Key Ingredients for the Screening of Single Atom Catalysts for the Hydrogen Evolution Reaction: The Case of Titanium Nitride

Clara Saetta, Ilaria Barlocco, Giovanni Di Liberto, Gianfranco Pacchioni

Dipartimento di Scienza dei Materiali, Università degli Studi di Milano Bicocca, Milano, Italy
c.saetta@campus.unimib.it, ilaria.barlocco@unimib.it, giovanni.diliberto@unimib.it, gianfranco.pacchioni@unimib.it

Single Atom Catalysts (SACs) are considered to be the new frontier in the field, since they establish a bridge between the homogeneous and heterogeneous catalysis. Computational chemistry shows the atomistic details of electrocatalytic processes, and it is extremely helpful to rationalize of even predict systems'properties. A lot of attention has been dedicated to the reactions of evolution and conversion of molecular hydrogen and oxygen from or to liquid water.[1] The Computational Hydrogen Electrode (CHE) approach is the key to predict the activity of SACs, where the free energy of key intermediates adsorbed is used to explain the catalytic activity[2], or even predict the reaction mechanism.

In this presentation we discuss a computational screening of Single Atom Catalysts (SACs) bound to titanium nitride (TiN), an emerging supporting matrix. The catalysts were tested against the Hydrogen Evolution Reaction (HER), based on density functional theory (DFT). We explore the role of fundamental ingredients to consider for a reliable screening of SACs. Namely, the formation of H_2-complexes[3] besides the classical H* one has an important impact on the predicted HER activity. Also, the results indicate that one needs to adopt self-interaction corrected functionals[4], including the Hubbard parameter on d electrons. Once unconventional intermediates in a self-interaction corrected scheme, the number of potential good catalysts for HER is strongly reduced, since the formation of unconventional intermediates lead to thermodynamic barriers. This study highlights the importance of including the key ingredients for the prediction of new systems, as the formation of unconventional intermediates and the adoption of self-interaction corrected functionals. Also, this study highlights some interesting candidate deserving more dedicate work.

References

1. L. Cao, Q. Luo, W. Liu, Y. Lin, X. Liu, Y. Cao, W. Zhang, Y. Wu, J. Yang, T. Yao, S. Wei, Nat. Catal. 2 (2019) 134.
2. J. K. Nørskov, T. Bligaard, A. Logadottir, J. R. Kitchin, J. G. Chen, S. Pandelov, U. Stimming, J. Electrochem. Soc. 152 (2005), J23.
3. G. J. Kubas, Chem. Rev. 107 (2007), 4152; R. H. Crabtree, Chem. Rev. 116 (2016), 8750.
4. I. Barlocco, L.A. Cipriano, G. Di Liberto, G. Pacchioni. Adv. Theo. & Simul. 6, 2200513 (2023)