# Supporting Information

# Design and Synthesis of Magnesium Modified Copper Oxide Nanosheets as Efficient Electrocatalyst for $CO_2$ Reduction

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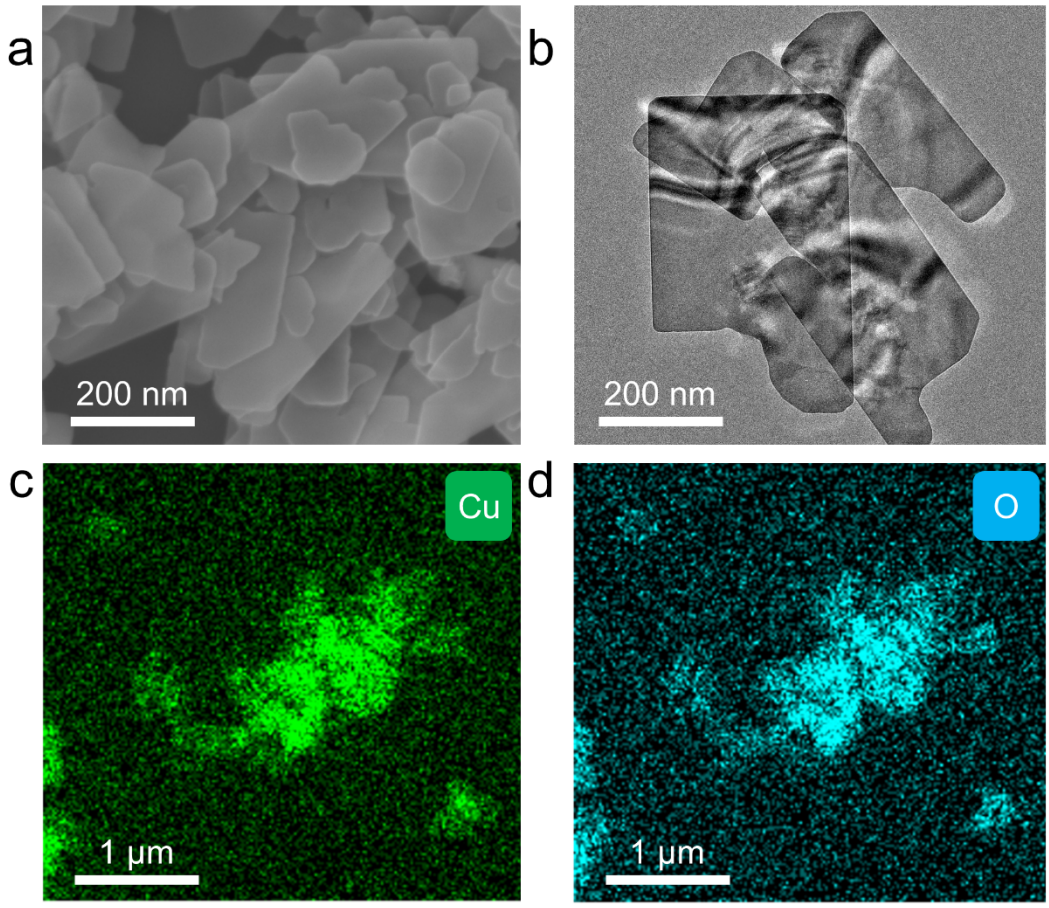


