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Electrolysis to Modulate the Carbon Dioxide Concentration on the Catalyst Surface

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DESCRIPTION

It has been a long-term objective to achieve the Electrochemical Reduction of Carbon Dioxide (CO_2R) with industrially significant current densities, good product selectivity, and long-term stability. Unfortunately, especially at high current densities, copper catalysts for generating important multicarbon compounds experience structural and morphological changes that quickly reduce product selectivity. Here, we describe a method of catalyst regeneration that makes use of an electrolysis technique that alternates between "on" and "off" operating regimes in order to improve the operating stability of a copper catalyst. Using a three-dimensional copper gas diffusion electrode, we also demonstrate CO_2R to ethylene, discovering that this system is rendered stable under these conditions for more than 36 hours. This work demonstrates that copper-based catalysts can be recovered to provide strong multicarbon selectivity even after they have reached the state that is typically thought to have reduced catalytic activity. The catalyst can operate for a longer period of time by combining brief electrolysis periods, which minimise morphological changes during "on" segments, with the gradual chemical oxidation of copper atoms on the catalyst surface during "off" segments, along with the additional effects of washing the accumulated salt and lowering the catholyte temperature.

Electrochemical reduction of carbon dioxide through the electricity generated from renewable sources (If CO_2 can be removed from the atmosphere at an acceptable cost, then CO_2R can be used to produce chemicals and fuels) contain carbon in a completely sustainable manner. Economic considerations dictate that CO_2R flux densities and selectivity for the target product should be high to reduce. Minimize separation costs. Industrially relevant operating conditions can be achieved using gas diffusion electrodes to maximize the transport of species to and from the cathode and by combining these electrodes with a solid electrolyte film eliminates the resistance loss associated with liquid electrolytes. In addition, high product selectivity can be achieved by carefully adjusting the microenvironment tissues near the catalyst surface (e.g. Ph, CO_2 and H_2O concentrations, and cation identification in the bilayer adjacent to the catalyst surface). With a discussion of our experimental and theoretical work to optimize the catalytic microenvironment for CO_2R. We first studied the effect of catalyst morphology on the formation of Multi-Carbon (C^{2+}) products *via* copper based catalysts, and then combined the kinetics of the buffering reaction with local concentration of CO_2 and ph to study mass transfer. This is followed by a description of how the local CO_2 concentration and ph depend on the CO_2R dynamics to form specific products on both copper and silver catalysts. Next, we examine the effect of electrolyte cation identity on CO_2R rates and product distributions. Next, we consider using pulsed electrolysis to modulate the local ph and CO_2 concentration on the catalyst surface. The final part of the discussion shows that combining ionomer-coated catalysts with pulsed electrolysis can achieve very high selectivities for C^{2+} products over copper in aqueous electrolytes. We then extend this part of the report to consider the differences between the catalytic nanoparticle microenvironment present in the catalytic layer of the membrane electrode assembly and the planar electrode immersed in the aqueous electrolyte.

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CONFLICT OF INTEREST

Authors declare no conflict of interest.

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