Electronic Supplementary Information

A high-performance alginate hydrogel binder for Si/C anode of Li-ion battery

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Si/C composite: Si nanoparticles (SiNPs) were purchased from Alfa Aesar (Tianjin, China) and used without any further treatment. In the preparation of Si/C composite material, 3.0 g citric acid was dissolved in 25 mL ethanol, and 0.3 g SiNPs were added into and stirred uniformly. The mixture after evaporation of ethanol was dried in a vacuum drier at 80 °C for 12 h. The Si/C composite material was obtained after pyrolysis of the dry mixture in a tubular furnace under a flow of argon first at 300 °C for 5 h then at 800 °C for 5 h with a heating rate of 5 °C min^{-1}. The morphology and composition of SiNPs and Si/C composite were characterized by scanning electron microscopy (SEM) (HITACHI S-4800), powder X-ray diffraction (XRD) (Philips X'Pert Pro Super X-ray diffractometer, Cu-Ka radiation). To determine the content of Si in the Si/C composite material, thermogravimetric analysis (TGA) (TG 209-F1) measurements were conducted in air condition. The samples were heated from 30 to 800 °C at a rate of 10 °C min^{-1}.

Alginate hydrogel binder: $CaCl_2$ was dissolved in deionized water to form a solution of 0.26 g L^{-1}. Then Na alginate (SA) with a mass of 100 times of $CaCl_2$ was added and magnetic stirred overnight at room temperature. It is worthy to mention that SA (Maichao, Shanghai, China) used here is commercially available with low viscosity (1100 cP at 1 wt%). The alginate hydrogel binder was dried in a vacuum drier at 80 °C for 12 h for XRD and FTIR analysis. FTIR spectra were recorded on a FTIR spectrophotometer (Nicolet 330) in the range of 4000–400 cm^{-1} using KBr pellets.

Electrochemistry analysis: To prepare working electrode of coin cell, the active material of Si/C composite, carbon black, and binder with a mass ratio of 53:18:29 were mixed into homogeneous slurry. The obtained slurry was pasted onto pure Cu foil and dried in a vacuum drier at 80 °C for 12 h. After drying, the working electrode was assembled, in glove box filled with argon, into 2025 coin cell using Li metal foil as counter electrode with a microporous polymer separator (Celgard 2400) and a liquid electrolyte mixture (TINCI, Guangzhou, China) containing 1 mol L^{-1} $LiPF_6$ in EC/DMC/DEC

(1:1:1 vol%) with 2 wt% VC. The electrochemical performance evaluation of the as-prepared Si/C composite anode was carried out at room temperature on a battery test system (LAND 2001, 5 V 5 mA, Wuhan, China) with constant current charge-discharge cycling. The testing voltage range of the coin cell was between 0.01 V and 1.2 V. All potentials presented in this study were quoted versus the Li/Li^+ scale.

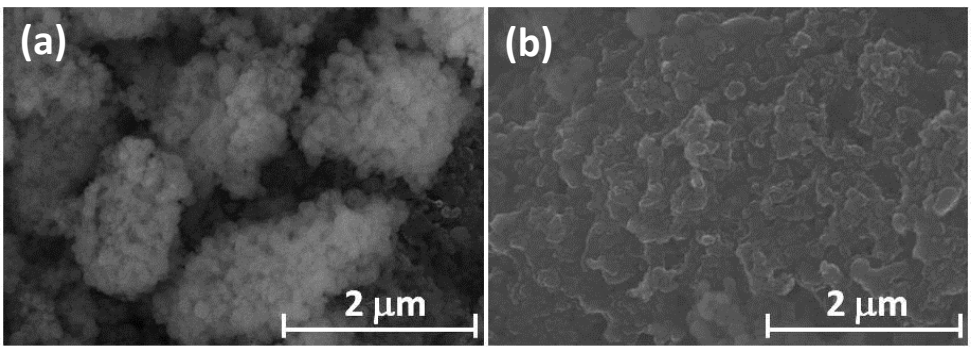

Figure S1 SEM images of pristine electrodes with (a) pure SA binder and (b) alginate hydrogel binder.

