

Supporting information for

## The synergistic effects of Al and Te on the structure and Li<sup>+</sup>-mobility of the garnet-type solid electrolytes

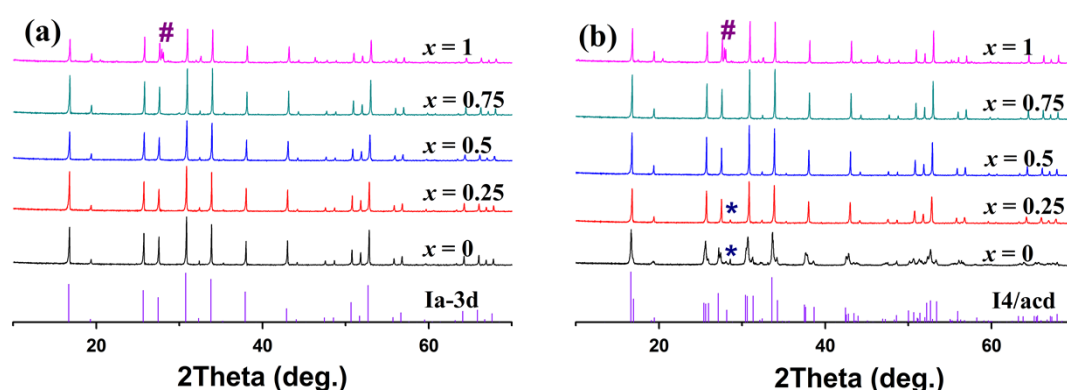
Dawei Wang<sup>a</sup>, Guiming Zhong<sup>a</sup>, Oleksandr Dolotko<sup>b</sup>, Yixiao Li<sup>a</sup>, Matthew J McDonald<sup>a</sup>, Jinxiao Mi<sup>c</sup>, Riqiang Fu<sup>d</sup>, Yong Yang<sup>\*a</sup>

<sup>a</sup> State Key Lab of Physical Chemistry of Solid Surface and Department of Chemistry, Xiamen University, Xiamen, 361005, China.

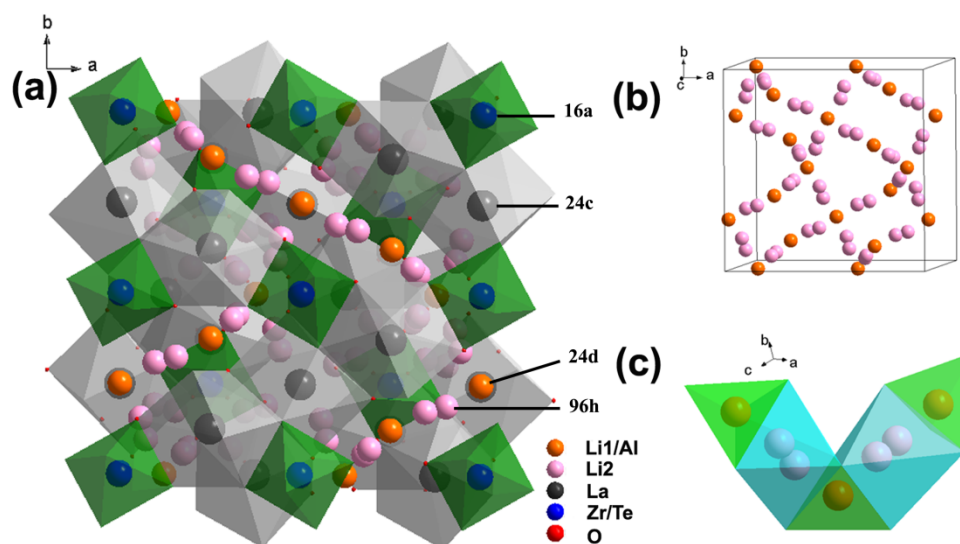
<sup>b</sup> Heinz Maier-Leibnitz Zentrum (MLZ), Technische Universität München, Lichtenbergstr. 1, D-85748 Garching, Germany.

