High-throughput Computational Screening of Perovskite Oxides and Related Compounds for Light Harvesting Applications

Dr Ivano Castelli

Theory and Simulation of Materials (THEOS), EPFL National Center for Computational Design and Discovery of Novel Materials (MARVEL), EPFL

Solar energy harvesting in a photoelectrochemical (PEC) cell, where water is split into hydrogen and oxygen by solar light, is an attractive and renewable contribution to our global needs of increasing energy demand and storage.

Here, we explore the possibility of identifying novel photocatalysts for PECs with the use of high-throughput quantum mechanical simulations, screening in particular novel compounds within the class of cubic perovskites.

First, we identify three screening criteria that are necessary for a potential material to be used as a light harvester in a PEC. These are: (i) a narrow band gap, allowing the utilization of a significant fraction of solar spectrum, (ii) well positioned band edges with respect to the red-ox levels of water, and (iii) chemical/structural stability under irradiation (in addition, high electron-hole mobilities, low cost and non-toxicity would all be desirable properties for an efficient and eco-friendly material).

We devised inexpensive approaches to calculate systematically the properties above, and applied this screening procedure to 19000 cubic ABX_3 perovskites, obtained by combining 52 possible metals as A- or B- cations, together with oxygen, nitrogen, sulfur and fluorine as anions. Out of this search, 32 promising materials have been identified for visible-light harvesting, and 20 and 12 for one-photon and two-photon water-splitting processes, respectively [1,2]. 9 of the 14 oxynitrides have already been synthesized and shown activity as PEC devices. The remaining 5 have not been synthesized yet, and could have additional potential as light harvesters.

Last, the problem of corrosion has been addressed for all the candidates for one-photon water splitting, determining their Pourbaix diagrams through a combination of experimental and computational data [3].

References

[1] I. E. Castelli, T. Olsen, S. Datta, D. D. Landis, S. Dahl, K. S. Thygesen, and K. W. Jacobsen, Energy Environ. Sci. 5, 5814 (2012).

[2] I. E. Castelli, D. D. Landis, K. S. Thygesen, S. Dahl, I. Chorkendorff, T. F. Jaramillo, and K. W. Jacobsen, Energy Environ. Sci. 5, 9034 (2012).

[3] I. E. Castelli, K. S. Thygesen, and K. W. Jacobsen, Topics in Catalysis 57, 265 (2014).