Role of the main adsorption modes in the interaction of a dye-sensitized solar cell [COOH-TPP-Zn(II)] on the periodic TiO$_2$ slab exposing rutile (110) surface
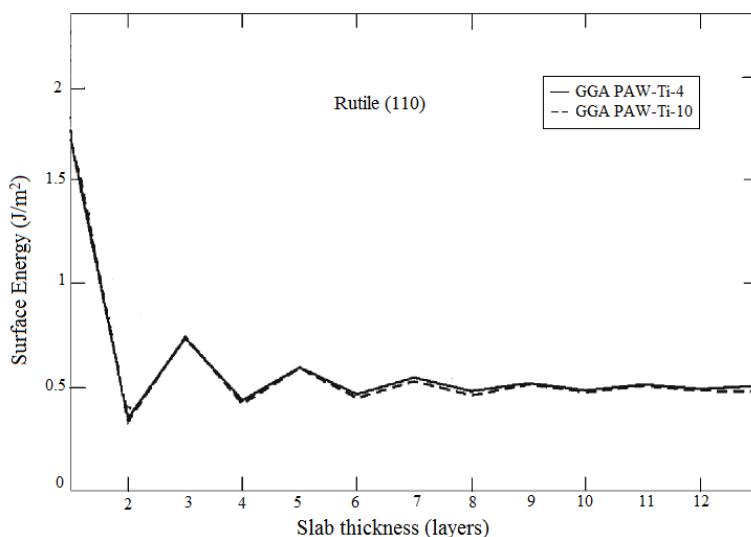
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Surface Model: Rutile TiO$_2$(110)

Our model consists of the molecule absorbed on a two layers periodically repeated TiO$_2$ slab. The slabs have been constructed using the lattice parameter optimized for the bulk and reported in a previous work[a]. The goal was to calculate geometric bulk parameters, bulk moduli, energy gaps and relative stabilities of the TiO$_2$ rutile determined from periodic DFT calculations to validate the methodology used and then to propose a realistic surface model, which was chosen as a compromise between the computational cost, the number of layers to describe the system and the idea of minimizing the interaction between repeated dye images along directions parallel to the surface. This surface model had to be adapted to the size of the molecules to investigate, which were of a large size. In Figure S1 shows that for the two-layer model of the surface there is not significant surface energy change respect to the models of 4, 6, 8, 10 and 12 layers.

Figure S1. Surface energy versus the number of layers in the slab calculated for fully relaxed slabs.

We have reported[a] the calculated band gap for the surface model of $TiO_2(110)$ as 1.35 eV, which is in agreement with other theoretical studies[b,c] but far from the experimental data (3.05 eV). It is known that GGA doesn't do a good job in describing the electronic properties of defective TiO_2 surfaces[d]. Therefore we have calculated the relaxed cell parameters for the bulk $TiO_2(110)$ again using different approaches (Tabla S1). All the functionals employed in this work provide results which are in agreement with what has been reported for rutile.

[a]X. Zarate, E. Schott, T. Gomez, R. Arratia-Perez, *J. Phys. Chem. A,* 2013, **117**, 430-438.

[b]H. Fox, K. E. Newman, W. F. Schneider, S. A. Corcelli, *J. Chem. Theory Comput.* 2010, **6**, 499–507.

[c]S. M. Baizaee, N. Mousavi, *Physica B* 2009, **404**, 2111–2116.

[d]E. Dagotto, *Rev. Mod. Phys.* 1994, **66**, 763−840.

The band gap value using GGA-type functionals lead to a underestimation which has been well documented in the literature, while hybrid functionals with exception of AM05 and PBE0, overestimate the band gap, in particular for the functional HSE06, is generally more accurate than GGA, while PBEsol+U performs slightly best than the other DFT schemes. In the present work, it appears that the PBE+U method and HSE06 provides a good compromise between accuracy and computational effort.

The U value has been chosen by comparing to experiment but constraining U(3d) and U(2p) to be the same, to avoid an excessive parametrization. In the case of rutile we varied U between 2 and 7 eV, and the band gap predicted varies between 1.34 and 2.09 eV.

Table S1. Relaxed cell parameters of rutile: Cell volume (Vol in Å^3), optimized lattice parameters (a and c) (in Å), bulk moduli B_0 (in GPa) and energy gaps (Gap in eV).

	Vol (Å^3)	a(Å)	c(Å)	B_0(GPa)	B_0'	Gap (eV)
AM05	62.9021	4.6053	2.9427	255.5592	4.4822	1.66
B3LYP	59.6675	4.5250	2.9340	341.1308	3.9750	2.88
HSE06	62.4149	4.5934	2.9523	276.9766	4.3034	3.13
PBE	59.6639	4.5249	2.9850	332.3897	4.0411	1.74
PBE0	59.6620	4.5248	2.9458	334.1002	4.0299	1.98
PBEsol	62.7564	4.6018	2.9552	259.1475	4.4671	1.66
PBEsol+U	63.1322	4.6109	2.9598	256.6894	4.5037	1.82
Exp		4.6020	2.9560	210-260		3.05

Optimized coordinates for TPP-Zn(II) on TiO_2(110) model (Figure 3 a, b and c)

Figure 3 (a and b)

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Figure 3 c

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