Energy Materials

Supplementary Materials

Residual alkali effect in LLZO-modified composite solid polymer electrolyte

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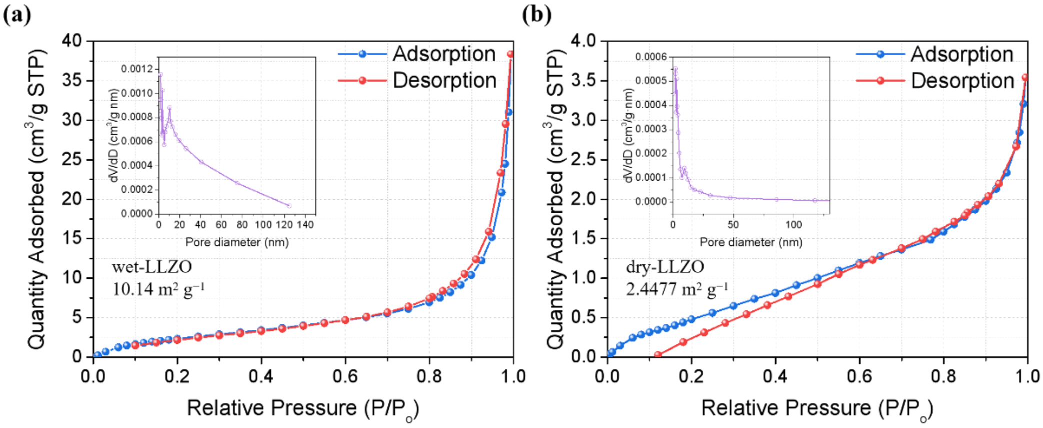
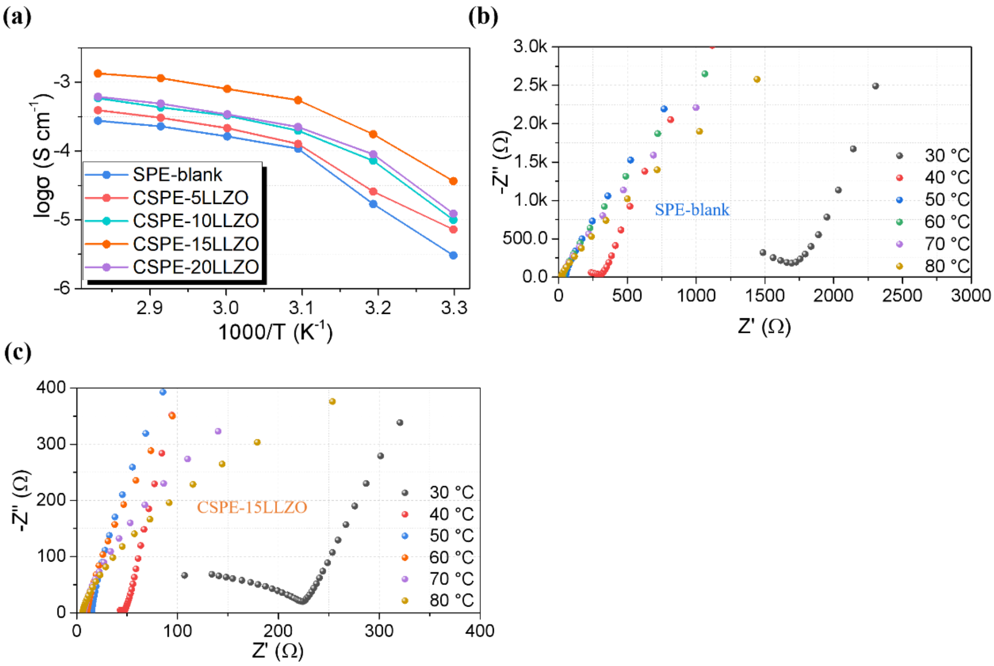
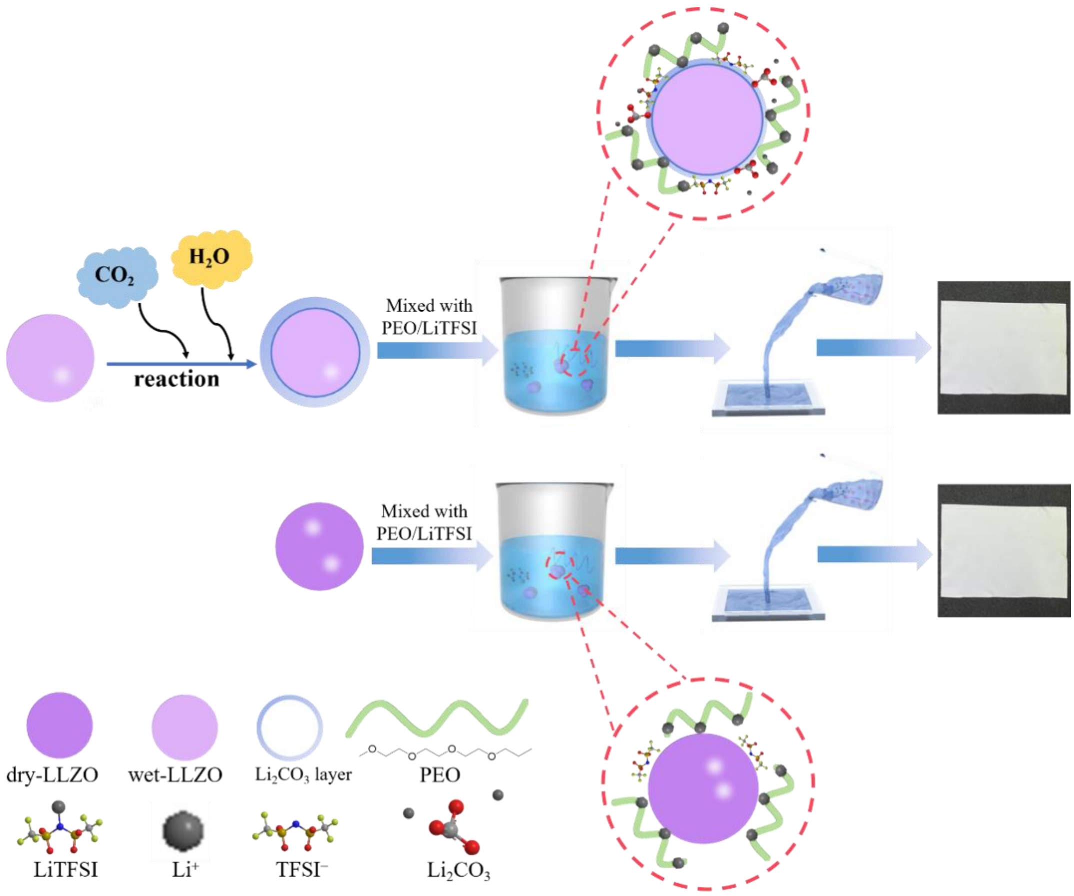


