Sol-gel synthesis of $Li_{3x}La_{2/3-x}TiO_3$ solid electrolyte using acetate precursor for all-solid-state battery

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The electrolytes for lithium secondary batteries are mainly based on organic solvent. Although present commercial liquid electrolyte provides fast lithium diffusion and less resistance, there are still many problems such as leakage, explosion, forming of dendrite from the organic solvent. Moreover, owing to the narrow operating voltage range, the decomposition of liquid electrolyte at high voltages restricts the development of high voltage batteries. [1] Therefore, it is necessary to develop a novel solid state electrolyte for overcoming the disadvantages of liquid electrolyte. In order to apply solid electrolyte to battery, it is essential to have not only high ionic conductivity as much as 10^{-2} S cm^{-1} equivalent to liquid electrolyte, but also stability under operating condition. Recently, thio-LISICON type Li-Ge-P-S solid electrolyte was reported to have the highest ionic conductivity of 10^{-2} S cm^{-1}. But the stability of thio-LISICON is very low when exposed to atmosphere due to the reaction with moisture in air. However, $Li_{3x}La_{2/3-x}TiO_3$ (LLTO) of perovskite type is stable against air and is a promising material for solid electrolyte due to the ionic conductivity in the order of 10^{-3} S cm^{-1}.

In the present study, LLTO was prepared by sol-gel reaction using acetate precursors. The XRD patterns (figure 1) confirmed the formation of LLTO with tetragonal structure. The reflection matched the P4/mmm space group as given in JCPDS card 87-0935. SEM images (figure 2) revealed the morphology of LLTO consisting of micron size particles. In order to measure the ionic conductivity each surface of LLTO pellet was sputtered with platinum, which acted as blocking electrodes. Conductivity measurements were carried out in the frequency range of 1MHz - 25Hz at a constant voltage of 0.1 V. In order to investigate stability, LLTO was immersed in various organic solvents such as NMP, DMC, etc for over a week and the pellets were tested for any structural deformation. Furthermore, a cell containing LLTO as electrolyte was tested using cyclic voltammetry between 0 – 5 V for checking the electrochemical stability. A detailed discussion based on the experimental results will be presented.

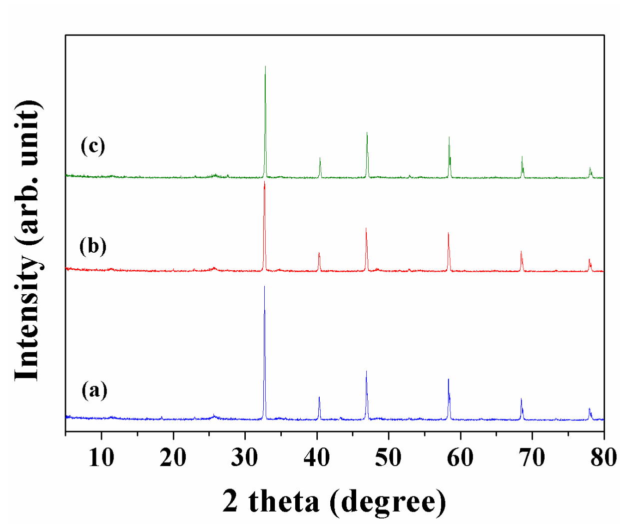

Fig. 1 XRD patterns of LLTO solid electrolyte calcined at (a) 1050 ^oC, (b) 1100 ^oC and (c) 1150 ^oC.

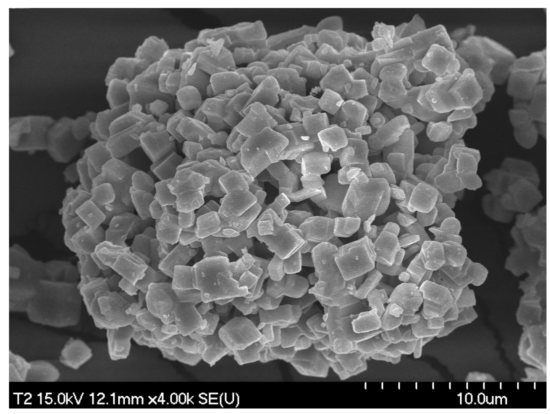

Fig. 2 SEM image of LLTO powder prepared by sol-gel method

References

[1] Danna Qian, Bo Xu, Hyung-Man Cho, Toru Hatsukade, Kyler J. Carroll and Ying Shirley Meng, *Chem. Mater.* (2012), DOI:10.1021/cm300929r