Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Polysulfide entrapment and retardation in gel electrolyte Li-S batteries: experiments and modeling

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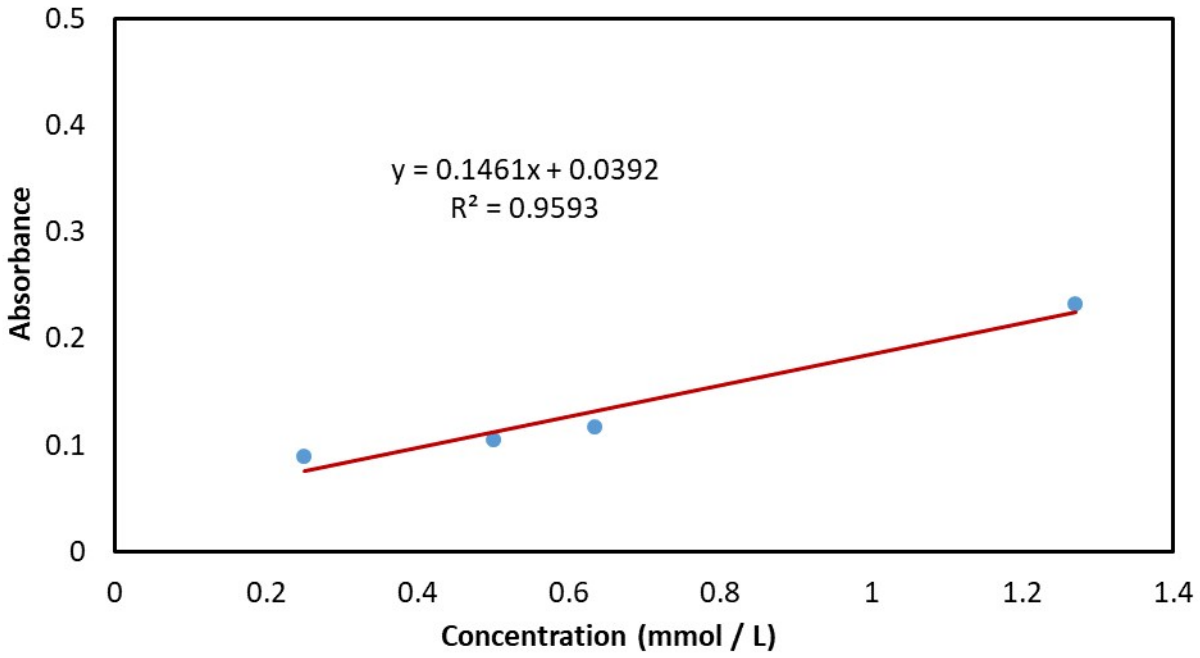
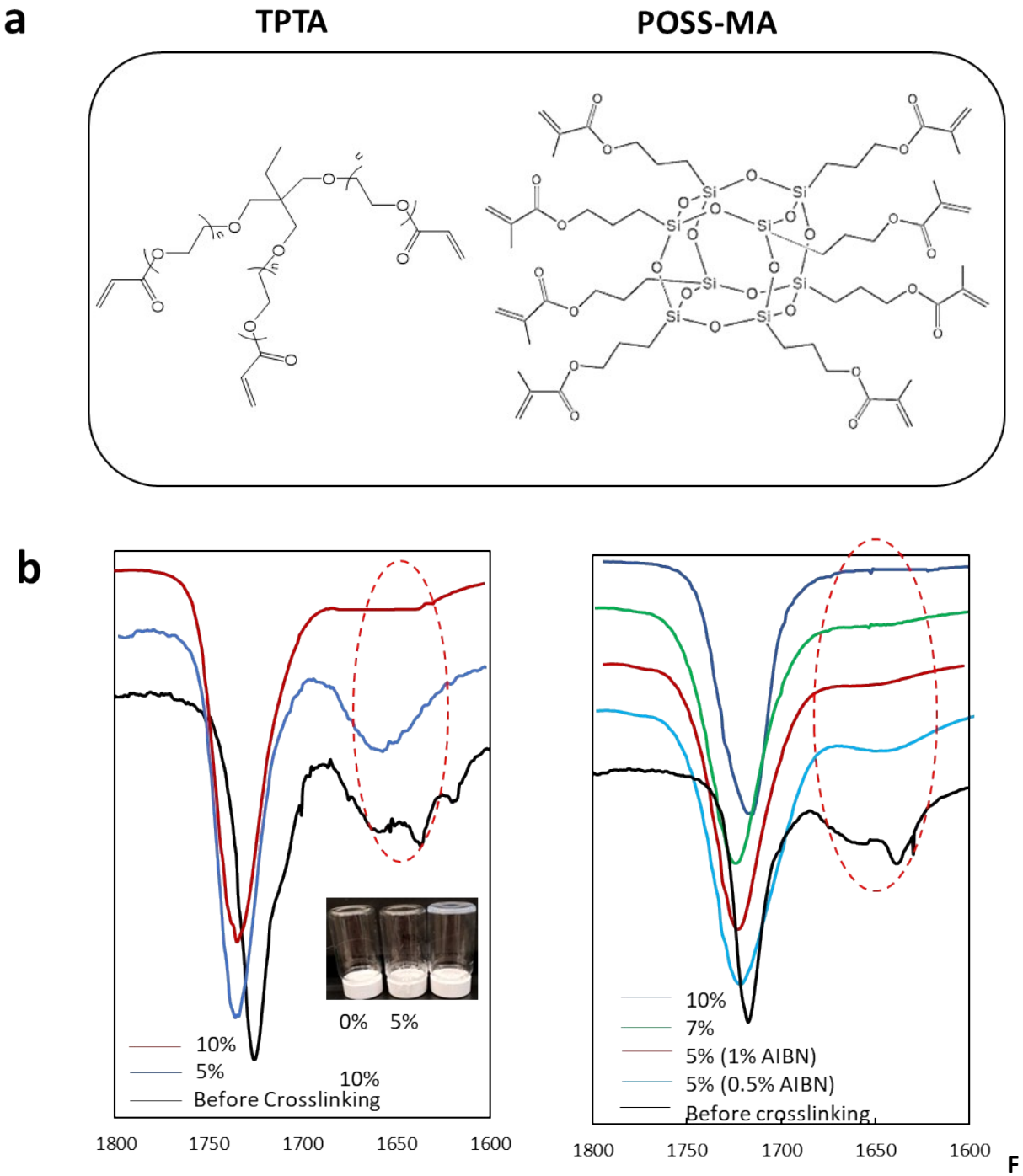


