

Tape-cast Ce-substituted $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ Electrolyte for Improving Electrochemical Performance of Solid-State Lithium Batteries

Purna Chandra Rath, Yu-Syuan Jheng, Cheng-Chia Chen, Chih-Long Tsai, Yu-Sheng Su,
Chun-Chen Yang, Rüdiger-A. Eichel, Chien-Te Hsieh, Tai-Chou Lee, Jeng-Kuei Chang

Table S1. Chemical composition of LLZO, LGLZO, and various LLZCO samples examined using ICP-MS.

Sample	Chemical formula	Li	La	Zr	Ce	Ga
LLZO	$\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$	5.16	3.000	1.97	NA	NA
LGLZO	$\text{Li}_{6.25}\text{Ga}_{0.25}\text{La}_3\text{Zr}_2\text{O}_{12}$	4.18	3.000	2.00	NA	0.28
LLZCO-0.05	$\text{Li}_7\text{La}_3\text{Zr}_{1.95}\text{Ce}_{0.05}\text{O}_{12}$	4.99	3.000	1.96	0.050	NA
LLZCO-0.10	$\text{Li}_7\text{La}_3\text{Zr}_{1.90}\text{Ce}_{0.10}\text{O}_{12}$	5.02	3.000	1.90	0.095	NA
LLZCO-0.15	$\text{Li}_7\text{La}_3\text{Zr}_{1.85}\text{Ce}_{0.15}\text{O}_{12}$	5.02	3.000	1.84	0.145	NA

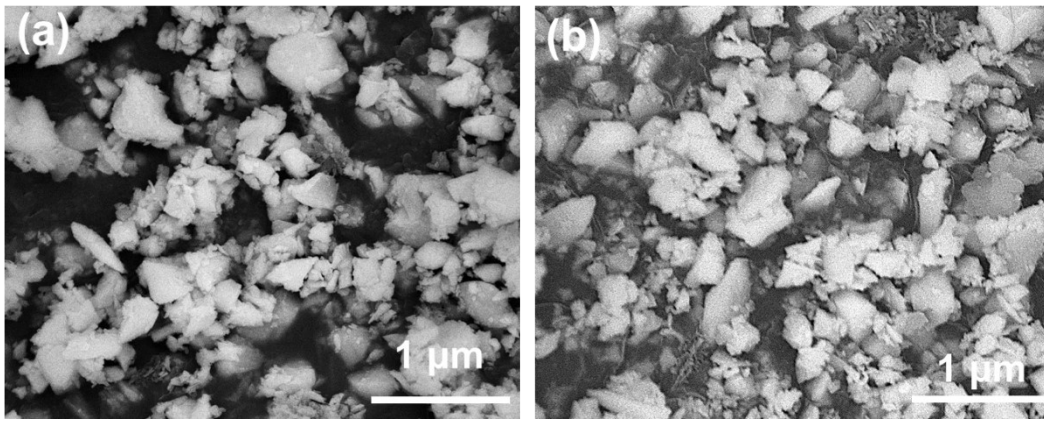


Fig. S1. SEM images of (a) LLZCO-0.05 and (b) LLZCO-0.15 samples.

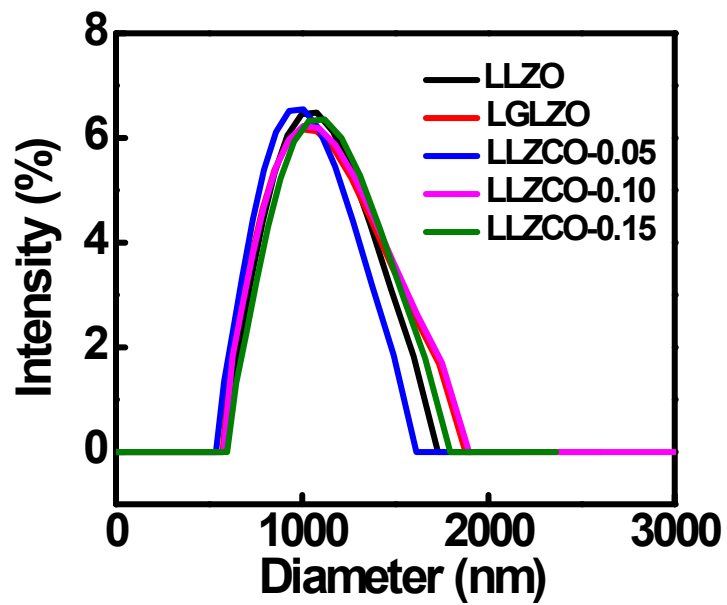


Fig. S2. DLS data of LLZO, LGLZO, and various LLZCO samples.

