Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information

Investigation of MgO additives on microstructure and properties of thin LLZO electrolytes for all-solid-state batteries

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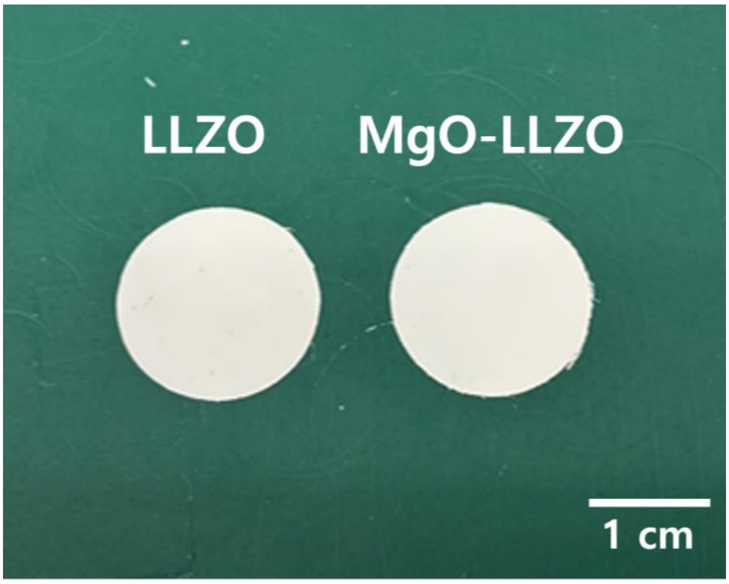

Figure S1. Digital photo of LLZO (left) and MgO-LLZO (right)..

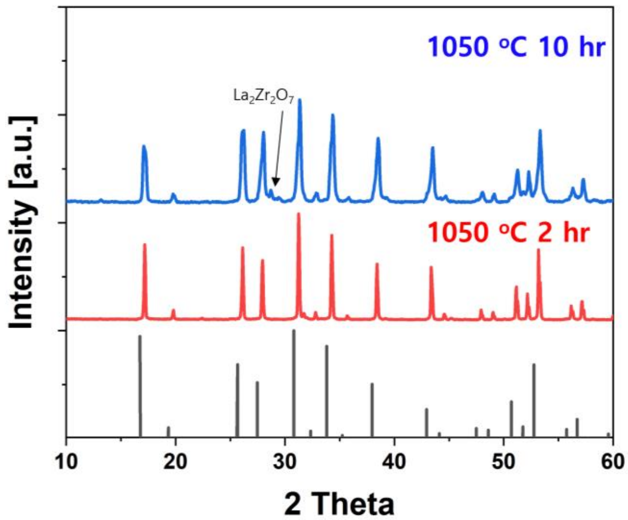

Figure S2. XRD peaks of LLZO at 2 hr (red) and 10 hr (blue).

