Supporting Information

High-Performance Solution-Processable Flexible and Transparent Conducting Electrode with Embedded Cu Mesh

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Experimental Section

Synthesis of Cu flake particles

Cu acetate ($Cu(CH_3COO)_2$, 98%), 1,3-dichlorobenzene ($C_6H_4Cl_2$, 99%), octylamine ($C_8H_{17}NH_2$, 99%), oleic acid ($C_{18}H_{34}O_2$, 90%), phenyl hydrazine ($C_6H_5NHNH_2$, 97%) and toluene ($C_6H_5CH_3$, anhydrous, 99.8%) were purchased from Sigma Aldrich. All chemicals were used without further purification. For synthesizing Cu particles, Cu acetate (38.9 g) and oleic acid (32.5 g) were dissolved in a solvent mixture of octylamine and dichlorobenzene (159.2 g) in a three-neck round-bottomed flask. Then, phenyl hydrazine (81.9 g) was added at an elevated temperature. The reactant mixture was heated to 150 °C and stirred with a magnetic stirrer under refluxing conditions. After 30 min, the reactant mixture was cooled to room temperature. All the synthetic processes were carried out under inert atmosphere. After the completion of the reaction, the resulting Cu particles were retrieved from the Ar-filled flask and handled in air without further chemical passivation procedures. The synthesized Cu particles were collected by centrifugation and washed with toluene. As much as ~10 g of the Cu particles were obtained by a single synthesis process without significant agglomeration. The Cu particles were dispersed in dichlorobenzene at a solid loading of 20 wt%, and then transformed into 2-dimensional flake-shaped particles by a ball-milling process.

Fabrication of Cu mesh TCEs

For the flexible substrate, a square trench pattern was transferred to the PDMS substrate by replica molding from an SU-8 mesh pattern. Then, a toluene suspension (10 wt%) of the synthesized flake-shaped Cu microparticles was dropped onto the PDMS substrate, followed by blading the suspension across the substrate using a glass slide to facilitate the filling of Cu

flakes into the square trenches with an inverse-mesh structure. To remove excess Cu flakes, a commercial adhesive was attached and detached repetitively, leaving the Cu flakes only in the square trenches. A subsequent electroless deposition process was performed with Copper(II) sulfate pentahydrate (≥ 98%, Aldrich), potassium sodium tartrate (99%, Aldrich), sodium hydroxide (99.99%, Aldrich), and formaldehyde (37 wt% in water, Aldrich) after exposing the substrate to oxygen plasma. Finally, the Cu TCE consisting of a Cu mesh embedded in a PDMS substrate was obtained after washing with DI water.

Fabrication of a tunable color reflector based on Cu mesh TCEs

A green-colored CLC solution was prepared by dissolving 25.7 *w/w*% chiral dopant (R-811, Merck) in a host nematic liquid crystal (BHR-59001, BaYi Space LCD Tech.) A slide glass was placed on the surface of a Cu TCE heater with a spacer of 50 μm, of which gap is infiltrated with the CLC solution. The CLC film was observed by optical microscopy in reflection mode (L150, Nikon) and the reflection spectra were acquired using a fiber-coupled spectrometer (HR 4000, Ocean Optics) mounted on the microscope (L150, Nikon) with a 10× lens (N. A. 0.30).

Characterization

The size and shape of the Cu mesh structures were examined by scanning electron microscopy (SEM) (JSM-6700, JEOL). The crystal structure of the Cu flake microparticles was analyzed using an X-ray diffractometer (D/MAX-2200V, Rigaku) in the range of $2\theta = 20–80°$ at 40 kV and 40 mA. The chemical structure of the Cu flake microparticles was analyzed by X-ray photo-electron spectroscopy (XPS) (K-Alpha, Thermo Fisher Scientific). The sheet resistance

and transmittance of the Cu mesh TCEs were measured using a noncontact eddy current probe system (EC-80P, Napson Corp., Japan) and an ultraviolet-visible spectrophotometer (Lambda 750S model, PerkinElmer Inc., USA). The electromechanical stability of the Cu TCE was evaluated as a function of bending radius using an automatic bending tester (PMC-1HS, Autonics). The joule heating characteristics of the Cu mesh TCE were examined with a DC power supply (E3634A, Keysight). The temperature and images of the heater based on Cu mesh TCE were captured in real-time using an IR camera (A600-series, FLIR Systems Inc., Sweden).

