A controllable atom-trapping strategy is developed to extract isolated Mo atom from the a-MoO$_x$@CoSe$_2$ pre-catalyst into CoOOH through an ultra-fast self-reconstruction process during the OER process. The single Mo-atom decoration shifts the oxidation cycle of Co sites towards a more energetically favored rate-limiting process, resulting in outstanding OER activity. Credit: Angewandte Chemie International Edition (2023). DOI: 10.1002/anie.202309341

Single-atom catalysts (SACs), representing the ultimate high atom
utilization efficiency and specific activity, have arguably become the
most active new frontier in heterogeneous catalysis. The technology of
atom trapping based on support effects is a viable method for
synthesizing SACs.

Because trapping isolated metal atoms is highly desired by researchers,
the structural dynamics of materials have been used during catalytic
conditions, especially at the dynamical generation stage of active moiety,
where the reconstructed atomic-precision catalyst surface usually
possesses thermodynamically stable stoichiometry.

In a study published in Angewandte Chemie International Edition, the
research group led by Prof. Zhang Jian from Fujian Institute of Research
on the Structure of Matter of the Chinese Academy of Sciences, in
collaboration with Prof. Zhang Huabin from King Abdullah University
of Science and Technology, Saudi Arabia, reversely trapped isolated
atoms in high oxidation to accelerate the oxygen evolution reaction
(OER) kinetics.

The researchers first studied the atomization effects under in-situ OER
conditions and elucidated how the incorporation of high-value metal
modulators promotes the OER activity of CoOOH. The developed Mo-
CoOOH demonstrated enhanced OER activity, kinetics and stability.

The team then revealed the origin and kinetics of the ultra-fast structural
reconstruction during the extraction of isolated Mo atoms from
a-MoO₅@CoSe₂ pre-catalysts by using multi-model operando
characterizations. The ultra-fast reconstruction process enabled the Mo
atoms to trap in the CoOOH lattice, forming higher covalency Co-O-Mo
bonds.

Electrochemical analysis and theoretical calculations verified that the
single Mo-atom decoration shifts the oxidation cycle of Co sites toward
a more energetically favored rate-limiting process.

In addition, the researchers found that the Mo-CoOOH only needs an extremely low overpotential of 297 mV to achieve a current density of 100 mA cm$^{-2}$ in alkaline media, exhibiting top-level catalytic activity among all reported electrocatalysts.

This study provides a fundamental understanding of the dynamic reaction pathways of the pre-catalyst and offers more possibilities of scalable single-atom extraction strategies for designing highly active electrocatalysts.


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