SCC-DFTB simulations of ceria surfaces and nanoparticles

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First principles modelling, using e.g. the density functional theory (DFT), has become a valuable tool in materials research. However, today's computer resources limit the size and time scales that can be studied with such techniques, thereby hindering the full utilization of computational chemistry for large-scale systems in practice. Thus, new developments of reliable approximate and/or parameterized methods are needed.

One promising approximate method, conceptually similar to the DFT, is the self-consistent charge density functional based tight binding method (SCC-DFTB). SCC-DFTB calculations are parameterized against DFT data (see illustration in Figure 1) and are at least two orders of magnitude faster than a standard semi-local DFT calculation. However, to obtain an accuracy comparable to DFT for complex oxides is a task that has proven to be a challenge.

In this talk, I will present our SCC-DFTB parameterization effort for the technologically important reducible oxide CeO_2.[1] I will discuss the strategy we have developed for the parameterization and the special complication that follows with reducible oxides. Furthermore, I will demonstrate the applicability of the generated parameters and show results from validation by comparing to data obtained from DFT calculations for CeO_2. I will show results for oxygen vacancy formation in various ceria structures of different dimensionality, ranging from 0D (nano) to 3D (bulk) and for oxygen adsorption on ceria nanoparticles and preliminary results regarding ceria nanoparticle agglomeration.

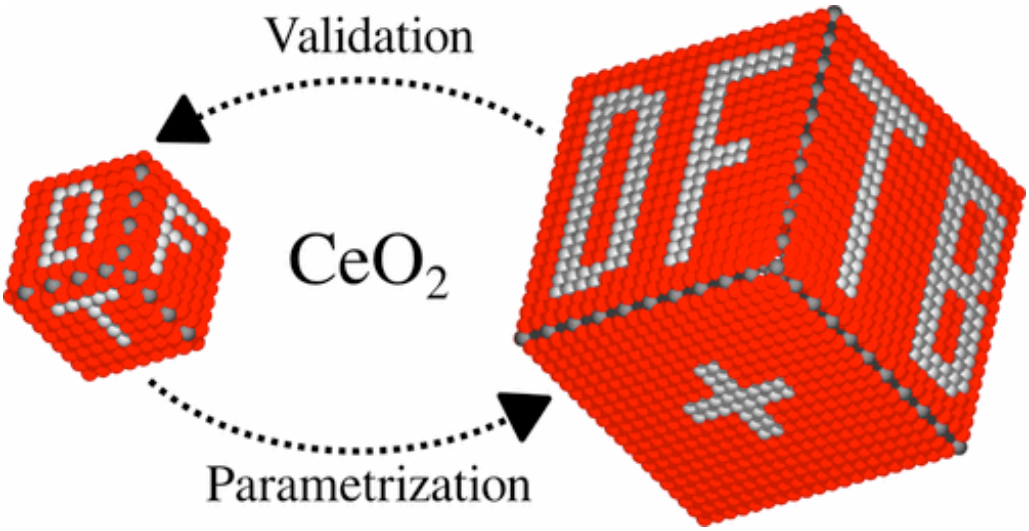

Figure 1. The SCC-DFTB method is parameterized from DFT data.

References

[1] J. Kullgren, M. J. Wolf, K. Hermansson, C. Köhler, B. Aradi, T. Frauenheim, and P. Broqvist, Journal of Physical Chemistry C, **121** 4593 (2017).