Fig. S1. UV-Vis calibration curve of PS solutions.



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ig. S2. a) Chemical structure of Trimethylolpropane Trimethacrylate, TPTA (left) and Polyhedral Oligomeric Silsesquioxane Methacrylate, POSS-MA (right). b) FT-IR spectra of crosslinked electrolyte with TPTA (left) and POSS-MA (right) at different concentration. The insets are photos of the crosslinked electrolyte samples.

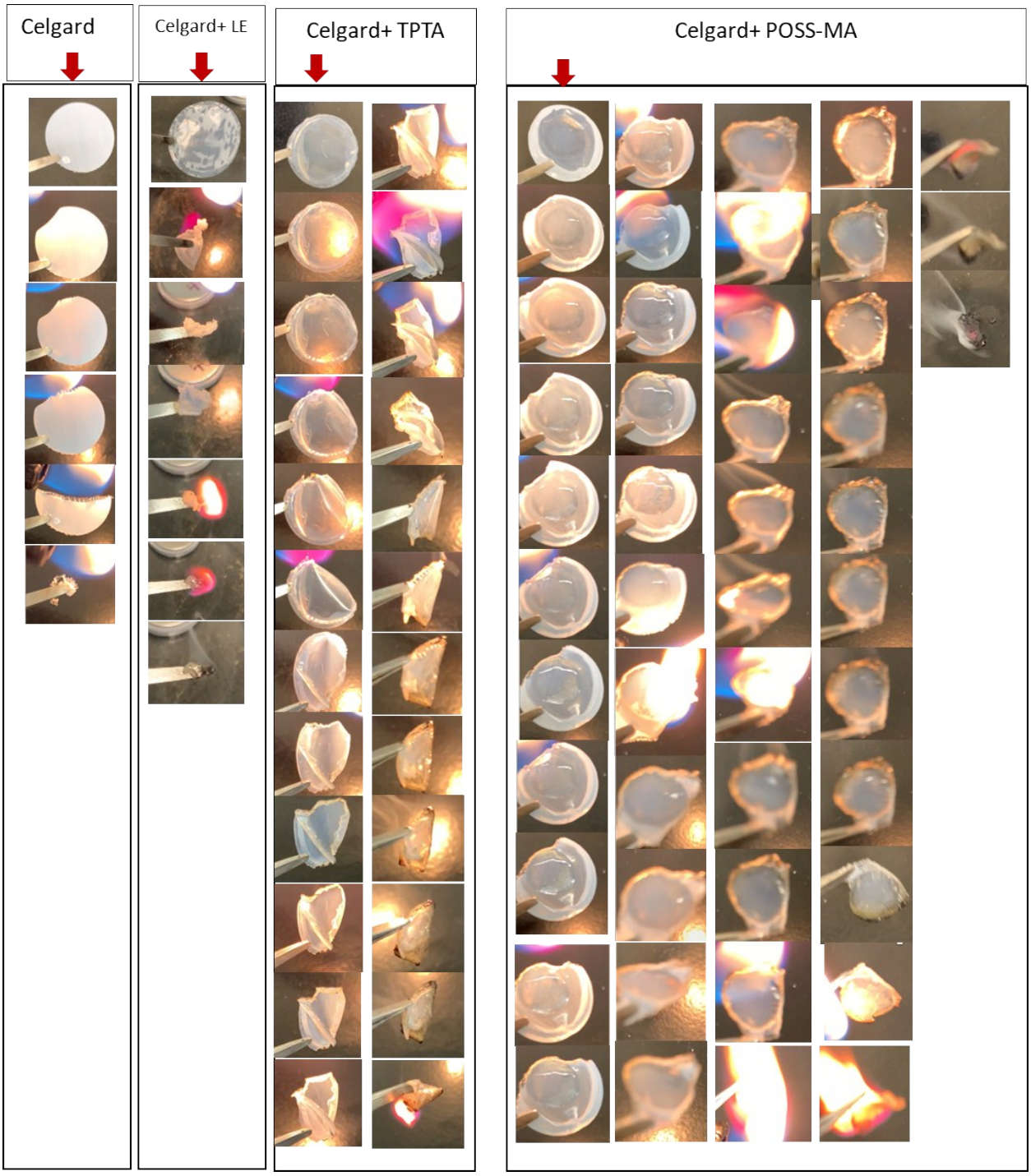

Fig. S3. Time lapse images of flammability test on a) dry celgard, b) Celgard soaked in LE, c) TPTA gelled Celgard, and d) MA-POSS gelled Celgard. Each image is after a three-second exposure of the separator to the flame.

Calculation of relative diffusion rate of Celgard 2400 to gelled Celgard with TPTA 10%:

The Fig S4. shows a schematic diffusion cell with the polysulfide solution in the cathode chamber that will diffuse to the anode chamber. The figure also compares the diffusion pass through the LE celgard and gelled one (GE).