<sup>c</sup> Department of Material Science and Engineering, Xiamen University, Xiamen, 361005, China.

<sup>d</sup> National High Magnetic Field Laboratory, 1800 E. Paul Dirac Drive, Tallahassee, Florida, 32310, USA.



**Fig. S1** XRD patterns of  $\text{Li}_{7-x}\text{La}_3\text{Zr}_{2-x}\text{Te}_x\text{O}_{12}$  ( $0 \leq x \leq 1$ ) samples sintered in (a)  $\text{Al}_2\text{O}_3$  crucibles, (b)  $\text{ZrO}_2$  crucibles. \*denotes  $\text{La}_{0.5}\text{Zr}_{0.5}\text{O}_{1.75}$  #01-075-0346, and # represents  $\text{La}_2\text{O}_3$  #01-089-4016.



**Fig. S2** (a) Crystal structure of the cubic garnet (refined from neutron diffraction data), (b) Lithium jump route proposed by Awaka *et al.*<sup>20</sup>, (c) Polyhedral coordination of Li1 and Li2.

Table S1. Fitting results of Nyquist plots for samples with  $x = 0.25$  sintered in  $\text{Al}_2\text{O}_3$  crucibles, with  $k = A/l = 5.91$ .

| T(°C) | L (H)         | R1 ( $\Omega$ ) | CPE1-T (F)      | CPE1-P      | R2 ( $\Omega$ ) | CPE2-T (F)     | CPE2-P      |
|-------|---------------|-----------------|-----------------|-------------|-----------------|----------------|-------------|
| -50   | 7.8E-7(N/A)   | 1.62E5(0.39%)   | 8.8E-11(3.16%)  | 0.87(0.26%) | 1E20(100%)      | 1.0E-8(3.68%)  | 0.87(0.56%) |
| -30   | 7.8E-7(N/A)   | 2.65E4(0.48%)   | 1.9E-10(7.04%)  | 0.83(0.57%) | 1E20(100%)      | 1.2E-8(5.55%)  | 0.88(0.69%) |
| -10   | 7.8E-7(N/A)   | 5.4E3(1.06%)    | 3.9E-10(20.96%) | 0.82(1.62%) | 1E20(100%)      | 1.7E-8(13.32%) | 0.87(1.42%) |
| 10    | 7.8E-7(N/A)   | 1.4E3(0.74%)    | 2.1E-10(22.61%) | 0.87(1.57%) | 1E20(100%)      | 3.1E-8(8.75%)  | 0.84(0.89%) |
| 25    | 7.8E-7(N/A)   | 4.24E2(1.40%)   | 5.6E-10(59.86%) | 0.82(4.19%) | 1E20(100%)      | 4.1E-8(23.75%) | 0.84(2.11%) |
| 50    | 7.8E-7(N/A)   | 1.74E2(0.72%)   | -               | -           | 1E20(100%)      | 8.2E-8(14.54%) | 0.79(1.27%) |
| 70    | 7.8E-7(N/A)   | 81(0.96%)       | -               | -           | 1E20(100%)      | 1.6E-7(11.25%) | 0.76(1.035) |
| 90    | 7.8E-7(N/A)   | 42(0.62%)       | -               | -           | 1E20(100%)      | 2.0E-7(4.99%)  | 0.76(0.45%) |
| 110   | 7.8E-7(N/A)   | 24(5.53%)       | -               | -           | 1E20(100%)      | 2.3E-7(32.04%) | 0.76(2.88%) |
| 130   | 7.8E-7(N/A)   | 16(1.96%)       | -               | -           | 1E20(100%)      | 1.2E-8(7.09%)  | 0.78(0.63%) |
| 150   | 7.8E-7(2.50%) | 11(3.42%)       | -               | -           | 1E20(100%)      | 2.4E-7(11.15%) | 0.77(1.02%) |

L was obtained by fitting Nyquist plot measured at 150 °C, and then fixed at this value while fitting the other data.