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NANOSTRUCTURED CATALYSTS FOR SUSTAINABLE ACETYLENE-BASED VINYL CHLORIDE PRODUCTION

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Project Summary: The replacement of toxic mercuric chloride in catalyzed acetylene hydrochlorination is imperative to reduce the environmental impact of this long-established industrial technology in the production of polyvinyl chloride. To date, carbon-supported precious metals, such as Au single-atom catalysts (SACs), and metal-free carbons are potential alternatives, but suffer from fast deactivation. Herein, synthetic strategies to yield supported metal-based and metal-free catalysts with fine control over the elemental speciation and environment of the active sites are developed, which, when complemented with kinetic evaluation, advanced characterization, and theoretical modelling, are a powerful tool to establish structure-performance relationships. Following this methodology, innovative catalyst architectures were created, including porous nitrogen-doped carbons that rival the activity of benchmark Au-based systems, Ru nanoparticles encapsulated into single-layer graphene with increased stability against redispersion into single atoms under reaction conditions, Au and Pt SACs with tunable coordination environment, leading to unparalleled stability in the latter case, as well as bimetallic SACs. Finally, we derive a global performance descriptor, based on competitive acetylene/HCl interaction, to rationalize the activity and stability hierarchies of the different metal nanostructures and their dynamic behavior as well as the role of the support in acetylene hydrochlorination.

CV: Selina Kaiser obtained her M.Sc. in Chemistry at the Free University of Berlin in 2016 and subsequently pursued her doctoral studies in the group of Prof. Pérez-Ramírez.