Monodisperse and homogeneous SiO$_x$/C microspheres: A promising high-capacity and durable anode material for lithium-ion batteries

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**ABSTRACT**

Monodisperse SiO$_x$/C microspheres with tunable size (300–1000 nm) and well-controlled carbon content (~20–60 wt%) have been fabricated through a facile sol-gel method. The judicious selection of silicon and carbon precursors (vinyltriethoxysilane and resorcinol/formaldehyde) enables the formation of an homogeneous SiO$_x$/C (x = 1.63) composite, in which the SiO$_2$ mainly exists as ultrafine nano-domains (<2 nm). Benefiting from the unique structural features, the resultant SiO$_x$/C microspheres demonstrate high capacity and outstanding cyclability. Specifically, a reversible capacity of 999 mA h g$^{-1}$ can be achieved at 100 mA g$^{-1}$, retaining 833 mA h g$^{-1}$ after 150 cycles. At 500 mA g$^{-1}$, the SiO$_x$/C delivers a specific capacity of 689 mA h g$^{-1}$ and maintains 91.0% of the capacity after 400 cycles. SiO$_x$/C/LiFePO$_4$ full cells are also assembled, leading to an energy density of ~372 W h kg$^{-1}$ based on the total mass of active materials. This work sheds light on the rational design of high-performance SiO$_2$ based anode materials for lithium-ion batteries.

1. Introduction

As a well-known energy storage device, lithium-ion batteries (LIBs) have been extensively applied in portable electronics because of their high energy density, long cycle life, and no memory effect [1–5]. However, the performance of current state-of-the-art LIBs cannot meet the increasing demand. Tremendous efforts have been dedicated to developing novel high-capacity electrode materials. As for the anode materials, silicon (Si) has attracted great interest as the prime alternative due to its high theoretical capacity (4200 mA h g$^{-1}$) and abundant reserves. However, two issues seriously hamper the practical application of Si: (1) remarkable volume change (>400%) during cycling, which leads to poor cycling performance; (2) low conductivity, which limits the rate capability [6–8]. A number of strategies, such as fabricating nanostructured Si and Si-based composite materials, have been developed to tackle these issues [9–12]. Despite the encouraging advances have been achieved, the high manufacturing cost of nanostructured Si still remains unaffordable for practical applications.

Silica, also known as silicon dioxide (SiO$_2$), is the major constituent of sand. Although bulk SiO$_2$ shows little activity towards lithium, nanostructured SiO$_2$ has been demonstrated to be electrochemically active. For example, Gao et al. found that commercial SiO$_2$ nanoparticles (7 nm) exhibited a reversible capacity of ~400 mA h g$^{-1}$ when reacting with lithium [13]. Yu et al. reported that carbon coated SiO$_2$ nanoparticles could deliver a discharge capacity of over 500 mA h g$^{-1}$ with good cyclability [14]. Hollow porous SiO$_2$ nanocubes prepared by Yan et al. exhibited a high reversible capacity of 920 mA h g$^{-1}$ [15]. Chang et al. reported that mechanically activated quartz (SiO$_2$) could deliver a reversible capacity of 800 mA h g$^{-1}$ [16]. Silicon sub-oxides (SiO$_x$) have also been studied as a promising substitute for Si. Compared to Si, the SiO$_x$ exhibits several advantages, such as lower cost, easier synthesis, and less volume expansion. In addition, the inert Li$_2$O and lithium silicates (Li$_4$SiO$_4$, Li$_2$Si$_2$O$_5$) generated in the initial lithiation process can buffer the volume variation and help to maintain the structural stability [17–19]. However, like elemental Si, the widespread application of SiO$_x$ is limited by its low conductivity and large volume change. A series of SiO$_x$/C composites, such as hierarchical SiO$_x$/C [20], SiO$_x$/C dual-phase glass [21], SiO$_x$@C microspheres [22], and SiO$_x$@C nanorods [23], have been designed to improve the electrochemical performance. Although significant improvements have been achieved, the facile synthesis of SiO$_x$/C composites with tunable structure and excellent lithium storage performance remains a great challenge.