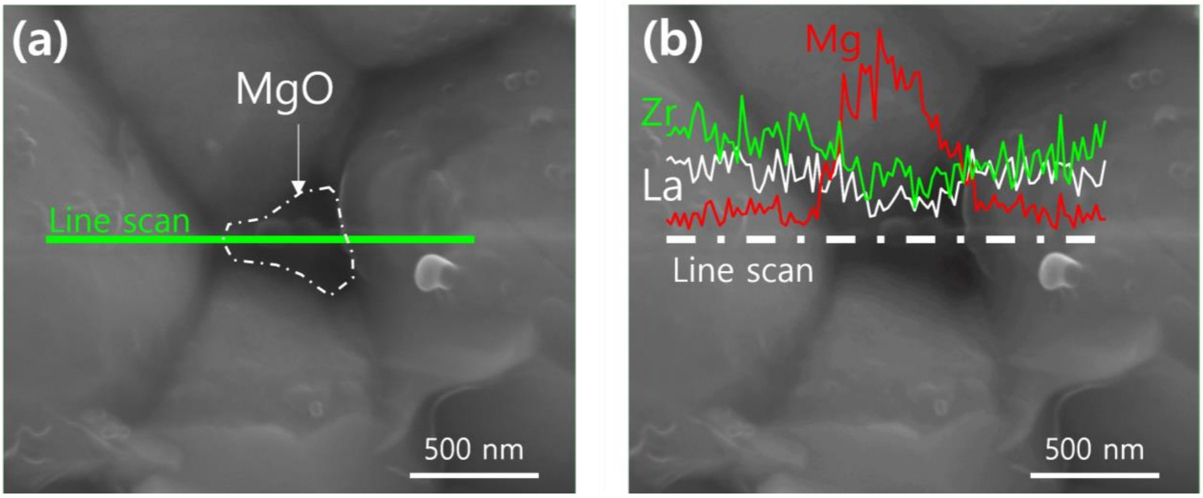

Figure S3. EDS line scan at LLZO and MgO grain boundary. (a) Location of line scan and (b) line scan results for Mg, Zr, and La elements.

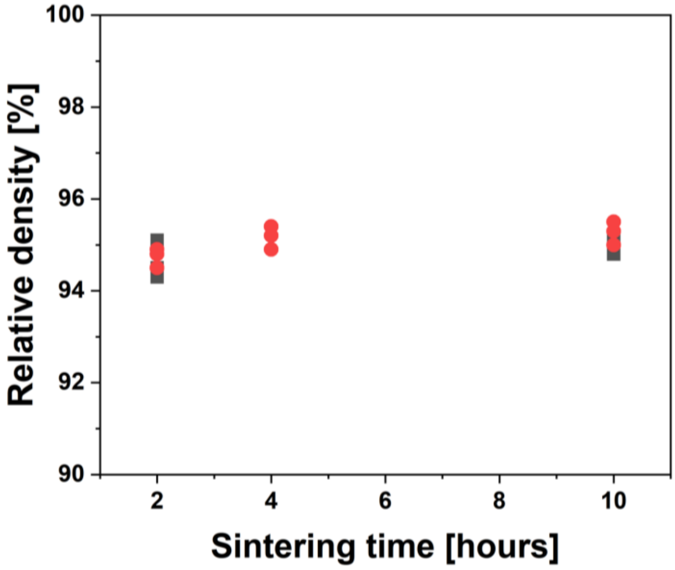

Figure S4. Densities of LLZO (Black) and MgO-LLZO (Red) sintered for different lengths of time at 1050 ℃.

