Computational modeling of complex physicochemical processes in energy- and sustainability-related catalysis

Timeline | 1/9/2024 to 31/8/2027

ICIQ People | Albert Solé-Daura

Budget | 297,900 €

Call | Junior Leader La Caixa

SUMMARY

Spin-forbidden reactions play a central role in a wide variety of fields, including catalysis, materials science, and photochemistry. However, very little is known about the mechanisms governing these processes at the atomic level, hindering the development of design rules to improve their efficiency. This knowledge gap arises from the prohibitively high computational cost associated with current simulation methods, which typically limits their scope to exceedingly small systems in the absence of an explicit environment. Consequently, the vast majority of systems suited for practical applications remain largely unexplored. Therefore, more affordable computational strategies are clearly needed to bridge the current gap between systems amenable to computational study and those employed in experimental applications.

This project aims to develop a novel, cost-effective computational strategy (SURFXsim) to unlock the atomistic simulation of spin-forbidden events in medium- to large-sized systems consisting of several hundred atoms. The SURFXsim method relies on the application of DFT-based Molecular Dynamics (MD) simulations, combined with enhanced-sampling techniques and in-house developed collective variables to bias chemical systems between two minima located on potential energy surfaces of distinct spin multiplicities.

In addition to offering atomically resolved mechanistic details, this technique will allow for the reconstruction of free-energy profiles along reactive events, providing access to estimates of kinetic and thermodynamic parameters. As part of this project, SURFXsim will be employed to investigate the reaction mechanisms underlying strategic processes in the fields of sustainability- and energy-related catalysis promoted by polyoxometalates.

The project that gave rise to these results received the support of a fellowship from "la Caixa" Foundation (ID 100010434). The fellowship code is LCF/BQ/PI24/12040012.