The 123rd GREEN Seminar

Electrochemical conversion of CO2 using single atom decorated catalysts

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The pursuit of a circular carbon economy and the need to surmount the constraints of CO2 electroreduction technology has prompted the development of single-atom catalysts (SACs) for electrocatalysts. SACs consist of isolated metal atoms dispersed on a support material. First, zeolitic imidazolate framework (ZIF)-8 containing various transition metal ions—Ni, Fe, and Cu—at varying concentrations upon doping were fabricated for the electrocatalytic CO2 reduction reaction (CO2RR) to CO without further processing. The electron-rich sp2 C atoms of optimized copper doping on ZIF-8, leading to a local effect between the Zn–N4 and Cu–N4 moieties, achieve a maximum Faradaic efficiency for CO2 to CO of 88.5% at -1.0 V (vs. RHE) with a stability over 6 h. Second, compared to nanoparticles, SACs have been shown to significantly enhance the efficiency of metal atom utilization by nearly 100%, which results in a higher concentration of active sites, leading to outstanding catalytic activity, excellent product selectivity, and stability. Among the diverse support materials, the ZIFs, a subclass of metal-organic frameworks (MOFs) with high nitrogen content, have been widely used to prepare metal-nitrogen-doped carbon (M-NC) SACs with dense active sites. Herein, we utilized an eco-friendly approach to produce two-dimensional ZIF-8 nanosheets (ZIF-8-NS) as an optimal support material for SACs. Additionally, we introduced Ni precursor into the synthesis process of Ni-ZIF-8-NS, which was then subjected to pyrolysis at 950 ° C under a N2 atmosphere to yield the final product, Ni-NC-NS. Ni-NC-NS demonstrated an outstanding CO2RR performance by exhibiting excellent Faradaic efficiency (FE) toward CO of ~100% both in the H-cell and flow-cell reactors as well as a remarkable turnover frequency (TOF) of 23,699 h−1. Finally, we will present a facile strategy for synthesizing M–NC SACs using metal-chelating ligands, eliminating the need for additional processing steps. Specifically, the method to use ethylenediaminetetraacetic acid as a strong metal-chelating ligand will be shown.

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