

**Fluidized bed reaction towards crystalline-embedded-amorphous Si anode with  
much enhanced cycling stability**

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## 2. Experimental

### 2.1 Preparation of material

Si nanoparticles were prepared by chemical vapor deposition in a modified fluidized bed reactor (Fig. S1). Silane and hydrogen (Zhongning Materials Technology Co., Ltd., Zhejiang, China) were mixed with a molar ratio of 3:20, which was injected into a tube reactor, and the pressure was kept at 0.3 MPa. Si solid particles formed the nucle and grew larger in the gas phase. The fluidizing gas carried the Si powder to the cooling and filtration systems. The decomposition temperature was controlled at 500 °C, and the corresponding products were denoted as crystalline-amorphous silicon nanoparticles (c-a-Si-500). Silicon nanoparticles with different crystalline degrees were prepared at 600 and 700 °C (c-a-Si-600 and c-a-Si-700). Crystalline Si nanoparticles (c-Si, Shuitian ST-NANO Science & Technology Co.,Ltd, Shanghai, China) were used for comparison purpose.

### 2.2 Characterization

Powder X-ray diffraction (XRD, Rint-2000, Rigaku) was used to identify the crystal structure. Raman spectra from 400 to 600  $\text{cm}^{-1}$  were obtained on a WiTec Alpha300 system applying 632.8 nm laser light. The microstructure of the sample was observed by scanning electron microscopy (SEM, MAIA3, TESCAN, USA) and transmission electron microscope (TEM, Tecnai, G12). The electronic conductivity of the prepared samples was measured using a multifunction digital four-probe tester (RTS-8). The chemical components were analyzed by X-ray photoelectron

spectroscopy (XPS, VG Multilab 2000).

### 2.3 Electrochemical tests

The electrochemical performance of the samples was evaluated via 2025 coin type cells. The working electrodes were prepared by mixing the active material, Super P, and sodium alginate (Sigma-Aldrich Co.) in a weight ratio of 6:2:2. The mixed slurry was spread onto copper foil and dried in a vacuum oven at 60 °C for 5 h. After that, the electrode was punched into disk with a diameter of 12 mm. The mass loading of the electrode was 0.6 mg cm<sup>-2</sup>. Lithium foil was used as the counter electrode and a Whatman GF/A glass microfiber filter was used as the separator. The electrolyte (Suzhou Fosai New Material Co., Ltd., China) consisted of ethylene carbonate (EC)/dimethyl carbonate (DMC) (1:4 in wt. ratio) containing 1 M LiPF<sub>6</sub> mixed with 2 wt. % fluoroethylene carbonate (FEC). The cells were assembled in an argon-filled glove box. Assembled half-cells were rested for at least 10 h until reaching a stable open-circuit voltage. Electrochemical discharge-charge tests were performed with a Neware battery cycler in the potential range of 0.01-2.00 V (vs. Li/Li<sup>+</sup>) at 25 °C. The electrochemical impedance spectra (EIS) experiments were performed with an Electrochemical Workstation (CHI604E, Chenhua, China) over a frequency range from 100 kHz to 0.01Hz. The cyclic voltammetry (CV) measurements were measured at a scanning rate of 0.1 mV s<sup>-1</sup>.

