

Enhanced thermoelectric power factor and low thermal conductivity in one-dimensional Te/Ag₂Te composites

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ABSTRACT

We reports the synthesis and characterization of Te/Ag₂Te nanorod composites with various Ag₂Te contents. The composite samples were prepared by mixing Te and Ag₂Te nanorods synthesized by polyvinylpyrrolidone (PVP)-assisted solution-phase mixing. The thermoelectric properties of the prepared composites vary according to the Ag₂Te content. Furthermore, the samples exhibit an enhancement in electrical conductivity and a reduction in the Seebeck coefficient with increasing Ag₂Te content. The maximum power factor (350.71 μV/K at room temperature) is observed for the sample containing 30% Ag₂Te. The samples were assembled as 1D nanostructures, which led to a decrease in thermal conductivity owing to the strong phonon scattering effect. The maximum thermoelectric figure of merit (ZT) at room temperature, 0.34 was obtained for the sample with 30% Ag₂Te content. This value is 480% larger than that obtained for the pristine Te nanorod samples.

1. Introduction

The rapid increase in the demand for energy in the past few decades requires the development of alternative energy sources. Some renewable energy conversion devices such as piezoelectric and thermoelectric (TE) devices, fuel cells, and solar cells have been developed. Among these alternative energy sources, TE materials are of particular interest, because they can convert heat into electricity, and vice versa. The performance of TE materials is determined by the dimensionless figure of merit $ZT = S^2\sigma T/\kappa$, where σ , κ , S , and T are the electrical conductivity, thermal conductivity, Seebeck coefficient, and absolute temperature, respectively. So far considerable efforts have been made forward enhancing the figure of merit values in other existing TE materials classes, including tellurides [1–3], half-Heuslers [4], and silicides [5]. To achieve high ZT values, materials with a high power factor ($PF = S^2\sigma$) and low thermal conductivity are needed. A high power factor is generally achieved by enhancing the Seebeck coefficient or electrical conductivity of the material.

One simple way to achieve a high Power factor involves using materials with a high Seebeck coefficient. Tellurium (Te) [6,7] and its alloys [8,9] have attracted significant attention for this purpose. Te is a p-type narrow-band-gap semiconductor, with a direct band gap of approximately 0.35 eV at room temperature [10] and a particularly high Seebeck coefficient (~ 400 μV/K at room temperature [11,12]). However, Te does not represent the best option for achieving a high power factor because of its low electrical conductivity (~ 10 S/m at

room temperature [12]) compared to other Te-based materials.

This limitation has prompted many studies aimed to determine methods of enhancing the electrical conductivity of Te [13]. One of the simplest ways to achieve this goal involves preparing composites with other materials [14]. Silver telluride (Ag₂Te) is an n-type narrow band gap semiconductor material with a high electrical conductivity and very high Seebeck coefficient [15,16]. Ag₂Te exists in two phases, α and β -Ag₂Te. The α -Ag₂Te phase, a face-centered cubic structure, is stable at temperatures below 473 K and exhibits better thermoelectric properties than to the monoclinic β Ag₂Te phase [17]. The electrical conductivity of Ag₂Te (~ 20,000 S/m at room temperature [15,16]) is higher than that of Te. Furthermore, Ag₂Te shows an outstanding Seebeck coefficient, which makes it a suitable material for enhancing the power factor of Te.

Another important requirement for achieving a high ZT is a low thermal conductivity. The unique performance and functionalities of one-dimensional (1D) nanostructures have attracted considerable interest [18,19]. The 1D nanostructure of a material can increase its lattice scattering of phonons [20], reducing the thermal conductivity. Therefore, a low thermal conductivity can be achieved by nanostructuring a thermoelectric material. Introducing nanostructures in bulk thermoelectric materials generates a large number of boundaries that can scatter phonons [21]. Previous studies showed that Te-based thermoelectric materials with 1D nanostructure had lower thermal conductivity than their bulk counterparts [22,23]. This suggests that nanostructured Te and Ag₂Te materials could exhibit low thermal

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conductivity, leading to high-performance thermoelectric composites. Yang et al. [24] reported a reduction in the thermal conductivity of Ag₂Te nanowires fabricated by solution-phase synthesis.

