



## Article

# Radical-Scavenging Activatable and Robust Polymeric Binder Based on Poly(acrylic acid) Cross-linked with Tannic Acid for Silicon Anode of Lithium Storage System

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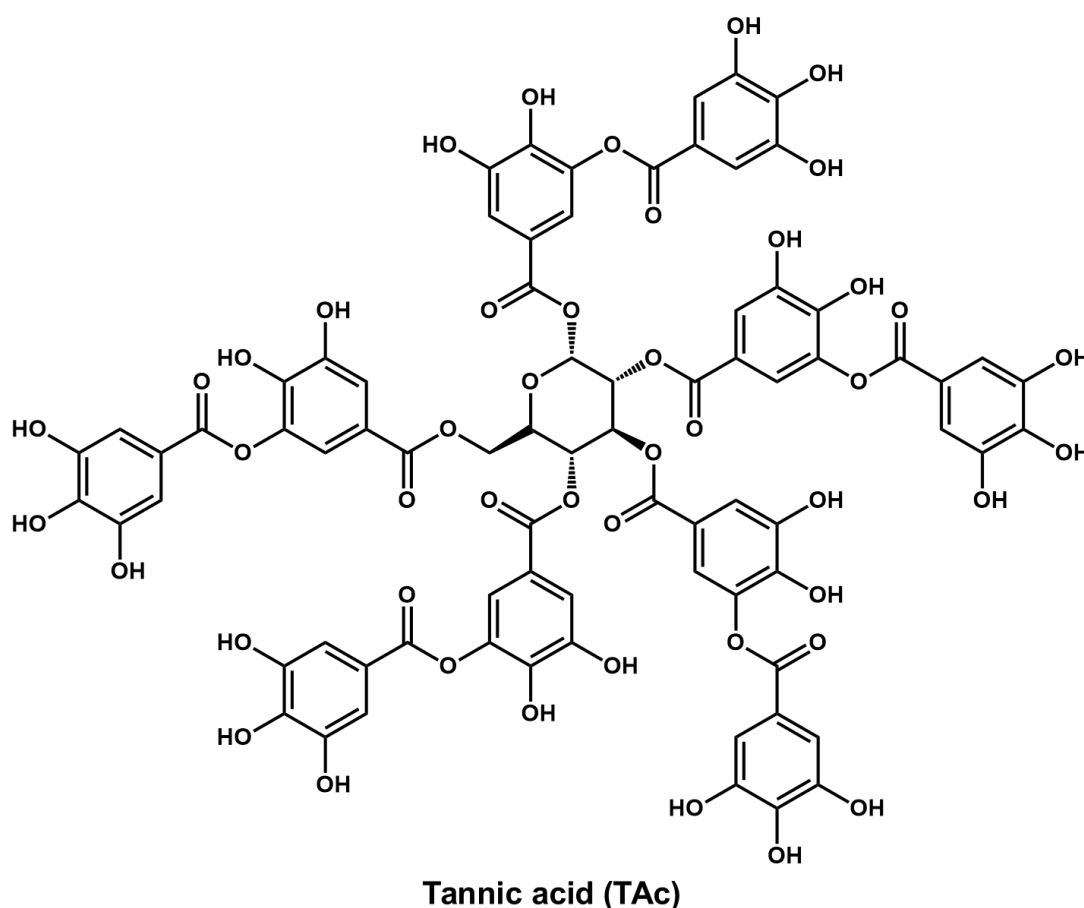
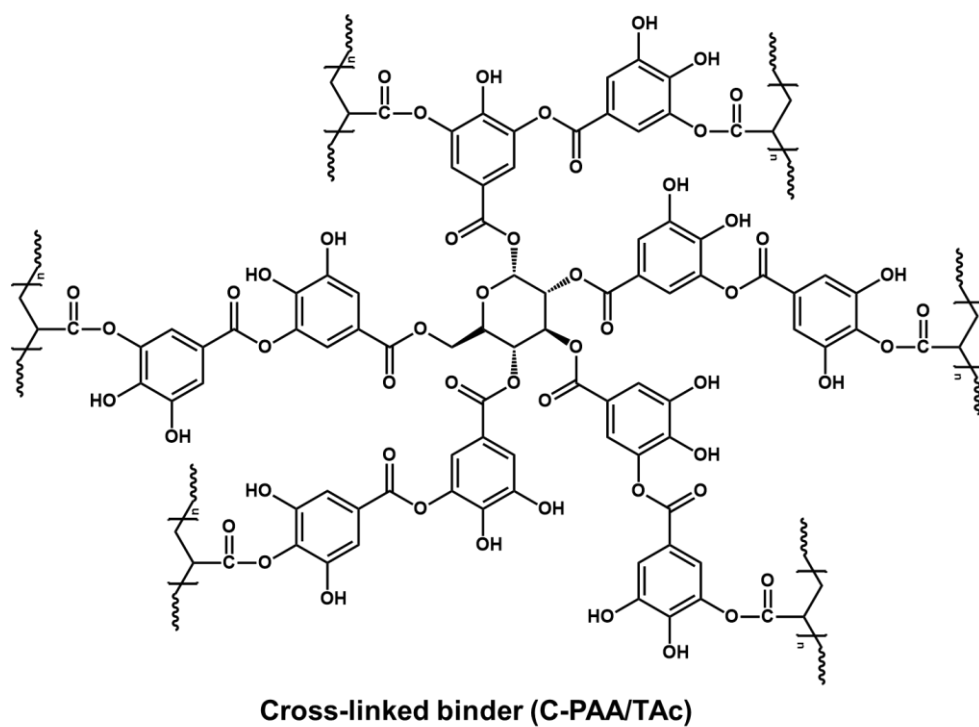
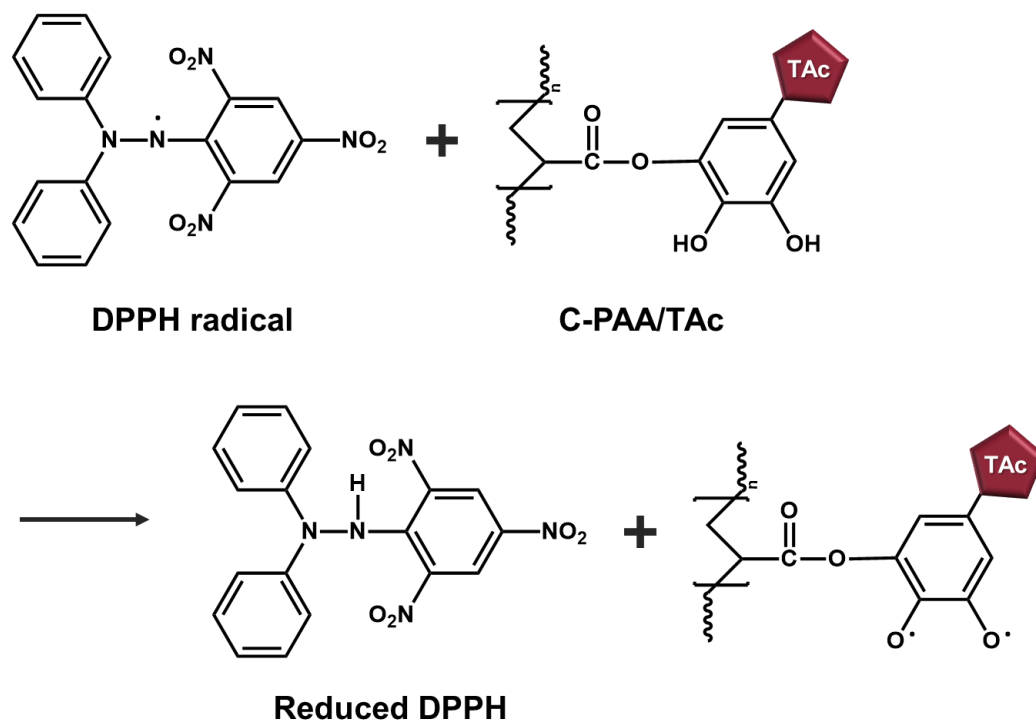


Figure S1. Molecular structure of tannic acid (TAc).



