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Interfacial insights into the efficient electroconversion of CO₂ to CO using Cu-based catalysts supported on Printex L6 carbon

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Carbon dioxide (CO₂) is the primary byproduct of fossil fuel combustion and is widely recognized as the foremost anthropogenic greenhouse gas contributing to global warming. In response to the environmental crisis, the electrochemical reduction of CO₂ (CO₂RR) has garnered significant attention as a promising approach to reduce carbon emissions and transform this greenhouse gas into valuable fuels and chemicals [1-3]. In this study, we investigated CO₂RR using a series of nanostructured electrocatalysts based on carbon black Printex L6 (PCL6) adorned with copper clusters. Five different Cu/PCL6 composites were synthesized through a hydrothermal process by varying the mass ratio between the copper precursor and carbon to optimize catalytic performance. The catalysts were characterized by using electrochemical techniques such as electrochemical impedance spectroscopy (EIS), cyclic voltammetry, and electrochemically active surface area (ECSA) determination, along with morphological and structural techniques including X-ray diffraction (XRD), transmission electron microscopy (TEM), and Raman spectroscopy. Following characterization, the materials were assessed for CO₂RR activity in 0.1 mol·L⁻¹ KHCO₃ electrolyte under CO₂-saturated conditions (pH 6.87). Rotating disk electrode (RDE) analyses conducted at 1600 rpm in CO₂-saturated solution indicated a positive shift in onset potential to -0.50 V vs. RHE for the catalyst with a Cu:C mass ratio of 1.5:1, compared to -0.79 V vs. RHE for pure PCL6, suggesting improved CO₂ reduction activity. Further evaluations using a rotating ring-disk electrode (RRDE) facilitated real-time detection and quantification of both soluble and volatile products. Among the synthesized materials, the catalyst with a Cu:C mass ratio of 1:1 showed the highest selectivity for carbon monoxide (CO), with production observed starting at -0.4 V vs. RHE at the disk. This indicates a favorable surface electronic structure and effective dispersion of Cu active sites in this composition. At more negative disk potentials (-1.2 V vs. RHE), formic acid production was observed at the ring at 1.15 V vs. RHE. The increased CO selectivity at the 1:1 ratio emphasizes the importance of optimizing metal content to balance conductivity, surface area, and the accessibility of active sites. These findings show that hydrothermal synthesis is an effective method for tuning the catalytic interface and facilitating the selective conversion of CO₂ into value-added products using materials that are abundant in nature.

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