

Title

“Computational Chemical Approaches to the Microscopic Mechanism of Anhydrous Proton Conduction Materials”

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Abstract

Anhydrous proton-conduction materials have been developed for use in high-temperature polymer electrolyte membrane fuel cells. In particular, imidazole (Im)-incorporated composite materials have gained considerable attention. However, the microscopic picture of the proton conduction has not yet been clarified. This is essential information for the design of future anhydrous proton-conduction materials.

In this work, we investigated the proton conduction mechanism in anhydrous imidazolium hydrogen succinate (Im-Suc) and poly(vinylphosphonic acid)-Im (PVPA-*x*Im) composites those are respective organic hydrogen-bond crystal and acid-base polymer composite by using quantum chemical calculations and molecular dynamics (MD) simulations.

In the Im-Suc system, we constructed a potential energy surface by quantum chemical calculations, which related the proton conduction mechanism to the proton transfer (PT) and molecular motions of Im. We found that proton conduction occurs when coupled with PT and librational motion of Im.

In the PVPA-*x*Im system, we investigated the local hydrogen bond structures and dynamics of Im combined with quantum chemical calculations and MD simulations. We discovered that PVPA-*x*Im has three types of hydrogen bond structures, which are related to the rate and molecular motions of Im. We revealed that the proton conduction process consists of (1) PTs between Im and PVPA or Im and (2) Grotthuss-type diffusion with the reorientation of Im molecules.

In summary, our calculations demonstrated that the hydrogen bond structures depended on the anhydrous proton-conduction materials dominates PT and molecular motions, which are key factors for controlling the proton conduction.