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Corrosion modeling of spent fuel – adsorption of O, O₂, H₂O and H₂O₂ on the UO₂ surface

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Uraniumdioxide (UO₂) is the main component of nuclear fuels. This compound is very susceptible to oxidation, which results in formation of the mixed-valence oxide U₃O₈. [1] From an application point of view, this interest relates to the safe and sustainable management of nuclear fuel. Our goal is an understanding of this oxidation process at the atomic level, using ab initio electronic structure calculations.

The focus in this work lies on the early oxidation of stoichiometric UO₂, where excess oxygen can be treated as interstitial point defects. In order to study this oxidation we investigate the adsorption of the O atom and oxygen containing molecules (O₂, H₂O, H₂O₂) on the three most stable UO₂ surfaces ((111), (110) and (100)). The calculations are performed within DFT, using PBE+U including spin-orbit coupling and taking into account the non-collinear 3k (transverse) magnetic ground state.

Dissociative adsorption is found for H₂O and H₂O₂ on the (111) and (100) UO₂ surface respectively, forming hydroxyl groups on the surface. Adsorption on the UO₂ surface lowers the barrier for dissociation of O₂. The defect formation energy is calculated for interstitial oxygen, up to several layers into the material starting from the surface. Additionally adsorption energies are studied as a function of temperature and partial pressure.

References

[1] E. De Bona, K. Pope, O. Walter, M. Cologna, C. Hennig, A. C. Scheinost, and D. Prieur., *Inorg. Chem.* **61**, 1843 (2020).

