Institut de Chimie des Milieux et Matériaux de Poitiers

funded 3-year PhD project, autumn 2024

Towards *in silico* prediction of two- and three-dimensional porous materials for catalysis

in *Applied Quantum Chemistry* group at IC2MP, Poitiers University – CNRS (France)

Advisors: Dr Gilles Frapper & Dr Frédéric Guégan

SUMMARY: the aims of this PhD project are the conception of an open-source code to build 2D or 3D networks from the only knowledge of three building blocks (metal center, ligand and counter ion/solvent, e.g., MOF design), coupled with (machine learning enhanced) quantum chemistry calculations and the application of this methodology to predict novel porous catalytic materials for NH_3 production as a proof-of-concept (heterogenous catalysis at interfaces).

Overview. Functional porous materials based on two- and three-dimensional coordination polymers are conceptually the result of a fantastic Lego game. Indeed, their crystalline framework is the result of the arrangement of elementary components: a metal site, associated with a (redox) ligand, and a counter ion inserted into the cavities of the metal-ligand framework. The intimate association of these three components metal, ligand, and counter-ions, in a key-lock-like game, modulates the physicochemical properties of the material: crystallinity, network topology, conducting behavior, magnetic order, catalytic activity, etc. Exploring the possibilities of such an ensemble by usual methods, whether experimental or theoretical, is dizzying. A change of paradigm is thus required.

Aims. This multidisciplinary project - materials sciences, heterogenous catalysis, quantum chemistry, programming -, aims to develop a methodology for designing and predicting functional Metal Organic Frameworks (MOFs, coordination polymers) through an intelligent exploration of all the possibilities offered by metal/ligand/counter ions combinations. Machine-learning techniques, metaheuristic algorithms such as genetic algorithms, and high-throughput first-principles calculations will help in the design and prediction of new porous materials. The focus of this project is on two- and three-dimensional networks with large accessible surfaces, to enable significant interfacial chemical reactivity of N_2 hydrogenation (NH_3 synthesis) as a proof-of-concept. Special attention will be given to the impact of anions and/or solvents on the dimensionality of the network (2D/3D), the shape of the cavities, and the resulting interfaces due to the expected template effect.

To quickly sort the large pool of MOF candidates, we aim to develop a dedicated tool based on reactivity descriptors obtained from quantum chemistry calculations that probe potential catalytic sites. The most powerful catalysts will undergo a thorough analysis, including refined crystal structure optimization through periodic DFT simulations, assessment of the viability of 2D or 3D materials in terms of thermodynamic, dynamical, mechanics, thermal, and chemical stabilities, and explicit computation of reaction profiles (such as H_2 and N_2 activation and NH_3 production). A comparative analysis with related catalysts will be conducted to rationalize the key factors governing interfacial activity.

Gilles FRAPPER, IC2MP Institut de Chimie des Milieux et Matériaux de Poitiers
UMR 7285 CNRS – Université de Poitiers – Bureau 116, Bâtiment B27
ic2mp.labo.univ-poitiers.fr – gilles.frapper@univ-poitiers.fr

cnrs Université de Poitiers

This 3-year PhD is funded by EUR Intree for 3 years (gross salary: 2100 €/month, net ~1700 €/month including social security etc… PhD student will be enrolled at Ecole Doctorale Rosalind Franklin of Poitiers University, ideally starting in October 2024. If the candidate is a French speaker, she/he may have the opportunity to teach undergraduates and to earn extra salary.

Consortium. This PhD research topic is related to our ANR project MAGDESIGN (2022-2027) and the High-Performance Centers TGCC/IDRIS/CINES GENCI project, which can provide the necessary technical and financial support (work environment, workshops and conferences, publication editing…) This research program involves collaboration with national and international experimental and theoretical experts, creating a fruitful scientific environment for the candidate e.g. in Crystal Structure Prediction (CSP) field, Machine-learning Potentials, DFT modelling of reactivity (XtalOpt, PyXtal, USPEX, AIRSS/EDDP, Autografs, DFTB+, VASP codes among others…).

Illustrative articles by supervisors in the field of "Computational Materials Discovery".

"Structurally Constrained Evolutionary Algorithm for the Discovery and Design of Metastable Phases", JOURNAL of CHEMICAL THEORY and COMPUTATION, 2023, 19, 21, pp. 7960–7971 DOI: 10.1021/acs.jctc.3c00594

"*Design of Metastable Oxychalcogenide Phases by Topochemical (de)intercalation of Sulfur in $La_2O_2S_2$*", NATURE COMMUNICATIONS, 2021, 12 (1), p. 3605. DOI: 10.1038/s41467-021-23677-w

"*Prediction of a New Layered Polymorph of FeS_2 with $Fe^{3+}S^{2-}(S_2^{2-})_{1/2}$ Structure*, JOURNAL OF PHYSICAL CHEMISTRY LETTERS, 2020, 11 (20), pp.8861-8866 DOI: 10.1021/acs.jpclett.0c02543

"*Predicted Polymeric and Layered Covalent Networks in Transition Metal Pentazolate $M(cyclo\text{-}N_5)_x$ Phases at Ambient and High Pressure: Up to 20 Nitrogen Atoms per Metal*", CHEMISTRY OF MATERIALS, 2021, 33 (13) , pp.5298-5307 DOI: 10.1021/acs.chemmater.1c01400

ORCID: Gilles Frapper https://orcid.org/0000-0001-5177-6691
Frédéric Guégan https://orcid.org/0000-0002-4932-8643

Our research group. The Applied Quantum Chemistry group comprises two academics, three post-doctoral researchers, five PhD students, and interns pursuing Bachelor's or Master's degrees.
IC2MP is the Chemistry Institute of Poitiers U. and CNRS. It has ~200 members including ~90 PhD students.
Poitiers is a charming Romanesque and quiet city, 1h20 from Paris and Atlantic Ocean by high-speed train.

Profile. The ideal candidate should possess a Master's degree in Materials Science or Quantum Chemistry, with expertise in molecular and/or periodic DFT (a plus: in crystal structure prediction methods, machine-learned potentials…). Proficiency in programming languages such as Python is also required.

How to apply: Interested candidate should send their meaningful applications in a single PDF file to gilles.frapper@univ-poitiers.fr . The file should content a CV, a motivation letter, transcript of degree certificates (ranking), and the contact details of at least two referees before may 1rst, 2024.

Applications will be examined until the position is filled.

Gilles FRAPPER, IC2MP Institut de Chimie des Milieux et Matériaux de Poitiers
UMR 7285 CNRS – Université de Poitiers – Bureau 116, Bâtiment B27
ic2mp.labo.univ-poitiers.fr – gilles.frapper@univ-poitiers.fr