWCNT filler. The high electrical conductivity will form an electrical network inside the polymer; this will give the charge carrier more chance to find an easy path to travel from hot to cold side thus increase the Seebeck coefficient.

Finally, FTEM 4 film shows the highest Seebeck coefficient value; the reason can be subjected to decrease of excess carrier concentration by the screening effect and enhance the flow of charge carrier.

The higher the capacitance (dielectric constant) will tend to increase the repulsive electrostatic force build up

from charges formed at the cold side thus increase number of difference in charged carrier between hot and cold side [i.e., electric field (Seebeck coefficient)]. This will lead to push larger number of charges back to the hot side (Yu and Chau 2009; Kittel and Kroemer 1980).

Figure of merit

Figure 6 shows the figure of merit for the four FTEM films. The value of the figure of merit show great enhancement by adding of conductive filler to the films as shown in FTEM 1, 2, 3, 4 films. The highest ZT value achieved is 3.134×10^{-8} . On the other hand, the decrease of the screening effect leads to great enhancement in the ZT value by order of 8 which can be considered new way to develop a high ZT value composite.

Dielectric constant

Figure 7 show the dielectric constant of FTEM films with frequency. In the low frequency region the dielectric constants have the maximum value and at high frequency region the value of the dielectric is the lowest. This behaviour can be explained by the relaxation time theory (Dutta et al. 2002) that states at the low frequency region there is enough time for the dipole to align themselves with the electric field before field goes opposite direction. For the high frequency region the decrease was due to the dipoles cannot align themselves probably as to keep tracking the change in electric field frequency.

The FTEM 1 film shows an increase in the value of the dielectric constant; this may attribute to formation of interfacial polarization between ZnO and the polymer and also the increase could be attributed to the higher dielectric constant of the ZnO compare to PDMS.

The FTEM 2 film shows a sharp increase in the dielectric constant, this may be subjected to the formation of mini capacitor inside the polymer matrix. The sharp increase may also attribute to the formation of p–n junction (between ZnO and PANI inside PDMS matrix) that will increase the dielectric constant by reducing screening effect (Ho et al. 2008).

The FTEM 3 film shows a great enhancement of the dielectric constant; this result can be assigned to the formation of mini capacitor by the MWCNT and the polymer matrix or the ZnO filler. Under the applied electric field the charge are accumulated on the nanotube where the MWCNT serve as a secondary electrode (Poh et al. 2016).

Finally the FTEM 4 film show a large enhancement in the dielectric constant, the reason also can be assigned to the increase in the number of the mini capacitor formed inside the polymer and the interfacial polarization of the filler–filler interaction and the polymer matrix–filler interaction.

Conclusion

Simple sol–gel method is used to synthesis of flexible thermoelectric material films consist of cheap and non-toxic materials. The thermoelectric and dielectric properties for the films were done. The influence of the screening effect on the thermoelectric parameter Seebeck coefficient and electrical conductivity was investigated for all the films. The Seebeck coefficient value and sign of the films show a great dependent on the filler type; the highest value reaches $280 \mu\text{V}/\text{K}^\circ$. The electrical conductivity of the films enhances dramatically with conductive fillers; the highest value achieved is 0.14 S/m ; on the other hand, the thermal conductivity is less influenced by fillers and maintains low thermal conductivity value of less than 0.5 W/mK° . The figure of merit also shows a dramatical enhancement though it still has very low value of about 3.134×10^{-8} ; the overall enhancement was very sharp. The dielectric results of the films show great enhancement by adding conductive filler to them due to formation of mini capacitor. On the other hand, it was suggested that another reason for increase in the Seebeck coefficient is the high capacitance of the FTEM films.

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