Supporting Information

Highly foldable and flexible films of PEDOT:PSS/Xuan paper composites for thermoelectric application

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Experimental section

Materials. Commercial product of PEDOT:PSS aqueous dispersion (Clevios PH1000) was purchased from Heraeus Deutschland Gmbh. The concentration of PEDOT:PSS is 1.1 wt%, and the mass ratio of PSS to PEDOT is 2.5. Three types of Xuan papers, including raw Xuan, PiMade Xuan, and Sized Xuan, were bought from Yutai Xuan Paper Art Co., LTD. (Anhui, China). Dimethyl sulfoxide (DMSO) was purchased from Aladdin (Shanghai, China). All of the materials were used as received.

Preparation PEDOT:PSS/Xuan paper composite films. The PEDOT:PSS/Xuan paper (P/Xuan) composite films were prepared by a simple dip-coating and subsequent drying process. Typically, 1 mL DMSO was added into 19 mL PEDOT:PSS aqueous dispersion and stirred for 30 min at room temperature. The Xuan paper was coated with PEDOT:PSS components by immersing the Xuan paper into DMSO-treated PEDOT:PSS dispersion for 20 min. For comparison, the dip-coating process is respectively processed in atmosphere and vacuum conditions, respectively. The vacuum dip-coating process was performed in a vacuum oven (Shanghai Yiheng Scientific Instrument Co. Ltd) at room temperature. After the Xuan paper was completely immersed into DMSO-treated PEDOT:PSS dispersion, the vacuum degree was slowly increased to -100 kPa and then kept for 20 min. The PEDOT:PSS/Xuan paper composite films via both atmosphere and vacuum dip-coating were obtained by drying at 60 ^{o}C.

Characterizations of the structures and morphologies. The film morphologies were observed by field-emission scanning electron microscope (FESEM, FEI-APREO-S). Contact angle measurements were carried out using a contact angle measurement equipment (JY-82, Dingsheng, China). The tensile properties were performed by a Universal testing machine (Suns EUT4103, Shenzhen, China). The crosshead tensile speed is 10 mm min^{-1}. Fourier transform infrared (FTIR) spectra were collected over a range of 4000–700 cm^{-1} on a PerkinElmer Spectrum 3 (USA). The UV-vis-NIR

spectroscopy measurements were performed with a spectrophotometer (UV, Shimadu 2600). Thermogravimetric analysis (TGA, TA Q50) was performed under air atmosphere. The temperature ranged from room temperature to 800 ^{o}C, and the heating rate was 20 ^{o}C min^{-1}. Optical and polarized optical micrographs (POM) were obtained using an optical microscope (OM, Leica 2700) equipped with a rotatable polarizer and analyzer. The X-ray photoelectron spectrometry (XPS) analysis was carried out on a Thermo Scientific K-Alpha XPS spectrometer (USA).

Measurements of thermoelectric (TE) performance. The TE performance at room temperature, including in-plane Seebeck coefficients and electrical conductivity, were measured by a commercial Thin-Film Thermoelectric Parameter Test System (MRS-3RT, Wuhan Joule Yacht Science & Technology Co., Ltd.), where a quasi-steady-state mode was employed. The electrical conductivities of the folding test samples were measured by the commercial instruments Keithley 2000 Multimeter (Keithley Instruments Inc., USA). Composite thicknesses were measured using a thickness gauge, and the widths and lengths were measured by a caliper ruler.

Table S1 Mass ratios of PEDOT:PSS to Xuan paper (wt%) in the P/Xuan composite films.

