Supporting Information

Humidity-Driven Bilayered Composite Nanofiber Textile Actuators for Smart Heat and humidity Management

Xi Yu,[a, c] Qiang Zhou,[a] Yingxin Zhao,[a] Guiying Xu,[a] Peimin Shen,[a] Yang Wang,*[, a, c] Chi-wai Kan,*[d] Hui Yu,[a, c] Lihuan Wang,[a, c] Jianhua Yan, [a, b] Xianfeng Wang [a, b]

2D AFM images of 6wt%PVDF membranes and 6wt%PVDF/2wt%FPU membranes (Fig. Sl); The peel strength between the hydrophilic and hydrophobic layers of the bilayered membranes (Fig. S2); Optical image of the bilayer membranes without window and with window (Fig. S3); Schematic diagram of the bilayered membranes fabrics promoting human sweat evaporation and cooling body (Fig. S4); Two-step fracture behavior of the 6wt% PVDF/2wt%FPU@16wt%PA bilayered membranes (Fig. S5).

Movie S1, impact dynamic behavior of a water drop on the 6wt% PVDF membranes;
Movie S2, impact dynamic behavior of a water drop on the 6wt% PVDF/2wt%FPU membranes;
Movie S3, humidity-responsive behavior of 16wt%PA @16wt%PA bilayered membranes in humidification processes;
Movie S4, humidity-responsive behavior of 8wt%PAN @16wt%PA bilayered membranes in humidification processes;
Movie S5, humidity-responsive behavior of 6wt% PVDF/2wt%FPU@16wt%PA bilayered membranes in humidification processes.

Experimental

Preparation of Polyamide Nanofiber Membranes

First, polyamide (PA) spinning solutions with different concentrations were prepared: 8g, 12g, 16g, and 20g of PA pellets were dissolved in mixed solvents of ethanol (80.5g, 77g, 73.5g, 70g) and deionized water (11.5g, 11g, 10.5g, 10g), respectively. The mixtures were stirred at a constant temperature below 55°C until complete dissolution to obtain 8wt%, 12wt%, 16wt%, and 20wt% PA spinning solutions.

Electrospinning was conducted in a constant humidity environment (90% RH) with the following parameters, the injection rate was set at 1.5 mL/h, the collection distance was set at 15 cm, the applied voltage was set at 30 kV, and the moving distance of the stage was set at 10 cm. The spinning solution was loaded into a 10 mL syringe and electrospun onto a silicon oil-coated paper substrate mounted on a rotating drum collector (50 rpm). Uniform nanofiber membranes with a thickness of approximately 100 μm were obtained.

Preparation of Hydrophobic Nanofiber Membranes

Four hydrophobic polymer systems were selected for electrospinning: polyacrylonitrile (PAN), aramid 1313 (PMIA), polyvinylidene fluoride (PVDF), and PVDF/FPU core-shell material. The spinning solutions were prepared as follows: PAN (8wt%), PVDF (6wt%), and PVDF/FPU (6wt%/2wt%) were dissolved in N,N-dimethylacetamide (DMAc), PMIA (10wt%) was dissolved in a mixed solvent of 85wt% DMAc and 5wt% lithium chloride. The hydrophobic layers were electrospun in situ onto PA nanofiber substrates.

For PAN, PMIA, and PVDF membranes, spinning was performed at 30-40% RH with the different parameters. For PAN and PVDF, the collection distance was set at 15 cm, the stage translation was set at 10 cm, the voltage was set at 30 kV, the injection rate was set at 1 mL/h. For PMIA, the collection distance was set at 10 cm, the voltage was set at 20 kV, the injection rate was set at 1.5 mL/h. All samples used a drum speed of 50 rpm and stage speed of 155 cm/min. For PVDF/FPU core-shell membranes, electrospinning was conducted at 80-90% RH, the collection distance was set at 15 cm, the moving distance of the stage was set at 7 cm, the voltage was set at 15 kV, with

injection rates of 1 mL/h (PVDF) and 0.35 mL/h (FPU). All hydrophobic layers had a controlled thickness of ~100 μm.

