Ionic Conductivity of Garnet-type Thin Film Li-La-Zr-O Electrolyte

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Among many research on all-solid-state lithium-ion batteries, attention has been directed to the solid-state electrolyte for solving the safety issue and increasing energy and power density. Regarding battery engineering, garnet-type $Li_7La_3Zr_2O_{12}$ (LLZO) solid electrolyte with cubic phase has drawn a lot of interest due to its reported chemical stability against lithium metal and the high ionic conductivity (10^{-4} to 10^{-3}S/cm).[1][2][3] Toward diverse applications of lithium-ion batteries, thin film scale provides a larger degree of freedom in the design of the battery structures. In this study, we used a simple sputtering process to fabricate LLZO thin film by co-sputtering LLZO and Li_2O targets with post-annealing at 700°C for 1 hour. By changing the amount of Li_2O addition and sintering agent, we can control the phase formation of garnet-type LLZO from tetragonal to cubic phase. Furthermore, to observe the density effect combined with phase transformation on ionic properties, the LLZO thin films without and with sintering agent were analyzed by scanning electron microscopy (SEM), grazing incident X-ray diffraction (GI-XRD) and electrochemical impedance spectroscopy (EIS). The Hebb-Wagner polarization method was also used to measure partial electronic conductivity. A conductivity of 2×10^{-5} S cm^{-1} at 298K by in-plane measurement on a mixed-phase thin film LLZO electrolyte (400 nm) was demonstrated. Our result reveals the key factor for improving ionic conductivities of LLZO thin film is not only about achieving cubic phase but also creating a denser thin film.

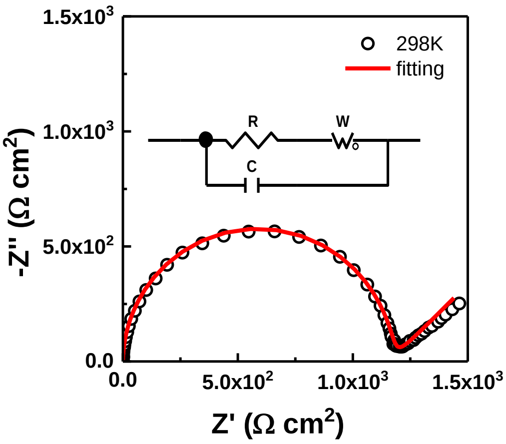

Figure 1. Nyquist plots of ac impedance spectra for the thin film LLZO.

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