Accurate phase transitions and ion conductivity in $Li_2B_{12}H_{12}$: MD study within machine learning interatomic potentials

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Accurate prediction of phase transitions, diffusion coefficients and ion conductivity is essential for the design of solid electrolytes derived from known metal borohydrides. MD with the use of machine learning interatomic potentials (MLIP) allow to simulate materials in a large time and space scale, combining accuracy of quantum-chemical calculations and efficiency of classical (empirical) molecular dynamics. MLIP based on different DFT data were built for the $Li_2B_{12}H_{12}$ solid electrolyte for the first time and using constructed MLIP's phase transition and ion conductivity were simulated. It was shown that rev-vdW-DF2 functional is the best in terms of phonon spectrum and lattice parameters, and rev-vdW-DF2-based MLIP is the best for phase transitions, lattice parameters and ion conductivity. MD simulations predict: 1) phase transition temperature of the system to be in excellent agreements with the experiment with deviation less than 50 K, and 2) lattice parameters with errors less than 0.05 at over the temperature range from 300 to 900 K. Ion conductivity of vacancy rich systems are also in a good agreement with the experimental data, as well as activation barriers of diffusion. The importance of the reorientational motion of the $[B_{12}H_{12}]^{2-}$ dianion was shown, and in vacancy rich structures a complete rotation was observed. Combined use of DFT with vdW corrections, active learning and MD might be helpful for research of related systems.We acknowledge funding from Russian Science Foundation (grant No. 22-73-00219)