Developing new catalysts for carbon dioxide electroreduction

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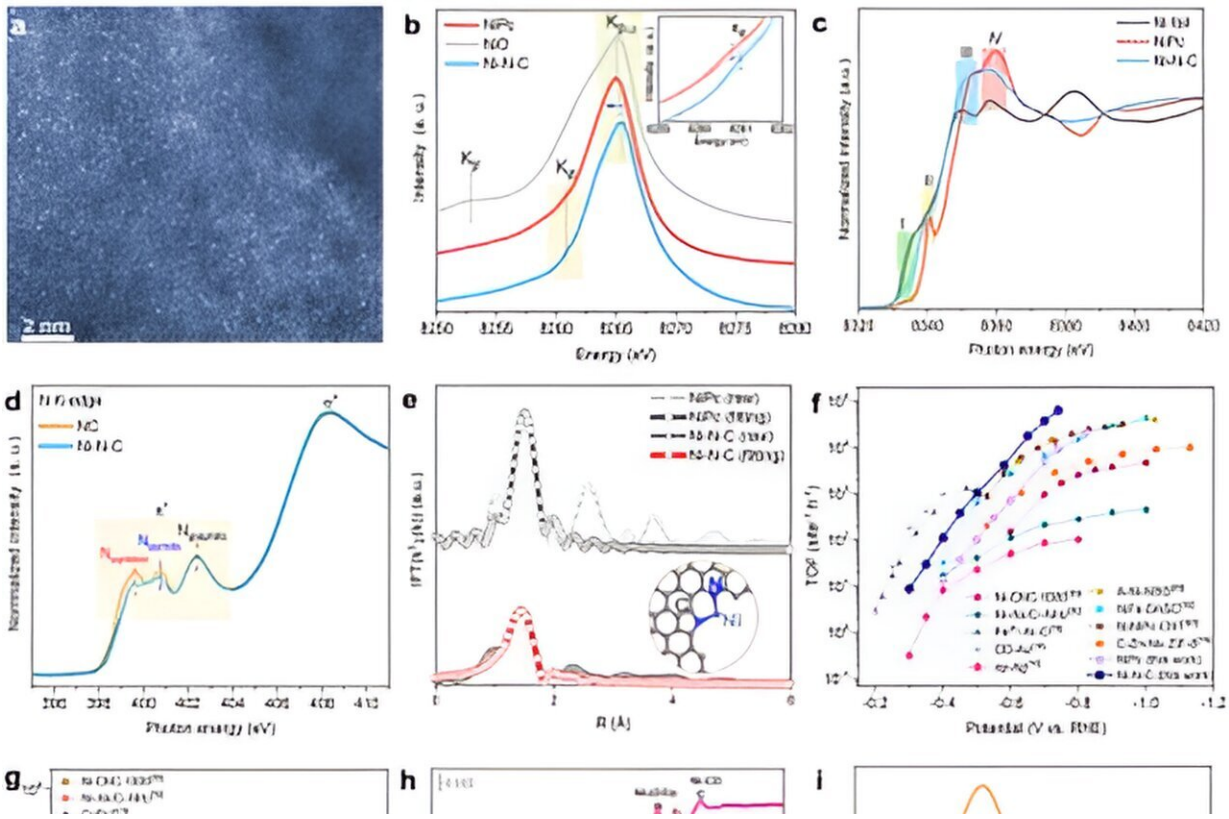

Analysis of fine structure, properties, and catalytic reaction mechanism of catalysts. Credit: Prof. Song's team

As a crucial part of Carbon Capture, Utilization, and Storage (CCUS) technology, CO_2 reduction reaction (CO_2RR) to carbon-based fuels and

chemicals presents broad application prospects in renewable energy storage and CO_2 negative emission.

Recently, a team led by Prof. Song Li and Associate Researcher He Qun from the National Synchrotron Radiation Laboratory of the University of Science and Technology of China (USTC) of the Chinese Academy of Sciences (CAS) put forth a novel understanding of the mechanism of CO_2RR on the nickel (Ni) single-atomic sites. Their study, titled "Asymmetric Dinitrogen-Coordinated Nickel Single-Atomic Sites for Efficient CO_2 Electroreduction," was published in *Nature Communications*.

An ideal CO_2RR catalyst requires low overpotential and high current density to products. However, former catalysts either are featured with high cost and low current density, such as gold (Au) and silver (Ag), usually exhibit much higher overpotentials than Au and Ag, such as Fe, Co, or Ni, limiting reaction efficiency.

Therefore, it is imperative to develop overpotential low, high current density, abundant 3d metal-based catalysts to replace precious metal catalysts for CO_2RR. To address those challenges, the researchers proposed an asymmetric dinitrogen-coordinated nickel single-atom catalyst (Ni-N-C). By utilizing the unsaturated and asymmetric characteristics of the sites, structural self-optimization during the electrochemical process is achieved, thereby enhancing the intrinsic activity of the sites in CO_2RR.

In the study, the team designed and synthesized Ni-N-C featuring dinitrogen coordination (pyridinic and pyrrolic nitrogen) and then utilized it for CO_2 electroreduction reactions in neutral and alkaline media. Synchrotron radiation X-ray absorption spectra and emission spectra revealed the local coordination structure of Ni sites in the catalyst. The electrochemical test results showed that the Ni-N-C catalyst

could achieve very high electrochemical performance in both neutral (H-type cell) and alkaline (gas diffusion electrode, GDE) electrolytes.

Especially in alkaline conditions, the catalyst could achieve a CO partial current density of 20.1 mA cm_{geo}^{-2} at -0.15 V vs. reversible hydrogen electrode (V_{RHE}), Faraday efficiency of over 90% for CO in the potential range of -0.15 to -0.9 V_{RHE}, and high turnover frequency (TOF) of over 274,000 $site^{-1}$ h^{-1} at -1.0 V_{RHE}, surpassing most reported catalysts.

This study offers a novel comprehension of the catalyst's role in the CO_2 electroreduction reaction and promises to shed new light on future CO_2 reduction technologies.

More information: Yuzhu Zhou et al, Asymmetric dinitrogen-coordinated nickel single-atomic sites for efficient CO_2 electroreduction, *Nature Communications* (2023). DOI: 10.1038/s41467-023-39505-2

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