In this work, we report the facile sol-gel synthesis of monodisperse SiO$_x$/C microspheres with tunable size and well-controlled carbon content. With the homogeneous dispersion of SiO$_x$, ultrafine nanodomains in an amorphous carbon matrix, the electronic conductivity of SiO$_2$ can be significantly enhanced and the large volume variation can...
be effectively buffered. The as-obtained SiO$_x$/C composite demonstrate high capacity and excellent cycling stability in lithium storage. A high reversible capacity of 999 mA h g$^{-1}$ can be achieved at 100 mA g$^{-1}$, maintaining 853 mA h g$^{-1}$ after 150 cycles. In addition, the SiO$_x$/C manifests a specific capacity of 689 mA h g$^{-1}$ at 500 mA g$^{-1}$ and a capacity retention of 91.0% after 400 cycles. The outstanding electrochemical performances enable robust full-cell operations in pairing with LiFePO$_4$, delivering an energy density of 372 W h kg$^{-1}$.

2. Experimental section

All the chemical reagents used were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

2.1. Sample preparation

In this work, SiO$_x$/C composite microspheres with carbon contents of ~ 20%, 40%, and 60% were designated as SiO$_x$/C-1, SiO$_x$/C-2, and SiO$_x$/C-3 respectively.

For the synthesis of SiO$_x$/C-1, 1 mL of aqueous ammonia (NH$_4$OH, 25 wt%) was dispersed in a solution containing 20 mL of deionized water and 10 mL of absolute ethanol. After stirring for 1 h, 1 mL of VTES (vinyltriethoxysilane) was added into the mixture. After a reaction time of 5 h at room temperature, the mixture was transferred into a 100 mL Teflon lined autoclave and hydrothermally treated at 100 °C for 12 h. The organosilica microspheres could be obtained after centrifugation and drying at 70 °C. After annealed at 800 °C (3 °C/min) for 3 h in Ar, the final product, SiO$_x$/C-1 was obtained.

For the synthesis of SiO$_x$/C-2, 0.3 mL of aqueous ammonia and 0.1 g of resorcinol was dissolved in a solution containing 20 mL of deionized water and 8 mL of absolute ethanol. After stirring for 1 h, 0.14 mL of formaldehyde was added into the mixture. Subsequently, 1 mL of VTES was added under stirring for 24 h at room temperature. The organosilica-polymer microspheres could be obtained after centrifugation and drying. After annealed at 800 °C (3 °C/min) for 3 h in Ar, the SiO$_x$/C-2 was obtained. The synthesis of SiO$_x$/C-3 is quite similar to that of SiO$_x$/C-2 except different amounts of ethanol (10 mL), resorcinol (0.3 g), ammonia (1 mL) and formaldehyde (0.42 mL) were used.

2.2. Materials characterization

X-ray diffraction (XRD) measurements were performed on a D8 Discover X-ray diffractometer with a non-monochromated Cu Kα X-ray source. Field emission scanning electron microscopy (FESEM) images were collected with a JEOL-7100F microscope at an acceleration voltage of 10 kV. Transmission electron microscopy (TEM) images were recorded by a JEM-2100F STEM/EDS microscope. Brunauer–Emmett–Teller (BET) surface areas were measured using a Tristar II 3020 instrument. Thermo gravimetric analysis (TGA) was conducted using a STA-449C thermogravimetric apparatus. X-ray photoelectron spectrum (XPS) measurement was performed using a VG Multi Lab 2000 instrument.

2.3. Measurements of electrochemical performances

The SiO$_x$/C composite (75 wt%), acetylene black (10 wt%), and sodium alginate (15 wt%) were dispersed in water to form a slurry. The slurry was spread onto a Cu foil by a doctor blade method, followed by drying in vacuum at 70 °C for 12 h. The mass loading of the active material was 1.5–2 mg cm$^{-2}$. 1.0 M lithium hexafluorophosphate (LiPF$_6$) in ethylene carbonate/dimethyl carbonate (50/50, v/v) was used as the electrolyte. The electrochemical performances were characterized by assembling CR2016 coin cells with lithium foil as the counter and reference electrode. Galvanostatic discharge/charge measurements were performed in a potential range of 0.01–3.0 V vs. Li+/Li using a multichannel battery testing system (LAND CT2001A). Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) measurements were performed with an electrochemical workstations (Autolab PGSTAT 302N and CHI660E).

