

Supporting Information

Compositional and structural control in LLZO solid electrolytes

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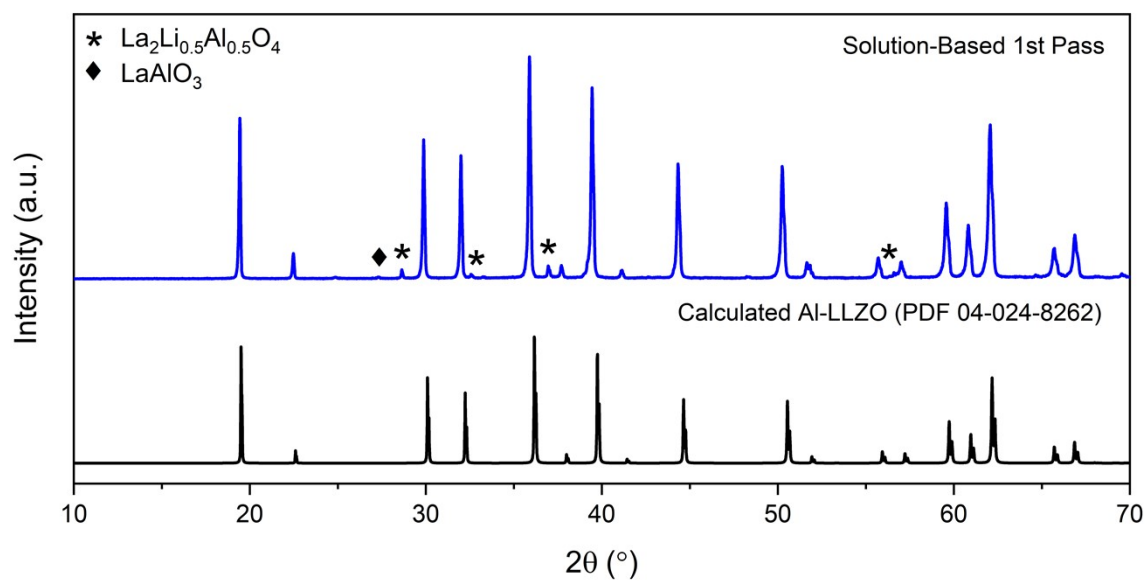


Figure S1. XRD pattern for calcined Al-LLZO powders for the SB sample with 0wt% excess Zr-precursor. The theoretical diffraction pattern of cubic Al-LLZO is included for reference.

Table S1. ICP-OES data for Al-LLZO samples with varying dopant contents where nominal stoichiometry was based on $\text{Li}_{7-3x}\text{Al}_x\text{La}_3\text{Zr}_2\text{O}_{12}$. Data has been normalized to La. Chemical formulae have been calculated from molar ratios and oxygen content has been estimated from charge balance calculations.

Nominal Al Content x (mol pfu)	Measured Al Content x (mol pfu)	Nominal Al wt%	Measured Al wt%	Li/La	Al/La	Zr/La	Chemical Formula
0	0	0	0	2.69	0	0.54	$\text{Li}_{8.08}\text{La}_3\text{Zr}_{1.62}\text{O}_{11.78}$
0.12	0.12	0.39	0.38	2.53	0.04	0.60	n/a [†]
0.18	0.18	0.58	0.59	2.47	0.06	0.61	$\text{Li}_{7.41}\text{Al}_{0.18}\text{La}_3\text{Zr}_{1.84}\text{O}_{12.16}$
0.24	0.25	0.77	0.80	2.39	0.08	0.63	$\text{Li}_{7.16}\text{Al}_{0.25}\text{La}_3\text{Zr}_{1.89}\text{O}_{12.23}$
0.3	0.32	0.96	1.02	2.37	0.11	0.63	n/a ^{††}
0.36	0.38	1.15	1.22	2.28	0.13	0.60	n/a ^{††}

[†]Composite of cubic and tetragonal LLZO phases

^{††}Impurities LaAlO_3 and $\text{La}_2\text{Zr}_2\text{O}_7$ present (see Figure S2)

