Supporting Information

Lithium-ion transport in inorganic active fillers used in PEO-based composite solid electrolyte sheets

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Figure S1. FE-SEM image and particle size distribution exhibited by milling-30min and milling-80min (a) and (b)

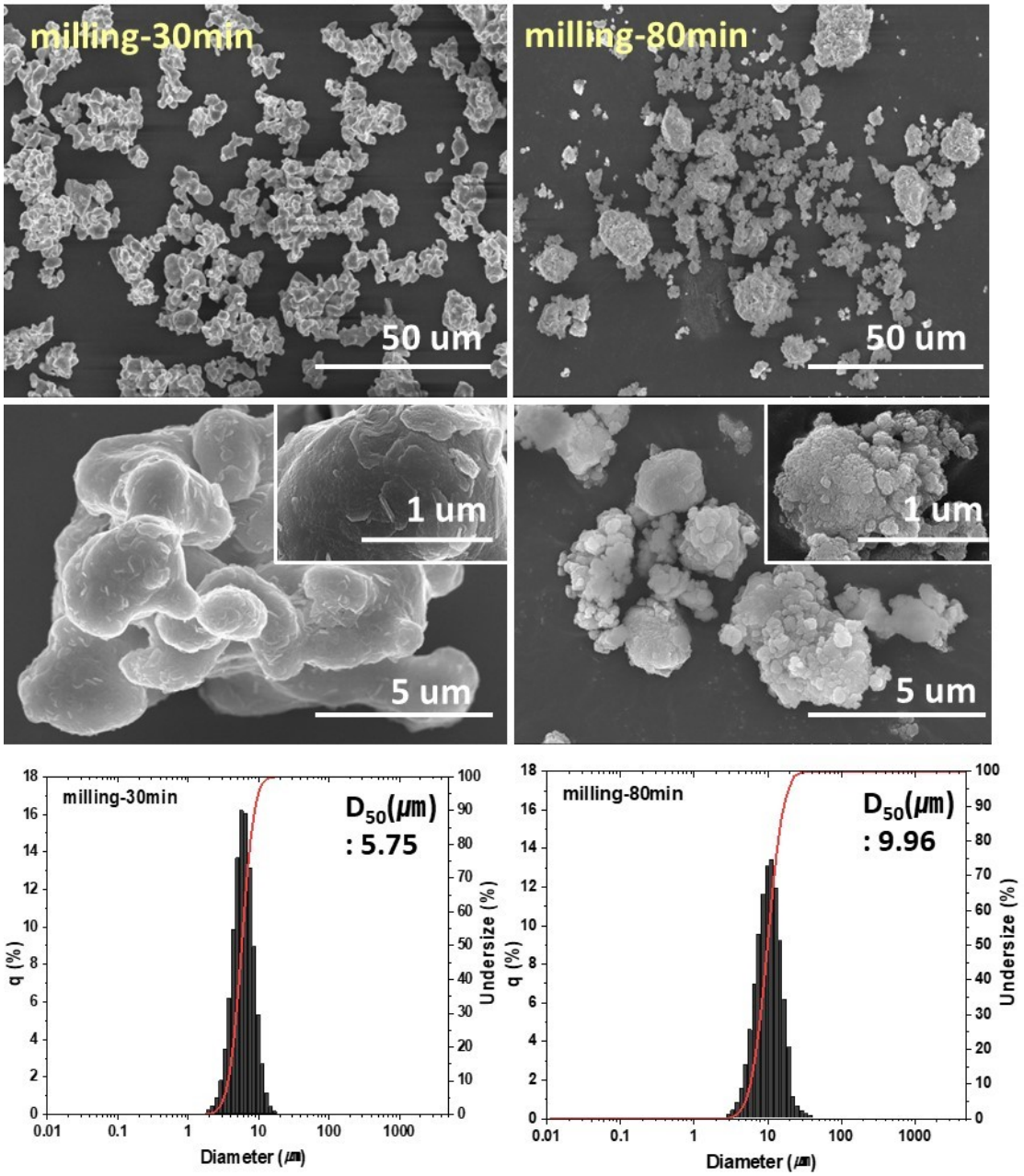

Figure S2. FE-SEM and EDX mapping images of LLZTO CSE and BM-LLZTO CSE

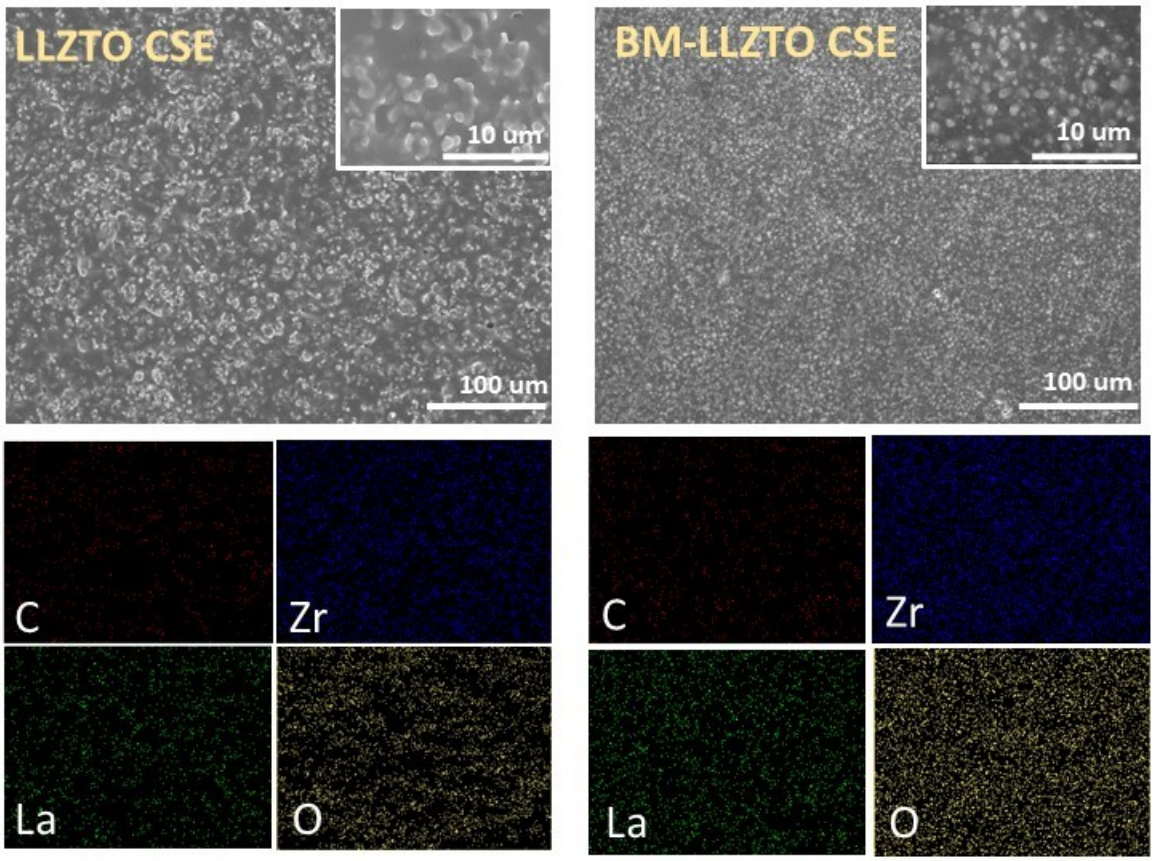

Figure S3. Stress–strain curves of PEO-Li salt and BM-LLZTO CSE

