P14A

Ab initio study for plasma facing components for ITER

A. Allouche

Physique des Interactions Ioniques et Moléculaires,
CNRS and Université de Provence (UMR6633)
Campus Scientifique de Saint Jérôme, service 242
13397 Marseille Cedex 20 - FRANCE

Motivations

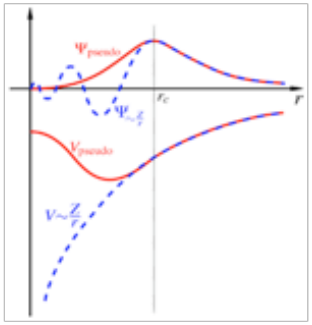
Quantum theory within the DFT frameworks provides very powerful tools in analyzing the fundamental chemical processes governing the reactivity of plasma facing surfaces in nuclear fusion devices. In this contribution we present *plane-waves* DFT calculations results on the reactivity of surfaces of interest in fusion devices.

Oxygen and water are always present in the tokamaks (because of accidental leaks) or in laboratory experiment. After beryllium oxidation by pure oxygen, we present here water molecule and water-oxygen adsorption and dissociation on Be(0001).

Deposition of a lithium thin layer onto graphite was found to considerably suppress physical sputtering. DFT calculations are developed on lithium interaction with pristine and defective graphite surfaces, oxidized or not, in order to evaluate hydrogen retention and restitution. Li is found to significantly enhance H/D adsorption. Oxygenation or/ and perturbation (single vacancies) is shown to give rise to complex structures modifying hydrogen trapping on surface and inducing Li diffusion towards the inner graphite layers.

Computational details

➳ *PBE* GGA functional + ultrasoft pseudopotentials and plane wave Bloch functions



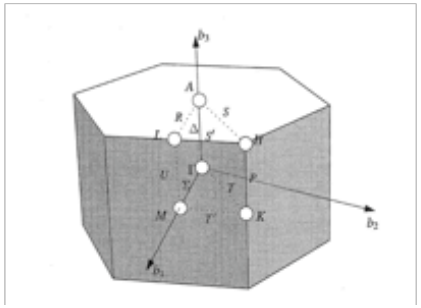
➳ Small systems ~ 100 atoms

➳ Only stationary states calculation ☝ ***No dynamics !***

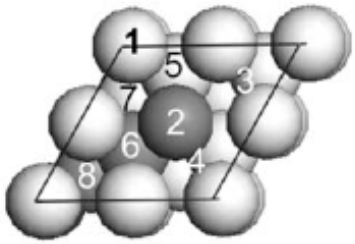
➳ Energy profiles (PES): each configuration is fully optimized (BFGS gradient optimization) excepted the selected reaction coordinates.

➳ The barriers of activation are calculated using the Nudged Elastic Band theory

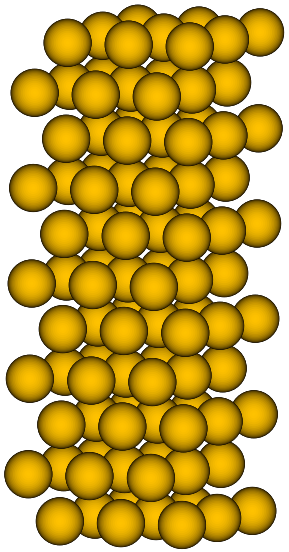
➳ BZ sampling 6x6x1 (surface) 6x6x6 (bulk)

