# Supporting Information:

# Polydopamine Wrapping Silicon Cross-linked with Polyacrylic Acid as High-Performance Anode for Lithium-Ion Batteries

Yitian Bie [†], Jun Yang [*, †], Xiaolin Liu [†], Jiulin Wang [†], Yanna Nuli [†] and Wei Lu [‡]

[†] School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai, 200240, China

[‡] Department of Chemical Engineering, University of Michigan, Ann Arbor, MI 48109, USA

*Tel./Fax:+86 2154747667.

E-mail:yangj723@sjtu.edu.cn

**Experimental Details**

*Preparation of Si@PD nanoparticles*

Firstly, 0.08g Si nanoparticles (50-200 nm, Alfa-Aesar) and 0.08g dopamine hydrochloride (Alfa-Aesar) were dispersed in 80 ml Tris-HCl (10mM, pH=8.5) buffer solution and then stirred for 2 h at room temperature (ca. 25°C). Si particles coated by PD (Si@PD) were collected by centrifugation and washed by water for further use.

*Electrode fabrication*

Si@PD particles were mixed with Super P (40nm, Timical) and PAA (Mv~450 000, Aldrich) in an 8:1:1 weight ratio in water. After stirred for 5 h, the slurry was coated onto a Cu foil current collector and then further dried at 80 °C in vacuum for 8 h. The foil was cut to Φ12 mm sheets to assemble cells. The electrode samples were designated as Si@PD/PAA.

For comparison, silicon nanoparticles were directly used for the electrode preparation with PAA, CMC (Aladdin) or SA (Aldrich) binder. The weight ratios of Si: binder: Super P were 8:1:1. These electrode samples were described as Si/PAA, Si/CMC and Si/SA, respectively.

*Morphology and structure characterization*

The morphologies and microstructures of the samples were investigated by a FEI Nova SEM 230 ultra-high resolution FESEM and a JEM-2100F TEM (JEOL Ltd, Japan). Fourier transform infrared (FTIR) spectra of samples were recorded on a FTIR spectrometer (Bruker VECTOR22). The polymer weight fraction in Si@PD sample was determined by comparing the mass loss of Si@PD and Si after calcination by thermogravimetric analysis (TGA, TA 2050). The X-ray photoelectron

spectroscopy (XPS) analysis was performed using a Kratos Axis UltraDLD spectrometer (Kratos Analytical-A Shimadzu Group Company) with monochromatic Al Kα source (1486.6 eV). A 20 mm wide and 100 mm long electrode sample was attached to 3M tape, and the peel strength of the electrode specimens was measured with a high-precision micromechanical test system (FMT-310A5, Alluris).

*Cells assembling and electrochemical tests*

The electrochemical performances of the as-prepared anodes were tested using two electrode coin-type cells. The CR2016 coin cells were assembled in an argon-filled glove box (MB-10 compact, MBraun) containing less than 2 ppm water and $O_2$ using 1M $LiPF_6$/EC+DMC (1:1 by volume, ethylene carbonate (EC), dimethyl carbonate (DMC)) as electrolyte, plus 10% Fluoroethylene carbonate (FEC), ENTEK ET20-26 as separator, and pure lithium foil as counter electrode. The cycling performances were evaluated on a LAND battery test system (Wuhan Kingnuo Electronics Co., Ltd., China) at 25 °C constant current densities. The cut-off voltage was 0.01 V versus $Li/Li^+$ for discharge (Li insertion) and 1.2 V versus $Li/Li^+$ for charge (Li extraction). The specific capacity was calculated on the basis of the weight of active materials. The impedances of cells were measured by Metrohm Autolab PGSTAT302N after charging to 1.2 V in the frequency range of 10 kHz to 0.1 Hz.

**Table S1.** XPS Peak Binding Energy Assignments for N 1s regions.

| Functional group | Binding energy(eV) |
|---|---|
| -NRCO- | 399.6±0.1 |
| -NHR | 400.5±0.2 |
| -NH2 | 402.0±0.1 |

