

Intitulé du Sujet de Thèse : Supported artificial metalloenzymes for aerobic oxidation of organic compounds

Laboratoire : iSm2

Equipe Biosciences

Directeur de thèse : Yasmina MEKMOUCHE

Encadrant (éventuellement) :

Co-encadrant (éventuellement) :

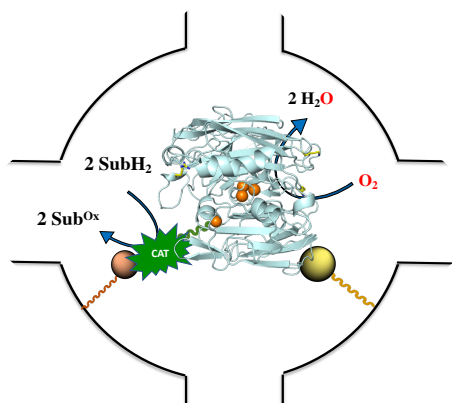
email : y.mekmouche@univ-amu.fr

Contexte de l'étude

The development of new processes for sustainable chemistry is at the heart of the great challenges of tomorrow. Regarding heterogeneous catalysis, major breakthroughs are expected in terms of functionalization and surface modification as well as in the development of nano-catalysts. Indeed, there is a growing interest on achieving a comprehensive view of processes that control all mechanistic aspects related to stability, efficiency and selectivity of reactions catalyzed by these systems. It is of great importance to understand fundamental synthesis and processing concepts for tailoring materials and controlling all involved mechanistic aspects.

Descriptif du projet

The aim of this project is to develop a new generation of supported chemoenzymatic catalyst for sustainable cooperative oxidation of organic compounds. It is based on the tandem use of a synthetic transition-metal-based catalysts with a robust multi-copper oxidase in order to couple the selective oxidation of organic compounds to a safe four-electron/four proton reduction of the renewable dioxygen to water. These catalysts are intended to be confined and immobilized into a tailor-made silica foam where the control of the orientation of the catalysts relative to each other and to the silica surface will define unique interfaces. Preliminary results suggest that the support is more than a matrix ensuring stability and reusability by playing a role in the catalytic process. Therefore, a high cooperativity is expected due to the vicinity and the organization of the components. These hybrid materials will be primarily optimized for alcohol oxidation and aza-Wacker type cyclisation reactions.



Schematic representation of an organized hybrid material composed of a porous silica foam functionalized with a chemoenzymatic catalyst for aerobic oxidation of organic compound

The project will stand on the exploitation and integration of different scientific approaches to build up and study the different components of the system: materials synthesis; production of the bio-catalyst; immobilization/orientation of the catalysts into the material; catalytic efficacy of the supported catalyst on model reactions; optimization of hybrid materials for continuous flow catalysis. This project is a collaborative project (R. Backov, CRPP Bordeaux and E. Deniau, UCCS Lille).

Keywords

Heterogeneous catalysis, artificial metalloenzyme, aerobic oxidation, sustainable chemistry.

Candidate profile and procedure

Successful applicants will have obtained excellent grades in his/her Bachelor and Masters degrees (or equivalent). He/she should also be well-motivated, hardworking, willing and able to work as part of a team. Candidates for the PhD position should have a Masters' degree Chemistry, Chemical Engineering or Materials Science. Background / experience in catalysis would be beneficial.

Applicants are invited to send their CV, a cover letter, their transcripts of academic records, and the contact information for at least two references to Yasmina MEKMOUCHE (y.mekmouche@univ-amu.fr) before April 30th.

Audition of the candidate by the scientific ED250 doctoral school committee will take place June 7th and 8th, 2021

Références bibliographiques

i) Y. Mekmouche, L. Schneider, P. Rousselot-Pailley, B. Faure, A. J. Simaan, C. Bochot, M. Réglie, T. Tron *Chem. Sci.* **2015**, 6, 1247 ; *ii)* A. Roucher, V. Schmitt, J.L. Blin, R. Backov *J Sol-Gel Sci Technol* **2019**, 90, 95 ; S. Gentil, P. Rousselot-pailley, F. Sancho, V. Robert, Y. Mekmouche, V. Guallar, T. Tron, A. Le Goff, *Chemistry - A European Journal*, **2020**, 26, 4798-4804 ; ; *iii)* K. Engström, E. V. Johnston, O. Verho, K. P. J. Gustafson, M. Shakeri, C.-W. Tai, J.-E. Beckvall *Angew. Chem. Int. Ed.* **2013**, 52, 14006–14010.