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Supporting Information

Bicarbonate Electroreduction to Multicarbon Products Enabled by Cu/Ag Bilayer Electrodes and Tailored Microenvironments

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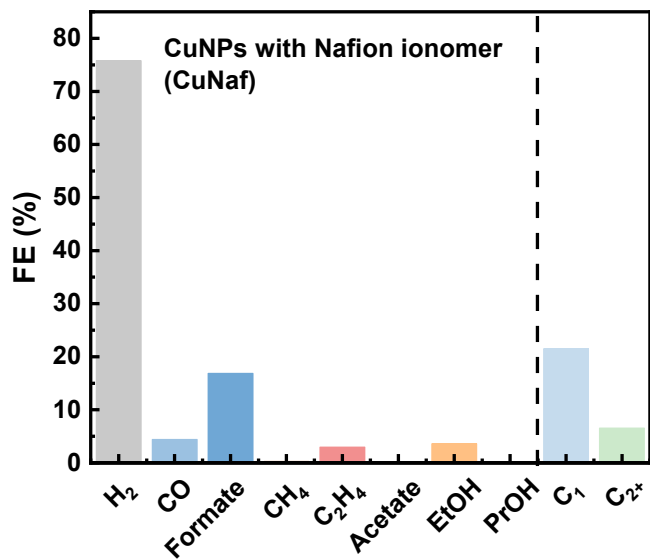


Figure S1. Product distributions of bicarbonate conversion of CuNaf catalyst on H23 carbon paper. The bicarbonate conversion was performed in MEA-based flow cell with BPM at a current density of 100 mA cm^{-2} for 1 h.

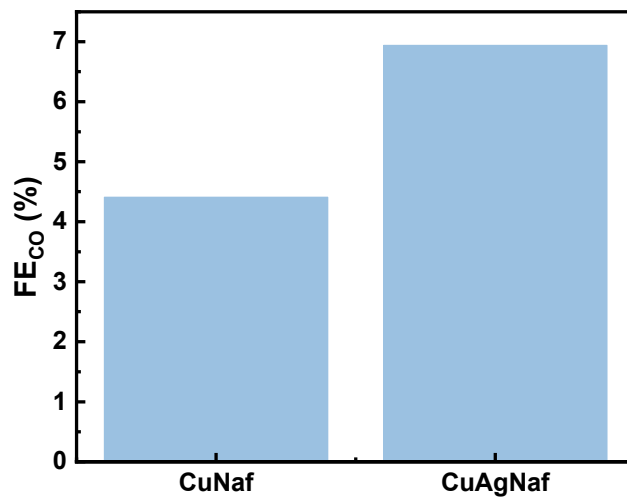


Figure S2. Comparison of FE_{CO} of CuNaf and CuAgNaf catalysts on H23 carbon paper. The bicarbonate conversion was performed in MEA-based flow cell with BPM at a current density of 100 mA cm⁻² for 1 h.

