In-situ Investigation of Chemical/Physical Dispersion of metal Complex on Copper Surface during Carbon Dioxide Reduction Reaction

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Abstract

Electrical conversion of CO_2 into chemical fuels provides a promising approach to store massive renewable energy. Through in-situ techniques including X-ray absorption spectroscopy and Raman, herein, we find that the dispersion means of porphyrin-based metallic complexes on the copper surface can determine the material structure and regulate the selectivity toward carbon dioxide reduction reaction (CO_2RR). The physical dispersion forms the molecule-metal catalyst interface, resulting in a reaction-intermediate-rich local environment and ethanol production. On the other hand, the chemical dispersion reconstructs the catalyst surface as the single-atom alloy during CO_2RR, which can migrate the reaction intermediates into the active single-atom iron, and thus facilitates the methane production. This finding unveils that the manners of surface modification manage the material systems and the CO_2RR selectivity. It also highlights the significance of in-situ investigation for structure identification during catalysis and catalytic mechanism.

Keywords – in-situ X-ray absorption spectroscopy, carbon dioxide reduction reaction, selectivity, molecule-metal catalyst interface, single-atom alloy

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