



# Increase in thermoelectric power factor of carbon-nanotube films after addition of polystyrene



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## ABSTRACT

We report the enhancement of the thermoelectric power factors of single-walled carbon-nanotube (SWCNT) films caused by the addition of polystyrene to the films as a binder. The Seebeck coefficient of the SWCNT films was increased by addition of polystyrene. On the other hand, the electrical conductivity gradually decreased with increasing amount of polystyrene. The power factor was maximum for a polystyrene concentration of 20 wt%; it was approximately 1.7 times higher than that of a pure SWCNT film. These results indicate that polystyrene is a superior binder polymer for synthesizing CNT/polymer thermoelectric composites.

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## 1. Introduction

Thermoelectric devices [1,2] have attracted considerable interest because of the increasing demand for energy harvesting from the ubiquitous waste heat in living areas. Most heat sources have curved surfaces; to place thermoelectric devices on such curved heat sources, it is essential that the devices be flexible. Thus, various flexible thermoelectric materials and devices have been researched extensively [3–15]. Composite materials consisting of carbon nanotubes (CNTs) and polymers [3,5,6,9] (i.e., CNT/polymer composite) are promising flexible thermoelectric materials because CNTs have a very high electrical conductivity [16] and exhibit a relatively high Seebeck coefficient in flexible materials [17]. At the same time, the polymer mixed with the CNTs acts as a binder, heightening mechanical flexibility and formability to the resulting composite material.

Considerable research efforts have been devoted to improving the thermoelectric performance of CNT/polymer composites by controlling the characteristics of CNTs and the binder polymer used [3,5,6,9]. Various types of polymers can be used for synthesizing CNT/polymer composites. In order to realize a high-performance composite, a polymer species that can fully bring out the superior thermoelectric properties of CNTs must be chosen. Currently,

conductive polymers, such as polythiophene derivatives [5], are being used actively for synthesizing CNT/polymer composites because conductive polymers result in composites with high electrical conductivities, which lead to high power factors (PF). On the other hand, we previously reported on a CNT/polystyrene composite with a PF of  $413 \mu\text{W}/\text{K}^2 \text{ m}$  [18], which is higher than that previously observed for many CNT/conductive polymer composites [5,6] or pristine CNTs [4,8]. Since polystyrene is a common insulating polymer, this high PF suggests that the use of an insulating binder, such as polystyrene, is a more effective way of bringing out the superior thermoelectric properties of CNT-dispersion materials. However, it was not revealed in the previous papers that why the CNT composites with polystyrene binders show such high power factors.

In this paper, the influence of polystyrene concentration on the morphological and electrical properties of the CNT/polystyrene composites was investigated, and a discussion on the mechanism that brings about the improved PF by the complexing of CNTs with polystyrene was presented. We found that the Seebeck coefficients was increased by using polystyrene binder. On the other hand, the electrical conductivity monotonically decreased with increasing amount of polystyrene. A maximum PF value was obtained at a polystyrene concentration of 20 wt%, and it was approximately 1.7 times higher than that of the pure CNT film. This increase in the Seebeck coefficient may be caused by the enhancement of energy filtering effects at the CNT/CNT contact points in the presence of polystyrene.

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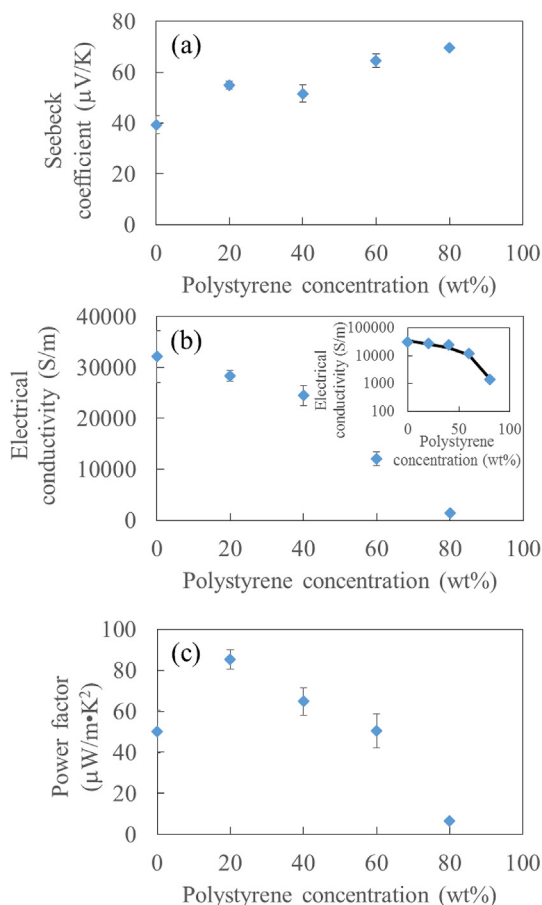
## 2. Experimental