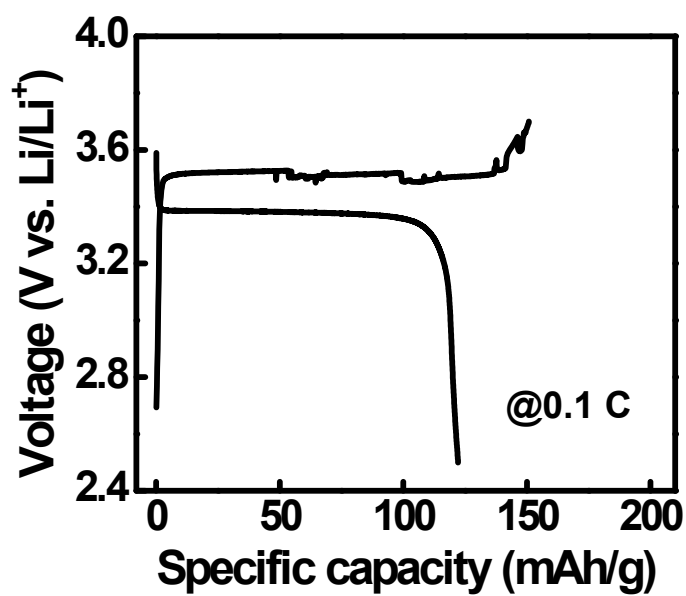


Fig. S3. Charge-discharge curves of Li//CSE//LFP cell with 25 wt.% LGLZO and EO/Li⁺ molar ratio of 5 in CSE.

