# Abstract

The use of molecular oxygen as an environmentally friendly and readily available oxidant is of great importance for technical applications. Due to the low reactivity of $O_2$ with organic substrates, activators are required. Nature has found an efficient solution for the oxygen activation and transfer in the form of the copper-containing enzyme tyrosinase. The development of functional model systems for the active site of tyrosinase targets the transfer of the working principle of the natural system to technical processes.

The biomimetic hybridguanidine ligands used in this thesis prove to be particularly suited for the chemical modeling of tyrosinase. As main reason the good donor properties of the guanidine function can be regarded besides the open coordination space that is created by the smaller amine function and facilitates the entry of a substrate to the $Cu_2O_2$ centre. The exceptional hydroxylation reactivity of hybridguanidine stabilised $Cu_2O_2$ complexes could be demonstrated in oxygen transfer studies to phenolic substrates. In particular, the superiority of the bis(μ-oxo) species $[Cu_2(TMGdmap)_2(\mu\text{-}O)_2][CF_3SO_3]_2$ compared to the analogue bisguanidine and bisamine systems $[Cu_2(btmgp)_2(\mu\text{-}O)_2][CF_3SO_3]_2$ and $[Cu_2(TMPDA)_2(\mu\text{-}O)_2][CF_3SO_3]_2$ has been studied intensively. Numerous experimental and theoretical studies have shown that the initially established working hypothesis, which associated the improved accessibility of the active $Cu_2O_2$ centre with an increase of the hydroxylation reactivity, was correct, which opens up new perspectives in the copper-oxygen hydroxylation chemistry. In this thesis, a ligand library was constructed and the influence of the substituents on the formation and stability of the $Cu_2O_2$ species as well as the resulting hydroxylation activity was examined in detail.

Furthermore, by means of combined EXAFS and Raman spectroscopy it was possible to investigate a bis(μ-oxo) dicopper complex at room temperature in an optically excited state for the first time, whereby new insights were obtained regarding the stability and reactivity of this system.

Moreover, a Cu(I)-hybridguanidine complex could be identified which is well suited as functional model system for type 1 copper centres as well as catalyst in the styrene ATRP due to its excellent electronic and steric properties.