Boosting CO₂ electroreduction to C₂₊ products on fluorine-doped

copper

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Experimental Section

Chemicals and materials: Potassium hydroxide (KOH, $\geq 85\%$) and sodium 2, 2-dimethyl-2-silapentane-5sulfonate (DSS, 99%) were purchased from Sigma-Aldrich. Cupric chloride (CuCl₂, $\geq 99.0\%$), sodium borohydride (NaBH₄, $\geq 99.0\%$), ammonium fluoride (NH₄F, $\geq 99.0\%$) were purchased from Sinopharm Chemical Reagent Co. Ltd. D2O (98%) and Ni foam were purchased from Beijing Innochem Science & Technology Co. Ltd. All the chemicals were used as received. N₂ (99.999%) and CO₂ (99.999%) were provided by Beijing Analytical Instrument Company. Deionized water was used in the experiments.

Synthetic procedures for F-Cu catalysts. The typical synthesis of F-Cu catalysts was conducted as below. First, 300 mg CuCl_2 were dissolved in 50 ml cold deionized water, followed by the addition of various amounts of NH₄F (0 mg for Cu particles, 20.7 mg for F-Cu). Then the solution was sonicated for a short time and frozen below -6 °C for 24h to form the solid ice cube. The ice cube was then added into the cold NaBH₄ solution (2M, 10 ml) in the ice-water bath and reacted for 30 min with stirring. The formed black powders were washed with the deionized water and acetone for 5 times and collected by centrifugation. The precipitates were dried in a vacuum freeze drying oven for 24h, and annealed at 500 °C under the N₂ stream for 2h with the heating rate as 10 °C/min. After cooling down into the room temperature, the F-Cu catalysts were obtained.

Characterization of the materials. X-ray photoelectron spectroscopy (XPS) analysis was performed on the Thermo Scientific ESCA Lab 250Xi using 200 W monochromatic Al K α radiation. The 500 μ m X-ray spot was used. The base pressure in the analysis chamber was about 3×10^{-10} mbar. Typically, the hydrocarbon C1s line at 284.8 eV from adventitious carbon was used for energy referencing X-ray diffraction (XRD) analysis of the samples and the experiment was performed on the X-ray diffractometer (Model D/MAX2500, Rigaka) with Cu-K α radiation at the scan speed was 5° min⁻¹. The morphologies of as-synthesized materials were characterized by a HITACHI S-4800 scanning electron microscope (SEM) and a JEOL JEM-2100F high-resolution transmission electron microscopy (HR-TEM).

Preparation of electrodes. To construct the cathode electrode, a catalyst slurry that contained 5 mg of obtained catalysts, 1 mL of methanol and 20 μ L of Nafion ionomer solution (5 wt% in H₂O) was first mixed and sonicated for 30 min. Then, the catalyst slurry (0.2 mL) was slowly drop cast onto a PTFE membrane (Fuel Cell Store) under vacuum to achieve a catalyst loading of ~1.0 mg cm⁻². Ni foam were used anode electrode.

Electrochemical study. Electrochemical studies were conducted in an electrochemical flow cell which including a gas chamber, a cathodic chamber, and an anodic chamber, as reported in our previous work.¹ An anion exchange

membrane (FumasepFAA-3-PK-130) was used to separate the anodic and cathodic chambers, and an Ag/AgCl electrode and Ni foam were used as the reference and counter electrodes, respectively. The electrolysis was conducted using a CHI-660e electrochemical workstation equipped with a high current amplifier CHI 680c. The measured potentials after *iR* compensation (3.6 ohm was used) were rescaled to the RHE by *E* (versus RHE) = *E* (versus Ag/AgCl) + 0.197 V + 0.0591 V/pH × pH. For performance studies, 1M KOH was used as the electrolyte, and it was circulated through the cathodic and anodic chambers using peristaltic pumps at a rate of 20 mL min⁻¹. The flow rate of CO₂ gas through the gas chamber was controlled to be 20 sccm using a digital gas flow controller.