SiO$_x$/C-2//LiFePO$_4$ full cells were also assembled. The weight ratio of cathode to anode was around 4:1, and the weight ratio of LiFePO$_4$/carbon black:PVDF was 80:10:10 in the cathode. The SiO$_x$/C-2 nanocomposite was firstly pre-lithiated in half cells and then taken out for full cell assembly. The SiO$_x$/C//LiFePO$_4$ full cells were charged/discharged galvanostatically at 0.2, 0.5, and 1.0 C (1 C = 170 mA h g$^{-1}$) in the electrochemical window of 2.2–3.75 V.

Fig. 1. Schematic illustration for the preparation of SiO$_x$/C microspheres. The synthesis relies on the simultaneous condensation of organosilica and RF.
3. Results and discussion

The synthesis of SiO\(_x\)/C microspheres includes two steps, i.e. the sol-gel synthesis of organosilica/polymer composite microspheres and the subsequent conversion into SiO\(_x\)/C microspheres (Fig. 1). The synthesis of the organosilica/polymer composite microspheres is carried out under Stöber reaction condition (water-ethanol-ammonia). Under such condition, vinyltriethoxysilane (VTES) is subjected to hydrolysis and condensation, yielding organosilicates; meanwhile, the resorcinol and formaldehyde polymerize, generating a resorcinol-formaldehyde (RF) polymer. Due to the matching reaction kinetics of the above two reactions as well as the strong hydrogen-bond interactions between the silanol (Si-OH) and phenol hydroxyl groups, the organosilica can co-condense with the RF polymer, forming homogeneous organosilica/polymer composite microspheres. During annealing, both the vinyl groups on organosilica and the RF polymer decompose into amorphous carbon, resulting in the formation of homogeneously distributed SiO\(_x\)-carbon composite (SiO\(_x\)/C) microspheres.

The three SiO\(_x\)/C samples show almost identical XRD patterns (Fig. S1). Only a number of very broad diffraction peaks can be observed, implying the amorphous feature of both SiO\(_x\) and carbon. According to literatures, the broad peak at ~ 23° can be ascribed to amorphous SiO\(_x\) [24], while the wide peak located at ~ 43° can be attributed to amorphous carbon [19,21].

Fig. 2a–f shows the morphology of the obtained SiO\(_x\)/C composites. All the samples exhibit regular spherical geometry with good monodispersity. The average diameters of SiO\(_x\)/C-1, SiO\(_x\)/C-2 and SiO\(_x\)/C-3 are determined to be ~ 1000, 400 and 300 nm, respectively. The elemental mapping results of a typical sample, SiO\(_x\)/C-2, are shown in Fig. 2g–j. It can be observed that the C, Si and O elements distribute homogeneously in the microspheres, resulting from the co-condensation of organosilica and RF frameworks. The key to the co-condensation is the matching polymerization rates of VTES and resorcinol/formaldehyde under Stöber reaction conditions. It should be pointed out that the co-condensation cannot be achieved if the VTES is substituted by tetraethyl orthosilicate (TEOS) or tetrapropyl orthosilicate (TPOS). According to the literatures, the polymerization of TEOS/TPOS always proceeds much faster than that of resorcinol/formaldehyde, leading to the formation of SiO\(_2@\)RF core-shell structures rather than homogeneous SiO\(_2@\)RF composites [25–28]. The elemental mapping images of SiO\(_x\)/C-1 (Fig. S2) and SiO\(_x\)/C-3 (Fig. S3) also show the uniform distribution of C, Si and O.

To further confirm the distribution of SiO\(_x\) and C in SiO\(_x\)/C-1, it was treated with HF (15 wt%) to remove the SiO\(_x\). Because no resorcinol/formaldehyde is introduced in the synthesis of SiO\(_x\)/C-1, the remaining carbon (Fig. S4) is derived from the vinyl group in VTES. The remaining carbon well inherits the micro-spherical morphology of SiO\(_x\)/C-1, suggesting that SiO\(_x\) species are uniformly dispersed in the carbon matrix. The SiO\(_x\)/C-2 and SiO\(_x\)/C-3 are obtained through the
co-condensation of VTES and resorcinol/formaldehyde, so the carbon is derived from both RF polymer and vinyl groups. Treating the organosilica/RF composite microspheres with HF can remove the organosilica while leaving the RF polymer. Annealing the remaining RF polymer in Ar yields carbon spheres (Figs. S5 and S6) with similar morphology and size to those of SiO$_x$/C-2 and SiO$_x$/C-3, demonstrating the even distribution of organosilica in RF framework (co-condensation). Etching the SiO$_x$ species from SiO$_x$/C-2 yields a microporous carbon (Fig. S7), suggesting the SiO$_x$ species mainly exists as ultrafine nano-domains (< 2 nm). Given the existence of Si–C bond in VTES, the homogeneous distribution of SiO$_x$ and carbon at nanoscale is not surprising.

