Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Achieving High Loading Si Anode via Employing Triblock Copolymer Elastomer Binder, Metal Nanowires and a Laminated Conductive Structure

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Table S1. Summary of recent researches that related to Si-based anodes with different Si loading.

Anode Material	Current density[mAg^{-1}]	Si areal loading [$mgcm^{-2}$]	Capacity retention[$mAhg^{-1}$]	Areal capacity[$mAhcm^{-1}$]	Reference& Year of publication
Si	300	3	400 at 60 cycles	1.2	Liu et al @ 2005[1]
Crystalline Si	150	1.957	1100 at 70 cycles	2.15	Li et al @ 2007[2]
Nano-Si	162	0.34-1.02	600 at 160 cycles	0.21-0.61	Beatti et al @ 2008[3]
SiO	100	0.89-1.7	700 at 50 cycles	0.62-1.19	Komaba et al @ 2011[4]
Si/graphite	50	1	870 at 30 cycles	0.87	Komaba et al @ 2012[5]
Si nanowires	300	1	2000 at 100 cycles	2	Chockla et al @ 2012[6]
Nano-Si	6000	0.2-0.3	540 at 5000 cycles	0.11-0.16	Wu et al @ 2013[7]
Nano-Si	375	0.22 or 0.3	~3000 at 50 cycles	0.66-0.9	Wu et al @ 2013[8]
Si microparticle	400	0.5-0.7	2094 at 90 cycles	1.05-1.47	Wang et al @ 2013[9]
Si/graphite	100	0.6	750 at 30 cycles	0.45	Han et al @ 2013[10]
Nano-Si	1720	0.2	3440 at 400 cycles	0.688	Ryou et al @ 2013[11]
Nano-Si	716	2.5	2000 at 100 cycles	5	Nguyen et al @ 2013[12]
Nano-Si/C fibers	60	1.5	1200 at 50 cycles	1.8	Radvanyi et al @ 2014[13]
Nano-Si	480	0.9-1	1200 at 680 cycles	1.08-1.2	Delpuech et al @ 2014[14]
Si/graphite	5600	1.5	526 at 300 cycles	0.79	Yim et al @ 2014[15]
Nano-Si	763	1.2	1834 at 100 cycles	2.2	Shao et al @ 2014[16]
Nano-Si	200	0.8-1	1450 at 100 cycles	1.16-1.45	Shen et al @ 2014[17]
Nano-Si	4000	1.5	1825 @ 300 cycles	2.74	Song et al @ 2014[18]
Si microparticle	329	1.0-1.1	2837 at 200 cycles	2.84-3.12	Zhang et al @ 2014[19]
Nano-Si	2142	0.6	1471 at 200 cycles	0.88	Jeong et al @ 2014[20]
Si/graphite	400	1	725 at 50 cycles	0.725	Chen et al @ 2014[21]
Nano-Si	480	0.9-1.1	2500 at 50 cycles	2.25-2.75	Hernandez et al @ 2015[22]
SiO	100	1.32	~1000 at 150 cycles	1.32	Zhao et al @ 2015[23]