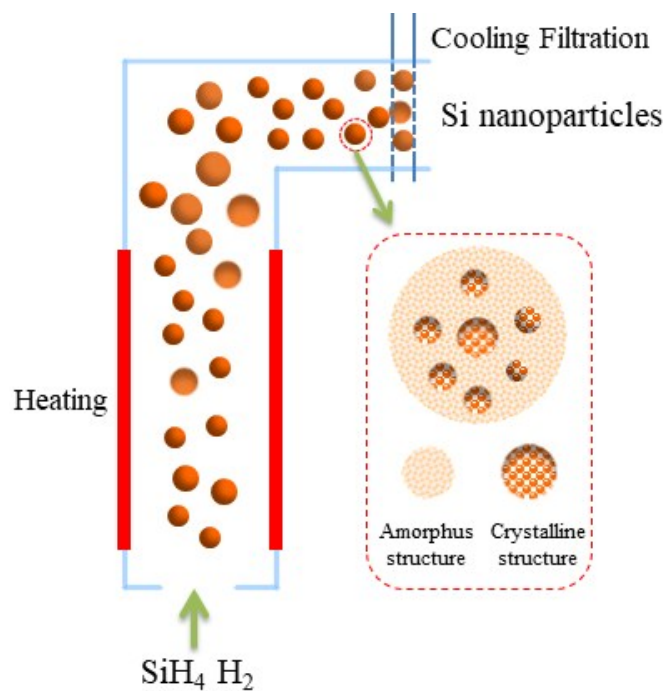


Fig. S1 Schematic illustration of the preparation process

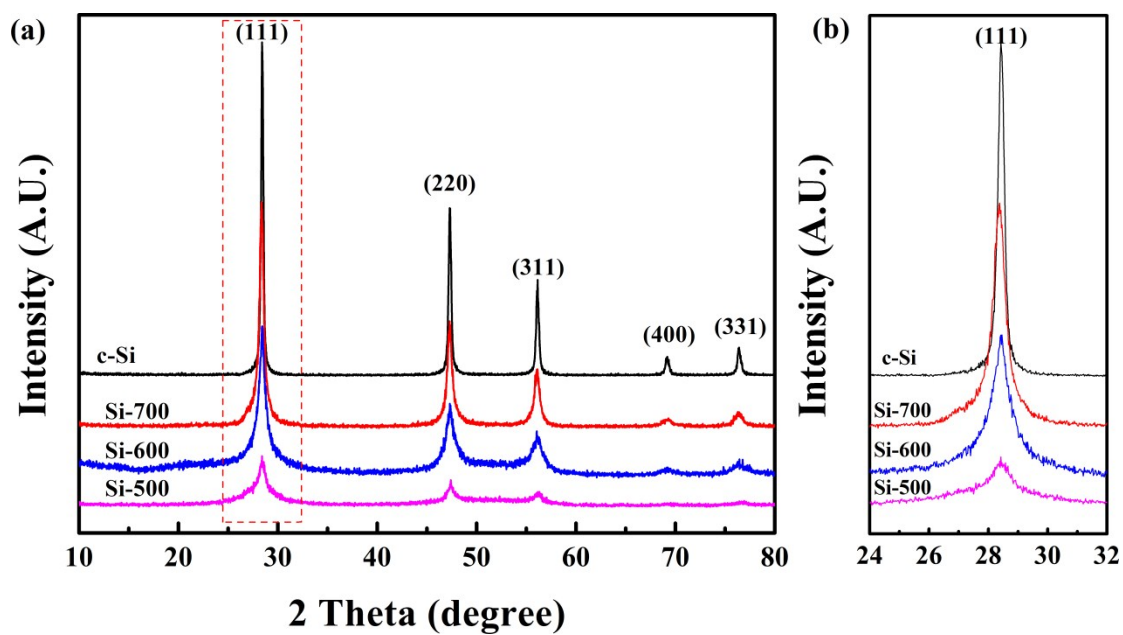


Fig. S2 (a) XRD patterns of c-Si and as-prepared samples; (b) The peaks of the (111) characteristic plane of the samples.

Table S1 crystal size and crystallinity of as-prepared Si samples

Samples	c-Si	c-a-Si-500	c-a-Si-600	c-a-Si-500
FWHM	0.18	0.73	0.58	0.47
D(nm)	43.96	10.83	14.39	16.83
$X_c$ (%) <sup>&amp;</sup>	100	20.6	29.7	62.8

(<sup>&</sup> $X_c = I_c / (I_c + I_a)$ , where  $X_c$  is the crystallinity;  $I_c$  is the integral strength of crystal diffraction peak;  $I_a$  is the integral strength of amorphous scattering peak.)