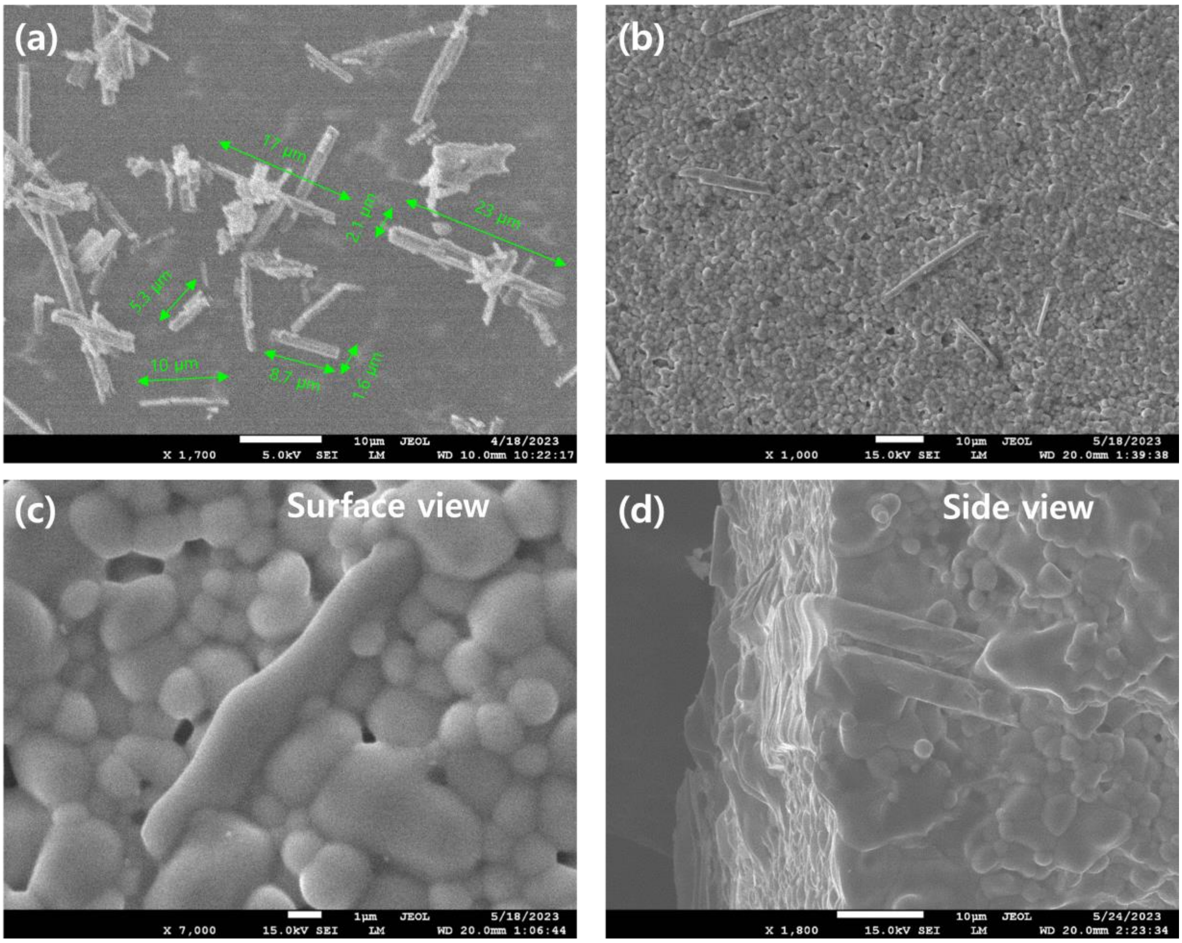

Figure S5. SEM images of (a) MgO fibers and (b-d) LLZO processed with MgO fibers sintered for 4 hours at 1050 °C (b-c surface views at different magnifications, d side view of fractured sample).

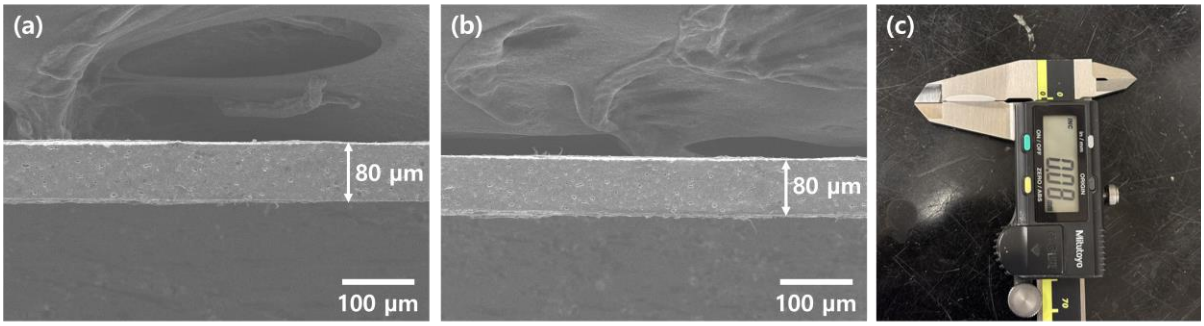

Figure S6. Cross-sectional SEM image of (a) LLZO and (b) MgO-LLZO. (c) Digital photo of LLZO showing its thickness using a vernier caliper.