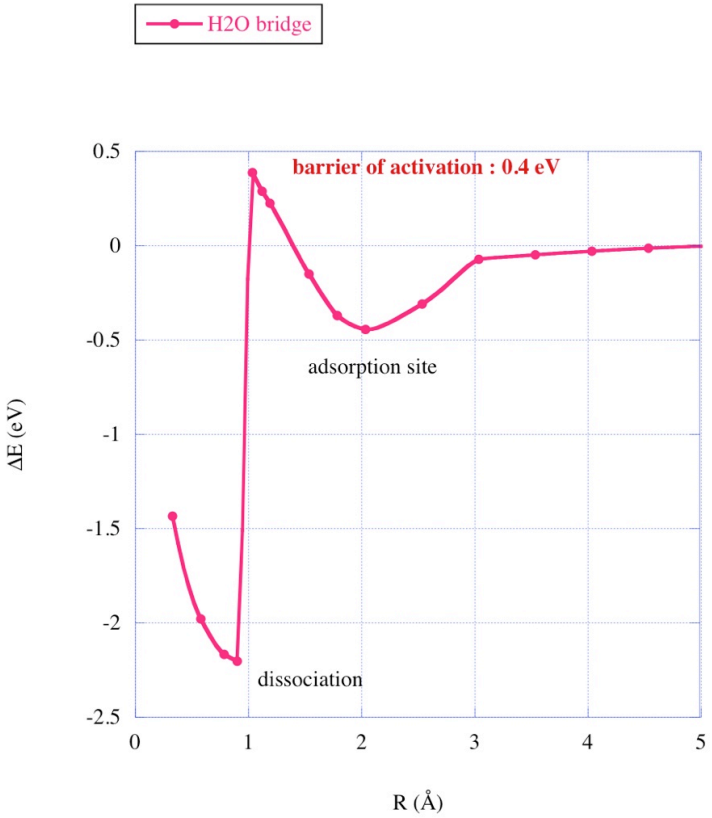


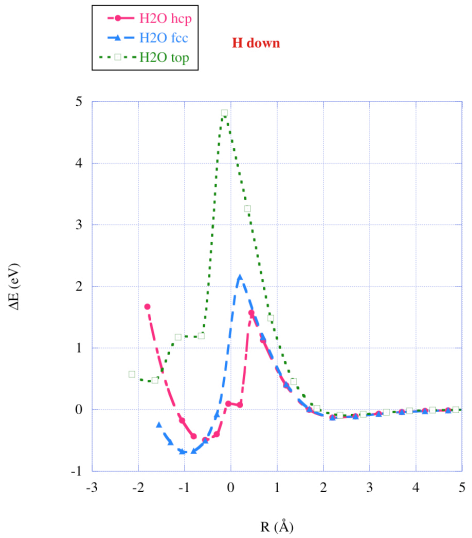
Single water molecule on pure Be



1,2: Top; 3: bridge,
5,6: hcp; 7,8: fcc

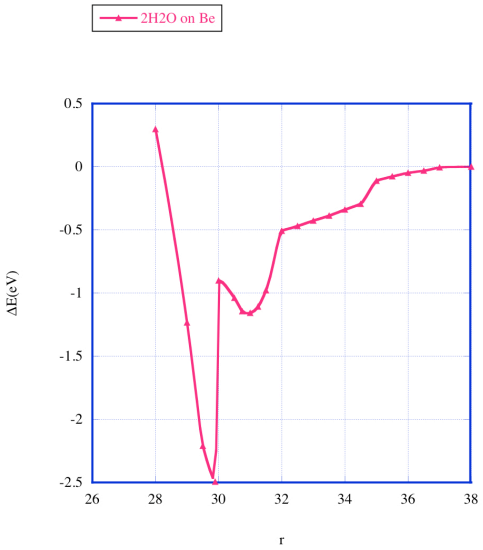


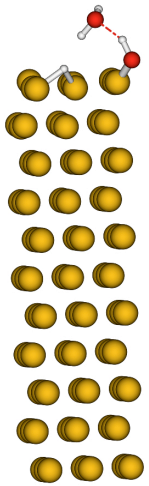




H_2O dissociates only on bridge site.
Other sites: high barrier of activation and weak adsorption.

