Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

SUPPORTING INFORMATION

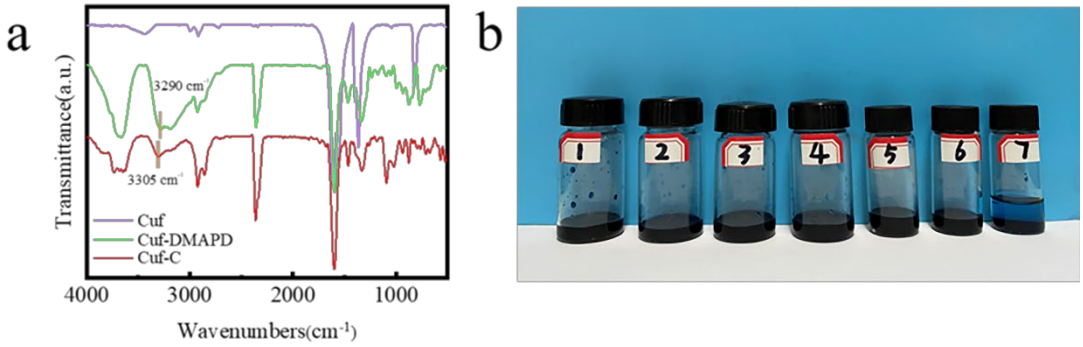

Figure S1. (a) Fourier infrared spectrum analysis of Cuf, Cuf-DMAPD and Cuf-C. (b) Solubility test of Cuf-C: (1) ethylene glycol, (2) isopropyl alcohol, (3) 1, 3-propylene glycol, (4) 1, 4-butanediol, (5) deionized water, (6) trichloromethane and (7) toluene.

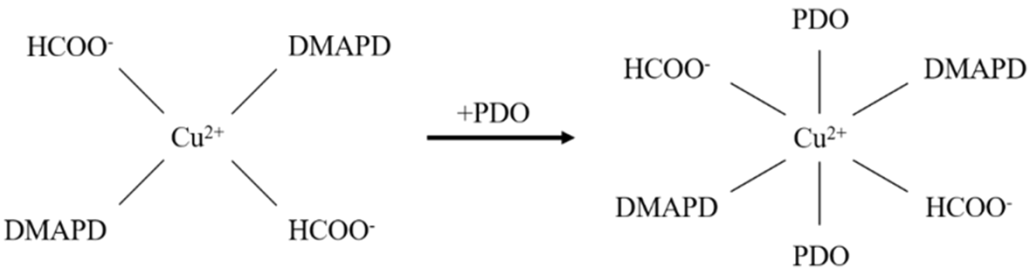

Figure S2. Schematic diagram of the coordination structure after adding PDO

The wide absorption band caused by the stretching vibration of O-H in DMAPD after the coordination of Cuf with DMAPD was shown at 3290 cm^{-1}, which proved the coordination of Cu^{2+} with DMAPD. After the addition of PDO, the absorption band shifted to 3305 cm^{-1}, showing the change of coordination structure after the addition of PDO. The 1336 cm^{-1} is the stretching vibration of C-N in DMAPD. The carboxyl group has no absorption peak at about 1710 cm^{-1}, but there is an asymmetric stretching vibration of the carboxyl group at 1595 cm^{-1}, indicating that there is a

coordination structure between the carboxyl group and the copper. With the addition of DMAPD and PDO, the absorption peak of Cuf at 1606 cm^{-1} moves to a lower wavenumber, indicating the difference in the coordination structure between Cuf and the latter two carboxyl groups and copper.

Table S1 The formulation of Cuf-C slurry and PVDF slurry

Reagent	Cuf-C slurry /g	PVDF slurry /g
Cuf	0.139	-
DMAPD	0.237	-
PDO	0.124	-
PVDF powder	-	0.5
NMP	10	10
Super-P	0.5	0.5
NCM811	4	4

Finite element simulation on polarization distribution of the composites

A single two-dimensional idealized model was established to simulate the distribution of polarization. The geometric models of Cuf-C/NCM811 cathode and PVDF/NCM811 cathode were established for simulation. The NCM811 particles located in the center were surrounded and tangent by Super-P particles. The peripheral Super-P particles kept point contact with each other. The geometric model of NCM811 was set as a circle with a radius of 6.35 μm, and the relative dielectric constant was 190. The geometric model of Super-P was set as a circle with a radius of 200 nm, and the conductivity was 1×10^5 S/m. The adhesive was set as a continuous square area with a length and width of 16 μm. The actual measured conductivity of the copper-based adhesive was 1.75 × 104 S/m. The relative dielectric constant of PVDF adhesive was 10. The remaining parameters referred to the material library data. A voltage of 2.5 V was applied in the

left and right directions of the composite model.

