Supporting Information

Overcome the fundamental challenge for PVDF binder to use with silicon anode with super-molecular nano-layer

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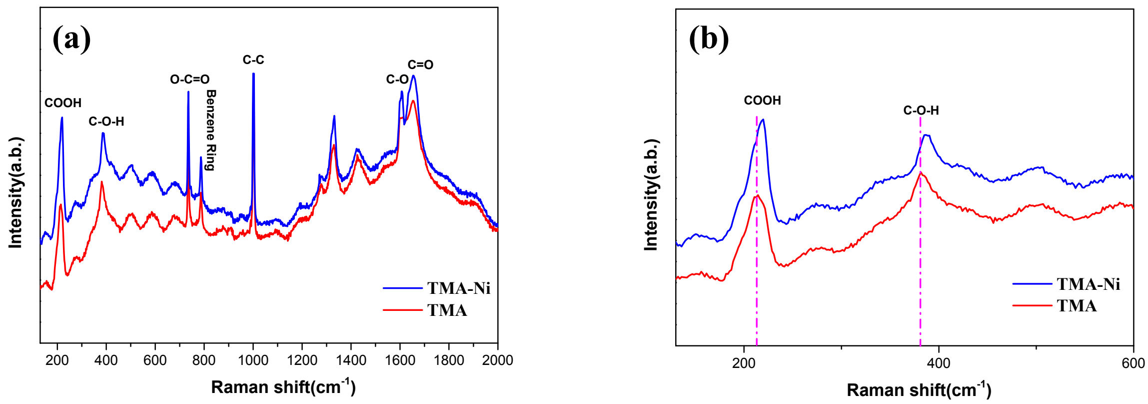

Fig. S1 Raman spectra of TMA and TMA-Ni compounds

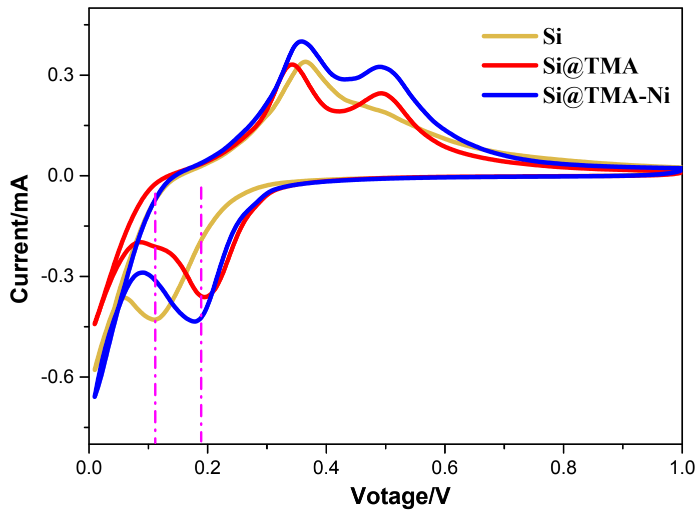

Fig. S2 The third CV curves for the pristine Si, Si@TMA, Si@TMA-Ni at a scan rate of 0.05 mV $s^{-1.}$ The enhanced peak intensity for the Si sample by TMA decoration shows the improved electrochemical activity for the electrode.

