Li-ion Conduction Measurement at Polycrystalline LLTO Grain Boundaries by Electrochemical Strain Microscopy

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The solid-state Li-ion electrolytes have been paid much attention as new electrolytes for Li-ion battery owing to their high safety [1]. Lithium lanthanum titanate (LLTO): $(Li_{3x}La_{2/3-x}\Box_{1/3-2x})TiO_3$ (0 < x < 0.16, \Box : A-site vacancy) [2] is one of the promising candidates for the next-generation solid-state Li-ion electrolytes because it shows high Li-ion conductivity inside the bulk. However, the Li-ion conductivity of LLTO is largely retarded at grain boundaries (GBs), which prevents the practical application of LLTO electrolytes. In the previous studies by electrochemical impedance spectroscopy, it has been impossible to reveal what kinds of GBs affect the Li-ion conductivity of LLTO. The purpose of this study is, therefore, to elucidate the effect of GBs on the Li-ion conductivity of LLTO and its conductive mechanism.

The LLTO polycrystalline samples were synthesized by the conventional sintering method. The high purity powders of Li_2CO_3 , TiO_2 , and La_2O_3 were mixed and then sintered at 1250 °C for 12 h to obtain LLTO polycrystalline samples. The local structure and the relative Li-ion conductivity at grain boundaries were measured by electron backscatter diffraction and electrochemical strain microscopy [3, 4]. It was found that the individual GBs show different Li-ion conductivity depending on their GB local structures.

References:

- [1] N. Kamaya, et al., Nat. Mater. 10 682 (2011) 682-686.
- [2] Y. Inaguma, et al., Solid State Commun. 86 (1993) 689-693.
- [3] S. Jesse, et al., MRS Bull. 37 (2012) 651-658.
- [4] S. Sasano, et al., Appl. Phys. Exp. 10 (2017) 061102.