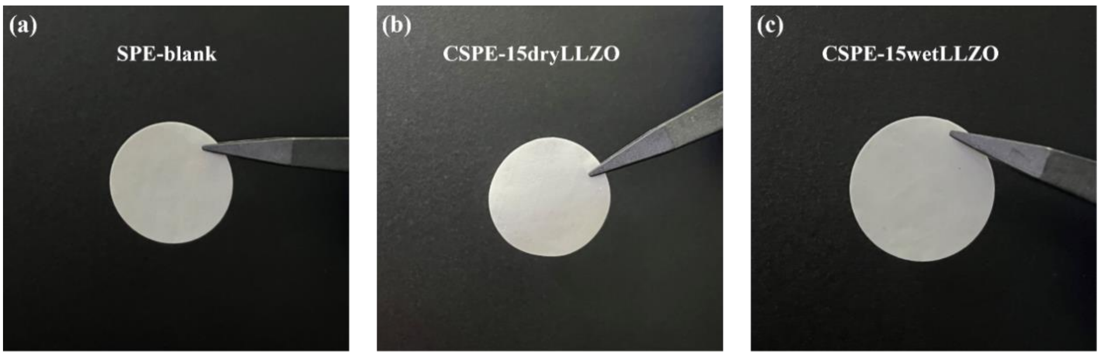
Supplementary Figure 1. (a) N_2 adsorption-desorption isotherm of wet-LLZO (the inset shows pore size distribution), (b) N_2 adsorption-desorption isotherm of dry-LLZO (the inset shows pore size distribution).



