Silicon monoxide (SiO) has been investigated as the next-generation anode material for lithium-ion batteries due to the high energy density it can offer. Compared to commercially used graphite, SiO has a theoretical gravimetric capacity of 1710 mAh g\textsuperscript{−1} and volumetric capacity of 1547 Ah L\textsuperscript{−1} and can offer an 18% increase in volumetric energy density on the cell stack level,\textsuperscript{1} making it a promising anode material for the next generation high energy density lithium-ion batteries.

SiO is composed of Si nanodomains in a SiO\textsubscript{2} matrix.\textsuperscript{2} An interphase region of transitional stoichiometry (SiO\textsubscript{x}, 0 < x < 2) is also present between Si and SiO\textsubscript{2} and takes up 20–25 at.% of the entire composition.\textsuperscript{2,3} During the lithiation and delithiation, the Si nanodomains react reversibly with lithium, similar to amorphous silicon, to give the reversible capacity. The SiO\textsubscript{2} matrix reacts irreversibly with lithium to form lithium silicates and lithium oxide.\textsuperscript{1} This microstructure shows many advantages. Firstly, small silicon nanodomains can better mitigate the stress during the lithiation and delithiation process. When silicon nanoparticles are smaller than 150 nm, Both pulverization and cracks are minimized during cycling.\textsuperscript{4} Secondly, compared to nano-sized Si, having Si nanoparticles embedded in a matrix may also decrease the contact of the electrolyte with the active material, resulting in less solid-electrolyte interphase (SEI) formation. Furthermore, SiO\textsubscript{2} matrix can act as a structural buffer, leading to lower overall volume change during cycling and better cycling stability.\textsuperscript{5,6}

Despite the advantages offered by its microstructure and the improved cyclability enabled by further modification and treatment,\textsuperscript{7–10} the cycle life of SiO is still far from being good enough for real-world applications. The primary focus of this work is to understand the major causes of capacity fade in silicon monoxide. To achieve this, we studied the evolution of the differential capacity of the SiO anode during cycling, as well as how the electrode composition and SiO annealing affect its electrochemical performance.

**Experimental**

**Materials and process.**—SiO from commercial vendor was used as received for studying the effect of electrode composition on cell performance. The C45 conductive additive was TIMCAL SUPER C45 conductive carbon black. Polyacrylic acid (PAA, M\textsubscript{w} = 450,000) was purchased from Sigma Aldrich. The PAA was then neutralized with LiOH to obtain a 9.8 wt% LiPAA aqueous solution with a pH of 6.8. To test the effect of annealing on cell performance, the SiO particles were placed in an alumina crucible and heated in a muffle oven at 600 °C for 18 hours in a glove box filled with argon. The sample was then allowed to cool down to room temperature before being transferred from the glove box for characterization and electrochemical testing.

**Instrumentation.—** Transmission electron microscopy (TEM) measurements were taken using a JEM-2100F field emission TEM (JEOL, Peabody, Mass.). Scanning electron microscopy (SEM) measurements were taken using an S-4700 SEM (Hitachi High Technologies, Tokyo). The X-ray diffraction was performed at Sector 11B of the Advanced Photon Source at Argonne National Laboratory. The wavelength of the X-ray was 0.1173 Å. The wavelengths were then converted to Cu Ka wavelength of 1.5406 Å for data presentation.

**Laminate fabrication and cell assembly.**—SiO, C45, and lithium polyacrylate (LiPAA) in weight ratios of 8:1:1, 7:1:2, and 7:2:1 were mixed using a planetary centrifugal mixer (Thinky Mixer, Thinky Corporation, Tokyo) at 2000 rpm until the slurry was uniformly mixed without apparent aggregates. The slurries were then coated onto a 10 μm thick copper foil. The laminates were dried in a 75 °C oven for 4 hours and then transferred to a 75 °C vacuum oven for overnight drying.

For 2032 coin type coin cell assembly, 15-mm-diameter electrodes were punched from the SiO laminates first. The electrodes were individually calendared to a porosity of approximately 45% and were dried overnight in a vacuum oven at 150–160 °C inside the glove box. The electrolyte used was Gen II (EC: EMC = 3.7, 1.2 M LiPF\textsubscript{6}) with 10 wt% fluoroethylene carbonate (FEC). For half-cell testing, lithium metal foil was used as the counter electrode.

