

Innovative Materials for Batteries

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In the struggle for the reduction of carbon emission, the use of energy storage systems is becoming more and more widespread, and the wider applications require a continuous upgrading of the batteries' performances.

The development of innovative materials for the main components of batteries, namely, electrodes and electrolytes, is key to the improvement of battery performance and safety. For different types of batteries, better performing materials allow for the improvement of an already well established chemistry. In this framework, this Special Issue collects several studies that present innovative materials for use in batteries, including their structural, thermal, dynamic, and electrochemical properties [1–7].

Among various energy storage systems, lithium batteries are currently the most efficient and diffused ones, but they present high costs due to use of expensive materials and safety issues. Most of the research ideas reported in this Special Issues focus on the study of new electrodes and electrolyte solutions for Li-ion batteries, aiming at reducing these drawbacks. In particular, one of these studies considers over-stoichiometric Li-rich nickel-manganese-cobalt layered oxides (LRLO) [1], a family of promising positive electrode materials characterized by an overstoichiometric lithium content, thought to improve lower-than-desired capacity and limited lifetimes of currently used cathodic materials. To make these new materials really applicative, a replacement of cobalt in their lattice is mandatory, and this work [1] suggests p-doping as a balanced strategy to remove cobalt from LRLO without massively deteriorating their structural integrity and the electronic properties. The main research efforts to tackle the safety issues address the exploitation of new electrolyte formulations to reduce or eliminate the flammable component of the electrolyte system. In particular, to avoid the use of carbonates, ionic liquids (ILs) are largely investigated as the solvent media or additives, and, in this Special Issue, there are two studies exploring the suitability of a series of ionic liquids with ammonium cations for potential battery applications [2,3]. One of them [2] reports the thermal and transport properties of ammonium-based ILs, focusing on the influence of the symmetrical imide-type anions bis(trifluoromethanesulfonyl)imide ([TFSI][−]) and bis(fluorosulfonyl)imide ([FSI][−]), of the side chain length and functionalization, as well as of the lithium salt content on the properties of the electrolytes. It shows that the ionic liquid electrolytes including [Li][TFSI] are able to successfully cycle in NMC/Li half-cells at elevated temperatures [2]. The other exploits the use of ammonium-based ILs with the asymmetric (fluoromethylsulfonyl)(trifluoromethylsulfonyl)imide (FTFSI)[−] anion [3], and it reports interesting ion transport properties, which could extend the operative temperature range down to low temperatures, as well as remarkable electrochemical stability, which make them appealing for realizing safer and highly reliable lithium battery systems operating at high voltages [3].

Moreover, IL-based electrolytes, in particular electrolytes containing 1-ethyl-3-methylimidazolium-FSI and N-trimethyl-N-butyl-ammonium-FSI, also show promising applicability for Na-ion batteries [4], which are currently studied as an alternative to Li-ion chemistry because of their much larger resource availability and lower cost.

Another approach to lower the electrolytes' reactivity and thus improve their stability and overall safety consists in the use of proper additives. In this framework, a



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symmetrical cell and galvanostatic cycling study carried out on different solvent systems [5] demonstrates that halogen-free closo-borate salts are interesting candidates as lithium salts for Li-ion batteries, with the best results obtained in the preliminary tests with the carbonate-based liquid electrolytes. Additionally, the addition of molecules with varying-sized polar sulfonyl groups attached to an octadecane-tail to an amphiphilic salt lithium trifluoromethanesulfonylimide octadecane (C18LiTFSI) was considered [6]. Indeed, the C18LiTFSI is considered as a possible single-component, ion-condensed electrolyte with a wide layered liquid crystalline phase regime, and the effect of different additives on the conductivity and the applicative temperature range of the obtained blended electrolytes were discussed [6].

Moreover, FSI-based ILs have also been demonstrated [7] as effective additives for inorganic solid electrolytes, insuring ion conduction. In particular, this conclusion is drawn by a paper [7], which reports an evaluation in terms of thermal, spectroscopic, and electrochemical properties of organic–inorganic hybrid electrolytes, based on Al-doped $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) and two different ionic liquids (ILs). This evaluation shows that a battery with favorable performance can be prepared through a simple cold-pressing procedure by using ILs as conductive binders for LLZO.

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