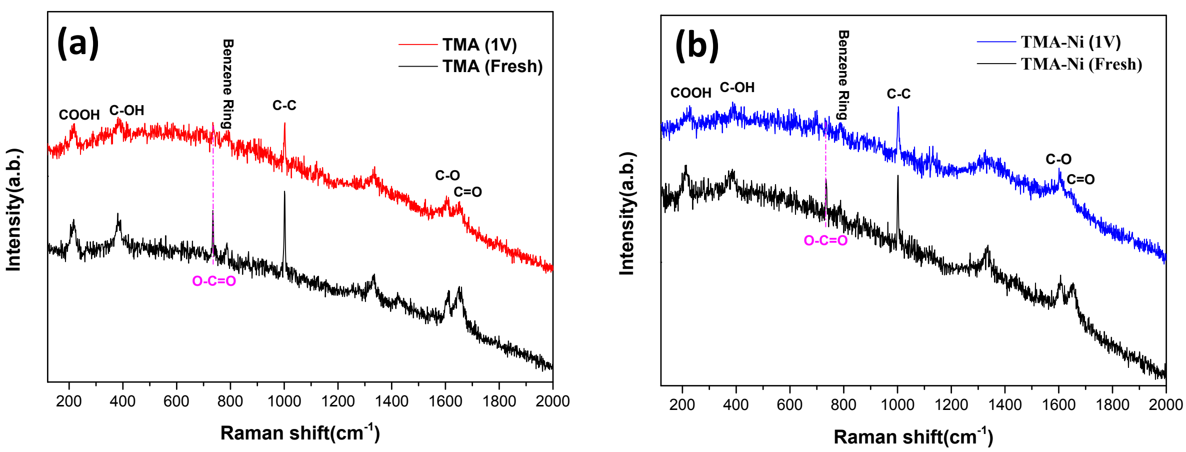

Fig. S3 Raman spectra for (a) the TMA anode and (b) the Ni-TMA anode

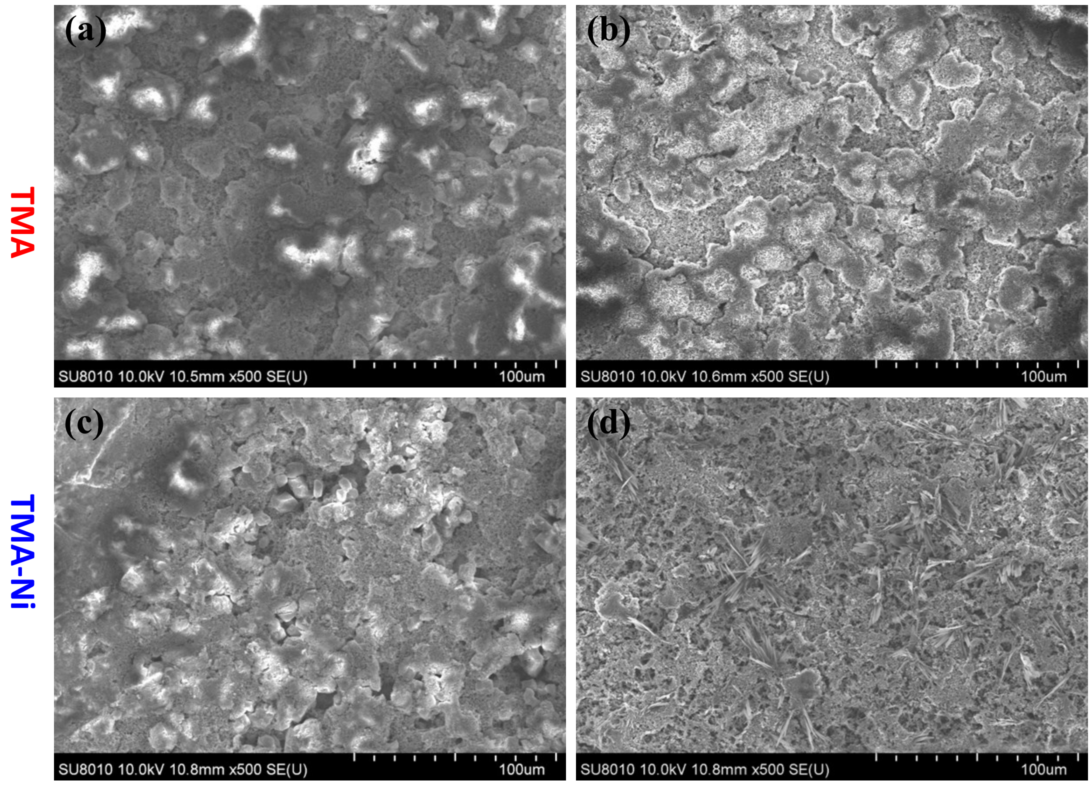

Fig. S4 The SEM images of the pure TMA and TMA-Ni electrodes: (a and c) at fresh state and (b and d) after CV tests.

