New approach developed for electrocatalytic H_2O_2 production and biomass upgrading

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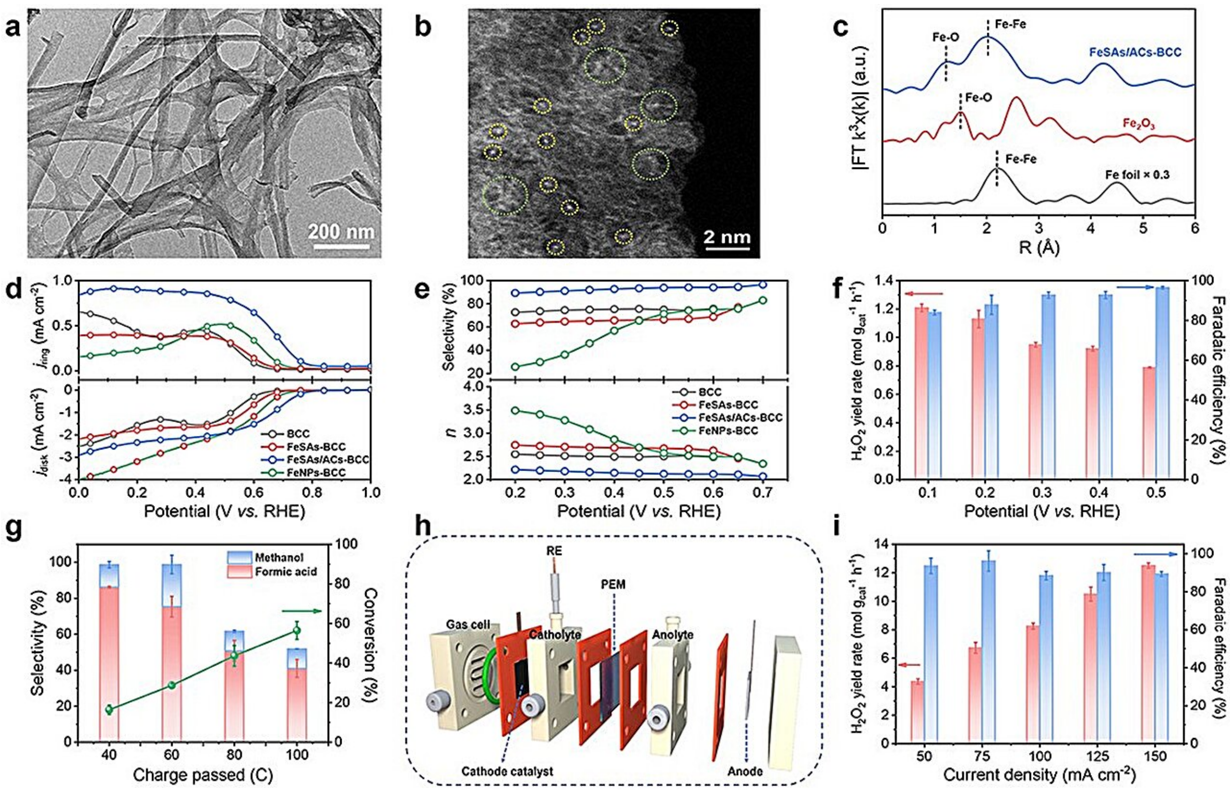


Characterizations, electrochemical H_2O_2 synthesis performance and coupled electro-Fenton process of FeSAs/ACs-BCC. Credit: Xu Hui

