

Supplementary Information

**Fusing Ta-Doped Li<sub>7</sub>La<sub>3</sub>Zr<sub>2</sub>O<sub>12</sub> Grains by Nanoscale Y<sub>2</sub>O<sub>3</sub> Sintering Aids  
for High-Performance Solid-State Lithium Batteries**

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**Table S1.** Stoichiometries of LLZTO pellets obtained from ICP-OES Tests.

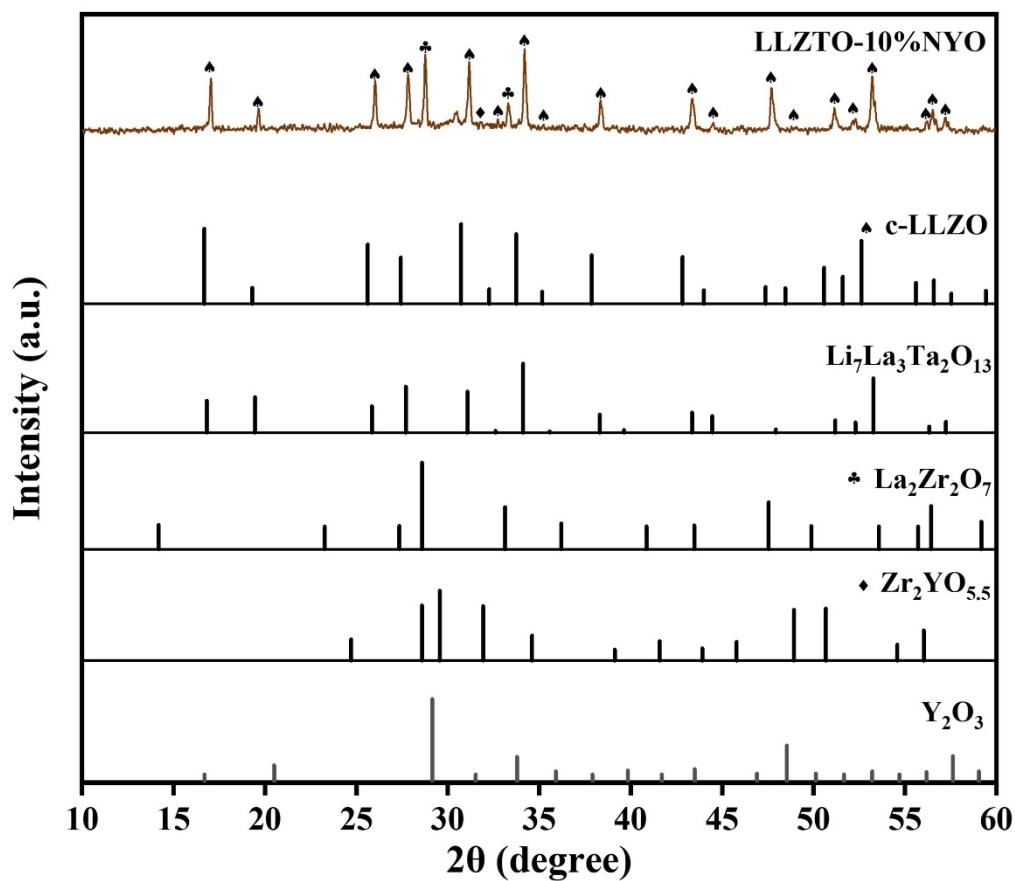
Samples	Li	La	Zr	Ta	Y	Stoichiometries
LLZTO-P	7.30	3.27	1.54	0.19	0.00	$\text{Li}_{7.3}\text{La}_{3.27}\text{Zr}_{1.54}\text{Ta}_{0.19}\text{O}_{12}$
LLZTO-1%NYO	6.83	3.23	1.54	0.23	0.08	$\text{Li}_{6.83}\text{La}_{3.23}\text{Zr}_{1.54}\text{Ta}_{0.23}\text{Y}_{0.08}\text{O}_{12}$
LLZTO-2%NYO	6.76	3.21	1.53	0.26	0.13	$\text{Li}_{6.76}\text{La}_{3.21}\text{Zr}_{1.53}\text{Ta}_{0.26}\text{Y}_{0.13}\text{O}_{12}$
LLZTO-5%NYO	6.84	3.19	1.52	0.30	0.22	$\text{Li}_{6.84}\text{La}_{3.19}\text{Zr}_{1.52}\text{Ta}_{0.30}\text{Y}_{0.22}\text{O}_{12}$
LLZTO-10%NYO	7.08	3.13	1.57	0.30	0.65	$\text{Li}_{7.08}\text{La}_{3.13}\text{Zr}_{1.57}\text{Ta}_{0.30}\text{Y}_{0.65}\text{O}_{12}$

