Synthesis and characterization of garnet-type of $Li_7La_3Zr_2O_{12}$ via modified polymer complex synthesis

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Up to now, much attention has been paid to lithium batteries with high energy density and high performance. There are, however, still remained unsolved with respect to safety issue since the organic electrolyte has been applied to lithium rechargeable battery. The battery safety should be more addressed in the first rank with higher energy density and larger sized battery. The solid electrolyte has been regarded as a basic solution to safety issues.

Several kinds of solid electrolytes (i.e., NASICON, Thio-LISICON, and Garnet types, etc.) have been mainly focused on getting high ionic conductivity comparable with liquid type of conventional electrolyte system. The NASICON type [$Li_{1+x}Ti_{2-x}Al(PO_4)_3$] of solid electrolyte shows high ionic conductivity of 10^{-3} S/cm, but instability at interface with anode (reduction potential range). The Thio-LISICON type [Li-Ge-P-S] system has been reported to be highest of ionic conductivity of 10^{-3} S/cm, but generate H_2S gas in the atmosphere due to high reactivity with residual water. On the contrary, the garnet type [$Li_7La_3Zr_2O_{12}$] system shows excellent electrochemical stability in wide potential range and very stable in atmosphere with relatively high ionic conductivity (~10^{-4} S/cm) [1-2].

To get ionic conductivity of garnet system comparable to sulfide system, the density of sintered membrane is demanded to be increased. In order to solve the ionic conductivity of garnet grain boundary in the sintered membrane, it is necessary to decrease thermal energy required for crystallization and sintering process.

In this work, we have applied newly modified polymer complex process to synthesize garnet type solid electrolyte with lower crystallization temperature and lower sintering temperature. The micro-structural and electrochemical characteristics of solid electrolyte are compared with that of conventional synthesis process. The decrease of crystallization temperature (Δ=~200^oC) has been identified from TG/DSC behavior and X-ray diffraction pattern with heat treatment temperature. The Li ionic conductivity of sintered membrane with cubic structure at lower sintering temperature is measured by AC impedance technique. Furthermore, the technical potential and issues of cathode material with artificial interface applying garnet type solid electrolyte will be discussed for all solid battery system.

References

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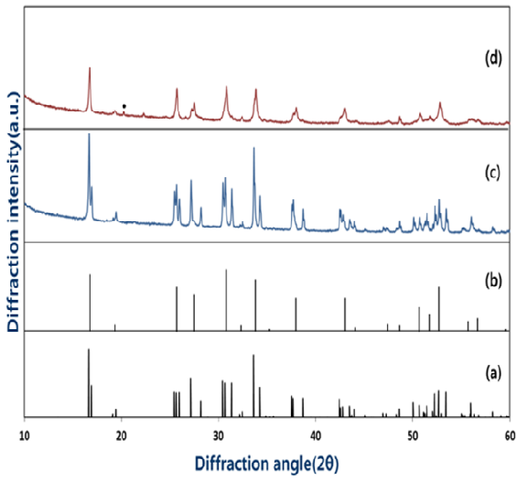

Fig. 1 XRD spectra of $Li_7La_3Zr_2O_{12}$: (a)simulated pattern of tetragonal LLZO, (b) simulated pattern of cubic LLZO, (c)LLZO calcined at 700°C for 5h, (d)LLZO calcined at 1100°C for 5h (•,Li_2ZrO_3)