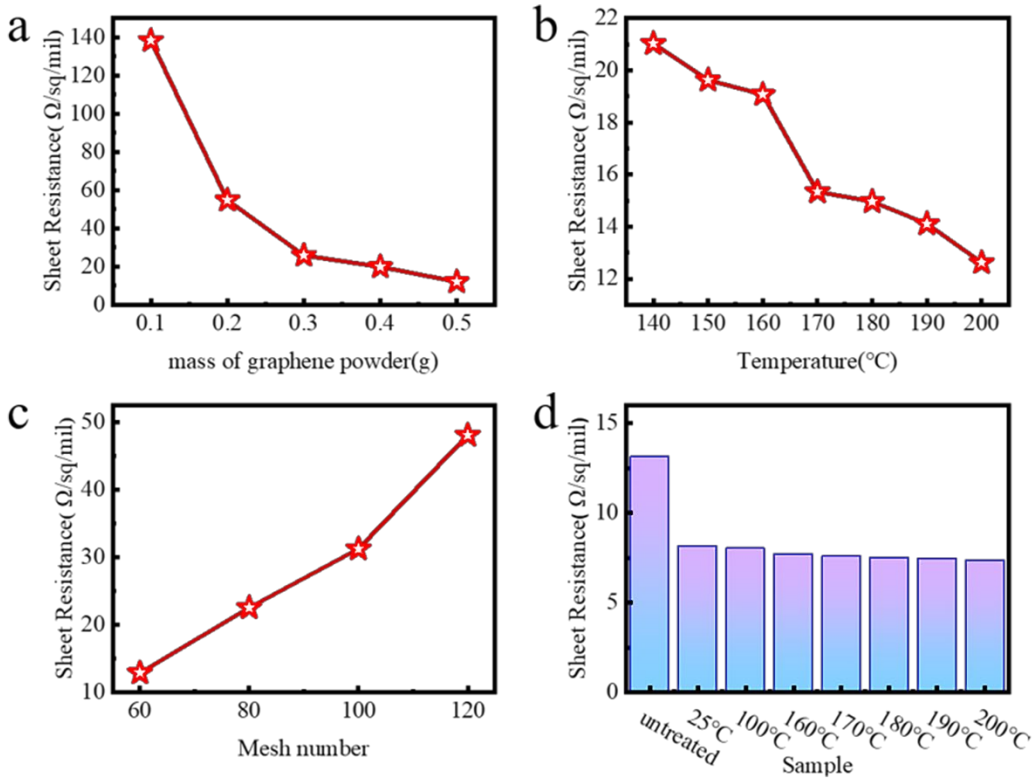

Figure S3. The effects of different (a) graphene powder addition amounts, (b) heat treatment temperatures, (c) screen mesh number, (d) post-treatment operations on the ink resistance .