Moisture Permeability Testing

The WVT rate of membranes was tested with YG 601H tester (China) to reflect their moisture transmission capacity, and the test condition was kept at 50%RH and 38 °C, with the value calculated by measuring the change in water vapor mass passing through the sample per unit time in the moisture transmission cup. For single-layer PA membranes, 34g water was firstly added to test cups, and membrane samples were cut to match cup openings and sealed, then the initial mass was recorded after 1h equilibration, and final mass was measured after 2h testing. For bilayered membranes (PA/PAN, PA/PMIA, PA/PVDF, PA/PVDF-FPU), non-windowed and windowed specimens were tested. The non-windowed specimens mimic the moisture permeability of the surface of the skin after perspiration. And the bilayered membranes with window mimic the moisture permeability of the skin surface after the bilayered membrane is bent in response to moisture. The moisture permeability of a non-window specimen is tested using a complete membrane, while a windowed sample needs to be cut along the three sides of the rectangle with a cutter to form an opening before the test. Specific testing process follows the same protocol as above.

Material Characterization

The PA nanofiber membranes with different concentrations, PAN, PMIA, PVDF nanofiber membranes, and PVDF/FPU core-shell nanofiber membranes were first sputter-coated with gold and then the surface morphology was observed by using scanning electron microscopy (SEM). The surface topography and properties of individual PVDF nanofibers and membranes, individual PVDF/FPU core-shell nanofibers and membranes were characterized by atomic force microscopy (AFM). Fourier transform infrared spectroscopy (FTIR) was performed on eight types of fiber membranes: PA, FPU, PVDF single membranes, PA/PAN, PA/PMIA1313, PA/PVDF bilayered membranes, PA/(PVDF/FPU) core-shell bilayered membranes. The spectral range was set from 600 to 4000 cm^{-1} with 64 scans and a resolution of 4 cm^{-1}. For bilayered membrane testing, we only analyzed the hydrophobic side. The pore size

distribution of polyamide nanofiber membranes with different concentrations was measured using a porous material pore size analyzer. The samples were placed in a petri dish, wetted with PMI solution, then clamped with tweezers and completely covered over the air-permeable aperture. The sample thickness was input before testing.

Mechanical properties of various nanofiber membranes (PA of different concentration, PAN, PMIA1313, PVDF, and PVDF/FPU core-shell membranes) were tested using a fiber tensile tester. The membranes were first cut into 3mm wide strips, then clamped and stretched until breaking. Twenty parallel samples were tested for each group. The peel strength between the hydrophilic and hydrophobic layers of the bilayered membranes was tested by tensile tester, and the peel strength P was calculated by the the formula $P = \frac{F}{W}$ (where F denotes the average peel force in cN and W represents the sample width in cm). Water contact angles of PA nanofiber membranes of different concentration and four types of bilayered membranes were measured using a contact angle goniometer. Samples were fixed on glass slides and placed on the instrument stage, with proper adjustment of the stage-to-syringe distance. The sessile drop method was employed, where 3μL droplets were dispensed while recording video. Contact angles were measured from captured images. Each sample was tested three times, with the average value reported.