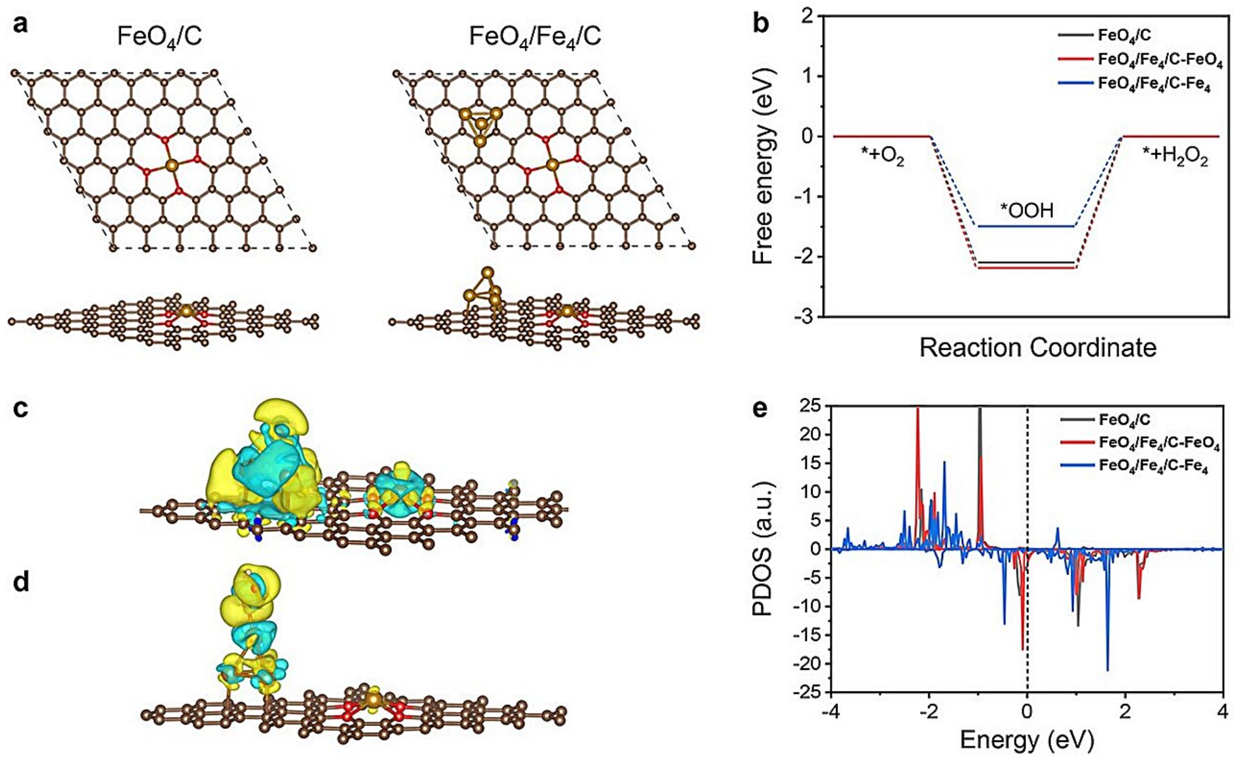
Scientists from the Hefei Institutes of Physical Science of the Chinese Academy of Sciences have synthesized an oxygen-coordinated Fe single atom and atom cluster catalyst that exhibits superior electrocatalytic performance for hydrogen peroxide (H_2O_2) production and biomass upgrading. The research is published in *Angewandte Chemie International Edition*.

H_2O_2 is a widely used chemical with applications in diverse fields such as environment, energy, and health care. While traditionally produced through energy-intensive processes, electrocatalytic synthesis offers a more environmentally friendly and efficient method using water and oxygen.

However, this approach requires advanced electrocatalysts for high-yield and selective H_2O_2 production, and further attention is needed to utilize the generated H_2O_2, particularly in electrochemical organic oxidation processes. This offers significant potential for value-added applications beyond environmental remediation.

For this study, the researchers used bacterial cellulose as an adsorption regulator and carbon source in combination with a multi-step approach involving wet chemical impregnation, pyrolysis, and acid etching processes to create a catalyst termed FeSAs/ACs-bacterial cellulose-derived carbon (BCC), consisting of oxygen-coordinated Fe single atoms (SAs) and atom clusters (ACs).

The presence of both Fe SAs and clusters was confirmed using advanced imaging techniques such as aberration-corrected scanning transmission electron microscopy. The atomic structure of Fe was also determined by X-ray fine structure absorption spectroscopy and X-ray photoelectron spectroscopy.



Theoretical calculations and mechanism elucidation toward $2e^-$ ORR. Credit: Xu Hui

This catalyst showed excellent electrocatalytic performance and selectivity for the 2-electron oxygen reduction reaction ($2e^-$ ORR) under alkaline conditions. Further H-cell experiments confirmed the accumulation of H_2O_2 in the electrolyte.

The researchers coupled the in situ generated H_2O_2 with the electro-Fenton process using ethylene glycol as the reactant and acidified 0.1 M Na_2SO_4 as the electrolyte. This resulted in a high rate of ethylene glycol conversion and high selectivity for formic acid, demonstrating that the electro-Fenton process has the potential to improve biomass-derived feedstocks through oxidative upgrading.

They also developed a three-phase flow cell based on the gas diffusion electrode to further improve the H_2O_2 yield.

Density functional theory analyses indicated that the actual catalytically active sites in the $2e^-$ ORR process were the Fe clusters, and the electronic interaction between Fe single atoms and Fe clusters could significantly enhance the electrocatalytic performance toward $2e^-$ ORR.

This work will be helpful for the design and development of atomic-level electrocatalysts for high-efficiency $2e^-$ ORR to H_2O_2 and biomass upgrading.

More information: Hui Xu et al, Atomically Dispersed Iron Regulating Electronic Structure of Iron Atom Clusters for Electrocatalytic H2O2 Production and Biomass Upgrading, *Angewandte Chemie International Edition* (2023). DOI: 10.1002/anie.202314414

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