The BET specific surface area and total pore volume of the SiO$_x$/C are investigated by nitrogen sorption (Fig. S8). All the SiO$_x$/C samples exhibit small surface areas (SiO$_x$/C-1: 2.8 m$^2$ g$^{-1}$, SiO$_x$/C-2: 15.4 m$^2$ g$^{-1}$, SiO$_x$/C-3: 37.3 m$^2$ g$^{-1}$) and low pore volumes (SiO$_x$/C-1: 0.005 cm$^3$ g$^{-1}$, SiO$_x$/C-2: 0.009 cm$^3$ g$^{-1}$, SiO$_x$/C-3: 0.023 cm$^3$ g$^{-1}$), indicating their nonporous feature. Such a dense microspherical structure is beneficial for achieving high tap density and thus high volumetric energy density in lithium storage.

Fig. 3a shows the TGA results of the three SiO$_x$/C composite samples. The carbon contents of SiO$_x$/C-1, SiO$_x$/C-2 and SiO$_x$/C-3 are calculated to be 21.06%, 39.75% and 59.38% respectively. Considering the SiO$_x$ species can be oxidized to SiO$_2$ during the combustion of carbon, the actual carbon contents of the samples should be slightly higher than the values determined from TGA. The XPS survey spectrum of the sample SiO$_x$/C-2 (Fig. 3b) confirms that the composite is composed of Si, C, and O. The C1s core level spectrum can be deconvoluted into three components: C in C–C bonds at 284.6 eV (Fig. S9), C in C=O bonds at 288.9 eV [29] and C in C–O bonds at 285.9 eV and C in C=O bonds at 286.8 eV [30]. The Si2p core level spectrum can be deconvoluted into three components, which are attributed to Si$^{4+}$, Si$^{3+}$, Si$^{2+}$ respectively [30]. From the high-resolution Si2p spectrum of SiO$_x$/C-2, the Si$^{4+}$: Si$^{3+}$: Si$^{2+}$ ratio is determined to be 35.05%:56.83%:8.12%, and the average valence state of Si is estimated to be ~ 3.27. This value is close to the theoretical valence state of Si (3.0) assuming most of the Si–C bonds in VTES are broken during carbonization [31]. As for SiO$_x$/C-1 and SiO$_x$/C-3, both samples show Si average valence states (3.15–3.16) and oxygen contents ($x = 1.58$) close to those of SiO$_x$/C-2 (Fig. S9, Table S1).

Fig. S10 displays the initial three cyclic voltammetry (CV) curves of the SiO$_x$/C-2 composite at a scanning rate of 0.1 mV s$^{-1}$. Several reactions would happen during the first cathodic process, such as the formation of SEI film, the conversion of SiO$_x$ to lithium silicates, Si, and Li$_2$O (SiO$_x$ + 2x Li$^+$ + 2x e$^-$ → x Li$_2$O + Si; SiO$_x$ + x Li$^+$ + x e$^-$ → 0.25x Li$_x$SiO$_{2x}$ + (1 – 0.25x) Si; SiO$_x$ + 0.4x Li$^+$ + 0.4x e$^-$ → 0.2x Li$_x$Si$_{1.5}$O$_3$ + (1 – 0.4x) Si), as well as the alloying of Si to Li$_x$Si (Si + x Li$^+$ + x e$^-$ → Li$_x$Si) [32,33]. The broad cathodic peak located at around 0.32 V in the second and third cycles is due to the formation of Li$_x$Si alloy, while the anodic peak at 0.42 V is corresponding to the transformation of Li$_x$Si alloy into Si. The second and third CV curves overlap well, demonstrating the reversible alloying/de-alloying reaction between the in-situ generated Si and Li$_x$Si (Si + x Li$^+$ + x e$^-$ → Li$_x$Si).

