

## **Supporting Information**

### The Study of the Binder Polyacrylic Acid and its Role in Concomitant Solid-Electrolyte Interphase Formation on Silicon Anodes

Katie L. Browning,<sup>‡†</sup> Robert L. Sacci<sup>†\*</sup>, Mathieu Doucet,<sup>§</sup> James F. Browning,<sup>§</sup> Joshua R. Kim,<sup>§</sup> and Gabriel M. Veith<sup>†\*</sup>

<sup>‡</sup> Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN 37996, United States

<sup>†</sup> Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States

<sup>§</sup> Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States

\*Corresponding author, email: [veithgm@ornl.gov](mailto:veithgm@ornl.gov)

Table S1. Tabulated values for NR refinements for the thickness, SLD, and roughness for the a-Si, interfacial layer, and PAA layers, respectively and goodness of fit ( $\chi^2$ ).

Potential	a-Si Thickness [nm]	a-Si SLD [ $10^{-6} \text{ \AA}^{-2}$ ]	a-Si Roughness [nm]	SEI Thickness [nm]	SEI SLD [ $10^{-6} \text{ \AA}^{-2}$ ]	SEI Roughness [nm]	PAA Thickness [nm]	PAA SLD [ $10^{-6} \text{ \AA}^{-2}$ ]	PAA Roughness [nm]	$\chi^2$
Air	58.81±0.04	1.76±0.02	1.52±0.03							4.2
OCV	56.38±0.16	1.73±0.03	1.31±0.41	4.33±0.36	1.38±0.17	3.25±0.12	36.14±0.16	4.33±0.02	1.12±0.18	4.7
1.5 V	56.54±0.15	1.79±0.03	1.81±0.37	4.31±0.41	1.48±0.21	3.08±0.14	36.19±0.30	4.31±0.02	1.39±0.18	4.4
1.2 V	56.08±0.39	1.78±0.03	1.40±0.71	4.75±0.65	1.42±0.21	3.08±0.16	36.30±0.28	4.13±0.02	1.66±0.18	7.2
0.8 V	56.08±0.51	1.78±0.03	1.30±0.65	4.97±1.02	1.30±0.40	3.56±0.27	36.12±0.54	4.41±0.02	1.30±0.1	6.4
0.4 V	58.19±1.77	1.25±0.04	3.29±1.15	5.12±1.7	1.25±0.31	3.03±0.11	33.87±0.10	4.55±0.02	0.85±0.14	3.4
0.15 V	108.43±0.51	-0.29±0.04	4.21±0.36	8.94±0.37	2.52±0.35	3.79±0.50	30.14±0.93	3.81±0.04	5.32±0.19	3.6
0.05 V	177.54±1.95	-0.64±0.04	4.67±0.95	9.94±0.30	1.52±0.10	4.96±1.31	20.16±2.35	3.56±0.09	6.36±0.42	7.8
1.5 V	61.51±0.10	-0.3±0.04	4.76±0.05	14.94±0.23	3.83±0.05	3.12±0.13	18.92±0.69	4.66±0.03	6.66±0.52	5.4
0.05 V	100.27±1.99	-0.65±0.04	5.81±1.29	10.96±1.22	1.13±0.61	4.71±0.53	19.68±0.89	3.42±0.07	6.93±0.34	5.4

