Summary

This thesis focuses on developing electrochemical strategies to convert CO_2 into syngas (CO and H_2), which can be further used in hydroformylation reactions to produce value-added aldehydes to support the electrification of chemical processes for a more sustainable industry.

Chapter 1 - Introduction presents the main topic of the thesis by explaining the broader context and motivations that drive the research.

Chapter 2 - Assessing the CO_2 capture and electro-reduction in imidazolium-based ionic liquids: role of the ion exchange membrane Explores strategies for stable electrochemical CO_2 reduction(eCO_2RR) to syngas using ionic liquid (IL)-based electrolytes. The influence of electrochemical cell design (single vs H-type) and membrane type (AEM vs BPM) on syngas stability and selectivity is studied through experimental work and COMSOL Multiphysics modeling.

Chapter 3 - Mechanistic insights and degradation pathways of Ionic Liquids at electrode-electrolyte interfaces: a combined operando-SEIRAS and electrochemistry study Investigates the mechanistic behavior and degradation pathways of ILs at the electrode-electrolyte interface, combining electrochemical data (collected at Politecnico di Torino) and SEIRAS spectroscopy (collected at Osaka University). The focus is on understanding selectivity mechanisms at the cathode and IL degradation at the anode.

Chapter 4 - Optimization of hydroformylation reaction at low pressure and temperature for the coupling with eCO_2RR Focuses on optimizing hydroformylation (HF) of 1-octene and limonene under mild conditions, making the reaction compatible with near ambient-condition eCO_2RR. Preliminary results show promising aldehyde yields, notably nonanal from 1-octene. The experiments were conducted in the laboratory of EURECAT, Tarragona, Spain. A first coupling experiment was conducted by combining the optimized conditions in Chapter 2 and Chapter 4, obtaining nonanal from 1-octene HF exploiting syngas from eCO_2RR.

Chapter 5 – e-Tandem catalysis towards chemical industry electrification: the hydroformylation challenges. Offers perspectives on integrating electrochemical and thermochemical processes, emphasizing the challenges of hydroformylation in the context of chemical industry electrification.

Thesis Keywords: CO_2 electroreduction, Ionic Liquids, IR spectroscopy, SEIRAS, Hydroformylation, Tandem Catalysis.