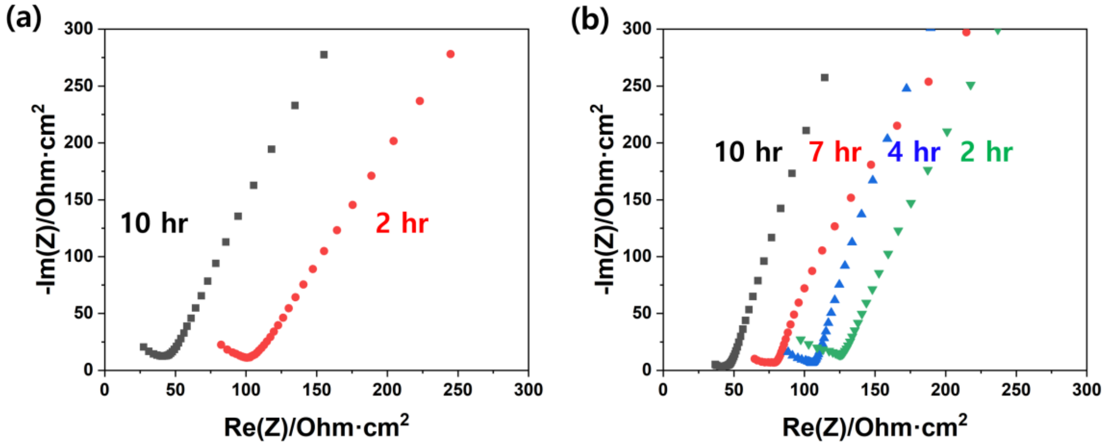

Figure S7. Nyquist plot of (a) LLZO and (b) MgO-LLZO

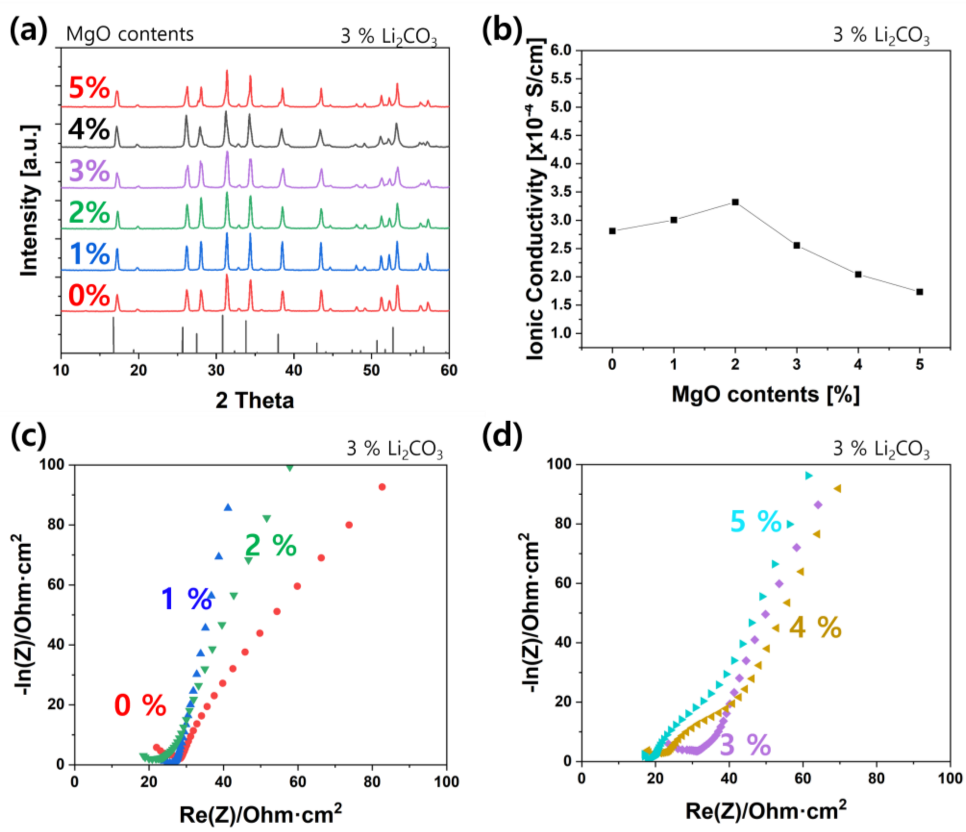

Figure S8. Characterization on LLZO processed with 3 % Li_2CO_3 and various amount of MgO (0-5 %) and sintered for 4 hours at 1080 °C. (a) XRD patterns, (b) Ionic conductivities, Nyquist plot of (c) 0-2 % MgO, and (d) 3-5 % MgO. This was a different batch of LLZO powder than that used in the main text, so the comparison is only qualitative.

