

HYBRID MOLECULE-METAL INTERFACES FOR ELECTROCHEMICAL CO₂ REDUCTION TO MULTICARBON PRODUCTS

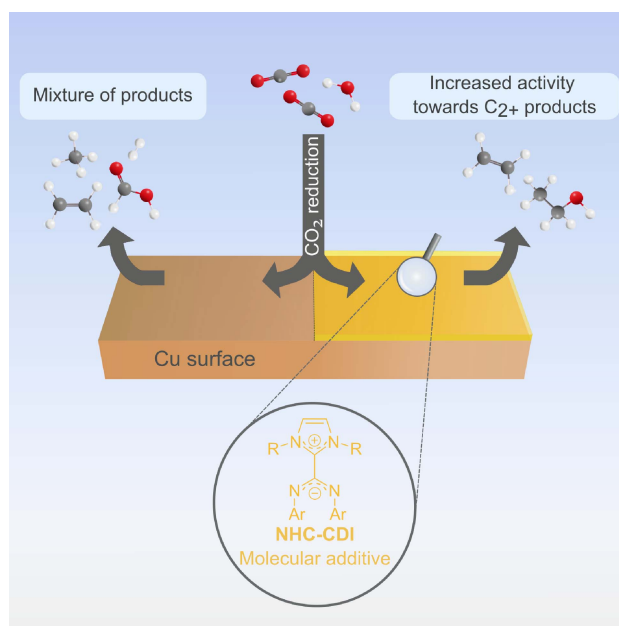
Kirstine Nygaard Kolding^{a,b}, Matias Bretlau^{a,b}, and Alonso Rosas-Hernández^{a,b}

^aInterdisciplinary Nanoscience Center, Aarhus University, Aarhus, Denmark

^bDepartment of Chemistry, Aarhus University, Aarhus, Denmark

The unprecedented and ever-increasing levels of greenhouse gases such as carbon dioxide in the Earth's atmosphere stemming primarily from the burning of fossil fuels have led to a global climate crisis.[1] For this reason, green technologies that accelerate the transition towards carbon-neutral energy sources are highly needed. The electrochemical CO₂ reduction reaction (CO₂RR), powered by renewable-derived electricity, is a promising approach for the seasonal storage of electrical energy by converting CO₂ into molecules that can be used as energy vectors and value-added chemicals.[2,3] Utilization of this reaction requires the development of efficient and selective catalysts and here copper-based catalysts show unique potential in the conversion of CO₂ into multicarbon products.[4,5]

In this work, we present the preparation, characterization, and performance evaluation of hybrid copper-based catalysts modified with *N*-heterocyclic carbene-carbodiimide (NHC-CDI) ligands in the CO₂RR. Prepared via an electrodeposition method, the catalysts exhibit up to a 10-fold increase in the activity for multicarbon products such as ethylene and ethanol depending on the NHC-CDI ligand employed. This work exemplifies the ability of molecular additives to alter the intrinsic catalytic activity of copper surfaces in the CO₂RR.



[1] Anderson T. R., Hawkins, E., Jones, P. D. *Endeavour* **2016**, *40*, 178-187.

[2] Appel, A. M.; Bercaw, J. E.; Bocarsly, A. B.; Dobbek, H.; DuBois, D. L.; Dupuis, M.; Ferry, J. G.; Fujita, E.; Hille, R.; Kenis, P. J. A.; Kerfeld, C. A.; Morris, R. H.; Peden, C. H. F.; Portis, A. R.; Ragsdale, S. W.; Rauchfuss, T. B.; Reek, J. N. H.; Seefeldt, L. C.; Thauer, R. K.; Waldrop, G. L. *Chem. Rev.* **2013**, *113*, 6621– 6658.

[3] Franco, F.; Rettenmaier, C.; Jeon, H. S.; Roldan Cuenya, B. *Chem. Soc. Rev.* **2020**, *49*, 6884– 6946.

[4] Hori, Y.; Kikuchi, K.; Murata, A.; Suzuki, S. *Chem. Lett.* **1986**, *15*, 897– 898.

[5] Kuhl, K. P.; Cave, E. R.; Abram, D. N.; Jaramillo, T. F. *Energy Environ. Sci.* **2012**, *5*, 7050– 7059.