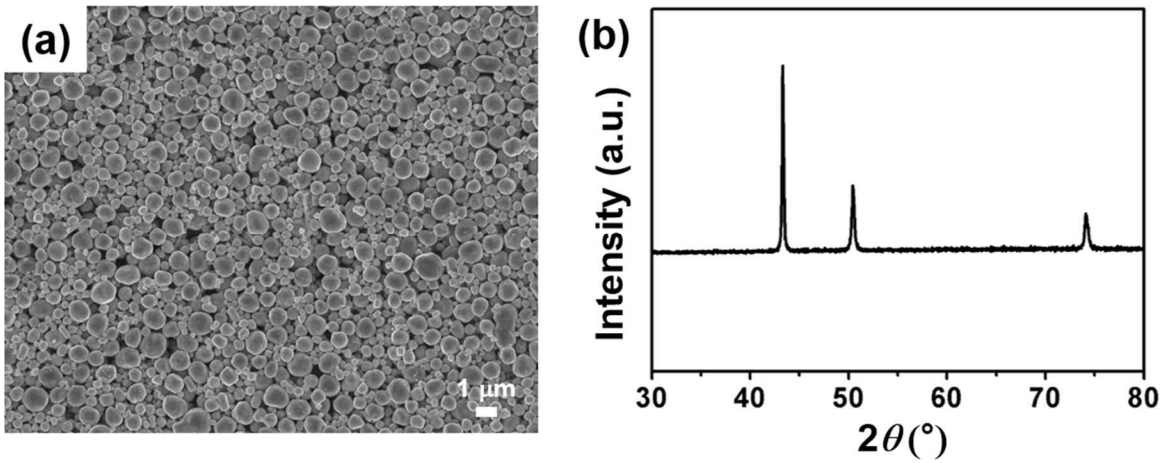

Fig. S1 (a) SEM images and (b) XRD pattern of surface-oxide-free Cu microparticles.

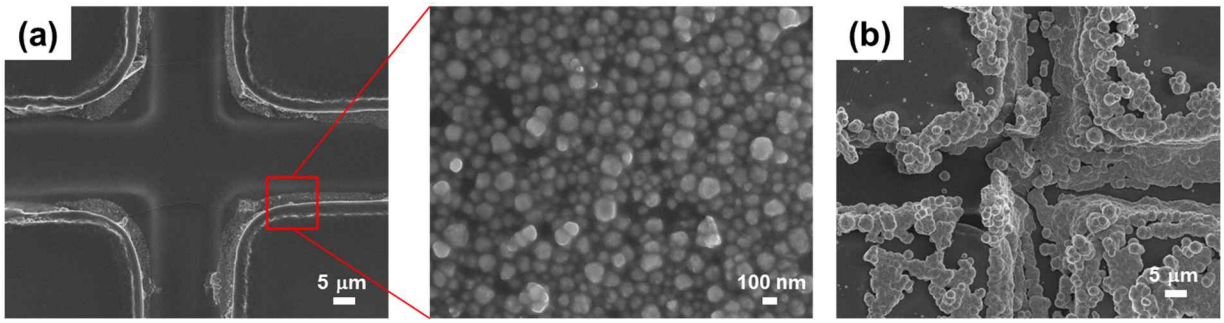

Fig. S2 (a) SEM images of Cu nanoparticles filled into mesh trenches on PDMS substrate. (b) SEM images of the plated Cu layer on nanoparticles after electroless Cu plating.

