

Supplementary Information for:

Siloxane incorporated copolymer as an in situ cross-linkable binder for high performance silicon anode in Li-ion battery†

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Supporting Information figures and text:

- **Table S1** Characterization of random copolymer (TBA-TEVS-n).
- **Fig. S1.** The first three cycle of cyclic voltammetry using a Si half-cell with TBA-TEVS-21 binder at a potential scan rate of 0.2mV/s.
- **Fig. S2** Nyquist plots of Si anode prepared with polymeric binders with various TEVS contents.
- **Fig. S3** TGA analysis of poly (tert-butyl acrylate) showing decomposition of TBA group at 220 °C.
- **Fig.S4** TGA analysis of TBA-TEVS-21 showing decomposition of TBA group at 215 °C.
- **Fig. S5** The FT-IR showing the formation of anhydride via cross linking of adjacent carboxylic acid functional group as a result of condensation reaction after heating at 150 °C.
- **Fig. S6** Hardness and Modulus measure for TBA-TEVS polymer series before (a), (c) and after (b), (d) cross linking.
- **Fig. S7** Nano indentation for PAA.

Table S1. Characterization of random copolymer (TBA – TEVS-n).

Polymer	TBA :TEVS ^a (x:y)	Mn(kDa) ^b
TBA-TEVS-9	91: 9	33
TBA-TEVS-21	79 : 21	15
TBA-TEVS-26	74 : 26	15
TBA-TEVS-34	66:34	13
TBA-TEVS-50	50 : 50	10
TBA-TEVS-100	0 : 100	6

^a The molar ratio were determined by comparing peak integrals of hydrogen attached to the tertiary carbons of tertiary butyl acrylate and triethoxyvinylsilanes. ^b Data based on GPC (THF, PMMA standard).

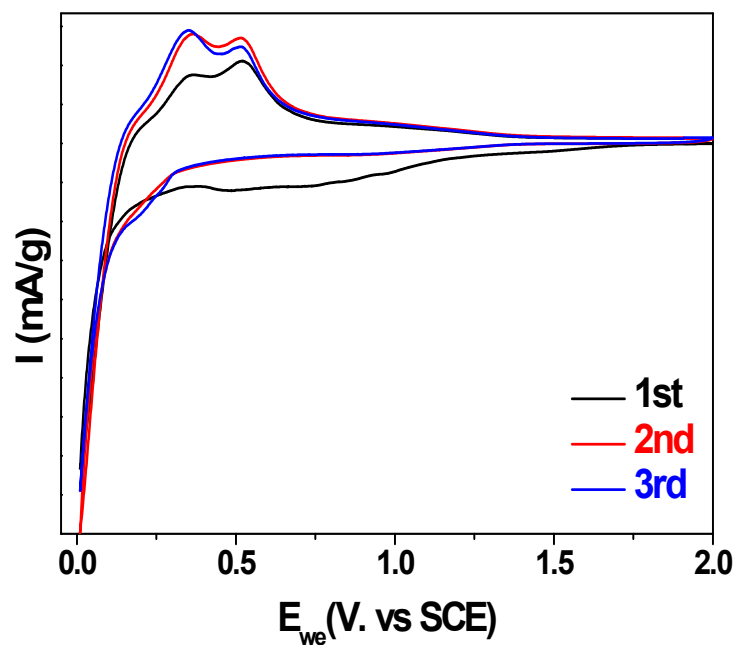


Fig. S1 The first three cycle of cyclic voltammetry using a Si half-cell with TBA-TEVS-21 binder at a potential scan rate of 0.2mV/s

