Supplementary Information

Key parameters for enhancing the thermoelectric power factor of PEDOT:PSS/PANI-CSA multilayer thin films

Hyung Myung Lee^{a‡}, Gopinathan Anoop^{a‡}, Hye Jeong Lee^a, Wan Sik Kim^a, and Ji Young Jo^{a,*}

^aSchool of Materials Science and Engineering, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea.

*Email:jyjo@gist.ac.kr

Experimental

Solution preparation. PEDOT:PSS (clevios PH1000) solution with 5 vol% dimethyl sulfoxide (DMSO) was stirred for 24 hrs at room temperature after filtering with a syringe filter with a pore size of 0.45 μ m. The PANI-CSA was mixed with m-cresol solvent and the solution was stirred at 50 °C for 24 hrs and filtered using polytetrafluoroethylene filters with a pore size of 2.7 μ m.

Multilayer deposition. The PANI-CSA solution was dropped onto a glass substrate with an area of $1 \times 1 \text{ cm}^2$ using a syringe filter, and then spin-coated at various spin speeds ranging from 4000 to 9000 rpm for 30 s. In order to remove the residual solvent, the PANI-CSA films were then dried at 50 °C for 1 hr in a glove box. The PEDOT:PSS solution was spin-coated onto a PANI-CSAcoated glass substrate at a similar spin speed and spin time as those used for the PANI-CSA layer deposition. The deposited PEDOT:PSS/PANI-CSA film was baked at 120 °C for 15 mins to remove the solvent. The ML thin films were prepared via alternately stacking PEDOT:PSS and PANI-CSA layers via repeated deposition. The as-deposited ML thin films were treated using DMSO solvent to enhance the electrical conductivity. In order to identify the number of layers in ML thin films, the films are labelled using "*n*". In our earlier study¹, the PANI-CSA was synthesized in house and the properties were reported elsewhere.² The PANI-CSA labelled SLIP-p1 was used to fabricate multilayer thin films in our earlier study. It is reported that the precursor solution is very critical in determining the thermoelectric properties of multilayer thin films.^{3,4} The SLIP-p1 PANI-CSA possess higher molecular weight than the conventional PANI-CSA. Therefore, the TE properties of the multilayer thin films fabricated in this study using conventional PANI-CSA is different from those reported in our earlier study, which utilized SLIP-p1 PANI-CSA.^{1,2}

Characterization. The thickness of the films was measured using a surface profiler (DEKTAK II). The electrical conductivity of the films was measured using the Van der Pauw geometry at room temperature. The experiment was repeated for various samples grown under similar conditions and an average value was taken for accuracy and reliability. Raman spectra were recorded using a Raman microscope (RENISHAW, inVia) with an excitation wavelength of 514 nm. The inplane *S* value was measured using a four-point probe SEEPEL thermoelectric measurement system (TEP 600, Seepel instrument), and the average of at least ten measurements results was calculated.

2



Figure S1. Fitted Raman spectra of PEDOT:PSS/PANI-CSA multilayer thin films having two and eight layers.



Figure S2. S2p XPS spectra of single-layer PEDOT:PSS thin films with respect to the deposition spin speed.



Figure S3. The Raman spectra of single-layer PEDOT:PSS thin films with respect to the thickness of each layer.

References

- 1 H. J. Lee, G. Anoop, H. J. Lee, C. Kim, J. W. Park, J. Choi, H. Kim, Y. J. Kim, E. Lee, S. G. Lee, Y. M. Kim, J. H. Lee and J. Y. Jo, *Energy Environ. Sci.*, 2016, **9**, 2806–2811.
- 2 C. Kim, W. Oh and J. W. Park, *RSC Adv.*, 2016, **6**, 82721–82725.
- 3 V. Andrei, K. Bethke, F. Madzharova, A. C. Bronneberg, J. Kneipp and K. Rademann, *ACS Appl. Mater. Interfaces*, 2017, **9**, 33308–33316.
- 4 Z. Fan, D. Du, H. Yao and J. Ouyang, ACS Appl. Mater. Interfaces, 2017, 9, 11732–11738.