Accelerated Discovery of Cathode Materials for Li-ion Batteries using General-Purpose Neural Network Potentials

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With the rapid development of mobile technology and electric vehicles, research and improvement of Li-ion batteries (LIBs) have recently garnered significant attention from engineers and scientists. A modern Li-ion battery consists of two electrodes separated by a non-aqueous liquid electrolyte. Typically, the anode is made of graphite, while the cathode is either a layered oxide or spinel. During charging and discharging, Li ions move between the electrodes. Despite their high energy density, great power, and long-life cycle, Li-ion batteries still face operational challenges such as electrode degradation. The cathode, in particular, encounters significant issues due to the insertion and removal of Li ions, leading to phase transformations, surface reconstruction, and electrolyte decomposition in contact with the cathode. Consequently, discovering new cathode materials is essential. Traditionally, materials discovery often consumes a vast amount of time and resources in experiments and computational modelling/simulation because of the limitations of experimental conditions or theoretical foundations. Thus, it is crucial to approach this task in a new way. In recent years, the availability of large experimental and simulation datasets combined with advanced algorithms and computational resources has heightened interest in data-driven and machine learning methods for materials discovery, achieving notable improvements in both time efficiency and prediction accuracy. One promising approach is the use of neural network potentials (NNPs), which model atomic interactions by mimicking neural networks in the human brain. These potentials retain the accuracy of quantum mechanical calculations while operating at the speed of force field calculations.

In this talk, we will present our current development of a high-throughput screening scheme for exploring multi-component cathode materials in Li-ion batteries using general-purpose NNPs [1], in combination with global optimization techniques. From this scheme, several candidate materials will be revealed. Additionally, we will discuss the effects of mixing entropy in multi-component cathode materials by employing the general-purpose NNP alongside the multi-canonical sampling-based Wang-Landau method [2-4] to study the thermodynamic properties and cation ordering within the transition metal layers of $LiNi_xCo_yMn_{1-x-y}O_2$ (NCM) layered oxides. With the acceleration provided by the general NNP, configurational spaces of several NCM compounds are explored, and configurational density of states are calculated. For each compound, different transition temperatures are identified from the calculation of configurational specific heats, depending on the compositions of metal components. The approaches introduced here not only accelerate the discovery of high-performance cathode materials but also provide insights into the thermodynamic behavior and stability influenced by cation mixing.

References

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