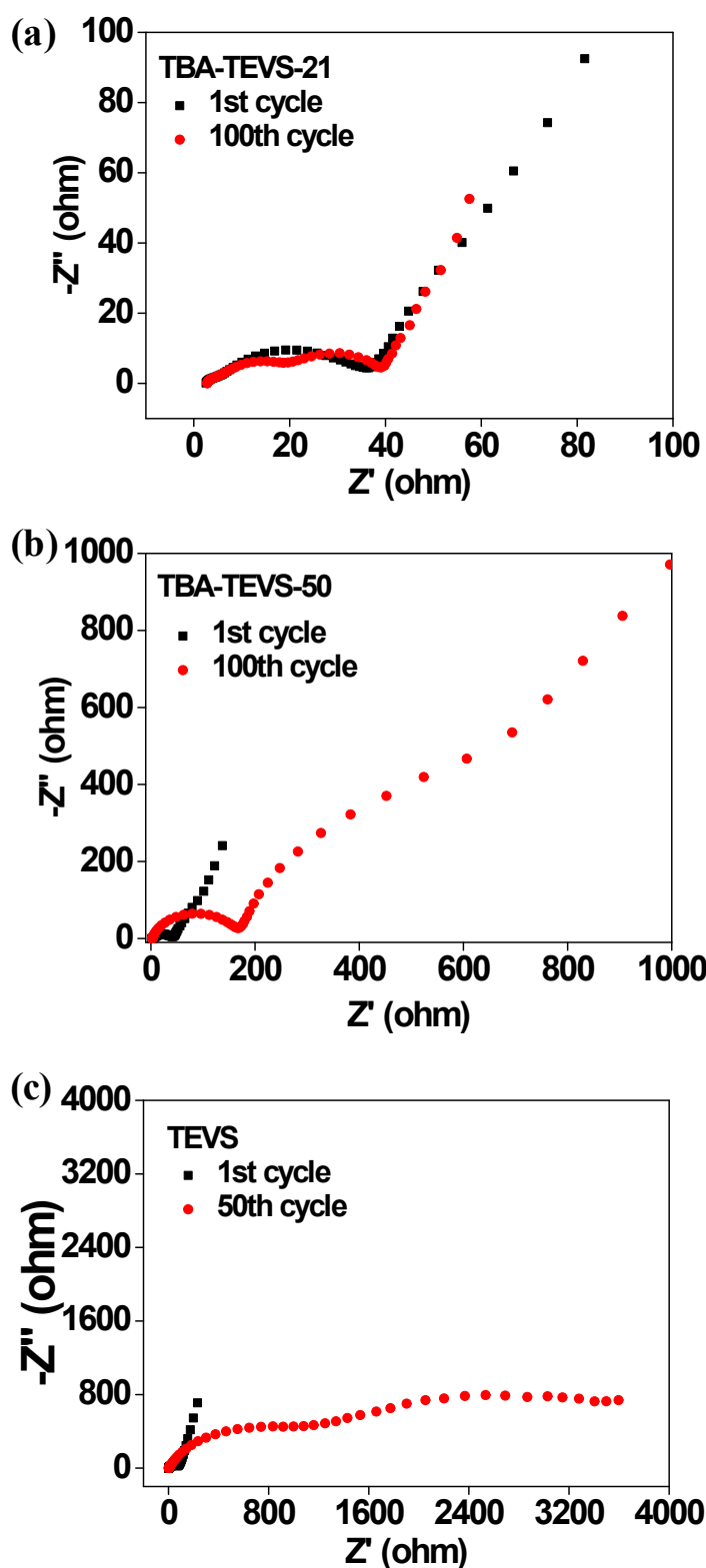


Fig. S2 Nyquist plots of Si anode prepared with polymeric binders with various TEVS contents. (a) TBA-TEVS-21, (b) TBA-TEVS-50, and (c) TEVS. TBA-TEVS-21 sample showed the lowest SEI resistance and charge transfer resistance values, and also maintained initial resistance after 100 cycles.

