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Large-Scale DFT Calculations of Electrochemical Systems

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Abstract:

Recent significant progress of massively parallel computational technology and linear-scaling (O(N)) DFT calculation methods have realized large-scale first-principles calculations. Such a progress has being opening the possibility of simulating various kinds of complicated systems.

We have developed a scheme of O(N) first-principles molecular dynamics (FPMD) calculation applicable to simulations and analyses on molecular-level structures of electrode-electrolyte interfaces and electrochemical reactions, which has been done based on the DFT calculation code, OpenMX[1]. The following developed items (a-c) and those applications on electrode-electrolyte interfaces of Li-ion battery systems (d-e) will be presented;

a. Hybrid scheme of O(N)-Krylov subspace method and effective screening medium (ESM) method[2,3]
b. Localized-orbital analysis method for large-scale FPMD simulations[4]
c. Introduction of bluemoon-ensemble scheme into OpenMX
d. Analysis on a mechanism of reduction decomposition reactions of organic electrolyte molecules[5]
e. Free-energy analysis on a Li^+ desolvation process under imposed bias[6]

References

[1] http://www.openmx-square.org/
[2] M. Otani and O. Sugino, Phys. Rev. B, 73, 115407 (2006).
[3] T. Ohwaki, M. Otani, T.Ikeshoji, and T. Ozaki, J. Chem. Phys. 136, 134101 (2012).
[4] T. Ohwaki, M. Otani, and T. Ozaki, J. Chem. Phys. 140, 244105 (2014).
[5][6] prepared to be submitted.