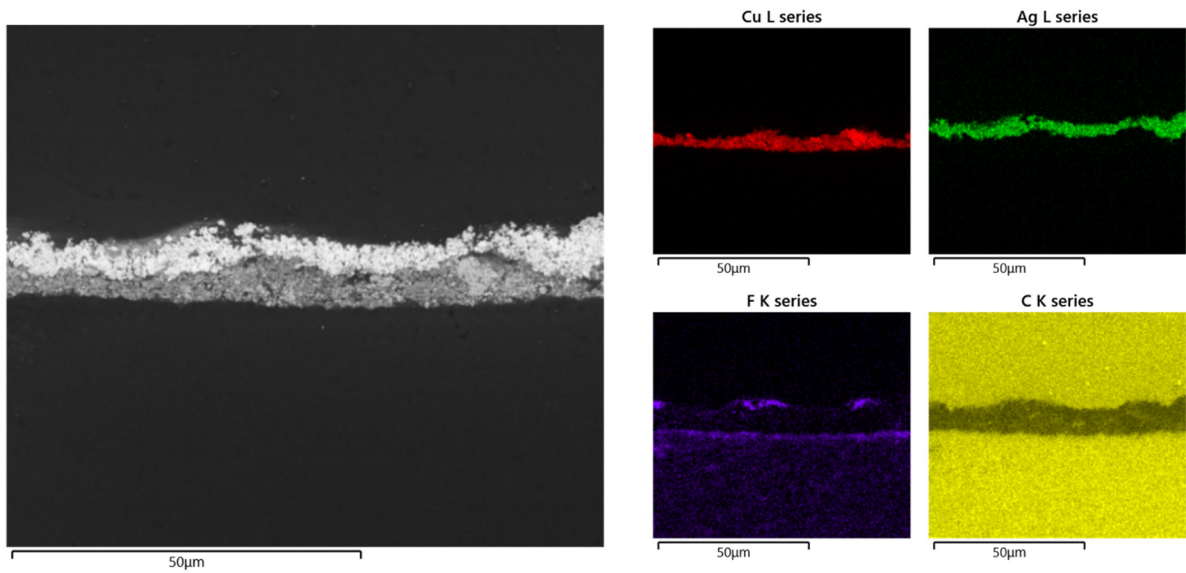


Figure S3. SEM image of bilayer CuSus/AgNaf electrode and corresponding EDX mapping. The carbon signal above the Ag layer is attributed to the epoxy matrix used for sampling.

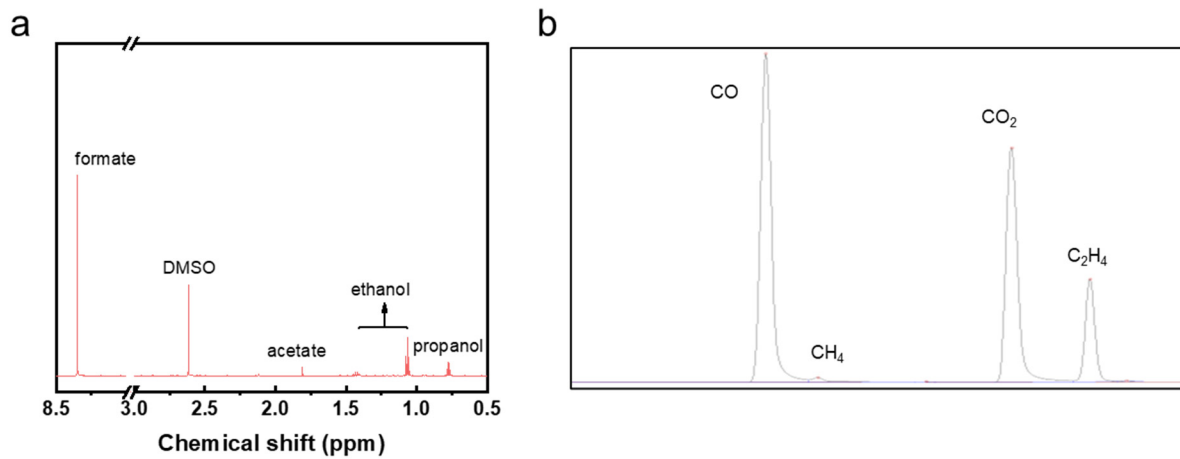


Figure S4. (a) ^1H NMR and (b) GC spectrum (FID signal) of the liquid and gaseous products, respectively.

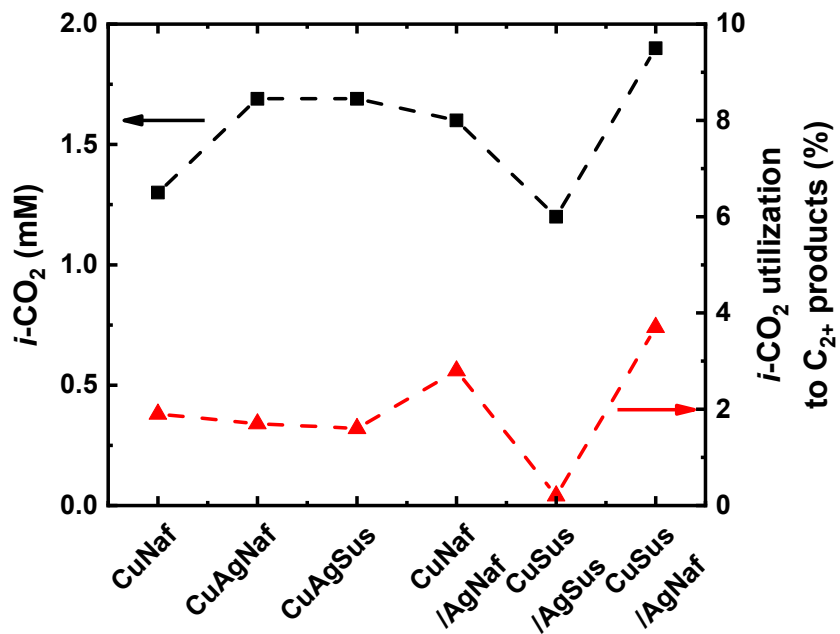


Figure S5. $i\text{-CO}_2$ formation and its utilization to C_{2+} products of six different electrode configurations during the bicarbonate conversion (100 mAcm^{-2} for 1 h).

