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Electrochemical reduction of carbon dioxide to value-added chemicals using copper-based catalysts

Dan Ren
École polytechnique fédérale de Lausanne, Switzerland

New energy technologies have attracted unprecedented research attention since the start of this century, as they will assist in reducing our dependence on fossil fuels. Meanwhile, the increase of the atmospheric carbon dioxide concentration, which caused severe climate changes, forces us to seek for solutions to alleviate carbon emissions. One promising technology is to design systems that convert carbon dioxide to carbon-containing fuels or chemicals using renewable energy sources, such as solar or wind. Among these systems studied, electroreduction of carbon dioxide driven by photovoltaics has achieved the highest efficiency so far. While many metals could reduce carbon dioxide to formate or carbon monoxide, copper is the only metal that catalyzes the formation of hydrocarbons and alcohols such as ethylene and ethanol, with appreciable amounts. However, the selectivity is rather poor. Here, we investigate different copper structures for the selective reduction of carbon dioxide to ethylene, ethanol and n-propanol. With the aid of operando Raman spectroscopy and extensive surface characterizations, we are able to reveal the active sites for the formation of different products and offer new insights into the mechanism. While CO dimerization is the key step for the formation of ethylene, a CO-insertion mechanism is proposed to be one of the possible routes to the formation of ethanol and n-propanol. Apart from the effect of surface structures, the influence of electrochemical factors such as overpotential and the mass transport of carbon dioxide on the catalysts' selectivity are also illustrated. We find that the selectivity of copper catalysts are largely dependent on the applied potential and limiting current of the system, by analyzing 24 different copper catalysts reported by >10 research groups worldwide, including metallic, oxide-derived and chloride-derived copper particles. These studies help us design better catalysts for this catalytic reaction. A prototype device combining a solar panel with an optimized electrolyzer will be introduced as well. This device is demonstrated to produce ethylene under the intermittent natural sunlight.

dan.ren@epfl.ch