Development of nanostructured and molecularly-defined base metal catalysts for sustainable organic synthesis

The dissertation is mainly related to the development of nanostructured and molecularly-defined base metal catalysts for sustainable organic synthesis of fine and bulk chemicals as well as molecules of interest for life science applications. More specifically, the detailed preparation of cobalt-nanocatalysts using MOFs as precursor is reported for reductive aminations to synthesize functionalized and structurally diverse amines and pharmaceuticals. Application of monosaccharide sugars is showcased for the preparation of monodisperse Ni-nanoparticles as sustainable catalysts for semihydrogenation of alkynes. In addition, molecularly-defined triphos ligated cobalt and nickel catalyst systems have been developed for the synthesis of primary amines via reductive amination and stereodivergent synthesis of alkenes by hydrogenation of alkynes, respectively. Further, cobalt-nanoparticles catalyzed hydrogenation of nitriles and nitro compounds has been performed for the preparation of aromatic, benzylc and aliphatic primary amines. Finally, we have demonstrated the detailed preparation of coordination polymer-derived cobalt nanoparticles-based catalysts for regio-, chemo-, and diastereoselective hydrogenation of various aromatic hydrocarbons to produce the corresponding cyclohexane-based products. The detailed characterization of catalytic materials by TEM, XPS, XRD as well as molecularly-defined complexes by NMR and X-ray crystal structure is presented. In order to design suitable catalysts and to accomplish these synthetic reactions, a number of optimization studies and control experiments including kinetic and mechanistic investigations have been performed.

Cyclohexane demonstriert. Die detaillierte Charakterisierung katalytisch aktiver Materialien durch TEM, XPS, XRD sowie von molekular-definierte Komplexen durch NMR- und Röntgenkristallstruktur wird ebenfalls vorgestellt. Um geeignete Katalysatoren zu entwerfen und diese Synthesereaktionen durchzuführen, wurden eine Reihe von Screening- und Kontrollexperimenten sowie kinetischen und mechanistischen Studien durchgeführt.