



PhD THESIS DEFENSE: Engineering Catalyst-Ionomer Interfaces for Carbon-Efficient CO₂ Electrolysis and Technology Prospects

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10:30

ICFO Auditorium

The electrochemical reduction of CO₂ (CO₂E) offers a promising route to convert greenhouse gas emissions into value-added chemicals and fuels. However, achieving performance metrics that enable the technoeconomic and sustainable viability of CO₂E remains challenging. This is especially acute in the case of multicarbon products (C₂+), important precursors for energy fuels and manufacturing, where achieving combined selectivity and carbon utilisation under industrially relevant conditions is challenged by undesired competing reactions. This thesis explores the design and implementation of new strategies to modulate electrochemical interfaces in CO₂E to overcome this barrier. These

are based on the implementation of ionomer coatings that specifically address key reactants and intermediates in CO₂E.

A key contribution is the development and mechanistic elucidation of ion management channels (IMCs), formed by co-distributing cation and anion exchange ionomers (CEIs and AEIs) within the catalyst layer. This architecture enables local regulation of hydroxide and cation populations, mitigating *OH poisoning and enhancing *CO adsorption, critical step for promoting C-C coupling and C₂+ product formation

The ionomer-catalyst interface is comprehensively characterised using SEM-EDS, FTIR, XPS, KPFM, contact angle measurements, cyclic voltammetry, and EIS. In situ Raman spectroscopy reveals the dynamic evolution of surface species, confirming that excessive *OH accumulation suppresses C₂+ selectivity, while IMCs restore favourable interfacial conditions. These insights are correlated with improved electrochemical performance, carbon efficiency, and stability across a wide range of operating conditions, including high acidic environments

The IMC concept is further implemented in membrane electrode assembly (MEA) device operating under neutral pH. Preliminary results demonstrate improved performance and reduced cell voltages for IMC-based electrodes, indicating compatibility with scalable reactor platforms and commercially viable components

The thesis concludes with a broader analysis of the challenges facing CO₂E at scale. Key bottlenecks, such as the reliance on iridium anodes and fluorinated membranes, are critically assessed, and material and performance targets for gigaton-scale deployment are proposed. A techno-economic and life-cycle analysis outlines trade-off between performance, cost, and sustainability, while global scaling efforts are reviewed. Benchmarking protocols are proposed to bridge the gap between laboratory research and industrial implementation

Together, this work advances a cohesive framework for interfacial engineering in CO₂E, linking molecular-level understanding to device-scale integration, and providing pathway toward industrial deployment

Friday February 20, 10:30 h. ICFO Auditorium

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