The present study describes the preparation of Te and Ag₂Te nanorods and Te/Ag₂Te composites, with the final goal of obtaining materials with enhanced the thermoelectric properties. Te nanorods were fabricated from a Te precursor solution by a polyvinylpyrrolidone (PVP)-assisted solution-phase synthetic process. Ag₂Te nanorods were also fabricated from the Te nanorods using a solution-phase synthetic route. After the synthesis of these materials, Te/Ag₂Te composites with various Ag₂Te contents were fabricated by ball milling. Thermoelectric properties such as S, σ , κ , the power factor and ZT were determined for each sample as a function of its Ag₂Te nanorod content. The combination of Te and Ag₂Te nanorods is expected to lead to enhanced thermoelectric properties in the composite materials.

2. Experimental

2.1. Materials

PVP [molecular weight (MW) = ~40,000] was purchased from Sigma Aldrich (St. Louis, USA). Tellurium (IV) oxide (TeO₂, 99%), ethylene glycol (EG, C₂H₆O₂, 99.5%), sodium hydroxide (NaOH, 98%), silver (I) nitrate (AgNO₃, 99.8%), and hydrazine monohydrate (N₂H₄·H₂O, 80%) were purchased from Daejung Chemicals & Metals Co (Seoul, Korea).

2.2. Synthesis of Te nanorods

Te nanorods were prepared by mixing 0.96g of TeO₂ (MW = 159.6), 0.4g of PVP, 1.2g of NaOH (MW = 40) and 40 ml of ethylene glycol in a 250 ml round-bottom flask under magnetic stirring. This solution was heated to 120 °C, and then 2.45 ml of N₂H₄·H₂O was injected into the solution. The color of the mixture became white within few seconds, indicating the formation of tellurium oxide colloids. The color of the tellurium oxide gradually turned to dark gray. After a reaction time of 90 min, a dark blue solution of Te nanorods was obtained. The resulting solution was poured into a deionized (DI) water solution containing 10 vol% N₂H₄·H₂O and vigorously stirred. Subsequently, the resulting solution was centrifuged with the addition of volumetric water twice, and the supernatant was decanted. Finally, the resulting solution was dried in a vacuum oven at 60 °C for 24 h.

2.3. Synthesis of Ag₂Te nanorods

The Ag precursor solution was prepared by dissolving 0.05g of AgNO₃ and 0.1g of PVP in 30 ml EG. The collected Te nanorods (0.02g) were redispersed in 100 ml EG. After the Ag precursor solution was poured into the EG, the reaction was carried out under magnetic stirring. The temperature was increased to 120 °C and maintained for 90 min. The resulting solution was poured into DI water, centrifuged and then dried in a vacuum oven at 60 °C for 24 h.

2.4. Fabrication of Te/Ag₂Te composite samples

Samples consisting of round disks of the Te/Ag₂Te composites were prepared by ball milling. The Te nanorods were ball-milled with various amounts of Ag₂Te nanorods using zirconia balls in an inert atmosphere for 2 h. The final product was loaded into a Fe mold and pressed at 200 °C under a pressure of 50 MPa for 10 min.