**Electrochemical testing protocol.**—Electrochemical testing was performed using a high-precision automated battery test system (Series 4000, Maccor Inc., Tulsa) in a temperature-controlled chamber at 30 °C. Half cells were first tested between 10 mV and 1.5 V for three formation cycles at a lithiation rate of C/10. After the formation cycles, the cells were tested for rate performance. The lithiation rate was set at C/5, while the delithiation rate was changed from C/5, C/3, C/2, 1C to 2C. Three cycles were performed for each delithiation rate. The cells then underwent cycle test at C/3, with every 10th cycle tested at C/10. For all cell testing, the active loading of the SiO electrode gave an initial reversible capacity of 1.3–1.6 mAh cm\textsuperscript{−2}.

In this study, we first investigate the major causes of capacity fading in silicon monoxide (SiO) anodes. Major capacity loss occurs in the early cycles followed by less capacity fading during the following cycles. Close examination of the electrochemical test results shows that similar to silicon, the major capacity loss of the SiO anode at the early cycles occurs at the earlier stages of lithiation (≥ 0.27 V vs Li/Li\textsuperscript{+}), suggesting the phenomenon of incomplete delithiation due to electrode volume expansion/contraction. Unlike silicon which shows continuous capacity loss toward lower voltages, the capacity retention of SiO stabilizes after the initial fast capacity loss, demonstrating the better cycling stability of SiO over Si. The capacity loss of SiO during long-term cycling has contribution from the whole voltage range, indicating loss of active material due to extensive cycling. The capacity loss can be mitigated with an optimized electrode composition. The long-term capacity retention of SiO is also found to be affected by the annealing treatment. The removal of the defects in the interphase region within the particle and the removal of the unfavorable surface groups from the surface are suggested as the possible reasons for the improved performance for annealed SiO.

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The AC impedance measurements were measured after the electrochemical testing of the half cells. The cells were charged to 50% state of charge and then rested for 4 hours. The AC impedance was recorded at open circuit potential at 50% SOC for frequency range of 50k Hz to 0.05 Hz with an amplitude of 5 mV.

Results and Discussion

Figures 1a and 1b show the SEM and high-resolution TEM images of the SiO particles. The SEM image shows that the majority of the particles are micron-sized, with some smaller particles of several hundred nanometers. The high-resolution TEM image shows crystalline domains in an amorphous matrix, as circled in Figure 1b. The lattice constant calculated from the high-resolution TEM image is 0.31 nm, corresponding to Si(111), confirming that the crystalline nanodomains are Si nanodomains.

In order to understand the major causes of capacity fading in the SiO anode, SiO anodes with 20 wt% LiPAA and 10 wt% carbon black were fabricated and cycled in the half-cell configuration. Figure 2a presents the typical cycling behavior of the SiO anodes. The cells were first cycled at C/10 rate between 1.5 V and 10 mV for 3 formation cycles, which were then followed by 15 cycles of rate test and 50 cycles of cycle test, as detailed in the Experimental section. The SiO anode starts with a reversible capacity of over 1500 mAh g\(^{-1}\), but fades rapidly to 2365 mAh g\(^{-1}\) and a delithiation capacity of 1578 mAh g\(^{-1}\), resulting in a 1st cycle coulombic efficiency of 66.7%. The low 1st cycle coulombic efficiency is typical for SiO due to the irreversible reactions between lithium and the SiO\(_2\) matrix as well as solid electrolyte interphase (SEI) buildup. As the cell cycles, the lithiation capacity above 0.27 V vs Li/Li\(^+\) gradually loses. Almost no capacity was obtained above 0.27 V at the start of the cycle test. Similarly during delithiation, while the voltage profile below 0.5 V remain almost unchanged during cycling, the capacity loss above 0.5 V was rapid in the initial cycles of formation and rate test. The rapid loss of capacity at higher voltages is also well-represented by the evolution of differential capacity plot in Figure 2c. The dQ/dV plot of SiO shows typical dQ/dV features of amorphous Si during cycling, showing two lithiation peaks at 0.23 V and 0.08 V and two delithiation peaks at 0.28 V and 0.48 V. No peaks corresponding to Li\(_{1-x}\)Si\(_x\) crystalline phase was observed. A major capacity loss at relatively high voltages (shaded area) occurred between the 3rd formation cycles and the 1st C/10 cycle in the cycle test. The change of the dQ/dV plot during the cycle test appears to be much less, with a gradual decrease of peak intensities observed from all redox peaks.

