Preparation of an ion-conductive LATP-based electrolyte thin film for lithum-ion microbatteries

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The progress in all-solid-state lithium-ion (Li^+) microbatteries, which is necessary for modern and future microelectronics, requires the development of high ionically conductive electrolytes. Among the various solid Li^+ electrolytes, the inorganic NASICON-type ceramic lithium aluminum titanium phosphate – $Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$ (LATP) has attracted high interest due to its air and water stability, high Li^+ conductivity and low cost. Though the ionic conductivity of crystalline bulk LATP is able to achieve ~10^{-3}–10^{-4} S/cm at room temperature (RT), the value for amorphous LATP film with thickness varies from 300 nm to 1000 nm currently does not exceed 10^{-6} S/cm and for crystalline 10^{-5} S/cm [1, 2, 3, 4].

Different techniques can be applied for synthesis of LATP thin films including sol-gel, magnetron sputtering (MS), pulsed laser deposition (PLD), spin-coating, aerosol deposition and etc. Among all methods, magnetron sputtering technique allows achieving thin films with smooth surface favorable for microbatteries due to its deposition feature. However, this synthesis method faces the problem of undesirable losses of lithium during sputtering and subsequent annealing.

Here we show that co-depositing of the film by two target sources, it is possible to reduce/compensate for the loss of lithium in the composition of LATP thin films during deposition. A series of depositions from LATP and Li_3PO_4 were carried out to study the effect of sputtering parameters, such as power (10-25%), deposition gas (Ar, N_2, O_2), on the composition and the ionic conductivity of the electrolyte film (Figure 1).

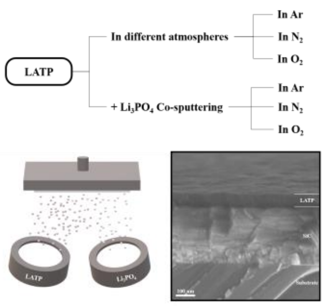

Figure 1. The schematically view of co-sputtering

Our results demonstrate how the deposition atmosphere affects the deposition rate and ionic conductivity of the thin films, as well as co-deposition with Li_3PO_4. In order to obtain the electrolyte films with the improved electrochemical characteristics, the parameters of the deposition process varied as well as the strategy to thin films' structure. Thus, the monolithic LATP sputtered in different gases, co-sputtered LATP and lithium phosphorus oxynitride (LiPON) with a general formula of $Li_xPO_yN_z$ were prepared and investigated. The film composition was determined by X-ray photoelectron spectroscopy (XPS) and inductively coupled plasma mass spectrometry (ICP-MS). A room-temperature conductivity σ_{RT} of ~10^{-6} S/cm is obtained for a thin LATP film obtained in nitrogen atmosphere. The compositional dependence of Li^+ transport mechanism is discussed.

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