**Table S2.** Size data for measuring relative density.

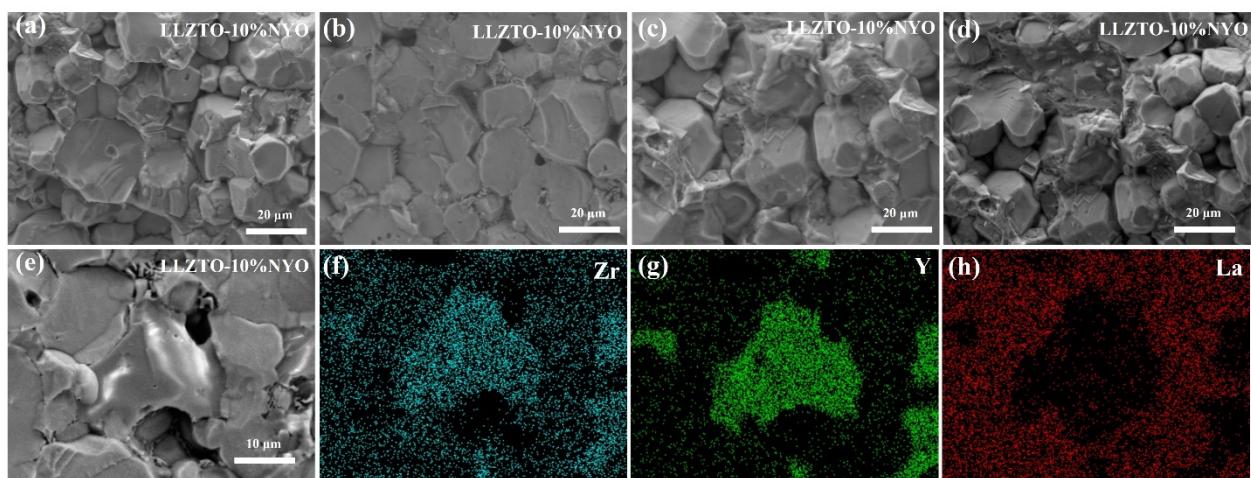
Samples	Quality (mg)	Thickness (mm)	Diameter (mm)	Density (g cm <sup>-3</sup> )	Relative density (%)
LLZTO-P	839.65	1.50	12.10	4.87	90.87
LLZTO-1%NYO	843.08	1.52	12.08	4.84	90.34
LLZTO-2%NYO	813.98	1.44	12.08	4.93	92.06
LLZTO-5%NYO	777.16	1.42	11.64	5.15	96.00
LLZTO-10%NYO	495.08	1.08	11.56	4.37	81.53