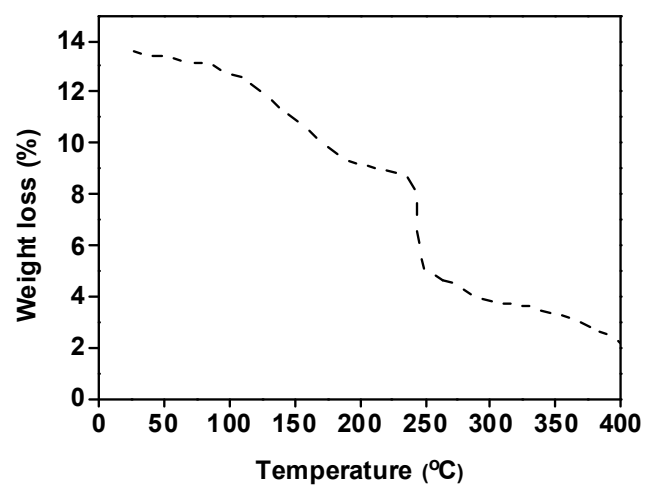


Fig. S3 TGA analysis of poly (tert-butyl acrylate) showing decomposition of TBA group at 220 °C.

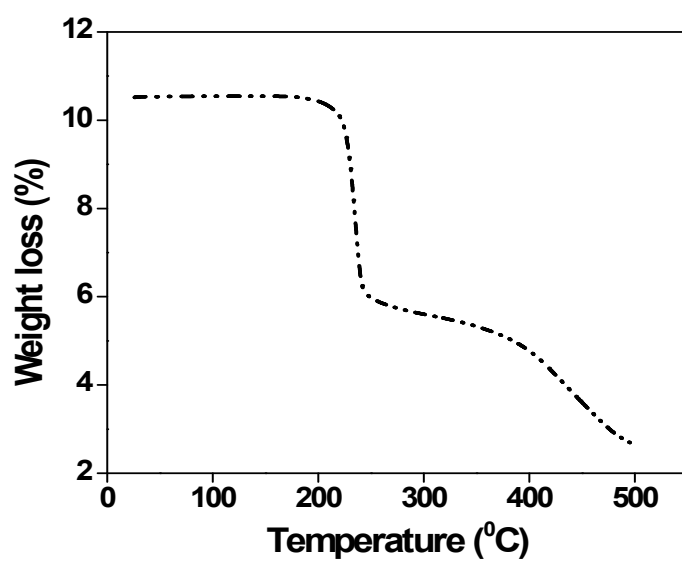


Fig. S4 TGA analysis of TBA-TEVS-21 showing decomposition of TBA group at 215 °C.

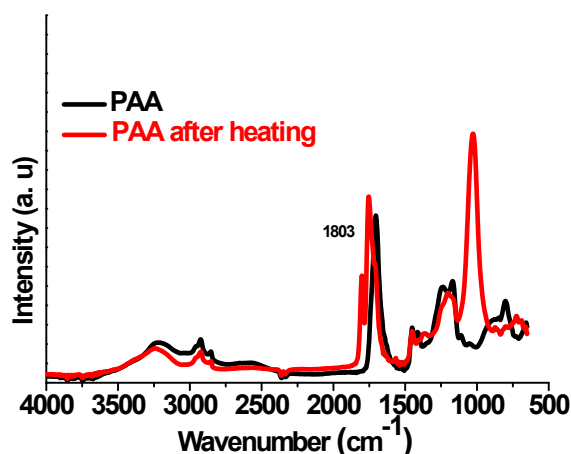


Fig. S5 The FT-IR showing the formation of anhydride via crosslinking of adjacent carboxylic acid functional group as a result of condensation reaction after heating at 150°C.

The strong and solid evidence for the formation of anhydride via the condensation of carboxylic acid could be provided by FT-IR spectroscopy which has reported in the literatures^{1, 2}. The peak appears around $\sim 1800\text{ cm}^{-1}$ in the IR spectroscopy is attributed specifically for the carbonyl stretching frequency of anhydride. For example, the poly(acrylic acid)(PAA) after heat treatment at 150 °C forms anhydride and shows a distinct peak appear around 1808 cm^{-1} . This peak is different from the carbonyl group of ester, acid, or carboxylate which usually appear less than 1750 cm^{-1} .

