



Invitation à la soutenance publique de thèse de  
Xiao MU

Master degree in the discipline of Inorganic Chemistry

Pour l'obtention du grade de Docteur en sciences

« Histidine Brace Containing Ligand Scaffolds for Developing  
Biomimetics  
of Lytic Polysaccharide Monooxygenases »

qui se déroulera  
le mercredi 09 décembre 2020 à 16h  
En visioconférence  
1348 Louvain-la-Neuve

#### Jury members :

Prof. Michael Singleton (UCLouvain), promoter  
Prof. Olivier Riant (UCLouvain), co-promoter  
Prof. Yann Garcia (UCLouvain), chairperson  
Prof. Tom Leyssens (UCLouvain), secretary  
Dr. Koen Robeyns (UCLouvain)  
Prof. Pedro Fardim (KU Leuven, Belgium)  
Dr. Victor Maurizot (Université de Bordeaux, France)

Lytic polysaccharide monooxygenases (LPMOs) have attracted considerable attention due to their ability to enhance enzymatic degradation of recalcitrant polysaccharide biomass via an oxidative cleavage mechanism. The active site of LPMOs contains a copper ion in a T-shaped N<sub>3</sub> coordination environment known as the histidine brace motif. Beyond this, the role of certain first and second coordination sphere features is unclear from the biological studies. Understanding the factors that contribute to the high oxidative capabilities of the enzyme can facilitate the design of future catalysts for biomass conversion. The aim of this research is the design and synthesis of new model complexes of the LPMO active site, specifically the histidine brace.

First, four small molecule ligands containing two imidazole groups connected by an aliphatic amide linker were designed and synthesized successfully. The amide ligands were used to coordinate with CuCl<sub>2</sub> to form the Cu(II) complexes in the presence of Et<sub>3</sub>N as base. The UV-Vis spectrophotometric titration, Job's plot analysis and MS data suggest that the stoichiometries between CuCl<sub>2</sub> and the ligands in MeOH and H<sub>2</sub>O were 1:1. The FTIR data indicate the participation of the amide for Cu(II) coordination, and the ligand could provide the N<sub>3</sub> coordination environment to copper.

In the second part, the use of conformationally stable and predictable folded aromatic oligoamides as ligand scaffolds can represent a powerful tool to design first and second coordination spheres. Based on this idea, two scaffold ligands, containing imidazole/histamine functionalized quinoline units and diazaanthracene were designed and synthesized successfully. The UV-vis spectra of the reaction of Cu(II) salts with scaffold ligands suggest that the scaffolds do coordinate a Cu(II) center. However, additional structural studies are needed to validate the exact coordination environment of Cu and see if it matches with the active site of LPMO.