Equilibrium/nonequilibrium electrochemistry of disordered solid-state interfaces

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Simulation of solid-state electrochemical systems from first principles is challenging due to several factors including control of the chemical potential and handling of the combinatorial explosion in the possible number of ion configurations. To tackle these issues, we coded several new features into our original abICS framework for combination of high-throughput ab initio calculations (VASP, Quantum Espresso, or OpenMX), machine learning surrogate model training (aenet, nequip/allegro, MLIP3), and ensemble sampling [1].

To speed up the sampling for large-scale many-component systems, we implemented the population annealing Monte Carlo method for massively parallel ensemble sampling. We were also careful to cut back on file IO to minimize the impact on the network file system.

We also enabled calculation and control of the chemical potential through the implementation of free energy integration and grand canonical Monte Carlo sampling at fixed chemical potential. Figure 1 shows the state of charge in $LiCoO_2$, an active material for Li-ion batteries, as a function of the Li chemical potential, which we calculated as a first test using the newly implemented grand canonical sampling. The results are in good general agreement with experiment. Further calculations on metal/solid electrolyte interfaces and electrocatalyst surfaces [2] are under way.
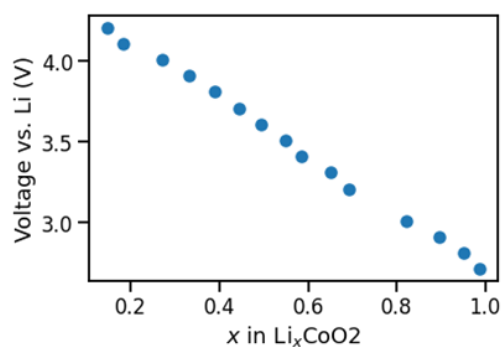

Fig. 1: Calculated state of charge vs. Li potential.

References

[1] S. Kasamatsu et al., Sci. Technol. Adv. Mater. Meth. 3, 2284128 (2023)

[2] A. Nakanishi et al., in preparation. arXiv:2307.11296