Support information

Carbonized polymer dots functionalized 3D conductive network binder for high-performance silicon anodes

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Experimental

Materials

Carboxylated chitosan (CS, carboxylation >80%) and Xanthan gum (XG) was purchased from Shanghai Yuanye Biotechnology Co. Epichlorohydrin (ECH) were purchased from McLean (Shanghai). Nanosilicon (80-100 nm), conductive carbon black (Super P), and negative cell shells were purchased from Keruder (Guangdong Candlelight New Energy Technology Co., Ltd.).

Synthesis of CX, CXD and CXDE Binder

Dissolve 1.2 g CS and 0.2 g XG in 26.6 g deionized (DI) water to form a 5wt.% CX binder. The 5 wt.% CX binder was then transferred to a polytetrafluorethylene (Teflon) -lined autoclave and heated at 150 °C for 12 hours. The obtained dark brown solution is CXD binder. Then CXDE binder was obtained by adding 0.05 g ECH to CXD binder and reacting at 80 ℃ for 1 h.

Preparation of nano silicon electrode and assembly of battery

The slurry is prepared by dispersing the silicon: binder in deionized water with a mass ratio of 9:1. The mixture was stirred by ball milling for 15 min (at a rotational speed of 300r/min and a ball-to-material ratio of 1:10), and then cast onto copper foil using traditional scraping techniques to prepare the anode. After vacuum drying the anode in a 90 °C vacuum oven for 12 hours, the anode was made into a 12mm diameter sheet, which was then made into a CR2032 button battery and placed in a glove box filled with Ar. The electrolyte contains an EC/DEC (v/v 1:1) solution of 1.0 M LiPF6 and a 10 wt% addition of fluorocarbon oxide ethane (FEC), polypropylene (PP) as a diaphragm, and lithium sheets as a counter electrode.

Characterization and Measurements

High-resolution transmission electron microscopy (HRTEM) was used to record the distribution of carbon points in the binder, and the particle size was estimated by Image J software. Scanning electron microscope (SEM) images were obtained using field emission scanning electron microscopy (Zeiss Utral 55 and Hitachi SU8010). The photoluminescence spectra of different binders were measured in the steady state/transient fluorescence spectrometer. Ultraviolet-visible absorption spectra are taken on an ultraviolet-visible infrared spectrometer. X-ray photoelectron spectroscopy (XPS) is implemented on the Thermo Scientific K-Alpha X-ray photoelectron spectrometer. FTIR spectra were obtained to examine changes in functional groups.

In order to characterize the binding of the binder to the collector, the contact Angle between the binder and the collector (copper foil) was measured. The contact Angle was measured statically on a surface photoelectric (SEO) instrument and calculated by SEO Surfaceware-9. The slurry with the same solid content was prepared, and the dispersibility of the slurry prepared with different binders was measured by qxd type scraper fineness meter. The hardness and elastic modulus of each type of electrode were obtained by measuring the electrode with nanoindentation instrument. The tensile test and 180° stripping test were carried out on the universal test machine. The tensile test was carried out at a strain rate of 20 mm/min. For the 180° peel test, a 2.4 cm wide and 5 cm long electrode sample was attached to a 3 M tape and the peel strength of each

electrode sample was measured at a rate of 20 mm /min and a tension of 5 N.

On the battery test system (LAND, Wuhan, China), the first five pre-cycle constant current charge-discharge tests were performed at a rate of 0.05 C from 0.01 V to 5 V. The subsequent cycle rate is 0.2 C. In the battery test system (Shenzhen Sunway, China), five multiple tests were carried out at 0.1 C, 0.2 C, 0.5 C, 1 C, 0.1 C. Different binders are applied directly to the copper foil, then the battery is assembled and CV tested. Cyclic voltammetry (CV) was tested from the open circuit voltage at a scan rate of 0.05 mV/s in the range of 0.01 to 1.5 V. The electrochemical impedance spectroscopy (EIS) data with a voltage amplitude of 5 mV from 0.01 Hz to 100 kHz were recorded using the CHI 660E electrochemical workstation. Long cycle tests were conducted at 0.2 ° C. The N/P ratio of the electrode under test was 1:1, the electrode density of silicon was 1.1g/cm^3, and the initial surface capacity was 2528.9 mA h g^{-1}.