**Fig. S1** (a) The SEM image, (b) The HRTEM image and (c-d) The mapping images of CuO.

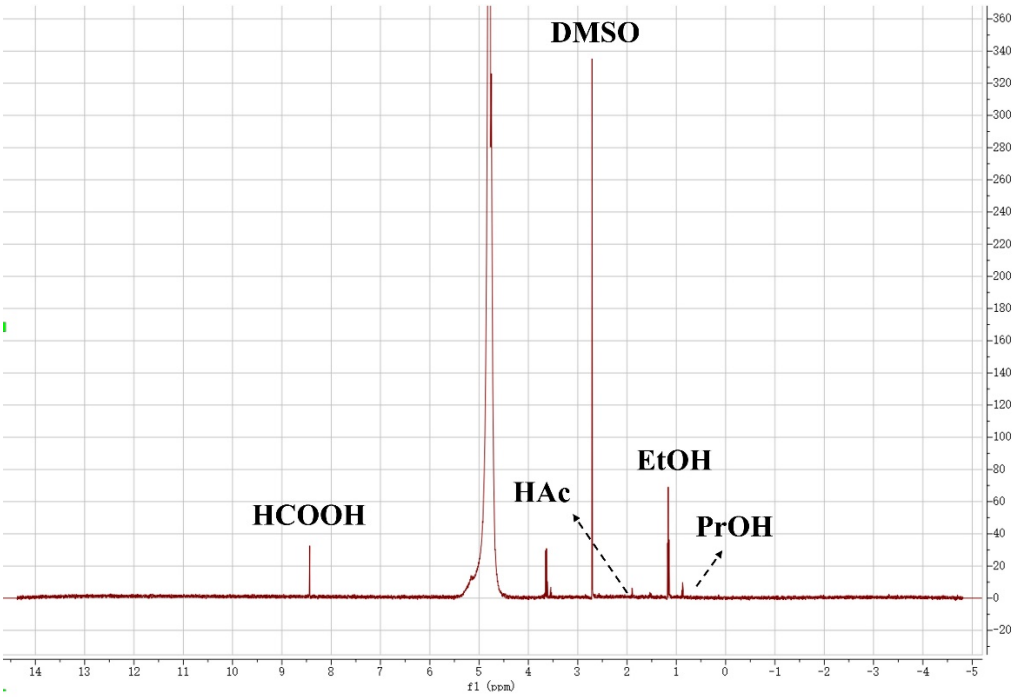


**Fig. S2** NMR spectrum of Mg-CuO-2.

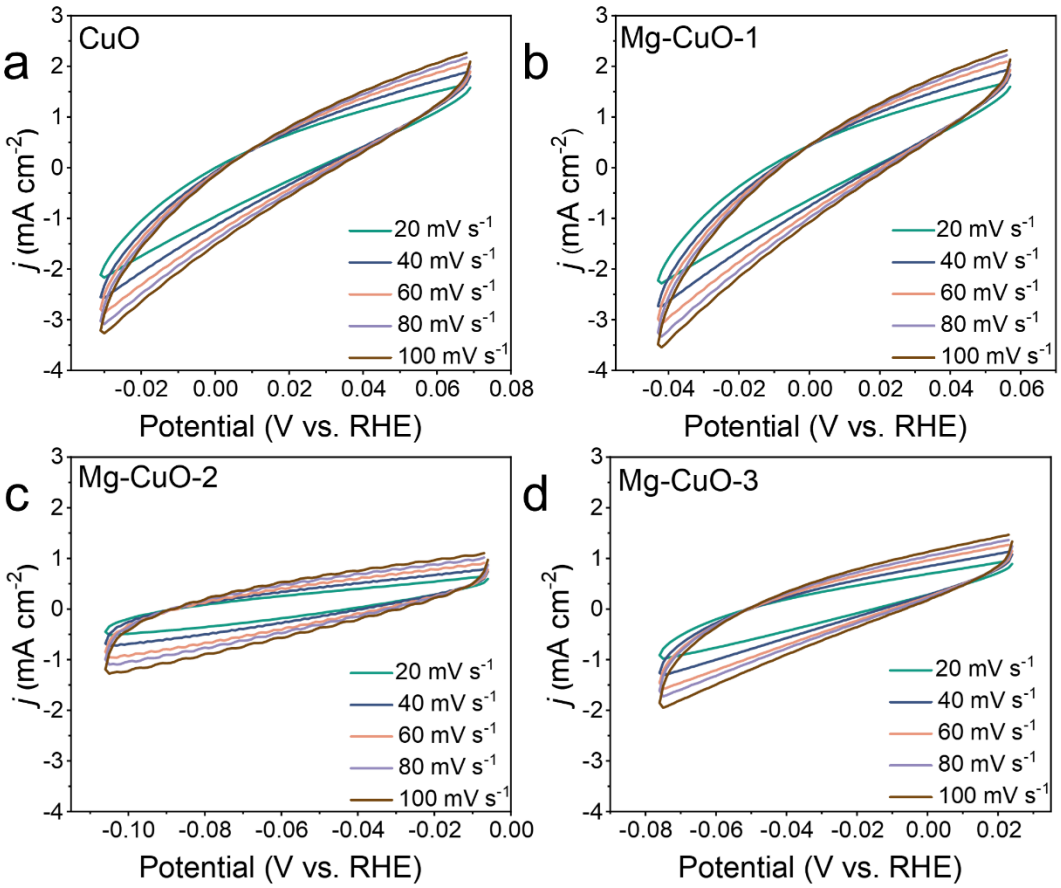


**Fig. S3** Cyclic voltammetry of (a) CuO, (b) Mg-CuO-1, (c) Mg-CuO-2 and (d) Mg-CuO-3 performed in $CO_2$-saturated 0.1 M $KHCO_3$ at various scan rates for measurement of double layer capacity. Cycle voltammetry was carried out under open circuit potential (OCPT) ± 0.05 V.

