Start : 1st October 2023

Keywords: Theoretical Modeling, Density Functional Theory, Single Atom Catalysis,

Candidate Profile : Strong background in physics or chemistry or materials science with an interest for atomistic simulations. Good knowledge of quantum mechanics. Experience with Linux environment and python programming will be appreciated. Please send a motivation letter, CV and transcripts, along with names and contact information of two referees.

Context :
Single-atom catalysis (SAC) has recently emerged as a new and promising field at the frontier between homogeneous and heterogeneous catalysis. A decisive challenge lies in the stabilization of single metal atoms while promoting their catalytic efficiency. Here, carbon matrix-isolated non-noble metal atoms are targetted for the selective hydrogenation of hydrocarbons (acetylene, butadiene) and oxygenates (levulinic and succinic acids). Those are important reactions for polymer and platform chemical synthesis, which are typically activated by expensive and insufficiently selective supported palladium nanocatalysts. The proposed knowledge-based approach will rely on advanced photon/photoelectron spectroscopy, electron microscopy, and theoretical modelling for the discovery of cost-effective SACs. The synthesis of phosphorus-doped carbon-based SACs will be developed in order to stabilize the metal atom and tune its coordination toward optimal performance for sustainable alkene and lactone production.

Objectives :
The thesis focuses on the theoretical modeling of the structures and properties of SACs, as well as the computation of spectroscopic signatures. Objectives include the investigation of electronic effects in selective hydrogenations by SACs, the design of optimal SACs within a given system, by tuning both the active metal M, its loading, and the support in interaction with M. Expected results include a thorough understanding of the relationship between the structure/composition and the catalytic properties, as well as the drawing of a general picture rationalizing the behavior of SACs in several systems to guide the discovery of new low-cost and efficient catalysts.

Methods :
Understanding the atomic-scale surface structure of single atom catalysts is a crucial step on the path toward designing optimal heterogeneous catalysts. We plan to combine spectroscopy and theoretical calculations to facilitate the design and development of single atom materials with fine-tuned catalytic properties. Density Functional Theory will be used to model the systems, guide the SAC selection through high-throughput predictions of SAC reactivity, and help in the interpretation of experimental data.

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Location :
The Institut Jean Lamour (IJL, Univ. Lorraine & CNRS) is a lab for fundamental and applied research in materials science; it brings together scientists from different disciplines. The lab is located in Nancy (France), a nice city in the heart of Europe (close to Germany, Belgium, Luxembourg) and direct from Paris by TGV. This project takes place within the ANR ISAC, gathering IJL Nancy and IRCELYON. It also has an international dimension and takes place within the *Integrated European Center for the Development of New Alloys and Metallic Compounds (https://ecmetac.eu/)*.