

1 Article

2 Te nanoneedles Induced Entanglement and 3 Thermoelectric Improvement of SnSe

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18 1. Experimental Methods

19 1.1. Preparation of SnSe Decorated with Te Nanoneedles

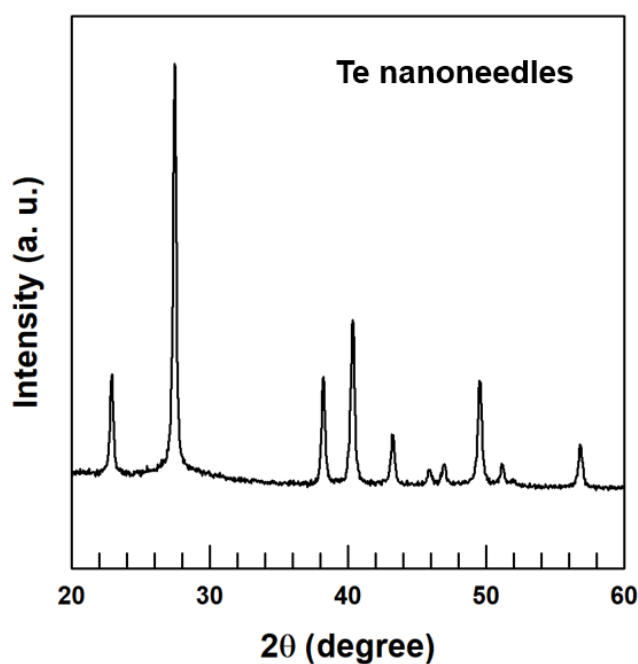
20 The SnSe crystals used in the present work were supplied in 99.999% purity by Alfa Aesar. The
21 TeO₂ (99%), polyvinylpyrrolidone (PVP, molecular weight ~40,000) and ethylene glycol (C₂H₆O₂,
22 99%) were supplied by Daejung Chemicals & Metals Co., Ltd. The SnSe crystals were ball-milled into
23 small particles using milling machine (HPM-700, Haji Engineering) with zirconia balls in an inert
24 atmosphere (N₂ gas). The rotation speed and milling duration of the planetary mill were set to 150
25 rpm and 6 h to generate a rolling action of the balls. The resulting SnSe powder (0.282 g) was added
26 to a round-bottomed flask containing 150mL of ethylene glycol and stirred for 10 min. TeO₂ (0.3 g),
27 and PVP (0.27 g) were then added to the solution, followed by stirring and gradual heating to 433 K.
28 This temperature was then maintained for various reaction times (0, 1, 2, 4, and 6 h) for the
29 optimization of the Te nanoneedle synthesis. The obtained samples are subsequently referred to as
30 Te nanoneedles(x)/SnSe, where 'x' indicates the reaction time (i.e. x = 0, 1, 2, 4, or 6 h). The resulting
31 product was collected and washed several times with water and ethanol, then dried overnight in a
32 vacuum oven. Finally, the product was pressed under 50 MPa at 823 K for 10 min, yielding pelletized
33 samples.

34 1.2. Characterization

35 The crystal structure of the materials was characterized by X-ray diffraction (XRD; New D8-
36 Advance/Bruker-AXS) at 40 mA and 40 kV, with Cu K α radiation (0.154056 nm) and a scan rate of
37 1°/s for 2 θ ranging from 5° to 70°. The morphologies were observed by field-emission scanning
38 electron microscopy (FE-SEM, SIGMA). The elemental mappings were performed by energy-
39 dispersive X-ray spectroscopy (EDS, NORAN system 7, Thermo Scientific). The binding energy peaks
40 were investigated by X-ray photoelectron spectroscopy (XPS, Thermo U. K. K-alpha) using a
41 monochromatic Al K α X-ray source (1486.6 eV) and a hemispherical analyzer. The thermal

42 conductivities were calculated from the formula $\kappa = \alpha \cdot \rho_d \cdot C_p$, where α is the thermal diffusivity, C_p
43 the specific heat, and ρ_d is the bulk density of the material. The thermal diffusivity was measured
44 using an LFA-457 Nanoflash (NETZSCH) and the specific heat was measured using differential
45 scanning calorimetry (DSC 131 Evo, Setaram Instrumentation). The DSC measurements described
46 the thermal transition behavior of the composites, with the samples heated from -20 to 120 °C at a
47 rate of 10 °C/min in a N₂ atmosphere. A four-point probe method with disk-shaped compressed
48 pellets was used to measure the electrical conductivity. A custom-made device containing a pair of
49 thermocouples and voltmeters was used to measure the Seebeck coefficient, S . The measurement was
50 repeated five times for each sample for reproducibility.

