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Supporting information for:

Polymerizable Deep Eutectic Solvent (PDES) Type Conductive Paper for Origami 3D Circuits

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EXPERIMENTAL SECTION

Materials Choline chloride (CCl, 98%, Shanghai Macklin Biochemical Co., Ltd), acrylic acid (AA, >99%, Macklin) and 2-hydroxy-4-(2-hydroxyethoxy)-2-methylpropiophenone (photoinitiator 2959, ≥98%, Tianjin Jiuri New Materials) were used as received.

Preparation of polymerizable deep eutectic solvent (PDES). CCl as ammonium salt should be dried under vacuum at 60 °C for 2 h, and AA as hydrogen bond donor molecules were dried over 4 Å molecular sieves before use. CCl and AA were mixed with different mole ratios. Then, the mixture was heated and stirred at 90 °C in a closed flask for around 4 hours until the appearance of a homogenous colorless solution. The prepared PDES was then kept in a vacuum desiccator with the silica gel for further use.

Fabrication of raw paper. The raw paper was made according to TAPPI (Technical Association of the Pulp and Paper Industry) standard method T205. The detailed papermaking procedure is provided elsewhere.¹

Fabrication of conductive paper. A typical procedure used as follows: PDES (10g) and photoinitiator 2959 (0.2g) were mixed thoroughly under ultrasonication. Then, the mixtures were transported onto the screen plate (mesh number = 240) and coated by a screen printing machine (blade angle = 60° , speed = 6 m/min, Shenzhen Kaimaojx factory, KM-SY3050). Finally, the reactions of coated paper were initiated by a UV source (RW-UVA- Φ 200U, Shenzhen Runwing Company, China) with a dominant wavelength of 365 nm for 10 seconds. The light intensity was 20 mW cm⁻² measured by a UV radiometer (type UV-A, Photoelectric Instrument Factory, Beijing Normal University).

Measuring the electrochemical properties. The coated paper (sandwiched by copper tapes) were measured by PGSTAT 302N (Princeton Applied Research) through the electrochemical impedance

spectroscopy (EIS) method. The applied frequency range in the electrical tests was from 0.01 to 10^5 HZ, the relative humidity was 35%, and the temperature was 25 °C.

Characterizations. Scanning electron microscope (SEM) images were obtained by using the HITACHI TM3030 Tabletop SEM. Fourier transform infrared (ATR-FTIR) spectra were recorded on a Bruker Vertex 33 spectrometer. Differential scanning calorimeter (DSC) was employed a 214 polyma NETZSCH tester. The PDES (CCI: AA = 1:2) was placed into aluminium pans and heated at 10 °C min⁻¹ from -150 to 120 °C under a nitrogen atmosphere. ¹H NMR spectra (400MHz) were tested using a Bruker spectrometer AVANCE III HD 400. Chloroform (CDCl₃) was used as external reference. The tensile testing was performed using a tensile machine (INSTRON 5565, 500N load cell). The samples were cut into 4×2 cm². Optical images were taken by a Nikon Digital Sight DS-Fil camera.



Figure S1. The physical and chemical properties of PDES. (a) The digital photography of PDESs with different mole ratios of CCI:AA. The top line (i) shows the initial condition at 90 °C. The bottom line exhibits the appearances after stored for 24 hours at room temperature. (b) Differential scanning calorimeter (DSC) and (c) nuclear magnetic resonance spectra (¹H-NMR) of as-prepared PDES. (d) Fourier transform infrared spectra (FTIR) of as-prepared PDES and its individual components. The mole ratio of CCI:AA is 1:2 for the PDES in (b-d).



Figure S2 Comparison of set width (solid grey) and actual printed width (shallow grey) of printed PDES lines on the paper. The blue curve is the dependence of diffusion extent of printed PDES lines on the pre-designed line width.

The diffusion extent can be calculated as below equation:

$$R_{diffusion} = \frac{W_{actual} - W_{set}}{W_{set}} \times 100\%$$



Figure S3 The cross-sectional scanning electron microscope (SEM) image of PDES type conductive paper.