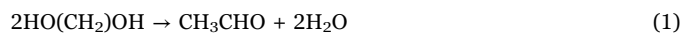
2.5. Sample characterization

The morphology and structural properties of the as-prepared products were analyzed by field-emission scanning electron microscopy (FE-SEM, SIGMA). Elemental mappings of the samples were analyzed

by energy-dispersion X-ray diffraction (EDS, NORAN System 7, Thermo Scientific), X-ray diffraction (XRD, New D8 Advance, Bruker AXS) analyses and X-ray photoelectron spectroscopy (XPS, VG-Microtech, ESCA2000). X-ray diffraction analysis was performed at 40 mA, 40 kV, and a scan rate of 1°/s, with 2 θ ranging from 5 to 70° using Cu K α radiation (λ = 0.154056 nm). The electrical conductivity of the samples was measured using the 4-point-probe method and the thickness of the sample was measured using a digital micrometer. The Seebeck coefficient S was analyzed using a custom-made device containing a pair of thermocouples and voltmeters, according to the linear relationship between the thermal electromotive force (ΔV) and the through-plane temperature difference (ΔT) of the composite films ($S = \Delta V / \Delta T$). The thermal conductivity of the samples was determined using the formula $\kappa = \alpha \rho C_p$, where α is the thermal diffusivity, ρ is the bulk density, and C_p is the specific heat of the materials. The α values were measured using an LFA 447 NanoFlash (NETZSCH), whereas the C_p values were measured using differential scanning calorimetry (DSC 131 Evo, SETARAM Instrumentation) under temperatures between -20 °C and 120 °C at a rate of 10 K/min under N₂ atmosphere.

3. Results and discussion

Te and Ag₂Te nanorods were prepared following a solution-phase synthetic route. The overall scheme for the synthesis of the Te/Ag₂Te composite is shown in Fig. 1. TeO₂ and NaOH were mixed with PVP in EG. N₂H₄·H₂O, was injected into the solution after raising the temperature to 120 °C. In this reaction, PVP plays a key role as a surfactant. Previous studies have reported that linear polymers can react with inorganic ions to form a chain-shaped intermediate [25,26]. PVP induces 1D growth and controls the growth rate of Te and Ag₂Te nanorods. Nucleation of Te²⁺ ions took place at this stage. Te²⁺ ions were reduced to elemental Te and solid crystal nuclei were formed. The reduction accelerated the growth of Te crystal nuclei into Te nanorods, and the Te²⁺ concentration in solution decreased. PVP also plays an important role in the Ag₂Te synthesis, where it protects the Te nanorods so that Ag₂Te nanorods can be produced in the second step. The following reactions occur during the Ag₂Te synthesis:



The synthesis occurs in three stages: (1) EG is dehydrated into acetaldehyde. (2) The aldehyde group of the acetaldehyde reduces the Ag⁺ ions of the Ag precursor to elemental Ag. (3) The newly formed Ag reacts with Te, producing Ag₂Te nanorods.

In order to confirm the formation of Te and Ag₂Te nanorods, XRD patterns were recorded. Fig. 2a shows the XRD patterns of Te and Ag₂Te nanorods. All peaks in the XRD pattern of the Te nanorods can be indexed to the hexagonal phase of Te (JCPDS no. 36-1452), whereas the XRD peaks observed for the Ag₂Te nanorods, also shown in Fig. 2a, can be indexed to Ag₂Te (JCPDS no. 34-0142). The absence of other peaks denotes the successful formation of pure Te and Ag₂Te.

XPS analysis was used to further confirm the successful synthesis of Te and Ag₂Te samples. The XPS data for the Te sample are shown in Fig. 2b and c. The two peaks at 572.5 and 582.9 eV correspond to the Te 3d_{5/2} and Te 3d_{3/2} states, respectively. The other two peaks at 586.4 and 576.1 eV can be assigned to the binding energy of Te(IV) 3d states indicating the oxidation of Te. This finding demonstrates that the outer surface of the synthesized Te sample is easily oxidized in air, because of its nanostructure; in fact, previous studies showed that nanomaterials are could be oxidized more easily in air [27]. The XPS spectra of the Ag₂Te nanorods are shown in Fig. 2d-f. The peaks at 368.3 and 572.5 eV correspond to Ag 3d_{5/2} and Te 3d_{5/2} states, suggesting that

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