Product analysis. The gaseous product of electrochemical experiments was collected using a gas bag (the first bag was collected from the 10th minute at every applied potential and each bag was collected for 15 minutes) and analyzed by gas chromatography (GC, HP 4890D), which was equipped with TCD detector using argon as the carrier gas. The liquid product was analyzed by ¹H NMR (Bruker Advance III 400 HD spectrometer) in deuteroxide. **Calculations of Faradaic efficiencies of gaseous and liquid products**

Calculations of Faradaic efficiencies of Liquid products: After electrolysis, a certain amount of internal standard solution was added to the electrolyte as the internal standard. Because the concentration of internal standard was known, the moles of liquid products can be calculated from integral areas and calibration curves. To accurately integrate the products in NMR analysis, two standards located in different regions were used in NMR analysis. The sodium 2, 2-dimethyl-2-silapentane-5-sulfonate (DSS) was the reference for n-propanol, ethanol and acetic acid, and the phenol was the reference for formate. 400 μ L catholyte after the reaction was mixed with 100 μ L 6 mM DSS solution, 100 μ L 200 mM phenol and 200 μ L D₂O, and then analyzed by ¹H NMR (Bruker Advance III 400 HD spectrometer).

The Faradaic efficiency of liquid product is:

$$FE = \frac{moles \ of \ product}{Q / nF} \times 100\%$$

(Q: charge (C); F: Faradaic constant (96485 C/mol); n: the number of electrons required to generate the product) **Calculations of Faradaic efficiencies of Gaseous products.** From the GC peak areas and calibration curves for the TCD detector, we can obtain the V % of gaseous products. Since the flow rate of the outlet was monitored to be constant, the moles of gaseous products can be calculated. The Faradaic efficiency of gaseous product is:

$$FE = \frac{moles \ of \ product}{Q / nF} \times 100\%$$

(Q: charge (C); F: Faradaic constant (96485 C/mol); n: the number of electrons required to generate the product)

Electrochemical impedance spectroscopy (EIS) study. The EIS measurement was carried out in 1 M KOH at an open circuit potential (OCP) with an amplitude of 5 mV of 10⁻¹ to 10⁶ Hz.

Double-layer capacitance (C_{dl}) measurements. The catalyst was dropped on the 6mm glass carbon electrode (GCE) as the work electrode. The electrochemical active surface area is proportional to C_{dl} value. C_{dl} was determined by measuring the capacitive current associated with double-layer charging from the scan-rate dependence of cyclic voltammogram (CV). The CV ranged from -0.28 V to -0.36 V vs. RHE. The C_{dl} was estimated by plotting the Δj (j_a - j_c) at -0.30 V vs. RHE against the scan rates, in which the j_a and j_c are the anodic and cathodic current density, respectively. The scan rates were 10, 20, 40, 50, 60, 80 and 100 mV s⁻¹.

In-situ Raman measurements. *In-situ* Raman measurements were carried out using a Horiba LabRAM HR Evolution Raman microscope in a modified flow cell (purchased from Gaossunion Technology Co., Ltd.). A 785-nm laser was used and signals were recorded using a 40 s integration and by averaging two scans. The catalysts were firstly detected as the "pre" line in Fig. 3d and 3e to confirm the structure properties after the pre-electrolysis. The *iR* compensation was conducted in the *operando* XAS experiment at each potential, and a 15-minute electrolysis was conducted to gain the steady state before the collection of Raman spectra with constantly flowed gaseous CO₂.

XAFS measurements

The X-ray absorption find structure spectra data (Fe K-edge) were collected at 1W1B station in Beijing Synchrotron Radiation Facility (BSRF, operated at 2.5 GeV with a maximum current of 250 mA). The data were collected in fluorescence excitation mode using a Lytle detector. All samples were pelletized as disks of 13 mm diameter with 1mm thickness using graphite powder as a binder.