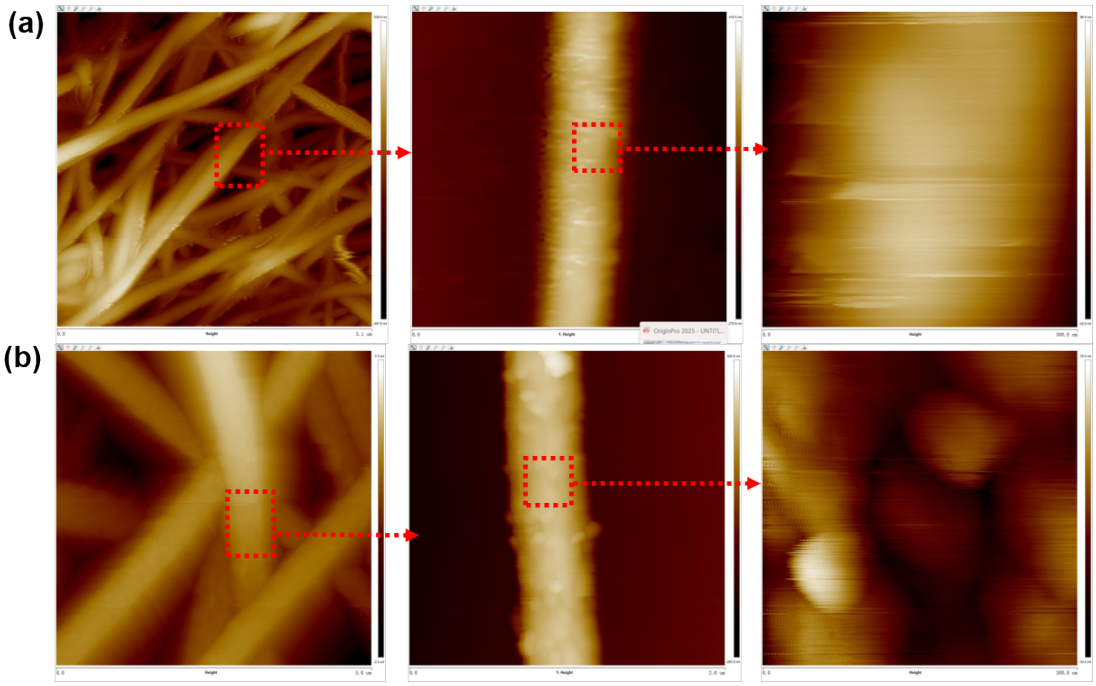

Fig. S1. (a) 2D AFM images of 6wt%PVDF membranes, (b) 2D AFM images of 6wt%PVDF/2wt%FPU membranes.

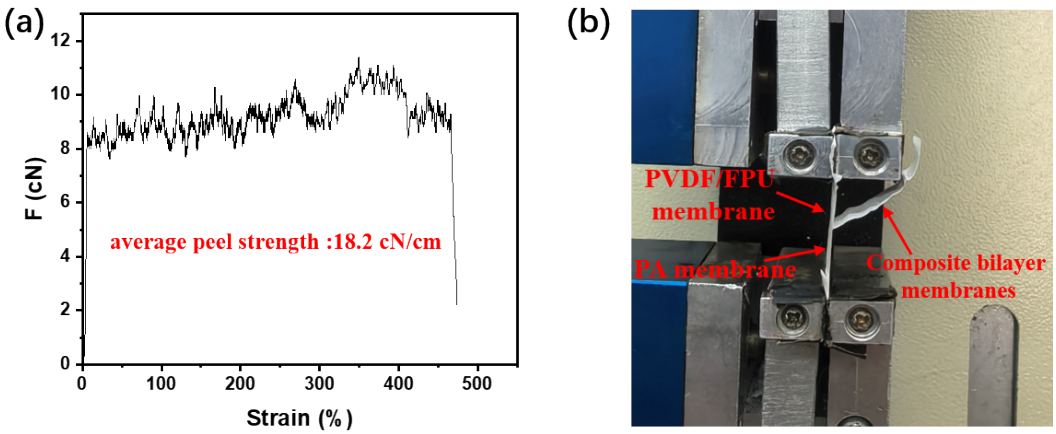

Fig. S2. (a) Peel curve of PA/PVDF/FPU composite membrane, (b) Schematic diagram of peel test for PA/PVDF/FPU membrane