Nano-Si	420	0.15-0.3	2300 at 180 cycles	0.345-0.69	Park et al @ 2015[24]
Nano-Si	326	0.46-0.7	2823 at 50 cycles	1.3-1.98	Kim et al @ 2015[25]
Nano-Si	283	0.5-0.6	2406 at 120 cycles	1.2-1.44	Chen et al @ 2015[26]
Si/graphite	575	2.9-3.1	~500 at 50 cycles	1.45-1.55	Nguyen & Oh @2015[27]
Si/graphite	1376.7	0.8	751 at 100 cycles	0.6	Lim et al @ 2015[28]
Si/graphite	100	0.48-0.8	~1040	0.5-0.83	Aoki et al @ 2015[29]
Nano-Si	144	0.75	2150 at 20 cycles	1.61	Dufficy et al @ 2015[30]
Si/graphite	22	1.7-1.8	~800 at 85 cycles	1.36-1.44	Klamor et al @ 2015[31]
Nano-Si	800	2	800 at 195 cycles	1.6	Uchida et al @ 2015[32]
Nano-Si	1200	0.2-0.6	1313 at 300 cycles	0.26-0.79	Choi et al @ 2015[33]
Nano-Si	120	0.5-0.54	1119 at 30 cycles	0.56-0.6	Zhu et al @ 2015[34]
Nano-Si	3500	0.3	~2500 at 200 cycles	0.75	Jeong et al @ 2015[35]
Nano-Si	1000	1.5	2000 at 100 cycles	3	Higgins et al @ 2016[36]
Nano-Si	434	0.4	~2000 at 100 cycles	0.8	Xu et al @ 2016[37]
Nano-Si	100	1.25-1.46	1639.6 at 50 cycles	2.05-2.4	Luo et al @ 2016[38]
Nano-Si	210	2	1711 at 50 cycles	3.42	Lee et al @ 2016[39]
Nano-Si	420	0.87	2907.6 at 100 cycles	2.53	Zhao et al @ 2016[23]
Nano-Si	300	1.5	2043 at 100 cycles	3.06	Zhang et al @ 2016[40]
Nano-Si	179	0.6	~3000 at 20 cycles	1.8	Nguyen et al @ 2016[41]
Nano-Si	280	0.4	~1100 at 100 cycles	0.44	Zhong et al @ 2016[42]
Nano-Si	1790	0.8-0.9	1200 at 100 cycles	0.96-1.08	Gao et al @ 2017[43]
Nano-Si	1193	0.4	1897 at 50 cycles	0.76	Xu et al @ 2017[44]
Nano-Si	1167	0.65	1958 at 80 cycles	1.27	Yoon et al @ 2017[45]
Si@TiO_2	2100	0.8	990 at 1500 cycles	0.79	Jin et al @ 2017[46]
Nano-Si	420	1	~2500 at 50 cycles	2.5	Karkar et al @ 2017[47]
Nano-Si	1500	0.4-0.5	1936 at 150 cycles	0.77-0.97	Bie et al @ 2017[48]

Nano-Si	840	2	2565 at 300 cycles	5.13	Wang et al @ 2017[49]
Nano-Si	1167	0.6	605 at 1000 cycles	0.36	Yuca et al @ 2017[50]
Nano-Si	1790	0.4	1597 at 200 cycles	0.64	Lü et al @ 2018[51]
Nano-Si	1000	2.2	~1840 at 60 cycles	4.04	Zeng et al @ 2018[52]
Nano-Si	100	0.6-1	2000 at 100 cycles	1.2-2	Shan et al @ 2018[53]
Nano-Si	8000	0.6	1350 at 1600 cycles	0.81	Lee et al @ 2018[54]
Nano-Si	480	2	1700 at 200 cycles	3.4	Hernandez et al @ 2018[55]
Nano-Si	~360	1.0-1.1	~2800 at 100 cycles	2.8-3.08	Zhu et al @ 2018[56]

Table S2. The number-average molecular weight (Mn) and Polydispersity (PDI) of each block.

	PS	SMA	SMAS
Molecule weight (Mn)	22429	121911	133826
Polydispersity (PDI)	1.3	4.2	4.8
Monomer conversion rate	92.1%	97.7%	95.6%

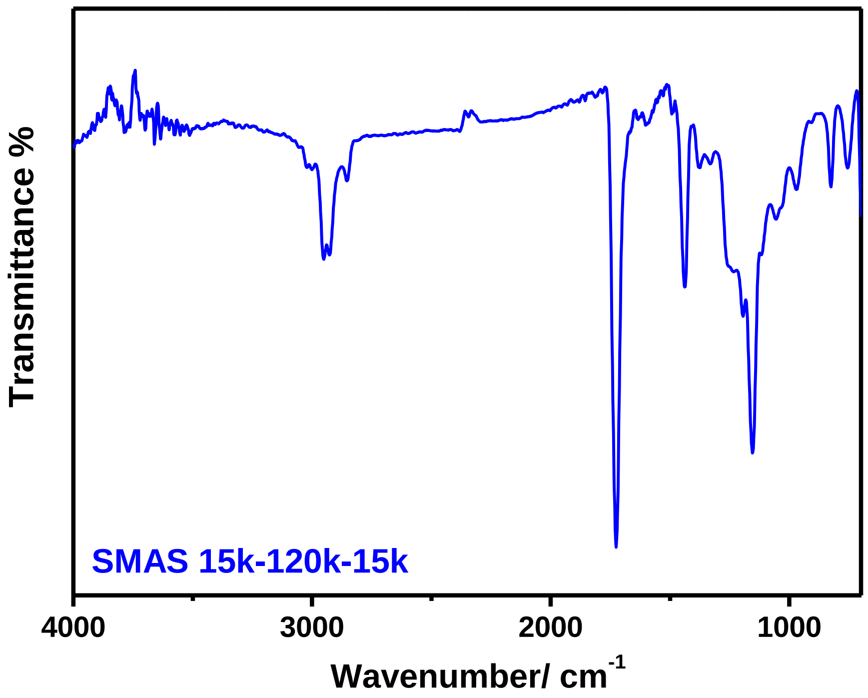

Fig. S1† The Fourier transform infrared (FTIR) spectroscopy test of SMAS.

