Construction of Neural Network Potential to Study Li-Ion Distribution near Au(111)/Li_3PO_4 Interface

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Li_3PO_4 based materials are used for solid electrolytes in thin film Li-ion batteries. Recently, its application as non-volatile memory devices is also being explored: Au/Li_3PO_4/Li as well as Ni/Li_3PO_4/Li stacked structures were found to exhibit a few different voltage states, which can be controlled by applied voltages [1]. To develop novel memory devices using this phenomenon, understanding on Li-ion distribution near the metal/Li_3PO_4 interfaces is crucially important. We had investigated these issues using density functional theory (DFT) based molecular dynamics (MD) calculations. However, their high computational costs hinder analysis of interface systems including an amorphous solid electrolyte. Considering this situation, we constructed the interatomic potentials for the Au/Li_3PO_4 interface system with adopting the framework of the high-dimensional neural network potential (NNP) [2, 3], which is expected to acheive high reliability and low computational costs simultaneously.

We constructed the four-elements NNP using the dataset obtained by DFT calculations, which includes bulk and surface structures of Au and Li_3PO_4, Au surfaces with Li adatoms, and Au/Li_3PO_4 interface. We created 236,322 structures in total, and these structures were randomly divided into the training (80%) and test (20%) sets. The root-mean-square-errors of the predicted total energies were 41.3 meV/atom and 41.4 meV/atom for the training and test sets, respectively. Next, we constructed the NNP using a little different procedure to explore the possibility of acceralating the construction process. First, we constructed two separate NNPs optimized for Au and Li_3PO_4, respectively. We then constructed the NNP for the Au(111)/Li_3PO_4 system starting from the above two NNPs and adding the interactions of Au-Li, -P, and -O. We found that the NNPs obtained from the two procedures show comparable accuracy, while the second approach required less computational time for NNP optimization. This work was supported by CREST, JST.

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