

A dual functional wrapping layer for enhanced cyclic performance of high-loading silicon anodes



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ABSTRACT

This work presents an efficient surface coating layer, a polyacrylic acid (PAA) layer doped with silica nanoparticles, providing highly enhanced cyclic stability of silicon anodes, without any loss in rate capability. Electrochemical characterizations show that the modified electrode retains 81% of the original capacity after 100 cycles, compared with 47% for the pristine electrode. The improved cyclic stability is ascribed to the porous and robust surface coating layer that makes the active material in intimate contact with both current collector and conductive additive while inhibits continuous parasitic reaction between silicon and electrolyte. In addition, the porosity of this coating layer provides the diffusion pathway for Li⁺ and thus ensures good rate capability.

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1. Introduction

Silicon is widely considered as the most promising anode material for the next generation lithium-ion batteries, due to its natural abundance, moderate working potential, and extremely high specific capacity of 3579 mAh·g⁻¹ at room temperature [1–3]. However, silicon anode material is still confronted with one major challenge prior to its commercialization, which is exactly the huge volume expansion upon its lithiation (*ca.* 300%) [4,5]. The volume expansion can easily bring about the mechanical fracture of silicon particles after several cycles, which further leads to the following two consequences: firstly, the crushed silicon particles will become electronically isolated and lose intimate contact with current collector and conductive additive, resulting in rapid capacity fading; secondly, as the silicon particles fractures, the continuous exposure of fresh silicon surface to the liquid electrolyte will allow for the continuous parasitic reaction between electrolyte and silicon surface, giving rise to a very low Coulombic efficiency and poor cyclic performance [6–9].

In an effort to mitigate this problem, a myriad of attempts have been resorted to, which can be mainly sorted into two categories:

one is designing unique and complicated silicon micro/nano-structures, such as Si nanowires, nanosheets, nanospheres or nanotubes, etc., to accommodate huge expansion rate [10–14]; the other is wrapping silicon particle with a conductive layer through chemical vapor deposition (CVD), atomic layer deposition (ALD) or molecular layer deposition (MLD) methods to increase electronic conductivity and simultaneously inhibit the volume expansion [15–17]. An outburst of research has been focused on the above-mentioned two kinds of strategies, and both of them have demonstrated excellent effects. Nevertheless, they are almost unbearable in terms of the complexity of production process and the unimaginably high production cost [18–20]. It is noted that there has been another interesting strategy using a kind of in-situ structural change of silicon into porous or wrinkled frameworks to show a good stability, simply by charge/discharge cycles [21–23].

In this study, a robust and porous layer, *i.e.*, the polyacrylic acid (PAA) layer doped with an appropriate amount of silica nanoparticles was closely coated on the surface of the prepared electrode. Moreover, the operation process is much simpler than the aforementioned two strategies. Electrochemical tests manifest that cyclic stability of Si anode is greatly enhanced without sacrificing any rate capability after the coating process when the loading of Si is as high as *ca.* 1.2 mg·cm⁻². Besides, we have also discussed and analyzed the influence of the added amount of silica in the coating layer on electrochemical properties.

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2. Experimental

2.1. Pristine electrode preparation

A mixture of nano Si (60 wt%, provided by Alfa Aesar, size <50 nm), conductive agent (20 wt%, super P) and binder (20 wt%, sodium alginate, NaAlg) was dispersed in an aqueous solution and stirred for *ca.* 5 h. The as-formed slurry was then coated onto a piece of copper foil, which was then dried in a vacuum oven. The Si loading was typically *ca.* $1.2 \text{ mg}\cdot\text{cm}^{-2}$ for thick electrodes and *ca.* $0.3 \text{ mg}\cdot\text{cm}^{-2}$ for flimsy electrodes. The pristine electrode was denoted as S0.

2.2. Surface modified electrode preparation (Coating procedure)

First, 0.1 g PAA and different amounts of SiO_2 (0 g; 0.01 g; 0.1 g, provided by Evonik company, product name: SIPERNAT 350, structure: porous, size: *ca.* $1 \mu\text{m}$) were dispersed in 2 mL ethanol, forming a uniform dispersion. Some of the viscous ethanol dispersion was then coated on the surface of pristine electrode, followed by drying in a vacuum oven at 50°C . According to the mass ratio of SiO_2 to PAA, the working electrode samples were

labeled as S1 (0%), S2 (10%), S3 (100%), additionally, the loading of the coating layer was *ca.* $0.1 \text{ mg}\cdot\text{cm}^{-2}$, $0.1 \text{ mg}\cdot\text{cm}^{-2}$, $0.13 \text{ mg}\cdot\text{cm}^{-2}$ for sample S1, S2 and S3, respectively.

