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Supporting Information

Hydrophilic Carbon Quantum Dots Assisting Porous P(VDF-HFP) film for Self-

powered Humidity Sensing with High Sensitivity and Low Hysteresis

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1. Experimental section

1.1 Synthesis of carbon dots

Citric acid (3.6 g), urea (1.5 g) and solvent (25 mL, H₂O or DMF) were heated at 200 °C for 12 h, and then cooled down to room temperature. The reacted solution was processed by centrifugation at 10000 rpm for 30 min, suction filtration with 0.22 μm membranes and dialysis with 1000 Da hydrolysis membrane for 24 h. The purified CDs solution was then freeze-dried into dark powder for late use.

1.2 Preparation of piezoelectric films

3 mg CDs was added into DMF (2 mL), and then stirred magnetically for 2 h to get CDs/DMF dispersion. PVDF-HFP (0.6 g), acetone (3 mL) and the CDs/DMF dispersion were stirred magnetically for 4 h to obtain PVDF-HFP/CDs solution. The solution was then spin-coated on the ITO-PEN electrode (20 mm \times 20 mm) at the rotating speed of 800 rpm for 60 s. After the solvent evaporated, the obtained P-Z-C film was annealed at the temperature of $120 \degree C$ for 1 h.

1.3 Assembly of flexible piezoelectric devices

The piezoelectric device was composed of a sandwich structure of the piezoelectric film attached to the bottom ITO-PEN electrode and a top porous copper foil. The top copper foil (20 mm \times 15 mm) was physically attached on the piezoelectric film to form an effective area of 15 mm \times 15 mm. The part of the piezoelectric film not covered by the top electrode was wiped with acetone to serve as a lead. The entire device was encapsulated with 3M polyimide tape.

1.4 Characterization and Measurement

Transmission electron microscopy (TEM, JEM-2100, JEOL) and scanning electron microscope (SEM, EVO MA10, Zeiss) were used to observe the morphologies of the CDs and films. The structural properties of the CDs and films were investigated by Fourier transform infrared spectroscopy (FT-IR, Nicolet IS50, Thermo Scientific), X-ray photoelectron spectroscopy (XPS, Nexsa, Thermo Scientific) and X-ray diffraction (XRD, D8 advance, Bruker). The contact angles of the films were tested by Theta Flex from Biolin. The dielectric constant of the films were determined using a broadband dielectric impedance spectrometer (concept 80, novocontrol). The piezoelectric properties of the devices were explored by an electromechanical platform from the Institute of Nano Science and Technology of the Chinese Academy of Sciences, an oscilloscope (TBS 1202B, Tektronix) and a charge-voltage converter (VK102, Guangdong Weijingyi).

2. Supporting figures

Fig. S1 TEM images of (b) C1 and (c) C2.

Fig. S2 Particle size distributions of (a) C1 and (b) C2

Fig. S3 SEM images of (a) PC0, (b) PC1 and (c) PC2.

Fig. S4 Pore size distributions of (a) PC0, (b) PC1 and (c) PC2.

Fig. S5 TEM images of (b) PC1 and (c) PC2.

Fig. S6 XPS high-resolution scans in O 1s of (a) C1 and (b) C2.

Figure S7 Real-time output voltages of (a) PC0, (b) PC1 and (c) PC2 under different forces, (d) applied force-voltage curves of the films

Fig. S8 Real-time output voltages of PC2 under different (a) forces, (b) frequencies and (c)

temperatures

Fig. S9 (a) Durability and (b) long-term stability of PC2

Figure S10 Real-time output voltages of (a,b,c) PC0, (d,e,f) PC1 and (g,h,i) PC2 under different ambient humidity

Fig. S11 (a) Applied force-current curves of the films, RH-current curves of (b) PC0, (c) PC1 and (d)

PC2

Fig. S12 RH-voltage curves of PC2 under (e) 5N, (f) 10N and (g) 15N

Fig. S13 Real-time output voltages of the applicable humidity sensor.

3. Supporting tables

Sample		C 1s (284.8 eV) N 1s (399.8 eV) O 1s (531.5 eV)	
C ₁	0.5868	0.1031	0.3101
C2	0.6302	0.1447	0.2251

Table S1 Element contents of CDs

Table S2 Bond contents of N 1s

Sample		$N-(C)_2$ (399.4 eV) $N-(C)_3$ (400.0 eV) $N-H$ (401.0 eV)	
C ₁	0.4555	0.3060	0.2385
C2	0.4755	0.4150	0.1095

Table S3 Bond contents of O 1s

Sample	$C=O(531.3 \text{ eV})$	$C-O (532.7 eV)$
C1	0.6226	0.3774
\mathcal{C}	0.6174	0.3826