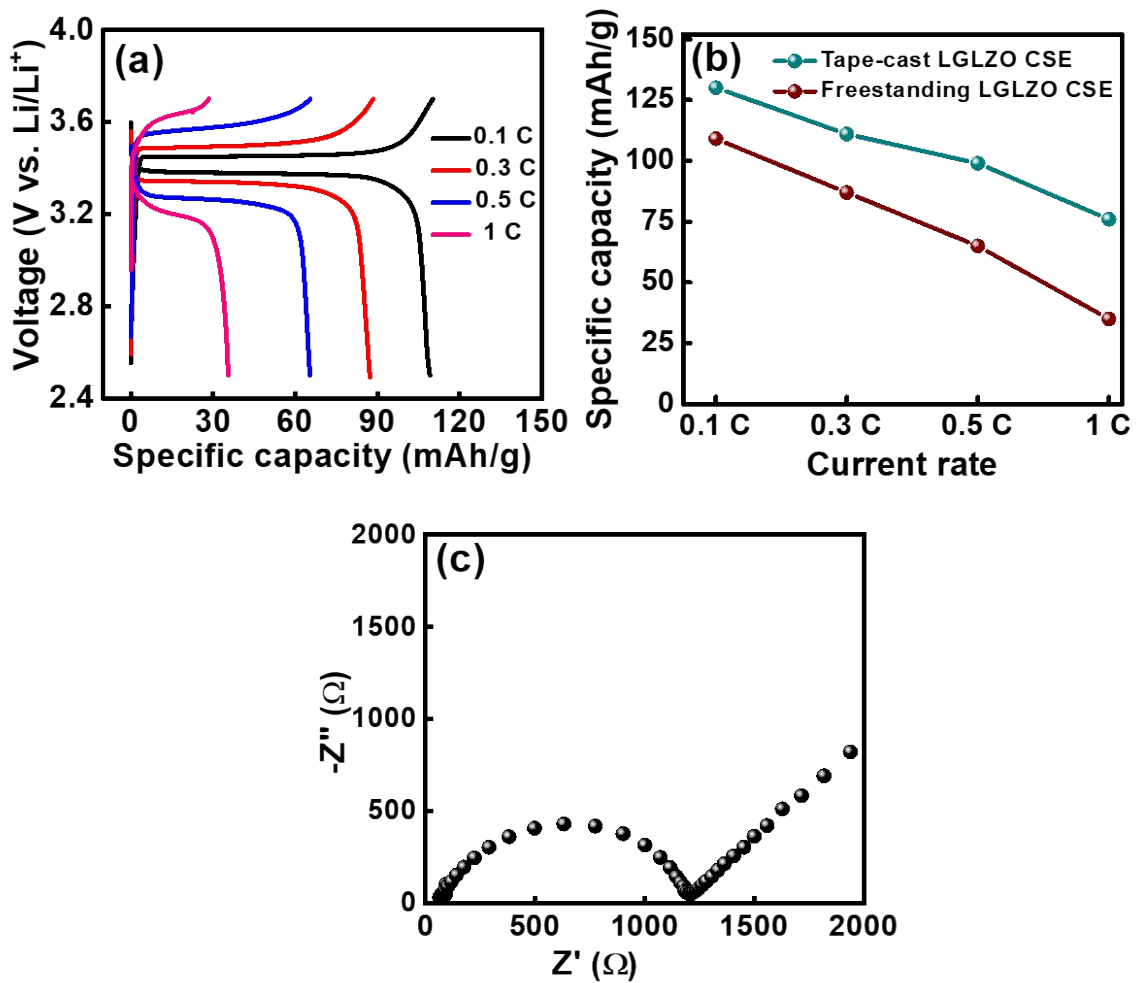


Fig. S4. (a) Charge-discharge curves of Li//LFP cell assembled with freestanding CSE. (b) Comparative rate performance of Li//LFP cells with freestanding and tape-cast CSE layers. (c) EIS spectra of Li//LFP cell assembled with freestanding CSE.

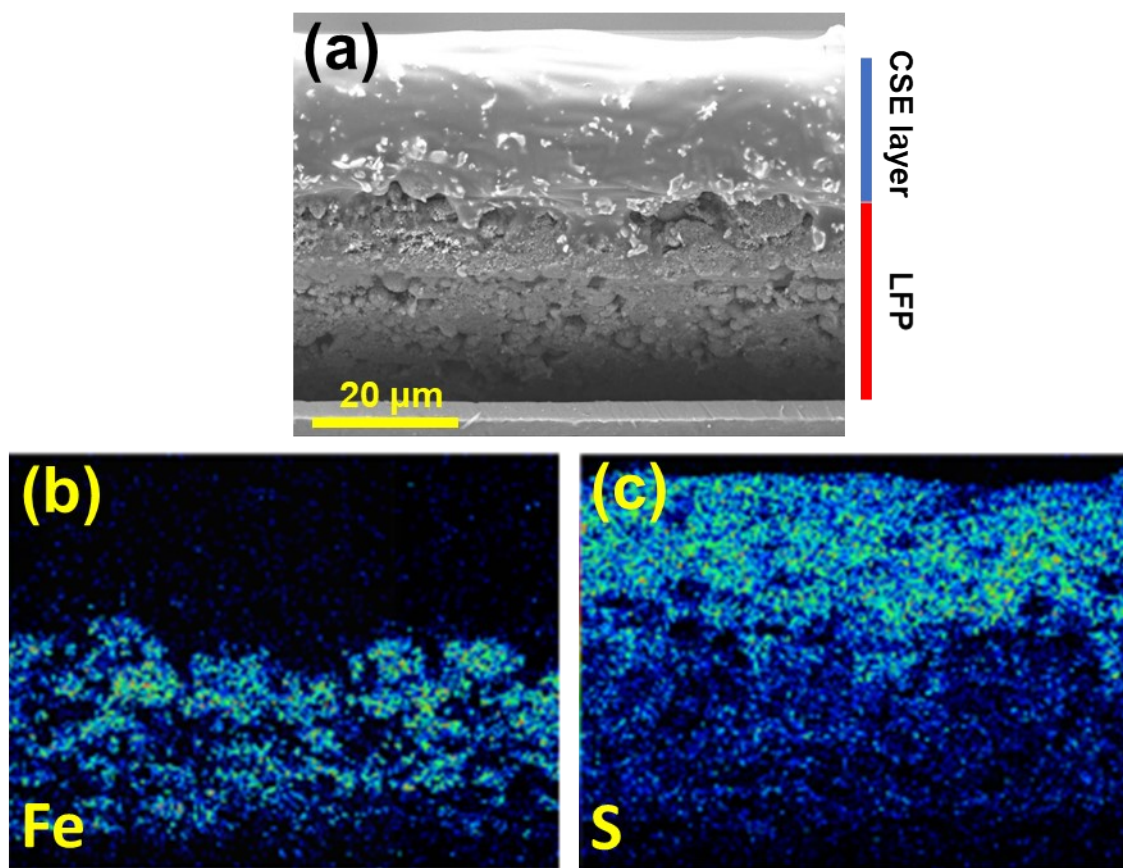


Fig. S5. (a) Cross-sectional SEM image of tape-cast CSE on LFP cathode. (b) The corresponding EDS Fe and S mapping in the same region.

