*Supplementary Information*

# Effect of crystal defects on the selectivity of a bulk Cu-Zn alloy for electrocatalytic $CO_2$ reduction

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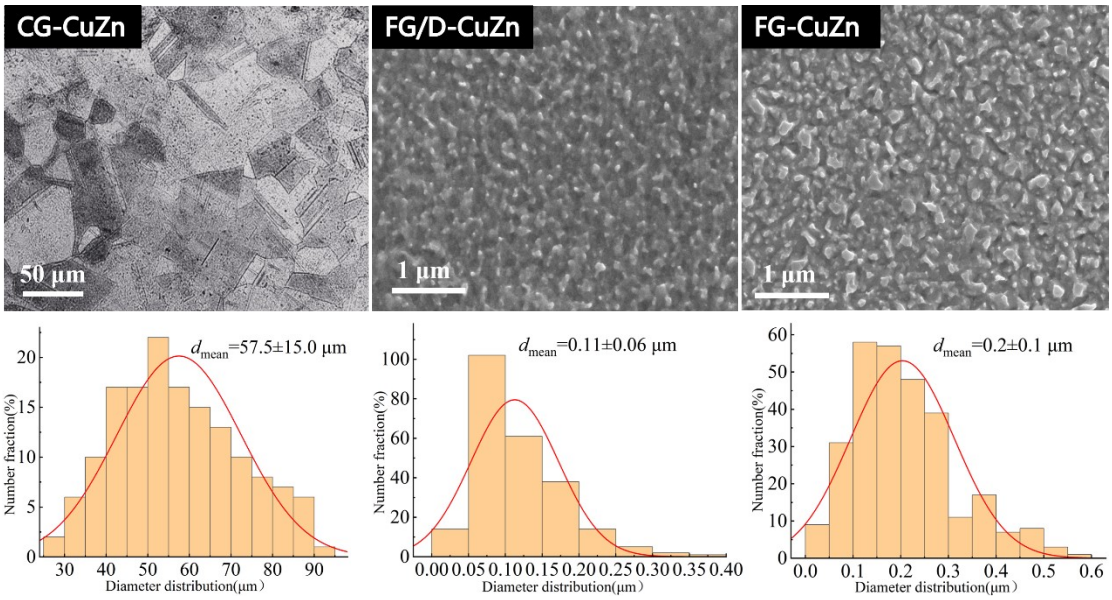


**Fig. S1** OM/SEM images of CG-CuZn, FG/D-CuZn and FG-CuZn, and the corresponding statistical chart of average grain sizes.

| | CG-CuZn | FG/D-CuZn | FG-CuZn |
|---|---|---|---|
| $D=\frac{\kappa\lambda}{\beta cos\theta}$(nm) | 98 | 12 | 25 |
| $\rho=\frac{2\sqrt{3}(\varepsilon^2)^{1/2}}{Db}$($m^{-2}$) | $0.87\times10^{14}$ | $86.55\times10^{14}$ | $6.51\times10^{14}$ |