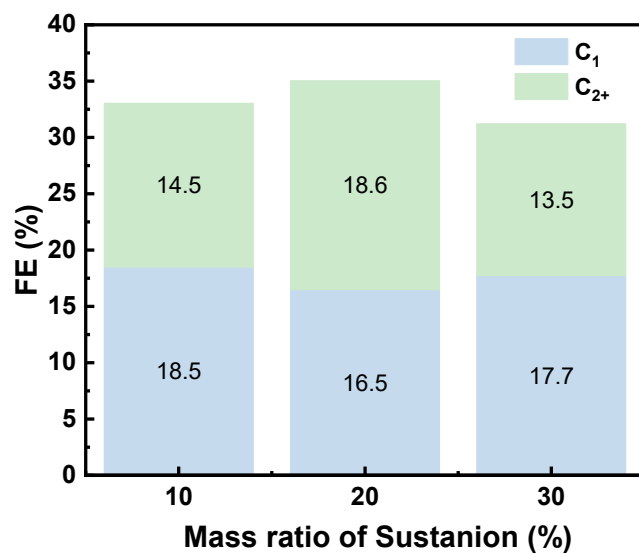


Figure S6. FE of C₁ and C₂₊ products as function of mass ratio of Sustanion on Cu layer. The Nafion content in Ag layer was maintained at 20 wt.%. The bicarbonate conversion was performed at a current density of 100 mA cm⁻² for 1 h.

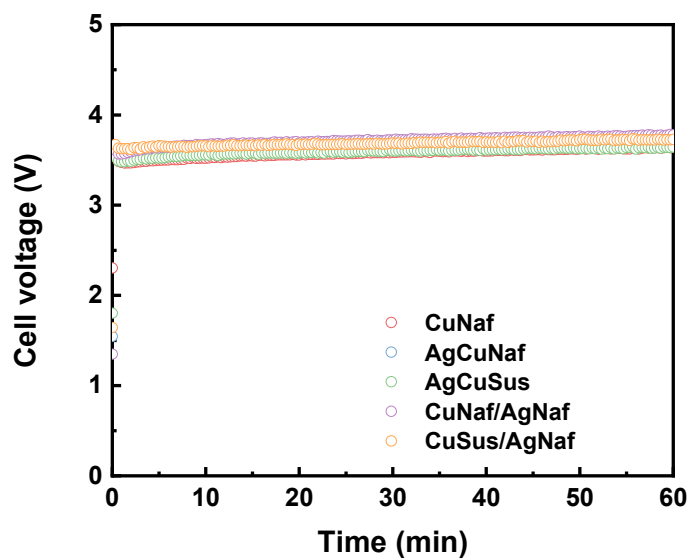


Figure S7. Cell voltage profiles of five different configurations of Cu and/or Ag catalysts with different ionomer (Nafion, Sustanion) on H23 carbon paper. The bicarbonate conversion was performed in MEA-based flow cell with BPM at a current density of 100 mA cm^{-2} for 1 h.