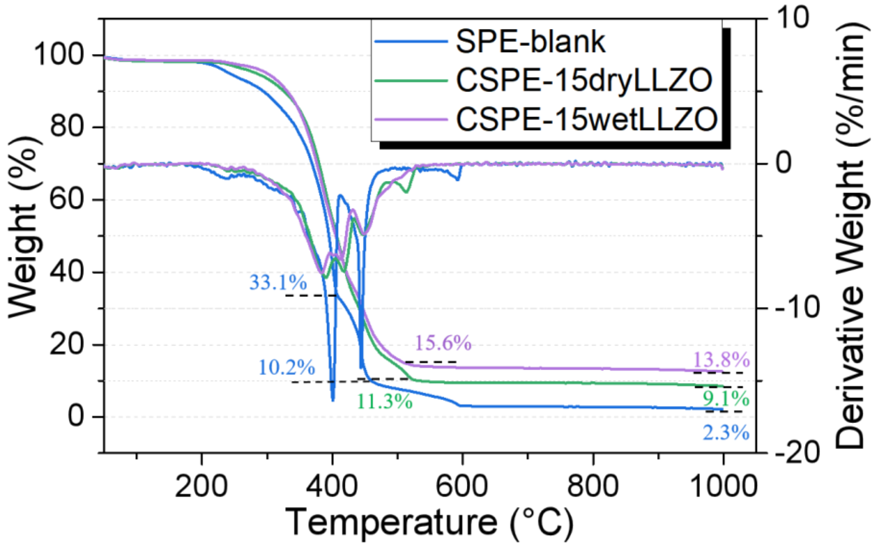
Supplementary Figure 2. (a) Ionic conductivity curves of CSPE-xLLZO (x=5, 10, 15 and 20), (b-c) EIS plots of SS/SPE-blank/SS, and SS/CSPE-15LLZO/SS (30 °C - 80 °C).



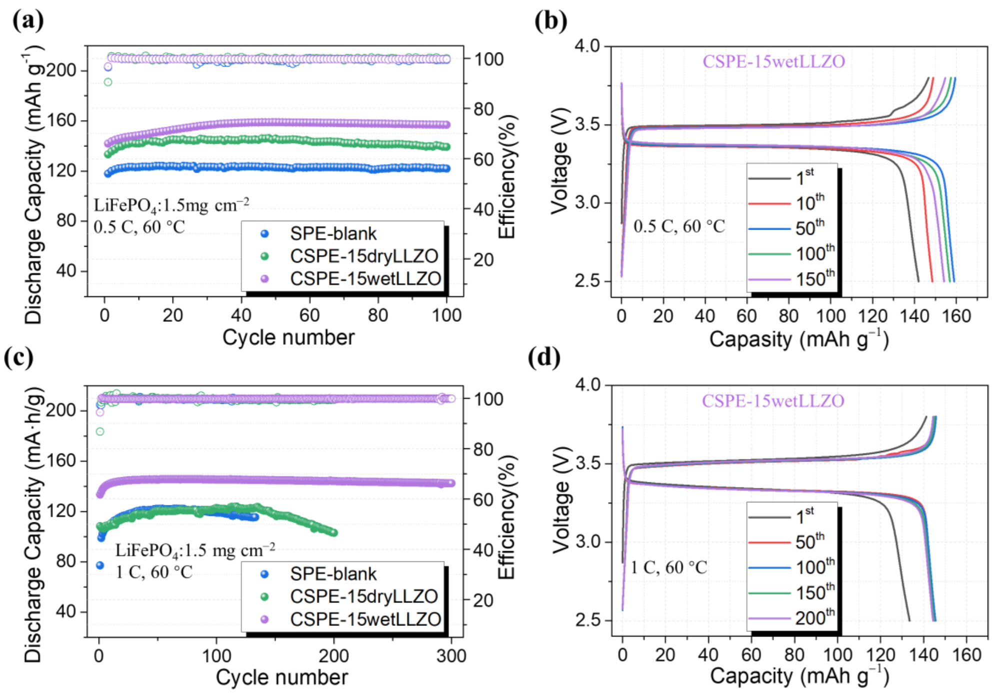
Supplementary Figure 3. Preparation process and mechanism diagram of PEO composite electrolyte with LLZO fillers containing different surface residual alkalis.



Supplementary Figure 4. (a-c) Digital photo images of SPE-blank, CSPE-15dryLLZO and CSPE-15wetLLZO.



Supplementary Figure 5. The TGA and DTG curves of SPE-blank, CSPE-15dryLLZO and CSPE-15wetLLZO.



Supplementary Figure 6. (a) Long-cycle performance of Li/SPEs/$LiFePO_4$ batteries at 0.5 C, (b) Charge-discharge curves of Li/CSPE-15wetLLZO/$LiFePO_4$ batteries at 0.5 C across different cycles, (c) Long-cycle performance of Li/SPEs/$LiFePO_4$

batteries at 1.0 C, (d) Charge-discharge curves of Li/CSPE-15wetLLZO/$LiFePO_4$ batteries at 1.0 C across different cycles.

Experimental Drug

The experimental drugs and chemical reagents used in this paper are shown in Table 1.

