

## Supporting Information

# Eco-Friendly Water-Processable Polyimide Binders with High Adhesion to Silicon Anodes for Lithium-Ion Batteries

Yujin So <sup>1,2,†</sup>, Hyeon-Su Bae <sup>3,†</sup>, Yi Young Kang <sup>1</sup>, Ji Yun Chung <sup>1,4</sup>, No Kyun Park <sup>1</sup>, Jinsoo Kim <sup>1</sup>, Hee-Tae Jung <sup>2</sup>, Jong Chan Won <sup>1,4,\*</sup>, Myung-Hyun Ryou <sup>3,\*</sup> and Yun Ho Kim <sup>1,4,\*</sup>

<sup>1</sup> Advanced Materials Division, Korea Research Institute of Chemical Technology (KRICT), Daejeon 34114, Korea; soyujin@kRICT.re.kr (Y.S.); yykang@kRICT.re.kr (Y.Y.K.); jjyjy@kRICT.re.kr (J.Y.C.); nkpark@kRICT.re.kr (N.K.P.); jinsoo@kRICT.re.kr (J.K.)

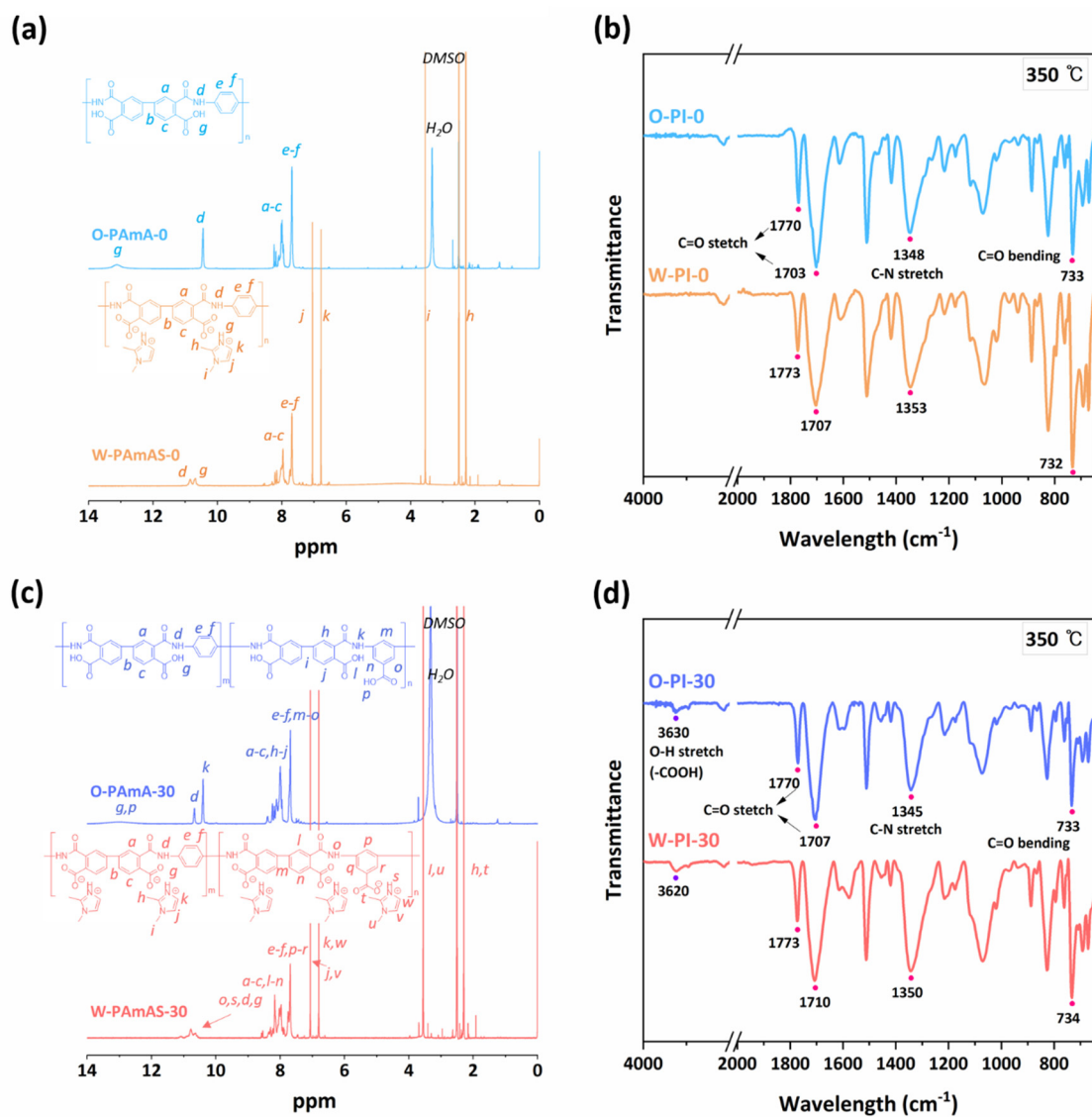
<sup>2</sup> Department of Chemical and Biomolecular Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Korea; heetae@kaist.ac.kr