**Table S3.** Comparison of ionic conductivity with reported LLZTO electrolytes.

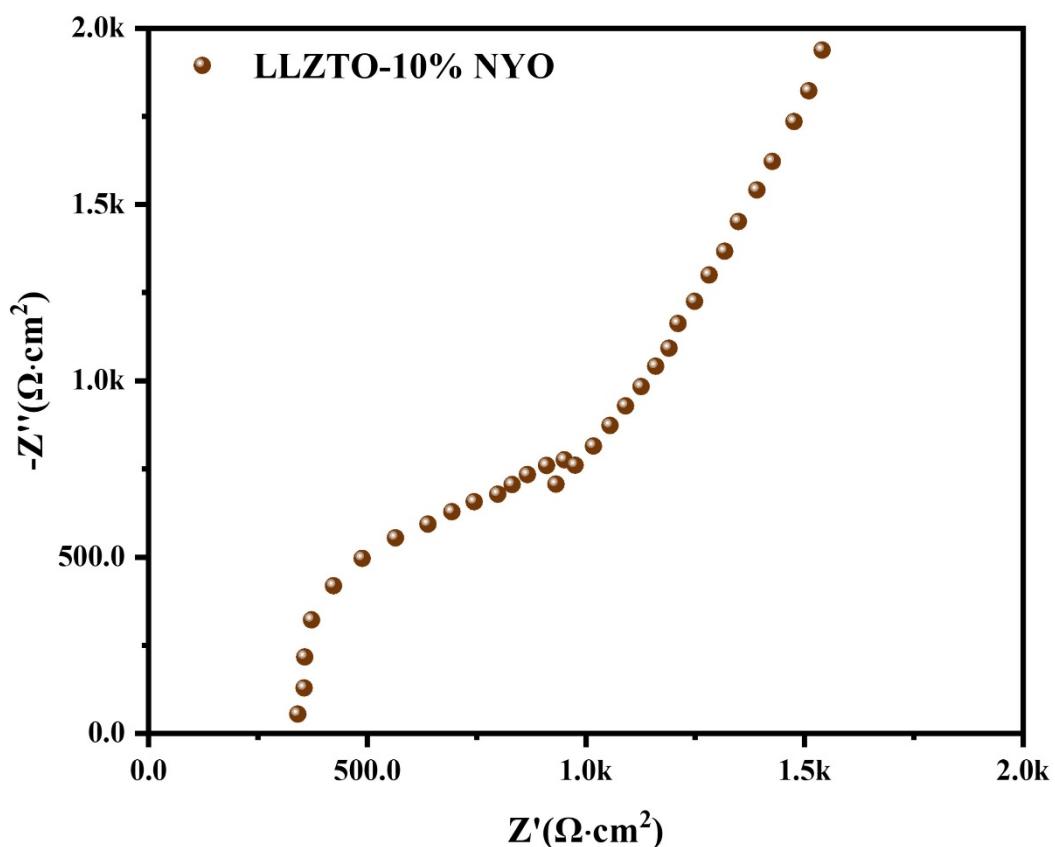
Composition	Conductivity (S cm <sup>-1</sup> )	Synthesis method	Reference
$\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$	$6.1 \times 10^{-4}$	Molten salt synthesis	1
10 mol% $\text{Al}_2\text{O}_3$ + $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$	$3.1 \times 10^{-4}$	$\text{Al}_2\text{O}_3$ sintering additive	2
5 wt% $\text{MgO}$ + $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$	$5.2 \times 10^{-4}$	$\text{MgO}$ sintering additive	3
6 wt% $\text{MgO}$ + $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$	$5.17 \times 10^{-4}$	$\text{MgO}$ sintering additive	4
1 mol% $\text{SiO}_2$ + $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$	$3.84 \times 10^{-4}$	$\text{SiO}_2$ sintering additive	5
$\text{SnO}_2$ coated $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$	$1.6 \times 10^{-3}$	Hot pressing and coating	6
$\text{Li}_{6.84}\text{La}_{3.19}\text{Zr}_{1.52}\text{Ta}_{0.30}\text{Y}_{0.22}\text{O}_{12}$	$7.39 \times 10^{-4}$	$\text{Y}_2\text{O}_3$ sintering additive	This work



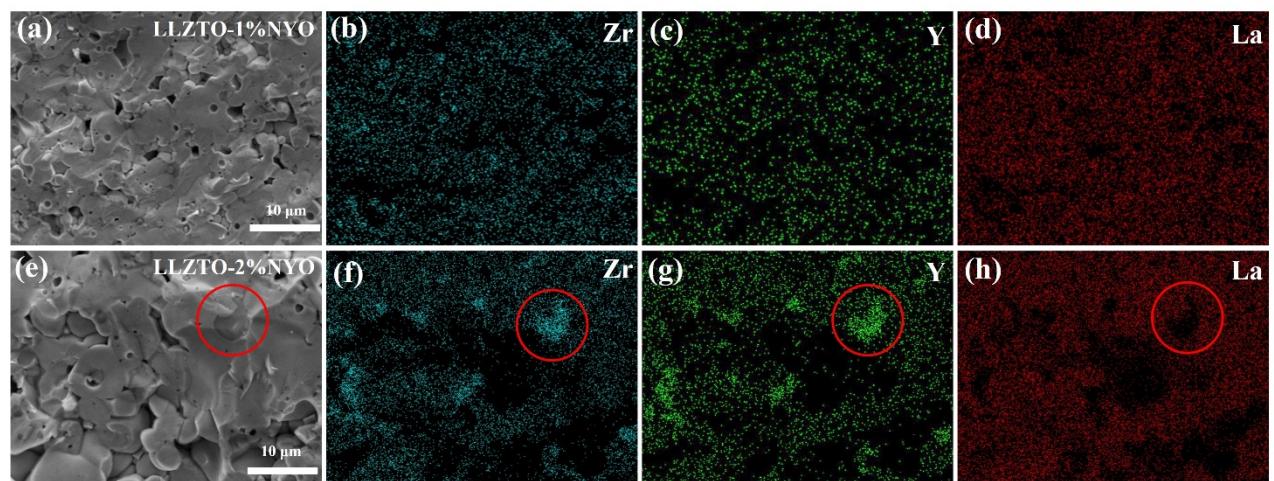
**Figure S1.** XRD patterns of LLZTO-10%NYO powder.