**Table S2.** Results of peeling tests for Si@PD/PAA electrode and Si/PAA electrode. (Speed: 100.0 mm $s^{-1}$)

| Sample | Force (N) |
|---|---|
| Si@PD/PAA | 0.614 |
| Si/PAA | 0.561 |

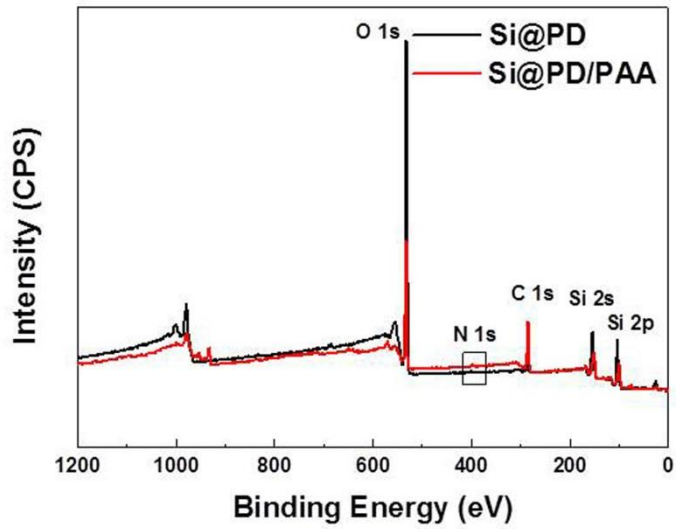


**Figure S1.** XPS spectra of all regions for Si@PD particles and Si@PD/PAA electrode. The high-resolution spectra of N 1s regions in the box are shown in Figure 2.c-d.