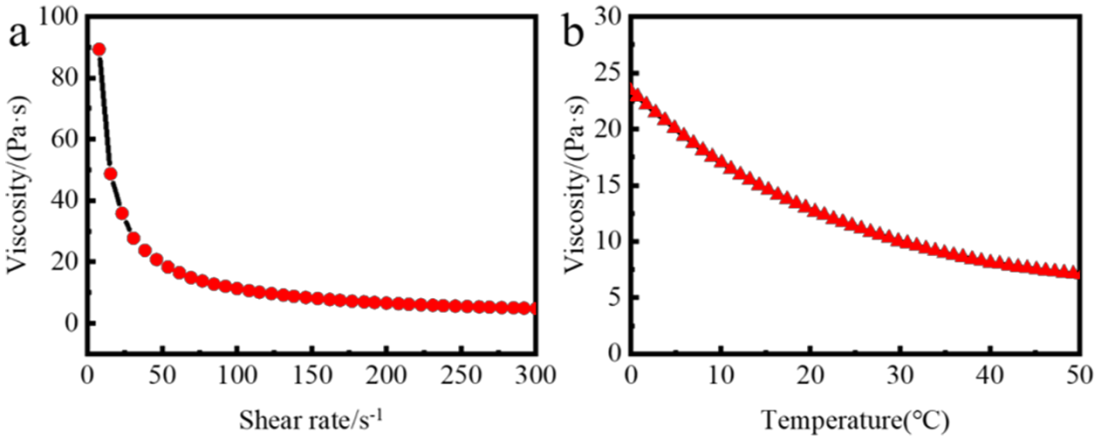

Figure S4. (a) Viscosity -shear rate curve, (b) Viscosity - temperature curve

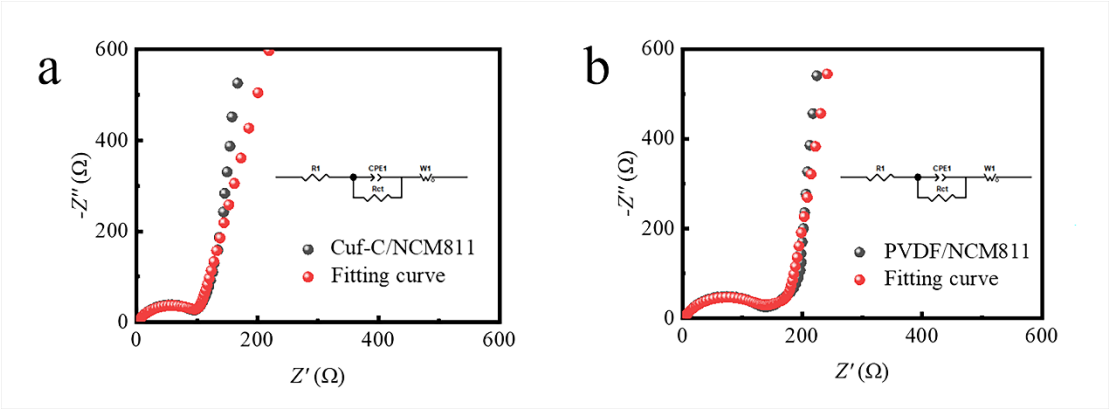

Figure S5. Equivalent circuit fitting data of (a) Cuf-C/NCM811 cell and (b) PVDF/NCM811 cell.

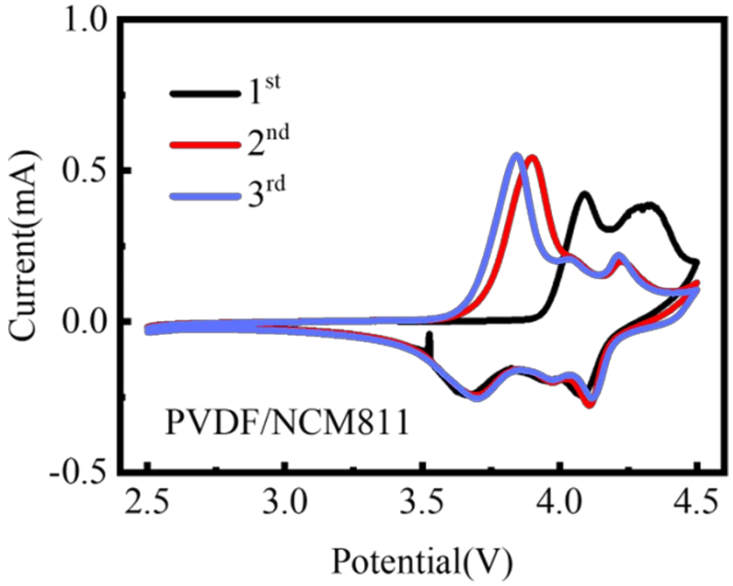

Figure S6. The consecutive CV curves of the first three cycles at a scan rate of 0.1 mV s^{-1} in the voltage range of 2.5-4.5 V for PVDF/NCM811 cell.

