Unintended Cation Crossover in CO_2 Conversion MEA Cells: Causes and Effects

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Abstract

Membrane electrode assemblies in gas diffusion cells enable CO_2 reduction at industrially relevant rates, yet their long-term operational stability is often limited by the formation of solid precipitates (e.g. K_2CO_3) in the cathode pores. This is a consequence of a combination of 1) local alkalization due to the electrochemical reaction, 2) generation of (bi)carbonate by chemical reaction of CO_2 with the alkaline electrolyte, and 3) the presence of alkali metal cations. In catholyte-free, zero-gap cells using anion exchange membranes, the presence of electrolyte cations at the cathode is the result of unintended crossover from the anolyte, and a detailed understanding of the factors enabling this crossover is lacking. Here we show that the anolyte concentration governs the flux of cation migration through the membrane, and this substantially influences the behaviors of copper catalysts in catholyte-free CO_2 electrolysers. Systematic variation of the anolyte ionic strength (using aqueous KOH or $KHCO_3$) correlated with drastic changes in the observed product selectivity – most notably, at low ionic strength, Cu catalysts produced predominantly CO, in contrast to the mixture of C_{2+} products typically observed on Cu. In this talk, we examine the factors influencing ion crossover and the resulting effects on catalyst structure and activity, under conditions of both CO_2 and CO reduction. Operando X-ray absorption spectroscopy and quasi in situ X-ray photoelectron spectroscopy were used to study how the catalyst is affected by operation conditions. Our results show that even in catholyte-free cells, cation effects (including unintended ones) can significantly influence reaction pathways, and this must be considered in future development of catalysts and devices.

El-Nagar, G. A., Haun, F., Gupta, S., Stojkovikj, S. & Mayer, M. T. Unintended cation crossover influences CO2 reduction activity in Cu-based zero-gap electrolysers. *Nature Communications*14, 2062 (2023) doi: doi.org/10.1038/s41467-023-37520-x

Figure 1

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