Supporting Information

Significantly Enhancement in Seebeck Coefficient and Power Factor of Thermoelectric Polymers by Soret Effect of Polyelectrolytes

Xin Guan, Hanlin Cheng and Jianyong Ouyang*
Department of Materials Science and Engineering, National University of Singapore, 7 Engineering Drive 1, Singapore, 117574
* Corresponding Author.
E-mail: mseoj@nus.edu.sg

Materials and Methods
Table S1

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Materials: PEDOT:PSS aqueous solution (Clevios PH1000) was purchased from Heraeus. The concentration of PEDOT:PSS was 1.3 wt%, and the weight ratio of PSS to PEDOT was 2.5 in solution. Other chemical including PSSH and PSSNa were obtained from Sigma-Aldrich.

Treatment of PEDOT:PSS films: PEDOT:PSS films were prepared by spin-coating the PEDOT:PSS aqueous solution on glass substrates which were pre-cleaned successively with detergent, deionized water, acetone and isopropanol (IPA). They were dried at 120 °C on a hot plate for 20 min. They were then treated with 1 M H_2SO_4 for 3 times as described in our previous works.[1] PSSH layer was prepared on the acid-treated PEDOT:PSS films by spin coating 300 μL of 1 wt% PSSH at 3000 rpm/min for 60 s. The PSSH/PEDOT:PSS films were dried overnight in air at room temperature. The procedure for the NaOH treatment of A-PEDOT:PSS is the same as our previous work.[2] Aqueous solution of 2.7 wt% PSSNa that has the same molar concentration of 1 wt% PSSH is also coated on A-PEDOT:PSS or BA-PEDOT:PSS.

Characterization: The electrical conductivities of the PEDOT:PSS films were measured by the van der Pauw four-probe technique using a Keithley 2400 source/meter. The thicknesses of the polymer films were determined with an Alpha 500 step profiler. The Seebeck coefficients were measured using a home-built system (the same as our previous works), which included two Peltier devices (TEC1-19906 by Beijing Geshang Electronic Pte. Ltd.) affixed on an alumina heat sink. [2] The temperature difference (ΔT) was detected with two T-type thermocouples of 25 μm in diameter (Omega, US), and the thermal voltage output (ΔV) was measured with a Keithley 2000 multimeter. For each sample, the ΔV values were measured at seven different ΔT values. The Seebeck coefficient of each film was extracted from the best linear fitting of the ΔV-ΔT plots, and the coefficient of determination (R^2) of fitting line is higher than 0.99. The output voltage was measured using a home-built system. The films for the output voltage measured had a length of 2.6 cm and a width of 1.5 cm. The distance between the two wires during voltage collection was 1.3 mm. The temperature gradient was 1 K. An external resistor that had a same resistance as the internal resistance of the sample was connected during the measurements. In-situ Raman spectra were collected in a back scattering configuration using a Jobin Yvon

Horiba LabRam HR 800 Raman system. A 514.5 nm Ar^+ was used as the excitation source.

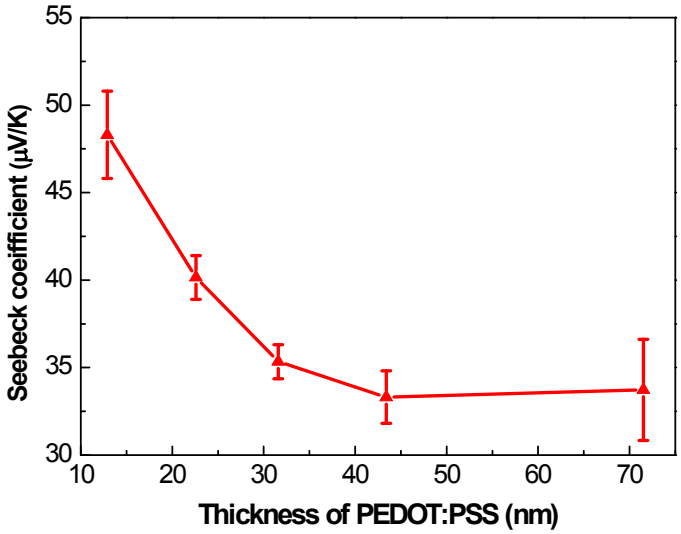

Figure S1. Variation of the Seebeck coefficient of PSSH/A-PEDOT:PSS films with the thickness of the A-PEDOT:PSS films.

Table S1. TE properties of A-PEDOT:PSS, BA-PEDOT:PSS, PSSH/A-PEDOT:PSS, PSSNa/A-PEDOT:PSS, and PSSNa/BA-PEDOT:PSS at the RH of 20%.

Sample	S (μV K^{-1})	σ (S cm^{-1})	PF (μW m^{-1} K^{-2})
A-PEDOT:PSS	18.9	3152	112.6
BA-PEDOT:PSS	36.1	2122	276.5
PSSH/A-PEDOT:PSS	28.3	2687	215.2
PSSNa/A-PEDOT:PSS	28.1	2403	189.7
PSSNa/BA-PEDOT:PSS	37.3	2163	300.9

S: Seebeck coefficient, σ: electrical conductivity, PF: power factor.

References

1. Y. Xia, K. Sun and J. Ouyang, *Adv. Mater.*, 2012, **24**, 2436-2440.
2. Z. Fan, P. Li, D. Du, J. Ouyang, *Adv Eenergy Mater.* **2017**, *7*, 1602116.