**Figure S2.** Cross-linked binder (C-PAA/TAc) formed by condensation between PAA and TAc.



**Figure S3.** Reaction of antioxidant and DPPH free radical.

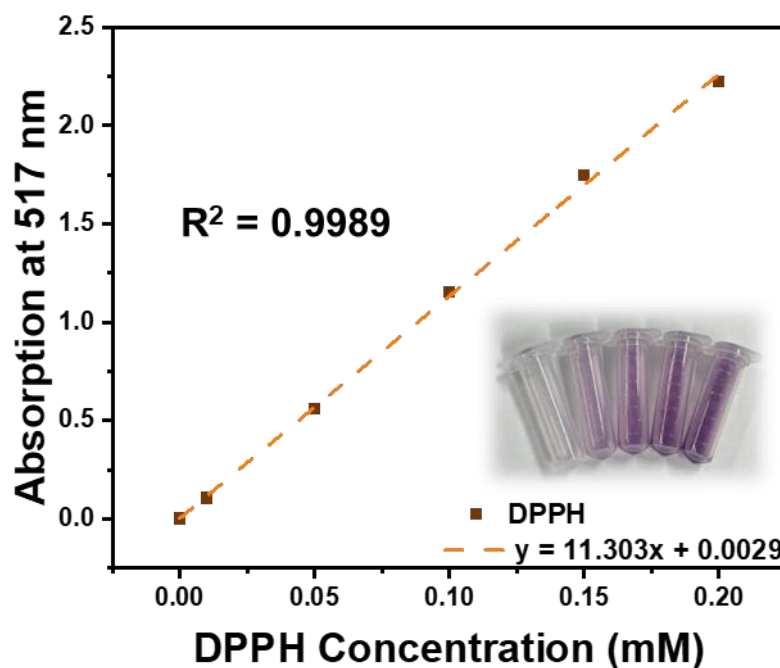


Figure S4. Optimization of DPPH assay: Color response as a function of the DPPH concentration.