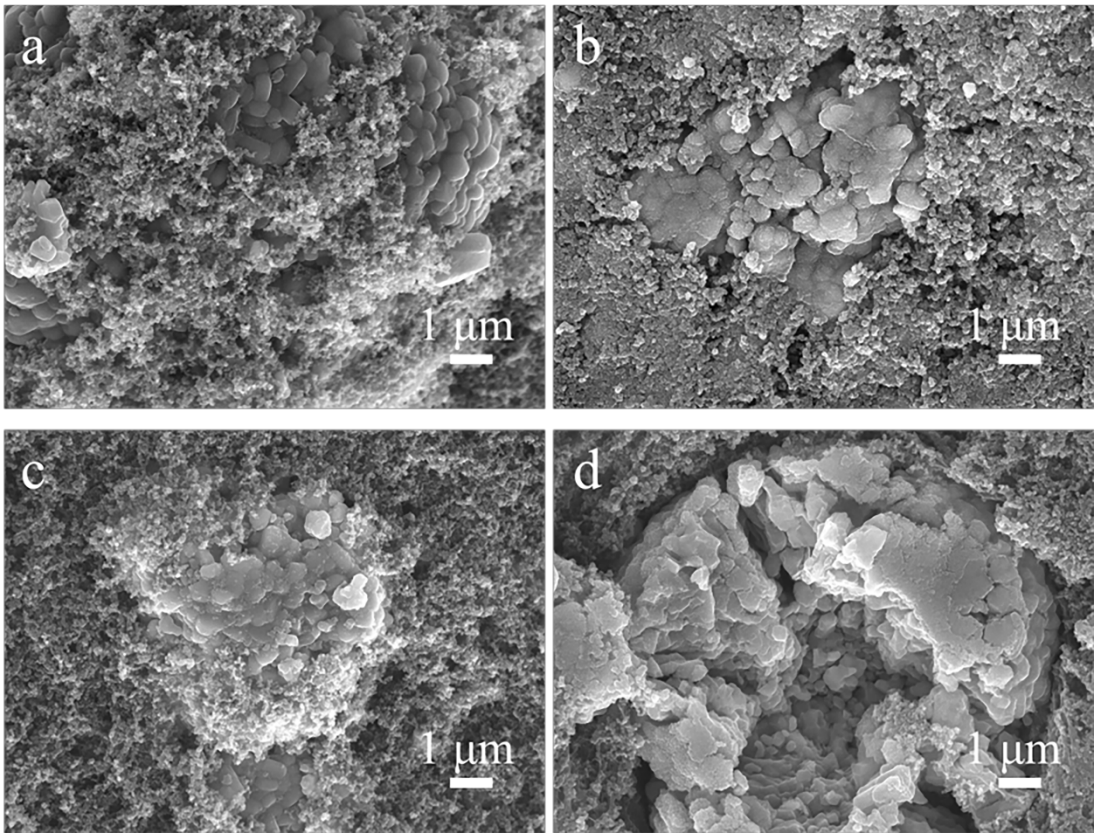

Figure S7. SEM images of Cuf-C/NCM811 cell (a) before and (b) after 100 charge-discharge cycles in the voltage range of 2.5-4.3 V at 1C. SEM images of PVDF/NCM811 cell (c) before and (d) after 100 charge-discharge cycles in the voltage range of 2.5-4.3 V at 1C.