Table 1. Experimental chemicals and reagents required for the experiment

Reagent Name	Simplest chemical formula	Specification	Manufacturer
Lithium Hydroxide Monohydrate	$LiOH{\cdot}H_2O$	Analytical- grade	Aladdin Reagent (Shanghai) Co., Ltd.
Zirconia	ZrO_2	99.5%	Nanjing Mingshan New Material Technology Co., Ltd.
Gallium oxide	Ga_2O_3	99.99%	Aladdin Reagent (Shanghai) Co., Ltd.
Tantalum oxide	Ta_2O_5	99.5%	Aladdin Reagent (Shanghai) Co., Ltd.
Lanthanum oxide	La_2O_3	99.5%	Sinopharm Chemical Reagent Co., Ltd.
Isopropanol	C_3H_8O	99.7%	Xilong Scientific

			Co., Ltd.
Magnesium oxide crucible	MgO	99.9%	Foshan Kesheng New Materials Co., Ltd.
Lithium bis (trifluoromethanesulfonyl) imide	LiTFSI	≥99%	Aladdin Reagent (Shanghai) Co., Ltd.
Poly (ethylene oxide)	PEO	AR, Mw=600000	Aladdin Reagent (Shanghai) Co., Ltd.
Acetonitrile	CH_3CN	99.9%, water≤10ppm	Anneji (Shanghai) Pharmaceutical Chemical Co., Ltd.
Polytetrafluoroethylene mold	PTFE	6 × 8 cm	Shenzhen Yimei Hardware & Plastic Products Co., Ltd.
Lithium foil	Li	battery-grade	China Energy Lithium Co., Ltd.
steel foil	S	Φ= 16 mm	Wuhan Lixing (Torch) Power Sources Co., Ltd.
battery case	CR2032、CR2016	99.99%	Wuhan Lixing (Torch) Power Sources Co., Ltd.
Lithium iron phosphate	$LiFePO_4$	Battery- grade	Canrd Technology Co., Ltd.

Conductive carbon black	Super P	Battery- grade	Shenzhen Kejing STAR Technology Company
Polyvinylidene fluoride	PVDF	Battery- grade	Zhangjiagang Guotai Huarong New Chemical Materials Co., Ltd.
N-Methyl-2-pyrrolidone	NMP	AR	Tianjin Kemiou Chemical Reagent Co., Ltd.
High-purity nitrogen	N_2	99.99%	Shenzhen Huatepeng Special Gases Co., Ltd.

Laboratory apparatus

Detailed information about the experimental equipment and instruments used in this paper is presented in Table 2.

Table 2 Experimental Instruments and Equipment

Instrument and Equipment Names	Model	Manufacturer
Glove box	Vigor V-lab	Vigor Gas Purification Technologies (Suzhou) Co., Ltd.
Blast drying oven	BPG-9040A	Shanghai Yiheng Technology Instrument Co., Ltd.
Muffle furnace	KSL-1400A3	Henan NOBODY Materials Science & Technology Co.,

		Ltd.
Planetary ball mill	MTR-YXOM-2L	Changsha Mitr Instrument Equipment Co., Ltd.
High-precision electronic balance	FA2004B	Shanghai Precision Instruments Co., Ltd.
Ultrasonic cleaner	KQ 3200DE	Kunshan Ultrasonic Instruments Co., Ltd.
Syringe	10 ml	Gan Food and Drug Administration Medical Device
Coin cell crimping machine	MSK-PN110-S	Shenzhen Kejing Star Technology Co., Ltd.
Electrode slicer	MSK-T10	Shenzhen Kejing Star Technology Co., Ltd.
Electrochemical workstation	IVIUMnSTAT	Ivium Technologies B.V., Eindhoven, Netherlands.
Battery test system	CT-4008-5V10mA 164	Neware Technology Limited
Differential Scanning Calorimeter	DSC-200 F3	NETZSCH Scientific Instruments (Shanghai) Trading Ltd.
Scanning Electron Microscope	TESCAN-MIRA3	TESCAN Brno, s.r.o., Brno, Czech Republic
Ion sputtering coater	JS—1600M	Nanjing Saintins Laboratory Equipment Co., Ltd.
Thermogravimetric Analyzer	STA 449F5	NETZSCH Scientific Instruments (Shanghai) Trading Ltd.

X-ray diffractometer	Rigaku Ultima IV	Rigaku Corporation, Tokyo, Japan
Fourier Transform Infrared Spectrometer	VERTEX 70V	Bruker Corporation
Raman spectrometer	alpha300R, 532nm laser excitation	WITec GmbH, Ulm, Germany
Electronic Universal Testing Machine	INSTRON 3343	Instron Corporation, Norwood, MA, USA.
X-ray photoelectron spectroscopy	PHI 5000 VersaProbeIII	ULVAC-PHI
Li NMR Spectrometer	Ascend 400	Bruker BioSpin GmbH & Co. KG (Germany)

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