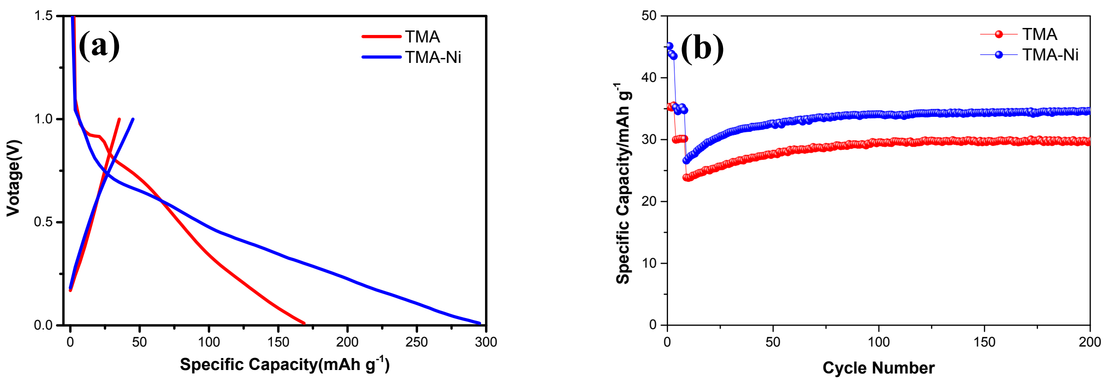

Fig. S5 The electrochemical properties for the TMA and TMA-Ni anodes: (a) the first charge-discharge profiles and (b) long-term cycling behavior. It shows the contribution of the TMA layer to the capacity of the Si anode can be neglected.

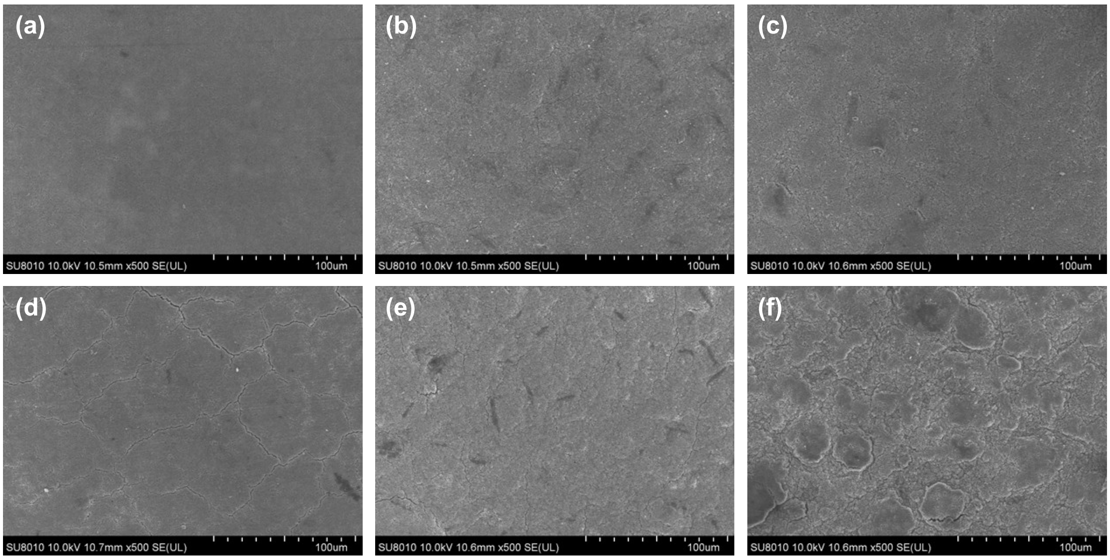

Fig. S6 The electrode integrity for the (a, d) Si, (b, e) Si@TMA and (c, f) Si@TMA-Ni electrodes: (a-c) after formation cycles, (d-f) after rate test.

