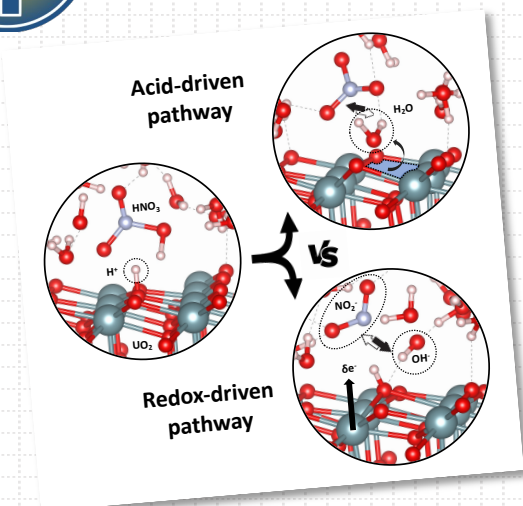




PhD DEFENSE



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Study of UO_2 dissolution using *ab initio* molecular dynamics simulations



**Tuesday,
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2026**



8:30 AM



**Auditorium
ICSM**

The dissolution of uranium dioxide (UO_2) in nitric acid is a key step in spent nuclear fuel reprocessing, yet the elementary mechanisms initiating this process remain poorly understood at the atomic scale. This thesis provides a unified description of these early stages, primarily based on *ab initio* molecular dynamics simulations within the framework of density functional theory (DFT). The characterization of hydrated actinide dioxide interfaces first establishes the roles of hydration and protonation in their reactivity. The study of nitric acid dissolution then reveals two coexisting pathways: a general acid-base pathway, common to metal oxides, and a UO_2 -specific redox path-way, in which uranium oxidation is coupled to acid reduction through interfacial electron transfer enabled by the partially occupied uranium 5f states. The simulations show that $\text{NO}_2^\bullet$ can accept an electron from UO_2 to form NO_2^- and U(V) , while extraction leads to a solvated uranyl(V) species. Combined with the equilibria between NO_2^- , HNO_2 and NO_x species, these results support a self-accelerating dissolution mechanism in which $\text{NO}_2^\bullet$ constitutes the active catalytic species sustaining the propagation process. They thus establish an atomistic link between interfacial electron transfer and the experimentally observed catalytic behavior.

Keywords: Dissolution; Uranium dioxide (UO_2); *ab initio* Molecular Dynamics (AIMD); Density Functional Theory (DFT); VASP



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