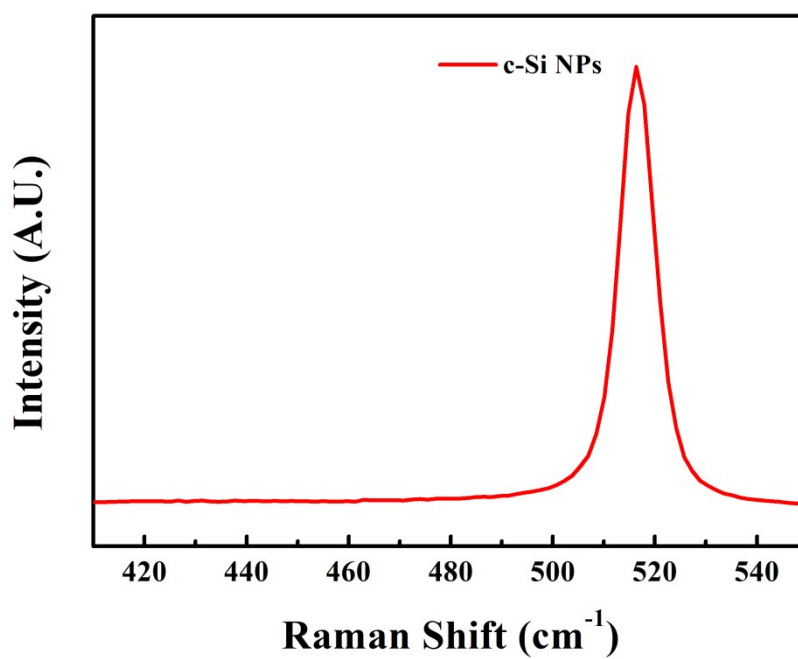


Fig. S3 Raman spectra of c-Si.

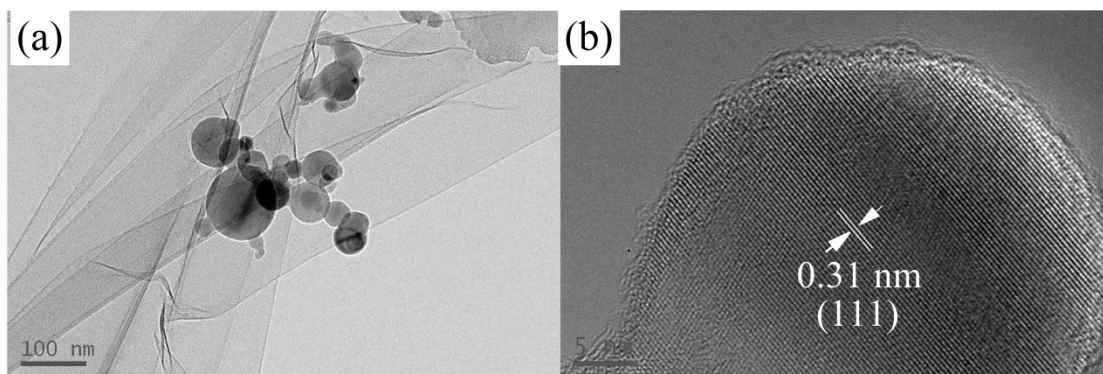


Fig. S4 TEM images of c-Si.

