Proceedings of the 9th International Conference of Recent Trends in Environmental Science and Engineering (RTESE 2025)
July, 2025 | Imperial College London Conference Center, London, United Kingdom
Paper No. 105
DOI: 10.11159/rtese25.105

Modulating the Coordination Environment of Single-Atom Catalysts for Energy and Environmental Applications

Xiaoqiang An[1]
[1]Tsinghua University
Center for Water and Ecology, School of Environment, Tsinghua University, Beijing,
100084, China.
xqan@tsinghua.edu.cn

Extended Abstract

Single atom catalysts (SACs) that integrate the merits of homogeneous and heterogeneous catalysts have been attracting considerable attention in recent years. A key challenge in this field is precisely manipulating the geometric and electronic structure of single metal sites. This study focuses on the catalytic structure-performance relationship of SACs in terms of coordination environment and metal-support interactions by advanced characterization and theoretical studies. Several emerging types of catalysts with synergistic multisites exhibiting intersite metal-metal interactions has been developed by the atom-level engineering of active sites.[1] By taking advantage of the mobile feature of single atoms, a universal photo-involved assembly strategy was explored to mediate the distribution of active sites for exceptional reactivity, selectivity, and stability. The significant role of sodium ions and oxygen vacancies in the migration of single atoms and charge transfer was fundamentally investigated, reveling a synergistic mechanism between multifarious active sites for the cascade catalysis of hydrogen evolution and the interfacial activation of reactive oxygen species.[2] The strategy was thereafter employed to construct a biotic-abiotic nanoarchitecture composed of Fe single-atom nanozymes combined with native nitrate reductases for the efficient and selective reduction of nitrate to nitrite via photobiocatalysis. The mechanistic studies reveal that Fe single atoms and cyano (-C≡N) groups on graphitic carbon nitride mimic the heme b_D and heme b_P in the NarI subunit and construct a unique electron transfer chain between the biotic-abiotic interface under visible light irradiation, realizing a high nitrate conversion rate of 1.64×10^{-2} min^{-1} with nearly 100% selectivity, which is a significant advance in both photocatalytic reduction and bioconversion of nitrate. This work represents a promising platform to address the bottleneck problems of SACs for new energy production and environmental remediation.

References

(1) J. Bian, Y. Liao, R. Liu, X. An, C. Hu, H. Liu, J. Qu, "Synergy of cyano groups and cobalt single atoms in graphitic carbon nitride for enhanced bio-denitrification," Water Res., vol. 218, pp. 118465, 2022.

(2) X. An, T. Wei, P. Ding, L. Liu, L. Xiong, J. Tang, J. Ma, F. Wang, H. Liu, J. Qu, "Sodium-directed photon-induced assembly strategy for preparing multi-site catalysts with high atomic utilization efficiency," J. Am. Chem. Soc., vol. 145, pp. 1759, 2023.

(3) J. Bian, X. An, J. Zhao, Y. Liao, X. Lan, R. Liu, C. Hu, J. Chen, H. Liu, J. Qu, "Directional Electron Transfer in Enzymatic Nano-Bio Hybrids for Selective Photobiocatalytic Conversion of Nitrate." Angew. Chem. Int. Ed. DOI: https://doi.org/10.1002/anie.202412194.