Interfacial Processes and Properties that Involve Electrolytes in Lithium-ion Cells

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It is generally regarded that many aspects of performance in Li-ion cells are governed by key interfacial regions. Two such regions are the solid-to-electrolyte interphase (SEI) and double layer regions adjacent to SEI, which are present at both the cathode and anode. Being a chemical system, a Li-ion cell and its interfacial regions can experience thermodynamic consequences that impact performance. One example is the possible formation of solid phases or pseudo-phases on the SEI surface at low temperatures due to a Gibbs Adsorption process. Another aspect is that electrolyte properties (viscosity, conductivity, density, etc.) in the double layer region can undergo marked changes under pulse conditions, and may become limiting in a local sense. This work considers how the electrolyte in a Li-ion cell can interact with the neighboring SEI to produce consequences of poor performance, particularly at low temperatures. Both laboratory data and modeling results will demonstrate various aspects of electrolyte behavior, which collectively lead to the conclusion that one or more interfacial thermodynamic processes are largely responsible for the drop in low-temperature performance in some Li-ion cells. Modeling of electrolyte properties is based on an associative form of the Mean Spherical Approximation (AMSA) description of molecular properties and interactions. Effects from ion solvation are explicitly considered as well as ion-ion interactions, such as ion pairs and triples. Complementing the AMSA framework are molecular-based governing equations that express the effect of the molecular environment on various transport properties, which are used within transport calculations that simulate cell performance during a pulse. The model applies equally to aqueous and nonaqueous systems, multi-solvent and dual-salt systems, and has shown impressive accuracy over wide ranges of solvent composition, solute concentration, and temperature. Although the immediate application of the modeling work is characterizing electrolytes for lithium-ion cells, other potential applications abound in industrial and medical processes.