We used single-walled CNTs (SWCNT) with an average diameter of 1.4 nm, and polystyrene (molecular weight of ~280,000). The SWCNTs and polystyrene were mixed through planetary ball milling at 600 rpm in 1, 2, 4-trichlorobenzene as a solvent. SWCNT/polystyrene composite films were fabricated by drop casting the mixed solution onto a glass substrate at a temperature of 50 °C. The Seebeck coefficients and electrical conductivities of the synthesized SWCNT/polystyrene composite films were measured along the in-plane direction by using a ZEM-3 system (ADVANCE RIKO, Inc.) The measurement temperature was 50 °C, and the measurements were performed in a low-pressure nitrogen atmosphere. Scanning electron microscopy (SEM) images of the films were obtained using a field-emission SEM system (S-4800, Hitachi High-Tech Science Corp.) To obtain clear SEM images, a Pt layer with a thickness of approximately 6.5 nm was coated on the sample films by using an ion-sputtering system (E-1030, Hitachi, Inc.).

## 3. Results

### 3.1. Thermoelectric characteristics of SWCNT/polystyrene composites

Fig. 1a,b and c show the values of the Seebeck coefficient, electrical conductivity, and power factor of the SWCNT/polystyrene



**Fig. 1.** (a) Seebeck coefficient, (b) Electrical conductivity, and (c) power factor of the synthesized SWCNT/polystyrene composites as functions of the polystyrene concentration. Inset of Fig. 1 (b) is a semi-logarithmic plot, where the solid line is a fit using Equation (2), and the values of  $\sigma_0$ ,  $V_0$ , and  $t$  were 95,000 S/m, 0.1, and 1.1, respectively.

composite films as functions of the polystyrene concentration. Interestingly, the Seebeck coefficient showed increasing tendency with the polystyrene concentration, indicating that the polystyrene causes an increase in the Seebeck coefficient of the SWCNT films, even though polystyrene is an insulating material.

The electrical conductivity decreased as the polystyrene concentration increased. However, the rate of decrease is low considering the volume ratio of SWCNTs and polystyrene. For instance, in the case of the composite with 40 wt% polystyrene, the volume ratio of SWCNTs:polystyrene is approximately 2:3. Although polystyrene occupied a 1.5-fold larger volume than SWCNTs did in the composite, the electrical conductivity of the composite with 40 wt% polystyrene was approximately 80% that of the film with a polystyrene concentration of 0 wt% (i.e., a film consisting only of SWCNTs). Here, for calculating the volume ratio, we assumed  $\rho_{PS}$  to be 1.04 g/cm<sup>3</sup> [19], while  $\rho_{CNT}$  (= 2.17 g/cm<sup>3</sup>) was calculated using the following equation [20].

$$\rho_{CNT} = \frac{4000}{1315} \left[ n/d - \left( 2ds \sum_{i=0}^{n-1} i \right) / d^2 \right], \quad (1)$$

where  $d$ ,  $n$ , and  $ds$  denote the diameter, number of walls, and intershell distance of the CNTs, respectively.

The composite films with a polystyrene concentration of 20–60 wt% showed higher power factors than those of pure SWCNT films. The power factor was maximum for a polystyrene concentration of 20 wt% and reached a value of 85  $\mu\text{W}/\text{m}^2\text{K}^2$ , which is approximately 1.7 times higher than that of the pure SWCNT film.

### 3.2. Microstructure of SWCNT/polystyrene composite films

Fig. 2 shows SEM images of SWCNT/polystyrene composite films with different polystyrene concentrations. In the case of the pure SWCNTs film, fiber-like structures (i.e., bundle of SWCNTs) and numerous voids several hundred nanometers in diameter were observed. The density of the pure SWCNT film was 1.49 g/cm<sup>2</sup>. By using this density value, the voids in the pure SWCNT film were calculated to occupy approximately 31 vol% of the film. The voids in the composites decreased in number with an increase in the polystyrene concentration, and they nearly disappeared for polystyrene concentrations greater than 80 wt% (Fig. 2), indicating that the spaces between the individual SWCNTs were filled by polystyrene.

## 4. Discussion

The power factors of the SWCNTs were improved by the addition of polystyrene as a binder polymer (Fig. 1c). This appears to be similar with many reports on the improvement of the carrier mobility of organic semiconductors owing to improvement in the organization and crystallinity in the materials, which is caused by mixing of the organic semiconductors with insulating polymers [21,22]. However, the origin of this improvement of power factor is mainly due to an increase in the Seebeck coefficient (Fig. 1a), which is a different phenomenon from the well-known mobility increase in organic semiconductors. In the SWCNT/polystyrene composites synthesized in the present study, charge carriers flow only in the SWCNTs, as the composites do not contain any other conductor except for SWCNTs. The electrical conductivity ( $\sigma$ ) of the SWCNT/polystyrene composites in this study can be fitted by Equation (2) (inset of Fig. 1b), which is derived from the percolation model [23].

$$\sigma = \sigma_0 (V - V_0)^t \quad (2)$$

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