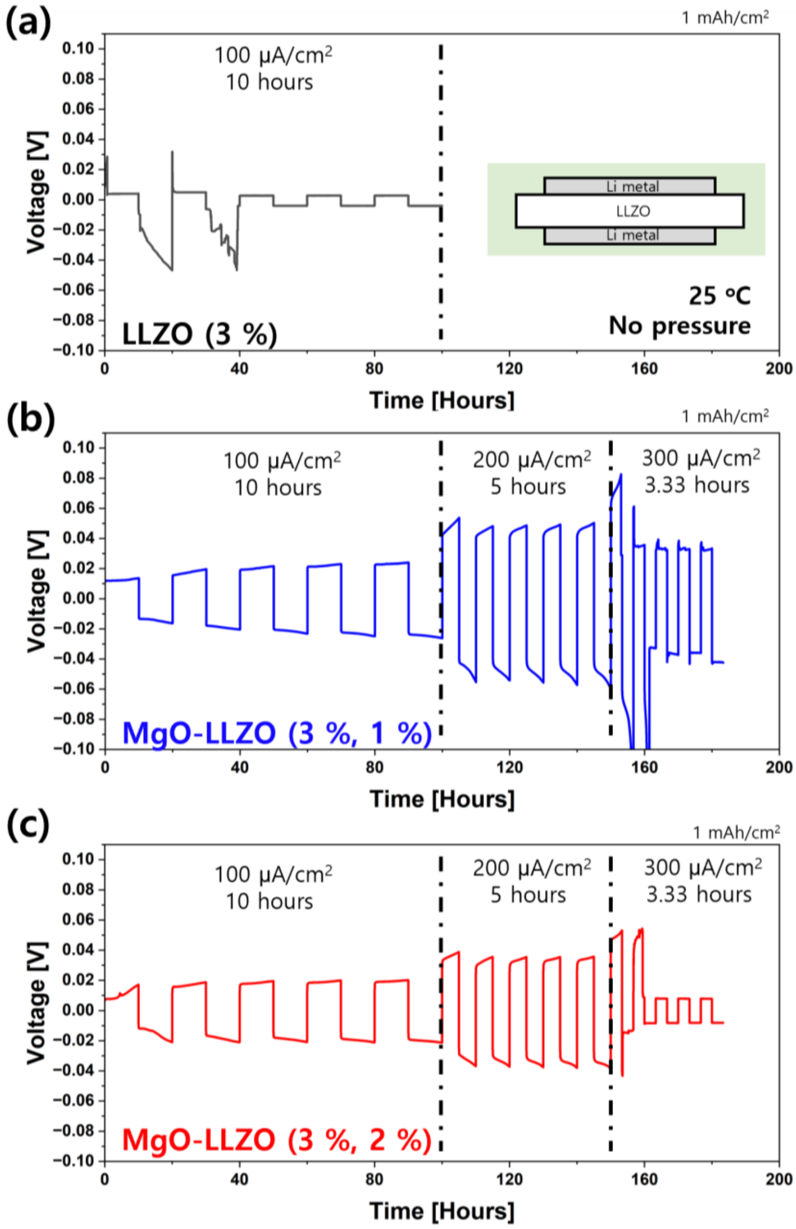

Figure S9. Symmetric cell cycling test using LLZO processed with 3 % Li_2CO_3 and various (0-2 %) MgO. (a) LLZO (Li_2CO_3 3%, no MgO), (b) MgO-LLZO (Li_2CO_3 3 %, MgO 1 %), and (c) MgO-LLZO (Li_2CO_3 3 %, MgO 2 %). Cycling at 25 ℃, no added pressure, no-liquid, and fixed areal capacity of 1 mAh/cm^2.

