

Operando Methodology Probing the Behaviors of Metal Centers during the Chemical Reactions

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ABSTRACT

Electrochemical reduction of CO₂ is heavily pursued as a potential solution of CO₂ recycling and realizes the high-density renewable energy storage. Among numerous types of catalysts, copper-based catalysts have been shown to perform interesting nature toward hydrocarbon products. Nevertheless, achieving practical CO₂RR selectivity toward desired products on the state-of-the-art copper-based catalysts is still facing great challenges. The great challenge for promoting the CO₂RR selectivity may arise to a fact that this electrochemical process is a multiple proton-electron-transfer step and highly surface-sensitive, implying that the surface state of electrocatalyst may be dynamic and unpredictable under practical situations. By employing the comprehensive in-situ techniques we developed during past few years, we have demonstrated the first empirical demonstration to track the dynamic structural reconstruction/transformation in a model bimetallic system, which establishes a good understanding of the correlation between catalyst surface structure and catalytic selectivity. Furthermore, we also realized a very important achievement to develop an operando seconds-resolved X-ray absorption spectroscopy to uncover the chemical state evolution of working catalysts. It's well-known that a chemical reaction driven by applied potential is extremely fast, which is difficult to track the electrochemistry with acceptable temporal-resolution. This work is able to offer scientific insights into the roles metal states serve in chemical reaction in electrolyte.