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Enhanced thermoelectric properties of the flexible tellurium nanowire film hybridized with single-walled carbon nanotube

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ABSTRACT

Thermoelectrics is a challenging issue for future energy harvesting and cooling technology. We here have demonstrated a new system of the tellurium nanowire (TeNW) films hybridized with single-walled carbon nanotube (SWCNT) as a flexible thermoelectric material and investigated their thermoelectric properties as a function of SWCNT weight ratio in the hybrid. The excellent mechanical stability and electrical conductivity of SWCNT enhance the flexibility and thermoelectric properties of the pure TeNW film. The addition of 2 wt% SWCNT into TeNW matrix significantly increases the electrical conductivity from 4 to 50 S m^{-1} while maintaining the high thermopower, thereby leading to one order of magnitude higher figure of merit (ZT) compared to the pure TeNW film. These results indicate that the SWCNT/TeNW hybrid film would be promising for a potential use as a flexible thermoelectric material.

1. Introduction

Thermoelectric (TE) materials have been attracted because of their large potentials to both energy harvesting and electronic cooling devices [1–4]. The performance of TE material is evaluated by Figure of merit, given by

$$ZT = (S^2\sigma)T\kappa^{-1} \quad (1)$$

where S, σ, T, and κ are the thermopower, electrical conductivity, absolute temperature, and thermal conductivity, respectively. Thus, the high S and σ with low κ are desirable for high ZT. However, the design to get high ZT is not easy because these three factors are interdependent and lie in a trade-off relation [5]. In general, inorganic semiconductors are promising TE materials due to their intrinsic high thermopower derived from their crystalline structure [6–9], so many studies on the inorganic semiconductors or their alloys with high ZT have been reported. However, they still have low electrical conductivity, and furthermore have some drawbacks which are the scarcity of raw materials, chemical/mechanical unstability, expensive cost and processing rigidity of brittle materials [6,10]. In this study, as one of the promising approaches to overcome these limitations, we suggest a flexible hybrid system including the 1-D inorganic semiconductor with high thermopower such as tellurium nanowire (TeNW) and single-walled carbon nanotube (SWCNT) with high electrical conductivity.

Several approaches for flexible TE materials have already been investigated in order to overcome the limitations of inorganic semiconductors. The early researches focused on the properties of homogeneous materials such as conducting polymers [11,12] or nanocarbons [13–16], and more recently the hybrid systems composed of more than two materials have been studied [17]. Some carbon nanotube (CNT)/polymer hybrid systems for flexible TE materials have been reported [18–20]. They have shown the light weight, flexibility and high electrical conductivity due to CNT, but their TE performance has been unsatisfactory due to the low thermopowers of CNT and polymers. Inorganic semiconductor/conducting polymer hybrid systems have also been suggested for improved TE performance [21–24]. TeNW/water-soluble PEDOT:PSS hybrid system has shown the enhanced thermopower and electrical conductivity, thereby leading to higher ZT compared to PEDOT:PSS or TeNW itself [23,24]. However, the mechanical/chemical stability of this hybrid film has not been satisfied for flexible TE devices [25].

Therefore, we have proposed a new method to fabricate a flexible SWCNT/TeNW hybrid film, in which SWCNT and TeNW have been selected due to their high electrical conductivity and thermopower, respectively. The effects of the weight ratio of SWCNT in hybrid system on the TE performance have been

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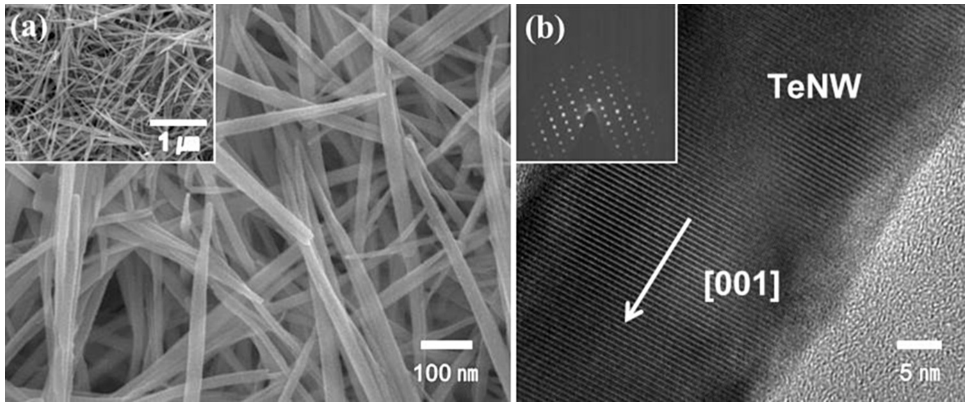

Fig. 1. (a) FE-SEM and (b) TEM images of the synthesized TeNW.

systematically studied. We have demonstrated that the 2 wt% addition of SWCNT into TeNW matrix results in the significant increase of the electrical conductivity maintaining the high thermopower and slight decrease of the thermal conductivity, thereby leading to one order of magnitude higher ZT compared to the pure TeNW film.

