Supporting Information

Metastable chloride solid electrolyte with high formability for rechargeable all-solid-state lithium metal batteries

Naoto Tanibata^{*1,2}, Shuta Takimoto¹, Koki Nakano¹, Hayami Takeda^{1,2}, Masanobu

Nakayama^{1,2,3}, Hirofumi Sumi⁴

¹ Department of Advanced Ceramics, Nagoya Institute of Technology, Gokiso, Showa,

Nagoya, Aichi 466-8555, Japan

² Elements Strategy Initiative for Catalysts and Batteries (ESICB), Kyoto University, 1-30

Goryo-Ohara, Nishikyo, Kyoto 615-8245, Japan

³ Frontier Research Institute for Materials Science (FRIMS), Nagoya Institute of Technology, Gokiso, Showa, Nagoya, Aichi 466-8555, Japan

⁴ Inorganic Functional Materials Research Institute, National Institute of Advanced Industrial

Science and Technology (AIST), 2266-98, Anagahora, Shimo-shidami, Moriyama-ku,

Nagoya 463-8560, Japan

*Corresponding Author: <u>tanibata.naoto@nitech.ac.jp</u>



Supporting Figure S1. DC polarization curve for the powder-compressed pellet of the mechanochemical synthesized LiAlCl₄ sample. The applied voltage is 1 V. The sharp drop in the current and the low steady-state current indicates the low contribution of electrical conductivity in the LiAlCl₄ material.



Supporting Figure S2. Arrhenius plots of the total conductivity for the powder-compressed pellet of the mechanochemical synthesized LiAlCl₄ sample.



Supporting Figure S3. Impedance plots of LiAlCl₄ green compacts at 25 °C sandwiched with

stainless steel (SUS) or Li metal electrodes.



Supporting Figure S4. Galvanostatic cycle performance of the Li symmetric cells with solid electrolyte of $Li_{6.6}La_3Zr_{1.6}Ta_{0.4}O_{12}$ green compacts at 25 °C at the current density of 0.1 mA cm⁻².



Supporting Figure S5. Impedance plots of the Li symmetric cells with solid electrolyte of $Li_{6.6}La_3Zr_{1.6}Ta_{0.4}O_{12}$ green compacts at 25 °C after the first cycle at the current density of 0.01 mA cm⁻².