

Safe Gel Polymer Electrolytes for High Voltage Lithium Batteries

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In the growing lithium ion battery (LIB) technology, safety of batteries has become a new concern in addition to the performance. LIBs possess some drawbacks, such as a thermal runaway, due to the presence of flammable organic liquid electrolytes. Consequently, there are strong requirements on the replacement of the flammable electrolyte with non-flammable ones. The use of ionic liquids and polymers as electrolyte components is an attractive approach to improve the safety of battery electrolytes and to allow efficient operations at high voltage [1,2]. In this work, safe and high-performance gel polymer electrolytes (GPEs) have been developed and used in lithium metal batteries having $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (LNMO) as a high voltage spinel cathode.

Dry polymer membranes based on poly(vinylidene fluoride-*co*-hexafluoropropylene) are immersed in the electrolyte mixture composed of 1M LiPF_6 in ethylene carbonate/dimethyl carbonate (LP30) and 1-butyl-1-methyl-pyrrolidinium hexafluorophosphate ($[\text{Py}_{14}]\text{PF}_6$) to form GPEs. Non-flammable character and good mechanical stability of the GPEs are demonstrated with flammability tests and rheological measurements, respectively. Lithium-ion conduction mechanism in the GPEs is evaluated by means of both impedance measurements and pulsed gradient spin-echo NMR technique. Galvanostatic charge and discharge cycles of lithium metal batteries with LNMO demonstrate improved electrochemical performance, specifically high discharge capacity above 110 mAh g^{-1} and coulombic efficiency above 99% throughout 150 cycles at C/5, when the GPE immersed in LP30/ $[\text{Py}_{14}]\text{PF}_6$ 7/3 wt% mixture is used. In addition, its capacity retention is sufficiently high compared to the GPE without $[\text{Py}_{14}]\text{PF}_6$, confirming that the ionic liquid is a highly promising additive for improving battery life.

[1] M.A. Navarra, A. Tsurumaki, F. M. Vitucci, A. Paolone, O. Palumbo, S. Panero, “A novel Li^+ -conducting polymer membrane gelled by fluorine-free electrolyte solutions for Li-ion batteries”, *Batteries&Supercaps*, 2020, 3, 1112-1119.

[2] A. Tsurumaki, M. Agostini, R. Poiana, L. Lombardo, E. Lufrano, C. Simari, A. Matic, I. Nicotera, S. Panero, M.A. Navarra, “Enhanced safety and galvanostatic performance of high voltage lithium batteries by using ionic liquids”, *Electrochimica Acta*, 2019, 316, 1-7.

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