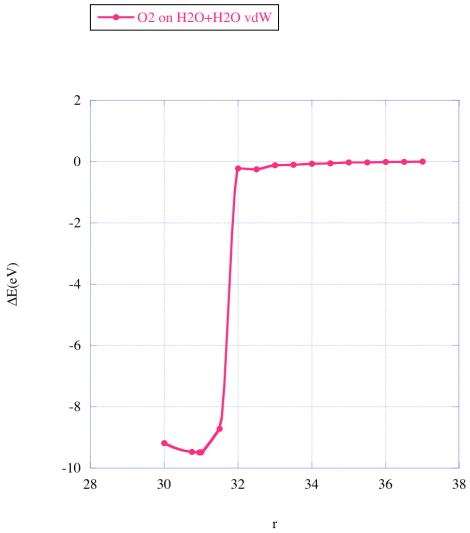
2 water molecules on pure Be

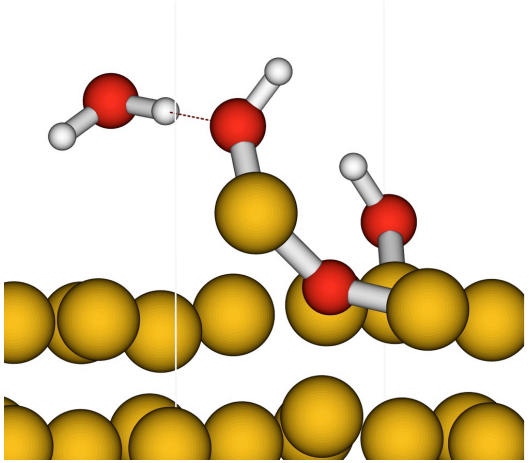




Dissociation + vdW bonding

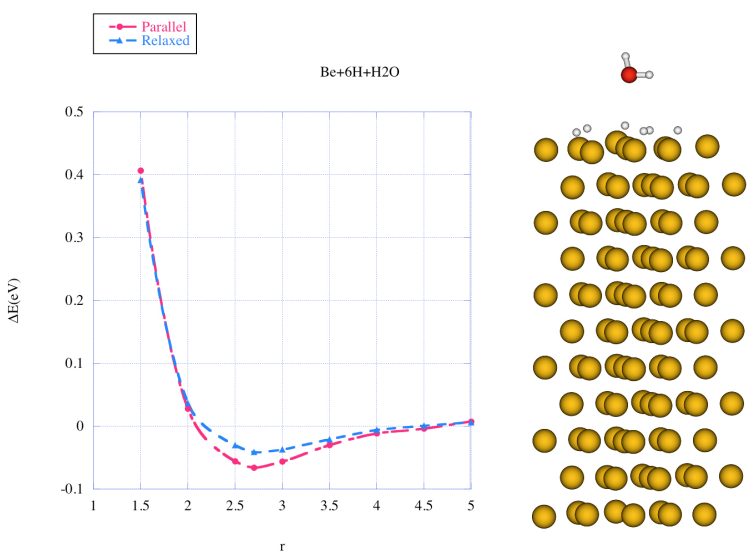
$O_2 + 2H_2O$ on pure Be

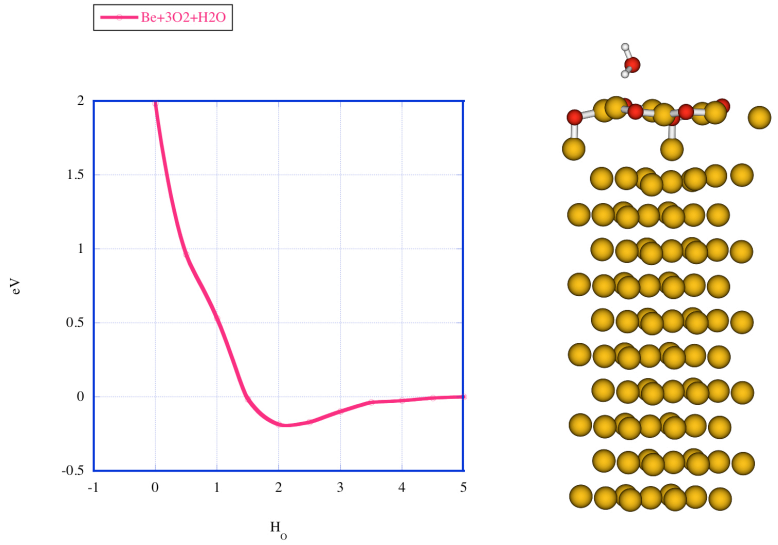




Barrierless dissociation
+ surface corrosion

Water molecules on pre-adsorbed H or O on Be





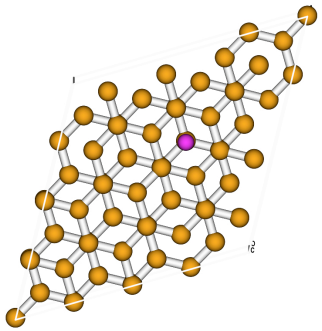
H passivation

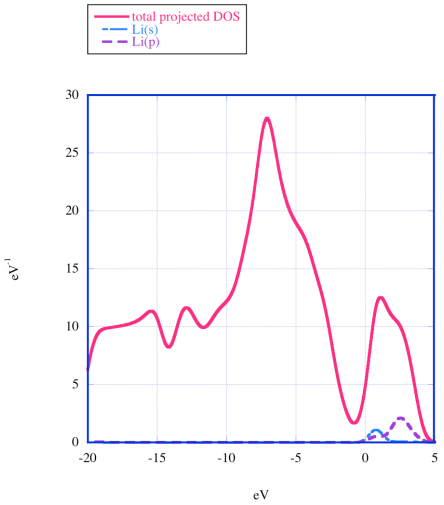
Weak interaction similar to on BeO crystal

Non definitive conclusion

1) Water molecule dissociates only on one symmetry site of the Be(0001) surface
2) An extra molecule favors dissociation
3) Hydrogenation and oxidation passivate the surface layer
4) O2 + H2O co-adsorption → exothermic dissociation and surface modification

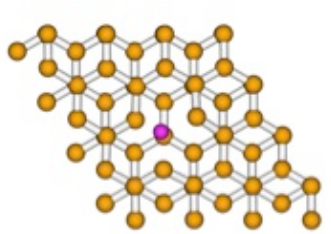
Lithium atom adsorption





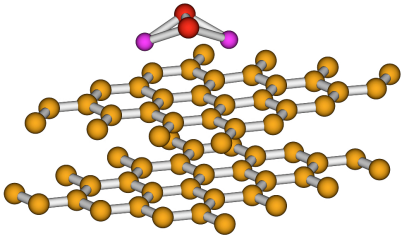
DOS of Li adsorbed on graphite

According to the Li position above the surface ➠ Li or Li^+

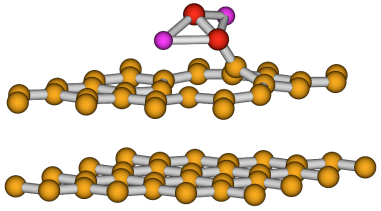


