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REVIEW HIGHLIGHT

Metal-support frontier orbital interactions in single-atom catalysis

Junling Lu

State Key Laboratory of Precision and Intelligent Chemistry, School of Chemistry and Materials Science, Collaborative Innovation Center of Chemistry for Energy Materials (iChEM), University of Science and Technology of China, Hefei, China

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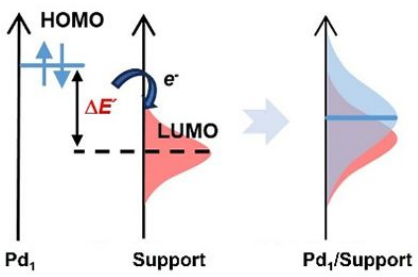
Author introduction

Professor Junling Lu is a Chair Professor at the University of Science and Technology of China (USTC). He received his Ph.D. in Physics from the Institute of Physics, Chinese Academy of Sciences in 2007. During his doctoral studies, he spent two years (2004–2006) as an exchange student at the Fritz-Haber Institute of the Max Planck Society in Germany. From 2007 to 2013, he conducted postdoctoral research at Northwestern University and Argonne National Laboratory in the United States. In March 2013, he joined the School of Chemistry and Materials Science at USTC as a full professor.

His long-term research focuses on the precise design of catalysts using atomic layer deposition (ALD) technology. He has achieved atomically precise controllable synthesis ranging from single atoms, diatomic, and triatomic structures to bimetallic nanoparticles. He has also pioneered the experimental validation and groundbreaking application of frontier molecular orbital theory in heterogeneous catalysis. He has published over 140 SCI-indexed papers in top journals including Nature (2), Science (1), Nature Nanotechnology (2), and Nature Catalysis (1), with his work cited over 20,000 times. His research achievements were selected among the Top Ten Scientific and Technological Advances in Chinese Universities in 2019 by the Ministry of Education. He has received several honors, including the China Catalysis Young Scientist Award in 2021 and the Chinese Chemical Society–Royal Society of Chemistry Young Chemist Award in 2019.

Highlights

- Corroborate of frontier molecular orbital (FMO) theory in single-atom catalysis.
- The stability and activity of SACs is fundamentally governed by metal–support and metal-adsorbate orbital interactions, respectively.
- Establish the support LUMO level as a robust, predictive descriptor for rationally designing metal–support pairs with tunable hydrogenation performance.



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Metal-support frontier orbital interactions in single-atom catalysis

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Abstract Single-atom catalysts (SACs), with their structurally simplified active sites, provide a unique platform for probing catalytic mechanisms at the atomic level. The performance of SACs—both activity and stability—is fundamentally governed by synergistic frontier orbital interactions at the metal–adsorbate and metal–support interfaces. Yet, a quantitative atomic-scale understanding of how these interactions determine catalytic performance remains elusive. Here, we show that the lowest unoccupied molecular orbital (LUMO) positions of pristine semiconducting supports, measured via Mott–Schottky analysis across 14 distinct materials, scale approximately linearly with the hydrogenation activity of supported palladium (Pd_1) atoms. Strikingly, downsizing the support to a few nanometers elevates its LUMO position, leading to record-high activity and outstanding stability in acetylene semihydrogenation. Theoretical calculations reveal that this LUMO elevation reduces the energy gap with the highest occupied molecular orbital (HOMO) of the Pd_1 atoms, which reinforces Pd_1–support orbital hybridization for enhanced stability; in turn, it sequentially tunes the LUMO of the anchored Pd_1 to amplify Pd_1–adsorbate interactions for improved activity. These findings are consistent with frontier molecular orbital (FMO) theory and establish the LUMO position of semiconducting supports as a general descriptor for the rational design of metal–support pairs with predictable hydrogenation performance.

Keywords single-atom catalysts, frontier molecular orbital theory, metal-support interactions, hydrogenation

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E-mail: junling@ustc.edu.cn

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