

Supporting Information

Controllable $\text{Li}_3\text{PS}_4\text{-Li}_4\text{SnS}_4$ solid electrolytes with affordable conductor and high conductivity for solid-state battery

Pingping Dong¹, Qing Jiao^{1*}, Zengcheng Zhang¹, Miao Jiang³, Changgui Lin¹, Xianghua Zhang², Hongli Ma², Baochen Ma¹, Shixun Dai¹, Tiefeng Xu⁴

¹ Laboratory of Infrared Material and Devices, Advanced Technology Research Institute, Ningbo University, Ningbo 315211, China

² Laboratory of Glasses and Ceramics, Institute of Chemical Science, UMR CNRS 6226, University of Rennes 1, Rennes, France

³ Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo, Zhejiang 315201, China

⁴ Ningbo Institute of Oceanography, Ningbo 315832, China

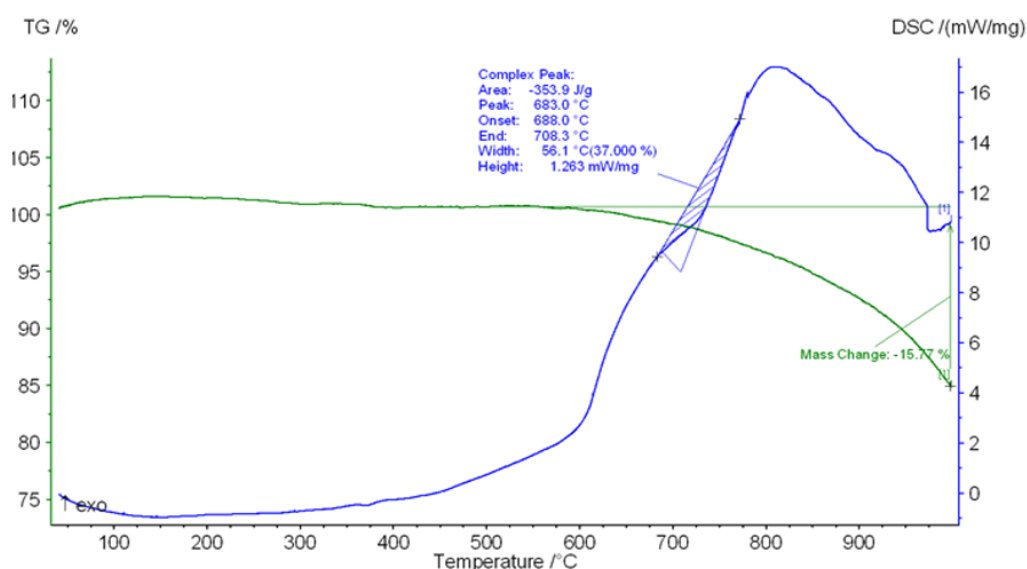


Fig. S1. Thermogravimetric/differential thermal (TG/DSC) graph of $2.5\text{Li}_3\text{PS}_4\text{-}0.5\text{Li}_4\text{SnS}_4$ SSE.

The crystallization temperatures (T_c) of amorphized samples were measured by Thermogravimetric/differential thermal (TG/DSC) to identify the optimum heat treatment temperature for glass-ceramics phases, as shown in supplement Fig. S1. The TG/DSC was performed for a powdered sample sealed in a titanium pan using a scanning calorimeter at a heating rate of 10 °C/min. It can be seen that the initial crystallization range of the $2.5\text{Li}_3\text{PS}_4-0.5\text{Li}_4\text{SnS}_4$ sample is about 400-600 °C, which is basically selected as the heat treatment temperature range in this experiment. And heat treatment of $2.5\text{Li}_3\text{PS}_4-0.5\text{Li}_4\text{SnS}_4$ SSE at different temperatures was carried out in this temperature range. The sample is partly to be melted at higher temperature over 700 °C, with obvious weight loss according to the TG result accompanied by the probable sulfur gasification loss.

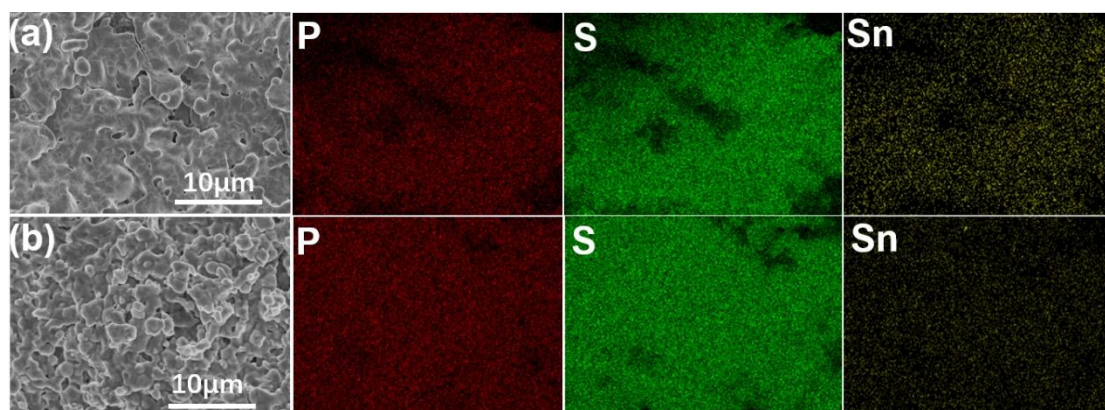


Fig. S2. Scanning electron microscope (SEM), Energy Dispersive Spectrometer (EDS) before (a) and after heat treatment (b) of $2.5\text{Li}_3\text{PS}_4-0.5\text{Li}_4\text{SnS}_4$ ($x = 1.5$).

The SEM/ EDS before and after heat treatment of $2.5\text{Li}_3\text{PS}_4-0.5\text{Li}_4\text{SnS}_4$ ($x = 1.5$) was shown in Fig. S2. For $2.5\text{Li}_3\text{PS}_4-0.5\text{Li}_4\text{SnS}_4$, a small number of crystals were precipitated by only ball milling was carried out without heat treatment, and the whole electrolyte was still in the glass state with tendency of phase separation. After 550°C

heat treatment, a large number of crystals were precipitated in the sample, the contact between grains was closer and the density was intensified, which reduced the grain boundary impedance and improved the ionic conductivity. In addition, the EDS spectra show that the elements are uniformly distributed inside the sample before and after heat treatment, even better after heat treatment. Therefore, it was believed that the conductivity was optimized by modification of the electrolyte component and further heat influence.