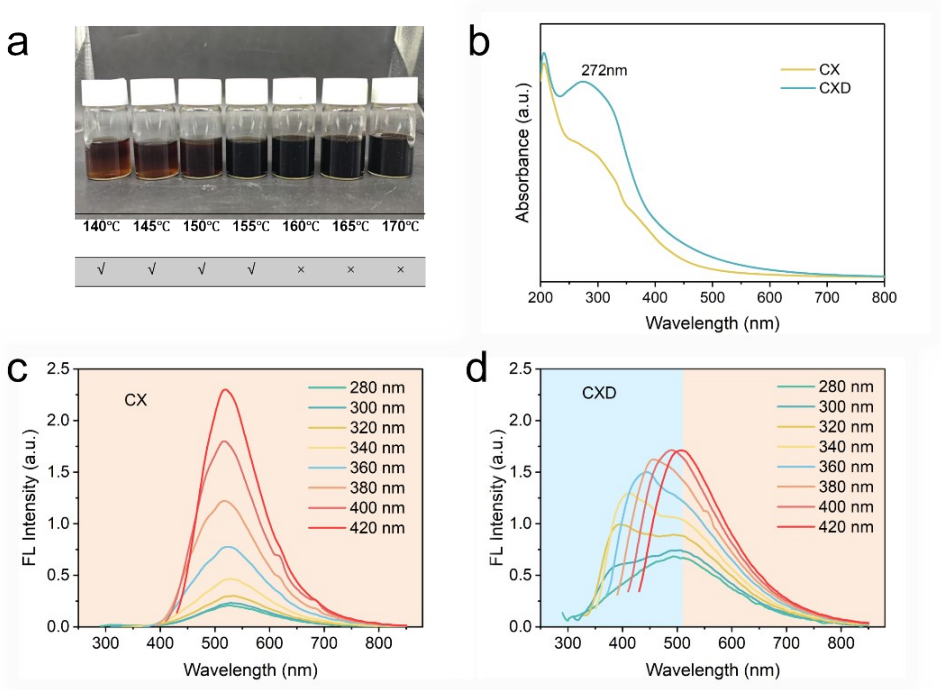

Figure S1 (a) Digital photos of CXD binder obtained by hydrothermal treatment at different temperatures and whether it is capable of chemical cross-linking. (b) UV-vis absorption spectra of CX and CXD. Photoluminescence emission spectra of CX (c) and CXD (d) after the same diluting in aqueous solutions under different excitation wavelengths display the contrast directly, the emission peak under 320 nm excitation with the highest emission intensity was used as the normalization reference. The light blue and orange background colors distinguished the different

parts of the the spectra (the light blue: excitation-independent emission, the light orange: excitation-dependent emission).

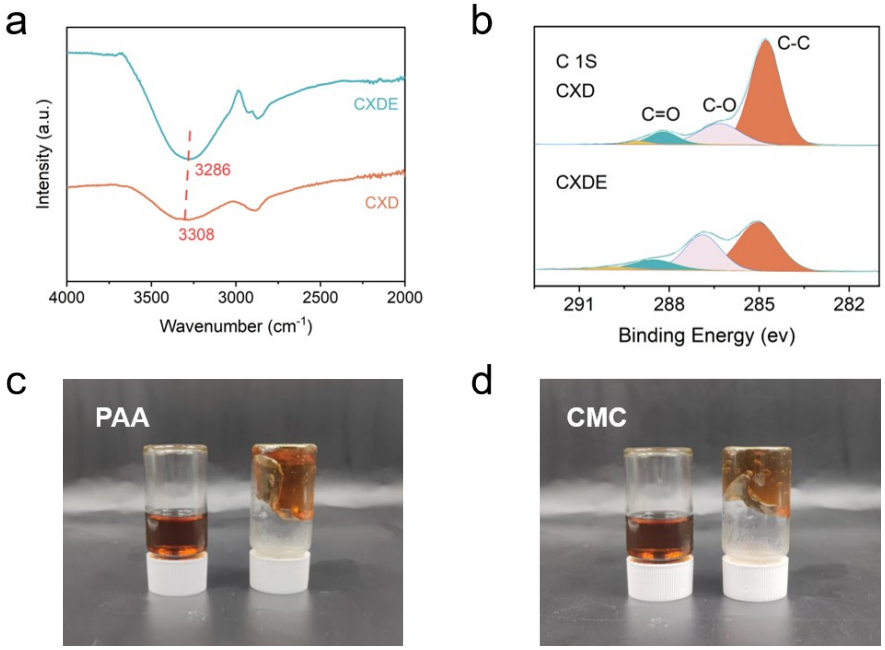

Figure S2 (a) FTIR spectra of the CXD and CXDE samples. (b) High-resolution XPS spectra of C 1s on the CXD and CXDE samples. (c) Digital photos of PAA after hydrothermal treatment and cross-linking. (d) Digital photos of CMC after hydrothermal treatment and cross-linking.

