Supporting information

High power factor Ag/Ag₂Se composite film for flexible thermoelectric generator

Qi Gao^a, Wu Wang^b, Yao Lu^{a,b}, Kefeng Cai^{a*}, Yating Li^a, Zixing Wang^a,

Miaomiao Wu^a, Changjun Huang^a, Jiaqing He^{b*}

Corresponding authors

*Email address: kfcai@tongji.edu.cn; hejq@sustc.edu.cn

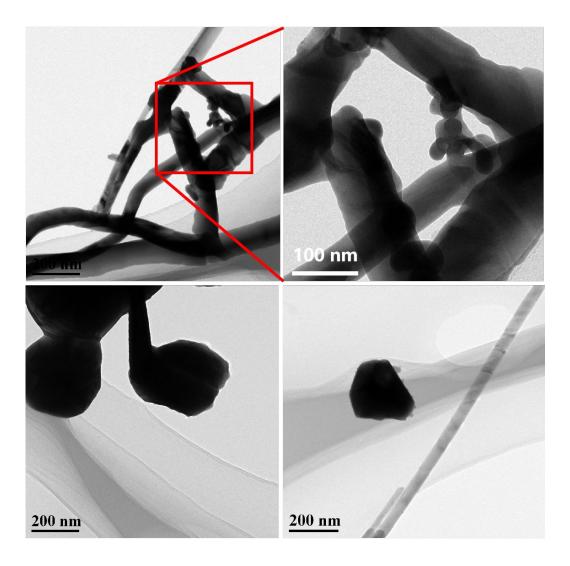


Figure S1 TEM image of the Ag/ Ag₂Se composite powders (Ag particles and Ag₂Se multiscale nanostructures)

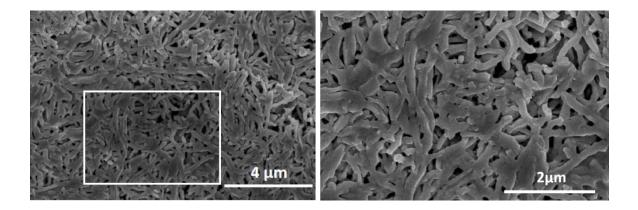


Figure S2 Surface FESEM image of the synthesized Ag/ Ag₂Se composite powders after vacuum filtration and then cold-pressing.

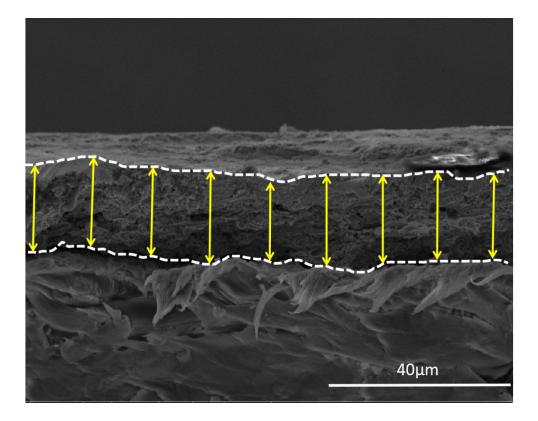


Figure S3 Fractured surface SEM image of the sample 3 on nylon membrane.

