Foundational MLIP: the Li-ion Battery

Ioan-Bogdan Magdău[1], I Batatia[2], P Benner[3], Y Chiang[4,5], AM Elena[6], DP Kovács[2], J Riebesel[2,4] ... & G Csányi[2].

[1]Newcastle University, NE1 7RU, UK
[2]University of Cambridge, CB2 1TN, UK
[3]BAM, D-12205, Germany
[4]University of California Berkeley, CA 94720, USA
[5]LBNL, CA 94720, USA
[6]STFC, WA4 4AD, UK

Ioan.Magdau@newcastle.ac.uk

The atomistic processes underpinning ion transport as well as the chemistry of the interface determine the performance of electrochemical devices such as batteries and fuel cells. Ideally, we would like to understand these mechanisms from first principles, however the time and length scales on which these processes play out are prohibitively expensive for ab initio methods.

The emergence of Machine Learning interatomic potentials (MLIPs) in the past decade, has made it possible to significantly extend the scales of molecular dynamics (MD) simulations to thousands of atoms and hundreds of nanoseconds while preserving the ab initio accuracy. In practice, however, training MLIPs can be a tedious task [1] plagued by many challenges: how to choose representative training data? what is the right model architecture? which hypermeters need tunning? etc. Recent advances in our MLIP understanding [2] have led to the development of Foundational ML (FML) models [3,4] which are trained on large datasets and show a great deal of out-of-the-box transferability across chemical systems and simulation conditions.

Here we demonstrate the performance of MACE-MP-0 [3] (a foundational model trained on the Materials Project database [5]) in simulating a full Li-ion battery system: Cu | H-capped graphite+Li | EC/EMC+$LiPF_6$ | NMC+Li. Previous work [1] has shown that modelling even the neat solvent can be a challenge to MLIPs owing to the weak, but crucially important, inter-molecular interactions. The out-of-the-box MACE-MP-0 (without any retraining) generates stable MD trajectories for the neat electrolyte, with both density and diffusivity reproducing the expected results. The full battery environment was simulated in the NVT ensemble to suppress issues arising from unphysical strains at the interfaces. All trajectories were stable at 500 K for the entire length of the simulation (100-200 ps). The Li-ions were found to spontaneously de-intercalate from both the graphite anode and NMC cathode and the entire electrolyte remained fluid. The H-capped graphite was found to be inert, whereas the cathode-electrolyte interface exhibited pronounced reactivity. Evident from the start of the simulation was the extensive proton transfer from the carbonate solvent (EMC in particular) to the oxygen atoms in NMC. This in turn led to continuous breakdown of solvent molecule (which became radicals), and chemisorption onto the cathode surface. Notably, substantial amounts of CO_2 and H_2O were generated in the process, and oxygen atoms were easily extracted from the cathode leaving behind binding sites for the oxygen-rich carbonate molecules.

These early simulations demonstrate MACE-MP-0 robustness in modelling a complex battery environment comprising four different materials, four interfaces and nine distinct chemical species in a single simulation. These tests also showcase the initial steps in modelling the SEI formation with ab initio accuracy – which has been a long-held dream of the scientific community.

References

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Figures

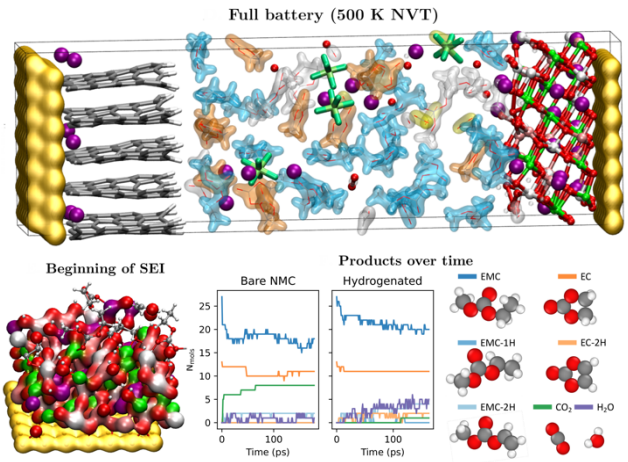

Figure 1. Full Li-ion battery simulation (Cu | H-capped graphite+Li | EC/EMC+$LiPF_6$ | NMC+Li) with MACE-MP-0