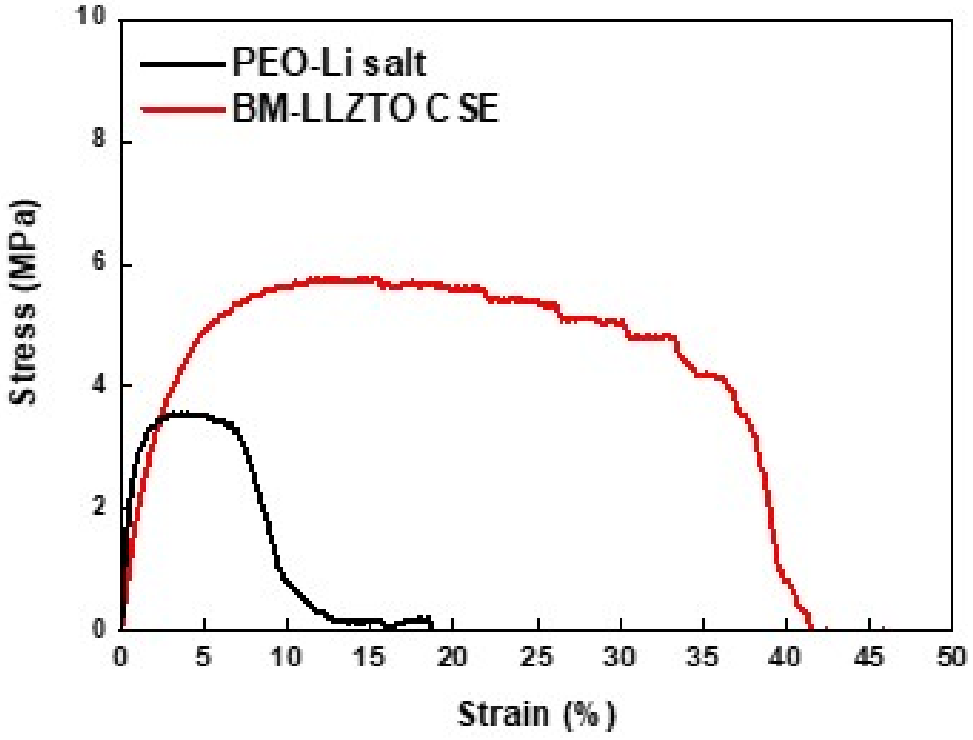

Figure S4. Current evolution under a polarization voltage of 10 mV milling-30min and milling-80min (a) and (b), total ionic conductivity, Lithium ionic conductivity and Lithium transference number curves at 70°C according to milling time

