Supplementary Information

Synthesis of Coordination Polymer Thin Films with Conductance-Response to Mechanical Stimulation

MATERIALS AND METHODS

Materials. Copper foils (purity of 99.9%, 0.1 mm in thickness), Potassium ferricyanide $(K_3[Fe(CN)_6] (AR, \ge 99.5\%))$, hydrochloric acid, acetic acid and Copper chloride dehydrate were purchased from Sinopharm Chemical Reagent Co.,Ltd without further purification.

Methods.

Cleaning procedure for copper substrates. All of the copper substrates were washed twice by sonication in 0.5 M hydrochloric acid, deionized water and acetone for 5 min alternately. The copper substrates were dried in air at the temperature of 40 °C.

Preparation of the copper oxide layer. The copper oxide layers were synthesized via calcination of copper substrates in air at the temperature of 300 °C for 50 min. The obtained copper oxide films were rinsed with deionized water and ethanol for six times alternately, and dried under vacuum at the temperature of 40 °C.

Preparation of the KCu[Fe(CN)₆] • **5H**₂**O films.** In brief, 0.25 mmol of K_3 [Fe(CN)₆] was dissolved in 50 mL of distilled water to form a clear solution first. After that, 0.1 mol/L of acetic acid (1 mL) was added to the solution. Then, the as-prepared copper oxide films were immersed in the solution. The solution was aged at 40 °C for 120

min. Finally, the films were washed alternately with pure water and ethanol 6 times, and dried at room temperature.

Preparation of the KCu[Fe(CN)₆] • 5H₂O powders. 700 mg of CuCl₂ was dissolved in 200 mL of pure water to form clear solution A. K₃[Fe(CN)₆] (1.32 g) and 8 mL of 0.1 mol/L acetic acid were dissolved in another 200 mL of pure water to form clear solution B. The solution A was mixed with the solution B under stirring for 5 min, then aged at room temperature for 36 hours. Then, the sample was collected by centrifugation, and washed alternately with deionized water and ethanol for several times. Finally, the samples were dried at room temperature for subsequent operation.

Device assembly operation. In brief, one piece of Cu sheet deposited with PBA thin film was cut into two square pieces with side lengths of 2 cm. The two pieces were assembled by touching the sides with PBA thin films in a face-to-face fashion. The copper sides were connected to an electrochemical station with a bias potential of 1 V. To make sure the thin films were fixed tightly, a pressure of 0.01 MPa was applied between two pieces. We should note that the pressure do not alternate the recorded current value because no significant current can be detected if the external pressure is less than 0.5 MPa.

Pressure tests. A bias potential of 1 V was applied to the assembled device. The applied pressure was changed from 0 to 12 MPa during the test. The current intensity was recorded under corresponding pressures.

Characterization. The SEM observation was performed by using a Hitachi 4800 FESEM. Wide-angle PXRD patterns were obtained with a Rigaku Smartlab

diffractometer using a monochromated Cu K α radiation (40 kV, 40 mA) at a scanning rate of 1° min⁻¹. The AFM characterization was performed on a Cypher ES AFM (Asylum Research). Commercial rectangular silicon cantilevers (OMCL-AC160TS-R3; Olympus) with a spring constant of 26 N/m was used for mechanical property measurements in AM-FM mode. Ti/Ir coated silicon tips (ASYELEC-01-R2; Asylum Research) with a spring constant of 2.8 N/m was used for conductivity measurements.The i—t curve measurements were performed on a CHI electrochemical workstation (CHI, 660E).



Fig. S1. The experimental scheme flow chart.



Fig. S2. SEM images of the copper sheets after annealing in air at 300 °C for 50 min.



Fig. S3. Cross-section SEM image of the Cu₂O film.



Fig. S4. XRD patterns of the copper sheet (a) before and (b) after surface oxidization.



Fig. S5. (a) Nitrogen adsorption-desorption isotherms of the PBAs powder. (b) Pore-size distribution calculated by the non-local density functional theory (NLDFT) method.



Fig. S6. (a) Nitrogen adsorption–desorption isotherms of the PBAs film. (b) Pore-size distribution of the PBAs film.



Fig. S7. SEM image of the thin films before (a) and after (b) detaching.



Fig. S8. Time-course monitoring of the coordination replication process by XRD.



Fig. S9. Top-view SEM images (**a-e**) of the PBA films after a conventional hydrothermal treatment at 40 °C for 120 min with different concentrations of the $K_3[Fe(CN)_6]$. (a) 2.5 mmol/L, (b) 5 mmol/L, (c) 7.5 mmol/L, (d) 10 mmol/L and (e) 12.5 mmol/L.



Fig. S10. XRD patterns of the PBA films after a conventional hydrothermal treatment at 40 °C for 120 min with different concentrations of the K_3 [Fe(CN)₆].



Fig. S11. (a) Height image and (b) current maps of the PBA films.



Fig. S12. The optical photos of the PBAs thin films after bending.



Fig. S13. The relationship between the maximum current (I_{max}) and the external pressure. The formula can be simulated as $I_{max} = 9.62 \times 10^{-4} \text{ P} + 1.57$, $R^2 = 0.97$ (P \geq 0.5 MPa).



Fig. S14. The *I-t* curves of the PBA films under different pressures. A bias potential of 1 V was applied during the test.



Fig. S15. The relationship between the maximum current (I_{max}) and the external pressure after repeating for multiple times. $I_{max} = 8.84061 \times 10^{-4} P+ 2.24284$, $R^2 = 0.978$.



Fig. S16. XRD patterns of the PBA films before and after pressure test.



Fig. S17. (a) Scheme for the conductance test of the PBA powders under external pressure. (b) The relationship between the external pressure and the maxima current intensity (I_{max} -t curve). The potential bias was 1 V.



Fig. S18. The relationship curves of (a) electronic current (I_e) with different applied pressures, $I_e = 3.8 \times 10^{-4} P + 0.89214$, $R^2 = 0.9734$. (b) Ionic current (I_i) with different applied pressures, $I_i = 9.2 \times 10^{-4} P + 0.93506$, $R^2 = 0.9871$. During the pressing test, the applied constant voltage was 1 V.



Fig. S19. The effect of external pressure on the contact area of the PBA films.



Fig. S20. (a, b) The optical photos and (c) SEM image of the PBAs thin films which were grown on non-flat substrates.