To further compare the capacity loss contributions from different voltage ranges, we divide the lithiation into three stages: 0.8–0.27 V(L1), 0.27–0.16 V(L2) and 0.16V–0.01V(L3), corresponding to the early stage of lithiation (Li\(_{0.99}\)Si),\(^{11}\) 1st redox peak (Li\(_{2.2}\)Si)\(^{11}\) and 2nd redox peak (Li\(_{1.3}\)Si). We then integrated the dQ/dV plot under the three lithiation voltage ranges. Figure 2d shows the capacity contribution from the three lithiation stages to the full lithiation capacity. It clearly shows that the rapid loss in total capacity in the early cycles is mainly from the early stage of lithiation (L1). The capacity from 0.8–0.27 V quickly dropped to close to zero after the formation and rate test. For the later cycles, capacity loss from both L2 and L3 contributed to the total capacity loss, indicating loss of active material from extensive cycling, which will be discussed in details later.

The similar rapid capacity loss in the early stage was also observed by Yoon and coworkers in their investigation of capacity fading mechanisms for the Si electrodes. They observed loss of capacity at high potentials (0.27–0.8 V vs. Li/Li\(^+\)), lithiation voltage) over cycles,\(^{11}\) which is attributed to the incomplete delithiation of Si caused by the substantial volume change during cycling as well as SEI resistance buildup. As the Si particles contract during delithiation, the electric contact between the particles and the binder/carbon black worsens, resulting in increase of contact resistance, which leads to incomplete delithiation. Since SEI is electronically resistive, continuous SEI buildup can also further increase the cell impedance, contributing to the complete delithiation. Since the active component of SiO is the Si nanodomains, SiO also experiences 117% reversible volume change during cycling. The volume change also leads to similar fading mechanisms in SiO. For the loss of SiO capacity at high potentials (> 0.27 V vs. Li/Li\(^+\)), it could also result from incomplete delithiation. As the lithiated SiO particles shrink during delithiation, contact resistance increases. The SEI layer outside particles may also peel off as the particles shrink, leading to new SEI formation on the newly exposed SiO surfaces. The SEI buildup also leads to impedance buildup. The impedance buildup due to contact resistance and SEI buildup in the electrode can lead to the observed incomplete delithiation. Figure S2 shows a schematic which helps to visualize the schematic. Unlike Si, where the capacity loss from L2 becomes significant when L1 loses all the capacity, the capacity loss of SiO slows down once the capacity from L1 is lost. This more stable cycling could be due to the less volume change of the SiO compared to Si. Despite the improvement in capacity retention, the slow capacity fading during long-term cycling is still observed and is found to be caused by loss of active material. The effect of particle annealing on the improvement of long-term cycling stability and its implication of the impact of the intrinsic chemical/mechanical stability of SiO on cycling stability is discussed in the annealing section.

The capacity loss in longer-term cycling shows contribution from all lithiation stages, suggesting loss of active materials due to extensive

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**Figure 1.** SEM and TEM images of pristine silicon monoxide.
As the SiO electrode is being cycled, the binder/conductive network experiences the repeating stress from the volume change of the active SiO particles. This may eventually lead to inability of the network to endure the large volume change. The continuous SEI buildup may also lead to degradation of the mechanical integrity of the electrode upon cycling. Furthermore, diffusion controlled lithium trapping may also take place during cycling, leading to capacity loss caused by the loss of active material.

The above findings suggest that good electric contact between the active material (SiO) and the binder (LiPAA)/conductive additive (C45) is essential. A good electrode composition with a proper binder and conductive additive content can lead to a better interaction between the active material and the conductive additive, which further results in better electric contact as the particles expand and contract during lithiation and delithiation, and may lead to less loss from both the incomplete delithiation and loss of active material from extensive cycling. To confirm the importance of proper electrode composition, we prepared SiO electrodes of different electrode compositions and tested the electrochemical performance.