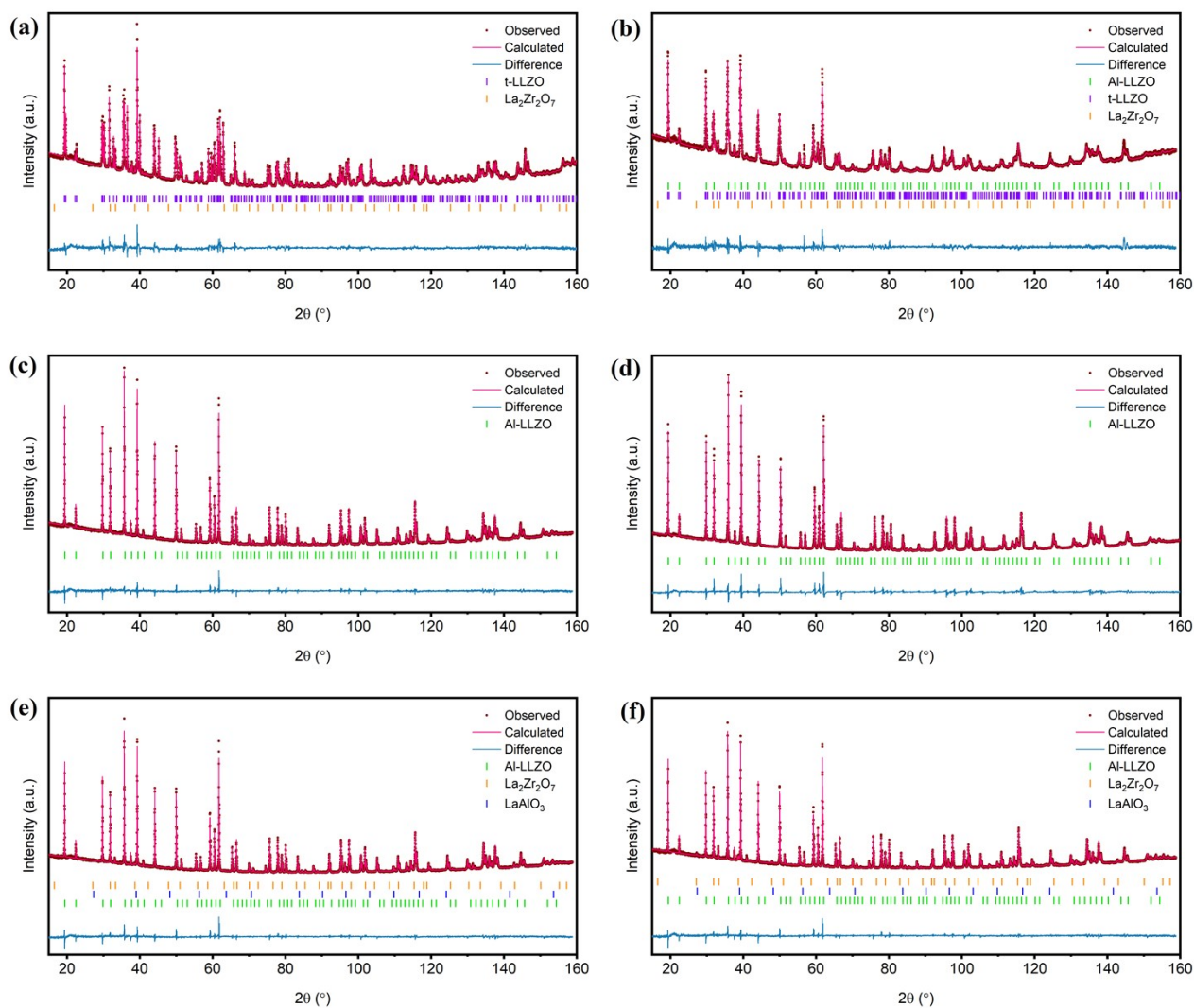


Figure S2. Rietveld refinement plots for calcined LLZO samples prepared by the SB method with an Al content (mol pfu) of (a) 0, (b) 0.12, (c) 0.18, (d) 0.24, (e) 0.3 and (f) 0.36.