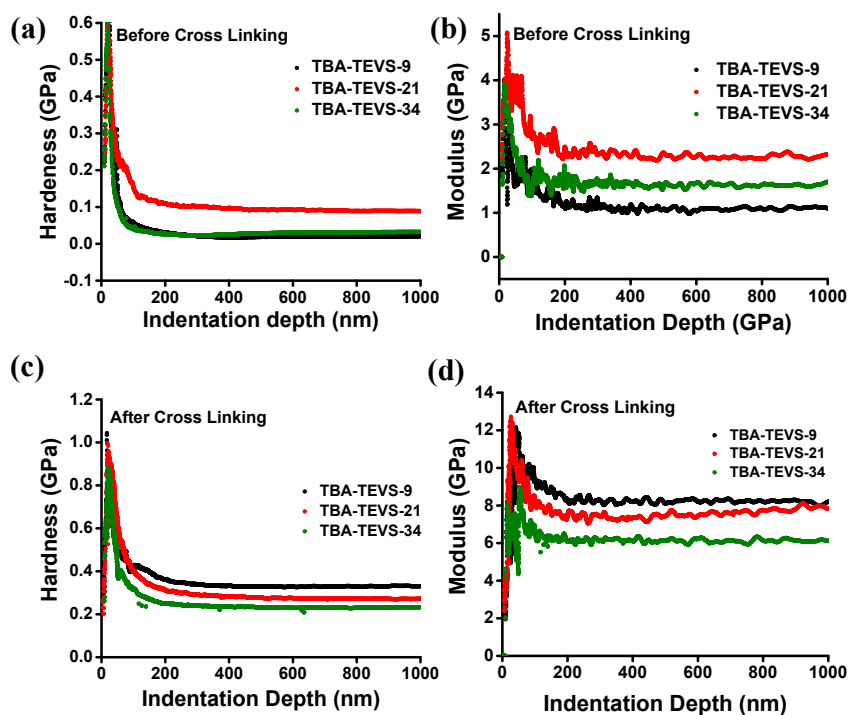


Fig. S6 Hardness and Modulus measure for TBA-TEVS polymer series before (a), (b) and after (c), (d) cross linking.

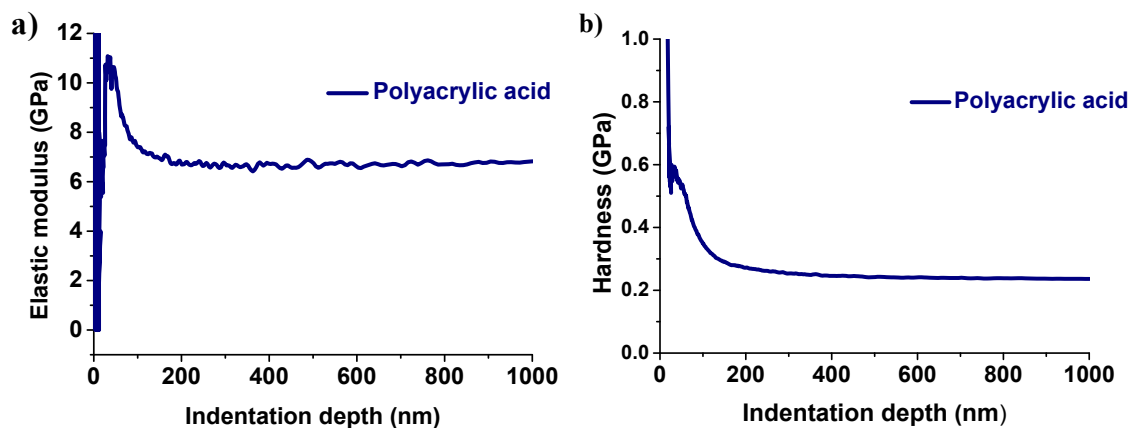


Fig. S7 (a) Elastic modulus and (b) hardness of PAA measured by nanoindentation.

Reference

1. B. I. Koo, H Kim, Y. Cho, K. T. Lee, N. S. Choi, J. Cho, *Angew. Chem. Int. Ed.* 2012, **51**, 8762–8767.
2. M. T. Jeena, J. I. Lee, S. H. Kim, C. Kim, J. Y. Kim, S. Park, J. H. Ryu, *ACS Appl. Mater. Interfaces*, 2014, **6**, 18001–18007.