Model of defective surface ➠ atomic vacancy A single vacancy on the surface is a good approximation of the reactivity of zigzag and armchair edges

Li peroxide formation

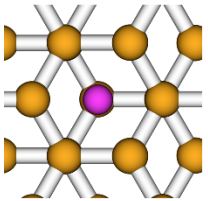


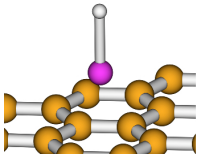
pristine surface



defective surface

	Configuration	ΔE	d(Li-H)	Q(Li)	Q(H)
①	Li adsorption on bare graphite surface (***h*** site)	-1.46		0.65	
②	H adsorption on bare graphite surface	-0.85			0.10
③	Pseudo LiH on Gr	-1.42	1.73	0.21	-0.28
③	H close to Li on Gr	-1.15	1.88	0.58	-0.03
③	H far from Li on Gr	-1.26	5.71	0.45	0.09
④	Li adsorption on V_c	-2.83		0.46	
④	H adsorption on V_c	-4.28			0.06
⑤	Pseudo LiH on V_c	-0.68	1.81	0.21	-0.20
⑤	H far from Li and V_c	-1.48	5.24		
⑤	H on V_c close to Li	-3.94	2.49	0.59	0.03





① ***Weak long range Li - C interactions***

② ***Excellent agreement between PBE-D and high level ab initio methods***

③ & ② ***Li always enhances H bonding, even at long distance***

④ ***Higher reactivity of the vacancy for individual atom adsorption***

⑤ ***Li adsorption counterbalance the vacancy***