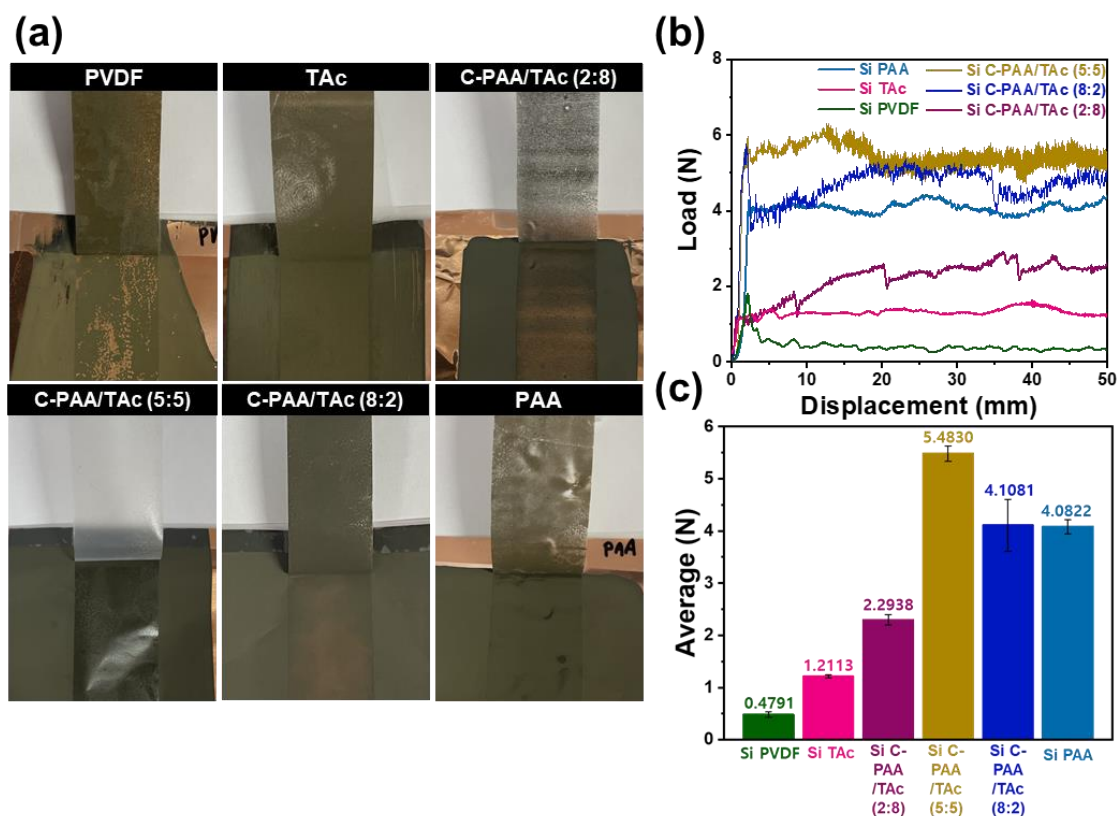


Figure S5. (a) Optical images of the tapes peeled from the Si PVDF, Si TAc, Si C-PAA/TAc, and Si PAA electrodes; (b) 180° peel-off test results of Si PVDF, Si TAc, Si C-PAA/TAc, and Si PAA electrodes EIS data of Si PAA, Si TAc and Si C-PAA/TAc; (c) the average peeling forces.

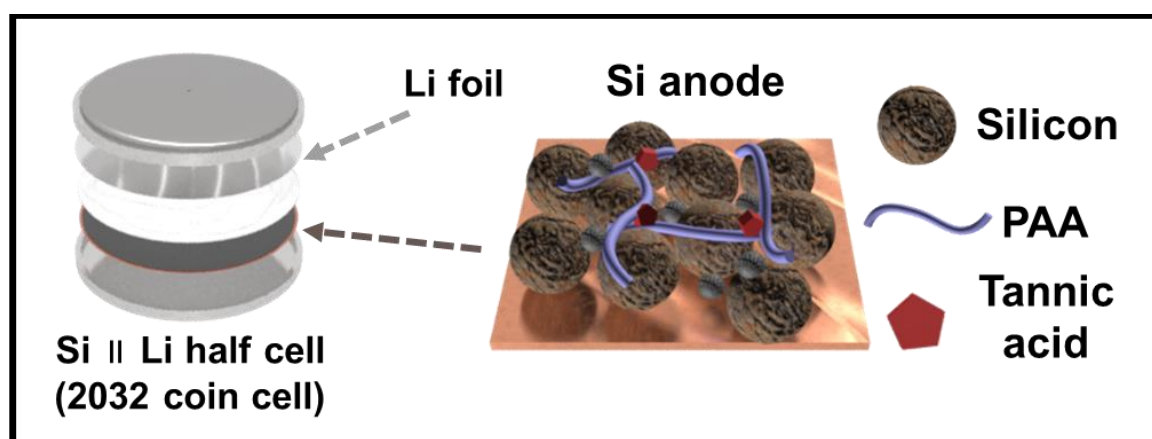


Figure S6. Schematic assembly illustration of Si || Li half cell configuration.

