IMLB 2014
The 17th International Meeting on Lithium Batteries June 10-14, 2014 | Como, Italy

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In-Situ gelled Polymer Electrolytes for Advanced Lithium Ion Batteries

Friday, 13 June 2014
Cernobbio Wing (Villa Erba)

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The activities aimed at reducing the safety hazard in lithium batteries have successfully considered the replacement of the conventional, volatile and flammable carbonate-based, liquid electrolytes with more stable polymer electrolytes, obtained in the form of free standing membranes. With respect to other solid polymer electrolytes, such as those based on poly(ethylene oxide), gelled polymer electrolytes (GPEs) have the great advantage of offering high conductivity, i.e. of the order of 10^{-3} S cm^{-1}, over a wide temperature range, combined with a large electrochemical stability, i.e. extending up to 5V versus Li^+/Li [1-3].
Poly(vinylidene fluoride) (PVdF)-based, gel-type membranes have been widely investigated as Li-conducting systems and prepared by conventional solvent-casting procedures [4] or electro-spinning techniques [5].
In this work we report on originally developed, PVdF-based GPEs obtained by *in-situ* formation of the gel matrix during cell assembly, followed by a lithium salt infiltration step [6]. Particular emphasis was devoted to the study and the optimization of the GPE composition. The substitution of the commonly used $LiPF_6$salt with the fluorine free, bis(oxalato)borate (LiBOB) was investigated.
A detailed thermal and dynamical mechanical characterization was adopted to elucidate the properties of the proposed systems over a wide temperature range. Vibrational spectroscopy revealed interesting molecular interactions among components, mainly. polymer and salt, strongly affecting the stability of the membranes. The electrochemical investigation confirmed high ionic conductivity, a controlled interfacial resistivity versus lithium metal and a good electrochemical stability window.
The feasibility of the *in-situ* gelled, PVdF-based polymer electrolyte containing the LiBOB salt was demonstrated by successful galvanostatic cyclations of the quoted membrane in different lithium-ion cell configurations, obtained by coupling a Sn-C alloying anode with an high voltage spinel or a $LiFePO_4$cathode.

Acknowledgments
The results of this work have been obtained by the financial support of the European Community within the Seventh Framework Programme APPLES (Advanced, High Performance, Polymer Lithium Batteries for Electrochemical Storage) Project (contract number 265644).

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