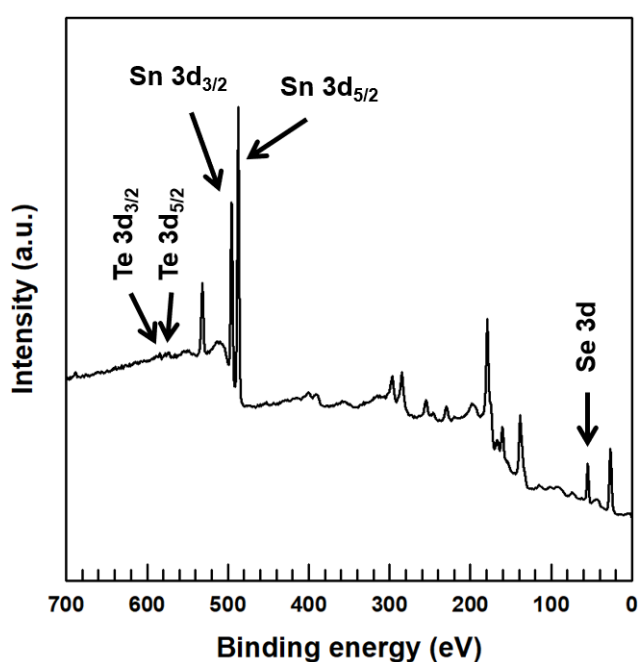
51 2. Figures



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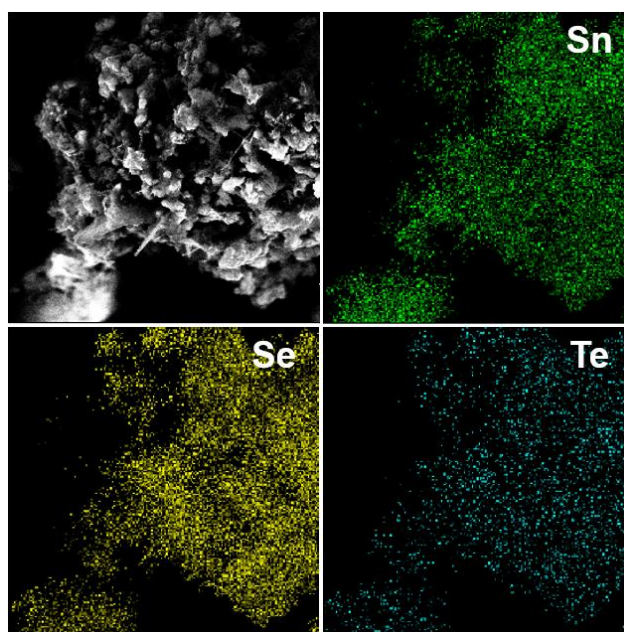
Figure S1. XRD patterns of the Te nanoneedles.



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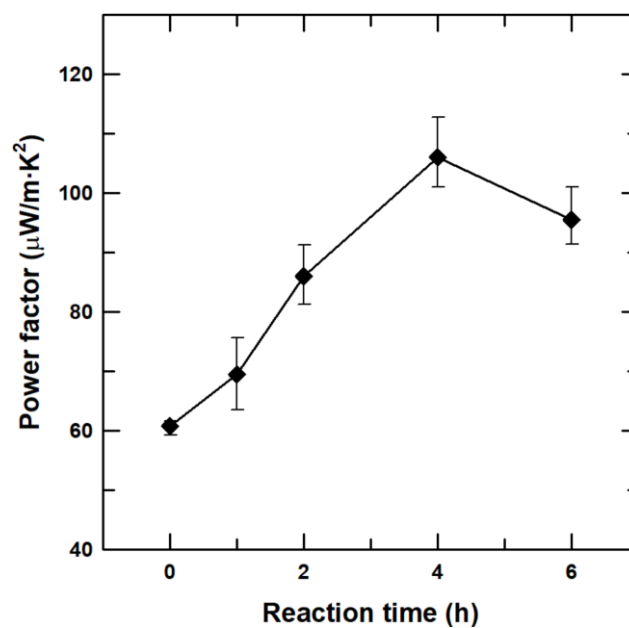
Figure S2. Survey scan XPS spectrum of the Te nanoneedles(4h)/SnSe composite.



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Figure S3. FE-SEM image of the Te nanoneedles(4h)/SnSe composite and the resulting EDS mappings of Sn, Se, and Te atoms.



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Figure S4. Power factor (σS^2) values of the Te nanoneedles(x)/SnSe composites as a function of reaction time.



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