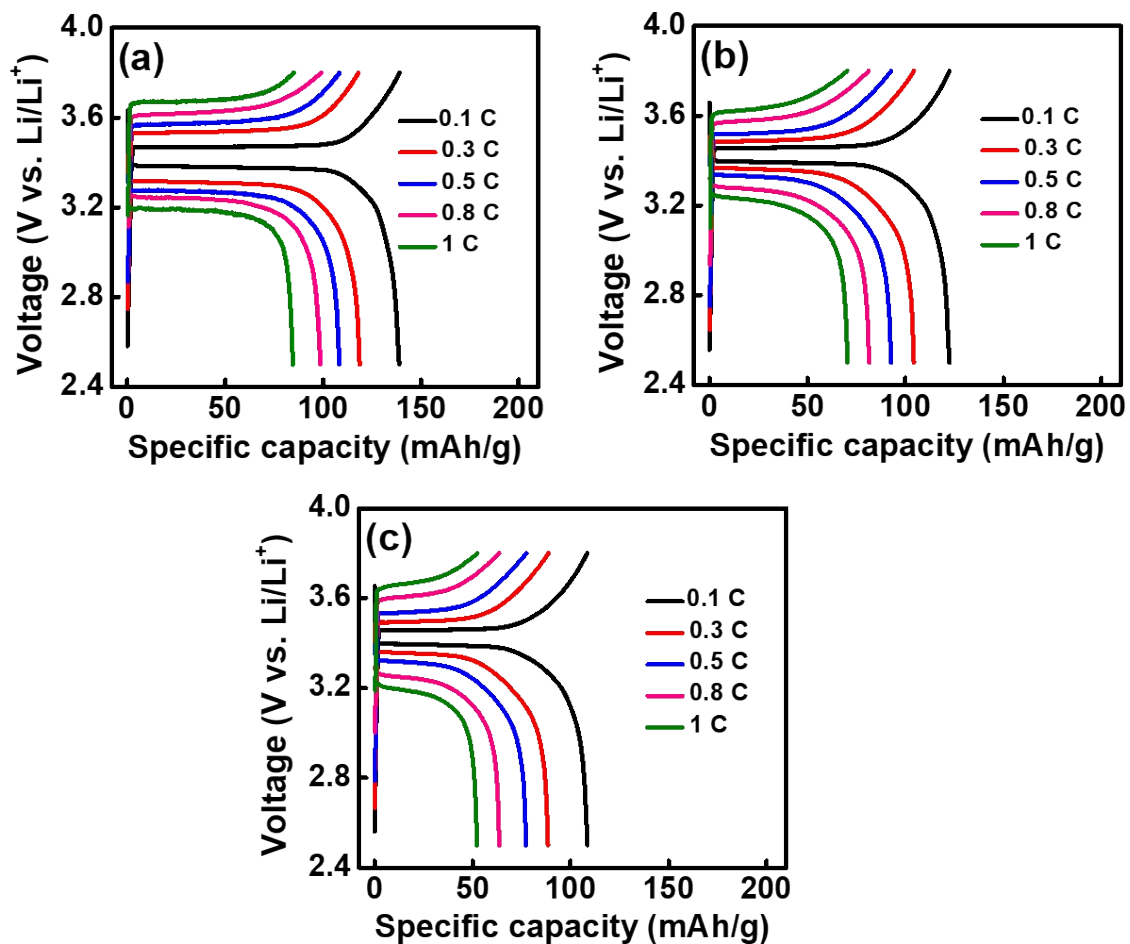


Fig. S6. Charge-discharge curves of Li//CSE (with EO/Li⁺ = 10 and 35 wt.% LGLZO)//LFP cell with CSE thickness of (a) 20, (b) 50, and (c) 80 μm.