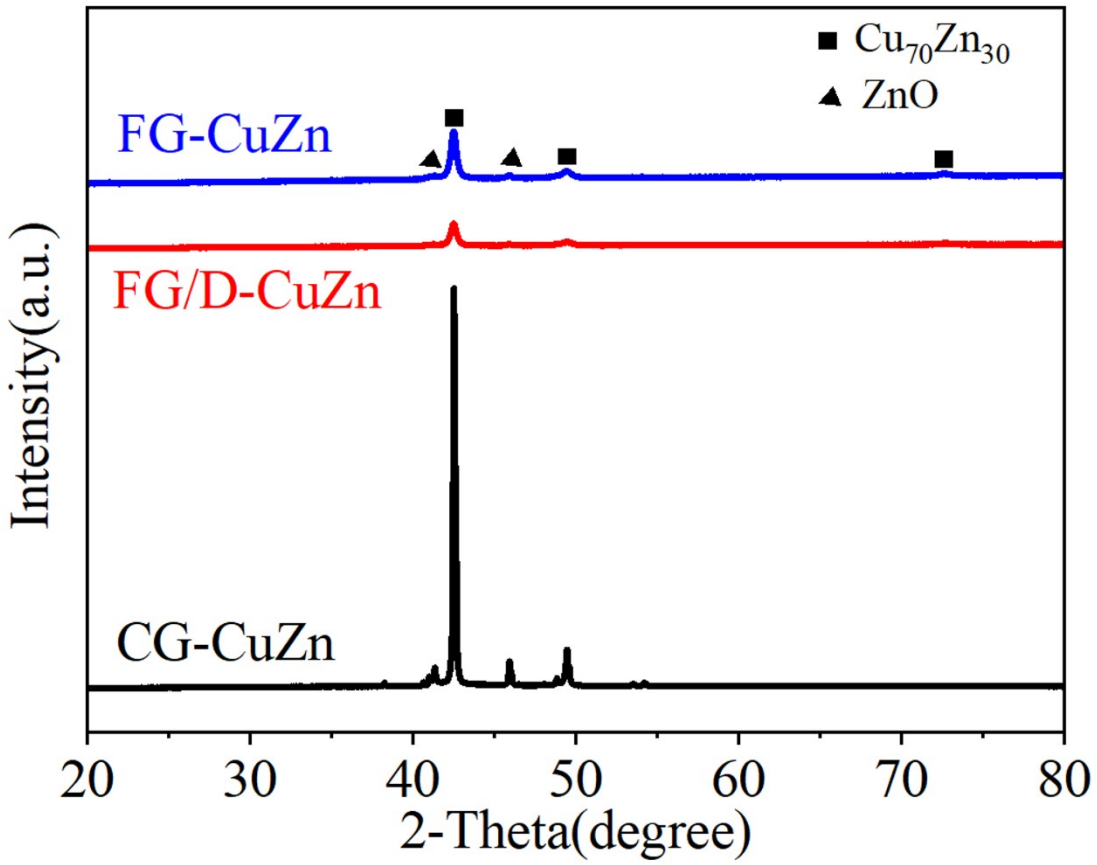


**Fig. S2** XRD patterns of CG-CuZn, FG/D-CuZn and FG-CuZn, and the corresponding calculation results of crystallite size ($D$) and dislocation density ($\rho$)

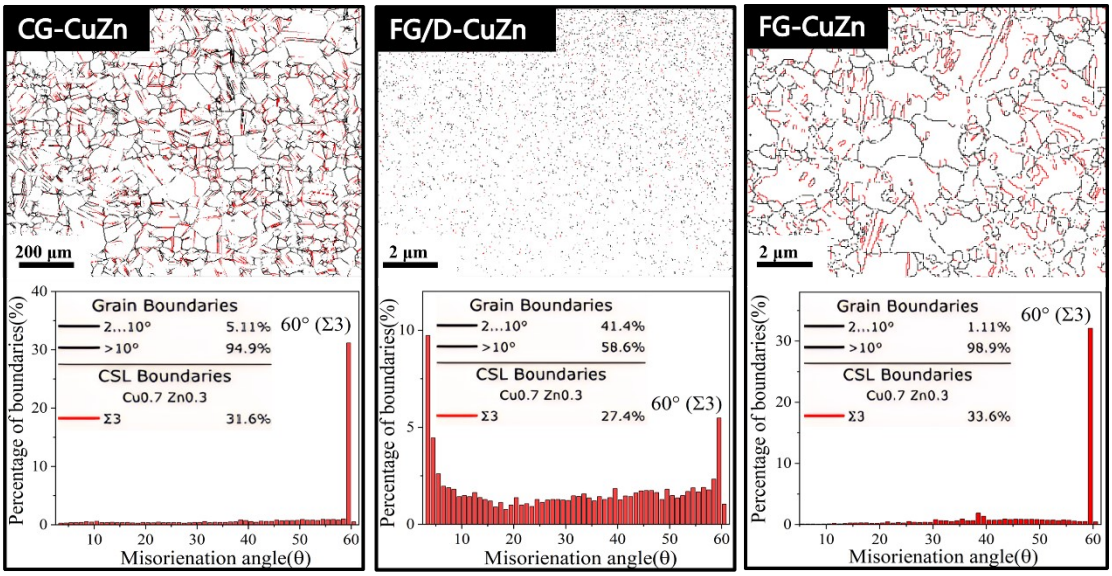


**Fig. S3** grain boundary diagrams and misorientation angle distribution diagrams of EBSD for CG-CuZn, FG/D-CuZn and FG-CuZn samples. The black lines represent general grain boundaries, and the red lines represent twin grain boundaries (∑3).