Fig. 4a shows the selected discharge-charge curves of SiO$_x$/C-2 over a potential window of 0.01–3 V at a current density of 100 mA g$^{-1}$. The initial discharge and charge capacities are 1460 and 965 mA h g$^{-1}$, respectively. The initial coulombic efficiency is 66.1%. The relatively large irreversible capacity (495 mA h g$^{-1}$) during the first cycle is due to the formation of SEI film, lithium silicates, and Li$_2$O [34-36]. From the 2nd to the 150th cycle, the discharge capacity decreases slightly from 999 to 854 mA h g$^{-1}$, corresponding to a capacity retention of 83.5%. Fig. 4b presents the cyclic performances of SiO$_x$/C microspheres at a current density of 100 mA g$^{-1}$. The initial discharge capacity of SiO$_x$/C-1 reaches 1500 mA h g$^{-1}$. However, the capacity fades continuously; a value of only 642 mA h g$^{-1}$ remains after 150 cycles, corresponding to a retention of 50.8% against the 2nd cycle. The SiO$_x$/C-3 delivers a stable discharge capacity of about 625 mA h g$^{-1}$ with negligible tendency of degradation. Several trends can be observed from Fig. 4c and Table 1: (1) the initial coulombic efficiency (CE$_{int}$) decreases with the increase of carbon content; the SiO$_x$/C-1 with a lowest carbon content of 21.06% has a highest CE$_{int}$ of 72.4%. This trend suggests the amorphous carbon matrix contributes an important share to the initial irreversible
capacity. Similar trend can be observed in the pomegranate-like Si/C nanocomposite and it is caused by the high density of Li trapping sites in amorphous carbon [37]. (2) The reversible capacity ($C_{rev}$) also decreases with increasing carbon content due to the high theoretical capacity of SiO$_x$. (3) The capacity retention increases significantly with the carbon content, which can be attributed to the volume change buffering ability of carbon. (4) The SiO$_x$/C-2 with a moderate carbon content of ~39.75% shows the best performance trade-off among CEint, $C_{rev}$ and capacity retention.

The rate performances of SiO$_x$/C are shown in Fig. 4c and d. With the gradual increase of current density from 100 to 600 mA g$^{-1}$, the SiO$_x$/C-2 exhibits a stable discharge capacity at each current density. A stable discharge capacity of ~620 mA h g$^{-1}$ can be achieved at 600 mA g$^{-1}$. When the current density is switched back to 100 mA g$^{-1}$, the capacity can be recovered to the original value. Due to the higher content of carbon and thus better electronic conductivity, the SiO$_x$/C-3 manifests much better rate performance than SiO$_x$/C-1, the SiO$_x$/C-2 demonstrates a specific capacity of 689 mA h g$^{-1}$ after 400 cycles, corresponding to 91.0% of the 2nd capacity (Fig. 4e). EIS measurements were performed in a frequency range of 100 to 0.01 Hz to explore the interfacial impedance of SiO$_x$/C with different carbon content (Fig. S11). The sample with higher carbon content shows lower charge-transfer resistance, demonstrating the role of carbon in improving the electrical conductivity.

To evaluate the possibility for practical applications in LIBs, the SiO$_x$/C-2 was assembled into full cells by pairing with a commercial cathode material, LiFePO$_4$. For anode materials with a low reversible capacity, an excess amount of cathode materials are required to compensate the irreversible capacity loss of the first cycle. To make full use of the LiFePO$_4$ cathode material in the full cells, the SiO$_x$/C-2 was pre-lithiated in a half cell for half a cycle and then took out for full cell assembly. Based on the mass of LiFePO$_4$, the SiO$_x$/C-2//LiFePO$_4$ full cell delivers specific capacities of 145 and 130 mA h g$^{-1}$ at 0.2 and 1.0 C (Fig. 5a), which are quite close to the capacities of LiFePO$_4$ in half cells (Fig. S12). The corresponding charge/discharge profiles (Fig. 5a inset) exhibit a typical discharge plateau between 2.8–3.3 V. The cycling performance of the SiO$_x$/C-2//LiFePO$_4$ full cell is shown in Fig. 5b. The full cell exhibits a reversible capacity of 147 mA h g$^{-1}$ at 0.2 C and maintained at 130 mA h g$^{-1}$ after 100 cycles with a capacity retention ratio of 88.4%. Taking an average voltage value of 3.1 V and an anode/cathode weight ratio of 4:1, the full cell is able to deliver an energy density of 372 W h kg$^{-1}$, and this value can be further enhanced by optimizing the cathode/anode weight ratio.

