Accurate Prediction of Voltage of Battery Electrode Materials Using Attention-Based Graph Neural Networks

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Abstract

Performing first-principles calculations to discover electrodes' properties in the large chemical space is a challenging task. While machine learning (ML) has been applied to effectively accelerate those discoveries, most of the applied methods ignore the materials' spatial information and only use predefined features: based only on chemical compositions. We propose two attention-based graph convolutional neural network techniques to learn the average voltage of electrodes. Our proposed methods, which combine both atomic composition and atomic coordinates in 3D-space, improve the accuracy in voltage prediction significantly when compared to composition-based ML models. The first model directly learns the chemical reaction of electrodes and metal ions to predict their average voltage, whereas the second model combines electrodes' ML predicted formation energy (E_{form}) to compute their average voltage. Our E_{form}-based model demonstrates improved accuracy in transferability from our subset of learned Li ions to Na ions. Moreover, we predicted the theoretical voltage of 10 Na_xMPO_4F (M = Ti, Cr, Fe, Cu, Mn, Co, and Ni) fluorophosphate battery frameworks, which are unavailable in the Material Project database. It could be shown that we can expect average voltages higher than 3.1 V from those Na battery frameworks except from the $NaTiPO_4F$ and $TiPO_4F$ pair of electrodes, which offer an average voltage of 1.32 V.