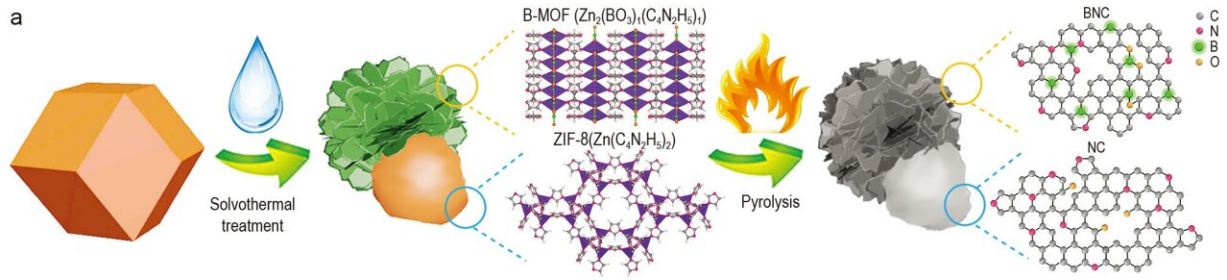
A Janus carbon electrocatalyst can balance intrinsic activity and electronic conductivity

December 7 2022



A Janus MOF heterostructure composed of ZIF-8 crystals and boron-containing MOF nanosheets (B-MOF) was constructed through a "molecular clipping and re-suturing" process. The pyrolysis of ZIF-8/B-MOF yielded Janus carbon structures consisting of nitrogen-doped carbon block and boron, nitrogen co-doped carbon nanosheets. Credit: Science China Press

Carbon-based electrocatalysts are considered as promising alternatives to the state-of-the-art precious metal catalysts. Heteroatom doping can effectively create highly active catalytic centers, but unfortunately, result in lower electronic conductivity and thus hinder the electrocatalysis process.

To address this issue, a team from South China University of Technology developed a Janus carbon electrocatalyst with different heteroatom doping levels between the two sides, which could resolve the conflict between intrinsic activity and electronic conductivity to boost

the performance in the electrocatalytic hydrazine oxidation reactions.

Electrocatalysis enables the transformation of electrical energy to chemical energy. The smooth proceeding of electrocatalytic reactions relies on the design of electrocatalysts with highly active centers and efficient electron conduction. Carbon materials represent an important class of electrocatalysts. The major barrier to performance improvement of carbon materials is the trade-off between intrinsic activity and electronic conductivity.

Now, a team led by Prof. Yingwei Li at South China University of Technology addressed this issue by developing a carbon-based catalyst with a Janus structure. The Janus carbon electrocatalyst consists of a conductive nitrogen-doped carbon block (NC) and catalytically active boron-, nitrogen co-doped carbon nanosheets (BNC).

"The design of Janus carbon nanomaterials is not an easy task. Carbon materials are usually prepared by the carbonization of carbon-containing precursors. However, conventional precursors lack the designability to synthesize carbon materials with tunable structures and compositions. Our group has been engaged in the development of efficient catalysts based on metal-organic frameworks (MOFs), a class of materials with high designability, tunable compositions, and ordered atomic distributions. The interesting properties of MOFs motivated us to design a Janus MOF as the precursor for Janus carbon nanomaterials," explained Yingwei Li.

The researchers developed a "molecular clipping and re-suturing" strategy for the construction of the Janus MOF. ZIF-8 crystals were heated in a methanol solution of boric acid. ZIF-8 was slowly etched by boric acid to release metal ions and ligands, followed by nucleation and growth of B-MOF on etched ZIF-8. ZIF-8/B-MOF was then employed as precursors for the synthesis of Janus NC/BNC.

The NC side displayed a lower doping level and thus a higher electronic conductivity compared with the BNC side. However, the BNC side possessed catalytically active BO_3 sites with higher intrinsic activity. The integration of NC with BNC could not only ensure high electronic conductivity of the hybrid, but also induce further charge delocalization of active sites on the BNC side with enhanced catalytic activity.

In the electrocatalytic hydrazine oxidation reaction, NC/BNC exhibited significantly improved activity than the single counterparts and simple physical mixtures.

In view of the big family of MOFs, the team believes that the proposed MOF-templated strategy can be extended to the synthesis of various Janus carbon materials with tunable compositions and structures. This will hopefully enrich the toolbox of tailorable chemistry and nanotechnology for potential applications in interfacial stabilizers, drug delivery, and phase-transfer catalysis.

The research is published in the journal *National Science Review*.

More information: Jieting Ding et al, A Janus heteroatom-doped carbon electrocatalyst for hydrazine oxidation, *National Science Review* (2022). DOI: 10.1093/nsr/nwac231

Provided by Science China Press