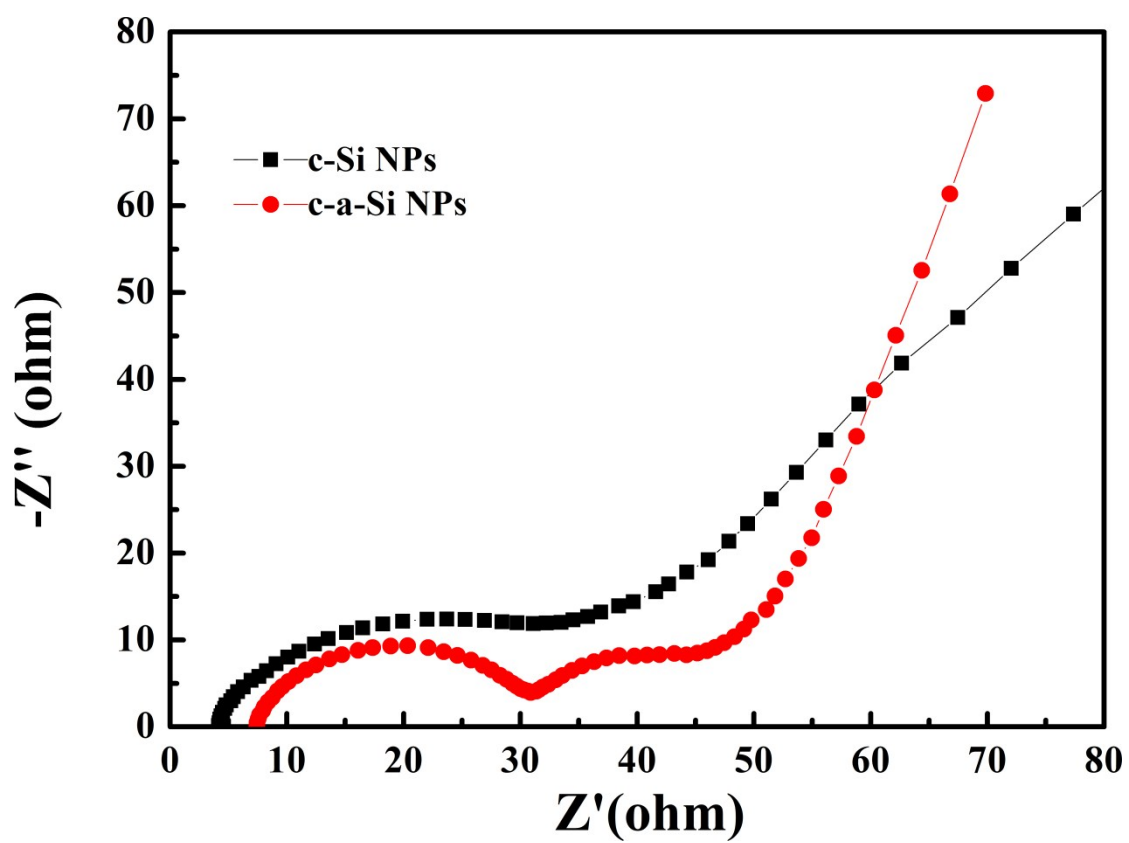


Fig. S5 EIS curves of c-Si and c-a-Si-500 at lithiation stage (0.12 V).

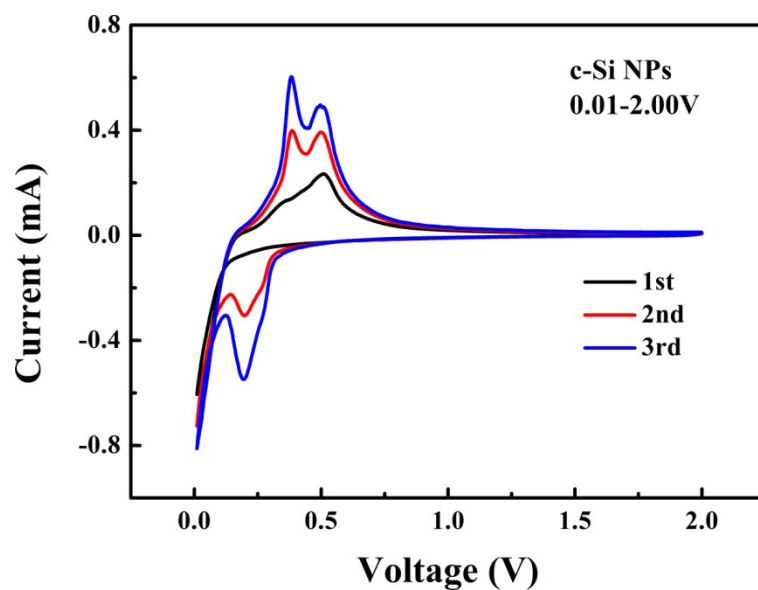


Fig. S6 The cyclic voltammety curves of c-Si electrode in the voltage range of 0.01-2.00 V

Table S2 The electronic conductivity of c-Si and c-a-Si

Sample	c-Si	c-a-Si-500	c-a-Si-600	c-a-Si-700
Electronic resistivity ( $\rho$ , $\text{k}\Omega\cdot\text{cm}$ )	0.592	4.078	2.586	1.015
Electronic conductivity ( $\sigma$ , $\text{S m}^{-1}$ )	0.169	0.025	0.039	0.098