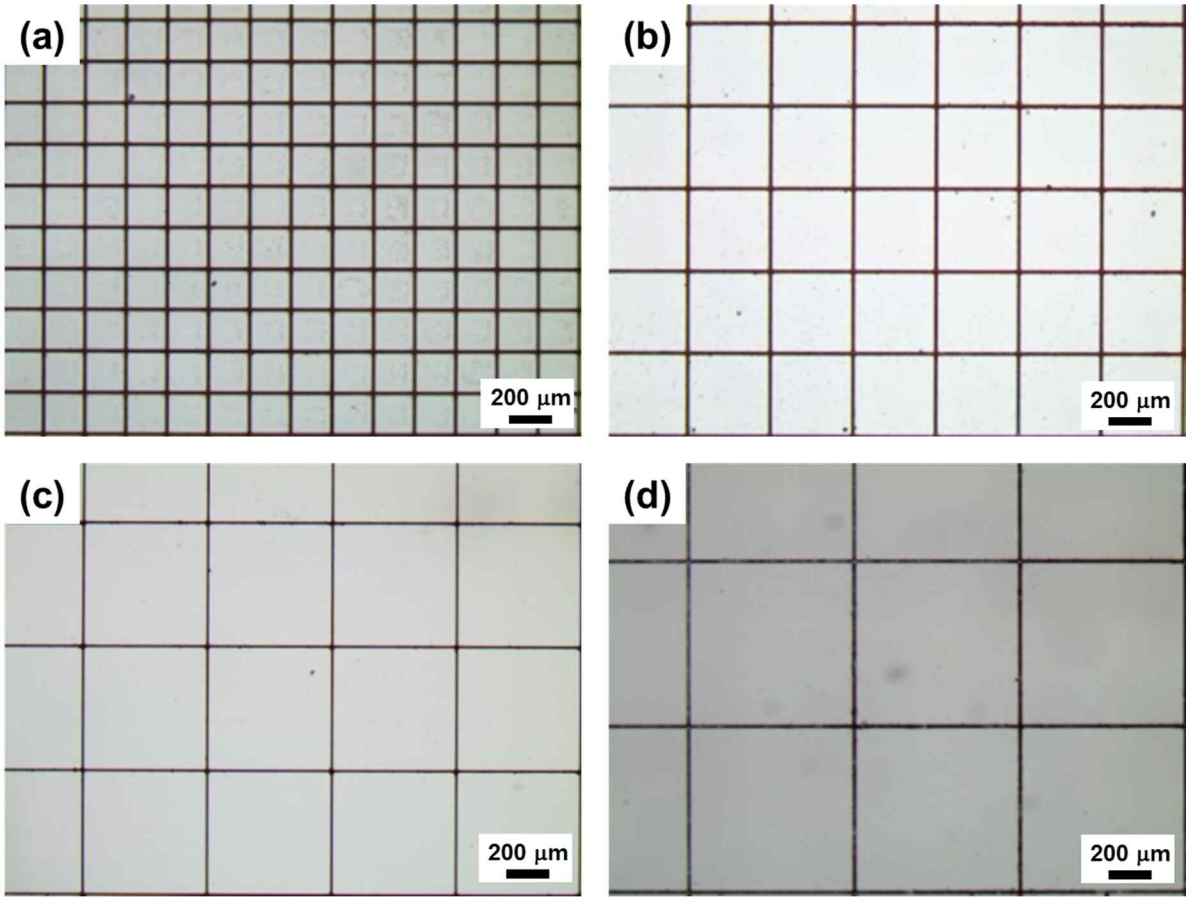

Fig. S3 OM images of Cu mesh structures on PDMS substrate with grid pitch of (a) 200, (b) 400, (c) 600, (d) 800 μm.