The structural integrity is an important parameter for anode materials, especially for high-capacity anode materials suffered from large volume variation. Without proper structural design, such materials would pulverize after repeated discharge-charge processes. For the SiO$_x$/C samples reported here, their micro-spherical morphology can be well preserved after 150 deep discharge-charge cycles at 100 mA g$^{-1}$ (Fig. 6), demonstrating their excellent structural stability. After 150 cycles, both SiO$_x$/C-2 and SiO$_x$/C-3 show a clear and smooth surface after cycling (Fig. 6c and d), suggesting the formation of uniform thin SEI layers on their surface. In contrast, the sample SiO$_x$/C-1 shows a vague and rough surface (Fig. 6a and b), indicating that the sample is buried under thicker SEI layers. This is reasonable because the SiO$_x$/C-1 with the lowest carbon content has the largest volume change; the repetitive volume expansion and contraction causes the repeated rupture and formation of SEI layers, resulting the build-up of SEI.

The high reversible capacity and excellent cyclability of SiO$_x$/C can be attributed to its unique structural features. In the constructed SiO$_x$/C

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Table 1

<table>
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<th>Sample</th>
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<th>1st coulombic efficiency</th>
<th>Reversible capacity</th>
<th>Capacity retention</th>
<th>Carbon content</th>
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<td>999</td>
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<td>SiO$_x$/C-3</td>
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<td>60.5%</td>
<td>713</td>
<td>88.1%</td>
<td>59.38%</td>
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</table>

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Fig. 4. Discharge-charge profiles of SiO$_x$/C-2 at a current density of 100 mA g$^{-1}$ (a); cycling performance and coulombic efficiency of SiO$_x$/C at 100 mA g$^{-1}$ (b); rate capability of SiO$_x$/C-2 (c); rate capability of SiO$_x$/C-1 and SiO$_x$/C-3 (d); long cycling stability and coulombic efficiency of the SiO$_x$/C-2 at 500 mA g$^{-1}$ for 400 cycles (e).
composites, the SiO<sub>x</sub> species mainly exist as isolated and ultrafine nano-domains (<2 nm), evenly distributed in an amorphous carbon matrix. The ultrafine SiO<sub>x</sub> is highly active towards lithium and acts as the primary active material, contributing a high capacity to the composites. Meanwhile, the amorphous carbon matrix serves multiple purposes: (I) The amorphous carbon functions as the secondary active component, contributing a certain capacity to the SiO<sub>x</sub>/C composite. (II) The amorphous carbon matrix acts as a continuous electron highway, wiring the whole SiO<sub>x</sub>/C composite. (III) The amorphous carbon matrix effectively buffers the large volume change, inhibits the aggregation, and prevents the pulverization of SiO<sub>x</sub>, resulting in excellent cycling stability. The rate performance of the SiO<sub>x</sub>/C composites is not comparable to state-of-the-art anode materials at present, which might be related to their non-porous characteristic. Further improvement of the rate capability is in progress by creating mesopores in the SiO<sub>x</sub>/C composites, aiming to boost the sluggish solid-state Li<sup>+</sup> diffusion. The initial Coulombic efficiency of SiO<sub>x</sub> based anode materials is generally low, and it might be further enhanced by decreasing O content of SiO<sub>x</sub>, improving the graphitization degree of carbon and employing pre-lithiation [38].

4. Conclusion

In summary, monodisperse SiO<sub>x</sub>/C composite microspheres with tunable size (300–1000 nm) and well-controlled carbon content (20–60 wt%) have been fabricated via a facile sol-gel co-condensation process using VTES and resorcinol/formaldehyde as precursors. With the homogeneous dispersion of SiO<sub>x</sub> ultrafine nano-domains in amorphous carbon matrix, the obtained SiO<sub>x</sub>/C composite microspheres manifest high specific capacity (999 mA h g<sup>−1</sup>), excellent cycling stability (85.5% capacity retention after 150 cycles), and outstanding structural integrity. The SiO<sub>x</sub>/C-2//LiFePO<sub>4</sub> full cell demonstrated an energy density of 372 Wh kg<sup>−1</sup> with good cycling performance, demonstrating the great potential of SiO<sub>x</sub>/C-2 in next-generation LIBs. This work sheds light on the rational design of cost-effective and high-performance SiO<sub>x</sub> based anode materials for LIBs.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ensm.2018.01.004.

References