Table S3. The electrolyte uptake of PAA and SMAS.

	m_1 (g)	m_2(g)	△m(g)	Weight increase (%)
PAA	1.1355	1.5666	0.4311	37.97
SMAS	1.0158	2.5964	1.5806	155.60

Detailed calculations about Specific energy density of Si/ $Li(Ni_xCo_yMn_z)O_2$ (x+y+z=1) (Si/NCM811) full cell at various capacity retentions.

Equation S1: $\boldsymbol{E = FQ_CU}$

E: The Specific energy density.

F: The estimate of the fraction of anode mass that is active material.

Q_C: Normalized to mass of active material (Si or graphite).

U: 3.6V for Si/ $Li(Ni_xCo_yMn_z)O_2$ (x+y+z=1) (Si/NCM811) full cell, 3.6V for graphite/$LiCoO_2$ (commercial LIB).

Equation S2: $\boldsymbol{F = \frac{t}{f_1 t + f_2}}$

t: The areal mass loading of active material in anode.

f_1: The unit mass of all cell components for each unit areal mass of active material loading.

f_2: The unit mass of cell components that scale without the mass of the active material.

Table.S4 Mass of battery components that scales with anode active material loading.

Battery component	$\boldsymbol{f_1: mg\ mg^{-1}}$	
	Si-NCM811	Commercial LIB
Electrolyte	0.35	0.35
Binder	0.35	0.11
Conductive additive	1.05	0.03

Cathode material	23.3	2.33
Anode material	1	1
Total f_1	26.05	3.82

Anode formula: Nano Si: Super P: (PAA: SMAS) =63%: 22%: (7.5%: 7.5%)

Cathode formula: NCM811: Super P: PVDF=92%:3%: 5%

Table S5. Mass of battery components that scales with anode active material loading.

Battery component	$f_2: mg\ cm^{-2}$	
	Si-NCM811	Commercial LIB
Al foil (16μm)	4.3	4.3
Cu foil (11μm)	9.9	9.9
PP separator (25μm)	1.0	1.0
Electrolyte in separator	1.3	1.3
Total f_2	16.5	16.5

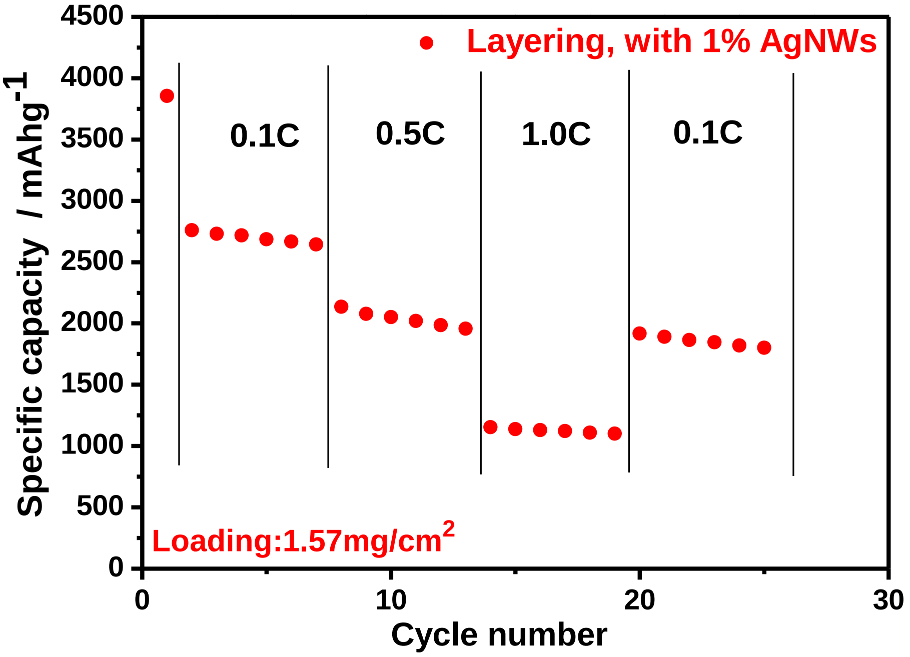

Fig. S2† The rate performance of composite Si anode with “laminated conductive structure” at different current densities (from 0.1 C to 1 C). The mass fraction of AgNWs in the whole electrode is 1%.