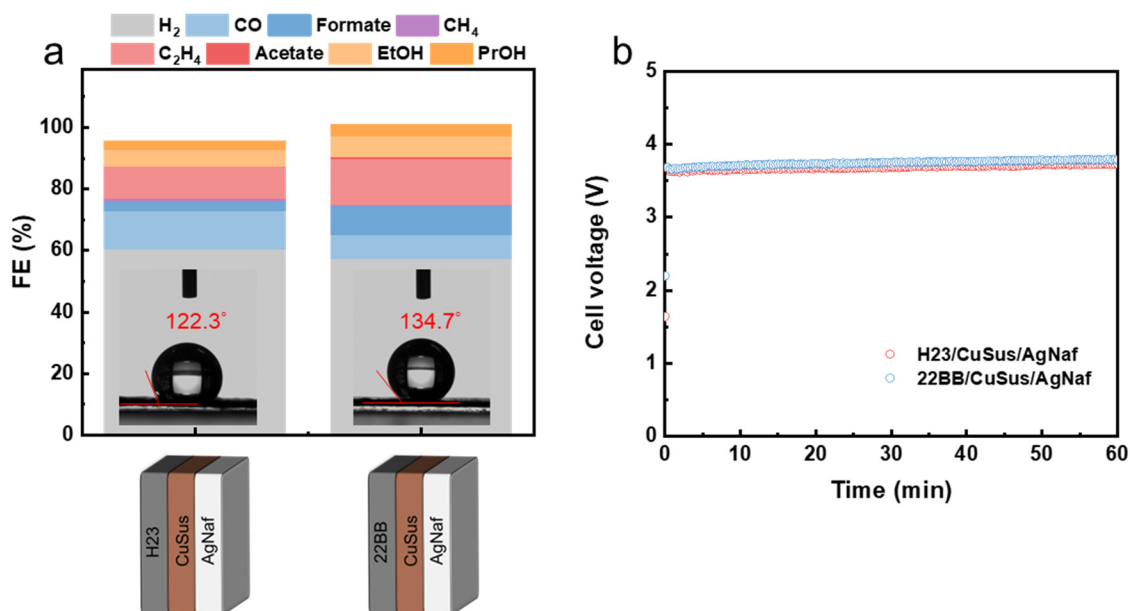


Figure S8. Comparison of (a) FE (inset: images of contact angle measurement on H23 and 22BB carbon paper without catalyst layer) and (b) cell voltages of bilayer CuSus/AgNaf electrode on H23 and 22BB carbon paper. The bicarbonate conversion was performed in MEA-based flow cell with BPM at a current density of 100 mA cm^{-2} for 1 h.

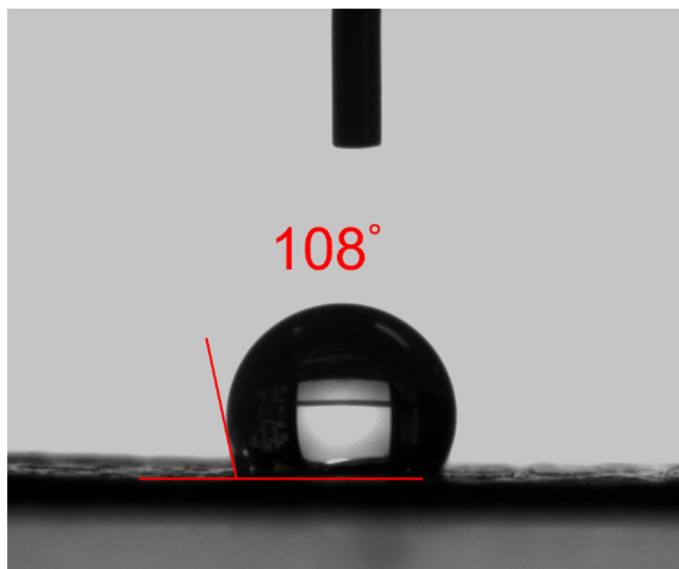


Figure S9. Contact angle measurement of CuSus/AgNaf-P50 after 1 h electrolysis.

Table S1. Comparison of eCO₂R performance of state-of-the-art coupled CO₂ capture and reduction systems.

Catalysts	CO ₂ capture solution	Membrane	products	FE (%)	Current density (mA cm ⁻²)	Cell voltages (V)	Ref
CuSus/AgNaf-p50	3 M KHCO ₃	BPM	C ₂ ⁺	41.6	100	3.9	This work
Cu nanoparticles	1 M K ₂ CO ₃	BPM	C ₂ ⁺	10.1	150	n/a	[1]
Cu foam	3 M KHCO ₃ with CTAB	BPM	CH ₄	34	120	> 6	[2]
Ag foam	3 M KHCO ₃	BPM	CO	59	100	3.6	[3]
Ag nanoparticles	3 M KHCO ₃	BPM	CO	37	100	n/a	[4]
Ag nanoparticles	3 M KHCO ₃	BPM	CO	82	100	n/a	[5]
Electrodeposited Bi	3 M KHCO ₃	BPM	HCOO ⁻	64	100	> 4	[6]
Ag nanoparticles	2M MEA with 2M KCl	CEM	CO	72	50	-0.8 vs. RHE	[7]

References

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