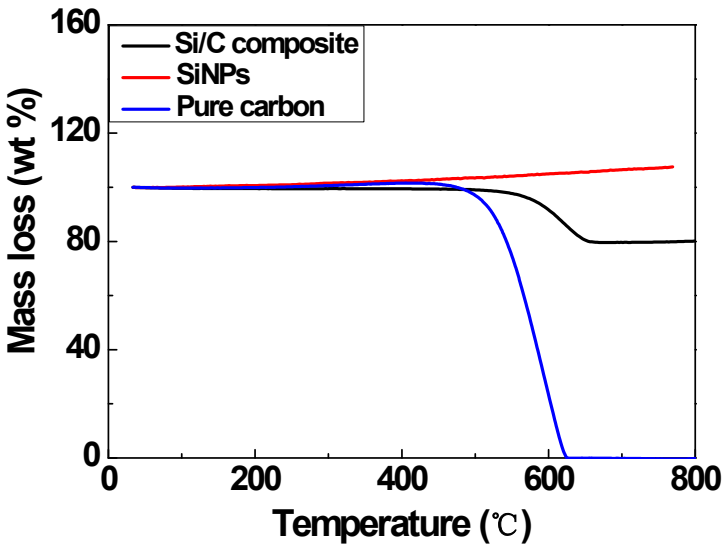

Figure S2 TGA curves of SiNPs, Si/C composite and pure carbon in air condition from 30 to 800 °C at a heating rate of 10 °C min^{-1}.

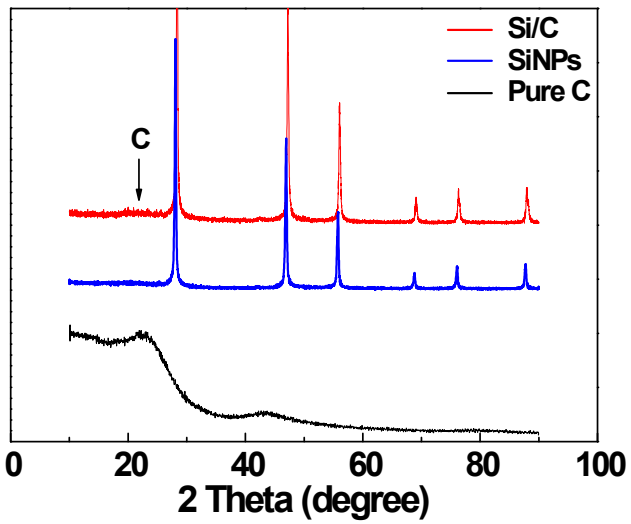

Figure S3 XRD patterns of SiNPs, Si/C composite and pure carbon.

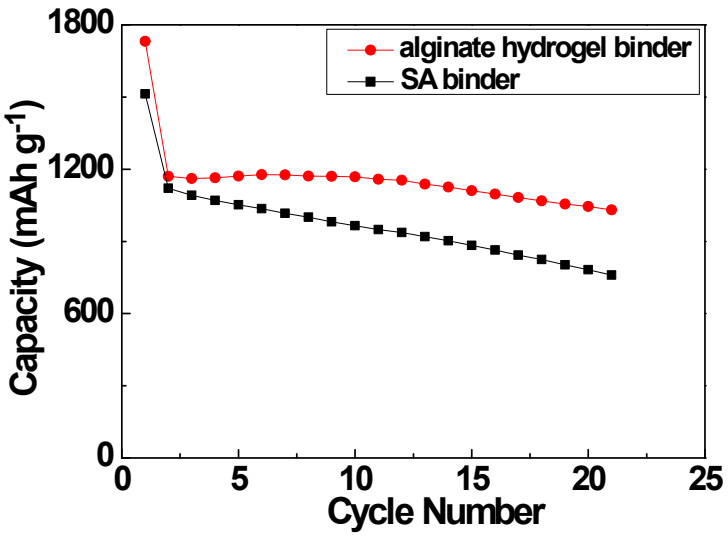

Figure S4 Cycle performances of Si/C composite electrodes with pure SA binder and alginate hydrogel binder at 840 mA g^{-1} .The mass ratio of active material of Si/C composite, carbon black, and binder is 70:15:15.