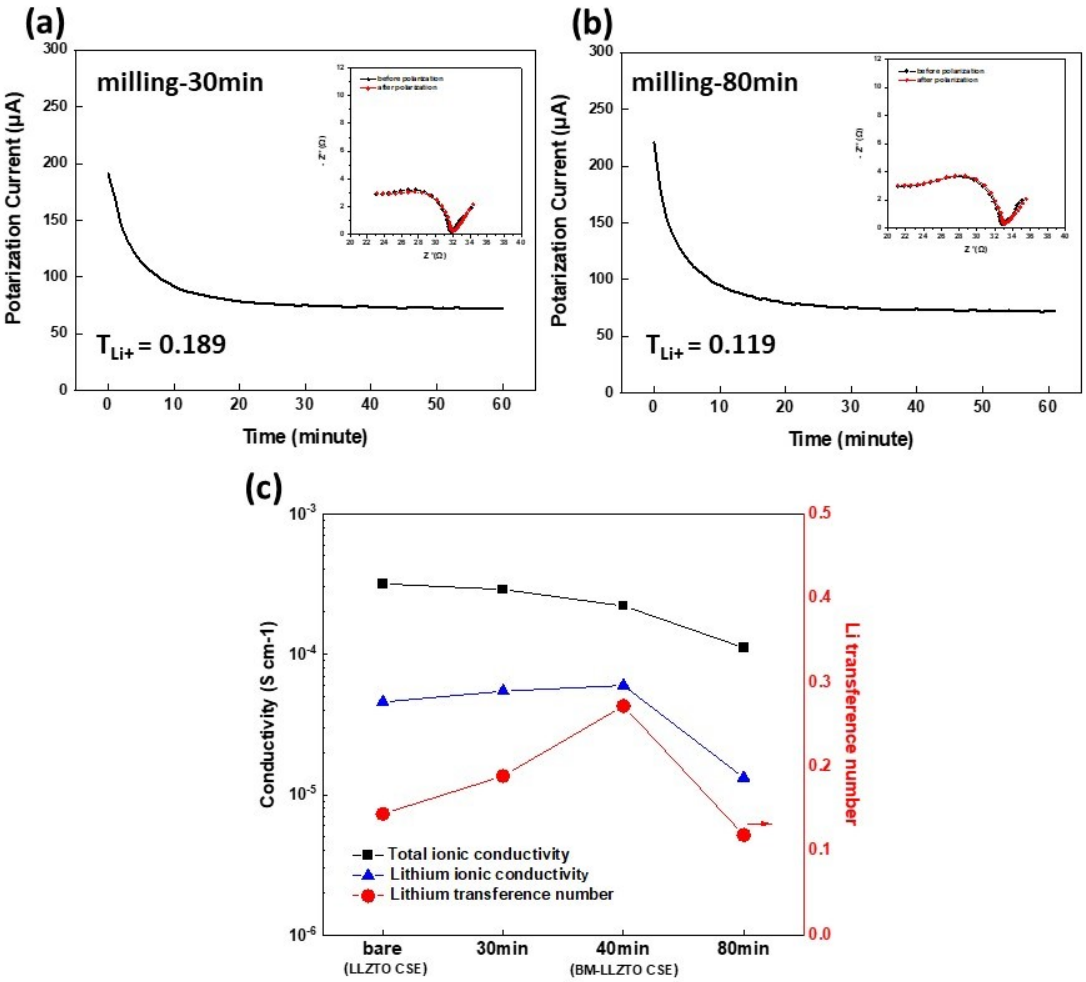

Figure S5. Characteristics comparisons of "Polymer-in-ceramic" CSE.

Composition of CSE		Active material	σ_{total} (S cm^{-1})	T_{Li+}	Potential range (V)	Capacity (mAh g^{-1})	Ref
Solid electrolyte	Polymer						
BM-LLZTO CSE ($Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ 70wt%)	PEO	$LiFePO_4$	2.2×10^{-4} (70℃)	0.272	5.0	153.3 (80μm) 144.4 (60μm)	This work
CSPE0 (Al-doped $Li_7La_3Zr_2O_{12}$ 70wt%)	PEO	$LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$	8.5×10^{-5} (60℃)	0.290	5.2	121 (160μm)	[S1]
$Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ (80wt%)	PEO/PEG	$LiFePO_4$	5.2×10^{-4} (80℃)	-	5.0	127.7 (100μm)	[24]
$Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ (20/80/20vol%)	PEO	LiFePO4	9.1×10^{-5} (55℃)	-	5.0	118.6 (N/A)	[S2]
LAGP-70 ($Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$ 70wt%)	PBA	$LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$	2.0×10^{-4} (65℃)	-	4.9	169.5 (N/A)	[S3]

[S1] J. H. Cha, P. N. Didwal, J. M. Kim, D. R. Chang and C. J. Park, *Journal of Membrane Science,* 2020, **595** 117538, DOI: 10.1016/j.memsci.2019.117538

[S2] H. Huo, Y. Chen, J. Luo, X. Yang, X. Guo and X. Sun, Adv. Energy Mater., 2019, **9**, 1804004, DOI: 10.1002/aenm.201804004

[S3] M. S. Park, Y. C. Jung and D. W. Kim, *Solid State Ionics,* 2018, **315**, 65–70, DOI: 10.1016/j.ssi.2017.12.007