Operando XAS measurements were conducted in a custom-designed flow cell as our previous reports.¹ Catalysts were filtered on the PTFE membrane as the working electrode and 1 M KOH was used as the electrolyte. The saturated KCl Ag/AgCl electrode was chosen as the reference electrode and the Ni foam was used as the counter electrode. The *iR* compensation was conducted in the *operando* XAS experiment at each potential, and a 15-minute electrolysis was conducted to gain the steady state before the collection of XAS spectra with constantly flowed gaseous CO₂.

Athena and Artemis software, parts of the Demeter package,² were used for analysis of the Extended X-ray Absorption Fine Structure EXAFS data. Pre-processing of data included alignment, edge calibration, deglitching, normalization, background subtraction, and conversion of data into a chi file for data fitting was performed with Athena. The edge energy at Cu K-edge, determined by the first inflection point of the absorption edge of the reference copper foil, was calibrated to the reported Cu K-edge energy, 8979 eV. The Fourier transformed (FT) data in *R* space were analyzed by applying the first shell approximation for the Cu-Cu shell. The amplitude reduction

factor, S_0^2 , were determined by fitting the experimental Cu foil data and fixing the Cu-Cu coordination number to be 12, and then fixed for further analysis of the measured samples. The parameters describing the local structure environment including coordination number (*CN*), scattering path distance between absorber and backscattering atoms (*R*), mean square relative displacement ($\Delta \sigma^2$, Debye-Waller factor) and inner potential correction (ΔE_0) were allowed to vary during the fit process. The fitting ranges in both *k* space and *R* space in analysis of data characterizing samples were determined by the data quality. These values were used with the Nyquist theorem³ to estimate the justified number of fitting parameters, which was not exceeded in the fitting. For the XANES part, standard procedures were used in the analysis of the XANES spectra.²

Supplementary Figure



Fig. S1. (a) The X-ray photo-electron spectra of Cu-P and F-Cu catalysts in the region of Cu 2p. (b) The X-ray photo-electron spectra of Cu-P and F-Cu catalysts in the region of Auger Cu LMM.



Fig. S2. The X-ray photo-electron spectra of Cu-P and F-Cu catalysts in the region of F 2p.



Fig. S3. The TEM image of Cu-P before CO₂RR.



Fig. S4. Fourier transforms of k^3 -weighted EXAFS data at Cu K-edge over Cu-P and F-Cu before CO₂RR.



Fig. S5. The XRD patterns of Cu-P after CO₂RR.



Fig. S6. *Operando* Fourier transforms of k^3 -weighted EXAFS data at Cu K-edge at -0.97 V (vs. RHE) over Cu-P and F-Cu during CO₂RR.



Fig. S7. The EXAFS data fitting results of Cu-P (a) and F-Cu (b) before CO_2RR .



Fig. S8. The EXAFS data fitting results of F-Cu at various applied potentials.



Fig. S9. The EXAFS data fitting results of Cu-P (a) and F-Cu (b) at -0.97 V.



Fig. S10. The EDX mapping of F-Cu after the electrochemical test.



Fig. S11. The X-ray photo-electron spectra of F-Cu catalysts in the region of F 2p after CO_2RR .



Fig. S12. A typical ¹H NMR spectrum of liquid products after electrolysis at -0.97 V (vs. RHE) over F-Cu.



Fig. S13. The ¹H NMR plot of the liquid products after electrolysis at -0.97 V (vs. RHE) over F-Cu using ${}^{13}\text{CO}_2$ as gas source.



Fig. S14. (a) LSV and (b) the total current density over Cu-P and F-Cu at various applied potentials in CO₂RR.



Fig. S15. The distribution of products over F-Cu at various applied potentials in CO₂RR.



Fig. S16. The distribution of products over Cu-P at various applied potentials in CO₂RR.



Fig. S17. The comparison for the partial current density of (a) C_2H_4 and (b) ethanol over Cu-P and F-Cu at various applied potentials in CO_2RR .



Fig. S18. The comparison for the FE of H_2 over Cu-P and F-Cu at various applied potentials in CO_2RR .



Fig. S19. Nyquist plots for Cu-P and F-Cu catalysts (a, b) and corresponding simulated circuit (c).