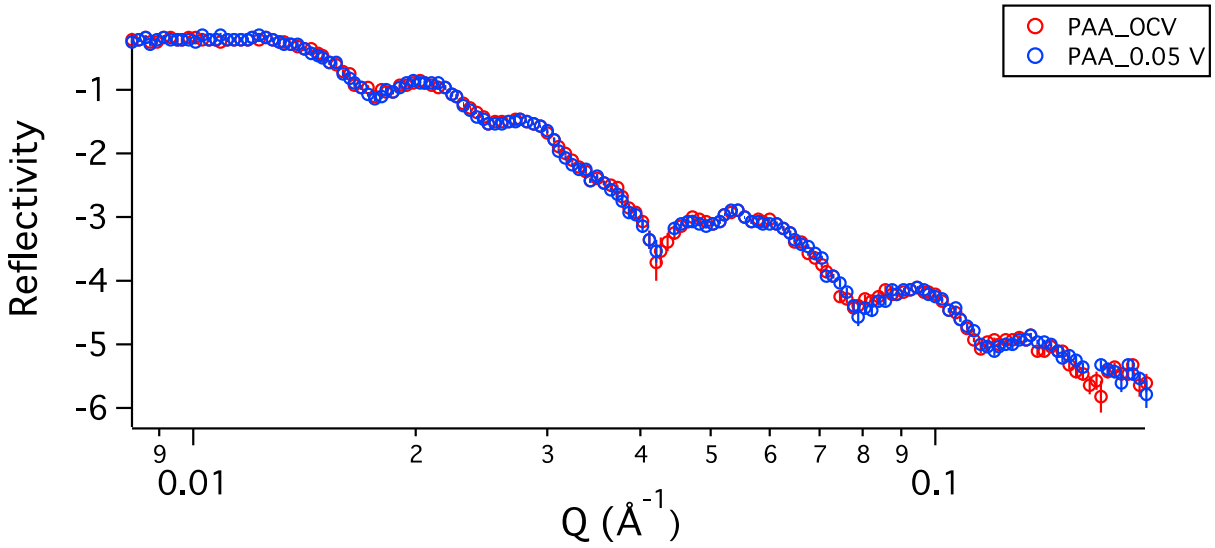


Figure S1. The reflectivity patterns, of PAA processed at 5 wt% and 200°C, that shows no change between the OCV measurement and 0.05 V.

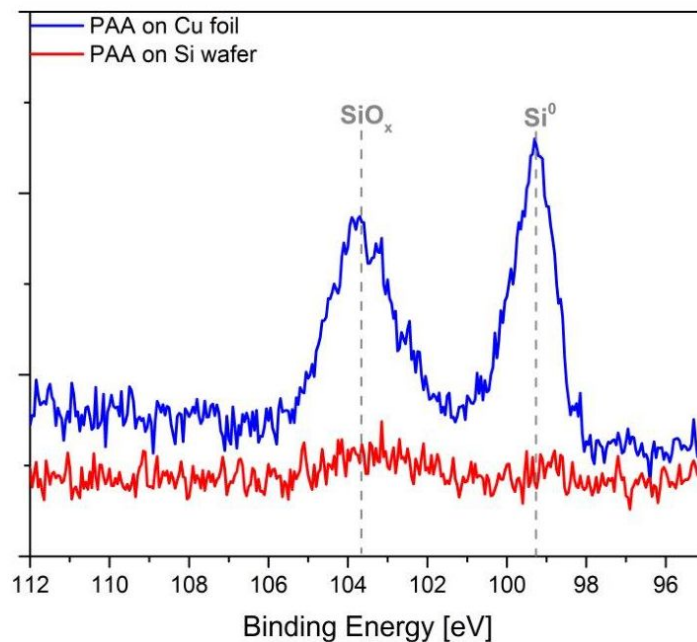


Figure S2. The Si2p from XPS of PAA coated on atomically smooth Si (red) and Cu foil

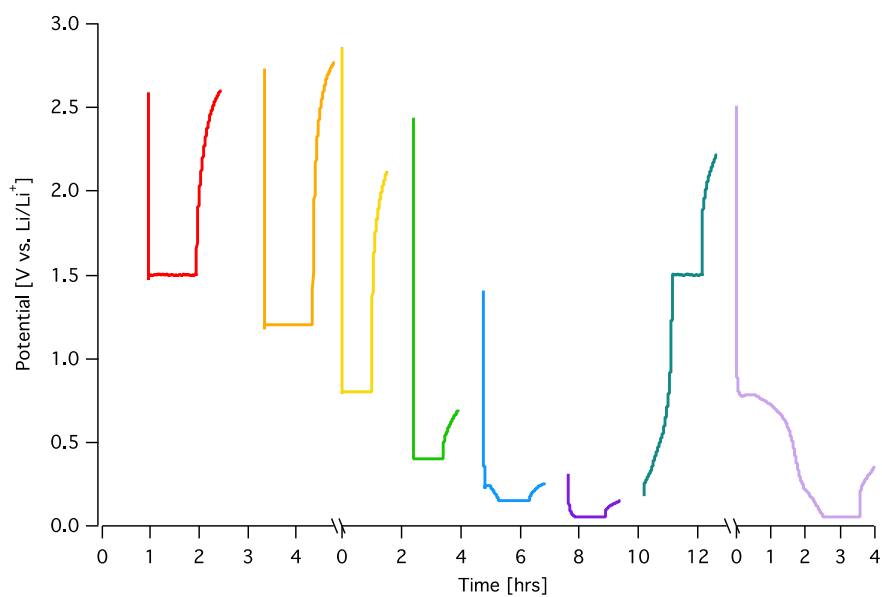


Figure S3. Voltage profile for the NR experiments. Gaps between potentials is when NR was collected as the cell relaxed back to a OCV.