Method	Raw Xuan	Sized Xuan	Pimade Xuan
Atmosphere	21.4±4.0	17.3±2.1	12.3±0.6
Vacuum	31.7±4.8	20.0±1.8	15.3±2.3

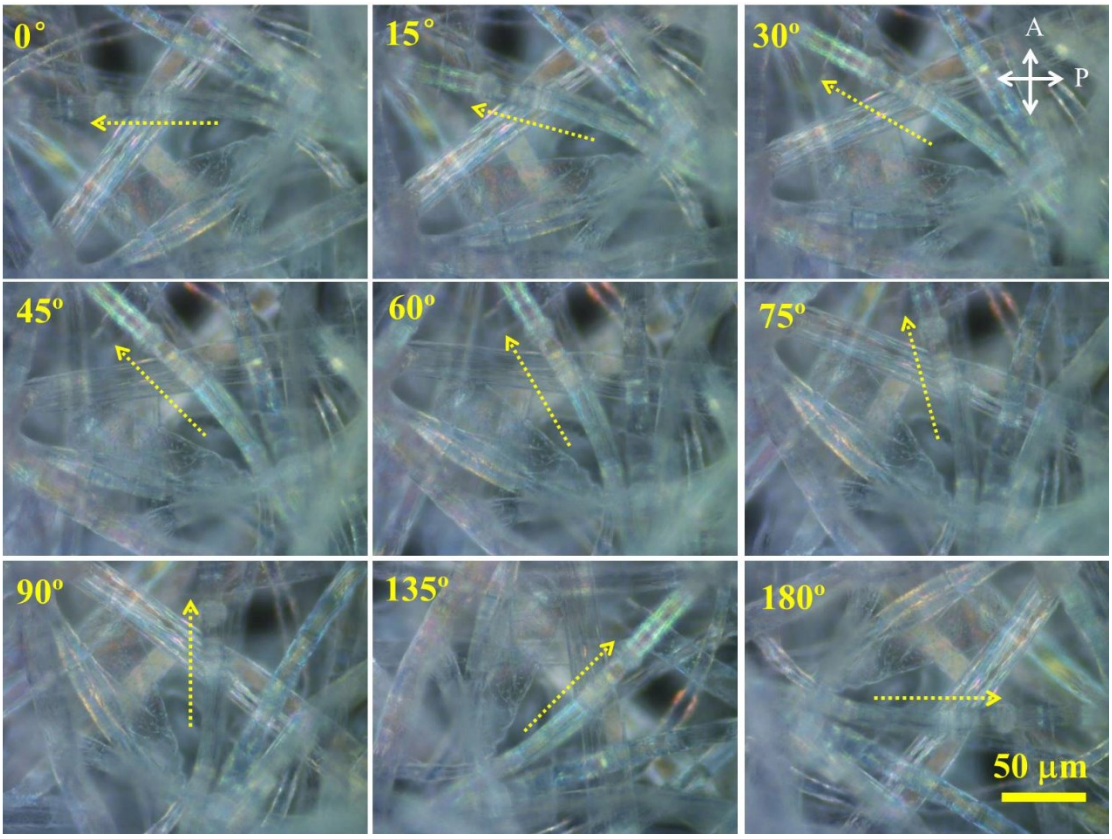

Fig. S1 POM micrographs of Raw Xuan, rotated between crossed polarizers. The scale bar in the bottom right micrograph represents 50 μm, and is applied to all other micrographs. The arrows indicate the rotating degree along clockwise direction, as shown in top left corner.

