Supporting Information

Fe-P-S electrodes for all-solid-state lithium secondary batteries using sulfide-based solid electrolytes

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Table S1. Properties of all-solid-state lithium sulfur batteries using sulfur or sulfur composite active materials.

Active material	Weight of active material / mg	Carbon content / wt%	Temperature / °C	C-rate / C	Discharge Capacity / mAh $g^{-1}_{(Active\ material)}$	Discharge Capacity / mAh	Energy density* / Wh $cm^{-3}_{(Composite\ electrode)}$	Reference
Cu-S	3.8	6	R.T.	0.02	650	2.5 (at 20th cycle)	~1.1 (at 20th cycle)	[1]
Sulfur	1.6	20	25	0.02	1050	1.6 (at 50th cycle)	~2.1 (at 1st cycle)	[2]
Sulfur	0.7	37	25	0.15	500	0.33 (at 10th cycle)	~0.90 (at 1st cycle)	[3]
Sulfur	3.8	10	25	0.18	~1500	~5.6 (at 100 cycle)	~1.5 (at 1st cycle)	[4]
Sulfur	1.5	10	25	0.1	1288	1.9 (at 1st cycle)	~2.1 (at 1st cycle)	[5]
FeS_2-S	6.6	15	25	0.02	~710	~4.7 (at 5th cycle)	~0.99 (at 5st cycle)	[6]
Fe-P-S	6.9	2	100	~0.1	625	4.3 (at 50th cycle)	~2.2 (at 50th cycle)	This study

*The energy density per the volume of the composite electrodes is calculated by assuming that each density of the solid electrolyte and the carbon additives was 1.9 g cm^{-3} and 1.8 g cm^{-3}, respectively, and molecular weight of the solid electrolytes is 180 g mol^{-1}. This molecular weight corresponds to that of Li_3PS_4. The average discharge voltages, which are used for the calculation, are assumed to be the voltages at the middle point of discharge capacity per the weight of the active materials. In the calculation of the energy density, we assumed lithium metal is used as the negative electrodes.

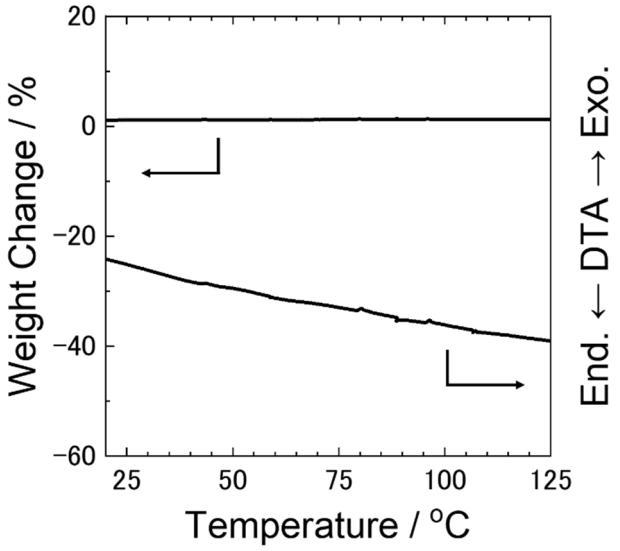

Figure S1. TG-DAT curves of the 70$FePS_3$·30S electrode measured at a heating rate of 5 ºC min^{-1}.

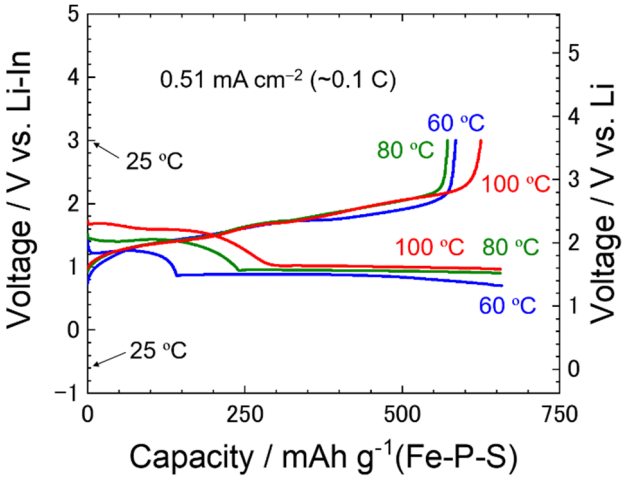

Figure S2. First discharge-charge curves of all-solid-state batteries using the $70FePS_3 \cdot 30S$ electrode at 25 °C (black lines), 60 °C (blue lines), 80 °C (green lines), 100 °C (red lines) under a current density of 0.51 mA cm^{-2} (~0.1 C). First, the battery was discharged to the capacity of 656 mAh g^{-1}; this was calculated based on the theoretical capacities of $FePS_3$ (220 mAh g^{-1}) and sulfur (1672 mAh g^{-1}). The cut-off voltage was set to –0.62 V vs. Li-In for discharging and 3.0 V vs. Li-In for charging.