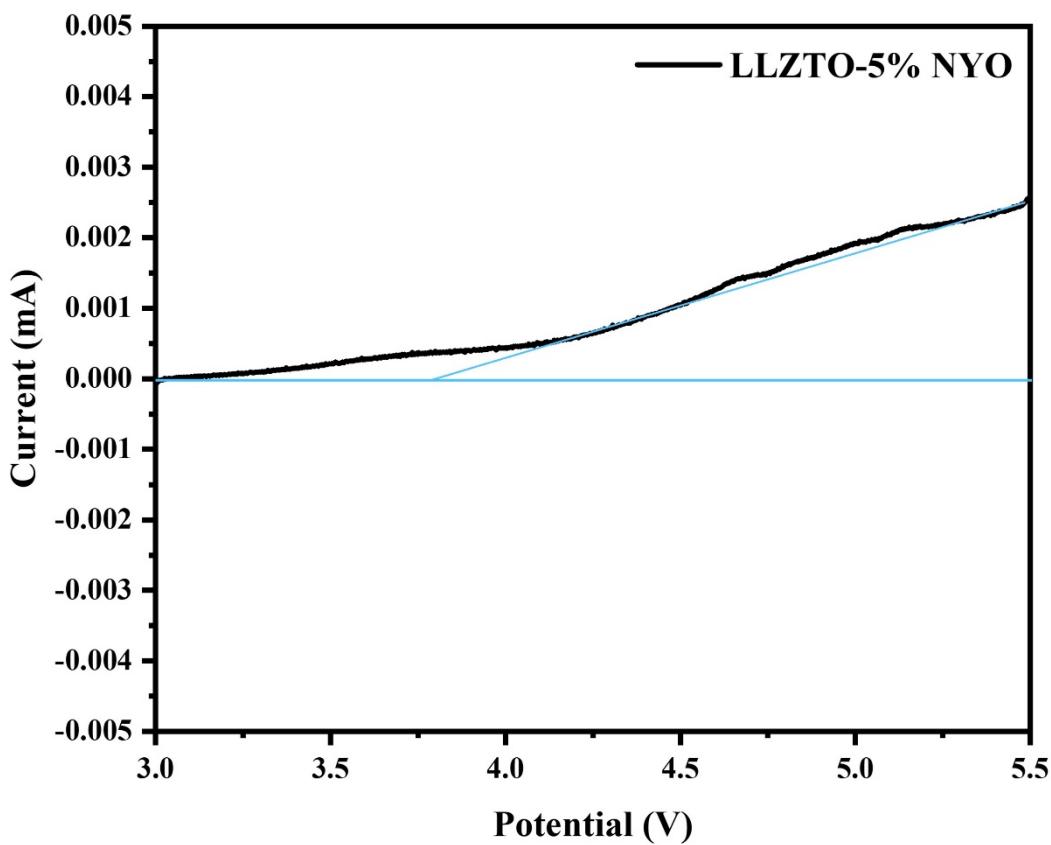
**Figure S2.** Cross-section SEM images (a-d) and elemental mappings (e-h) of LLZTO-10%NYO.