To further verify the effect of coating layer, an extra amount of PAA, which is equal to the amount in the coating layer, was directly mixed into the slurry, to form the electrode S4. In the electrode S4, the mass ratio of silicon nanoparticle to PAA binder was *ca.* 2: 1.

2.3. Characterization and electrochemical tests

The electrode morphologies before and after cycling were observed by a NOVA NanoSEM 230 field-emission scanning electron microscope.

Electrochemical properties were measured using CR2016-type coin cells. The coin half-cells were assembled in an argon-filled glove box (MBRAUN Unilab, Germany), using 1 M LiPF_6 dissolved in the mixture of dimethyl carbonate (DMC) and ethylene carbonate (EC) (with 1:1 volume ratio), plus 10% fluoroethylene carbonate (FEC), as the electrolyte, polyethylene (PE) membrane as the separator, and lithium metal as the counter electrode. Cyclic performance was evaluated on a LAND CT2001 battery test system within the potential window of 0.01–1.2 V (the initial three cycles

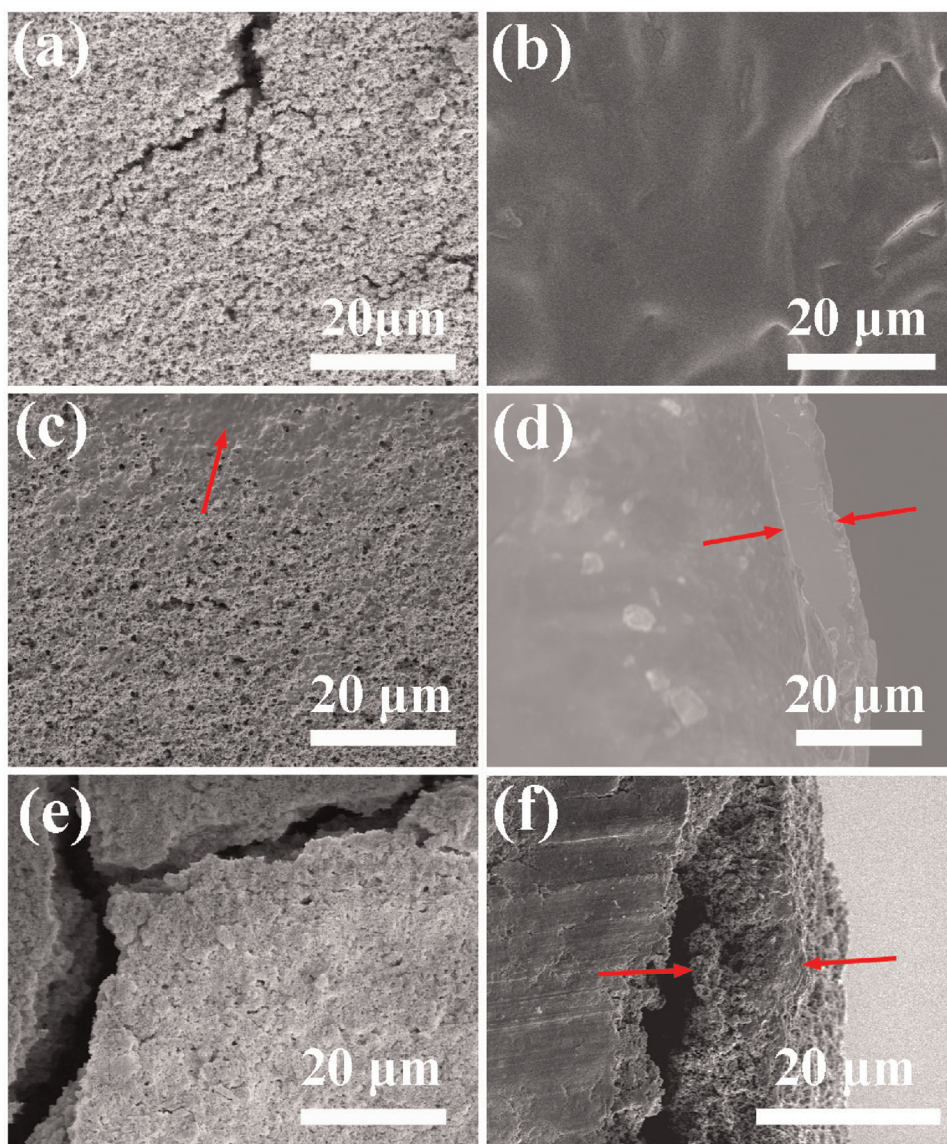


Fig. 1. SEM images of electrode S0 (a), S1 (b), S2 (c, d) and S3 (e, f) before cycling (mass loading was *ca.* $1.2 \text{ mg}\cdot\text{cm}^{-2}$). (d) and (f) are the side views.

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