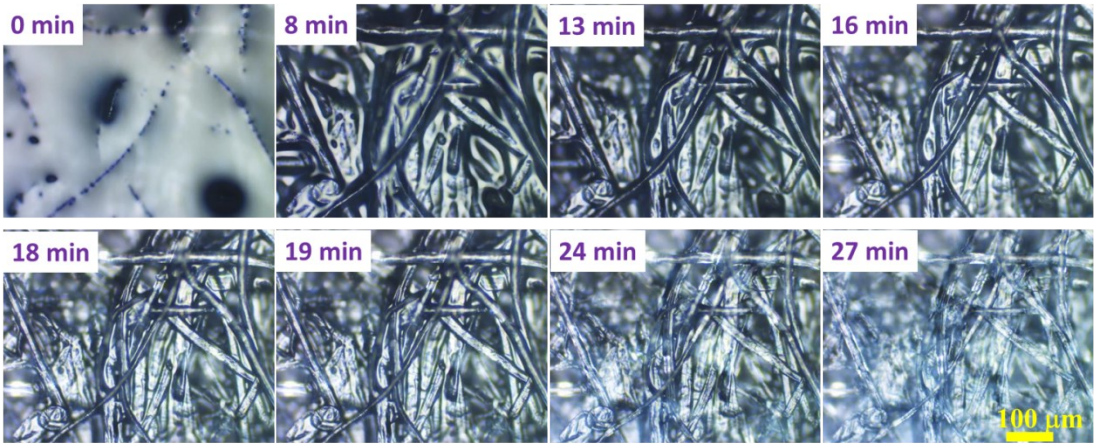

Fig. S2 Optical micrographs of the evolution process of PEDOT:PSS coating on the cellulose fibers of P/Raw-V composite film.

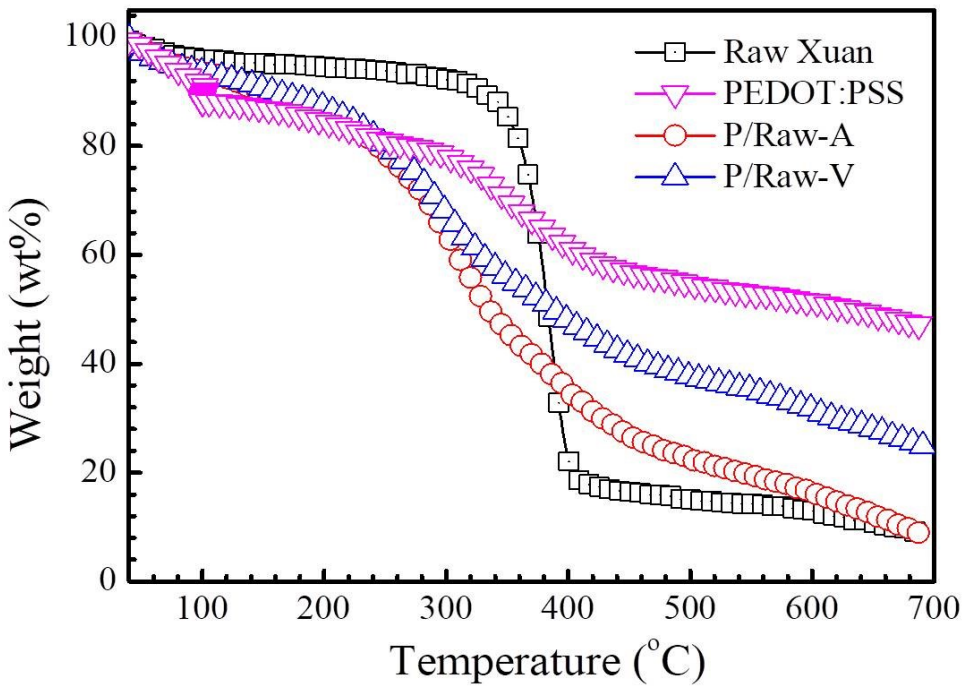

Fig. S3 TGA curves of Raw Xuan, P/Raw-A, P/Raw-V, and the neat PEDOT:PSS films.