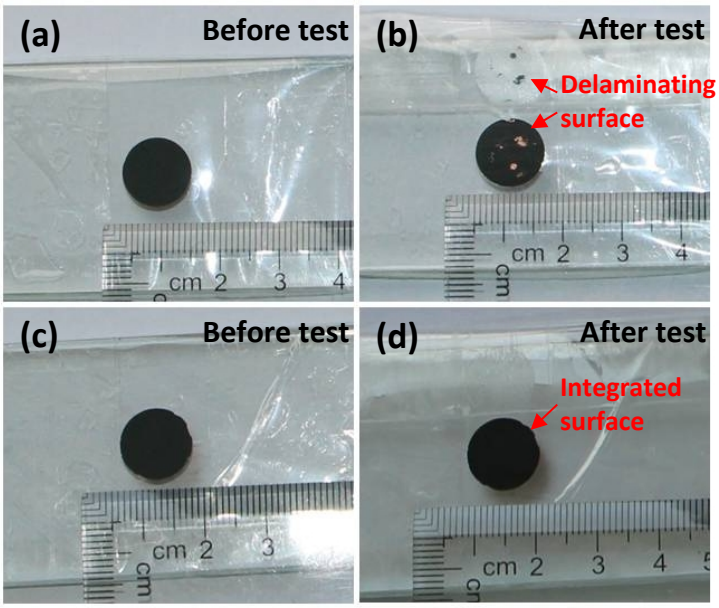

Figure S5 Morphology changes on Si/C electrodes with SA binder (a, b) and alginate hydrogel binder (c, d) after the adhesion tests. The back of Si/C electrode was pasted on sellotape, and the other face of Si/C electrode was also pasted using sellotape, and then the sellotape was peeled off to observe whether the Si active materials were peeled off. An integrated surface instead of delaminating surface is observed on Si/C electrode with alginate hydrogel binder.