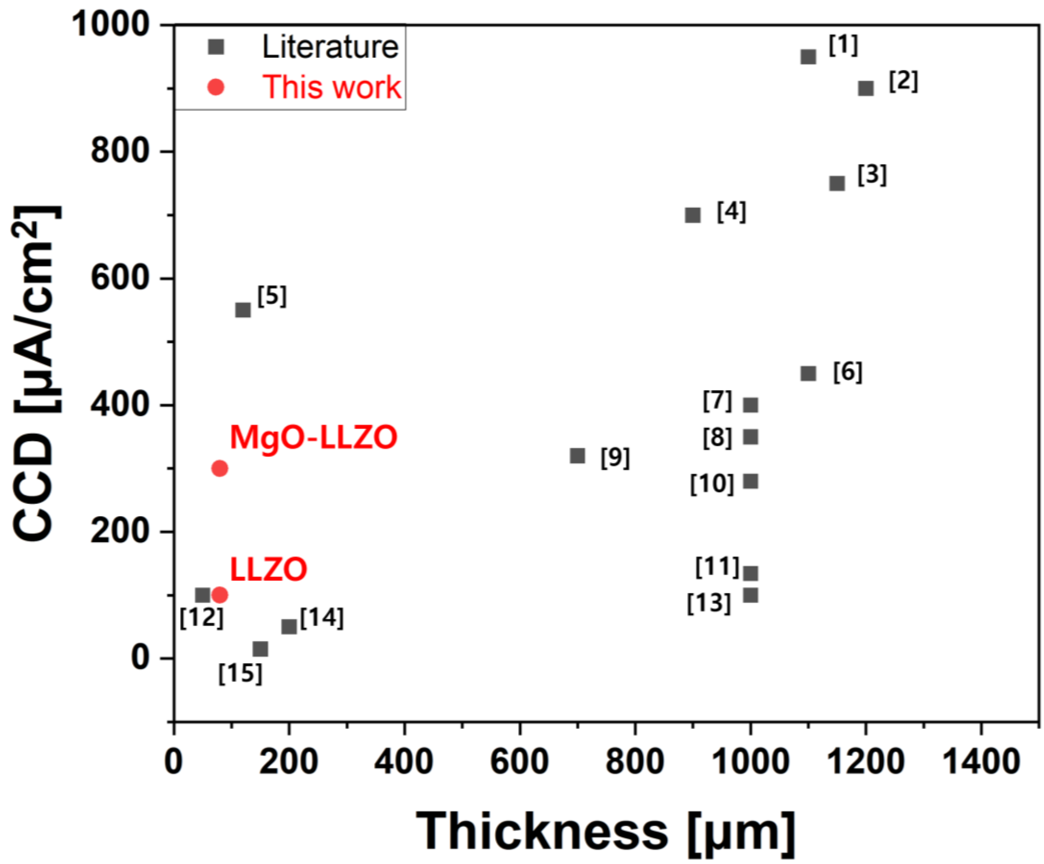

Figure S10. Summary of thicknesses of LLZO and CCD

Note that Ref [5] is previous work from our group, and the LLZO prepared there also included MgO powder. The higher CCD achieved in that work was due to the addition of surface texturing to increase the Li/LLZO contact area.

Table S1. Critical current density values reported in the literature.

Ref	CCD [μA/cm^2]	Areal Capacity [mAh/cm^2]*	Pressure [MPa]	Thickness [μm]**	Temp. [℃]
1	950	0.475	-	1000-1200	25
2	~900	0.2	3.4	1200	25
3	750	0.375	-	1100-1200	25
4	700	0.7	1.0	900	R.T.
5	500~600	0.5~0.6	No pressure	120	25
6	300~600	0.15~0.3	3.5	1100	R.T.
7	400	0.2	1.4	~1000	R.T.
8	350	0.175	0.4	1000	25
9	320	0.1	1.25	700	25
10	280	~0.14	No pressure	1000	20
This Work (with MgO)	**300**	**0.3**	**No pressure**	**80**	**25**
	200	**1.0**	**No pressure**	**80**	**25**
This Work (with MgO fiber)	**200**	**0.2**	**No pressure**	**80**	**25**
11	134	0.067	0.2	~1000	R.T.
This Work (no MgO)	**100**	**0.1**	**No pressure**	**80**	**25**
12	>100	0.1	No pressure	50	R.T.
13	100	0.05	0.32	1000	R.T.
14	50	0.05	-	200	60
15	15	0.075	-	150	80

* For each cycle, ** Thickness of LLZO electrolyte

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