

## Supporting Information

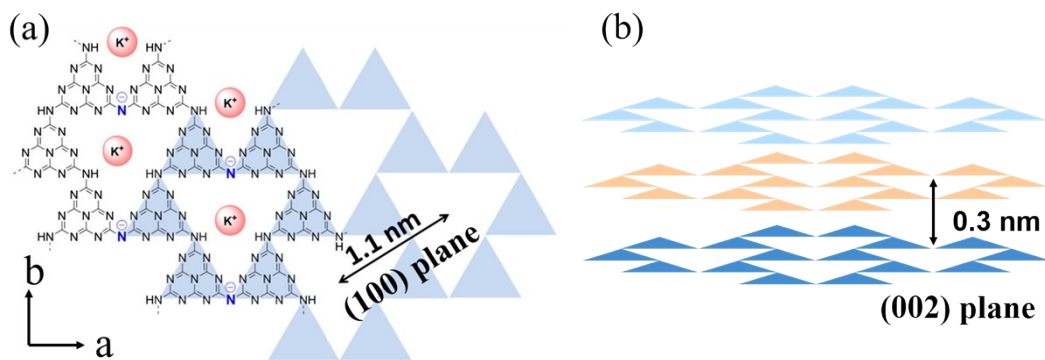
### **Visible light-induced enhancement in the Seebeck coefficient of PEDOT:PSS composites with two-dimensional poly-(heptazine imide)**

Binrong Li,<sup>‡a,b</sup> Shizhong Yue,<sup>‡a</sup> Hanlin Cheng,<sup>a</sup> Chundu Wu,<sup>\*b</sup> Jianyong Ouyang<sup>\*a</sup>

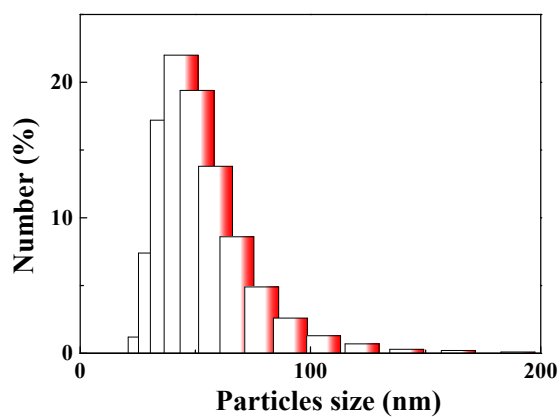
*<sup>a</sup> Department of Materials Science and Engineering, National University of Singapore, Singapore 117575. Email: mseoj@nus.edu.sg*

*<sup>b</sup> School of Environment and Safety Engineering, Jiangsu University, Zhenjiang 212013, China. Email: wcdujs@126.com*

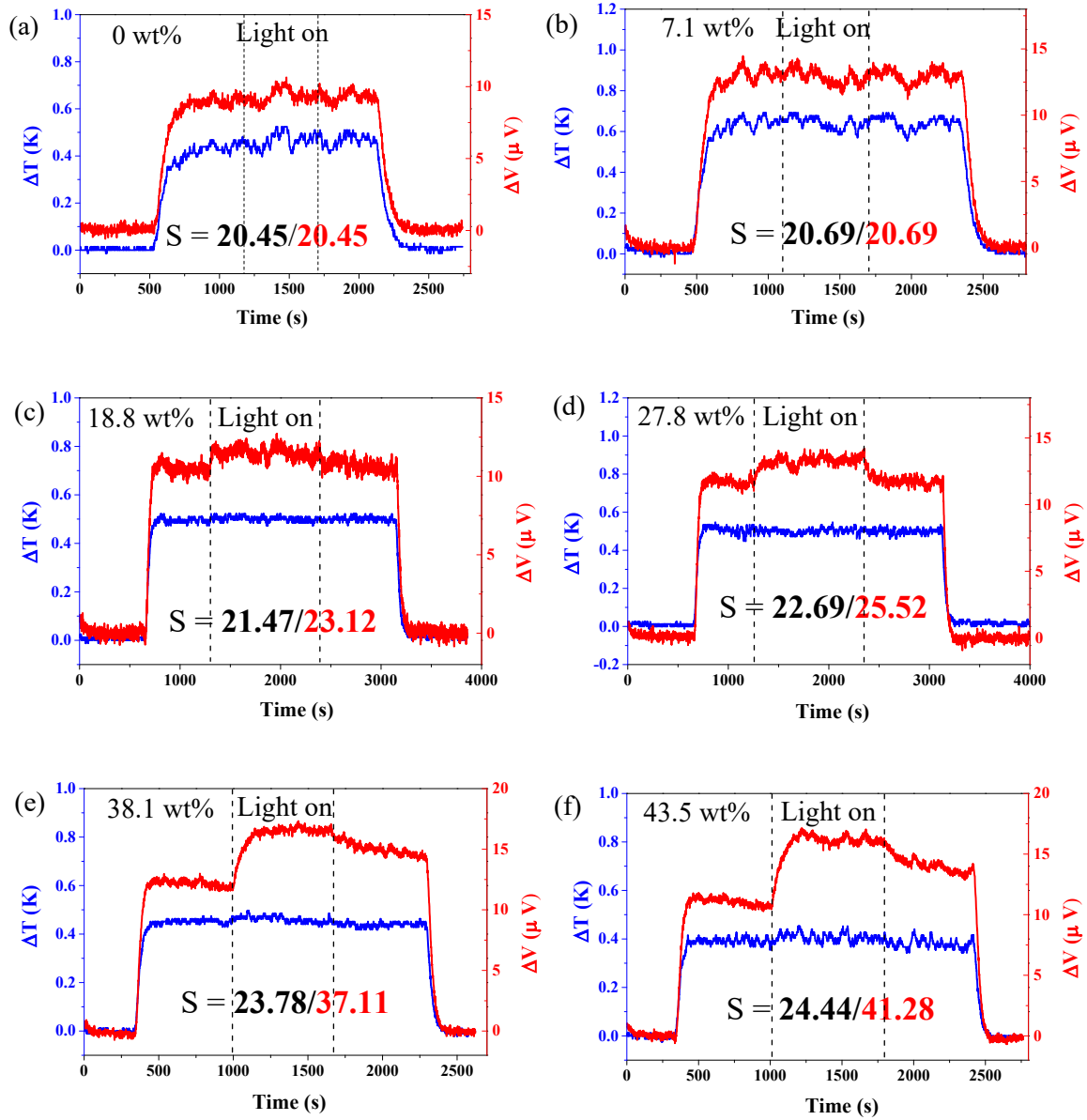
<sup>‡</sup> Binrong Li and Shizhong Yue contributed equally to this work.