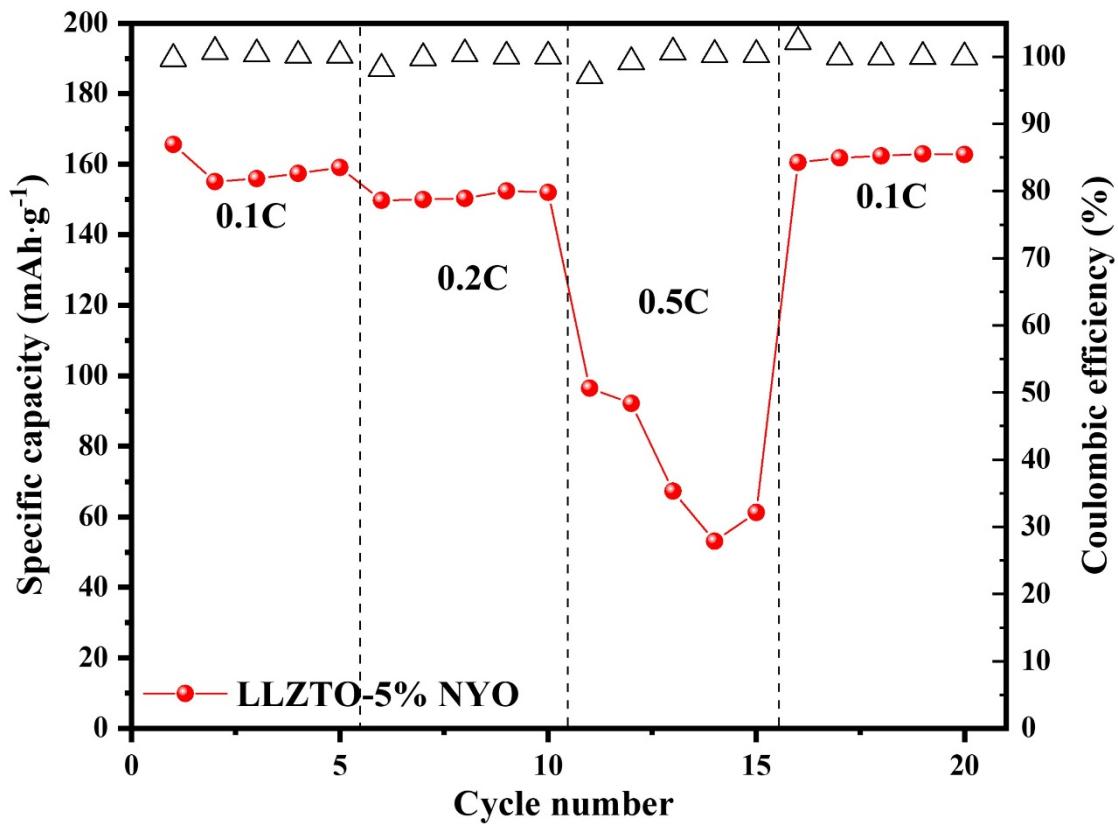
**Figure S3.** EIS plots of Ag|LLZTO-10%NYO|Ag cell.



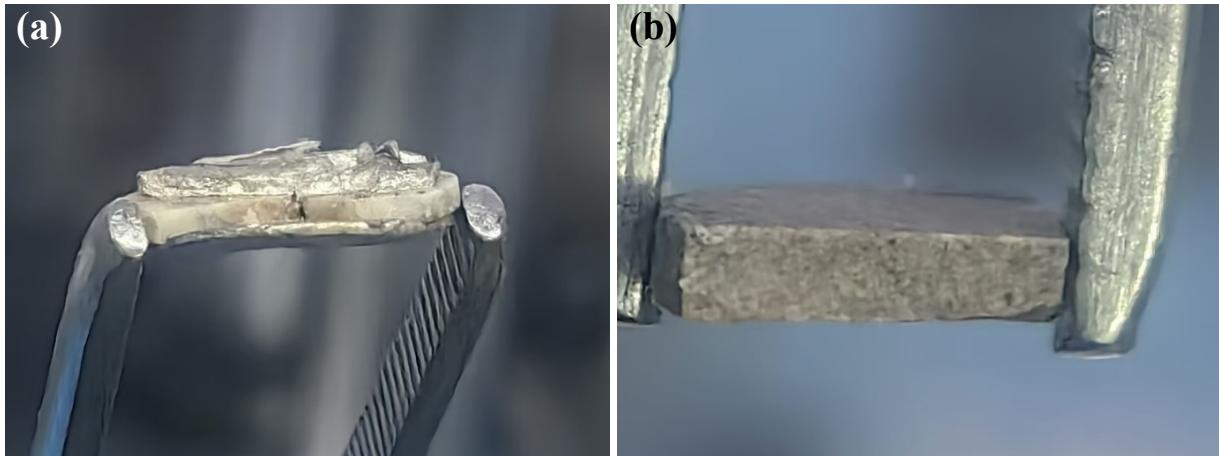
**Figure S4.** Elemental mappings of (a-d) LLZTO-1%NYO and (e-h) LLZTO-2%NYO pellets.



**Figure S5.** Linear scanning voltammetry (LSV) curve of SS|LLZTO-5%NYO|Li cell.



**Figure S6.** Rate Capability of LiFePO<sub>4</sub>|LLZTO-5%NYO|Li full cell.



**Figure S7.** Cross section photos of (a) LLZTO-P and (b) LLZTO-5%NYO pellets after cycling.

## References

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