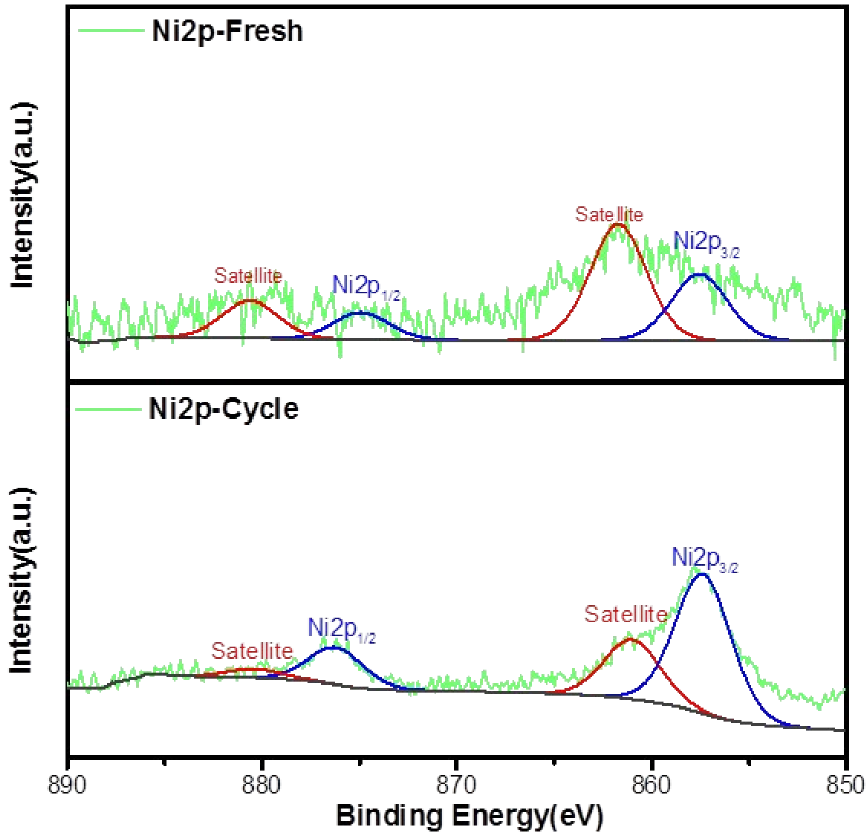

Fig. S7 Ni2p XPS spectra of Si@Ni-TMA anode at fresh state and after 200 cycles.

Table S1. Comparison of the electrochemical performances between the Si@TMA-Ni anode in this work and other reported Si-based anodes.

Sample	Binder	First coulombic efficiency	Current density ($A\ g^{-1}$)	Reversible capacity ($m\ Ah\ g^{-1}$)	Rate capability ($m\ Ah\ g^{-1}$)	Reference
Si@TMA-Ni	**PVDF**	**75.32%**	**0.84**	**1900.6(200th)**	**1685.2 (21 $A\ g^{-1}$)**	**This work**
AMPSi@C	SA	80.3%	2.1	1271 (1000th)	619 (12.6 $A\ g^{-1}$)	1
Void@SiOx@C	c-PAA-DS	61.3%	0.5	696 (500th)	664 (2 $A\ g^{-1}$)	2
	PVDF	52.3%		157 (500th)	531(2 $A\ g^{-1}$)	
Si@GC	-	85.3%	1	1447 (250th)	1613 (5 $A\ g^{-1}$)	3
Onion-like Si/C	CMC	84.5%	0.2	1391 (400th)	793.4 (2 $A\ g^{-1}$)	4
Si@$LiAlO_2$	CMC	83.6%	4	1106 (500th)	788.3 (8 $A\ g^{-1}$)	5
3D-Si@SiOx/C	PAA	80.1%	0.2	1635 (100th)	1342 (4 $A\ g^{-1}$)	6
mpSi/C	PVA	73.7%	0.5	729 (200th)	1036 (5 $A\ g^{-1}$)	7
N-G@Si@HSi	-	74.9%	2.1	1286 (1400th)	1036 (21 $A\ g^{-1}$)	8
Si@PPBT/CMC	CMC/PAA	84.39%	1.26	1793 (200th)	1036 (8.4 $A\ g^{-1}$)	9
	PANi-100	70.4%		1400 (200th)	680 (4 $A\ g^{-1}$)	
Si@SiO_x	CMC	-	1	750 (200th)	-	10
	PVDF	-		100 (10th)	-	
Si@CTSC	-	81%	1	1324 (200th)	1423 (5A g^{-1})	11
	PVDF	63%		234(20th)	0 (5A g^{-1})	
SiNDs@SSHC	CMC	88%.	0.3	1350 (500th)	512 (3.2 $A\ g^{-1}$)	12

References in Table S1

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