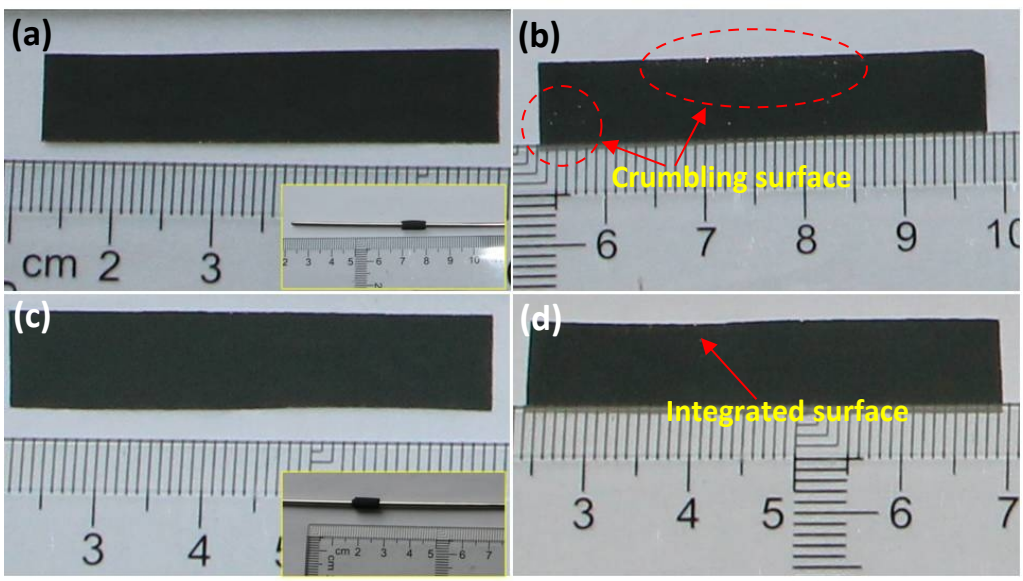

Figure S6 Morphology images of Si/C electrodes with SA binder (a) before winding test, (b) after winding test, and with alginate hydrogel binder (c) before winding test, (d) after winding test. Insets are images of Si/C electrodes with two kinds of binders wound on reels with a diameter of 1.90 mm,

respectively. No distinct cracks is observed on the electrode with alginate hydrogel binder.

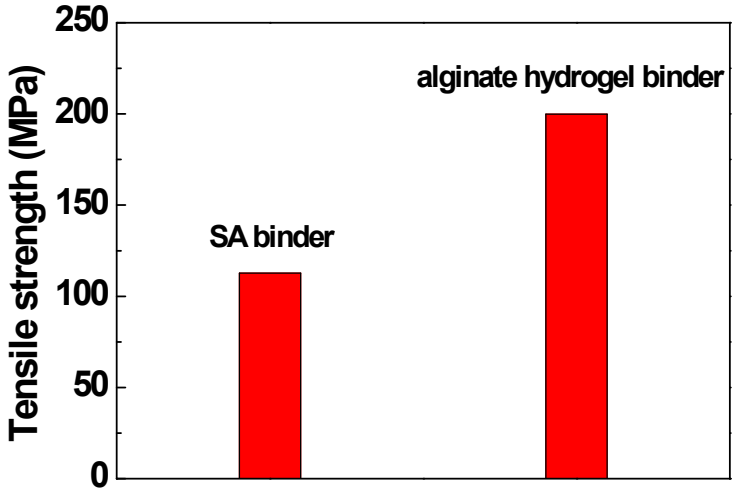

Figure S7 Tensile strength tests of SA binder and alginate hydrogel binder on a tensile strength tester (UTM4102, Sansi Zongheng, Shenzhen, China) with a drawing speed of 100 mm min^{-1}. It indicates that the alginate hydrogel binder shows a tensile strength of 199.9 MPa, which is 1.77 times that of SA binder (112.7 MPa).

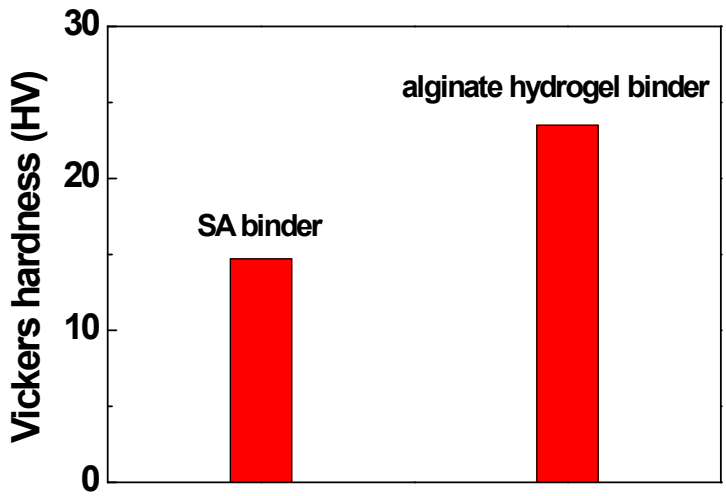

Figure S8 Vickers hardness tests of SA binder and alginate hydrogel binder on a stiffness tester

(MHV-20002C, Taiming, Shanghai, China) under the conditions of force = 25 gf, dwell = 15 s. It indicates that the dried alginate hydrogel binder shows a higher Vickers hardness (23.5 HV), which is 1.60 times that of SA binder (14.7 HV).