Master thesis project – Process and Energy department

Topic : Sequential cascade electrocatalytic CO_2RR at high pressures

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In a greener future, society's power will come from an increased share of sustainable but fluctuating energy sources, like solar or wind. To enable this future, energy storage on a large-scale is necessary. In this context, the conversion of electricity into energy stored in chemical bonds is key since the scale will be huge compared to other forms of energy storage (e.g. batteries). Fluctuation in power will lead to periods of cheap and abundant electricity, and here electrochemical conversion comes into the picture. In this way electricity can be used to convert CO_2 (or other sustainable feed stocks) into useful products - for the storage and later recovery of that energy, but also for industrial usage.

Cascade catalysis is a multistage process where reactants are converted to intermediates in the first stage(s) that are then consumed under the optimal conditions to the desired products in the final stage. One of the primary focuses in electrochemical CO_2RR has been in the development of suitable catalysts to produce multi-carbon products like ethylene, ethanol, propanol etc, to name a few. However, this route is limited by the scaling relations (competing binding energies of different intermediates) which prevent the optimization of every step in the entire mechanism resulting in poor selectivity for anyone particular product. Recently cascading has become interesting as a method to break the scaling relationship, when we look at the mechanism of CO_2RR with CO as a primary intermediate as demonstrated by Gurudayal et al.[1]. There are several highly selective CO_2RR catalysts for CO production (Ag, for example) and then for the further reduction of CO into the target products Cu is highly tuneable and active electrode. With improved selectivity, conversion and reduced potentials, this new route is promising but it is still in infancy. Parameters like operating conditions, catalyst configurations, flow behavior etc. can still be improved and studied to understand the underlying phenomena.

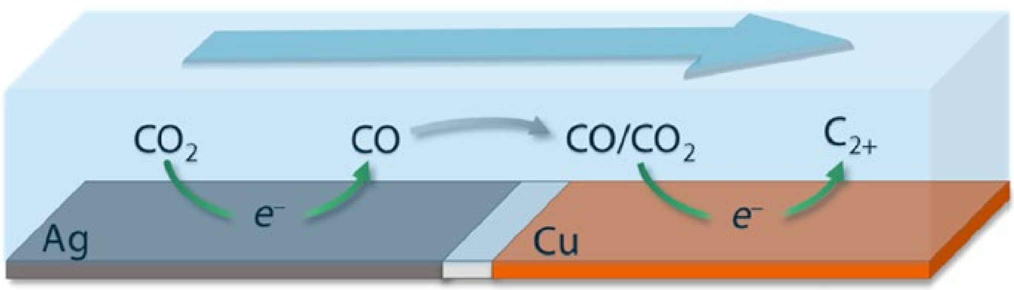
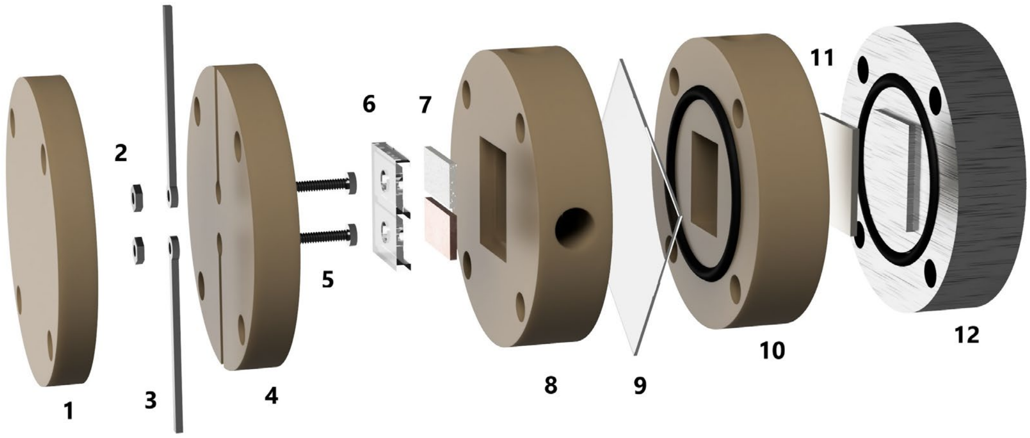


Figure 1: Sequential cascade process

Overall, within this project scope there are several opportunities available to work upon: A kinetic mass transport modelling to determine the effect of pressure on cascade catalysis to determine the optimal operating conditions (Project 1); An experimental work to help validate the model (Project 2); Or a combination of both (Project 3). We already have a novel design that exists and a preliminary model. **So you will not be starting from scratch (you are**



expected to build upon the existing work).

Requirements**:** The ability to program especially in MATLAB or Python. Knowledge of electrochemical systems will be essential for this project, but can be learned as the project progresses. Finally, strong knowledge of reactor modelling techniques will be useful. Familiarity with mechanical design and manufacture. Knowledge of electrochemical systems would be an asset or at least a desire to learn. Finally, strong project management skills are an asset because this work will involve managing lead times for ordered and manufactured parts.
This masters project is part of the ongoing activities at the Process and Energy Department (P&E) as part of collaborations with industrial partners focused on the development of a reactor system to study and understand cascade catalytic CO_2 electrolysis.

The main objectives of this project are as follows:

- Literature study to come up with ideas to further optimize the preliminary system/model available.

Figure 2 : Existing high pressure reactor with multiple cathode

- Project 1: Modelling to provide key insights into the behaviour of the system under optimized conditions.
- Project 2: Experimental validation of the model. As a first step, you will have to reproduce the results of the previous work [1].
- Critical analysis backing the obtained results and insights into the challenges that remain with future outlook.

The final thesis report should comply with the guidelines of the Large Scale Energy Storage section of Process and Energy department.

References:

1. G. Gurudayal, David Perone, Saurabh Malani, Yanwei Lum, Sophia Haussener, and Joel W. Ager. Sequential Cascade electrocatalytic Conversion of Carbon Dioxide to C-C Coupled Products. *ACS Applied EnergyMaterials*, 2019.
2. Quincy Wols, Design of a High-Pressure CO 2 Reduction Reactor System Design of a CO 2 Reduction Reactor System Master’s Thesis, TU Delft, 2019.
3. Andrew R. T. Morrison, Vincent van Beusekom, Mahinder Ramdin, Leo J. P. van den Broeke, Thijs J. H. Vlugt, and Wiebren de Jong. Modeling the Electrochemical Conversion of Carbon Dioxide to Formic Acid or Formate at Elevated Pressures. *Journal of The Electrochemical Society*, 166(4):E77–E86, 2019.