Advanced Materials for the Energy Transition

Machine Learning Interatomic Potentials for Li-ion Diffusion in Batteries

In this practical, we have worked through the full pipeline of training machine-learned interatomic potentials (MLIPs) on Density Functional Theory (DFT) data, to then run fast molecular dynamics (MD) simulations to study ion transport in a solid-state electrolyte:

1. **Explored** a dataset of DFT calculations for a lithium thiophosphate (Li-P-S) system.
2. **Trained** a MLIP on the DFT data, and examined/**validated** its accuracy.
3. **Ran** accelerated MD simulations with the trained ML potential (orders of magnitude faster than would've been possible with DFT, without a big supercomputer).
4. **Extracted** the Li-ion diffusion coefficient from the MD trajectory.

Solid-state electrolytes are of great interest for next-generation lithium batteries with no liquid electrolyte components, offering improved safety and energy density. Understanding Li-ion transport at the atomic scale requires long molecular dynamics simulations that are often too expensive with DFT-based MD (AIMD) alone. MLIPs bridge this gap: we train a ML model on a relatively small set of DFT data, which we can afford to compute, then use this ML potential to run simulations orders of magnitude faster — at near-DFT accuracy.

The same simulation procedure can be applied to analyse ionic transport in other solid electrolytes or electrode materials, to analyse phase transitions and structural dynamics in other materials systems (e.g. molecular rotations in perovskites or melting of organic crystals as we saw in lectures), thermal conductivity etc. Here we have seen that Li-ion diffusion is indeed very fast in this system, corroborating the fact that it is a promising candidate for a solid-state electrolyte for advanced Li-ion batteries!

The assessment of this practical comprises 3 equally-weighted components:

1. Follow-on simulation task: Determining D_0 and E_a in the Arrhenius relationship $D(T) = D_0\exp(=E_a/k_BT)$ for the $LI_{27}P_{12}S_{44}$ system, and generating 'learning curves' for our MLIP – following Sections 5 and 6 in the Extensions notebook.
    a. You can similarly run this notebook in Google Colab, and save your progress by downloading the .ipynb file with File > Download > Download .ipynb.

   b. This task will involve multiple MD simulations / model training runs. You should try to avoid overwriting notebook cells or data when doing this, so starting each new MD simulation in a new notebook cell, to retain the full track record of your computational experiments.
   c. It is recommended to download your trained models and MD trajectories to your local PC as you progress through this follow-on task, to avoid any lost progress from restarting the Colab session (e.g. if you complete this task over multiple sessions).
2. Discussion questions: In the Extensions notebook with these follow-on simulation tasks, there are bullet-point questions include. Please answer these questions concisely as you progress through this simulation task. You can double click on the cell to edit the text.
   a. When submitting the notebook with the simulation task and discussion questions, **please provide both the notebook file (.ipynb) and the notebook PDF**, which you can get by doing File > Print > Save as PDF.
3. The final component is a written report on this practical, describing the work completed during the practical session *and* the follow-on simulation task. Some guidance on this is included below, following a similar format to other practical reports.

Note: The content you will cover in your lectures for the final part of the course, with Dr Shijing Sun, will be very useful for understanding the architecture of the MLIPs we have been using in this practical, how we can expect their accuracy to change with different model settings (hyper-parameters) and more.

Practical Report Guidance

The write up component of this practical should be concise, with a maximum of ***1500 words*** excluding figures and references. The report should include:

- an abstract,
- an introduction,
- a computational methods section,
- results and discussion section,
- conclusion.

The write up should focus on what you have learnt about the solid-state electrolytes and how to model their ionic transport properties with (MLIP-accelerated) MD during the practical and follow-on task. Advice on writing practical reports can be found in the "**Keeping Notes and Writing Up Experimental Work**" document on Moodle. You do not need to reproduce detailed information from the simulation notebook, which may be referred to in your write up.