Theoretical Study of Aqueous Eutectic Lithium-Ion Electrolyte for Wide-Temperature Operation

이경민, 신은혜, 김홍이, 이상영, 곽상규†
울산과학기술원
(skkwak@unist.ac.kr†)

Conventional liquid electrolytes commonly used in rechargeable batteries have narrow-temperature operating range and safety issues. To overcome this weakness, aqueous eutectic electrolyte (AEE), which is high-concentrated (i.e., 5.2 m) LiTFSI aqueous electrolyte, is proposed. In this study, via molecular dynamics simulation and density functional theory calculation, we have investigated good operability of AEE in wide-temperature range and its high performance at low temperature. First, to investigate the cause of wide-temperature operation of AEE, configurations of AEE were compared with dilute aqueous LiTFSI (i.e., 1 m) at the temperature range from 233K to 373K. It was observed that AEE had higher average number of water-$TFSI^-$ hydrogen bond (HB) and lower number of water-water HB, indicating that icing effect of water molecules was inhibited in AEE. Secondly, high performance at low temperature was elucidated by estimating solvation free energy of Li^+ and the interaction energy between LiTFSI and water. It was found that Li^+ desolvation was thermodynamically more favorable in AEE. Thus, in AEE Li transfer at interface is facilitated and high performance is achieved.