Fig. S20. The capacitance current density versus scan rates (10, 20, 40, 50, 60, 80 and 100 mV s⁻¹) plot over Cu-P and F-Cu (b) after reaction.



Fig. S21. The measured specific double layer capacitance and fitted results for Cu-P (a) and F-Cu (b) after CO_2RR .



Fig. S22. The ECSA corrected partial current density of C_{2+} products over Cu-P and F-Cu at various applied potentials in CO_2RR .



Fig. S23. The SEM images of Cu-P and F-Cu after CO₂RR.



Fig. S24. The TEM images of (a) Cu-P and (b) F-Cu after CO₂RR.



Fig. S25. The HR-TEM image of F-Cu after the electrochemical test.



Fig. S26. The contact angle of electrolyte droplets on Cu-P/GDE before CO₂RR.



Fig. S27. The contact angle of electrolyte droplets on F-Cu/GDE before CO_2RR .



Fig. S28. The contact angle of electrolyte droplets on Cu-P/GDE and F-Cu/GDE after CO₂RR.



Fig. S29. In-situ electrochemical spectral cell for SERS test.



Fig. S30. In-situ SERS over Cu-P and F-Cu after CO₂RR.

Supplementary Tables

	Cu (<i>at</i> %)	O (<i>at</i> %)	F (<i>at</i> %)
Cu-P	34.5	65.4	0.12
-Cu	41.9	57.1	1.07
F-Cu (after CO ₂ RR)	53.4	45.6	1.18

Table 1. Elemental distribution of F-Cu catalysts based on XPS results.

 Cu (wt%)
 O (wt%)
 F (wt%)

 Cu-P (after CO₂RR)
 75.33
 24.67

 F-Cu
 64.97
 31.53
 3.50

 F-Cu (after CO₂RR)
 80.04
 15.35
 4.60

Table 2. Elemental distribution of Cu-P and F-Cu catalysts based on EDX results.

Table 3. EXAFS fit parameters characterizing reference copper foil.	

Sample	Shell	CN^{a}	R (Å)	$\Delta\sigma^{2\times}(\text{\AA}^{2})$	S_0^2	$\Delta E_0 (\mathrm{eV})$	R factor
Cu Foil	Cu–Cu	12.0	2.54 ± 0.01	0.009 ± 0.001	0.86 ± 0.06	5.0 ± 0.7	0.004

^aNotation: *CN*, coordination number; *R*, scattering path distance between absorber and backscattering atoms; $\Delta\sigma^2$, mean square relative displacement (Debye-Waller factor); S_0^2 , amplitude reduction factor and ΔE_0 , inner potential correction. R factor is used to value the goodness of the fitting.

Table 4. Summary of EXAFS fit parameters characterizing F-Cu catalysts during CO_2RR . The range in *k* was 3.9–11.6 Å⁻¹, and the range in *r* was 1.4–3.0 Å.

Sample	Shell	CN^{a}	R (Å)	$\Delta\sigma^{2\times}(\text{\AA}^2)$	$\Delta E_0 (eV)$	R factor
F-Cu-0.57	Cu–Cu	6.5 ± 1.1	2.54 ± 0.01	0.009 ± 0.001	4.7 ± 1.8	0.01
F-Cu-0.77	Cu–Cu	5.0 ± 0.7	2.54 ± 0.01	0.006 ± 0.001	5.3 ± 1.5	0.01
F-Cu-0.97	Cu–Cu	5.6 ± 0.8	2.54 ± 0.01	0.007 ± 0.001	4.8 ± 1.6	0.01
Cu-P-0.97	Cu–Cu	5.9 ± 0.7	2.54 ± 0.01	0.007 ± 0.001	4.5 ± 1.4	0.008

^aNotation: *CN*, coordination number; *R*, scattering path distance between absorber and backscattering atoms; $\Delta\sigma^2$, mean square relative displacement (Debye-Waller factor); ΔE_0 , inner potential correction. R factor is used to value the goodness of the fitting. S_0^2 was found to be 0.86 based upon fitting copper foil to bulk coordination values (Table S3, Fig. S7 and S8).

