Electrochemical CO_2 reduction at room temperature and mild pressures

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Abstract

Carbon capture and utilization technologies (CCU) and electrolytic hydrogen production are closely interconnected technologies and necessary for a sustainable energy system. This work describes the development of a process for room temperature co-electrolysis of CO_2 and water to produce syngas, at mild pressures. The influence of several parameters in the performance of the process is reported.

1. Introduction

Electrochemical reduction of carbon dioxide into chemicals powered by renewable energy is a promising technology for the production of green chemical building blocks. Following previous research by the authors described elsewhere [1], this work describes the further development of a process for room temperature co-electrolysis of CO_2 and water to produce syngas at mild pressures. This gaseous mixture (H_2 + CO) is an important intermediary for the production of gaseous, liquid fuels and several chemicals. However, this technology still presents significant challenges, namely the need to apply high overpotentials due to stability of the CO_2 molecule, poor stability of catalytic electrodes and mass transfer limitations. The performance of this process is influenced by many parameters such as type of catalytic electrodes, membrane separators and electrolyte composition. The operational conditions such as temperature, pressure, flow rate, electrochemical cell design also play an important role in determining process efficiency. The use of electrolytes based on ionic liquid 1-ethyl-3-methyl-imidazolium trifluoromethane (EMIMOTf) sulfonate is reported as a way to integrate CO_2 capture and conversion [2]. The influence of pressure, a poorly studied process parameter in syngas productivities is also investigated [3]. Crossover rates of the electrolysis gases obtained with different type of membranes will be analysed and the effect of flow rates on productivities is reported.

2. Experimental

An electrochemical cell was designed and built, with two compartments separated by a membrane, allowing the recirculation of electrolyte. The cell accommodates 6 x 6 cm^2 electrodes, a planar ionic exchange membrane between electrodes and it has a pressure rating of up to 100 bar. It works in the temperature range from room temperature up to 80ºC. Carbon dioxide from Air Liquide (N45 purity 99.995%) and distilled water were introduced in the cathode. Electrolysis was carried out using a commercial anode optimized for oxygen evolution in water electrolysis. The electrolysis gaseous products were analysed by gas chromatography using a 3000 MicroGC from Agilent, equipped with a thermal conductivity detector (TCD).

3. Results and discussion

The use of EMIMOTf electrolytes at pressures higher than atmospheric pressure (10 bar) increased syngas productivities, as CO_2 solubility is pressure dependent. As expected, overpotentials could be reduced through the use of these types of electrolytes [4]. Membrane thickness was the major factor in determining crossover of hydrogen and CO produced at the cathode into the anolyte compartment. In the conditions studied negligible crossover of CO were obtained with membranes thickness of 120 µm. Hydrogen crossover was higher and amounted to ca. 10% molar.

4. Conclusions

A significant research effort is still needed to increase the maturity level of electrochemical CO_2 reduction to fuels. Breakthroughs are needed in the development of membranes customized for CO_2 electroreduction. The tuneable properties of ionic liquids encloses the potential of further process optimization.

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