2. Experimental details

TeNW was synthesized by the typical procedure reported in the previous literature [26]. All chemicals were purchased from Sigma–Aldrich and used without further purification. 1.0 g of ascorbic acid ($C_6H_8O_6$) and 0.1 g of cetyltrimethylammonium bromide (CTAB, 0.2 mmol) were dissolved in distilled water (40 mL). And 0.052 g (0.25 mmol) of sodium tellurite (Na_2TeO_3) was added to the solution under vigorous stirring, forming a white suspension. The suspension was heated to 90 °C and reacted for 20 h, followed by several washings with DI water and ethanol to remove the excess CTAB. Synthesized TeNW was dried in oven for overnight. For hybrid film preparation, 10 mg of TeNW was re-dispersed in DI water with each volume of SWCNT (AST-100F grade, Hanwha nanotech.) solution. 1 wt% of sodium dodecyl sulfate was added to the solution with ultra-sonification for 10 min for well dispersion. Then, the hybrid film was fabricated under vacuum filtration of the mixed solution on 0.22 μm PVDF filter, washed intensively to remove the used surfactant and dried in a vacuum oven for overnight.

The morphology of the hybrid film was characterized by FE-SEM (JSM-6701F, JEOL) and HR-TEM (Tecnai F20, FEI). The distribution of SWCNT in the TeNW matrix was characterized using Raman

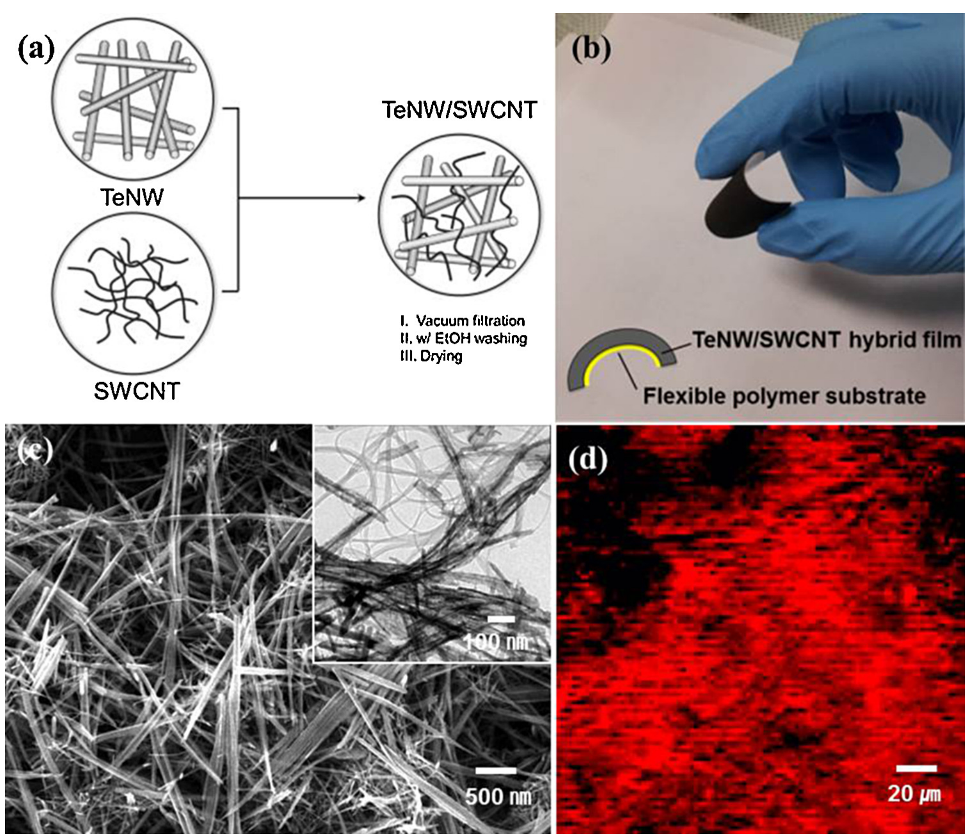

Fig. 2. (a) Schematic illustration for the preparation of SWCNT/TeNW hybrid film, and (b) Optical, (c) FE-SEM (Inserted image: TEM), and (d) Raman mapping images of SWCNT/TeNW film hybridized with 2 wt% of SWCNT.