Fabrication of LiCoO₂ Composite Electrode on Li_{6.25}Al_{0.25}La₃Zr₂O₁₂ Solid Electrolyte by Aerosol Deposition Method

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All-solid-state rechargeable lithium batteries using non-flammable ceramic solid electrolytes are promising as next-generation batteries with high safety. In general, high temperature sintering is carried out for those batteries to form an electrochemical interface since both electrode and electrolyte are solid. However, the high temperature heat treatment limits the combination of electrode and electrolyte materials because of a formation of impurities through their chemical reactions at high temperatures. Aerosol deposition (AD) method is one of new dry processes which can form a ceramic layer on a substrate without high temperature heat treatment. In this study, we tried to optimize AD conditions to obtain a thick LiCoO₂ layer on a Li_{6.25}Al_{0.25}La₃Zr₂O₁₂ (LLZ) solid electrolyte for all-solid-state rechargeable lithium batteries. In addition to LiCoO₂ particles, its composite particles with Li⁺-conducting Li₃BO₃ were synthesized and tested. Fig. 1 shows the cross-sectional SEM images of electrodes formed using LiCoO₂ particles and LiCoO₂/Li₃BO₃ (7/3 in weight) composite particles. They indicate that soft Li₃BO₃ works like binder to provide a thick electrode. Fig. 2 shows the charge-discharge curves of LiCoO₂ electrode and LiCoO₂/Li₃BO₃ composite electrode on LLZ pellets at 60 °C. The initial discharge capacity and utilization of LiCoO₂ were estimated to be 12.8 mA h g^{-1} and 9.3 % in the LiCoO₂ electrode, 55.5 mA h g^{-1} and 40.1 % in the LiCoO₂/Li₃BO₃ composite electrode, respectively. This result indicates that Li₃BO₃ works as not only binder but also Li⁺-conducting pathways to improve the utilization of LiCoO₂. However, the obtained electrochemical performance was still low. Thus, the optimization of Li₃BO₃ content, electrode densification and so on are now underway for further improvement of LiCoO₂ utilization.





Fig.1 Cross-sectional SEM images of (a) LiCoO₂ electrode and (b) LiCoO₂/Li₃BO₃ composite electrode formed on LLZ pellets by AD method.

Fig.2 1st charge-discharge curves of (a) $LiCoO_2$ electrode and (b) $LiCoO_2/Li_3BO_3$ composite electrode on LLZ pellets, measured at 60 °C and 0.1 C-rate.