Graph Neural Network Based Parameter Design Method for Liquid, Ice, and Clathrate Hydrate Phase Analysis

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A wide variety of crystal structures are composed of water molecules, and 17 crystal polymorphs have been discovered in ice crystals. In systems containing guests, a crystal structure called a clathrate hydrate is formed, characterized by storing guest molecules in a cage structure. Phase states in aqueous molecular systems are closely related to liquid, ice, and clathrate hydrate. Clathrate hydrates are also an important research target because of their ability to contain gases and liquids, which has led to the investigation of various applications. Molecular simulations are widely used for microscopic analysis, and molecular dynamics and Monte Carlo methods are useful. These simulations provide numerical data such as coordinates, velocities, and forces. The phase analysis from the simulation data conventionally uses a parameter that indicates the phase state, called the order parameter. Although these parameters have been handcrafted for each system, it has not been easy to design optimal parameters manually for liquid, ice, and clathrate hydrate crystalline polymorphs. In our previous study, we proposed a machine learning method that uses graph neural networks as direct input coordinate data obtained from simulations to design optimal parameters for a desired system through a structure classification task without prior processing or domain knowledge. The method is applied to liquid, ice, and clathrate hydrate systems to verify the generality of the parameter design method and to analyze clathrate hydrate nucleation systems using the designed parameters. This study will enable accurate classification of liquid and hydrate phases, detection of nuclei, and detection of crystal structures.