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Description of the Catalytic Microenvironment for the Electrochemical Reduction of Carbon Dioxide

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DESCRIPTION

Electrochemical carbon dioxide reduction with industrially relevant current densities, high product selectivity and long-term stability is a long-awaited goal. Unfortunately, copper catalysts for producing valuable multi-carbon products under ECR conditions undergo structural and morphological changes, especially at high current densities, leading to a rapid loss of product selectivity. Here we report a catalyst regeneration strategy using an electrolysis process involving alternating 'on' and 'off' operating regimes to increase the operational stability of copper catalysts. Lifetime is extended several times and ethylene selectivity greater than 40% is maintained when electrolysis is performed using a flow cell in a neutral ph medium at a current density of 150 ma cm^{-2}. We also demonstrated electrochemical carbon dioxide reduction to ethylene at a current density of 1 a cm^{-2} using a three-dimensional copper gas diffusion electrode with ethylene selectivity greater than 40%, demonstrating that this system can perform well under these conditions. This study shows that copper-based catalysts can be regenerated to achieve high multi-carbon selectivities once they enter a state generally considered to have reduced catalytic activity. A combination of short-term electrolysis, which minimizes morphological changes during the 'on' segment, and progressive chemical oxidation of the copper atoms on the catalyst surface during the 'off' segment, allows the collected salt and together with the reduction in catholyte temperature, it extends the operating life of the catalyst.

Electrochemical reduction of carbon dioxide by renewable power (such as solar and wind) is a promising potential for reusing carbon dioxide released in the production of cement, steel, aluminum, and in the production of ammonia and methanol. If carbon dioxide can be removed from the atmosphere at an acceptable cost, electrochemical reduction of carbon dioxide can be used to produce carbonaceous chemicals and fuels in a fully sustainable manner. Due to economic considerations, electrochemical reduction of carbon dioxide current densities should be in the range of 0.1 a/cm^2-1 a/cm^2, and high selectivity to desired products is required to minimize separation costs. Industrially relevant operating conditions are the use of gas diffusion electrode to maximize species transport to and from the cathode, and the combination of such electrodes with solid electrolyte membranes to reduce the ohmic losses associated with liquid electrolytes. Can be achieved by eliminating furthermore, high product selectivity can be achieved by carefully tuning the microenvironment near the catalyst surface We first studied the effect of catalyst morphology on the formation of multi-carbon products *via* cu-based catalysts, and then combined the kinetics of the buffering reaction with local concentration of carbon dioxide and ph to study mass transfer. The local carbon dioxide concentration and ph depend on the electrochemical reduction of carbon dioxide dynamics to form specific products on both copper and silver catalysts, the effect of electrolyte cation identity on the electrochemical reduction of carbon dioxide rates and product distributions, pulsed electrolysis to modulate the local ph and carbon dioxide concentration on the catalyst surface. The combining ionomer-coated catalysts with pulsed electrolysis can achieve very high selectivities for multi-carbon products over Copper in aqueous electrolytes.

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CONFLICT OF INTEREST

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