Death and Life of Homogeneous Carbonyl Reduction Catalysts: Navigating Condition Space Towards Superior Catalytic Performance

Abstract: Catalytic hydrogenation chemistry offers a greener and more sustainable alternative to prevalent stoichiometric reduction methodologies in the production of high-value chemicals. In recent years, traditional Ru and Ir catalysts have been complemented by novel first-row transition metal catalysts due to their favorable sustainability, economics, and reduced toxicity. The practical implementation of such catalysts based on e.g. Fe or Mn is often hampered by their limited stability under the catalytic conditions. Understanding the catalytic and deactivation paths for such systems is key to tailoring their properties towards enhanced and sustained catalytic performance. In this lecture, I will discuss the importance and challenges of understanding the chemistry of catalyst deactivation for the development of practical catalytic technologies with the examples of our recent studies on Mn catalysis for selective reduction of carbonyl-containing compounds. While the performance of homogeneous catalysts is typically interpreted based on the molecular structures and electronic properties of the originating organometallic compounds, these catalyst systems are complex and multifunctional. Their behavior is influenced by a wide range of parameters, such as activation procedure, presence of promotors, solvent type, and selected conditions. Insights into the underlying mechanisms and their condition-dependencies can be obtained through a combination of operando spectroscopy, kinetic studies, and automated computational mechanistic analysis. This approach can help navigate this vast condition space and enhance the efficiency and lifespan of 3d metal-based catalyst systems.

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