S4. Calculation of a sheet resistance of Cu-grid

Cu-grid patterns can be defined by a filling factor (FF) following equation:

$$FF = \frac{(p \times w) + [(p - w) \times w]}{p^2}$$

where p and w are the Cu-grid pitch and the width, respectively. From the calculated value of FF, we are able to predict the sheet resistance by following equation:

$$R_{s,\text{Cu grid}} = \frac{\rho_{\text{Cu grid}}}{t_{\text{Cu grid}}} \frac{1}{FF}$$

where $\rho_{Cu\ grid}$ is the resistivity of the Cu ink and $t_{Cu\ grid}$ is the thickness of the Cu-grid.

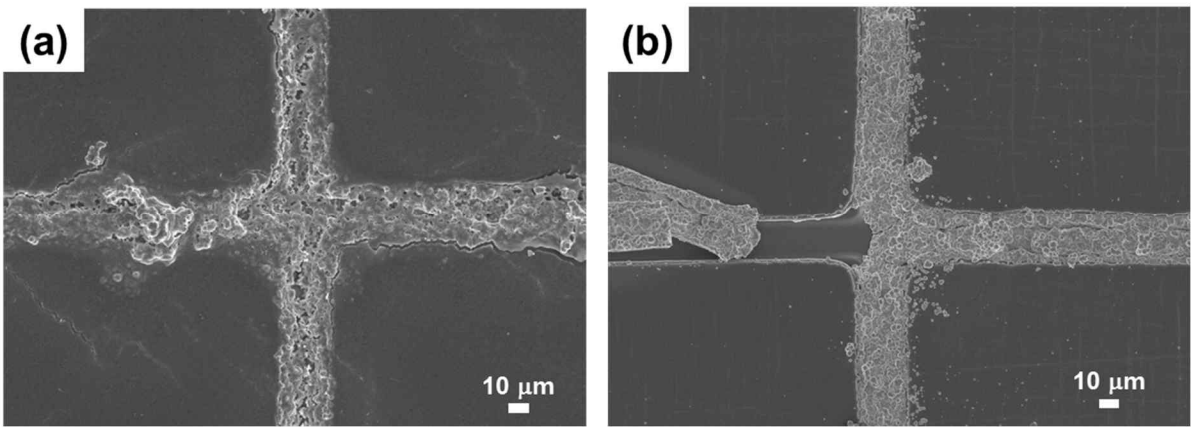

Fig. S5 SEM images of the Cu mesh electrodes with (a) and without (b) a layer of PEDOT:PSS after bending test.

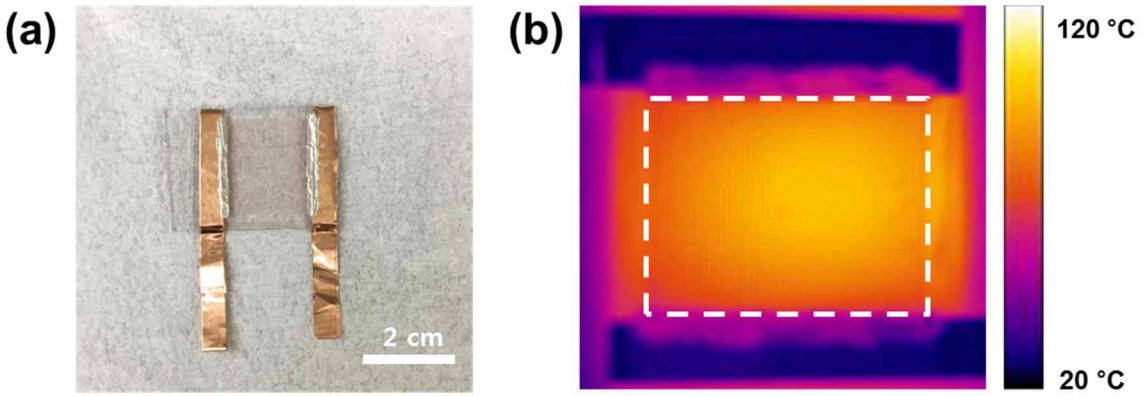

Fig. S6 (a) Photograph of Cu-mesh heater. (b) IR thermal image of the Cu-mesh heater showing uniform temperature distribution over the entire area.