FIRST-PRINCIPLES SIMULATIONS ON INITIAL STAGE OF URANIUM NITRIDE SURFACE OXIDATION

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As a promising fuel material for the Generation-IV fast reactors, uranium mononitride (UN) reveals unwanted oxidation in air, which greatly affects its properties, in contrast to a "traditional" UO_2 nuclear fuel. Thus, it is important to understand the mechanism of UN oxidation and possible steps for inhibition of this process. In this study, we present results of large-scale first-principles atomistic simulations of oxygen adsorption [1], dissociation [2] and diffusion upon the UN(001) surface (both perfect and defective, with regularly distributed single vacancies [3]).

The plane-wave DFT spin-polarized calculations (using the VASP computational package [4]) for basic properties, *e.g.*, reactivity, of the UN(001) surface have been performed on various 3D slab models [1-3]. Obtained results clearly demonstrate: (*i*) metallic-covalent inter-atomic bonding inside the substrate, (*ii*) possibility of spontaneous dissociation of oxygen molecule adsorbed upon the appropriate surface sites, (*iii*) further localization of oxygen adatoms, released after dissociation, upon the surface U atoms, (*iv*) high mobility of O adatoms along the surface, due to low migration barriers (~0.5 eV) between the two neighbouring adsorption sites upon the surface uranium atoms. The oxygen adatom atop the surface U atom nearest to the N vacancy can be spontaneously captured by the latter. Possibilities of further oxygen adatom migration between the adjacent vacancies on the UN(001) surface are discussed too. The results for O atom penetration into UN bulk [5] and surface layer are compared and verified (using experimental UPS data).

References

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