

Understanding the origin of products selectivity on copper catalysts during electrochemical CO₂ reduction

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Electrochemical CO₂ reduction has been considered as a potential route to convert harmful CO₂ to valuable hydrocarbons for sustainable carbon cycles. Since late 1980s, copper catalyst has been recognized as one and the only catalyst which produced hydrocarbons as primary products;¹ however, the poor selectivity obstructs the applications. According to DFT calculations, surface CO is the most common and important intermediate during CO₂ reduction reaction,² but further reaction mechanisms are still unclear, e.g. protonation and dimerization of CO. Cu(I) oxide was correlated to the formation of C₂ products during reactions,³ yet there is lack of solid intermediate evidence to distinguish different reaction routes.

In this talk, we used in situ surface enhanced infrared absorption spectroscopy, in situ X-ray absorption spectroscopy (Cu L-edge) and on-line GC to study the electrochemical CO₂ reduction mechanism occurred in different copper catalysts. We observed various CO intermediates such as CO_{atop} and CO_{bridge}, on copper surface during electrochemical CO₂ reduction and the formation of CO_{atop}/CO_{bridge} can be correlated to the existence of surface Cu(I) and Cu(0) sites, respectively. Our results suggested that the existence of CO_{atop} can result in the formation of methane during further reduction. In addition, ethylene can be produced in the presence of CO_{atop} and CO_{bridge}. Our results both prove theoretical predictions⁴ and meet the gap to illustrate the detail reaction mechanisms from CO₂ to hydrocarbons. We next propose a method to create Cu(I) sites efficiently on copper catalysts.

References

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