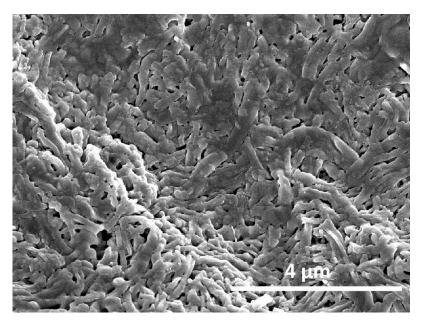


Figure S4 surface SEM image of sample 1

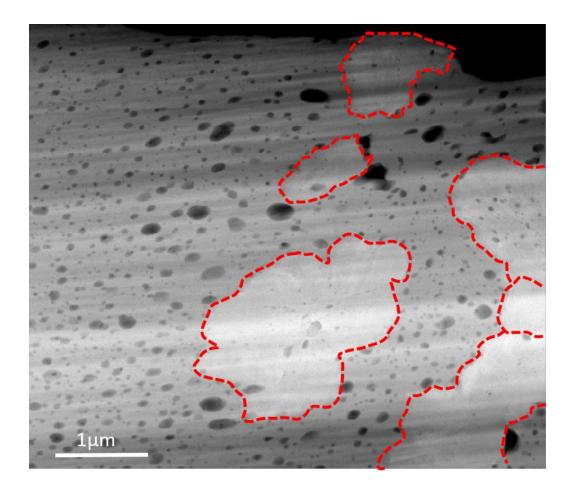


Figure S5 A TEM image of the sample 3. The Ag grains are marked by red dotted lines.

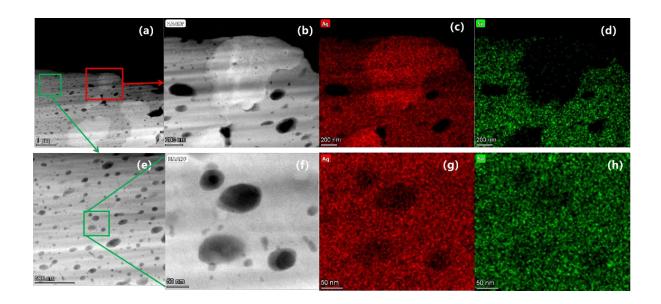


Figure S6 (a) overview TEM image of sample 3. (b) HAADF image corresponding to the area marked by a red square in (a) and corresponding EDS mappings of the elements Ag (c) and Se (d). (e) overview image of the Ag₂Se phase corresponding to the green square in (a). (f) HAADF image corresponding to the square in (e) and corresponding EDS mappings of the elements Ag (g) and Se (h).

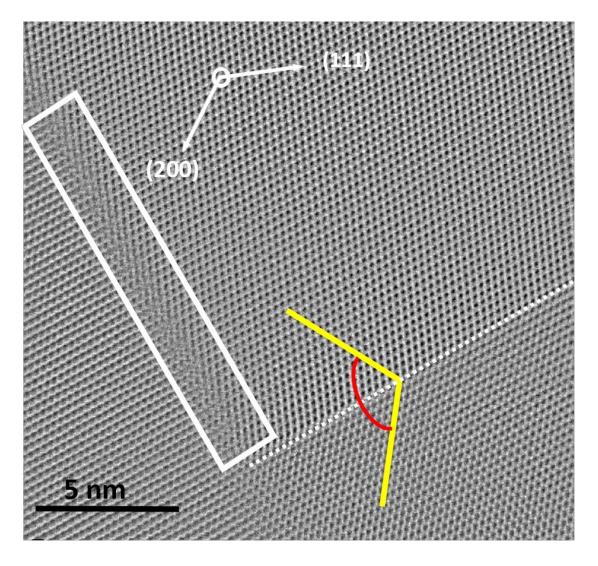


Figure S7 A HRSTEM image of Ag grains in sample 3.

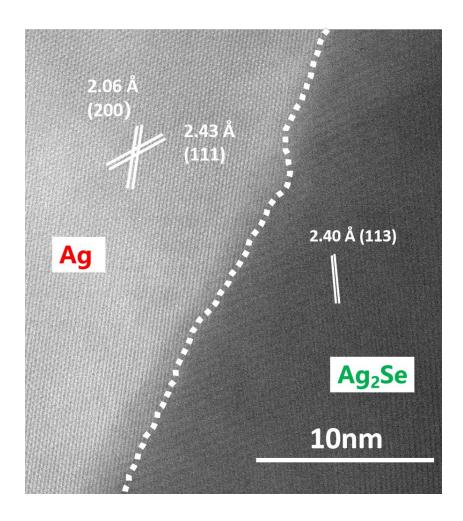


Figure S8 HRSTEM image of a heterointerface between a Ag grain and Ag₂Se grain in sample3.

