Supporting Information For

Facile synthesis of SiO_x/asphalt membrane for high performance lithium-ion battery anode

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Experimental section

Materials synthesis

Unless otherwise specified, all materials used in the experiment were of analytical grade, and were used without any pretreatment. SiO_x powders were purchased from Hengshui Chaofan New Energy Materials Co. Ltd. Emulsified asphalt was composed of asphalt (60 wt%), water and emulgator. The porous microspheres were synthesized through a combined process of ball-milling and spray drying. Firstly, SiO_x powders (150 g) and water (850 g) were added into ball mill, followed by ball milling 6 h under N_2 atmosphere and low temperature (10 °C) to fabricate nano-sized SiO_x particles. Then, the slurry was utilized to produce porous SiO_x particles by spray drying process. The detailed parameters were presented as follows: inlet temperature (190 °C), outlet temperate (100 °C), feeding speed (5 L h^{-1}) and rotate speed of nebulizer (30000 r min^{-1}). The obtained SiO_x microspheres (20 g) were dispersed in water (15 mL) and ethyl alcohol (10 mL) under stirring. Then the SiO_x aqueous was mixed with emulsified asphalt aqueous (17 g) to prepare SiO_x/asphalt composite. SiO_x/asphalt membrane was obtained by hot pressing SiO_x/asphalt composite at 80 ^oC. The resultant SiO_x/C@Ni binder-free anodes were fabricated by preforming SiO_x/asphalt membrane and nickel foam and high-temperature pyrolysis process. SiO_x/C composites were prepared by carbonizing SiO_x/asphalt membrane. All high-temperature pyrolysis process was performed under Ar atmosphere at 900 °C for 3h. SiO_x/C/G composites were composed of SiO_x/C and artificial graphite with the mass ratio of 7: 13. For comparison, SiO_x/G anodes with the mass ratio of 1: 3 presented the same reversible capacities with SiO_x/C/G anodes.

Characterizations

The morphologies of all materials were investigated by a field-emission SEM (JEOL 6701F). XRD patterns were collecteded by a Rigaku D/max 2500 diffractometer using Cu Kα radiation. Raman spectra was obtained using a Digilab FTS3500 system. Thermogravimetric (TG) analysis was performed on a TA-Q60 instrument from 50 °C to 1000 ^oC at a heating rate of 5 ^oC min^{-1} under air. The electrochemical performance of anodes was investigated using CR2032 coin cell, which was composed of Li foil as

the counter electrode, a celgard 2500 as separator, electrolyte (1M $LiPF_6$ in a mixture of ethylene carbonate (EC) diethyl carbonate (DEC) and dimethyl carbonate (DMC) (1:1:1, by volume) containing 5% fluoroethylene carbonate (FEC)) and the working electrode. For binder-free anodes, SiO_x/C@Ni was used as the working electrode. For other anodes, the working electrode was prepared through coating a homogeneous slurry consisting of active materials, Super P, sodium carboxymethyl cellulose (CMC) and styrene butadiene rubber (SBR) at a mass ratio of 90:5:2.5:2.5 on the C-coated copper foil, and then dried in a vacuum oven at 60 °C for 12 h. For SiO_x anodes, the mass ratio of active materials, Super P, CMC and SBR was 80:10:5:5. The mass loading of all active material was ~3 mg cm^{-2}. The charge and discharge measurements were performed between the voltage range of 0.005-2.0 V vs. Li^+/Li.

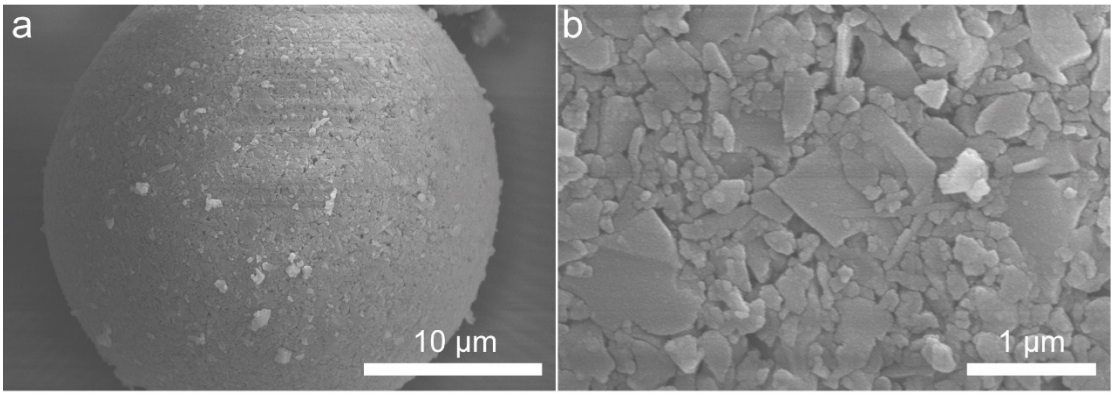

Fig. S1 The high magnification SEM images of porous SiO_x microspheres.

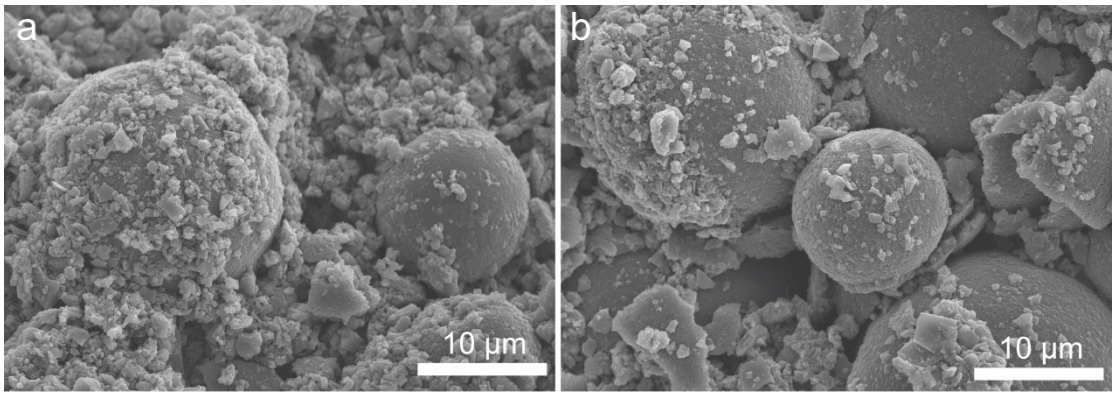

Fig. S2 The high magnification SEM images of (a) SiO_x/asphalt membrane and (b) binder-free SiO_x/C@Ni after high temperature (900 ^oC) pyrolysis.

Table S1. Summary of electrochemical performances of different SiO_x-based anodes.

Sample	Mass loading (mg cm^{-2})	Initial charge capacity (mA h g^{-1})	ICE	Reversible capacity (mA h g^{-1}) after (X) cycles	Capacity retention	Reference
SiO_x/CNTs	1	687		453 (100)	66%	Ref. S1
SGF	2.4	477	62%	448 (500)	94%	Ref. S2
SiO_x	0.8	1087	62%	760 (400)	70%	Ref. S3
SiO_x/C	-	647	44%	540 (200)	83%	Ref. S4
SiO_x/C/G	3	600	85%	541 (600)	90%	Our work

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