<sup>3</sup> Department of Chemical and Biological Engineering, Hanbat National University, Daejeon 34158, Korea; hsbae1234@gmail.com

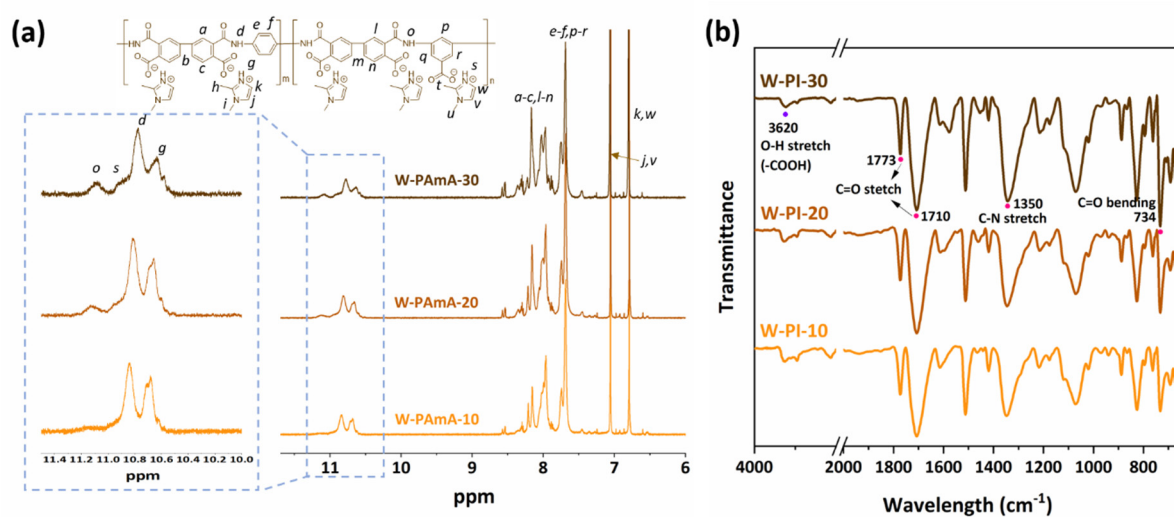
<sup>4</sup> Korea Research Institute of Chemical Technology (KRICT) School, University of Science and Technology, Daejeon 34113, Korea

\* Correspondence: jcwon@kRICT.re.kr (J.C.W.); mhryou@hanbat.ac.kr (M.-H.R.); yunho@kRICT.re.kr (Y.H.K.); Tel.: +82-860-7294 (J.C.W.); +82-42-821-1534 (M.-H.R.); +82-42-860-7274 (Y.H.K.)

