Electrochemical properties of $LiCoO_2$-Li_3BO_3 composite cathode layer formed on Al-doped $Li_7La_3Za_2O_{12}$ solid electrolyte by aero-sol deposition

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All-solid-state batteries using ceramic electrolytes are promising candidates for next-generation energy storages due to their attractive properties, such as high safety, high energy density and long lifetime. In order to develop practical all-solid-state rechargeable lithium batteries, thicker electrodes with higher capacity should be utilized in the battery. Aerosol deposition (AD) method by using an impact consolidation of ceramic particles is one of coating processes to fabricate a ceramic layer onto a target substrate with a direct deposition of ceramic particles with very high deposition rate, so that it is desirable to fabricate thicker electrodes for all-solid-state rechargeable lithium batteries[1)]. The continuous conductive pathways of both Li^+ion and electron inside the thick electrode are required to achieve high utilization of electrode materials. $LiCoO_2$ (LCO) has high Li^+ion and electron conductivities among various cathode materials. However, the Li^+ion conductivity of LCO is not enough to achieve high electrode utilization in the thick electrode. In order to solve this problem, the composite particles of LCO and Li_3BO_3 (LBO) were synthesized. LBO in the thick cathode layer works as a Li^+ion conducting binder due to its soft nature. For further improvement of LCO utilization, a heat treatment at 750 °C for 1 hr was carried out after the cathode layer formation with LCO-LBO composite particles on an Al-LLZ (Al doped $Li_7La_3Zr_2O_{12}$ ($Li_{6.25}Al_{0.25}La_3Zr_2O_{12}$) pellet. The melting point of LBO is around 700 °C, so that more uniform and continuous Li^+ion conducting pathway was formed due to homogeneous distribution of LBO in the cathode layer. In this study, the electrochemical performance of the LCO-LBO composite cathode formed on Al-LLZ ceramic electrolyte by AD method was investigated by using the full cell with lithium-metal anode.

The charge and discharge curves of the full cell (LCO-LBO composite cathodes on Al-LLZ pellets) were obtained at 60 °C. The specific discharge capacity of LCO was 100 mA h g^{-1}. The charge and discharge cycle performance of the full cell was improved by the heat treatment. Interfacial impedances between LCO and LBO particles and between LCO-LBO composite cathode and Al-LLZ pellet with/without the heat treatment were measured after the first charge (Fig. 1). The impedance with the heat treatment was much smaller than that without the heat treatment. This results suggest that the AD method using composite particles is one of promising ways to fabricate the thick cathode with higher utilization on Al-LLZ electrolyte.

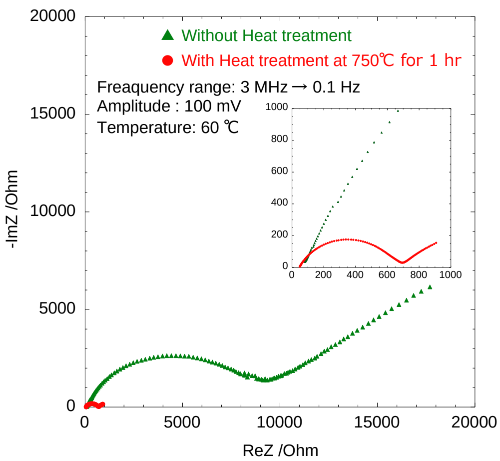

Fig. 1 The impedance spectra of the LCO- LBO / Al-LLZ pellet with/without heat treatment after AD.

References:

[1] T. Kato et al., J. Power Sources, **303** (2016) 65-72.