ADVANCED ELECTRONIC MATERIALS

Supporting Information

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High-Performance Solution-Processable Flexible SnSe Nanosheet Films for Lower Grade Waste Heat Recovery

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Methods and Materials

Synthesis of SnSe Nanostructures

A modified hydrothermal growth method is used to synthesize the SnSe nanostructures. First, 1.4 g of $SnCl_2 \cdot 2H_2O$ crystals (Sigma-Aldrich) are added to a 50 mL beaker filled with 50 mL of deionized water, and the contents are sonicated for 10 minutes in 2 L of cold water (Jeken PS-40A). Next, 3.6 g of NaOH pellets (Sigma-Aldrich) are added to the beaker, and the contents are sonicated for 10 minutes. In a separate, Teflon coated container, 0.24 g of Se pellets (Sigma-Aldrch) are added together with a magnetic stir bar that can fit circumscribed inside the container. The contents of the 50 mL beaker are poured into the Teflon container, and the container is placed inside a stainless steel autoclave vessel; the nutrients $SnCl_2$ and Se begin to dissolve in the H_2O and NaOH solvent solution. The vessel is then sealed, and heated on a preheated hot plate at 180^oC for 5 hours at 400 RPM surrounded by ambient conditions.

The vessel is allowed to cool to ambient conditions after 3 hours under 400 RPM stirring, and the contents are collected and cleaned; the SnSe nanosheets precipitate out of solution. To clean and purify the sample, the reaction mixture is transferred to a 200 mL beaker, and gravitational sedimentation is utilized in 30 minute increments. After the initial decant, 100 mL of DI water is added for each subsequent decant for a total of 4 cleaning cycles and 5 decanting processes. The residual products are then dried in the vacuum oven for 18 hours at 333 K.

Figure S1: Stainless steel autoclave vessel with Teflon coated container inside.

Thin Film Sample Preparation

Thin film samples are prepared for direct drop casting, 150 μL of DI water is added to 0.025 g of SnSe product in a 0.6 mL microcentrifuge tube. After sonicating the solution for 1 hour, followed by vortex mixing the sample for 10 seconds, 7 μL of solution is drop cast using a 2-20 μL pipet on copper tape on either a polyethylene terephthalate or glass substrate. The samples are heated at 313 K for 15 minutes on a hot plate until dry.

Seebeck Coefficient and Electrical Conductivity Measurement

The Seebeck coefficient measurement is conducted through standard setup consisting of an IOtech Personal DAQ 3005, sbRIO 9602, DL Instruments 1201 Low Noise Voltage Preamplifier, and E-type thermocouples integrated in a LabView environment. Two thermocouples serve the dual purpose to measure both the thermopower voltage and temperature gradient simultaneously. Multiplexing and line cycle reject are additionally implemented taking data 10 data points at 10 Hz using the IOTech DAQ 3005 together with Ithaco 1201 Low Voltage Noise Preamplifer with 10 Gain, differential DC voltage input with 30 Hz low-pass filtering.

The Seebeck coefficient is computed by fitting the slope (α) of temperature dependent voltage change[1]:

$$\alpha = -\frac{\Delta V}{\Delta T} + \alpha_{wire}(T) = -\frac{\Sigma x_i \Sigma y_i - N\Sigma x_i y_i}{(\Sigma x_i)^2 - N\Sigma x_i^2} + \alpha_{wire}(T) \qquad [1]$$

where x_i is the temperature gradient, y_i is the voltage gradient, and N is the number of samples. The slope method employed removes the contact voltage offset, and the reference probe correction is assumed to be temperature independent over the 300 – 400 K region. Electrical transport is performed in ambient conditions through standard two- and four-terminal measurements using a high-precision voltage meter (Keysight 2902a) and analyzed by Quick IV software.

Chemical Synthesis and Measurement of SnSe Single Crystals

We synthesized single crystal SnSe by chemical vapor transport method. High purity of tin and arsenide powder were grind together by mortar and pestle. The mixed powder was placed in quartz tube. The quartz tube is evacuated and flame sealed under high vacuum. The quartz tube is placed on the three zone furnace. Reaction temperature was set to the 950°C, 925°C, and 900°C for hot zone, middle zone, and cold zone, respectively. The temperature of furnace was monotonically heated from 25 ˚C to the reaction temperature for 24 hours. The reaction temperature was held for 4 days and slowly cooled down to the room temperature. The single crystal is used as a calibration for the system, and is recorded with a Seebeck coefficient measured to be 809 $\pm$ 55 μV/K.

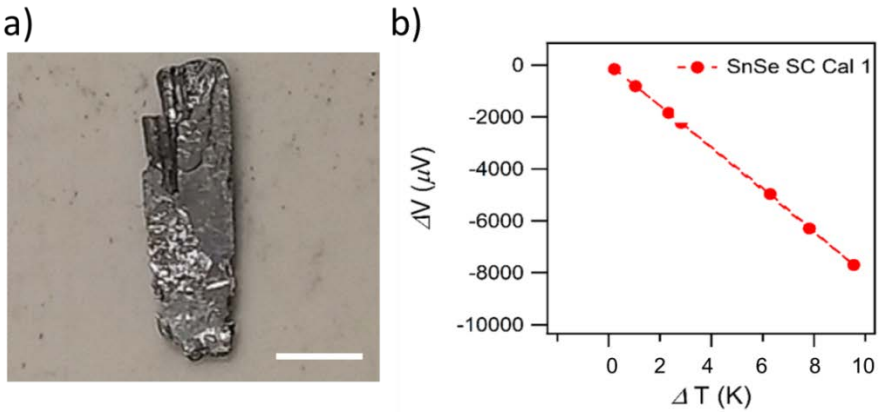

Figure S2. (a) A digital image of the SnSe single crystal synthesized using chemical vapor deposition method. The scale bar is 1 cm. (b) The Seebeck coefficient measurement of a SnSe single crystal.

Table S1: Summary of literature room-temperature thermoelectric performance.

σ (S/cm)	α (μV/K)	κ (W/mK)	PF (μW/mK^2)	ZT	Morphology	Ref.
0.1	900	0.5	8.1	0.005	$Sn_{0.9}Ge_{0.1}Se$	[2]
1.11	900	1.7	89.9	0.016	SnSe Single Crystal	[3]
8	320	0.85	81.9	0.029	Polycrystalline SnSe (with rock salt phase)	[4]
10	525	0.6	275.6	0.138	c axis, Single Crystal	[5]
320	110	0.36	387.2	0.323	Polymer Nanosheets, PEDOT:PSS-SnSe	[6]

880	73	0.33	469.0	0.426	PEDOT:PSS Dedoped with EG	[7]
800	155	1.1	1922.0	0.524	α-MgAgSb	[8]
1000	165	1.35	2722.5	0.605	c axis, Doped Single Crystal	[9]
455	220	0.75	2202.2	0.881	$MgAg_{0.97}Sb_{0.99}$	[10]
1250	185	1.1	4278.1	1.167	$Bi_{0.5}Sb_{1.5}Te_3$ Alloy P-Type	[11]
1200	185	1	4107.0	1.232	$Bi_2Te_{2.79}Se_{0.21}$ N-type	[12]
720	235	0.7	3976.2	1.704	$Bi_{0.5}Sb_{1.5}Te_3$ Alloy P-Type Engineered	[13]

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