The electrode composition for SiO testing reported in the literature usually consists of 10–20 wt% binder and 10–20 wt% conductive carbon additives. Accordingly, our SiO sample was made into electrodes with three different compositions to test for electrochemical performance:

- 10 wt% LiPAA and 10 wt% C45 (denoted as 10B–10C in the discussion below)
- 10 wt% LiPAA and 20 wt% C45 (10B–20C)
- 20 wt% LiPAA and 10 wt% C45 (20B–10C)

The electrochemical performance of SiO indeed appears to be strongly dependent on the electrode composition. Figure 3a shows the complete electrochemical performance of the SiO anode with the three different electrode compositions. For all electrode compositions, rapid loss of capacity was observed in the early cycles, with the most capacity loss observed for 10B–10C electrodes. Regarding long-term cycling performance, 10B–10C SiO electrode also shows the worst performance of the three, with capacity retention of approximately 64% obtained in the cycle test (capacity retention of the 50th cycle to the 3rd cycle in the cycle test). Adding 10 wt% more carbon black (10B–20C) improved the specific capacity at the start of the cycle test by 200 mAh g$^{-1}$ and the capacity retention from 64% to 75%, indicating adding more C45 improves the interaction between the SiO active material and the conductive additive/binder network. Keeping C45 content unchanged and adding 10 wt% more binder (20B–10C) leads to significant improvement of capacity retention to 90%, as well as an electrode specific capacity of 1300 mAh g$^{-1}$ at the start of the cycle test. While adding more C45 improves the electronic network and therefore improves the electrochemical performance, the electrochemical performance comparison shows that the binder plays a more important role since larger improvement was achieved when more binder is added. Figure S2 shows the capacity contribution from different lithiation stages for the SiO electrodes with 10B–20C and 10B–10C compositions. Comparing also with Figure 2d which presents the capacity contribution for pristine SiO electrode with 20B–10C electrode, it can be concluded that while an optimized electrode composition improved the cycling performance in the later stages, we did not observe an improvement of the overall capacity loss from L1 due to incomplete delithiation. 10B–20C sample showed gradual

Figure 2. a) electrochemical performance of SiO anode. b) voltage profile change of the SiO anode, c) dQ/dV change of the SiO anode over cycles, and d) contribution from different lithiation voltage ranges to the total capacity.
the SEM and TEM images of the particles after the 600 °C treatment. The SEM image shows no noticeable change in particle size, suggesting that there’s no significant sintering at this temperature. The high-resolution TEM image shows that the size of the Si nanodomains remains mostly unchanged. Figure 4c shows the X-ray diffraction patterns of the pristine and 600 °C-treated SiO samples. Calculated from Scherrer Equation, the average Si domain size increased slightly from 3.36 nm to 3.54 nm. Annealing at high temperatures can lead to growth of Si domains in SiO, which can also affect the cycling performance of SiO. Park and coworkers \(^{10}\) reported that by annealing SiO at 1000 °C, the initial cycling performance was improved. However when annealed at 1200 °C, the electrochemical performance was significantly worsened. The poor electrochemical performance of SiO after annealing at 1200 °C was attributed to the change of the SiO structure to spawn-like structure with well-developed Si nanodomains, which are much bigger than the Si nanodomains after annealing at 1000 °C. The lithiation of big Si nanodomains may lead to increased stress inside the SiO particles during cycling. In this work, we tried to avoid the crystallinity effect on the electrochemical performance. At an annealing temperature of 600 °C, the change of Si domain size is very minimal, therefore we do not expect the change of Si nanodomains to significantly affect the electrochemical performance of the SiO.

Figure 4d compares the electrochemical performance of the 600 °C-treated SiO sample with the untreated pristine SiO. The electrochemical performance of the two electrode compositions (20B–10C and 10B–10C) for 600 °C-treated SiO are shown to compare with the optimal electrochemical performance of pristine SiO electrode achieved with 20B–10C electrode composition. The rapid loss at the initial cycles was still observed for the 600 °C-treated SiO sample. However, unlike pristine SiO where continuous fast capacity fade was observed in the rate test, the 600 °C-treated SiO only showed very rapid capacity loss in the formation cycles, with no obvious capacity fade observed in the rate test. This different fading behavior is not clear at this moment. We believe that the initial fast capacity loss during formation cycles are still caused by the volume expansion. This difference in capacity stabilization may be a result of annealing and further study is needed to understand this phenomenon. The capacity retention during the cycle test was significantly improved after the annealing treatment. For both electrode compositions of 20B–10C and 10B–10C, close to 100% capacity retention was observed considering cycle test alone. The less dependency of the electrochemical performance of the 600 °C-treated SiO on electrode composition suggests the improvement of the intrinsic stability of the particles from the annealing treatment. Since SEM/TEM/XRD suggests minimum change in SiO particle size as well as Si nanodomain size, it is more likely that the improvement of the stability results from the improvement of the interphase region between the Si domains and the SiO\(_2\) matrix, or the surface of the SiO. Annealing may remove defects in the interphase region or unfavorable surface groups from the surface, and therefore result in improved mechanical property of the particles. Further study is currently undergoing to fully understand the effect of annealing on the electrochemical performance of SiO.