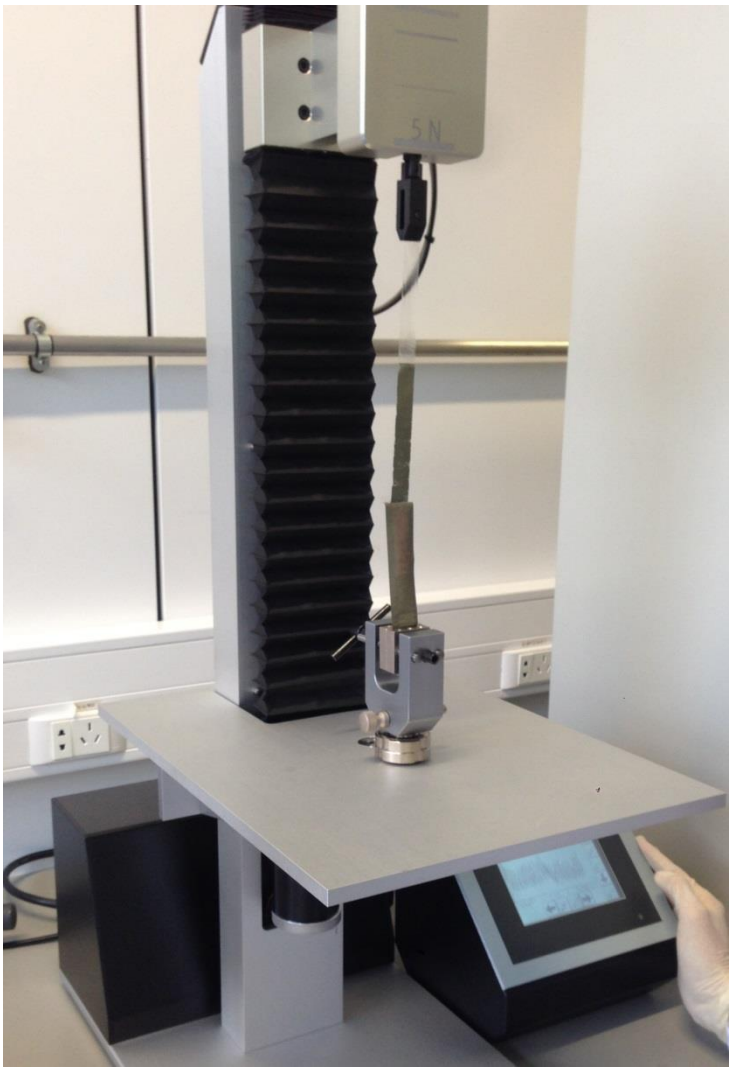


**Figure S2.** A photograph of the peeling test setup.

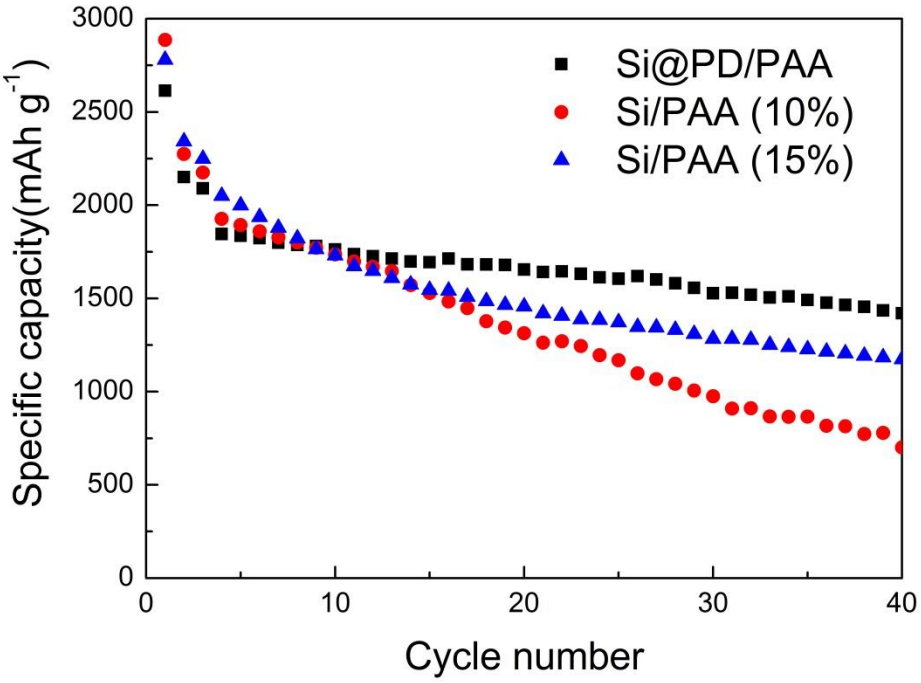


**Figure S3.** The cycle performance of Si@PD/PAA and Si/PAA electrodes. (Active materials loading: 2.0 mg $cm^{-2}$ for Si@PD/PAA and Si/PAA (10%), 1.3 mg $cm^{-2}$ for Si/PAA (15%))

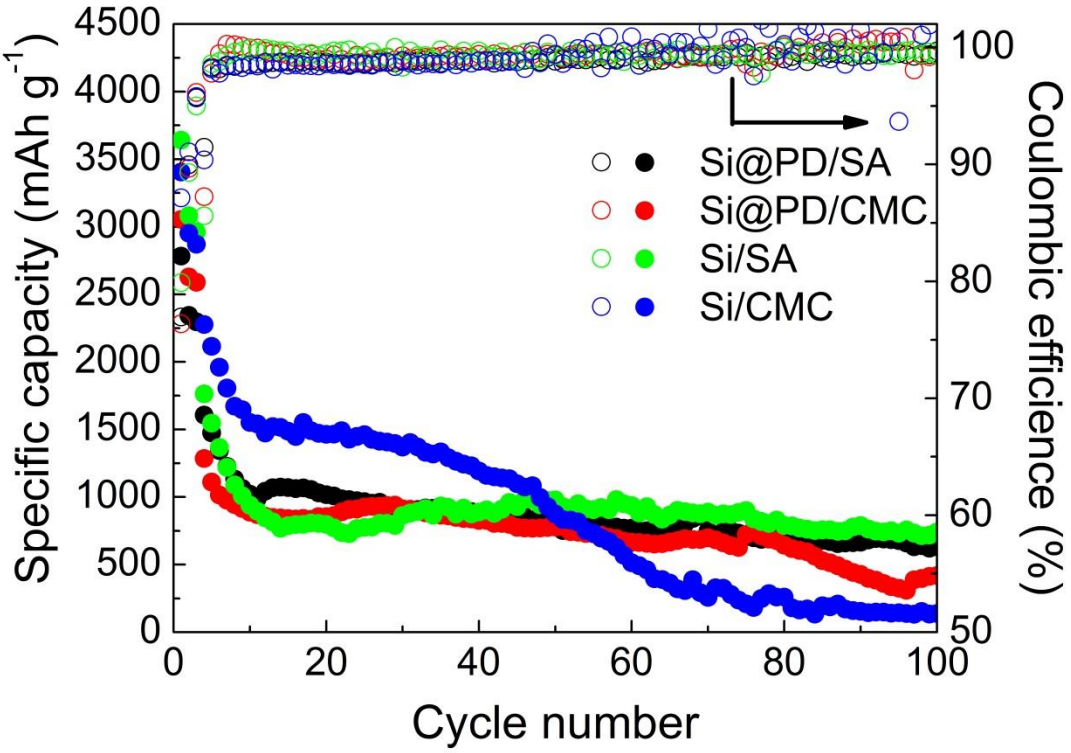


**Figure S4.** Cycling performance and Coulombic efficiencies of Si@PD/SA, Si@PD/CMC, Si/SA and Si/CMC electrodes at 1.5 A $g^{-1}$. (Si or Si@PD: Super P: binder=8:1:1, active materials loading: 0.5 mg $cm^{-2}$)