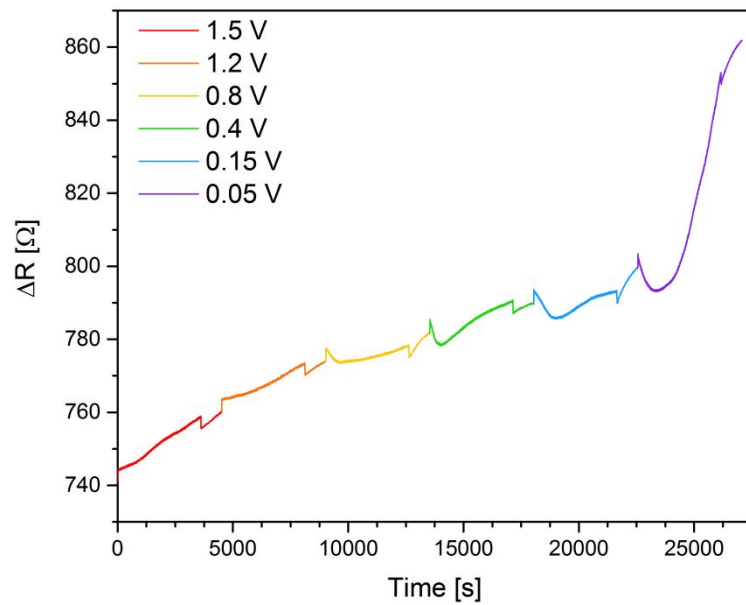


Figure S4. The change in resistance (in ohms) at the different potential holds.

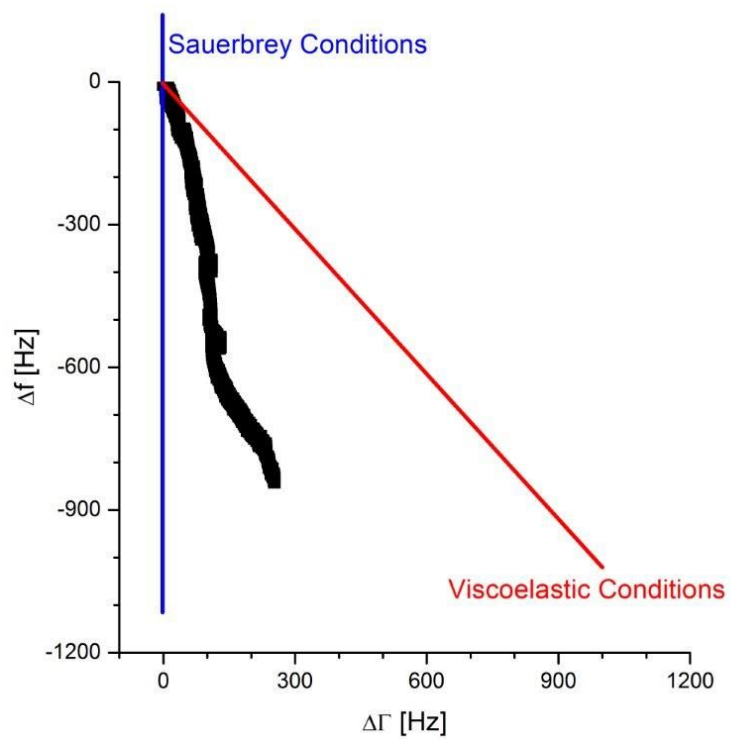


Figure S5. Sauerbrey versus viscoelastic conditions as a function of lithiation.

