Supplemental Information

Oxidized Starch as a Superior Binder for Silicon Anodes in Lithium-ion Batteries

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Experimental

Preparation of electrodes

Silicon nanoparticles (50-200 nm, Alfa-Aesar), Super P (40nm, Timical) and binders were mixed in a 6:2:2 weight ratio in water. After stirred for 5 h, the slurry was coated on a Cu foil current then further dried at 70 °C in vacuum for 8 h. The foil was cut to Φ 12 mm sheets to assemble cells. CMC was purchased from Aladdin and SA from Aldrich. OS was purchased from Dongguan Dongmei Starch Co. Ltd. For OS, polymer was firstly dissolved in water at 75 °C for 5 min before adding silicon nanoparticles and Super P.

Cells assembling and electrochemical tests

The electrochemical performances of the as-prepared anodes were tested via CR2016 coin cells with ENTEK ET20-26 as separator, and pure lithium foil as counter electrode. The cells were assembled in an argon-filled glove box (MB-10 compact, MBraun) using 1M LiPF₆/EC+DMC (1:1 by volume, ethylene carbonate (EC), dimethyl carbonate (DMC)) as electrolyte, including 10% Fluoroethylene carbonate (FEC). The cycling performances were evaluated on a LAND battery test system (Wuhan Kingnuo Electronics Co., Ltd., China) at 25 °C constant current densities with the cut-off voltage of 0.1/1.2 V vs Li/Li⁺. The specific capacity was calculated on the basis of the weight of silicon nanoparticles in electrodes. *Morphology and structure characterization*

The SEM images of the electrodes were observed by a FEI Nova SEM 230 ultra-high resolution FESEM. The Fourier transform infrared (FTIR) spectrum of the sample was recorded on a FTIR spectrometer (Bruker VECTOR22). To evaluate the binder strength of electrode film, an electrode sample in 20 mm width and 100 mm length was attached to 3M

tape (12mm wide), and the peeling strength of the electrode specimens was measured with a high-precision micromechanical test system (FMT-310A5, Alluris).



Figure S1 Nyquist plots of OS electrode after 5, 20 and 35 cycles with 0.5 A g⁻¹ lithiation and 2.0 A g⁻¹ delithiation.

Table S1 Comparison of the electrochemical performances for Si electrodes with
different binders. (Si is short for commercial silicon nanoparticles)

Type of binder	Electrode	Active	Electrode	Areal	Ref.
	composition	materials	performance	capacity	
		loading			
OS	60Si:20Super	0.8mg cm ⁻²	1904 mA h g ⁻¹ @ 0.5	1.52 mA h	Our
	P:20binder		A g ⁻¹ lithiation+2 A	cm ⁻²	work
			g ⁻¹		
			delithiation,120th		
Ca ²⁺ + SA	53Si-C:18carbon	Unknown	1822 mA h g ⁻¹ @	unknown	1
	black:29binder		0.42A g ⁻¹ , 120th		
Self-healing	50SiMP:6.5carbon	0.5-0.7 mg cm ⁻²	~2000 mA h g ⁻¹ @	1.0-1.4 mA h	2
binder	black:43.5binder		0.4 A g ⁻¹ , 100th	cm ⁻²	
NaPAA-g-CMC	60Si:20Super	0.45 mg cm ⁻²	1816 mA h g ⁻	0.817 mA h	3
	P:20binder	_	¹ ,100th	cm ⁻²	
Gum arabic	50Si:25Super	0.3 mg cm ⁻²	~2500 mA h g ⁻¹ @	0.75 mA h	4
	C65:25binder	_	0.4 A g ⁻¹ , 100th	cm ⁻²	
Pectin	60Si:20Super	1 mg cm ⁻²	~1500 mA g ⁻¹ @ 3 A	1.5 mA h	5
	P:20binder	_	g ⁻¹ , 100th	cm ⁻²	

Guar gum	52Si:18conductiv	1.1 mg cm ⁻²	~2000 mA g ⁻¹ @	2.2 mA h	6
	е		0.84 A g ⁻¹ , 30th	cm ⁻² , 30th	
	additive:29binder				
PAA/pullulan	60Si:20carbon	0.6 mg cm ⁻²	~1600 mA g ⁻¹ @	0.96 mA h	7
	black:20binder		0.2C lithiation+0.5C	cm ⁻²	
			delithiation, 100th		
Xanthan Gum	60Si:20Super	0.9 mg cm ⁻²	~1600 mA g-1@	1.44 mA h	8
	P:20binder		3.5A g ⁻¹ , 12th	cm ⁻² , 12th	
hyperbranche	60Si:20Super	0.8 mg cm ⁻²	~1500 mA g ⁻¹ @1.5	1.2 mA h	9
dβ-	P:20binder		A g ⁻¹ , 120th	cm ⁻²	
cyclodextrin					
polymer					

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