Tuning Distribution of Products Obtained by Pulsed Electrochemical CO₂ Reduction on Cu Catalyst

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Abstract:

Pulsed electrolysis can minimize hydrogen evolution and maximize C₂⁺ production even though it is consisting of potentials incapable of surface reconstruction of Cu catalyst. The temporal analysis using differential electrochemical mass spectroscopy (DEMS) reveals that product concentrations near the cathode stays in phase at the initial but out of phase with extended time, increasing the concentration of C₂H₄ at the expense of CO and H₂. We attribute these trends to an increased ratio of adsorbed CO to H on the Cu surface. Simulation of pulsed electrolysis also shows that the local concentration of CO₂ near the cathode builds up during anodic period that allows electrolysis with a higher CO₂ concentration during the cathodic period than could be achieved for static electrolysis.

Introduction

Pulsed electrolysis has been reported that it can enhance C₂⁺ production. But most of previous reports have focused on oxidation of Cu surface during anodic period although enhanced C₂⁺ production was also observed at anodic potentials incapable of surface oxidation. This study aims to developing a detailed understanding of how pulsed electrolysis impacts on evolution of products over Cu and in particular increases the formation of C₂⁺ products.

Results, Highlights, and Accomplishments

Variables for square-wave potential pulse

Potential for cathodic and anodic polarization: Hysteresis in DEMS analysis during CV. Local concentration of C₂H₄ & CO ↑ while H₂ ↓ during anodic sweep.
- Ea : on-set E for hysteresis of CO₂
- Ec : E with maximum (anodic+cathodic) of C₂H₄

Duration for cathodic and anodic polarization: DEMS analysis during stepped CA
- Around 25sec, system approaches to the steady state

Effect of anodic duration

Product distribution at bulk electrolyte (GC and HPLC) after pulsed electrolysis with different ta: For ta < 10 sec, HER ↑, C₂H₄ ↑, C₁ ↑ (optimum at 10 sec) in comparison with static electrolysis

Effect of cathodic duration

Pulsed electrolysis with different tc: For 10 sec of tc, HER ↓, C₂H₄ from 20 to 11%, C₂⁺ ↑ from 6% to 79% in comparison with static electrolysis

Outlook

Our study provides a detailed understanding of the CO₂RR under conditions of pulsed electrolysis using DEMS to reveal the temporal evolution of products at a Cu cathode.

The pulsed CO₂RR can minimize hydrogen evolution and maximize C₂⁺ production when duration at each potential was shorter than that requires to reach the steady state.

This study provides an insight for the unsteady state behavior during CO₂RR.

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