Supplementary information for

Cellulose Nanocrystal Composite Films for Contactless Moisture-Electric Conversion

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Fig. S1. SEM image of the composite film with varying PVA content: (a) 20% PVA, (b) 30% PVA, (c) 40% PVA, and (d) 50% PVA.



Fig. S2. (a) Fourier infrared spectrum (FTIR) analysis of the CNC, PVA and composite film. (b) Water contact angle on CNC and PVA films, respectively.

As shown in Fig. S2a, the infrared spectra reveal several characteristic peaks. For the CNC, a broad peak near 3333.15 cm⁻¹ indicates the presence of hydroxyl (-OH) groups. The symmetric stretching vibration absorption peak of -CH- appears at 2905.54 cm⁻¹. C-H bending vibration is observed at 1432.58 cm⁻¹. The S=O stretching vibration peak, derived from sulfuric acid hydrolysis and grafted onto CNC, is present at 1316.67 cm⁻¹. The stretching vibration of the C-C skeleton is detected at 1166.83 cm⁻¹. Ether C-O stretching vibration appears at 1108.87 cm⁻¹. A strong S-O stretching vibration peak of the sulfonic acid group is evident at 1033.95 cm⁻¹.

For the PVA, the -OH stretching vibration absorption peak is located at 3309.83 cm⁻¹. The C-H symmetric stretching vibration peak of methylene (-CH₂-) appears at 2922.50 cm⁻¹. The C-H bending vibration peak is at 1720.95 cm⁻¹. Additional C-H bending vibration peaks are observed at 1426.93 cm⁻¹ and 1316.67 cm⁻¹. The C-O bond stretching vibration peak is prominent at 1050.91 cm⁻¹. While in the CNC/PVA composite film, the -OH stretching vibration peak redshifts to 3304.17 cm⁻¹, indicating hydrogen bond formation between CNC and PVA. The symmetric C-H stretching vibration peak appears at 2916.85 cm⁻¹, intermediate between those of CNC and PVA. C-H bending vibration peaks are observed at 1646.03 cm⁻¹ and 1472.87 cm⁻¹. The C-O bond stretching vibration peak is detected at 1062.92 cm⁻¹, also intermediate between CNC and PVA. The characteristic sulfonic acid group peaks in CNC are preserved, with S=O and S-O stretching vibrations at 1311.01 cm⁻¹ and 1033.94 cm⁻¹, respectively.

These results confirm the physical mixing of CNC and PVA within the composite, with the

sulfonic acid group (-SO₃H) contributing mobile protons (H⁺), critical for the material's properties. The abundant hydroxyl groups (-OH) in PVA enhance the composite's hydrophilicity and improve the film flexibility.



Fig. S3. Influence of the size of the film on the electric output.

Fig. S3 shows that the composited film area minimally affects the voltage signal intensity. The film generates an electric signal as ions dissociate within the material and migrate along the concentration gradient created by unidirectional moisture absorption. Electrodes on either side harness the potential difference, producing a stable voltage signal. The sensor operates as a network of miniature electricity-generating units connected in parallel. According to Ohm's law, the voltage in a parallel configuration matches that of a single unit. Thus, increasing the composite film area does not enhance voltage output.



Fig. S4. Electric output of the composite film at varying CNC/PVA mass ratios (a) and under

different humidity conditions (b).



Fig. S5. Asymmetric and symmetrical electrode configurations. (a) Asymmetric distribution: The upper silver electrode is smaller than the lower conductive carbon strip, ensuring the lower side is sealed and the upper side absorbs moisture, creating a water content gradient within the film. (b) Symmetric distribution: The upper silver electrode matches the size of the lower conductive carbon strip, allowing uniform moisture absorption on both sides, resulting in no water content gradient inside the film.



Fig. S6. Photographs of the experiment visualizing dynamic H^+ diffusion: A CNC/PVA film was placed between two glass tubes containing a methyl orange/ethyl alcohol solution. Upon adding 5 mL of water to the upper cylinder, the color changes in the tubes reflected the dynamic variations in H^+ concentration.



Fig. S7. Photos of the respirator mask used in the experiment. The device is composed of three main components: an upper silver electrode, a middle composite film layer, and a lower conductive carbon tape. When attached to the mask's inner layer and connected to an electrochemical workstation via a conductive clip, the device detects biological signals generated by breathing.