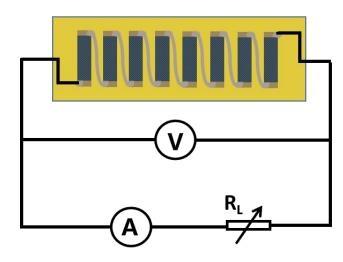


Figure S9 Schematic illustration of the performance measurement of the TEG

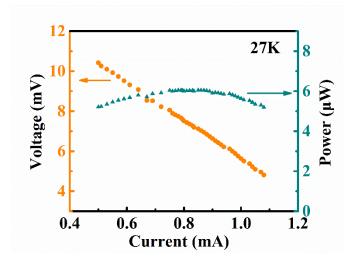


Figure S10 Output voltage and output power versus current at ΔT of 27 K of the TEG after having been stored in a drying cabinet (humidity 20~30%) for three months

Note 1 Synthesis of Se NWs

In a typical procedure, 1 g SeO₂ and 1 g β -cyclodextrin were added into 200 mL distilled water slowly in a glass beaker to form solution A with stirring. Then, 2 g L-ascorbic acid was slowly added into 200mL distilled water to form solution B with stirring. After fully dissolving, the solution A was slowly mixed with solution B under continuous agitation. After stirring for about 4 h at room temperature, the product underwent a process of centrifugation, alternating washing with distilled water and ethanol for several times. The prepared Se NWs were dispersed in ethanol for 2 days. Finally, the prepared Se NWs were redispersed into 400 mL EG and a suspension was formed for further synthesis reaction.

Note 2 Assembly of the flexible TE generator

The optimized film was cut into strips (6 mm \times 20 mm) and pasted on a polyimide (PI) substrate with an interval of ~ 5 mm. Then, a layer of Au was evaporated on two ends of each strip and subsequently Ag adhesive (SPI# 04998-AB) was painted to connect the 8 legs in series.

Note 3 Characterization and measurement

Electrical conductivity at RT was measured using a steady-state four-probe technique with a square wave current (~ 10 mA in amplitude) by Ecopia HMS-3000. The Seebeck coefficient at RT was determined by the slope of the linear relationship between the thermal electromotive force and temperature difference (~ 5 K) between two ends of each film. The temperature-dependent TE properties were measured by a Cryoall CTA-3 instrument in He atmosphere, with an instrument test error of $\pm 5\%$ for both σ and S. Hall measurement was performed using the Van der Pauw method (The Lake Shore 8400 Series).

The phase composition of the samples was examined by X-ray diffraction (XRD) (D/MAX 2550VB3+/PCII). The morphology of the samples was observed by field-emission scanning electron microscope (FESEM) (FEI Nova NanoSEM 450). The internal nanostructure of the film was examined by transmission electron microscope (TEM, Titan Themis G2 60-300, Thermo Fisher Scientific). The sample for TEM analysis was fabricated with a focused ion beam (FIB, Helios Nanolab G3 UC, Thermo Fisher Scientific). During FIB processing, in order to avoid ion damage, a thin layer of Pt with thickness of around 2 µm was firstly

deposited on the surface of the thin film. A cross-sectional lamella beneath the Pt layer with size of 10 μ m × 5 μ m × 1 μ m was cut by high-energy Ga ion beams and was bonded to a post of copper lift-out grid with the FIB easy-lift system. The center area of the lamella was further thinned with low-energy Ga ion beams until the thickness was suitable for TEM observation. The contamination of Ga on the surface lamella was carefully removed after a shower of low-energy Ga ion beam. The film thickness was determined by FESEM observation. The thickness is around 20 μ m and the details can be seen in the fracture surface SEM image in Figure S3. The bending test was performed using a home-made apparatus around a rod with a diameter of 4 mm.

For the TE power generator, its output performance was measured by a self-made simple circuit. Briefly, the value of load resistance in the circuit is adjusted to measure the output characteristics of the device when a certain ΔT is applied at both ends of the device (Figure S9).