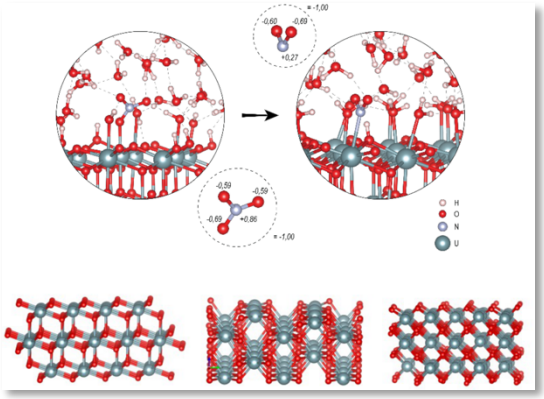
Open PhD thesis at
Institut de Chimie Séparative de Marcoule (ICSM), Bagnols-sur-Cèze, France
Ecole doctorale 459 Sciences Chimiques Balard (University of Montpellier)

Quantum dynamics for the dissolution of UO_2

Starting date: September 2023



The **dissolution of UO_2** in nitric media has been studied for decades in order to understand, control, and optimize the recycling of spent nuclear fuel. The **reaction mechanisms are complex to understand** by experimental methods and therefore remain very hypothesized even today. Although atomistic simulations can provide answers, dissolution is a **complex physico-chemical process** and must be treated by considering the electrons – thus at the **quantum level** – because the kinetically determining steps are surface chemical reactions.

Recent computing resources and the advent of HPC[1] make *ab initio* molecular dynamics (AIMD) simulations capable of describing systems large enough to determine **macroscopic thermodynamic data**. **AIMD simulations** have been expanding rapidly in recent years and are now commonly applied to liquid/solid interfaces. At present, very few publications dealing with the dissolution of mineral species by atomistic methods exist in the literature and none on UO_2. Proving, in the case of UO_2, that AIMD is a routine tool for dissolution, in general, would be a **considerable methodological advance**.

Preliminary results obtained at ICSM have made it possible to observe the spontaneous transformation, during the AIMD simulation, of the nitrate ion into nitrite ion on contact with the UO_2 surface. The PhD student will **establish the reaction mechanisms**, **their equilibria**, and **their properties**. The influence of crystal orientations, surface defects, and impurities on the reaction mechanisms will be explored. The work will mainly consist of the use of **AIMD simulations**, using the VASP code, on large national computers. Biased AIMD simulations, such as Blue-Moon or Slow-Growth, will be required to determine the **kinetic and thermodynamic quantities related to dissolution**. The **interface between molecular simulations and thermodynamics** requires the student to have a good knowledge of both. This is a little-explored field, but one whose scientific and industrial development in the coming years is certain to bring many opportunities to the future graduate.

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Net salary: ~1650 € / month (Gross salary: ~2100 € / month)

Further information: The successful candidate will join the LMCT group of ICSM and will be enrolled at the doctoral school ED459 Sciences Chimiques Balard of the University of Montpellier (France). The thesis will be done in collaboration with ENS Geology Nancy.

Contact: To apply, please send a cover letter, and a detailed CV to Dr. Bertrand Siboulet (bertrand.siboulet@cea.fr) et Pr. Jean-François Dufrêche (jean-francois.dufreche@icsm.fr).

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[1] HPC: High Performance Computing