Table 5. Summary of EXAFS fit parameters characterizing F-Cu-bulk. The range in k was 3.1–12.0 Å⁻¹, and the range in r was 1.0–3.3 Å.

Sample	Shell	CN^{a}	<i>R</i> (Å)	$\Delta\sigma^{2\times}(\text{\AA}^2)$	$\Delta E_0 (\mathrm{eV})$	R factor
E Cu	Cu–O	3.6 ± 0.4	1.96 ± 0.01	0.004 ± 0.001		
F-Cu- bulk	Cu–Cu ₁	4.9 ± 1.8	2.90 ± 0.01	0.009 ± 0.003	12.6 ± 1.0	0.01
Jun	Cu–Cu ₂	4.7 ± 1.8	3.10 ± 0.02	0.009 ± 0.003		

^aNotation: *CN*, coordination number; *R*, scattering path distance between absorber and backscattering atoms; $\Delta\sigma^2$, mean square relative displacement (Debye-Waller factor); ΔE_0 , inner potential correction. R factor is used to value the goodness of the fitting. S_0^2 was found to be 0.86 based upon fitting copper foil to bulk coordination values (Table S3, Fig. S6).

Table 6. Summary of EXAFS fit parameters characterizing Cu-P-bulk. The range in k was 3.1–12.0 Å⁻¹, and the range in r was 1.0–3.3 Å.

Sample	Shell	CN^{a}	<i>R</i> (Å)	$\Delta\sigma^{2\times}$ (Å ²)	$\Delta E_0 (eV)$	R factor
Cu-P- bulk	Cu–O Cu–Cu ₁ Cu–Cu ₂	2.2 ± 0.2 3.0 ± 1.1 3.2 ± 1.1	1.95 ± 0.01 2.90 ± 0.02 3.10 ± 0.01	0.003 ± 0.001 0.008 ± 0.003 0.008 ± 0.003	12.2 ± 0.9	0.01

^aNotation: *CN*, coordination number; *R*, scattering path distance between absorber and backscattering atoms; $\Delta \sigma^2$, mean square relative displacement (Debye-Waller factor); ΔE_0 , inner potential correction. R factor is used to value the goodness of the fitting. S_0^2 was found to be 0.86 based upon fitting copper foil to bulk coordination values (Table S3, Figure S6).

Catalysts	E (V) vs. RHE	$FE_{C2^{+}}$ (%)	$j_{\rm C2^+} ({ m mA~cm^{-2}})$	References
F-Cu	-0.97	70.4	250.4	This work
B-Cu	-1.05	48.2	16.1	S4
Cu_KI	-1.09	72.58	29.03	S5
FeTPP[C1]/Cu	-0.82	85	257.04	S6
I-modified Cu	-0.9	80	31.2	S7
Cl-doped Cu	-1	53.8	~10	S8
Multihollow Cu ₂ O	-0.61	75	267±13	S9
NGQ/Cu-nr	-0.9	80.4	226.81	S1
OD-Cu crystal	~ -1.05	74.9±1.7	224.7	S10
Reconstructed Cu	-0.68	64±1.4	255±5.7	S11
100-cycle Cu	-0.963	60.5	41	S12
OBC	-1	45	44.7	S13
Cu nanocubes	-0.8	60	44.4	S14
CC20	-1.12	75.2	913.2	S15
3D-CIBH	-0.91	91.7	1210	S16

Table 7. The Comparison of C_{2^+} products in CO_2RR on various Cu-based catalysts.

Sample	Amount of NH ₄ F (mg)	Cu ratio (at %)	F ratio (at %)	$\mathrm{FE}_{\mathrm{C2^+}}$ at -0.97 V
F-Cu-10	9.2	43.4	0.88	56.2
F-Cu	20.7	41.9	1.07	70.4
F-Cu-30	35.4	34.0	1.20	51.3

Table 8. The performance of F-Cu catalysts with various NH_4F amount in CO_2RR .

Notes and References

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