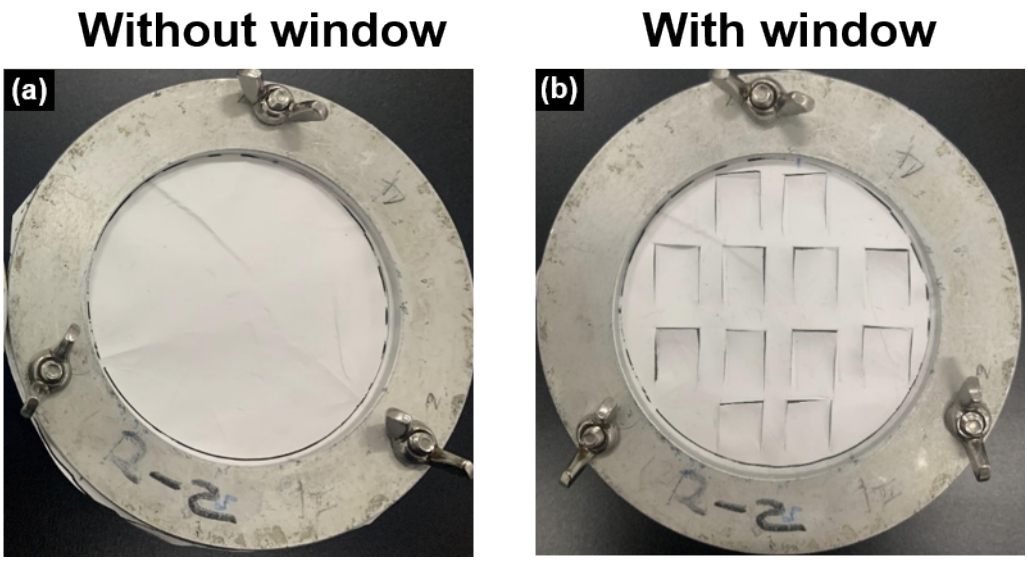

Fig. S3. Optical image of the bilayer membranes (a) without window and (b) with window.

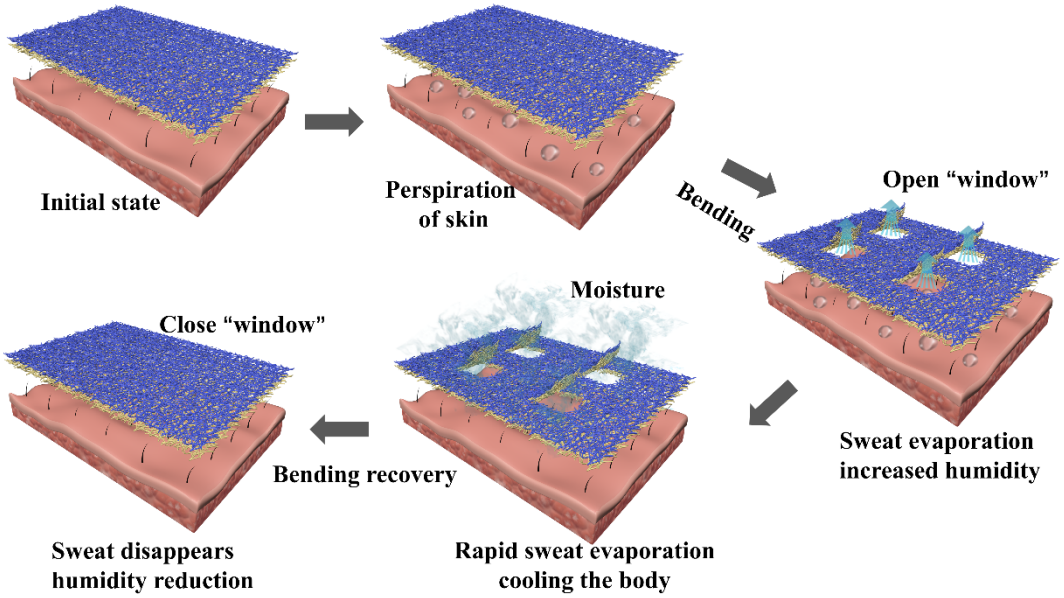

Fig.S4. Schematic diagram of the bilayered membranes fabrics promoting human sweat evaporation and cooling body.

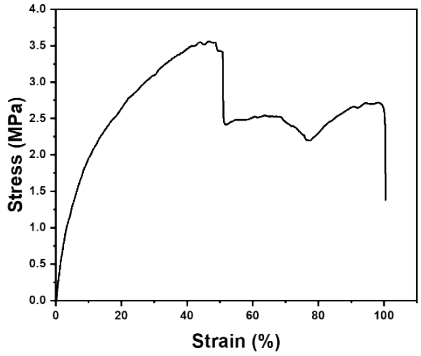

Fig.S5. Two-step fracture behavior of the 6wt% PVDF/2wt%FPU@16wt%PA bilayered membranes.