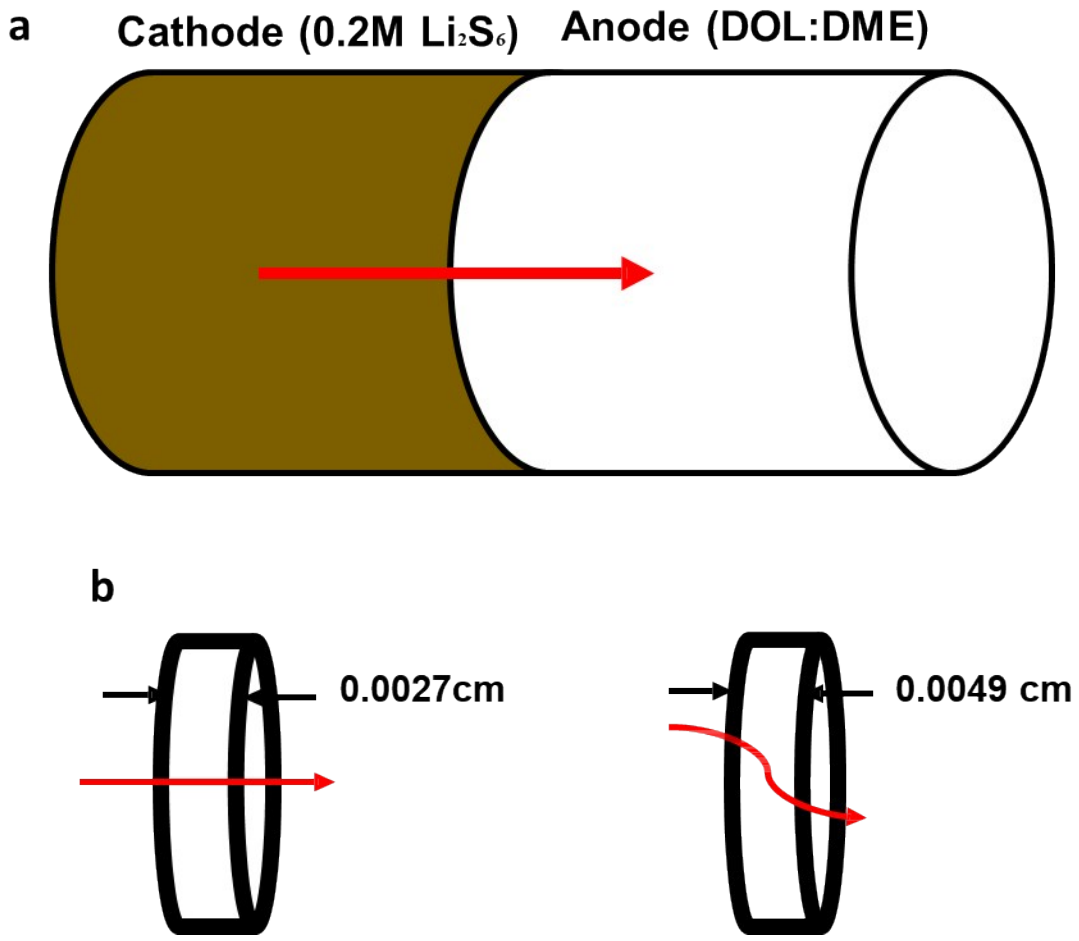

Fig S4. Schematic diffusion cell (a) and diffusion path and thickness of the two separators.

Assuming a well-mixed system, and a tortuosity factor of 2 and 1.5 for TPTA and Celgard 2400, respectively:

$$J_{TPTA} = -\frac{D_{TPTA}\,\varepsilon dC}{\tau \quad dx} = mol\ cm^{-2}\ s^{-1} \qquad \varepsilon = 0.15 \qquad \text{Eq. S1}$$

$$J_{Celg} = -\frac{D_{Celg}\,\varepsilon dC}{\tau \quad dx} = mol\,cm^{-2}\,s^{-1} \qquad \varepsilon = 0.4$$ Eq. S2

Where J is the diffusion molar flux [mol cm^{-2} s^{-1}], D_{TPTA} and D_{Celg} are the diffusion coefficients [cm^2 s^{-1}], ε is the porosity of the separator, τ is the tortuosity factor of the separator, C is the concentration of the polysulfide