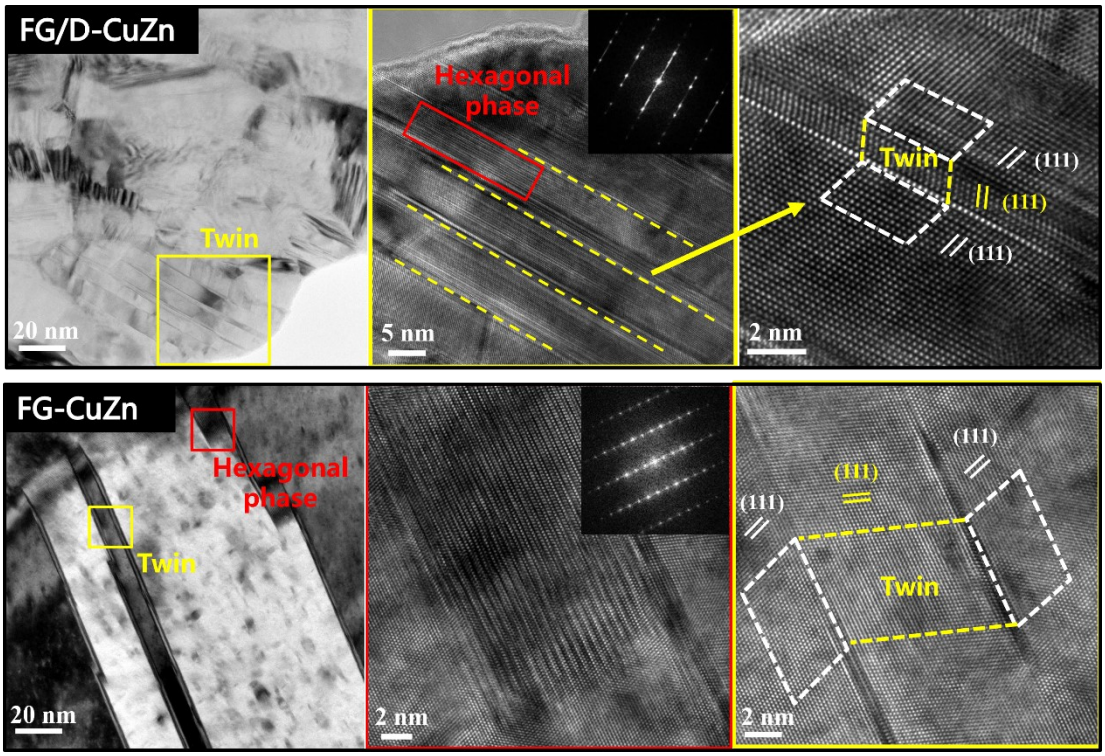


**Fig. S4** HRTEM images of the twins in FG/D-CuZn and FG-CuZn.

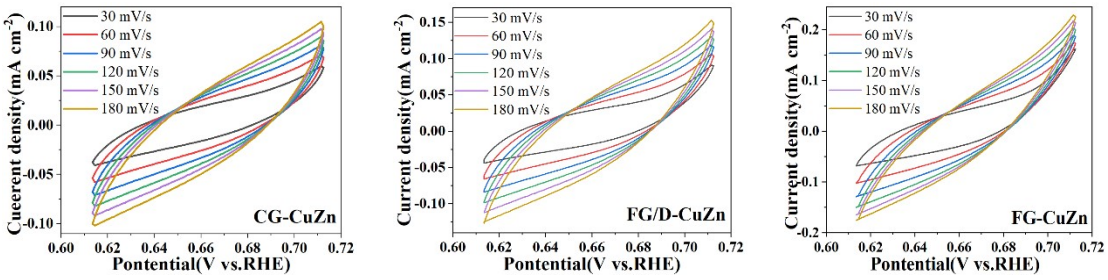


**Fig. S5** The CV of CG-CuZn, FG/D-CuZn and FG-CuZn at different scan rates.

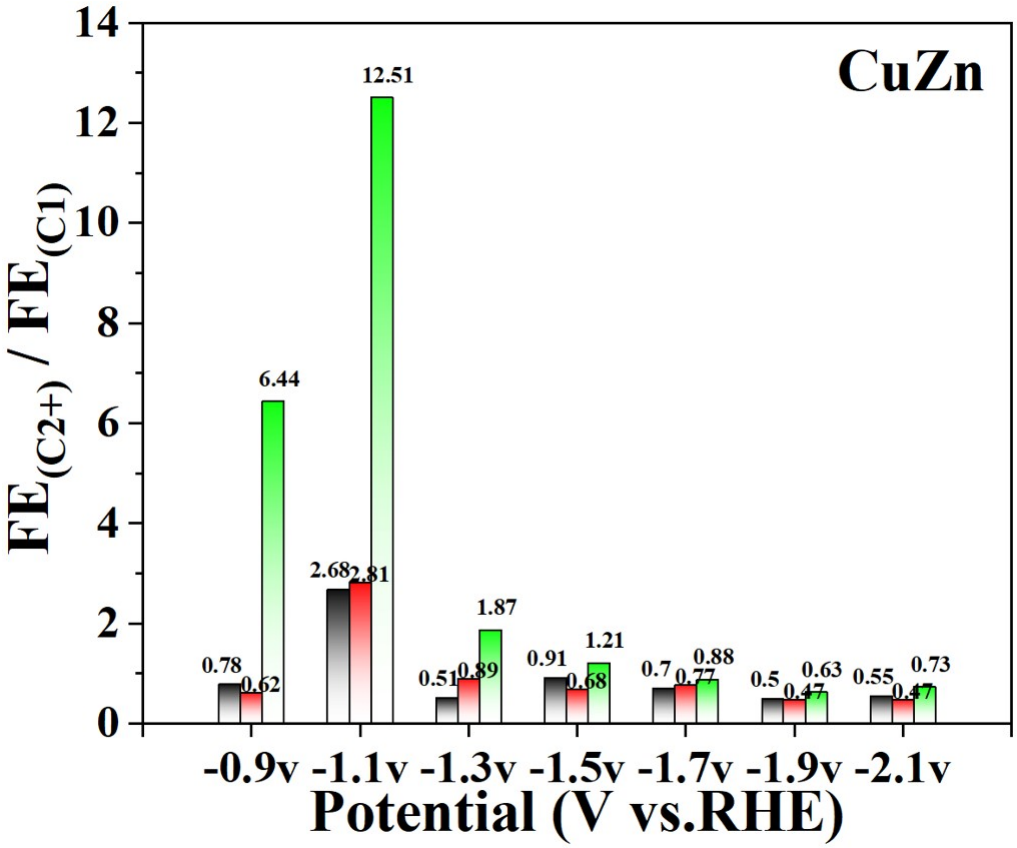


