DFT-MD study of highly concentrated Li-salt electrolytes for Lithium-ion batteries KEITARO SODEYAMA, National Institute for Materials Science (NIMS)/JST-PRESTO, YOSHITAKA TATEYAMA, National Institute for Materials Science (NIMS) — Li-salt concentration has been recently proposed as an important control parameter of reduction stability of electrolytes and high ion conductivity in Lithium-ion batteries. For example, highly concentrated (HC) Li-FSA salt in acetonitrile (AN) shows strong electrochemical stability against the reductive decomposition, though in low concentration (LC) solution AN is easily reduced and decomposed. In this study, we investigated the mechanism of the improvement of the reduction stability and Li-ion diffusion mechanism depending on the salt concentration by using DFT-MD simulations. We also calculated the diffusion coefficients of the Li-ions, anions, and solvents in the LC and HC electrolytes to elucidate how Li-ion diffusion was affected by concentration. For the reduction stability, we found that TFSA anion sacrificially accepts reductive electron and decomposed in the HC systems, because specific chained network structure is formed and the electron affinity of the anion shifts lower. For the diffusion mechanism, we analyzed the motions of individual Li ions in HC system, and found Li-ion hopping between the oxygen atoms of the anions. We concluded that change of the diffusion mechanism can be an origin of the high Li-ion conductivity in the HC electrolytes.

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