Single Atoms of Metals in Carbon-Supported Catalysts

Catalysis is an important part of numerous chemical processes ranging from synthesis of materials and chemicals to reducing harmful chemical emissions and energy conversion processes. Supported catalysts, i.e. nanoparticles of metals or metal oxides attached to the surface of a supporting material, are a common type of heterogeneous catalysts. The support slows down the process of nanoparticle sintering and can in some cases directly or indirectly influence the catalytic process.

Single atom catalysts are supported heterogeneous catalysts in which the dispersed metal does not form a separate phase. Instead, it is atomically distributed on the surface of the support. In the past such materials were occasionally reported in selected narrow fields of heterogeneous catalysis. However, with the increasing availability of atomic resolution electron microscopy, which enables visualization of single atoms on the support, single atom catalysis has become a thriving research field.

Single atoms can be atoms of any of the transition metals in various oxidation states dispersed on the surface of many different substrates (e.g. oxides, metals or carbon materials). Therefore, the properties of single atoms, their type of bonding with the support and their catalytic activity can vary strongly.

In this dissertation I studied single atom catalysts supported on carbon materials. By choosing four commercial catalysts with platinum-group metals (Ru, Rh, Pd, Pt) I showed that single atoms are present alongside supported nanoparticles. This finding suggests that single atoms may be present in numerous supported metal catalysts on carbon and are not just a special case as often suggested in the literature.

Secondly, I describe a porous nitrogen-doped carbon (NC) material with high porosity that due to the ionothermal synthesis procedure contains atomically distributed Zn remains. Electrochemical characterization of the material showed that the Zn atoms influence the electrochemical activity of the NC material even though they are not themselves catalytically active.

Finally, I discuss the preparation and properties of single atoms of Pt on NC support material. As opposed to Pt nanoparticles, the single atoms did not display any activity for electrochemical reactions of oxygen and hydrogen in aqueous electrolytes.

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