

Dye-electrode interfacial electronic features in p-type photoelectrochemical cells: a first-principles perspective

Michele Pavone and Ana B. Muñoz-García

Department of Chemical Sciences, University of Naples Federico II

DATE / TIME: Friday, October 16th 2015, 09:00 a.m.

LOCATION: Erwin Schrödinger Institute, Boltzmanngasse 9, 1090 Vienna

Recent interests in photo-electrochemical catalysis of water splitting have boosted studies on p-type dye-sensitized photo-cathodes for the hydrogen evolution reaction [1]. In these devices, the sunlight harvesting is carried out by p-type dye-sensitized solar cells (p-DSSCs), which are the complementary photocathodes to well-studied n-type DSSCs (Grätzel cells) [2], and are mostly built on nickel oxide (NiO) electrodes [3]. Their low performances, however, have hindered the development of convenient tandem solar cells based on coupled cost-effective n- and p-type DSSCs [3]. Experimental investigations have demonstrated that electronic processes at the dye-electrode interface are responsible for such low p-DSSC efficiencies [4]. Indeed, interfacial properties have been shown to be highly dependent on the dye anchoring group and binding modes on NiO surfaces [5].

Here, we report state-of-the-art first-principles calculations on the interfaces between NiO (001) surface and two different sensitizers: a prototypical coumarin-based dye (C343) and a recently proposed push-pull dye [6]. From our results, we derive structure-property-function relationships that can help to develop further p-type DSSC photocathode materials with improved performances.

[1] (a) J. M. Gardner, M. Beyler, M. Karnahl, S. Tschierlei, S. Ott and L. Hammarström, *J. Am. Chem. Soc.*, 2012, 134, 19189; (b) W. Hamd, M. Chavarot-Kerlidou, J. Fize, G. Muller, A. Leyris, M. Matheron, E. Courtin, M. Fontecave, C. Sanchez, V. Artero and C. Laberty-Robert, *J. Mater. Chem. A*, 2013, 1, 8217

[2] B. O'Regan and M. Grätzel, *Nature*, 1991, 353, 737

[3] A. Hagfeldt, G. Boschloo, L. Sun, L. Kloo and H. Pettersson, *Chem. Rev.*, 2010, 110, 6595

[4] F. Odobel, L. Le Pleux, Y. Pellegrin and E. Blart, *Acc. Chem. Res.*, 2010, 43, 1063–1071

[5] A. B. Muñoz-García and M. Pavone, *Phys. Chem. Chem. Phys.*, 2015, 17, 12238–12246

[6] J. Warnan, Y. Pellegrin, E. Blart, L. Zhang, A. Brown, L. Hammarström, D. Jacquemin and F. Odobel, *Dyes and Pigments*, 2014, 105, 174