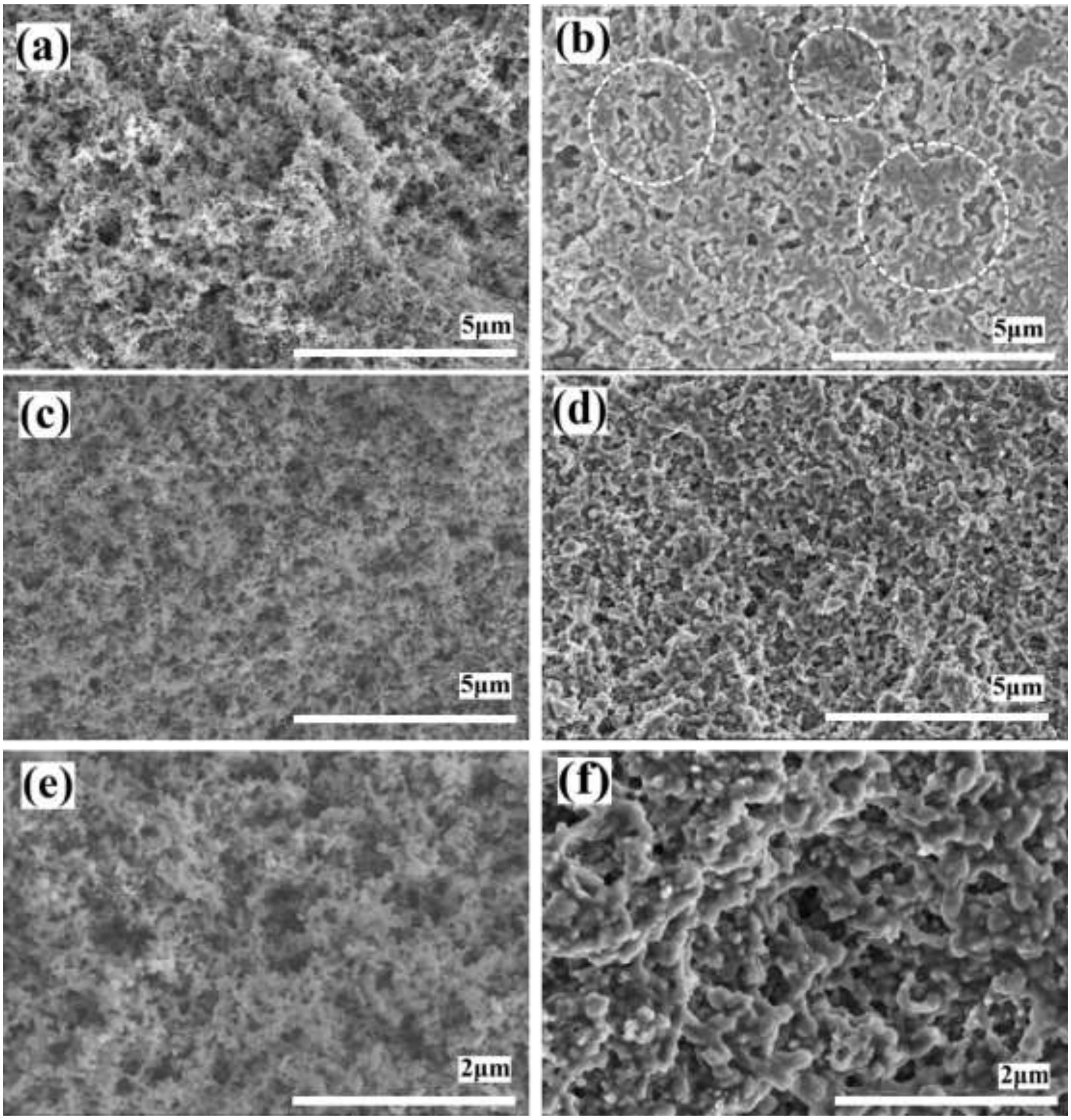

Fig. S3† SEM images of the surface morphology of the Si anode at a relatively high Si loading (about 1.5 mg cm^{-2}) (a)mixed with AgNWs before cycles, (b)mixed with AgNWs after 100cycles, (c) layered with AgNWs before cycles, and (d) layered with AgNWs after 100 cycles. The SEM images of the surface morphology of the Si anode layered with AgNWs at high Si loading (5.31 mg cm^{-2}) (e)before and (f) after 100 cycles.

