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

BLANCA M. BELSA CARNE

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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

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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 managemen channels (IMCs), formed by co-distributing cation and anion exchange ionomers (CEIs an AEIs) within the catalyst layer. This architecture enables local regulation of hydroxide an 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 Rama spectroscopy reveals the dynamic evolution of surface species, confirming that excessiv *OH accumulation suppresses C2+ selectivity, while IMCs restore favourable interfacia conditions. These insights are correlated with improved electrochemical performance carbon efficiency, and stability across a wide range of operating conditions, including highl acidic environments

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

The thesis concludes with a broader analysis of the challenges facing CO2E at scale. Ke bottlenecks, such as the reliance on iridium anodes and fluorinated membranes, are criticall 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, an sustainability, while global scaling efforts are reviewed. Benchmarking protocols ar 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

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Thesis Director: Prof. Dr. Francisco Pelayo Garcia de Arquer
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