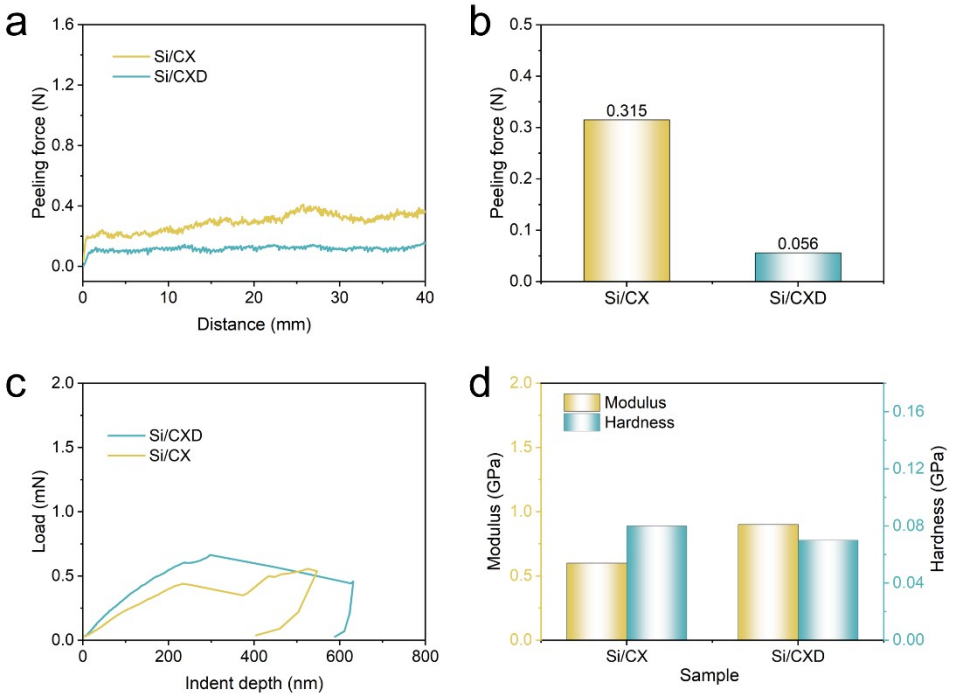

Figure S3 (a) Force–displacement curve. (b) Peeling strength. (c) Load–displacement curve of electrode. (d) Graphs of electrode strength and modulus of elasticity.

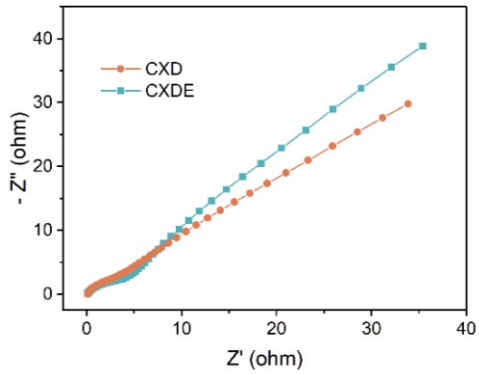

Figure S4 EIS spectra of Si electrodes with various binders after 3 cycles.

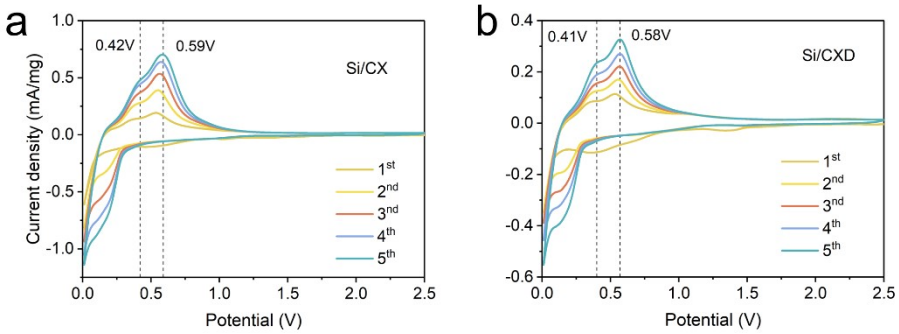

Figure S5 (a) Cyclic voltammetry curves of Si/CX. (b) Cyclic voltammetry curves of Si/CXD.

