

Improving the performance of Li/graphite half-cells at low temperatures through the modification of electrolyte

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The automotive industry has increasingly favored lithium-ion batteries (LIBs) with high energy density as a reliable source of power for electric and hybrid electric vehicles. LIBs have become an attractive energy storage option due to their high energy density, minimal self-discharge, negligible memory effect, high open circuit voltage, and long lifespan. Despite their advantages, experts worldwide are working to address potential concerns associated with their use, including their performance under different environmental conditions. One such condition is low temperature, which can cause significant capacity loss and rapid self-discharge in LIB-powered devices [1-3].

Researchers are still debating the main reasons for the drop in performance of lithium-ion batteries (LIBs) at sub-zero temperatures. Several studies have concluded that the ionic conductivity of the electrolyte does not have a significant impact on battery performance in cold temperatures since electrolytes with the same conductivity can exhibit different low-temperature characteristics. Instead, the slow desolvation at the liquid-solid interface, reduced lithium migration in the active material, and the formation of an unfavorable solid electrolyte interface (SEI) layer are believed to be the primary factors contributing to this problem. The high energy barrier of Li⁺-desolvation on the electrode surface causing significant charge-transfer resistance has been identified as the primary cause of reduced discharging capacity when the temperature drops below 0 °C [2, 3].

Graphite has been the primary material used as the anode in LIBs for several decades now due to its high theoretical specific capacity of 372 mAh g⁻¹, which makes it a desirable choice for negative electrodes of LIBs. However, at temperatures below -20°C, the capacity of graphite is only 12% of its room temperature capacity. The low-temperature electrochemical performance of LIBs is mainly hindered by factors such as difficult Li⁺ ion intercalation into graphite, high R_{ct} between the electrode and electrolyte interface, reduced SEI conductivity, and reduced Li⁺ ion diffusivity within graphite sheets.

The electrolyte in LIBs consists of lithium salts and solvents, which play a crucial role in the battery's performance. The

type of solvent, its compatibility with lithium salt, melting point, dielectric constant, and viscosity have a significant impact on the battery's operating temperature. However, currently available commercial carbonate-based electrolytes have high liquid-solid transition temperatures, making them unsuitable for use in cold environments. To address this issue, researchers have attempted to modify the electrolyte's composition using various methods such as enhanced solvent mixtures, new co-solvents, electrolyte additives, new electrolyte salts, and a combined approach to increase the cell's resistance to low temperatures and widen the operating temperature range of the battery. The best results have been achieved through a comprehensive solution that involves the modification of both the salt and solvents in the electrolyte composition.

In this work, the authors aimed to develop an optimal electrolyte system for the Li/graphite half-cell that can operate under low temperatures by applying a complex modification of the lithium salt and solvents. Various concentrations of commercial and alternative lithium salts were mixed with a combination of solvents to identify the optimal interaction with a graphite electrode at room and low temperatures, and the electrochemical and physical properties of cells using the newly designed electrolytes were studied in contrast to commercial electrolyte.

References

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