Calculations about Li-ion diffusion coefficient of the Si electrodes with different binders (using PAA/SMAS, or PAA solely).

Equation S3: $\boldsymbol{Z' = R_s + R_{ct} + \sigma\omega^{-1/2}}$

$\boldsymbol{Z'}$: imaginary resistance; $\boldsymbol{\omega}$: angular speed; $\boldsymbol{\sigma}$: Warburg factor.

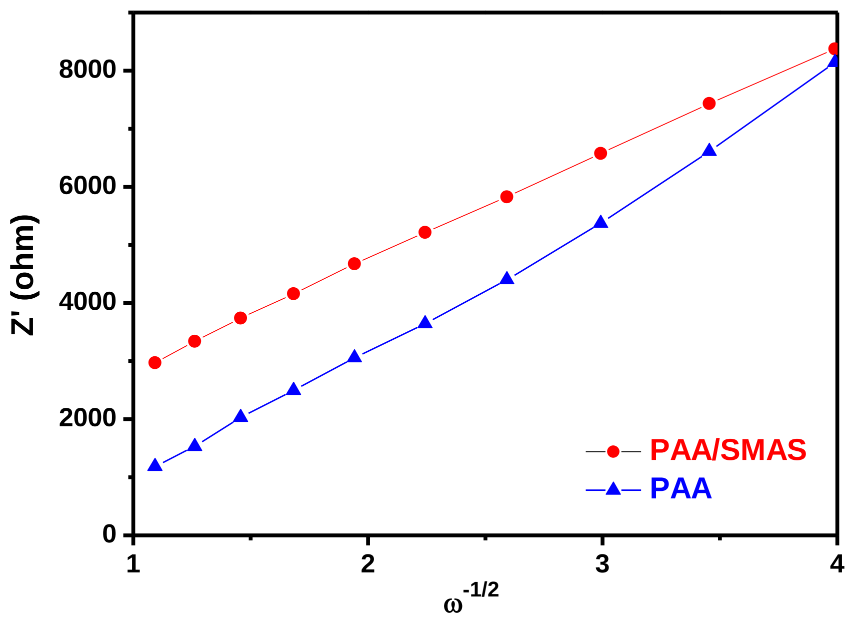

Fig. S4† Plot of imaginary resistance as function of inverse square root of angular speed for Si anodes with different binders (using PAA/SMAS, or PAA solely) after 100 cycles.

Warburg factor ($\boldsymbol{\sigma}$) for Si anodes with PAA binder and PAA/SMAS binder is 1528.82, and 2345.41, respectively.

Equation S4: $\boldsymbol{D_{Li^+}} = \frac{\boldsymbol{R^2T^2}}{\boldsymbol{2A^2n^4F^4c^2\sigma^2}}$

Table S6. The ion diffusion coefficient of the Si electrodes with different binders (using PAA/SMAS, or PAA solely) after 100 cycles.

Sample	$\boldsymbol{\sigma}$	$\boldsymbol{D_{Li^+}}$ **(cm^2 s^{-1})**
Electrodes with PAA binder	2345.41	~5.18×10^{-12}
Electrodes with PAA/SMAS binder	1528.82	~1.22×10^{-11}