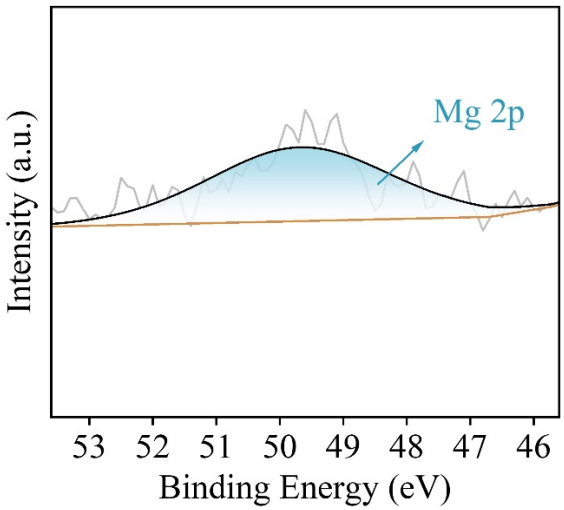


**Fig. S4** XPS spectra of Mg 2p for Mg-CuO-2 after the stability test.

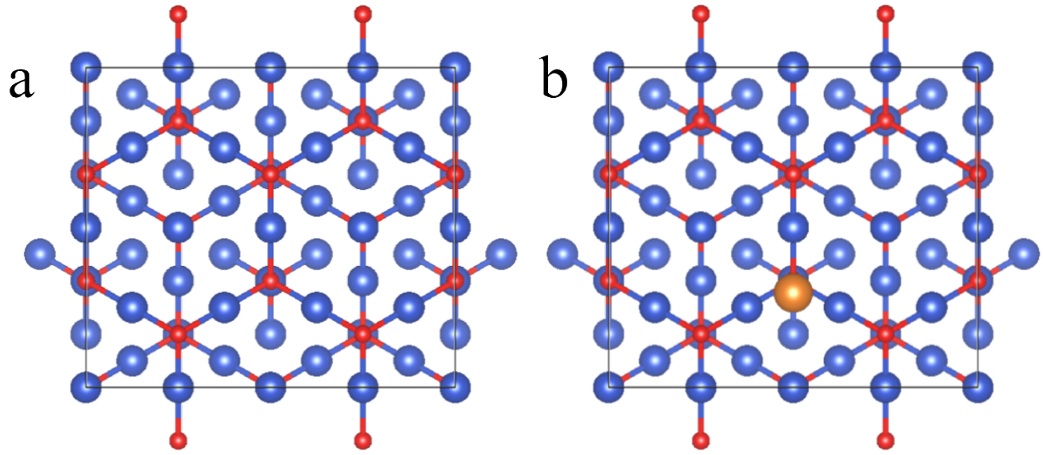


**Fig. S5** Models of (a) $Cu_2O$ and (b) Mg-$Cu_2O$-2 before structural optimization.

**Table S1** The $FE_{C2+}$ of Mg-CuO-2 at different potentials.

| Potential | $FE_{H2}$ (%) | $FE_{CO}$ (%) | $FE_{CH4}$ (%) | $FE_{HCOOH}$ (%) | $FE_{C2H4}$ (%) | $FE_{C2H5OH}$ (%) | $FE_{CH3COOH}$ (%) | $FE_{C3H8O}$ (%) | $FE_{C2+}$ (%) |
|---|---|---|---|---|---|---|---|---|---|
| -1.1 V | 29.10 | 0.04 | 0.85 | 7.54 | 32.51 | 8.21 | 2.14 | 1.85 | 44.71 |
| -1.2 V | 29.65 | 0.28 | 2.03 | 8.44 | 37.47 | 7.31 | 1.84 | 2.04 | 48.66 |
| -1.3 V | 25.32 | 0.24 | 1.92 | 8.94 | 46.34 | 13.32 | 1.71 | 1.27 | 62.64 |
| -1.4 V | 36.23 | 0.37 | 1.46 | 2.87 | 40.36 | 11.48 | 2.57 | 2.41 | 56.82 |
| -1.5 V | 43.07 | 0.09 | 1.14 | 2.29 | 33.41 | 12.47 | 2.45 | 3.52 | 51.85 |

