Improving the Physics and Parametrization of ReaxFF: Lessons from the Past and Insights for the Future

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ReaxFF is one of the most popular models for reactive molecular dynamics simulations. [1] For heterogeneous and condensed-phase systems where many chemical events occur simultaneously, reactive force fields are attractive compared to periodic Density Functional Theory (lower cost) or QM/MM (many events). In many respects, machine learning potentials (MLPs) are competing on the same selling points, and it can be expected that ReaxFF will eventually become obsolete as MLPs continue to advance. An overview of our improvements to ReaxFF and the protocols to parametrize it will be presented. Our first contribution was to introduce non-metallic screening in the electrostatic interactions, by replacing electronegativity equalization with atom-condensed Kohn-Sham DFT. [1,2,3] In collaboration with the company Software for Chemistry and Materials, ParAMS was developed to facilitate the training process, including an extensible format for archiving and exchanging training data. [4,5,6] Recently, a new loss function has been developed, BalancedLoss, to manage expectations about model performance and to compensate for data imbalance. [7] The utility of these developments goes beyond the scope of ReaxFF. In addition, these works have revealed strengths and weaknesses of ReaxFF, which may inspire the development of future machine learning potentials.

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