Can Quantum Chemistry Help to Understand Electrochemistry?

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Electrochemical reactions in batteries are complex processes characterized by significant changes in electronic configuration and ions movement. This complexity often results in modeling with simplified and less accurate macroscopic models. Solid-state batteries, with their complex chemical composition and porous structure featuring multiple channels, present significant challenges for accurate modeling.

Modern quantum chemistry has the capability to solve a wide range of problems, including the modeling of solids and liquids. However, in the context of battery processes, the field often resorts to simplistic models for the material and approximate Hamiltonians [1-2]. Such approaches overlook the critical role of environmental effects in accurately describing electronic structure and identifying potential pathways for ionic transfer.

We have recently developed a novel procedure for representing a crystalline environment through an embedding method [3-4]. This technique utilizes ab initio model potentials, optimized for specific crystal structures, in combination with the electrostatic potential. The quality of the embedding can be validated by comparing the electron density of a larger cluster or a periodic crystal. The cluster model enables the use of the most precise quantum chemistry methods, including multiconfigurational techniques [5], which are crucial for capturing the dynamic changes in the electronic configuration of ions during battery charging and discharging cycles.

This presentation will introduce our protocol, designed for modeling materials commonly used as solid-state electrolytes, and will share some preliminary results that underscore the potential of our method.

References

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