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TOWARDS ATOMIC PRECISION IN HETEROGENEOUS CATALYST DESIGN

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ETH Hönggerberg, HCI D2 and on Zoom
(https://ethz.zoom.us/j/61720220862)

Project Summary: Single-atom heterogeneous catalysts (SACs) represent a paradigm shift in metal utilization with opportunities for unique reactivity. Still, after a decade of research, our ability to precisely engineer active sites and understand their dynamics during reactions remains limited. This presentation showcases concepts for establishing robust synthesis-structure-performance relations in carbon-based SACs. The work focuses on halogen chemistry applications, owing to their sensitivity to catalyst nanostructure, with industrial relevance. Several aspects of SAC design are tackled by combining synthetic methodologies, kinetic evaluation, advanced characterization, and theoretical modeling. First, strategies to control the metal nanostructure are presented, encompassing i) nanoparticle redispersion in reactive environments as a scalable synthesis approach for in situ formation of SACs with high metal content, and ii) carbon engineering guidelines to optimize metal coordination and oxidation states for enhanced performance, as shown for Pd SACs in dibromomethane hydrodebromination. Next, the importance of monitoring working active sites is demonstrated for Pt SACs in acetylene hydrochlorination. This establishes the distinct catalytic functions of metal atoms and carbons, and uncovers the restructuring of metal-ligand architectures – offering new perspectives on metal precursor selection for catalyst synthesis sustainability and performance. Finally, aiming to unlock new reactivity patterns, the thesis introduces principles of carbon functionalization to control metal-metal interactions in bimetallic SACs, demonstrating distinct intermetallic synergies in catalyzing a probe reaction. These findings establish atomically-precise design criteria for SACs and tools to investigate active site dynamics, with applicability across various catalytic processes.

CV: Vera Giulimondi obtained her MSc. in Chemical and Biotechnology from EPFL in 2020 and subsequently started her doctoral research at aCe-Catalysis Engineering, led by Prof. Javier Pérez-Ramírez, and within the SNSF-funded NCCR Catalysis.