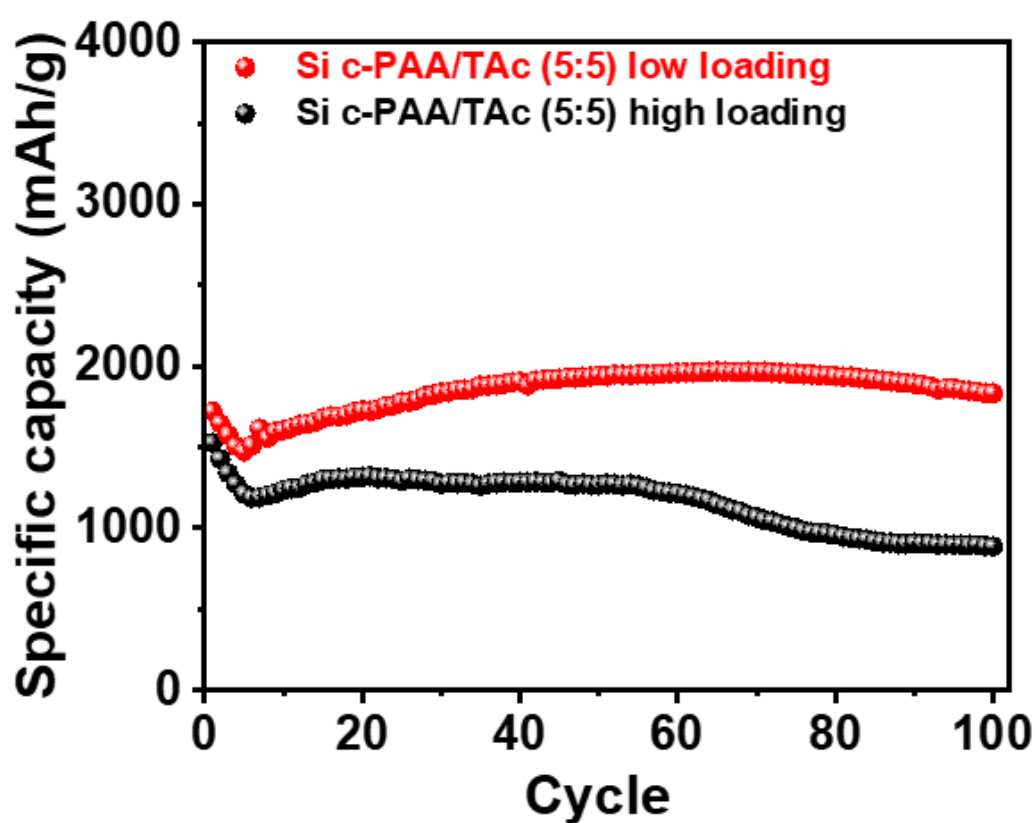


Figure S7. Cycling performance of Si C-PAA/TAc low ( $\sim 0.35 \text{ mg cm}^{-2}$ ) and high ( $\sim 1.1 \text{ mg cm}^{-2}$ ) loading at 0.5 C-rate.

Table S1. Electrochemical performances of Si PAA, Si TAc and Si C-PAA/TAc.

	1st discharge capacity (mA h g <sup>-1</sup> )	1st charge capacity (mA h g <sup>-1</sup> )	1st cycle efficiency (%)	100th cycle capacity (mA h g <sup>-1</sup> )	Capacity retention (100th cycle)
Si PAA	2583	2126	82.30	1072	67.61
Si TAc	2205	1617	73.33	1112	61.74
Si C-PAA/Tac (5:5)	2692	2195	81.53	1833	106.5

Si C- PAA/Tac (8:2)	2706	2235	82.59	1545	90.35
Si C- PAA/Tac (2:8)	2595	2061	79.42	1193	69.36

**Table S2.** EIS data of Si PAA, Si TAc and Si C-PAA/TAc after 100th cycle.

	<b>R<sub>s</sub> at 100th cycle</b>	<b>R<sub>SEI</sub> at 100th cycle</b>	<b>R<sub>CT</sub> at 100th cycle</b>
Si PAA	8.57	11.23	144.33
Si TAc	3.01	7.17	85.76
Si C-PAA/Tac (5:5)	2.84	5.98	85.10
Si C-PAA/Tac (8:2)	6.11	5.99	70.27
Si C-PAA/Tac (2:8)	4.82	7.69	116.27