† These authors contributed equally to this work.



**Figure S1.** <sup>1</sup>H NMR spectra of (a) O-PAmA-0 and W-PAmA-0 and (c) O-PAmA-30 and W-PAmA-30 in DMSO-d<sub>6</sub>. FT-IR spectra (b) O-PI-0 and W-PI-0 and (d) O-PI-30 and W-PI-30 after imidization at 350 °C.



**Figure S2.** (a)  $^1\text{H}$  NMR spectra of W-PAmAS-# and (b) FT-IR spectra of W-PI-# in proportion to the DABA content.

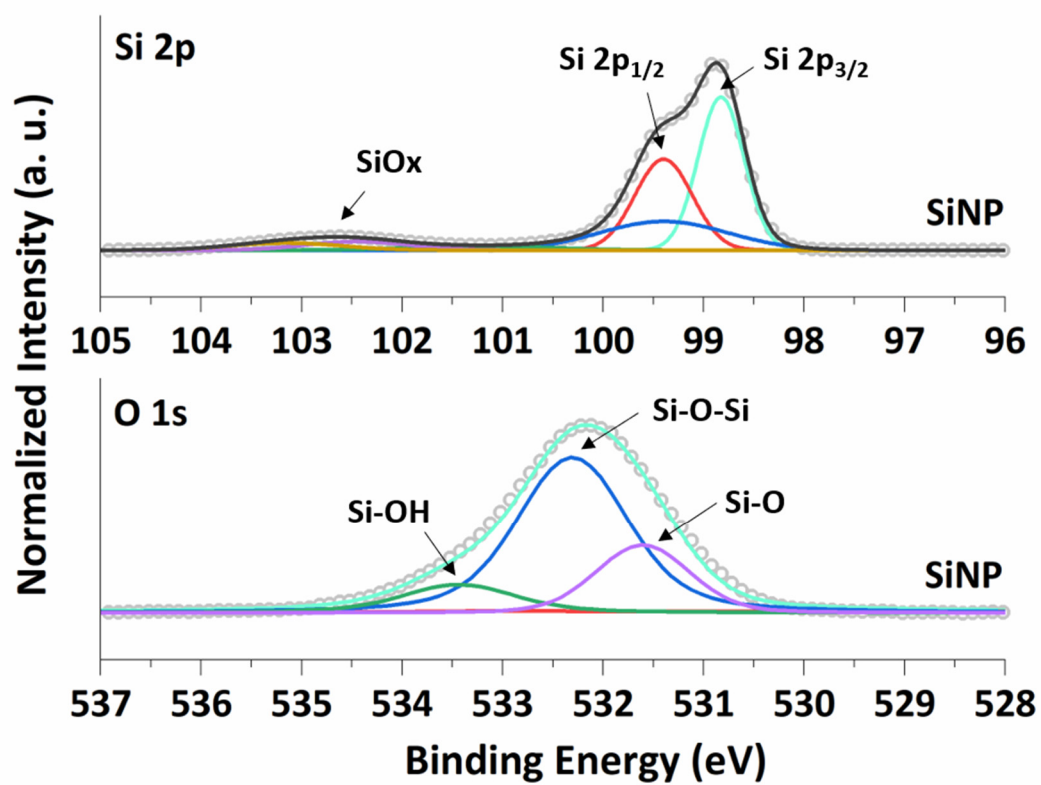
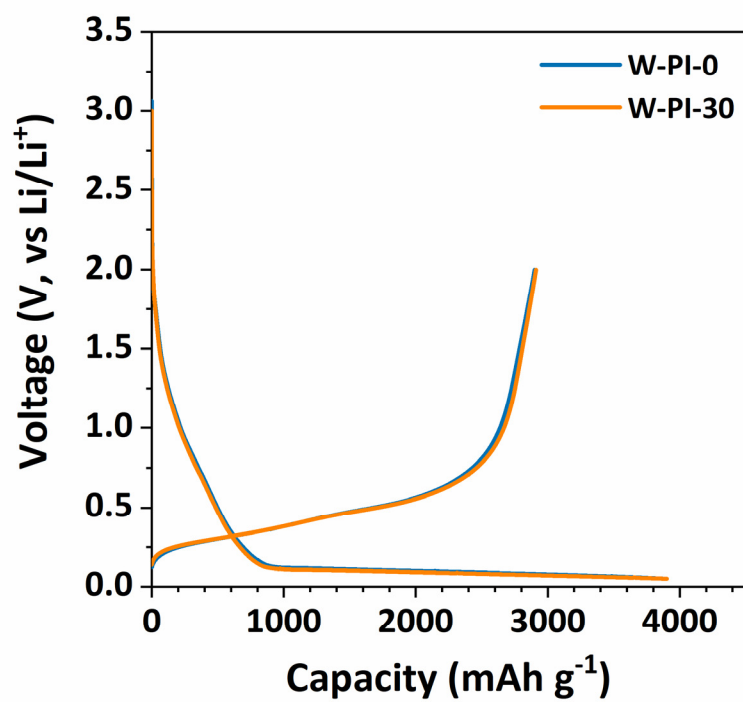


