Reactive Force-Field Parameter Optimization for Fe/P/C/O/H Systems

Vjeran Gomzi,[a] **Iva Movre Šapić**[b] **and Andrej Vidak**[b]

vjeran.gomzi@fer.unizg.hr

[a]*University of Zagrebu Faculty of Electrical Engineering and Computing, Unska 3, Zagreb*
[b]*University of Zagreb Faculty of Chemical Engineering and Technology,*
Trg Marka Marulića 19, Zagreb

Mesoscale modeling of processes in $LiFePO_4$ batteries should ideally involve molecular dynamics methods available to describe bond rearrangement and preferably also electron transfer reactions. Reactive force field approach as implemented in *ReaxFF* code has potential for satisfying both the above requirements [1]. In a recent work, we developed Li/O *ReaxFF* force field and implemented the optimization protocol relying on the *Optuna Python* package [2]. In this work we proceed to use this protocol to develop reactive force field (ie. optimize parameters) for systems containing Fe/P/C/O/H atoms. The extensive training set of more than 400 structures is used, of which about 10% is selected for validation and excluded from parameter training. A number of these is evaluated using *Gaussian 16* commercial code at the density-functional (DFT) level of theory thus yielding charges and energies for additional training. We validate the force field by the quality of reproduction of the (crystal) structures on a set of 30 crystal structures and 21 molecules including DFT calculated energies and charge distributions for comparison. Finally, a validation is also performed comparing equation-of-state like energy-volume diagrams of six simple iron-phosphorus crystals. Based on these results, the performance of the developed force field, its quality, behavior and stability are evaluated. This is to the best of the authors' knowledge the first Fe/P *ReaxFF* force field available.

References:

[1] T. P. Senftle, S. Hong, M. M. Islam, S. B. Kylasa, Y. Zheng, Y. K. Shin, C. Junkermeier, R. Engel-Herbert, M. J. Janik, H. M. Aktulga, T. Verstraelen, A. Grama, A. C. T. van Duin, *npj Comput. Mater.* **2** (2016) 15011.

[2] V. Gomzi and J. Juvančić, *J. Phys. Chem. A* **129** (2025) 10003–10013.