Heterostructured Electrolyte for High Energy Lithium Ion Batteries

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With the rapid development of smart portable devices, electric vehicles, and energy-storage systems, lithium-ion batteries (LIBs) with high energy density, power density, high safety, and long cycle life have been highly demanded. A viable electrolyte is of primary importance for the high performance Li-ion battery since it controls the operation voltage, rate capability, life spun and safety. Lithium bis(trifluoromethanesulfonyl imide) (LiTFSI) and lithium bisfluorosulfonyl imide (LiFSI) salt based electrolytes are better than LiPF6/EC-based electrolyte in term of ionic conductivity, thermal safety, stability against electrodes at high charge state, though the $LiPF_6$/EC-based electrolytes have dominated the electrolyte market of 4 V-class LIBs over the past 25 years. However, the operation voltage of LIBs using LiTFSI- or LiFSI-based electrolytes have to below 3.7 V vs. Li/Li^+ due to its corrosivity against Al current collector.

It is gradually recognized that solvation plays an important role in determining the electrochemical and chemical behaviours of an electrolyte. The electrochemical window of aqueous solution can expand from 1.23 V to 3.0 V by developing a “water in salt” electrolyte, where almost all H_2O molecules are involved in solvation sheath for Li^+ ions. It is the strong bonding between H_2O and Li^+ that enhances electrochemical oxidation resistance of H_2O. The impact of solvation on electrochemical behaviour of an electrolyte is not limited to tuning oxidation resistance. In recent years, it was reported that concentrated electrolytes using LiTFSI and LiFSI salts also showed superior capability in suppressing corrosion of Al foil, expanding electrochemical stability window, forming stable SEI on lithium metal and graphite anode. Moreover, the cycling performance of Li-S battery can also be greatly improved in the concentrated electrolyte because the dissolution of lithium polysulfides can be effectively suppressed owing to common ion effect. However, using excessive lithium salt also cause high viscosity, low conductivity and high cost. If the unique solvation sheath structure in concentrated electrolyte can be obtained in a dilute electrolyte, both advantages of dilute and concentrated electrolytes can be obtained, which will significantly enhance the electrochemical performance of LIBs.

Here a “pseudo-concentrated electrolyte” combining advantages of both concentrated and diluted electrolytes is described as a brand-new approach for high performance lithium-ion batteries. By designing a heterogeneous liquid structure for electrolyte, we made it possible for Li^+ to form a solvation sheath structure that is only attainable in concentrated electrolytes while using low salt concentration. Such pseudo-concentrated electrolyte, though with Lithium bis(trifluoromethanesulfonyl imide) as lithium salt and carbonates as solvents, demonstrates high electrochemical oxidation resistance, high anti-corrosion capability towards Al current collector that are usually the characteristic of concentrated electrolytes, as well as high ionic conductivity, and low viscosity that are only available from diluted electrolytes. And This innovative approach provides a brand new avenue to tailor-design electrolyte properties for advanced battery chemistry applications.