Figure S3. XPS spectra of Si 2p and O 1s in SiNPs.



**Figure S4.** Precycling curves of Si anodes with W-PI binders (0.005–2.0 V vs. Li/Li<sup>+</sup>, 200 mA g<sup>-1</sup> at 25 °C).

**Table S1.** Cycle performance of Si-based anodes with different polymer binders

Binder	Anode material	Specific Capacity (mAh g <sup>-1</sup> )	Retention (%)	Current rate	Content of binder (%)	Ref.
<i>Our polyimide (W-PI-30)</i>	<i>Nano-Si</i>	<i>2061</i>	<i>1883 mAh g<sup>-1</sup> (91.3 %) (200 cycles)</i>	<i>1200 mA g<sup>-1</sup></i>	<i>20</i>	-
Carboxymethyl chitosan	Si	1990	950 mAh g <sup>-1</sup> (50 cycles)	500 mA g <sup>-1</sup>	8	[22]
PVA-PEI	Nano-Si	3072.9	1063 mAh g <sup>-1</sup> (300 cycles)	1 A g <sup>-1</sup>	20	[23]
PAA-PANI	Nano-Si	1979	56.5 % (300 cycles)	1.0 C	25	[24]
PPyMAA	Nano-Si	3928.8	2200 mAh g <sup>-1</sup> (over 180 cycles)	420 mA g <sup>-1</sup> (0.1 C)	10	[25]
Gum arabic (GA)	Nano-Si	4056	2708 mAh g <sup>-1</sup> (75.7%) (100 cycles)	420 mA g <sup>-1</sup> (0.1 C)	25	[26]
Polyimide	Si	~800 (after 2 <sup>nd</sup> cycle)	800 mAh g <sup>-1</sup> (195 cycles)	800 mA g <sup>-1</sup>	15	[41]
Polyimide	Si	2077	75.9 % (20 cycles)	200 mA g <sup>-1</sup>	10	[42]
Polyimide	Si	1195.6	93.6 % (30 cycles)	0.1 C	25	[43]
Copolyimide (P84)	Si	1929	1313 mAh g <sup>-1</sup> (300 cycles)	1.2 A g <sup>-1</sup>	20	[4]
Copolyimide (PI-200)	Si	2989.7	2235.5 mAh g <sup>-1</sup> (200 cycles)	420 mA g <sup>-1</sup> (0.1 C)	20	[44]
Polyamide imide (PAI)	Nano-Si	~2000	1700 mAh g <sup>-1</sup> (20 cycles)	0.56 mA cm <sup>-2</sup>	10	[45]