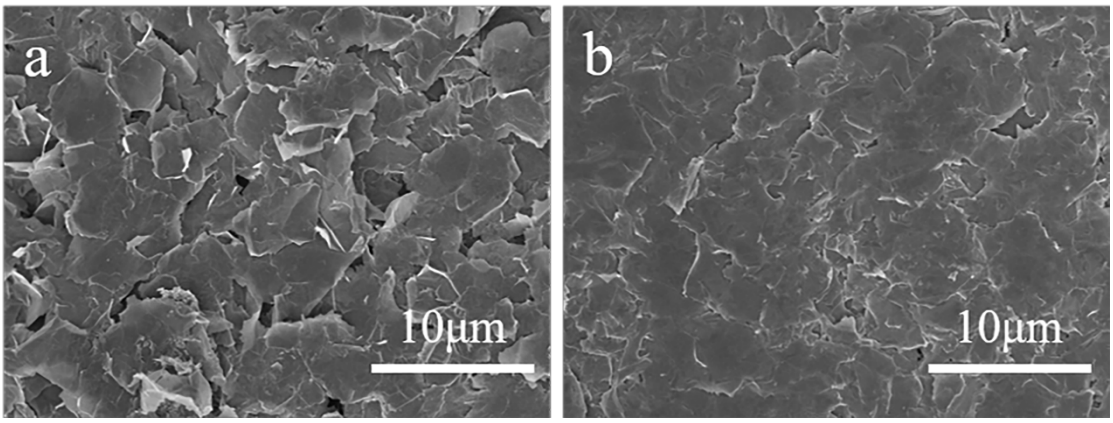

Figure S8. SEM images (a) before and (b) after hot pressing of Cuf-C/G ink

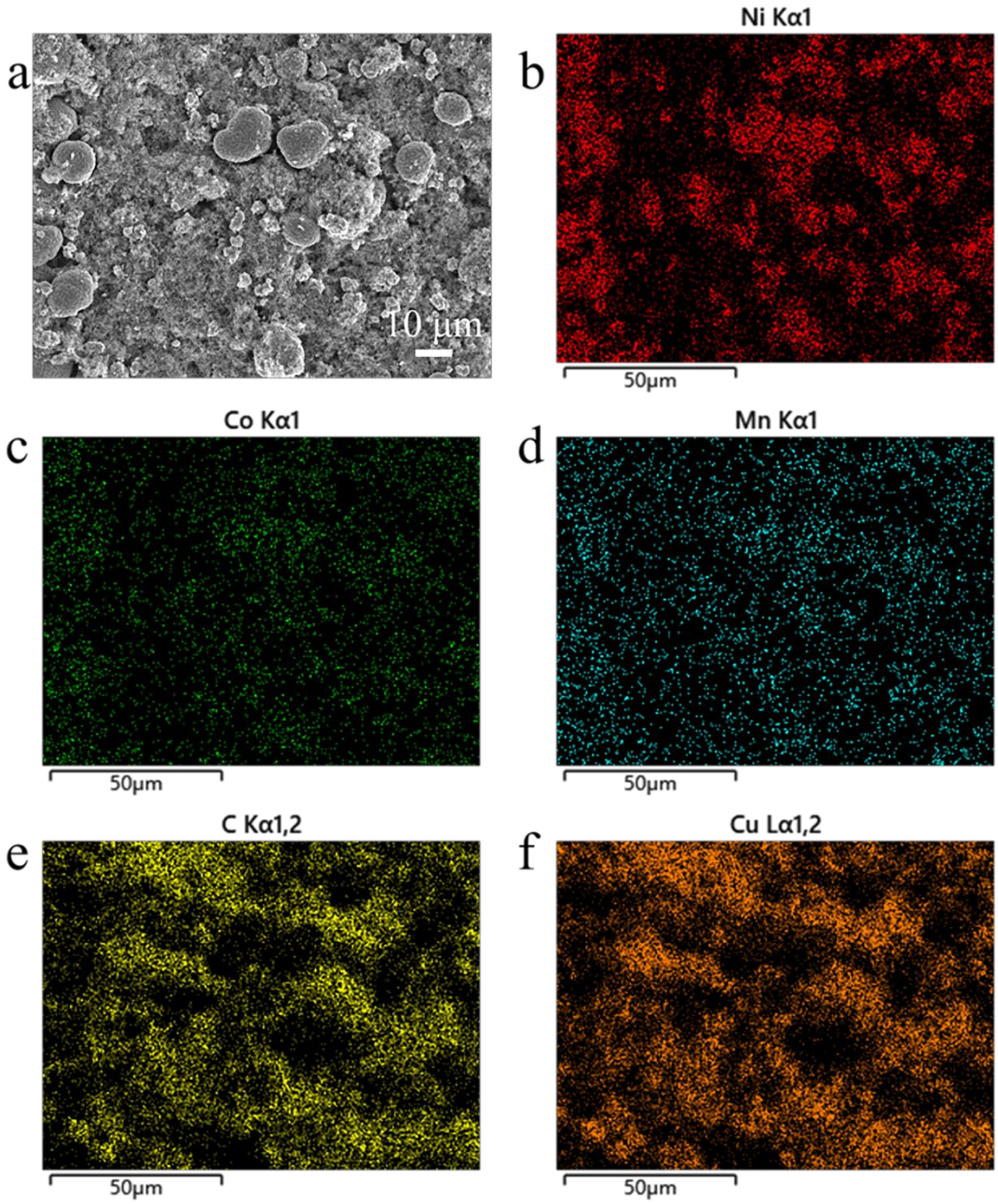

Figure S9. (a) SEM image and (b-f) EDS images of Cuf-C/NCM811. The Cuf-C/NCM811 cathode has not been rolled.