species [mmol L^{-1}], and x is the thickness of the separator [cm]. At 5 minutes, the concentrations for TPTA-10% and Celgard 2400 were evaluated. Due to the UV-vis's concentration limit, we analyzed the polysulfide concentration at earlier times in order to minimize concentration dilution issues.

TPTA-10%			Celgard 2400		
Absorbance	**Concentration (mM)**	**Time (min)**	**Absorbance**	**Concentration (mM)**	**Time (min)**
0.08	**0.28**	**5**	**0.272**	**1.04**	**5**
0.116	0.53	10	0.298	1.42	7.5
0.16	0.83	15			
0.212	1.18	20			

Calculated relative diffusivity of Celgard to TPTA = **23.86**

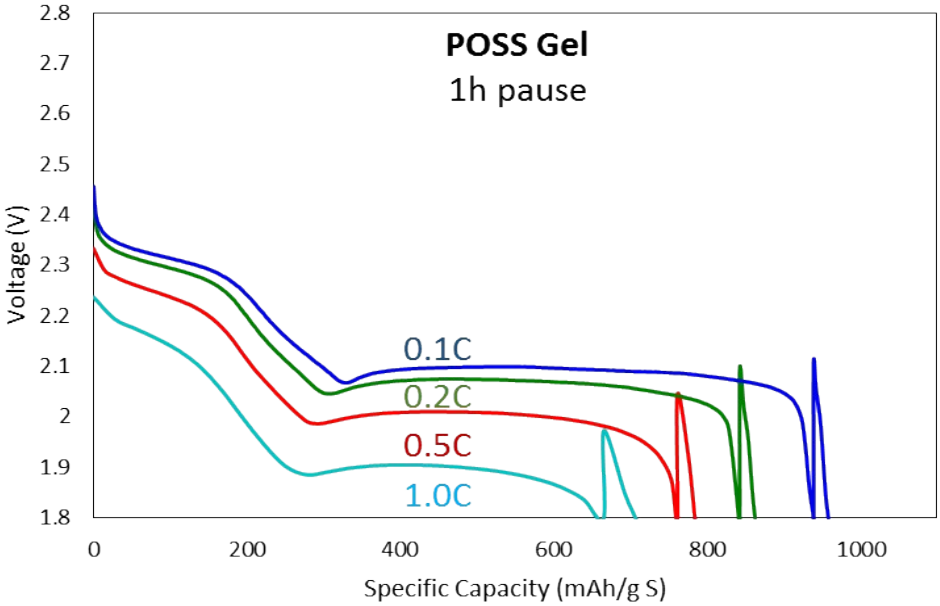

Fig. S5. POSS-MA gel electrolyte coin cell discharge curves with 1hr pause for four discharge rates.

