

Decarbonization through Conversion of CO₂ and Renewable Energy to Chemicals Fuels via Synthesis Gas

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Background and Motivation

- Whilst hydrogen shows promise as the energy source of the future, the success of global efforts to achieve net-zero CO₂ emissions is contingent on multiple methods of closing the carbon loop^[1]
- Utilizing renewable energy to drive the electrochemical reduction of CO₂ (CO₂RR) to value-added chemicals (i.e. renewable Power-to-X) offers the dual advantage of converting intermittent renewable energy to a stable chemical form, whilst simultaneously reducing the emission of waste CO₂ to the atmosphere^[2]
- A highly favourable product of CO₂RR is synthesis gas (or syngas), generated by the conversion of CO₂ and water (present as humidified CO₂) to CO and H₂
- Syngas is a key building block for the production of a wide range of chemicals, including synthetic liquid fuels such as ethanol and methanol, and plastic packaging materials^[3,4]
- The use of gas diffusion electrodes in an electrolyzer system allows for vapour-phase reactants, high pressures, and enhanced current densities, offering a promising route for the industrial application of syngas production from waste CO₂ and water^[5] (**Figure 1**)

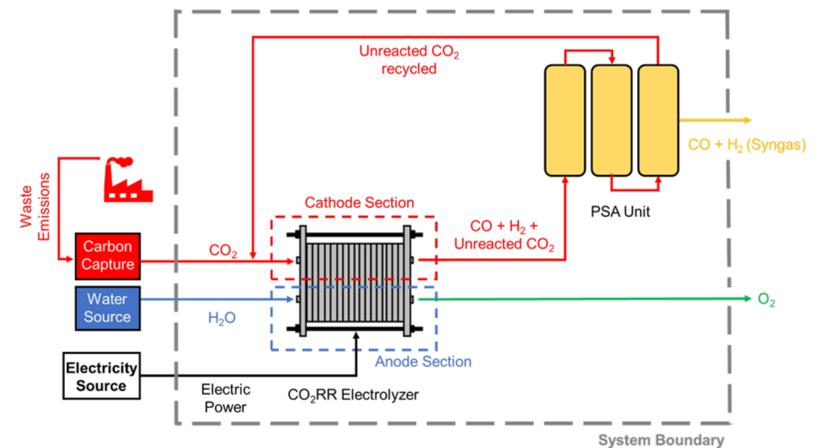


Figure 1: Envisioned commercial CO₂RR to syngas system. Waste CO₂ emissions are captured from industry and fed into the cathode section of the CO₂RR electrolyzer (as humidified CO₂) to generate syngas.

Methods and Experimental Setup

- Design and synthesis of a range of metal, metal-carbon, and single atom catalysts for CO₂RR to syngas^[6-8] (**Figure 2**)
- Macroscale electrode engineering, involving fabrication of gas diffusion electrodes exhibiting specialised components devoted to charge transfer, liquid inhibition, vapour transport, and catalyst bonding (**Figure 3**)
- System development and optimisation of a cell setup capable of continuous, vapour-fed CO₂RR, as well as investigation of parameter tuning for industrial implementation (**Figure 4**)

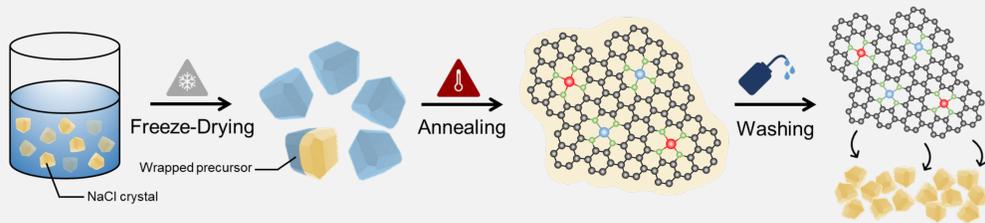


Figure 2: Synthesis approach for confinement of Ni and Fe single atoms in a N-doped graphene nanosheet structure. Freeze-drying avoids the aggregation of metal atoms into nanoparticles during the drying process, yielding a high single atom loading

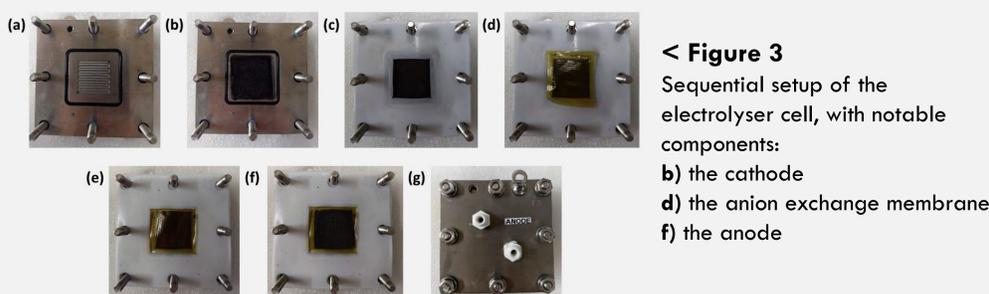
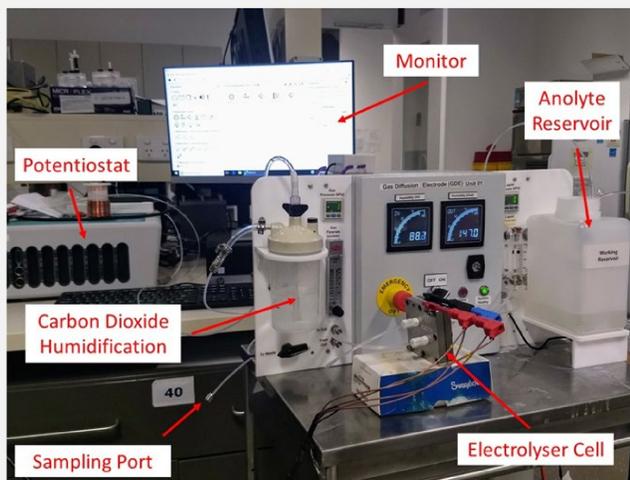


