Adsorption and Dissociation of Iodine Molecules on a Zr Surface

Iodine is a fission product of uranium. It can attack the inner side of zircaloy cladding in nuclear power reactors leading to cracking and fracture. Computations show that iodine molecules adsorb and dissociate on a zirconium surface without an energy barrier. The binding energy of iodine on this surface is large (nearly 300 kJ/mol per iodine atom), but the barriers for surface diffusion is only 6.8 kJ/mol. This gives rise to rapid surface diffusion allowing iodine atoms to reach the crack tips faster than the propagation of cracks.

Keywords: adsorption, dissociation, iodine, zirconium, computation

Experimental facts
Iodine has been reported to increase brittle fracture of zirconium with an increase of intergranular cracking [1]. This is a safety concern in nuclear power reactors since this type of fuel-cladding interaction can cause a weakening of zircaloy thus being one of the factors which limit the life time of a fuel assembly.

Computed results
Computations of the adsorption and dissociation of iodine molecules on the most stable zirconium surface, namely Zr(0001) surface reveal that this process occurs without any energy barrier as illustrated in Fig. 1. The adsorption energy is large (nearly 300 kJ/mol per I atom), while the diffusion barriers are very low, namely 6.8 kJ/mol as given in [2].

Figure 1. Computed energy profile of the adsorption and dissociation of an iodine molecule on a Zr(0001) surface. Note the slight elevation of the surface zirconium atoms at point (C), indicating a strong Zr-I affinity.
Significance

The quantitative understanding and description of interaction of fission products with cladding materials is of fundamental importance for the safe operation of nuclear power reactors. First-principles electronic structure calculations give detailed insight into these interactions and provide quantitative thermodynamic and kinetic data which can be used to model and predict the life-time behavior of fuel rods in nuclear power reactors. This type of simulations can help to increase the safety while potentially allowing a prolongation of the life time of the fuel elements.

MedeA modules used for this application

The present calculations were performed with the MedeA platform using the following integrated modules of the MedeA software environment:

- MedeA framework including crystal structure builders and geometric analysis tools
- JobServer and TaskServers
- VASP 5.2 and its graphical user interface as integrated in MedeA
- MedeA Transition state search (TSS) module

References
