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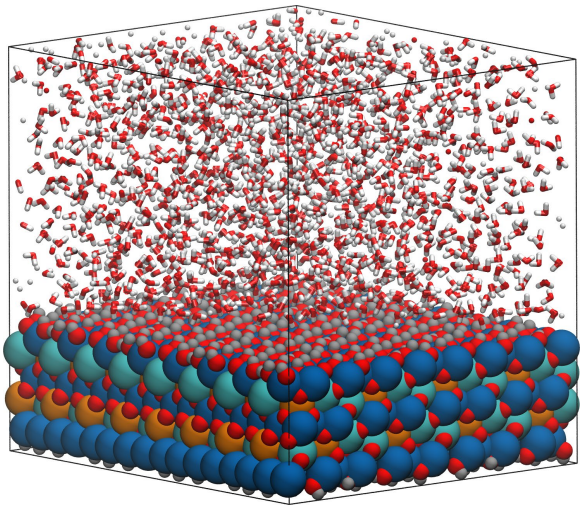

Ab initio modelling of magnetite surfaces for radionuclide retention

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Thick steel casks are used for radioactive waste disposal in deep geological repositories. However, it is expected that steel corrodes over time. The corrosion products are expected to form mixed iron oxides, mainly magnetite. After tens of thousands of years, casks may breach allowing leaching of the radiotoxic elements, such as plutonium and technetium, by host rock pore-water. The dissolved radionuclides can then interact with the steel corrosion products and be adsorbed or incorporated into the solids [1]. But since these interaction mechanisms are poorly understood at the atomistic scale, our goal is to better understand them by using computer simulations alongside experiments [2].

In this computational study we identified the dominant low index surfaces on nano-magnetite particles and their termination at the relevant conditions based on Kohn-Sham density functional theory (DFT). This was done using the open-source code CP2K. The DFT+*U* method was employed for the strongly correlated *3d* and *5f* electrons of iron and plutonium, respectively. The Hubbard *U* parameter was determined by comparing experimental cell parameters and band gaps to our results [3]. With this revised model, we examined the preferential magnetite crystal orientation plane (111) with different surface terminations as a function of oxygen and water fugacity. Based on our modelling, we found the most stable magnetite (111) surfaces under real repository conditions being Fe_{oct1}-O-H and Fe_{tet1}-O-H. Furthermore, we used classical and *ab initio* MD simulations to investigate the behaviour of radionuclides at the water-magnetite interface in deep geological repositories.

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