Hybrid organic-inorganic material for interfaces in all-solid-state lithium battery based on oxide solid electrolyte

Nataly C. Rosero-Navarro[a], Ryunosuke Kajiura[b], Akira Miura[a] and Kiyoharu Tadanaga[a]
[a] *Division of Applied Chemistry, Faculty of Engineering, Hokkaido University, Sapporo 060-8628, Japan*
[b] *Graduate School of Chemical Sciences and Engineering, Hokkaido University, Sapporo 060-8628, Japan*

E-mail: rosero@eng.hokudai.ac.jp

All-solid-state lithium batteries (ASLBs) based on oxide solid electrolytes (SE) are a practical proposal for large-scale energy storage applications because of chemical stability and ease to handle of oxide SE (e.g. $Li_7La_3Zr_2O_{12}$). Electrode/electrolyte interfaces have been improved using sintering additives (acting as binder) or incorporating an organic-liquid lithium conductor phase [1,2]. These processes still have important issues such as unfavorable reactions at the interface layer or the presence of porosity due to the deficient sintering.

In this work, a hybrid organic-inorganic material is proposed for the formation of suitable interface for ASLBs based on oxide SE. Hybrid network was prepared using TEOS (tetraethyl orthosilicate), GPTMS (3-glycidoxypropyltrimethosysilane) and EGDE (ethylene glycol diglycidyl ether) with different $LiClO_4$ content. Hybrid material was used as interface between $Li_7La_{2.95}$ $Ca_{0.05}Zr$ TaO_{12} pellet (LLCZT, 1 mm) and $LiCoO_2$ composite (LCO, 6 µm). Low heat treatment at 100 ºC was used to prepare the interface.

The obtained hybrid precursor was transparent and yellowish solution with a conductivity of ~ 0.7 $\times 10^{-4}$ S cm^{-1} at R.T. Hybrid membranes, prepared by casting and drying of the precursor solution, showed homogeneity/flexibility and thicknesses of ~800 µm. Fig 1a shows ionic conductivity of membranes with different lithium content. Conductivity up to 10^{-6} S cm^{-1} at R.T. ($\sigma \sim 10^{-4}$ S cm^{-1} at 100 ºC) with an activation energy of ca. 0.8 eV was obtained. Lithium amount higher than 0.4 mol lead to the drop of the conductivity because of overlapping charges. TGE-0.4Li membrane showed a Li^+ transport number of 0.4, indicating a good Li^+ mobility through ether oxygen's network. SEM-EDS analysis (Fig 1b) of LLCZT/LCO interface using TGE-0.4Li shows an intimate contact between LLCZT and LCO.

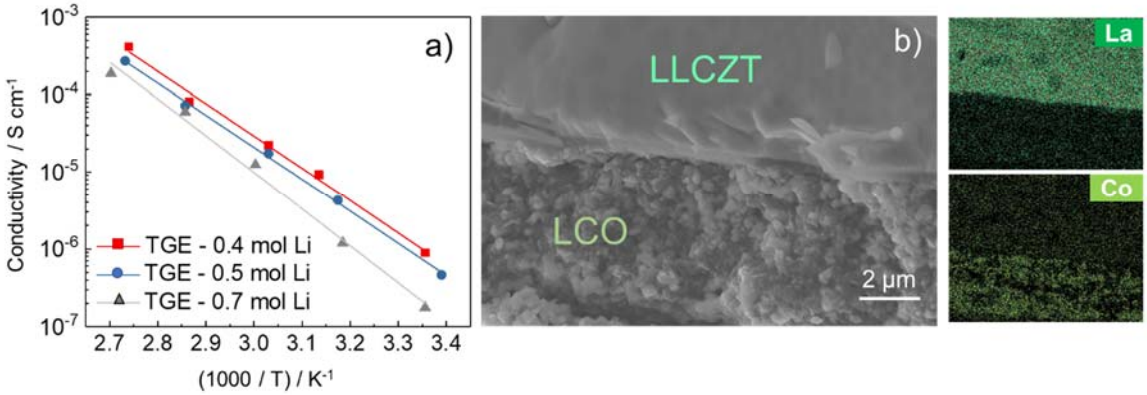

Figure 1. a) Ionic conductivity of hybrid organic-inorganic materials and b) SEM-EDS analysis of LLCZT/LCO interface using hybrid material.

References:

[1] J. Wakasugi, H. Munakata, K. Kanamura. Electrochemistry, 85 (2017) 77-81.
[2] F. Du, N. Zhao, Y. Li, C. Chen, Z. Liu, X. Guo. J. Power Sources 300 (2015) 24-28.

Acknowledgment. The present study is supported by KAKENHI Program, Japan. Reference Number 50791381.