

Extra Session of Focus Materialchemie – Tuesday, 23.01.2024 11:00 – @ Seminarraum Lehar 03 (TU-Wien, Getreidemarkt 9, BC, OG. 03) – [join us](#) on ZOOM (ID: 983 0066 2349)

Dynamic catalysts to improve eCO₂RR efficiency: Tandem Electrocatalysis vs Direct Carbonate Reduction.

E. Ibáñez-Alé^{a,b}

^a Institute of Chemical Research of Catalonia (ICIQ-CERCA), The Barcelona Institute of Science and Technology (BIST), Av. Països Catalans, 16, 43007 Tarragona, Spain.

^b Universitat Rovira i Virgili, Avinguda Catalunya, 35, 43002 Tarragona, Spain.

eibanez@iciq.es

The electroreduction of CO₂ driven by renewable electricity is a sustainable way to store energy by generation of synthetic fuels. Copper is one of the most promising pure metals for generation of multicarbon products [1]. However, the variety of product distributions is not easily controllable, which makes necessary a high understanding of the reaction mechanisms to improve the catalyst performance. One of the challenges in CO₂ electroreduction reaction (eCO₂RR) on Cu is the rapid reaction of dissolved CO₂ with hydroxide (OH⁻) to form carbonates (CO₃²⁻), which compromises the efficiency of the catalyst [2]. To bypass the carbonate formation the tandem catalyst approach has showed high selectivity towards C₂₊ products by decoupling the reaction into two steps, CO₂-to-CO and CO-to-C₂₊[3]. For this reason, we aim to achieve a better understanding of CO producing catalysts such as atomically precise gold nanoclusters, focusing on the different features that can tune their selectivity. Moreover, we found an alternative by applying a dynamic operation approach that can turn inert CO₃²⁻ into reactive. Herein, we combined Raman spectroscopy and DFT simulations to disclose a pulse potential enhanced pathway for direct carbonate reduction to formate during eCO₂RR on a Cu catalyst [4]. Thus, this prove of concept opens the possibility of reducing these CO₃²⁻ species and improving the eCO₂RR efficiency.

References

- [1] Nitopi, S.; Bertheussen, E.; Scott, S. B.; Liu, X.; Engstfeld, A. K.; Horch, S.; Seger, B.; Stephens, I.; Chan, K.; Hahn, C.; Nørskov, J. K.; Jaramillo, T. F.; Chorkendorff, I. *Chem. Rev.* **2019**, *119*, 7610–7672.
- [2] Rabinowitz, J. A.; Kanan, M. W. *Nat. Commun.* **2020**, *11*, 5231.
- [3] Chen, Y.; Li, X.-Y.; Chen, Z.; Ozden, A.; Huang, J. E.; Ou, P.; Dong, J.; Zhang, J.; Tian, C.; Lee, B.-H.; Wang, X.; Liu, S.; Qu, Q.; Wang, S.; Xu, Y.; Miao, R. K.; Zhao, Y.; Liu, Y.; Qiu, C.; Abed, J.; Liu, H.; Shin, H.; Wang, D.; Li, Y.; Sinton, D.; Sargent, E. H. *Nat. Nanotech.* **2023**, 10.1038/s41565-023-01543-8.
- [4] Ma, H.; Ibáñez-Alé, E.; Ganganahalli, R.; Pérez-Ramírez, J.; López, N.; Yeo, B. S. *J. Am. Chem. Soc.* **2023**, *145*, 45, 24707-24716.