Reducing Systematic Errors in Oxide Species with Density Functional Theory Calculations

Density functional theory calculations can be used to gain valuable insight into the fundamental reaction processes in metal–oxygen systems, e.g., metal–oxygen batteries. Here, the ability of a range of different exchange-correlation functionals to reproduce experimental enthalpies of formation for different types of alkali and alkaline earth metal oxide species has been examined. Most examined functionals result in significant overestimation of the stability of superoxide species compared to peroxides and monoxides, which can result in erroneous prediction of reaction pathways. We show that if metal chlorides are used as reference structures instead of metals, the systematic errors are significantly reduced and functional variations decreased. Using a metal chloride reference, where the metal atoms are in the same oxidation state as in the oxide species, will provide a computationally inexpensive and robust approach to significantly improve accuracy. The approach can potentially be applied to improve accuracy of calculations more generally.

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