**Supplementary Materials:** 

# Facile fabrication of poly(3,4ethylenedioxythiophene):poly(styrenesulfonate)-coated selenium nanowire/carbon nanotube composite films for flexible thermoelectric applications

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#### Note 1. Expeimental Methods

#### 1.1. Materials

A PEDOT:PSS aqueous solution (CLEVIOS PH1000) was purchased from Heraeus Clevios GmbH. The multi-walled carbon nanotubes (MWCNTs) used in this study were supplied by LG electronics, Korea. Selenium (IV) oxide (SeO<sub>2</sub>, 99.5%), L(+)-ascorbic acid (C<sub>6</sub>H<sub>8</sub>O<sub>6</sub>, 99.5%),  $\beta$ -cyclodextrin (C<sub>42</sub>H<sub>70</sub>O<sub>35</sub>, 98%), ammonium hydroxide (NH<sub>4</sub>OH), and ethanol (C<sub>2</sub>H<sub>5</sub>OH, C<sub>2</sub>H<sub>5</sub>OH) were purchased from Daejung Chemicals & Metals Co. (Seoul, Korea).

#### 1.2. Synthesis of PEDOT-coated Se NWs

PEDOT:PSS (4 mL), SeO<sub>2</sub> (1 g), and  $\beta$ -cyclodextrin (1 g) were dispersed in 100 mL of DI water in a round-bottom flask with stirring. In another beaker, ascorbic acid (1 g) was dissolved in 100 g of water and then stirred. After few minutes, the ascorbic acid solution was poured into the round-bottom flask. After reacting for 4 h, the precipitate was collected by centrifugation and washed alternately in DI water and ethanol. The fabricated powder was then stored in 200 mL of ethanol. Then, ammonium hydroxide (2 mL) was added to the solution, and the solution was stored at room temperature for 48 h, during which flocculated precipitate (PEDOT-Se) was formed. The final product was centrifuged and washed with DI water and ethanol sequentially.

#### 1.3. Fabrication of PEDOT-coated Se NW/MWCNT composites.

To prepare the PEDOT-Se/MWCNT composites, certain amounts of PEDOT-Se and MWCNTs were dispersed in 10 mL of ethanol. The total weight of the solid materials containing various properties of MWCNTs (1, 2, 3, and 5 wt.%) was controlled to 0.01 g. The resulting solution was ultrasonicated for 30 min. After ultrasonication, the solutions were then

filtered through a nylon membrane filter paper to obtain the composite samples, which were dried at 333 K for 12 h.

#### 1.4. Characterization

X-ray diffraction (XRD, New D8 Advance, Bruker AXS) was used to characterize the crystal structures of the synthesized materials. XRD was performed at 40 mA, 40 kV, and a scan rate of 1°/s, with 20 ranging from 5° to 70° using Cu K $\alpha$  radiation ( $\lambda$  = 0.154056 nm). The binding energy peaks of the synthesized materials were investigated with X-ray photoelectron spectroscopy (XPS, VG-Microtech, ESCA2000). The microstructure was observed with field-emission scanning electron spectroscopy (FE-SEM, SIGMA) and field-emission transmission microscopy (FE-TEM, JEM-2100F). Elemental mappings of the samples were analyzed by energy-dispersive X-ray spectroscopy (EDS, NORAN system 7, Thermo Scientific). The electrical conductivity ( $\sigma$ ) of the composites was investigated with the four-probe method. The custom device consisted of a thermocouple, and voltmeters were used to measure the Seebeck coefficient (*S*), which was determined based on the linear relationship between the thermal electromotive force ( $\Delta V$ ) and the temperature difference ( $\Delta T$ ) of the composite ( $S = \Delta V/\Delta T$ ). Five replicates of the composite samples were used for each test to verify the reproducibility of the experiments.

## 2. Figures



Fig. S1. (a) FE-SEM image and the (b-c) corresponding EDS mapping images of PEDOT-

coated Se NWs.



Fig. S2. Digital photograph of PEDOT:PSS-coated Se NW/MWCNT composite film with 2

wt.% of MWCNT.



**Fig. S3.** Demonstration of ratiof of Seebeck coefficient (*S*/*S*<sub>0</sub>), electrical conductivity ( $\sigma/\sigma_0$ ),

and PF  $(PF/PF_0)$  as a function of bending cycles.

	σ	S	PF	D. (
	(S/cm)	(µV/K)	$(\mu W/m \cdot K^2)$	Ref.
MWCNTs	896.2	12.2	13.34	1
PEDOT:PSS	620	23.1	33	2
Se NWs	1.4×10 <sup>-7</sup>	1000	1.4×10 <sup>-5</sup>	3
PEDOT:PSS- Se NWs	4.89	91.84	4.09	This study
PEDOT:PSS-Se/MWCNT with 1 wt.% of MWCNT	81.79	88.21	63.64	This study
PEDOT:PSS-Se/MWCNT with 2 wt.% of MWCNT	114.21	80.46	73.94	This study
PEDOT:PSS-Se/MWCNT with 3 wt.% of MWCNT	125.21	68.59	58.91	This study
PEDOT:PSS-Se/MWCNT with 5 wt.% of MWCNT	151.37	45.90	31.89	This study

**Table S1.** Thermoelectric properties ( $\sigma$ , S, PF) of MWCNT, PEDOT:PSS, Se NW,

PEDOT:PSS-coated Se NW and PEDOT:PSS-coated Se NW/MWCNT with various

MWCNT contents

	Contents (wt.%)			
	Se NW	PEDOT:PSS	MWCNT	
PEDOT:PSS-coated Se NW	~90	~10	-	
1 wt.% MWCNT	~89.1	~9.9	~1	
2 wt.% MWCNT	~88.2	~9.8	~2	
3 wt.% MWCNT	~87.3	~9.7	~3	
5 wt.% MWCNT	~85.5	~9.5	~5	

 Table S2. Weigh ratio of Se NW, PEDOT:PSS, and MWCNT in PEDOT:PSS-coated

Se/MWCNT composites with various MWCNT contents.

### References

- 1. C. Meng, C. Liu and S. Fan, Adv. Mater., 2010, 22, 535-539
- 2. G-H. Kim, L. shao, K. Zhang and K. P. Pipe, *Nat. Mater.*, 2013, **12**, 719-723
- 3. C. Kim, J. Hong and J-W. Park, *Polymers*, 2019, **11**. 1052