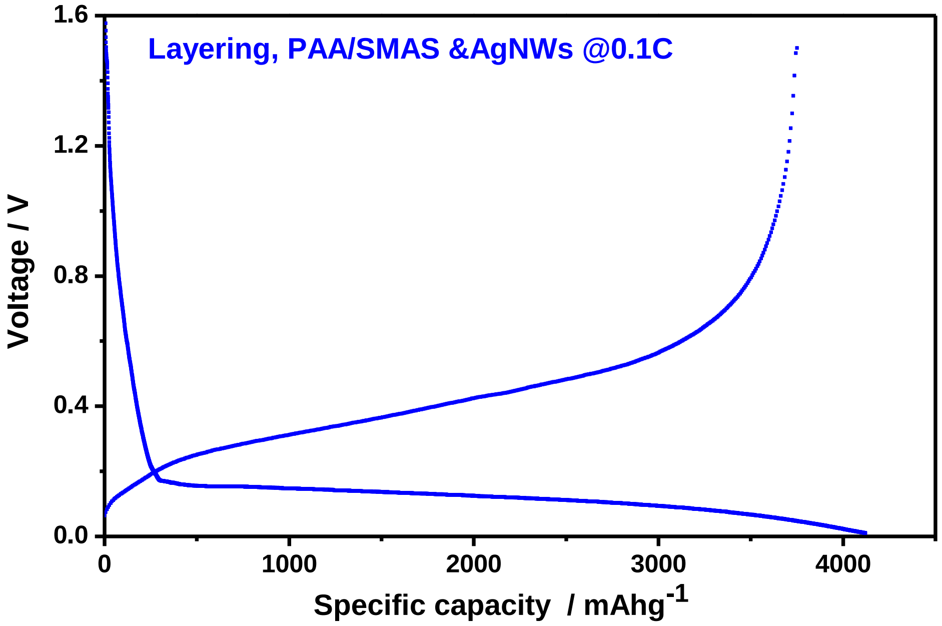

Fig. S5† The voltage profiles of composite Si anode with "laminated conductive structure" at loading of 5.31 mg cm^{-2} for the first discharge-charge cycle. The initial coulomb efficiency is 91.1%.

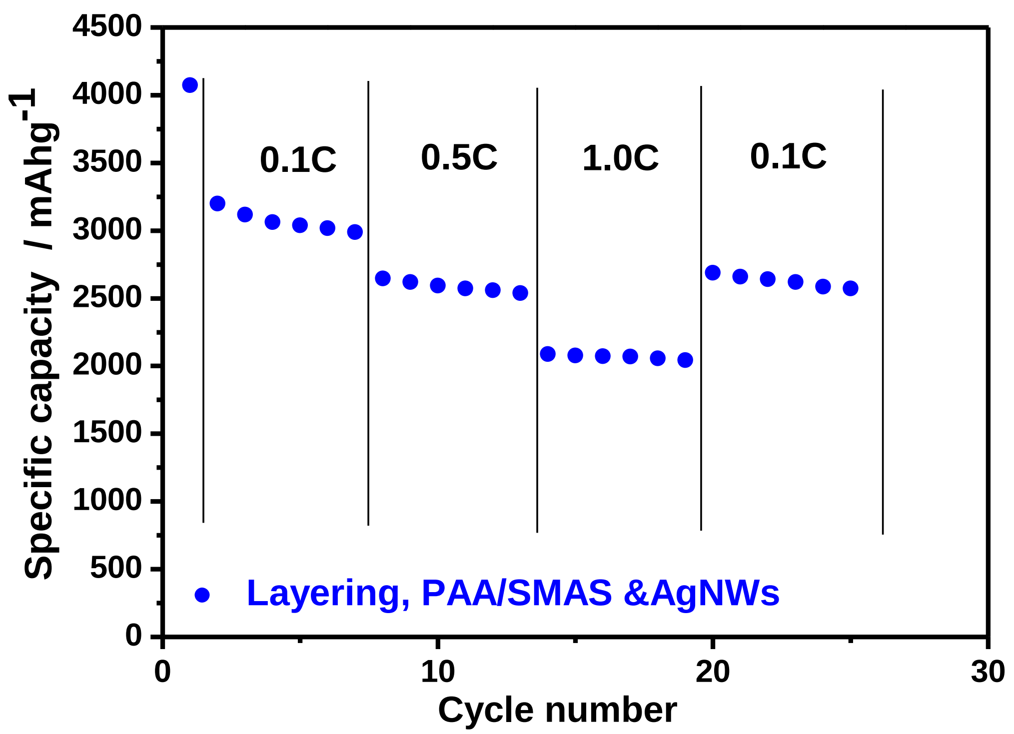

Fig. S6† The rate performance of composite Si anode with "laminated conductive structure" at loading of 5.31 mg cm^{-2} at different current densities (from 0.1 C to 1 C).

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