Targeted Design and Preparation of Atomic-Level Catalysts

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Abstract:

As metals are reduced to the atomic level, they assume the form of single atoms or double atoms dispersed on the carrier surface. This leads to a substantial enhancement in the stability of metal sites due to their strong interaction with the substrate, resulting in a near 100% utilization efficiency of metal atoms, showcasing the catalytic charm induced by the size effect. However, the field of Single-Atom Catalysts (SACs) faces two critical challenges: the rational design of single-atom localized structures at the atomic level and the realization of high single-atom loading and large-scale synthesis of SACs.

To address these challenges, we are developing an atomic-level catalyst database specifically for the selective catalytic reduction of NO_x using NH_3. Our approach involves employing machine learning techniques to predict and screen potential highly active atomic-level catalyst configurations, followed by catalyst preparation, in-situ characterization, and first-principles calculations to explore structure-activity relationships and catalytic mechanisms. Initially, we conducted DFT calculations, with a particular focus on Cu_1/CeO_2 SACs, to screen the type and configuration of single atoms. Theoretical calculations revealed that the construction of Cu single atoms promoted the adsorption of reactant molecules and kinetically facilitated the SCR process.

Moreover, we are dedicated to developing and optimizing low-cost large-scale preparation processes. To this end, we propose an electron-driven strategy for synthesizing SACs with high loadings and yields, successfully applying it to Cu_1/CeO_2 SACs. This single-atom synthesis method is easily scalable to industrial levels, allowing us to achieve kg-level synthesis with high single-atom loading.

Our ultimate goal is to achieve the targeted design of atomic-level catalysts through the integration of machine learning and theoretical calculations, surpassing the traditional trial-and-error development approach. We want to provide a reference paradigm for the design and development of catalytic materials, opening up new possibilities for efficient and sustainable catalysts.

Keywords: Atomic-level catalysts, SACs, NH_3-SCR, DFT calculations, Large-scale synthesis

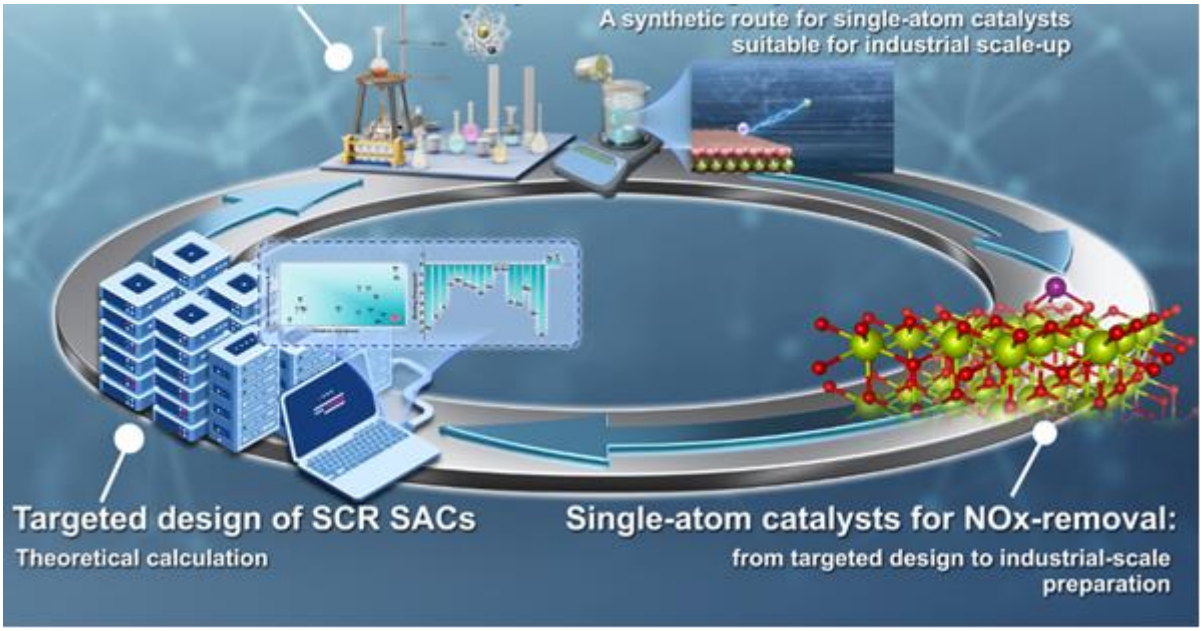

Figure 1 Targeted Design and Large-Scale Preparation of Atomic-Level Catalysts