Optuna Package in Reactive Force-Field Optimization: the Li/O Case Study

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Reactive force fields are used in molecular dynamics to model reactivity of large structures with reasonable accuracy. Development of the force field comes down to finding the set of parameters for modeling structures consisting of specific atom species. Reactive force field such as the used in ReaxFF package involves optimization of considerable number of parameters, making the optimization process lengthy and laborious, even for the systems of just two atom species. The standard protocol implemented offers single-parameter parabolic minimization, a process which is robust, although does not account for possible parameter dependencies [1]. Here we use a novel *Python* set of routines implemented in the *Optuna* package to gain more control and speed up the optimization. Force-field for lithium and its oxide species, of interest in modeling of lithium-air batteries, has been developed and has been used with considerable success [2]. This force-field, however, is based on electronegativity equilibration method (EEM) used to model atomic charges. A novel method that resolves two major theoretical problems of the previous EEM approach is based on Kohn–Sham density functional theory approximated to the second order (ACKS2) for modeling the charge distribution. This method for Li/O has recently been developed although some shortcomings of the optimized force field are observed [3]. Here we develop protocol for the use of *Optuna* capabilities in the optimization of the ReaxFF force field, and apply it in development of the improved ACKS2 reactive force field for Li/O systems. We validate the force field by the quality of reproduction of the (crystal) structures and DFT charges.

References:

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