**Table S2** The products formation rate of Mg-CuO-2 at different potentials.

| Potential | $H_2$ (mol/h) | CO (mol/h) | $CH_4$ (mol/h) | HCOOH (mol/h) | $C_2H_4$ (mol/h) | $C_2H_5OH$ (mol/h) | $CH_3COOH$ (mol/h) | $C_3H_8O$ (mol/h) |
|---|---|---|---|---|---|---|---|---|
| -1.1 V | 1.46*10^-4 | 2.01*10^-7 | 1.07*10^-6 | 3.79*10^-5 | 2.72*10^-5 | 6.88*10^-6 | 1.79*10^-6 | 1.16*10^-6 |
| -1.2 V | 1.26*10^-4 | 1.20*10^-6 | 2.16*10^-6 | 3.60*10^-5 | 2.66*10^-5 | 5.20*10^-6 | 1.31*10^-6 | 1.09*10^-6 |
| -1.3 V | 9.39*10^-5 | 8.90*10^-7 | 1.78*10^-6 | 3.31*10^-5 | 2.86*10^-5 | 8.23*10^-6 | 1.06*10^-6 | 5.88*10^-7 |
| -1.4 V | 1.13*10^-4 | 1.15*10^-6 | 1.13*10^-6 | 8.92*10^-6 | 2.09*10^-5 | 5.95*10^-6 | 1.33*10^-6 | 9.37*10^-7 |
| -1.5 V | 1.30*10^-4 | 2.71*10^-7 | 8.58*10^-7 | 6.89*10^-6 | 1.68*10^-5 | 6.26*10^-6 | 1.23*10^-6 | 1.32*10^-6 |

**Table S3** Comparison of electrocatalytic $CO_2RR$ performance with reported catalysts.

| Catalyst | Electrolytic cell | Electrolyte | Potential (V vs. RHE) | Electrode area | $FE_{C2+}$ (%) | $FE_{C2H4}$ (%) | Reference |
|---|---|---|---|---|---|---|---|
| Mg-CuO -2 | H-cell | 0.1 M CsI | -1.30 | 1.00 $cm^2$ | 62.64 | 46.34 | This work |
| $B_1$-CuO NS-2 | H-cell | 0.1 M $KHCO_3$ | -1.20 | 1.00 $cm^2$ | 54.78 | 38.56 | [1] |
| CuO/NG_AN | H-cell | 0.1 M $KHCO_3$ | -1.30 | Ø=10 mm | ~34.00 | ~30.00 | [2] |
| 60-CuO/$CeO_2$ | H-cell | 0.1 M $KHCO_3$ | -1.27 | 1.00 $cm^2$ | ~60.00 | 44.80 | [3] |
| ON−CuO | H-cell | 0.1 M $KHCO_3$ | -1.10 | 0.20 $cm^2$ | 77.00 | 56.00 | [4] |
| CuO spray | H-cell | 0.1 M $KHCO_3$ | -1.00 | 16.00 $cm^2$ | ~65.00 | 48.70 | [5] |
| CuO/$CeO_2$ | Single cell | 0.1 M $KHCO_3$ | -1.40 | Ø=12 mm | 62.20 | ~35.00 | [6] |
| B−CuO | Flow cell | 1.0 M $KHCO_3$ | -1.01 | - | 55.00 | 40.00 | [7] |
| $Cu_2O$ film | Flow cell | 0.1 M $KHCO_3$ | -0.99 | Ø=10 mm | 47.88 | 38.79 | [8] |
| Cu@$Cu_2(OH)_3NO_3$ | H-cell | 0.1 M $KHCO_3$ | -1.213 | 2.00 $cm^2$ | 41.80 | ~31.00 | [9] |
| Cu@Cu NS-12 | H-cell | 0.1 M $KHCO_3$ | -1.357 | 32 × 28 $cm^2$ | 63.93 | 40.00 | [10] |

**Table S4** The calculated ECSA of CuO and Mg-CuO-*x*.

| Catalyst | $C_{dl}$ (mF) | ECSA ($cm^2$) |
|---|---|---|
| CuO | 2.80 | 70.00 |
| Mg-CuO-1 | 2.96 | 74.00 |
| Mg-CuO-2 | 3.89 | 97.20 |
| Mg-CuO-3 | 3.74 | 93.50 |

**Note for Table S4:**

Electrochemical active surface areas (ECSA) are calculated by the following formula: $ECSA=C_{dl}/C_s$ where $C_{dl}$ corresponds to the slope of the double-layer charging current versus the scan rate (ν) plot, we use a specific capacitance ($C_s$) value of 40 μF $cm^{-2}$.

**Table S5** Adsorption energy of *CO on $Cu_2O$ and Mg-$Cu_2O$-2.

| *CO | $Cu_2O$ (eV) | Mg-$Cu_2O$-2 (eV) |
|---|---|---|
| $E_{CO\text{-}total}$ | -308.22 | -308.15 |
| $E_{bare}$ | -292.62 | -292.38 |
| $E_{CO}$ | -14.8 | -14.8 |
| $E_{CO\text{-}ad}$ | -0.8 | -0.97 |

## Reference


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