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Deep learning force filed for lithium thiophosphates solid state electrolytes

All-solid state (ASSB) batteries are one of the most promising alternatives of Li-ion liquid technology. ASSB have several advantages1, such as greater stability, better safety, and higher energy density2, but also an intrinsic, and still unsolved, problem caused by their solid nature, the low ion mobility. Amorphous Lithium thiophosphates, LPS (xLi2S-(1-x)P2S5) are among the most promising3 electrolytes. Optimizing their performance by tuning their composition in order to achieve high ion conductivities (at least 10-3 S/cm to be competitive with liquid electrolytes) is an extremely complicated task because we do not yet know the structure-properties relationship and the optimization process requires laborious trial-and-error laboratory procedures. Using molecular modelling we can help make this process much faster. Typically, computational approaches to electrochemistry involve ab-initio molecular dynamics (AIMD) methods, which despite their accuracy, are very slow and computationally intensive, compared to classical molecular dynamics (MD), that instead relies on parametric force fields (FF). Classical MD calculations on LPS are, at the moment, difficult to perform because they would need the development of a suitable FF whose analytical form is complicated because of the different states of chemical coordination that Li+ assumes in the amorphous solid. Nevertheless, it is important for the electrochemical community to have access to a quick and efficient way to model electrolytes such as LPS. To solve these issues, we have used deep learning molecular dynamics4, and generated a new deep learning force field (DLFF)7-8. The deep neural network5 has been trained using our ab-initio data, this way the DLFF has the high accuracy6 of the ab-initio level chosen for the training set and, being parametric, a high efficiency (classical MD level). Using this DLFF, we have performed fast ed accurate simulations on model systems beyond the actual limits of AIMD in size and time scales. This way we determined the electrochemical properties of different compositions of LPS in a quick and accurate way, rarely reached before. This approach will be crucial to determine electrochemical properties of LPS oxides (LPSO) and optimize their composition as well, making this technology competitive in a large-scale industrial environment.

[1] Sun, Y.-K., ACS Energy Lett. 2020, 5, 3221–3223. [2] Zhang, Z. et al., Energy Environ. Sci. 2018, 11, 1945–1976 [3] Grady, Z. A. et al., Front. Energy Res. 2020, 8. [4] Unke, O. T. et al., Chem. Rev. 2021, 12, 10142–10186. [5] Mo, Y. et al., Chem. Mater. 2012, 24, 15–17. [6] He, X. et al., npj Comput Mater 2018, 4, 1–9. [7] Wang, H. et al., Comp Phys. Comm. 2018, 228, 178–184. [8] Zhang, L. et al., Phys. Rev. Lett. 2018, 120, 143001.

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