**Fig. S6** The ratio of the C2+ to C1 for CG-CuZn, FG/D-CuZn and FG-CuZn.

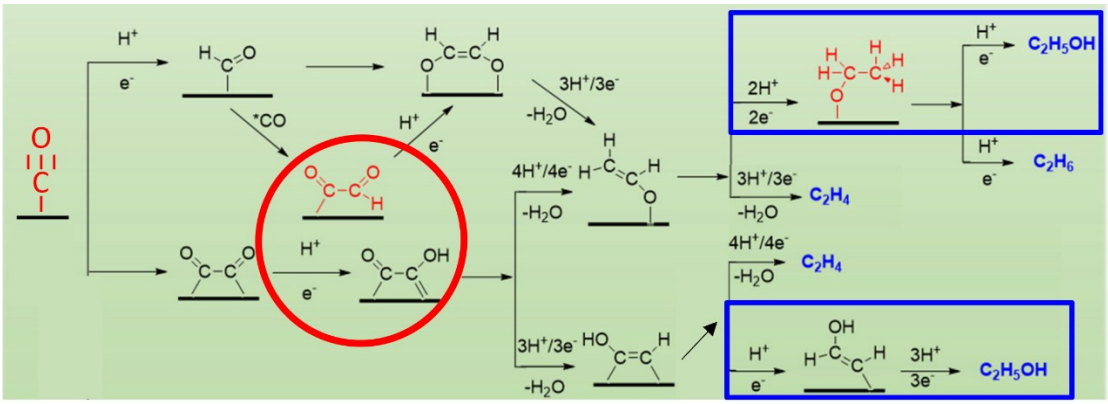


**Fig. S7** The generation pathway diagram of $C_{2+}$ products ($C_2H_4$, $C_2H_5OH$, $C_2H_6$) in the electrocatalytic $CO_2RR$.