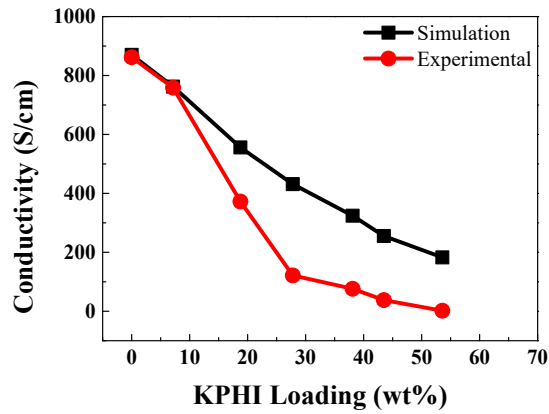
**Fig. S1.** (a) A holey framework of PHIK in the top view and (b) a stacked structure in the side view.



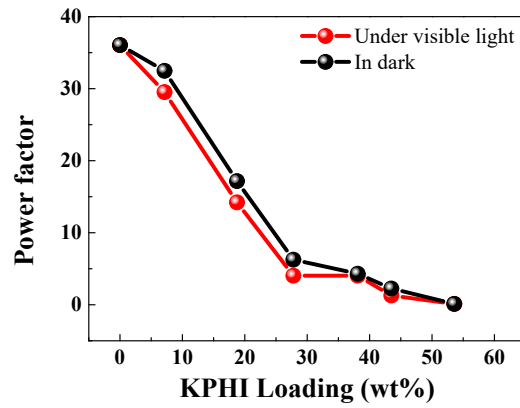
**Fig. S2.** Particle size distribution of an aqueous suspension of KPHI nanosheets by dynamic light scattering (DLS).



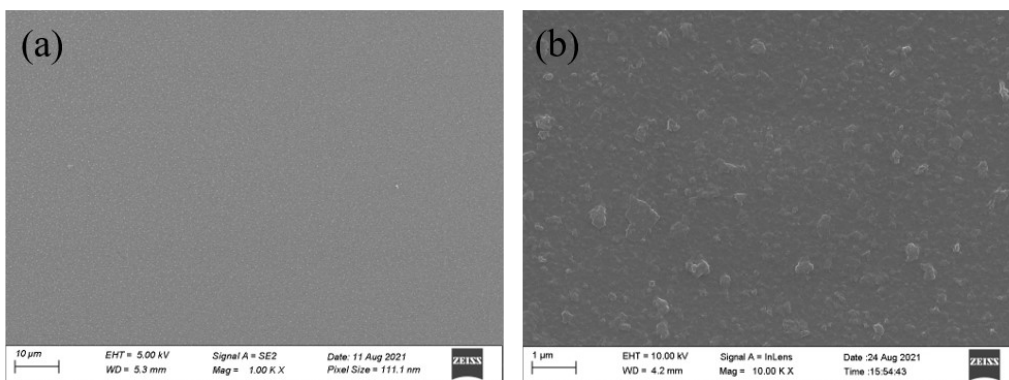
**Fig. S3.** The thermovoltage profiles of KPHI/PEDOT:PSS composite films with different KPHI loadings. The black and red Seebeck values ( $S$ ) are the ones in dark and under the visible light, respectively.



