Multiscale Modelling of Heterogeneous Functional Materials for Energy Conversion and Storage: Challenges and Perspectives

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The current high demand for sustainable energy solutions has motivated intense research for advanced and efficient energy conversion and storage technologies, including batteries, fuel cells and electrolyzers, solar cells and photo-electrochemical devices. Despite significant progresses, fundamental challenges remain, particularly for the optimization of the several different materials that operate in these devices. The usual design strategies based on trial-and-error are time consuming, costly and not very effective. A new rational design paradigm is needed, and it must rely on a solid understanding of the subtle physico-chemical processes that determine the device stability and performance.

Computational modelling provides a powerful tool to characterize and predict these processes for functional materials and heterogeneous interfaces in energy conversion devices. These conversion devices rely on electrochemical processes that involve key electrocatalytic events plus charge and mass transport across several materials' layers. For these reasons, an effective modelling approach should be based on first-principles methods that account for the quantum mechanical nature of interacting electrons and nuclei. However, conventional first-principles methods often fall short in capturing the realistic operating conditions of these devices. A major challenge lies in the absence of a unified framework capable of simultaneously addressing the diverse factors influencing interfacial reactivity, including electronic correlations, complex chemical environments, and applied electric bias.

To bridge this gap, novel multiscale modelling strategies are required, integrating quantum mechanical accuracy with larger-scale methodologies. In this contribution, we discuss few case studies that illustrate the current challenges in modelling electrochemical interfaces and explore advanced ab initio approaches that incorporate key external variables. We focus on the role of electronic correlations, electrolyte dybamics and chemical reactivity for extended materials and complex interfaces [1,2,3,4]. These findings emphasize the need for a bottom-up multiscale modeling paradigm, bridging different scales in length and time. Such an approach will enable not only the predictive design of novel energy materials but also the development of computational frameworks applicable beyond energy conversion, impacting diverse fields of materials science and catalysis.

References

[1] Massaro, A.; Muñoz-García, A. B.; Prosini, P. P.; Gerbaldi, C.; Pavone, M. *ACS Energy Lett.* **2021**, *6*, 2470.
[2] Fasulo, F.; Massaro, A.; Muñoz-García, A.B.; Pavone, M. *J. Chem. Theory Comput.* **2023**, *19*, 5210.
[3] Fasulo, F.; Piccini, G.M.; Muñoz-García, A. B.; Pavone, M.; Parrinello , M. *J. Phys. Chem. C* **2022**, *126*, 15752.
[4] Fasulo, F.; Muñoz-García, A. B.; Massaro, A.; Crescenzi, O; Huang, C.; Pavone, M. *J. Mater. Chem. A* **2023**, *11*, 5660.