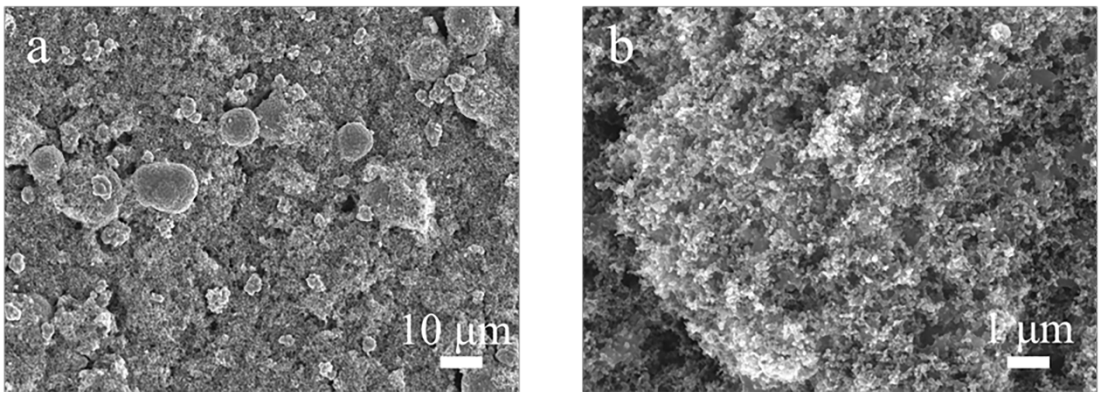

Figure S10. SEM images of Cuf-C/NCM811 cathode after soaking in a commercial electrolyte for 24 h.