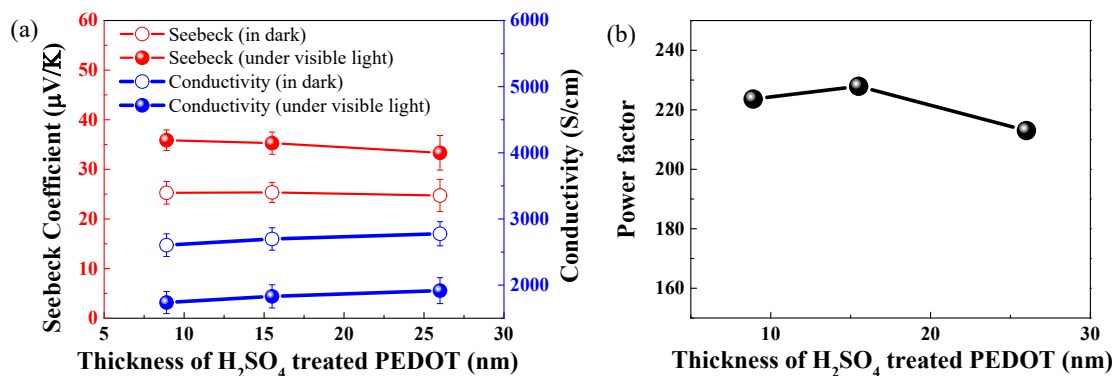
**Fig. S4.** Variations of the conductivity of KPHI/PEDOT:PSS composite films in dark by experiments and simulations.



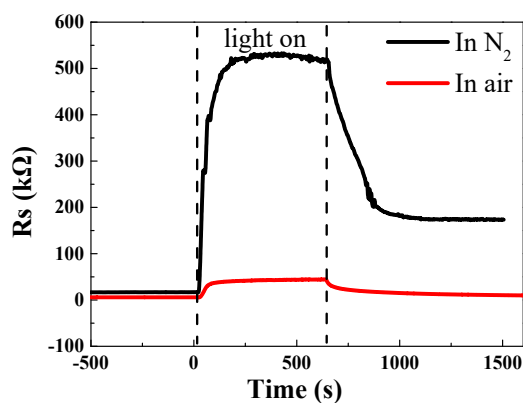
**Fig. S5.** Variations of the power factor of KPHI@PEDOT:PSS composites in dark or under the visible light with the KPHI loading.



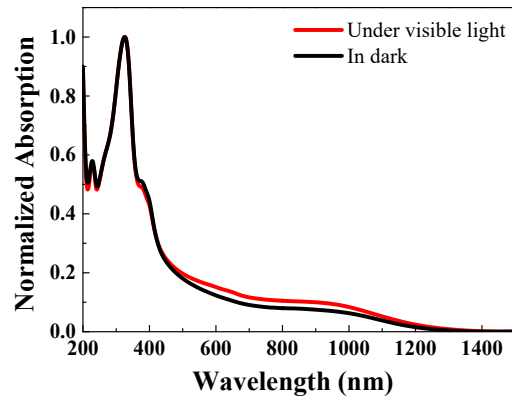
**Fig. S6.** SEM images of (a) the bottom PEDOT:PSS layer and (b) the top KPHI layer of a double-layer KPHI/PEDOT:PSS structure.



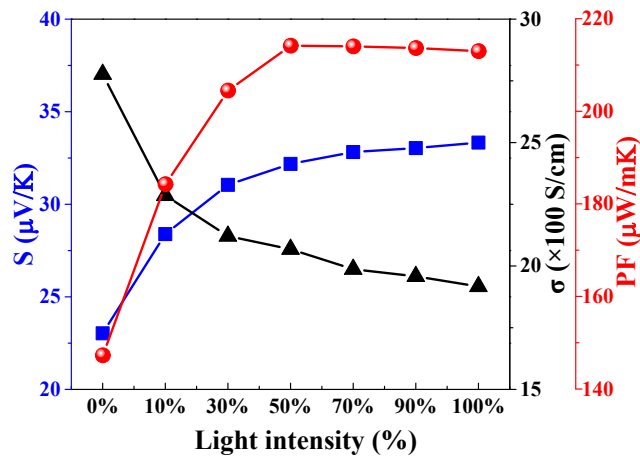
**Fig. S7.** Variations of (a) the Seebeck coefficient and conductivity and (b) power factor of the bottom PEDOT:PSS layer with the thickness of the PEDOT:PSS layer in the double-layer KPHI/PEDOT:PSS structures. A dispersion of 50 mg/L KPHI was used to coat the KPHI layer on the bottom PEDOT:PSS layer..