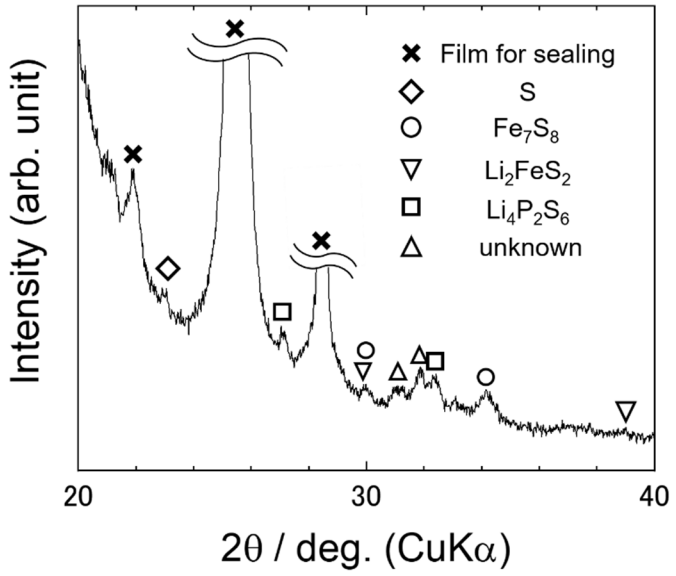

Figure S3. XRD pattern of the $70FePS_3 \cdot 30S$ composite electrode after the charge cycles.

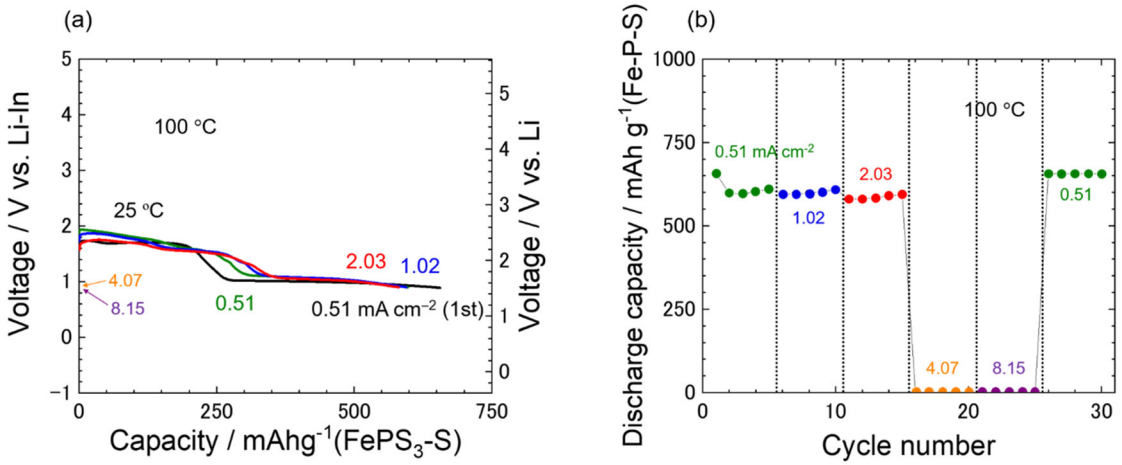

Figure S4. (a) Discharge curves and (b) cycle performance of all-solid-state battery using the $70FePS_3 \cdot 30S$ electrode at 100 °C under the constant current densities from 0.51 to 8.15 mA cm^{-2} for discharging. First, the battery was discharged to the capacity of 656 mAh g^{-1} under the cut-off voltage of 0.9 V vs. Li-In. For charging, the current density was set to 0.51 mA cm^{-2}, and the cut-off voltage was set to 3.0 V vs. Li-In.

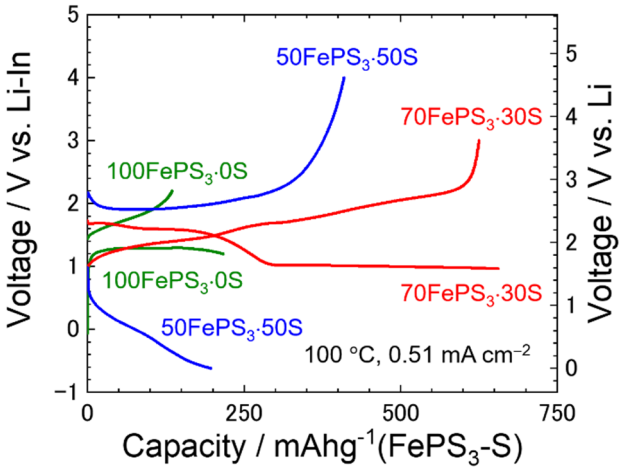

Figure S5. First discharge-charge curves of all-solid-state batteries using the Fe-P-S electrodes of 100$FePS_3$·0S (green lines), 70$FePS_3$·30S (red lines), and 50$FePS_3$·50S (blue lines) at 100 °C under a current density of 0.51 mA cm^{-2}. First, the batteries of 100$FePS_3$·0S, 70$FePS_3$·30S and 50$FePS_3$·50S were discharged to the theoretical capacities of 220, 656, and 946 mAh g^{-1}, respectively; these were calculated based on the theoretical capacities of $FePS_3$ (220 mAh g^{-1}) and sulfur (1672 mAh g^{-1}). The cut-off voltage was set to 0.62 V vs. Li-In for discharging. For charging, the cut-off voltages of 100$FePS_3$·0S, 70$FePS_3$·30S and 50$FePS_3$·50S were set to 2.2, 3.0, and 4.0 V vs. Li-In respectively.

References

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