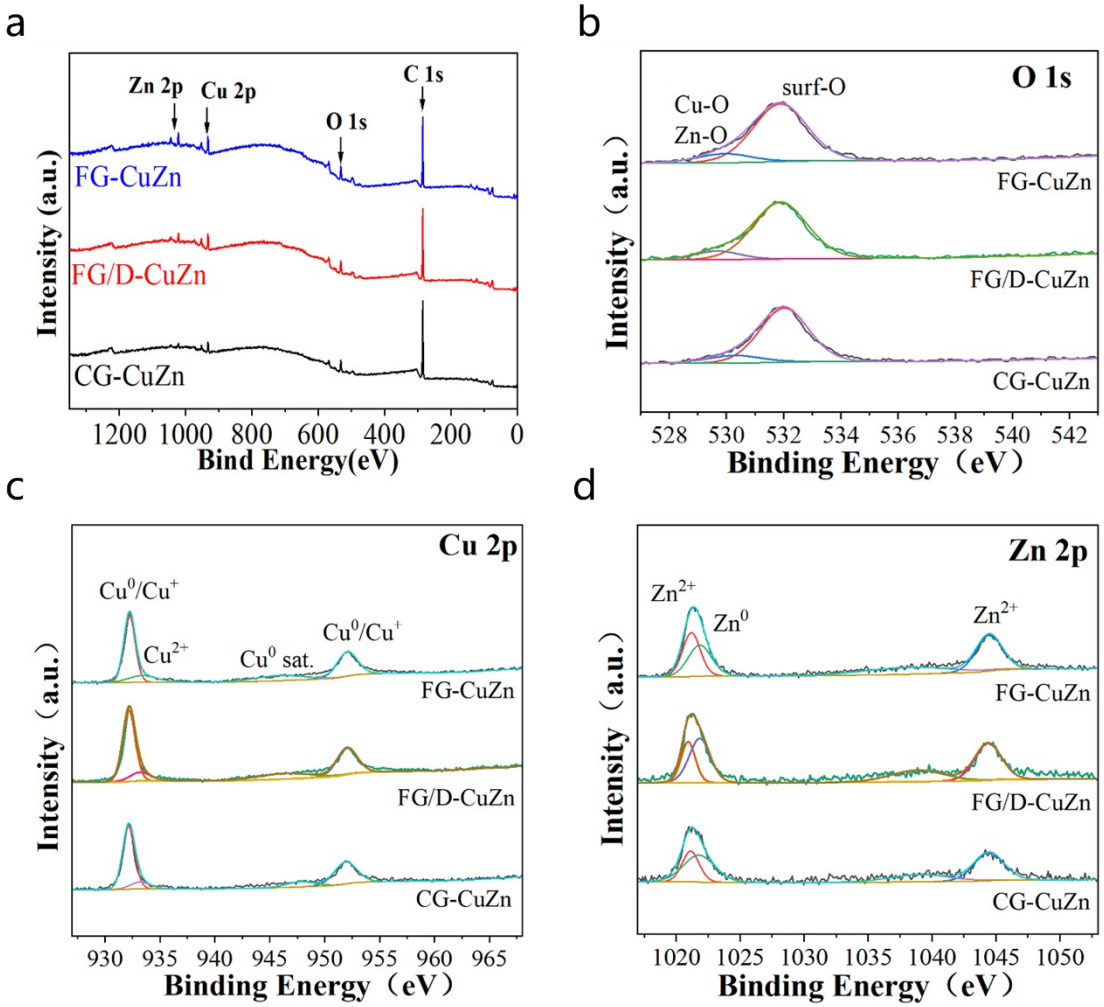


**Fig. S8** XPS of CG-CuZn, FG/D-CuZn and FG-CuZn: (a) survey spectrum; (b) O 1s; (c) Cu 2p; (d) Zn 2p.

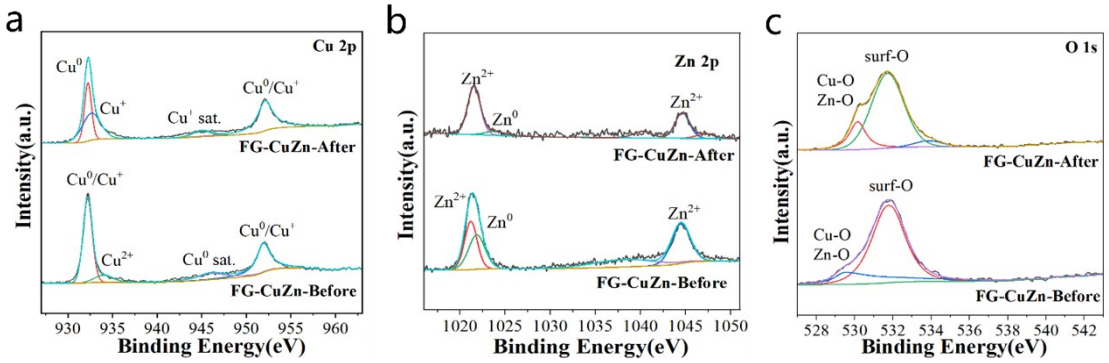


**Fig. S9** XPS spectra of FG-CuZn before and after 30 min of CO2RR: (a) Cu 2p; (b) Zn 2p; (c) O 1s.