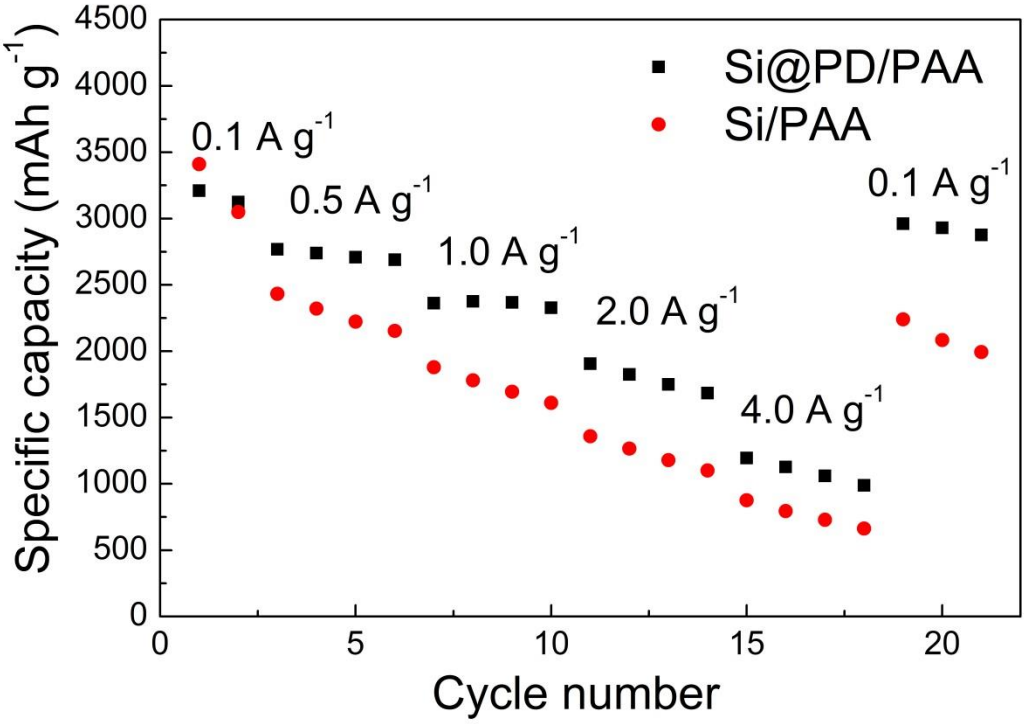


**Figure S5.** The rate performance of Si@PD/PAA and Si/PAA electrode (Si or Si@PD: Super P: binder=8:1:1, active materials loading: 0.5 mg $cm^{-2}$)

