Static or Dynamic? A First-Principles Multiscale Modeling Perspective on the Nature of the Surface of Operating Catalysts

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First-principles electronic structure calculations have emerged as a key contributor in modern heterogeneous catalysis research. Next to the dedicated computation of thermostability, spectroscopic signals and reactivity descriptors, they are increasingly used as basis for predictive-quality multiscale modeling approaches [1]. Corresponding approaches additionally account for meso- and macroscopic aspects like statistical interplay within the catalytic cycle or heat and mass transport in the reactor. This provides unprecedented insight into the catalytic function, be that the actual reaction mechanisms or the nature of the surface of the operating catalyst. Particularly with respect to the latter, this insight points at substantial deviations from the standard picture that analyzes catalytic function merely in terms of properties of and processes at active sites as they emerge from a crystal lattice truncation of the nominal catalyst bulk material [2]. Instead, a highly dynamic picture is suggested with continuous reaction-induced complex (surface) morphological changes at (near-) ambient conditions or an evolving, possibly liquid-like phase behavior due to limited heat dissipation channels. Advancing multi-scale catalysis modeling methodology to scrutinize such a picture is the next grand challenge. In my talk I will sketch some first steps in this direction, in particular to describe the atomistic mechanics of morphological transitions between commensurate lattices [3,4] and a non-adiabatic dissipation of released reaction heat [5].

References

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