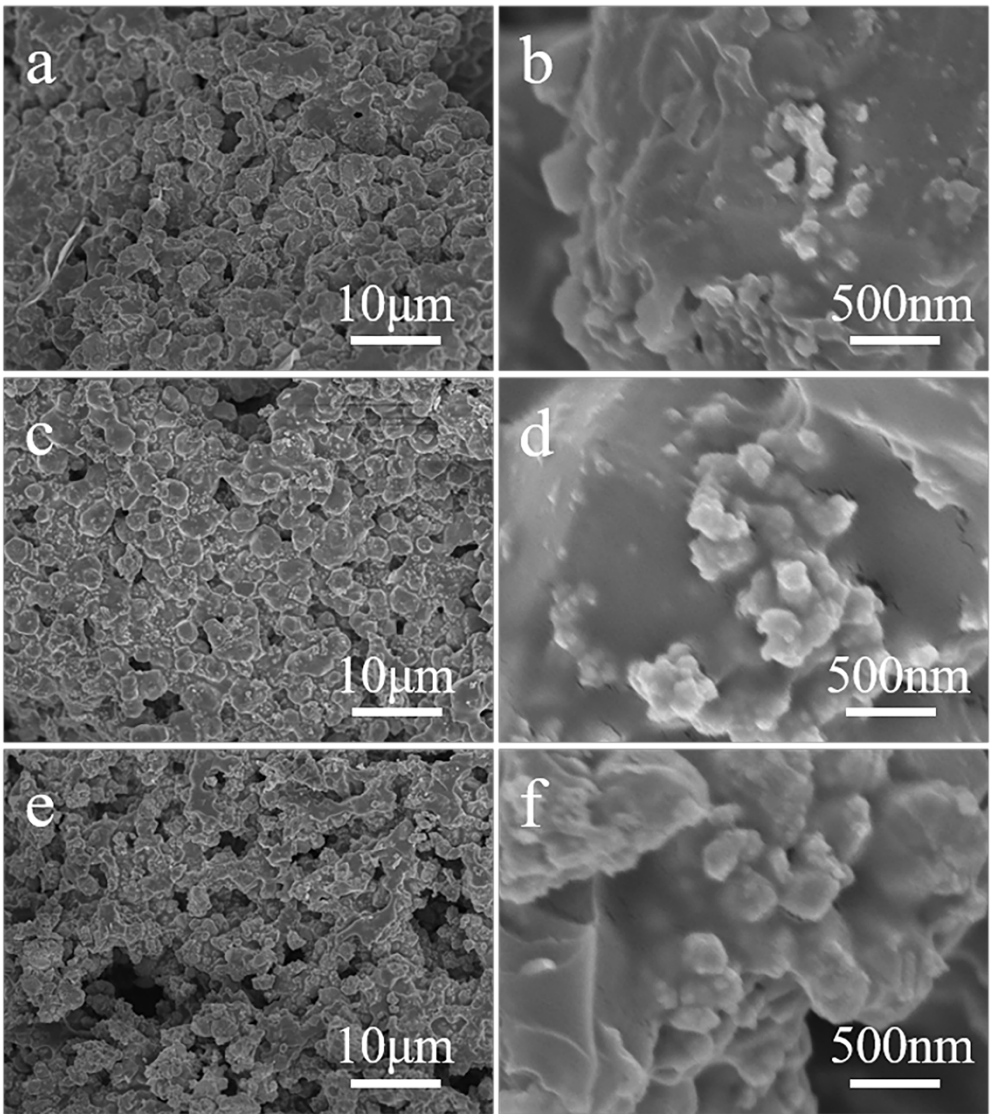

Figure S11. SEM images of Cuf-C pyrolysis products

(a-b) 140℃/30 min; (c-d) 160℃/30 min; (e-f) 180℃/30 min

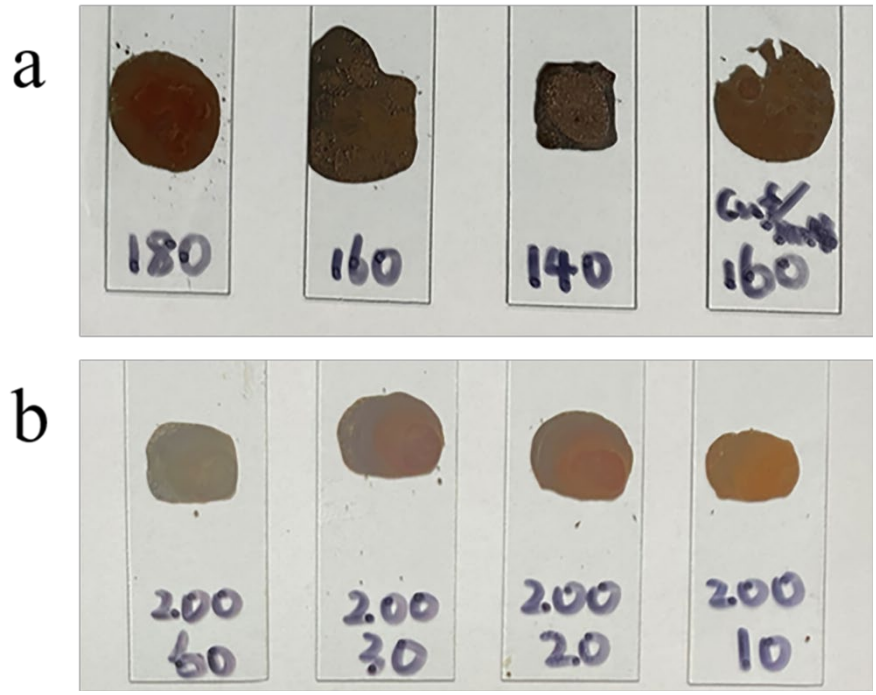

Figure S12. Sintered patterns

(a) Cuf-C at 140℃, 160℃, 180℃ for 30 min and Cuf-DMAPD at 160℃ for 30 min

(b) Cuf-C at 200℃ for 10 min, 20 min, 30 min and 60 min

Table S2. Ink formulation

	Cuf /g	DMAPD /g	PDO /g	Graphene /g	Super-P /g	Copper powder/g
Cuf-C/G	1.92	3.28	1.71	0.5	-	-
G	-	3.28	1.71	0.5	-	-
Cuf-C/SP	1.92	3.28	1.71	-	0.5	-
SP	-	3.28	1.71	-	0.5	-
Cuf-C/CP	1.92	3.28	1.71	-	-	0.5
CP	-	3.28	1.71	-	-	0.5