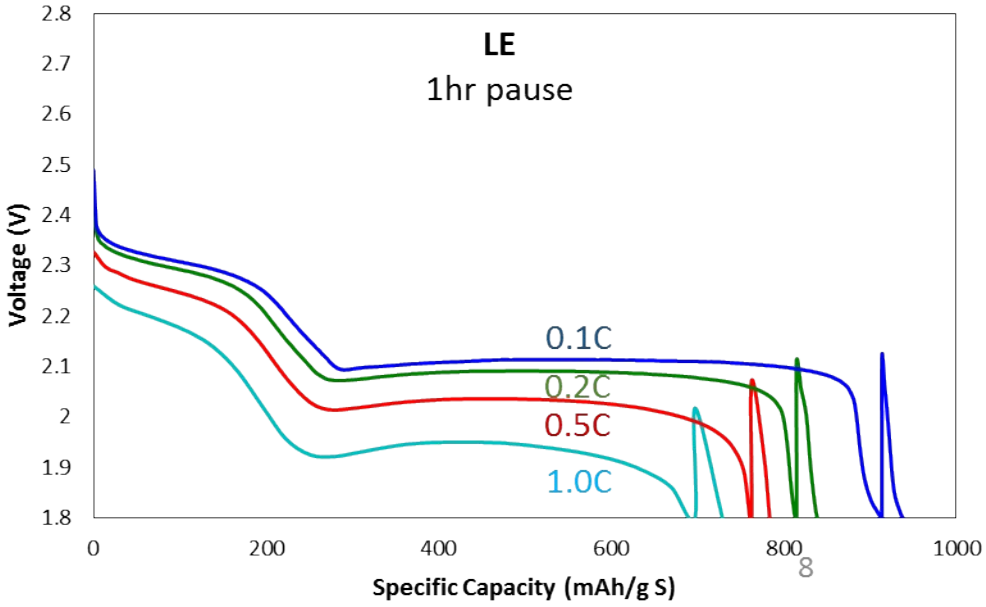

Fig. S6. Liquid electrolyte coin cell discharge curves with 1hr pause for four discharge rates with increased LiTFSI concentration (1.0M to 1.5M).

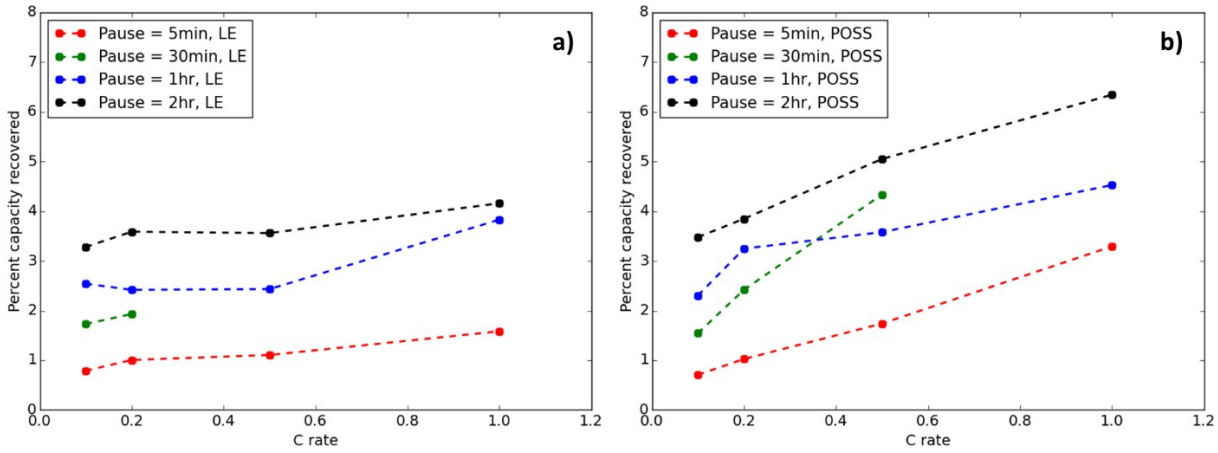

Fig. S7. Coin cell results for the effect of pause time on percent capacity recovered after pause for liquid electrolyte (a) and POSS-MA gel electrolyte (b).