It can also be found from Figure 4d that 600 °C-treated SiO electrode with 20B–10C composition showed less capacity loss from the initial cycles compared to 600 °C-treated SiO electrode with 10B–10C composition. Figure S4 further compares the capacity contribution from different lithiation stages for 600 °C-treated SiO electrodes at the 3\(^{rd}\) formation cycle where the cycling has become stable. The comparison between 20B–10C and 10B–10C electrodes shows that the capacity contribution from L2 and L3 are comparable for both electrode compositions. The contribution from L1, however, is almost completely absent for 10B–10C electrodes. The lack of contribution from L1 for 10B–10C composition further supports the important role of the binder at minimizing the incomplete delithiation. The less dependency of the 600 °C-treated SiO on electrode composition suggests the improvement of the intrinsic stability of the particles from the annealing treatment. Since the improvement

Figure 3. a) The electrochemical performance of SiO anodes with different electrode compositions, and b) Nyquist plots for cycled SiO electrodes with different compositions.

loss of capacity from L1 over the electrochemical test and 10B–10C sample showed complete loss of capacity from L1 at the 3rd formation cycle. While the rate of capacity loss from L1 (0.27–0.8 V vs. Li/Li\(^+\)) is different for the different electrode compositions, capacity from L1 is eventually all lost.

Figure 3b shows the Nyquist plots for the three electrodes after the cycle test. While 20B–10C cells showed the smallest semicircles, 10B–10C cells showed the largest semicircles, suggesting significant impedance rise from cycling due to the inability of the electrode composition to accommodate the volume change of the active material. SEM further shows that the electrode composition affects the mechanical property of the electrodes. Figure S3 shows the SEM images of the cycled electrodes of different compositions. While the microstructure of 20B–10C electrode and 10B–20C electrode showed few or no obvious cracks after cycling, the 10B–10C electrode showed many cracks, indicating the poor mechanical property of the 10B–10C electrode.

Despite the improvement in capacity retention with optimized electrode composition, the slow capacity fading during long-term cycling is still observed and is found to be caused by loss of active material. The fast capacity loss in the initial cycles is also still observed for the different electrode compositions. While adding more binder and carbon black may further improve the cycling stability, the energy density of the lithium-ion batteries will be adversely affected when too much inactive content is present. The loss of active material from extensive cycling can often be mitigated by improving the stability of the active materials. With the intention to minimize defects in the particles and improve structural stability, we annealed the pristine SiO particles under inert argon atmosphere for 18 hours at 600 °C. The weight change of the sample after the annealing treatment was –0.74%, indicating a minimum change in Si/O ratio in the sample. Figures 4a and 4b show the
in initial-cycle capacity retention was not observed for the pristine SiO with different electrode compositions, we suggest that to mitigate the capacity loss in the initial cycles, both improvement of particle intrinsic stability and optimization of the electrode composition are needed.

**Conclusions**

We investigated the major causes of capacity fade in SiO anode. The SiO anodes show a rapid capacity loss in the initial cycles followed by much more stable cycling. A careful examination of the differential capacity plots showed that the initial capacity loss is mainly from early stages of lithiation (>0.27 V vs Li/Li⁺), suggesting incomplete delithiation. Dramatic electrochemical performance observed with pristine SiO anodes with different electrode compositions suggest the important role of binder/conductive additive network to minimize capacity loss. The low capacity fading during cycle test can be further improved by annealing the SiO particles in inert atmosphere. We suggest that the annealing process may modify the interphase and the surface of SiO and improve the intrinsic stability of SiO particles for better electrochemical performance.

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Figure 4. a) SEM image, b) TEM image and c) XRD characterization of 600°C-treated SiO material. d) Electrochemical performance of 600°C-treated SiO electrode with electrode composition of 20B–10C (20% LiPAA, 10% C45) and 10B–10C (10% LiPAA, 10% C45), in comparison with the pristine SiO anode with optimal 20B–10C electrode composition.
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