Operando X-ray Absorption Spectroscopic Studies during CO_2 Reduction Reaction in various reactors.

Sung-Fu Hung

Department of Applied Chemistry, National Yang Ming Chiao Tung University, Hsinchu 300, Taiwan

Abstract: Converting CO_2 and H_2O into value-added chemical feedstocks and fuels, powered using renewable electricity, offers a carbon-neutral approach to tackling global energy and climate concerns. Nowadays, various reactors have been employed for CO_2 Reduction Reaction (CO_2RR), such as the electrocatalytic H-cell reactor, the flow reactor, and the photocatalytic gaseous-catalytic reactor. Catalysts undergo different material evolutions and show diverse catalytic activities in each reactor. Toward this end, we have developed various operando techniques for these reactors. For the H-cell reactor, we successfully unveiled the atomically dispersed nickel on nitrogenated graphene as the active site for CO_2RR. For the flow reactor, we monitored the carbon nanoparticle hybrid with the copper complex catalyst for CO_2RR.[1] It showed that increased carbon nanoparticle loadings could reduce the metallic copper coordination number unveiled by operando XAS, and the CO_2-to-methane selectivity of 62% can be achieved at a copper coordination number of 4.2.[2] For the photocatalytic gaseous-catalytic reactor, we probed the dynamic evolution of the atomically dispersed Cu species for CO_2 photoreduction to solar fuels. The results unveil that the initial atomically dispersed Cu(II) undergoes reduction to Cu(I) and ultimately to Cu(0); the Cu(I)/Cu(0) mixture is proposed to be more effective for CH_4 formation.[3] We hope that developing these operando X-ray absorption spectroscopic techniques can promote the applications of X-ray absorption spectroscopy.

References:

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