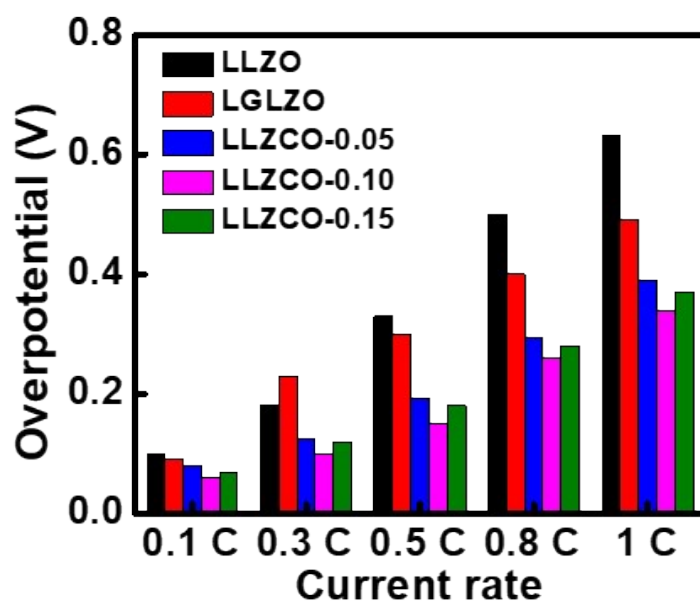


Fig. S7. Overpotential derived from charge-discharge curves (in **Fig. 7**) of Li//CSE//LFP cells with various CSEs.

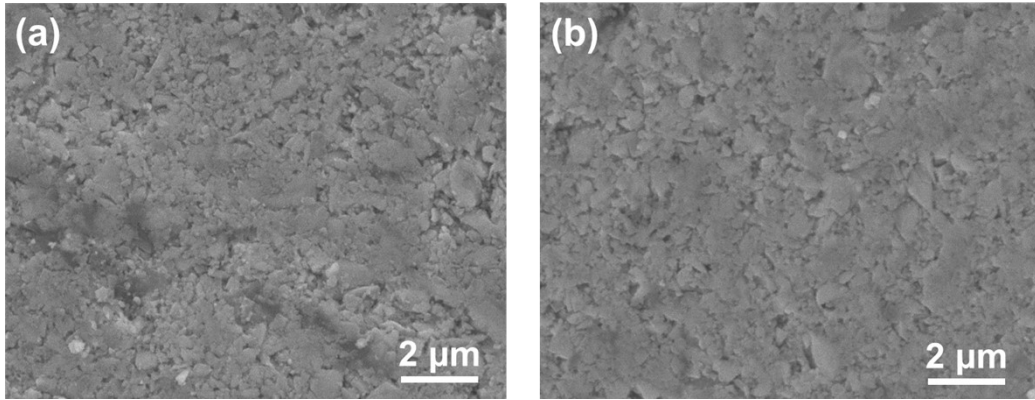


Fig. S8. SEM images of (a) LLZO (b) LLZCO-0.10 pellets after Li contact for 48 hours at 70 °C.

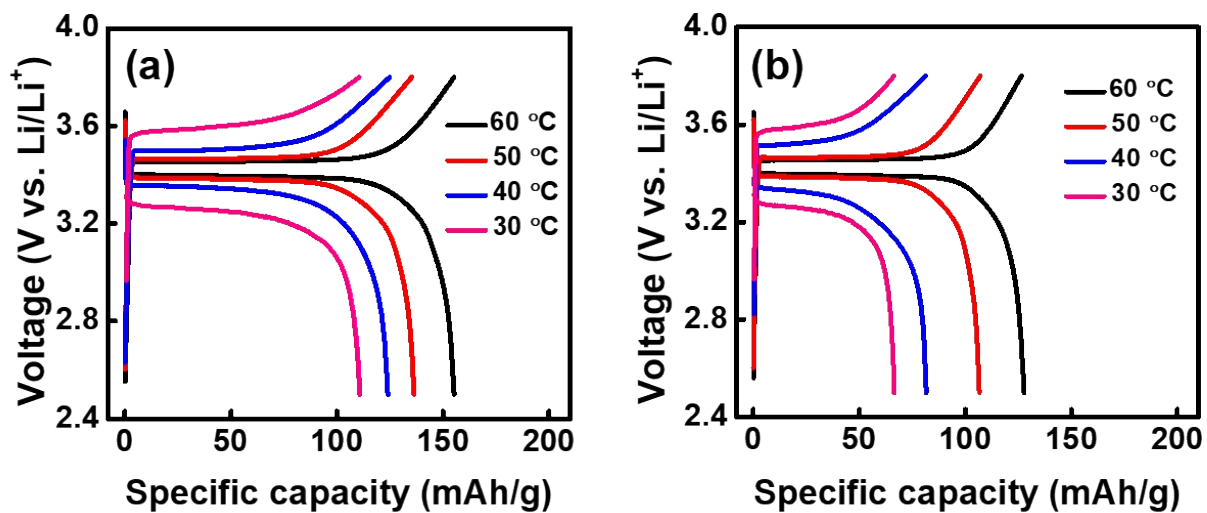


Fig. S9. Charge-discharge curves of Li//LLZCO-0.10 CSE//LFP cell measured at (a) 0.1 C and (b) 0.5 C at various temperatures.