

Suppressing Li-solid electrolyte interface resistance in solid-state Li-metal battery

Syed Atif Pervez^a, Maximilian Fichtner^a
^a*Helmholtz Institute Ulm*
Helmholtzstraße 11, 89081 Ulm, Germany

E-mail: syedatif.pervez@partner.kit.edu

Li metal solid-state batteries continue to attract great interest, especially for electric vehicle application. These batteries employ Li metal, conventional cathodes as electrodes and solid-state Li-ion conductors as electrolytes. The prominent features of the solid-state electrolytes (SSEs) are high ionic conductivity at operating temperature (preferably room temperature) and negligible electronic conductivity over the employed range of lithium activity and temperature. In principle, SSEs can address the issues associated with liquid electrolytes due to various advantages such as higher Li transference number ($t_{Li^+} \sim 1$) owing to single ion nature, wide electrochemical potential windows (0-6 V vs. Li^+/Li), non-flammability and suppression of Li dendrites due to high shear modulus [1]. In recent times, a significant focus has been on crystalline inorganic SSEs. Such structures can offer enhanced ionic mobility than amorphous or polymer type, presumably due to preferential pathways offered by the crystalline phase. Inorganic SSEs include oxides such as garnet-type, perovskites, LISICON-type, NASICON-type, sulfides such as LGPS, $Li_2SP_2S_5$ and argyrodites such as Li_6PS_5X ($X = Cl, Br, I$) [1]. Among them cubic phase garnet-type Li-ion conductors ($Li_5La_3M_2O_{12}$ ($M = Nb, Ta, Zr, etc.$)) are considered promising SSEs due to their high stability in contact with Li, high Li^+ transference number, high ionic conductivities (10^{-4} - 10^{-3} S.cm⁻¹ at 25 °C), and wide potential window (0-6V vs. Li^+/Li). However despite the aforementioned advantages, integrating garnet-type SSEs with Li and other electrodes have not shown satisfactory device performance. This may be due to the high electrode/electrolyte interface resistance originating from poor wet-ability of garnet electrolytes by Li metal and inability of rigid solid-state conductors to adhere well to the moving boundaries of the battery electrode during charge/discharge process. To tackle the issue, various approaches have been adopted such as pressing Li against SSEs at high pressures, electrolyte surface modification, tuning chemical composition of the SSEs and so on [2].

In this work, we have introduced a polyethylene oxide (PEO) and lithium bis(trifluoromethanesulfonyl)imide based thin polymer membrane between Li metal and garnet SSE pellet to improve their solid-solid electrode/electrolyte contact and reduce the interfacial resistance. Owing to the flexible and adhesive nature of the PEO polymer, better electrode/electrolyte contacts are achieved which assist in regulating the Li^+ flux at the interface resulting in significant improvement in the area specific resistance. Compared to other interface modification techniques the proposed strategy is simple, cost-effective and easily adoptable in current battery manufacturing processes.

References:

- [1] J. C. Bachman, S. Muy, A. Grimaud, H.-H. Chang, N. Pour, S. F. Lux, O. Paschos, F. Maglia, S. Lupart, P. Lamp, L. Giordano, Y. Shao-Horn, Chem. Rev. 116 (2016) 140–162.
- [2] C. Yang, K. Fu, Y. Zhang, E. Hitz, L. Hu Adv. Mater. 36 (2017) 170169.