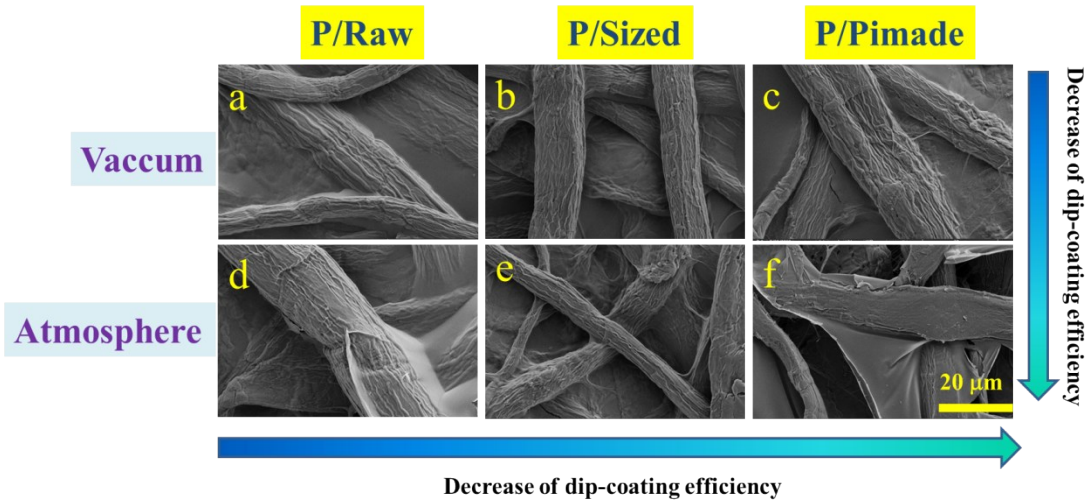

Fig. S4 FESEM images of (a, d) P/Raw, (b, e) P/Sized and (c, f) P/Pimade Xuan composite films by (a–c) vacuum and (d–f) atmosphere dip-coating processes.

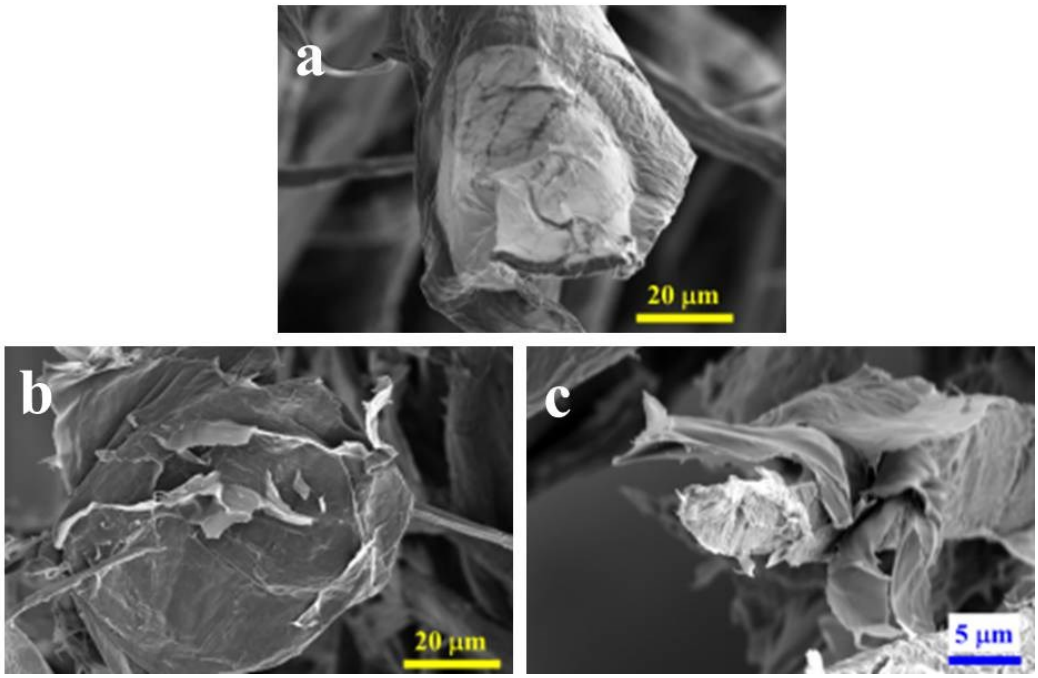

Fig. S5 SEM images of the cross-section of (a) Raw Xuan paper, (b) P/Raw-A and (c) P/Raw-V composite films after tensile tests.