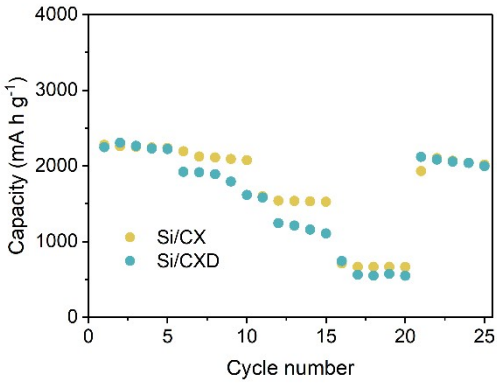

Figure S6 The rate capacity of Si/CX and Si/CXD.

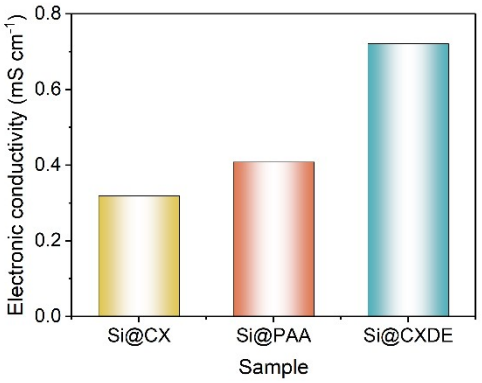

Figure S7 The electrical conductivity of the electrode sheet prepared by binder and silicon.

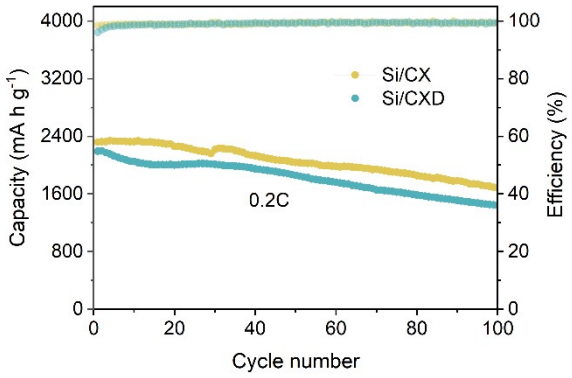

Figure S8 Cycling performance of Si/CX and Si/CXD at 0.2C.

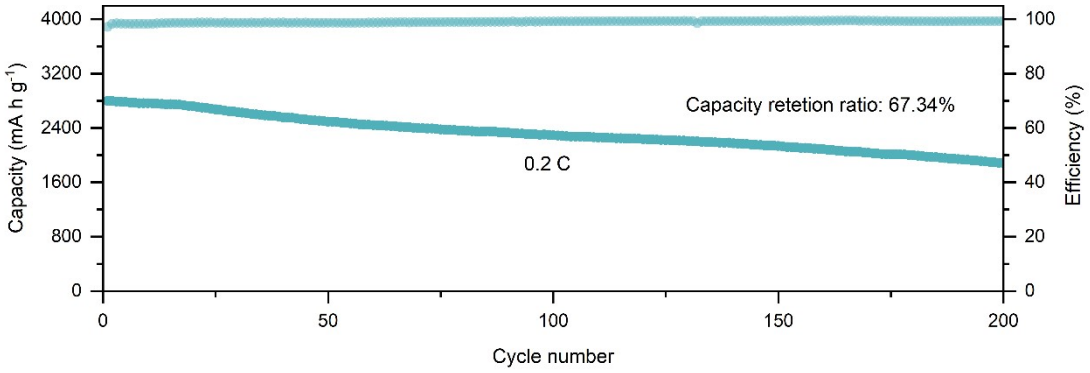

Figure S9 Cycling performance of Si/CXDE at 0.2C.

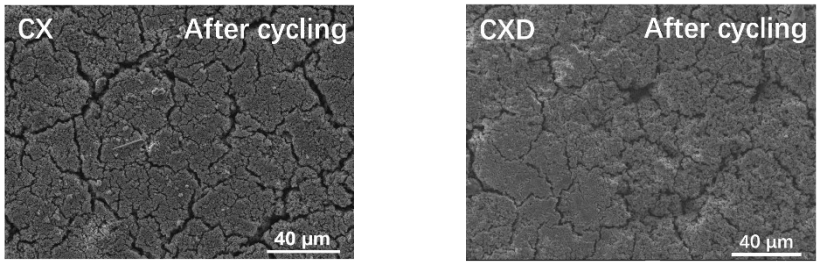

Figure S10 Surface SEM images of Si/CX (a) and Si/CXD (b) after 50 cycles.