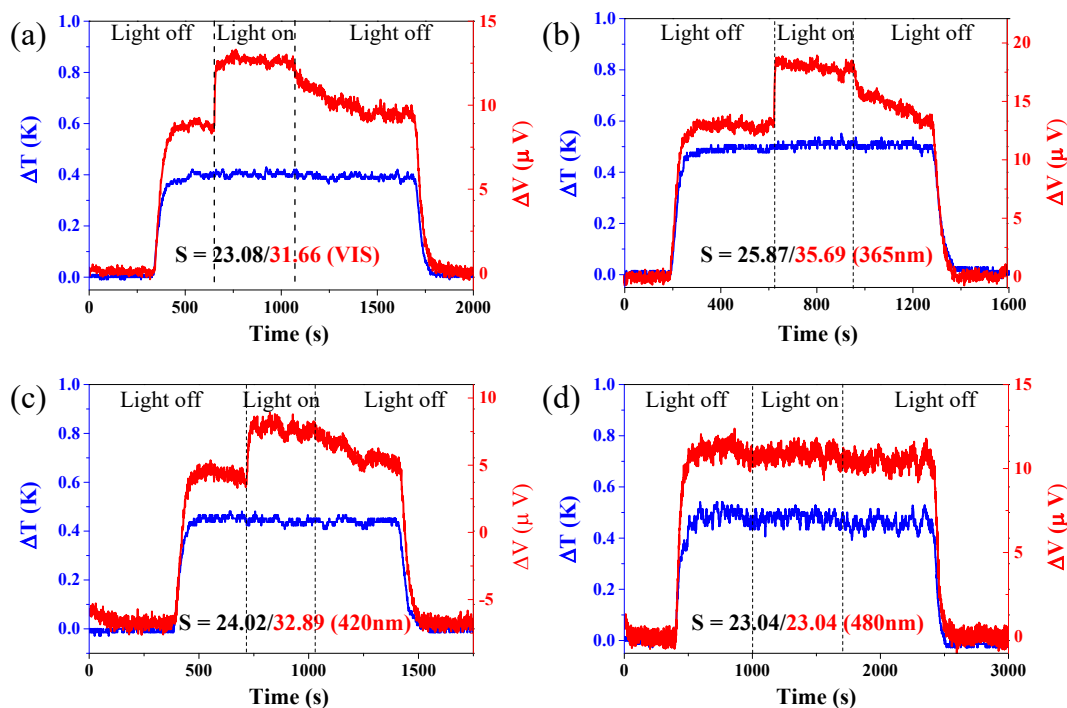
**Fig. S8.** The resistance evolutions of KPHI(53.6%)@PEDOT:PSS samples in  $\text{N}_2$  or in air.



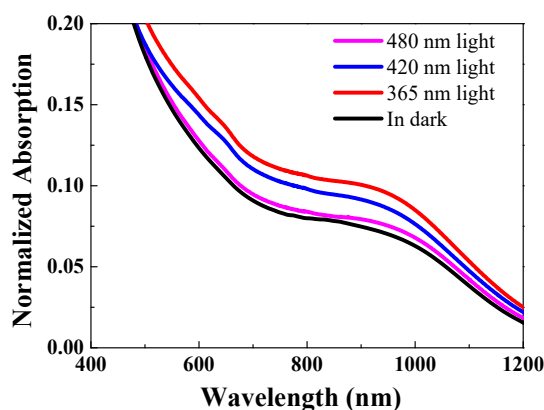
**Fig. S9.** UV-vis-NIR absorption spectra of KPHI(53.6%)@PEDOT:PSS before and after the visible light exposure.



**Fig. S10.** Dependences of the thermoelectric properties of a double-layer KPHI/PEDOT:PSS structure on the light intensity. The KPHI/PEDOT:PSS structure was exposed to the light with the wavelength of 420 nm. The intensity of the light was controlled by using optical filters.



**Fig. S11.** The thermovoltage profiles of double-layer KPHI/PEDOT:PSS structures exposed to (a) the visible light, (b) UV light of 365 nm, (c) violet light of 420 nm, and (d) blue light of 480 nm. The black and red Seebeck values (S) are the ones in dark and under the exposure of lights of different wavelengths, respectively. The KPHI layer was deposited from the dispersion of 50 mg/mL KPHI.



**Fig. S12.** UV-vis-NIR absorption spectra of KPHI(53.6%)@PEDOT:PSS without light exposure or exposure to the lights with the wavelengths of 365 nm (UV light), 420 nm and 480 nm, respectively.