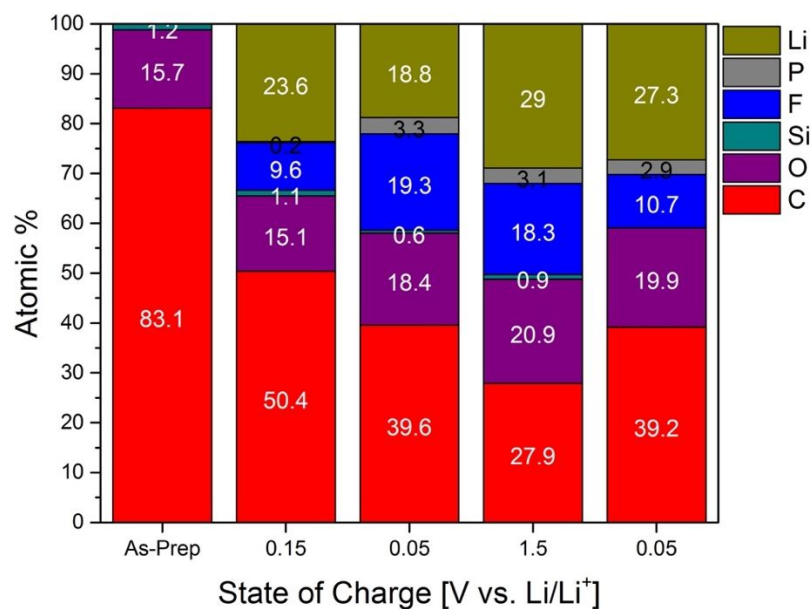


Figure S6. Atomic percent from XPS fits for 0.15, 0.05, 1.5, and 0.05 V

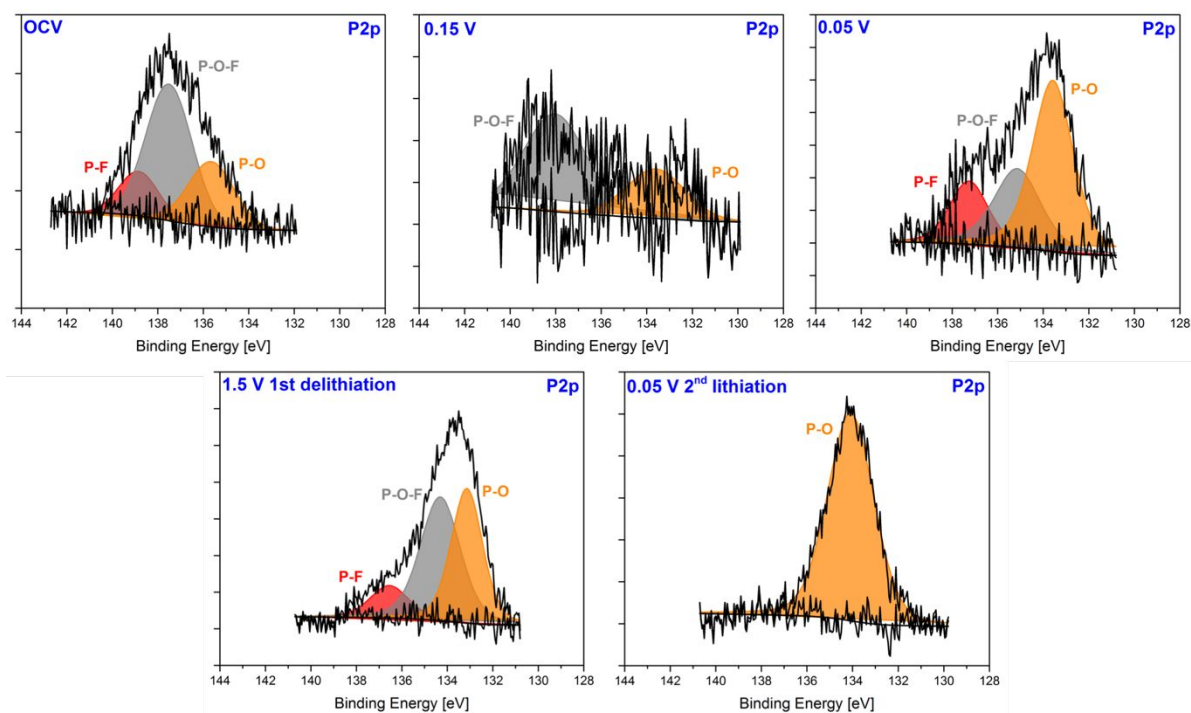


Figure S7. The P2p spectra.

Table S2: Tabulated XPS peak assignments.

Sample	C1s	C1s	C1s	C1s	C1s	O1s	O1s	O1s	O1s	O1s	Si2p	Si2p	Si2p	Si2p	Si2p
	C-C, C-H	C-O	C=O	O-C=O	-CO <sub>3</sub>	Li <sub>2</sub> O	Li-O	C=O	C-O, Si-O	C-O-C, P-O-F	Si <sup>0</sup>	Li-Si-O	Si <sup>2+</sup>	Si <sup>4+</sup>	Si-O-F
PAA	285.82		287.5	289.4				531.61	533.22	534.91	99.41		102.9	103.86	
OCV	284.97	286.29	287.46		290.06				533.41	534.63	99.38			103.99	105.55
0.15 V	284.16	285.25		288.21			529.42	531.33	532.72	534.43		101.51			
0.05 V 1st	284.79	285.95		288.78			530.33	531.93	533.65			101.06			104.78
1.5 V	284.77	286.67			289.18			531.7	533.49						104.72
0.05 V 2nd	284.02	285.05	287.63		290.06	528.79	530.55	532.17	533.8			101.49			105.67

Table S2 cont.: Tabulated XPS peak assignments.

Sample	F1s	F1s	F1s	P2p	P2p	P2p	Li1s	Li1s	Li1s	Li1s
	LiF	P-O-F	P-F	P-O	P-O-F	P-F	Li-Si	LiOH	Li-PF <sub>6</sub>	LiF
PAA										
OCV		687.87	690.22	135.61	137.46	138.8			57.87	
0.15 V	686.98		689.25	133.56	137.98		54.51	57.05		
0.05 V 1st	685.51	687.22	688.55	133.62	135.18	137.3	55.67			56.15
1.5 V	685.38	687.13	688.76	133.17	134.34	136.58	55.18			
0.05 V 2nd	686.02	687.94	688.98	134.1			55.07			56.16