Figure 3
Sequential setup of the electrolyser cell, with notable components:
b) the cathode
d) the anion exchange membrane
f) the anode

Figure 4 >

- Syngas generation was undertaken in the electrolyzer system as shown. The key components of this system include:
- A 5cm² electrolyser cell
 - A potentiostat providing the applied voltage and recording data
 - Gas pressure and liquid flowrate controllers
 - Flowrate, pressure, humidity, and liquid sensors
 - Product sampling port



Aims and Objectives

- To develop highly-active catalysts for the one-step conversion of waste CO₂ and water to industrially-applicable syngas ratios
- To develop a high-throughput electrolyzer system for the continuous reduction of CO₂ and H₂O to synthesis gas
- To investigate the reduction of operational expenditure, through methods such as application of waste heat to increase production rate

Results

- Catalytic performance of Ni,Fe-hG single atom catalyst in the electrolyser system^[7]
- Modifying parameters such as cell temperature and electrode surface hydrophobicity were investigated to tune performance

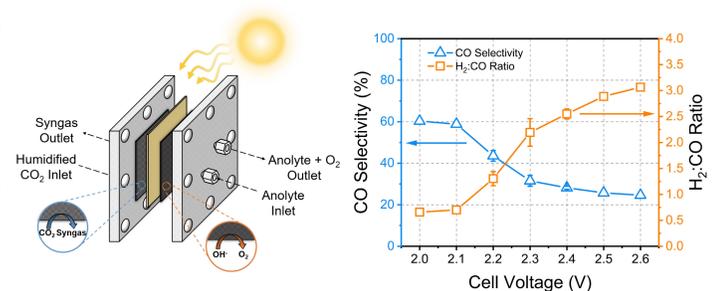


Figure 5: Membrane electrode assembly setup and dependence of CO selectivity and syngas ratio with applied cell voltage with Ni,Fe-hG catalyst. Syngas ratio increases as the cell voltage is increased.

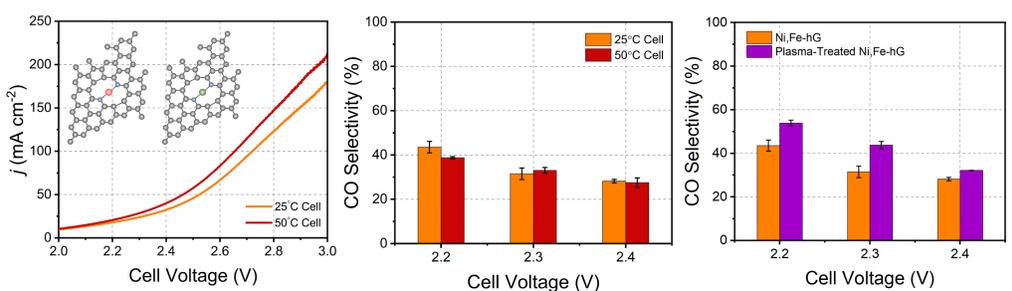


Figure 6: Activity and selectivity changes with membrane electrode assembly at 25°C and with cell heating (50°C). An increase in cell temperature leads to improved activity with minimal change in syngas ratio produced. Effect of plasma treatment on Ni,Fe-hG electrode shows that electrode surface hydrophobicity modification can be used to tune the product selectivity

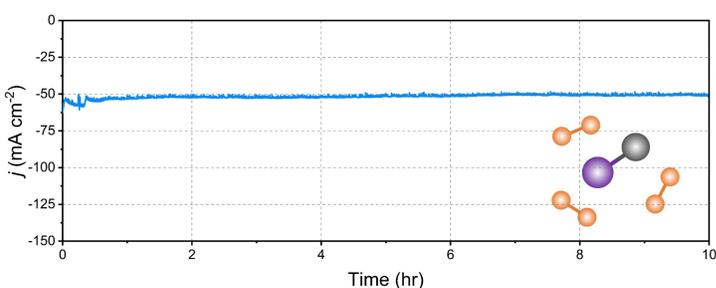


Figure 7
Stable performance of Ni,Fe-hG over a ten hour period in the membrane electrode assembly. A consistent syngas ratio of 3.0 was produced

Conclusions

- The membrane electrode assembly system is successful in allowing the continual conversion of CO₂ and H₂O into synthesis gas at high reaction rates
- The tuning of active sites in a range of metal, metal-carbon, and single atom catalysts can be achieved, in order to generate industrially applicable syngas ratios
- Further investigation of operating parameters and electrode engineering can push this solution to future large-scale application

References

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