Table S3. Preparation process of inks

	Heat treatment	Post-processing
Cuf-C/G	200℃, 10 min	-
G	200℃, 10 min	-
Cuf-C/G(P)	200℃, 10 min	200℃, 10 MPa, 10 min
G(P)	200℃, 10 min	200℃, 10 MPa, 10 min
Cuf-C/SP	200℃, 10 min	-
SP	200℃, 10 min	-
Cuf-C/SP(P)	200℃, 10 min	200℃, 10 MPa, 10 min
SP(P)	200℃, 10 min	200℃, 10 MPa, 10 min
Cuf-C/CP	200℃, 10 min	-
CP	200℃, 10 min	-
Cuf-C/CP(P)	200℃, 10 min	200℃, 10 MPa, 10 min
CP(P)	200℃, 10 min	200℃, 10 MPa, 10 min

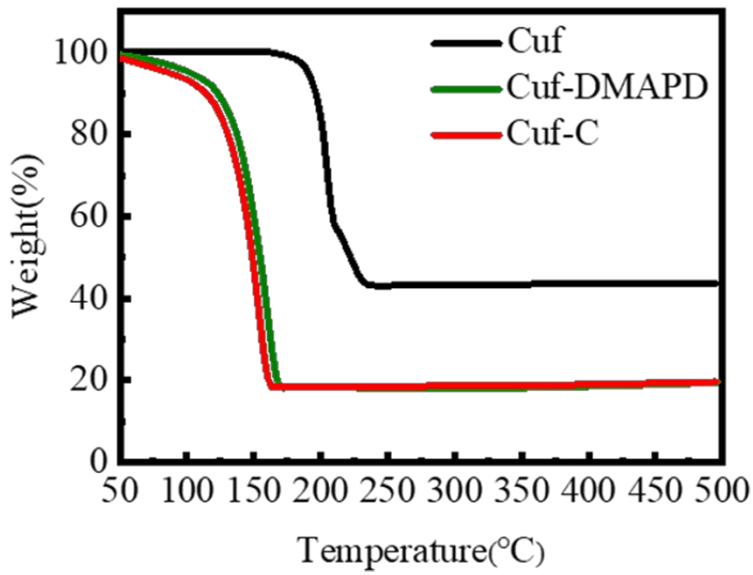

Figure S13. TG curves of Cuf, Cuf-DMAPD and Cuf-C under an air environment

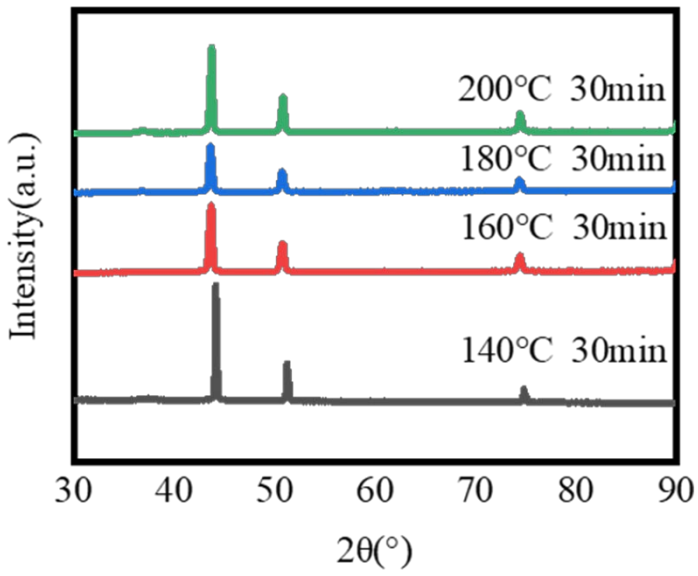

Figure S14. XRD pattern of Cuf-C annealed at different temperatures for 30 min