Abstract

Sodium-ion batteries (SIBs) offer a cost-effective solution for supercapacitors, electric vehicles, grid storage, and consumer electronics, thanks to the high natural abundance of sodium-ion resources. In any battery system, the electrolyte plays a crucial role in ion transport between the electrodes, maintaining high ionic conductivity, preventing side reactions, and ensuring stable performance. In this regard, in the past, various solvent media, including organic solvents, aqueous solvents, and ionic liquids, have been explored to achieve high stability and electrochemical performance in SIBs comparable to those of lithium-ion batteries (LIBs). In addition, a high-performance battery electrolyte should meet specific criteria, such as high ionic conductivity, a wide electrochemical potential window, a low melting point, and a high boiling point to enable operation across a broad temperature range, and be environmentally friendly and non-toxic. In this regard, novel electrolytes such as ionic liquid solvents can play a pivotal role due to their wide electrochemical window and minimal flammability and volatility risks. However, the high cost of these solvents hampers their commercialization in battery technology. To circumvent the cost issue, organic and aqueous solvent-based sodium electrolytes can be a cheaper option for developing SIBs. But one significant concern with the use of organic solvents is their volatility and flammability at high temperatures. Moreover, aqueous solvents suffer from undesired hydrogen evolution reaction, which are hazardous to electrode materials. To mitigate this issue, high-salt-concentration electrolytes are used, where the activity of water molecules is significantly reduced by dissolving a higher concentration of a sodium salt with a chaotropic anion or by adding ionic liquids, known as hybrid electrolytes. In this thesis, we have thoroughly studied the structural and dynamic properties of sodium-ion electrolytes based on ionic liquids, aqueous solvents, and their mixtures (aqueous and ionic liquid) using classical molecular dynamics simulations. We have also delineated the interfacial structure of ionic liquid-based sodium electrolyte near the graphite electrode under different applied voltages. The sodium salts containing fluorinated anionic groups, such as bis(trifluoromethanesulfonyl)imide ($TFSI^-$), bis(fluorosulfonyl)imide (FSI^-), and fluorosulfonyl(trifluromethylsulfonyl)amide (FTA^-), are examined because of their capacity to boost the formation of a solid electrolyte interface (SEI) and to improve overall stability of the electrolyte.