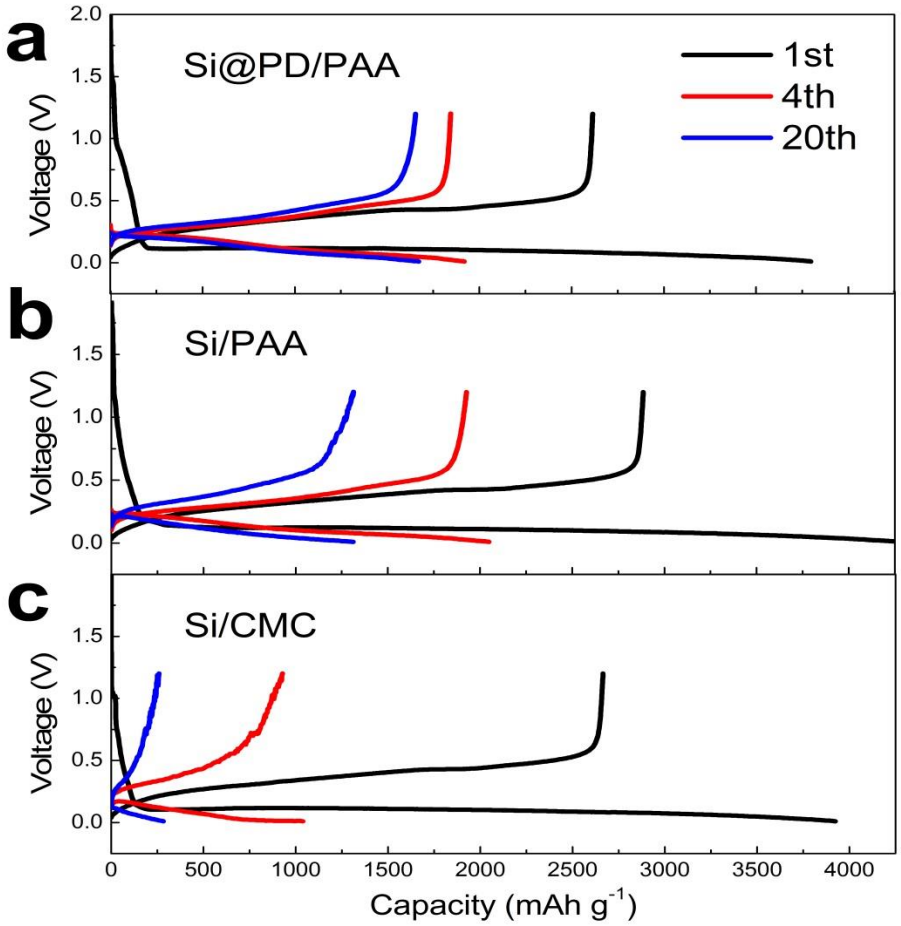


**Figure S6.** The charge and discharge curves of Si@PD/PAA (a), Si/PAA (b), Si/CMC (c) electrode with 2.0 mg $cm^{-2}$ active material loading. Current density: 0.1 A $g^{-1}$ for the first cycle and 0.5 A $g^{-1}$ for other cycles.

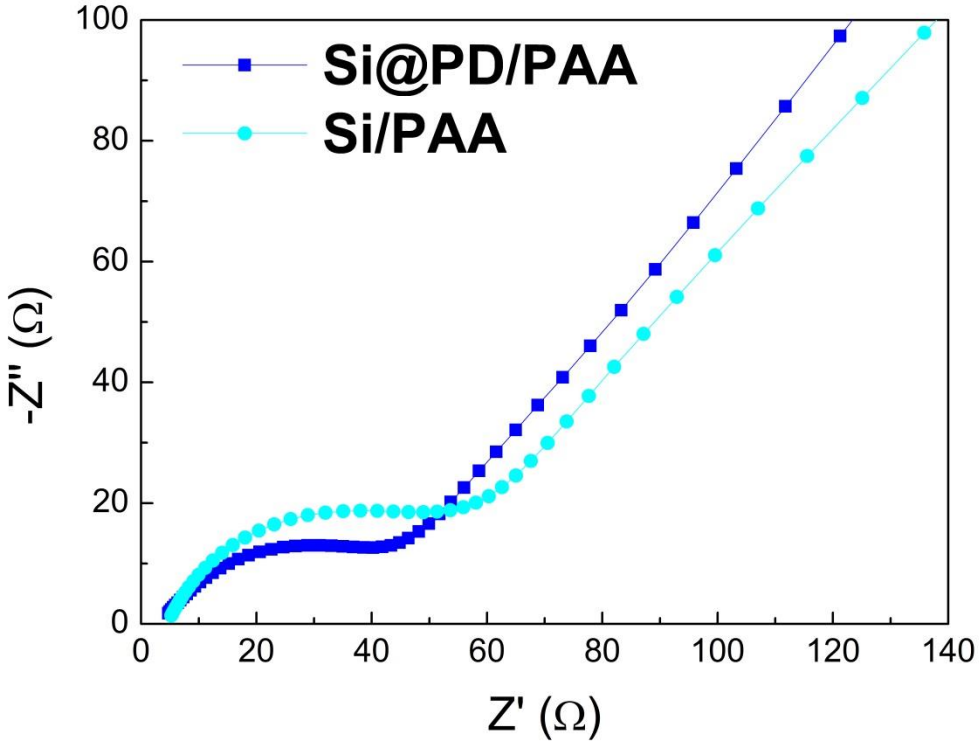


**Figure S7.** Nyquist plots of Si@PD/PAA and Si/PAA electrodes with 2.0 mg $cm^{-2}$ active materials loading after the fourth cycles.