Enhancing the Electrochemical Performance of Aqueous Processed Li-Ion Cathodes with Silicon Oxide Coatings

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Abstract: Lithium-ion battery cathode materials suffer from bulk and interfacial degradation issues, which negatively affect their electrochemical performance. Oxide coatings can mitigate some of these problems and improve electrochemical performance. However, current coating strategies have low throughput, are expensive, and have limited applicability. In this article, we describe a low-cost and scalable strategy for applying oxide coatings on cathode materials. We report synergistic effects of these oxide coatings on the performance of aqueously processed cathodes in cells. The SiO_2 coating strategy developed herein improved mechanical, chemical, and electrochemical performance of aqueously processed Ni-, Mn- and Co-based cathodes. This strategy can be used on a variety of cathodes to improve the performance of aqueously processed Li-ion cells.

Cathode materials such as $LiNi_xMn_yCo_zO_2$ (x + y + z = 1; NMC) are highly desirable for achieving high–energy density Li-ion batteries. However, most of these cathode materials are prone to structural, chemical, and electrochemical degradation under long-term cycling.[1,2] This degradation results in capacity fade and decreased cyclic stability of these materials, making them less attractive as high–energy density cathode materials.[1,2] Cathode reaction with a liquid electrolyte at higher voltages is a key contributor to the observed capacity fade. These parasitic reactions can be minimized using two strategies: modifying the electrolyte composition or applying a thin coating of materials such as SiO_2, TiO_2, Al_2O_3, ZrO_2, AlF_3, and LiF.[3,4] In particular, SiO_2 coating on cathode materials is highly desirable because of its low cost and high electrochemical stability.[4b] Several methods of applying SiO_2 coating have been explored, including ball milling of SiO_2 particles with cathode materials and sol-gel deposition of SiO_2 using protic solvents (e.g., EtOH).[5] However, these methods have their own limitations. For example, ball milling applies incomplete coating, and the high mechanical forces used in this process can result in the breaking of cathode secondary particles.[6] Although the sol-gel process can apply quite uniform and scalable coatings, the use of protic solvents such as EtOH and NH_3 results in the leaching of Li-ions out of the cathode (e.g., NMC) particles. This leaching lowers the capacity of the coated active material. Additionally, the sol-gel process requires removal of unused coating precursors and solvents and thus results in large quantities of waste.[7] Unused coating solution is removed mainly by filtration and less commonly by drying the unused solvents. However, the latter method leaves the solvents and unused coating precursors inside the reaction mixture. These dried reaction precursors can further reduce the overall capacity of the coated material and can lead to additional reactions with the electrolyte. Therefore, even though the sol-gel method can provide quite uniform coatings, it is not suitable because of its high cost, long process times, and the possible loss of capacity during coating. Similarly, oxide coatings can be applied using chemical vapor deposition, physical vapor deposition, or atomic layer deposition. However, these vapor-phase techniques require sophisticated instrumentation, high temperatures, and low pressure.[8,9,10] Additionally, they apply coatings only on the exposed surface and have very low throughput.

Therefore, a high-throughput, low-cost, environmentally benign strategy is needed that can apply a uniform coating without causing Li-ion leaching. This need is even more urgent when the cathode electrodes are prepared by aqueous processing, in which water is used to make the cathode slurries instead of organic solvents such as N-Methyl-2-pyrrolidone.[11] Aqueous processing has several advantages; for example, it uses a less costly and more environmentally friendly solvent (water) and eliminates solvent recovery in drying.[11] However, water interacts with cathode materials in aqueous processing, resulting in leaching of Li-ions, forming LiOH, and increasing the slurry pH. The resulting basic medium corrodes the Al current collector. The leaching of Li-ions results in initial loss of capacity.[11] Additionally, aqueously processed cathodes have large cracks due to lack of fully optimized binders. All these issues make aqueous processing disadvantageous compared with the nonaqueous processing of cathodes.[11f]

Most of the work on SiO_2 coatings has addressed nonaqueous processing of cathodes, whereas little work has been done investigating how SiO_2 coating affects the performance of aqueously processed cathodes in cells. In this study, we examined a low-cost, vapor-phase SiO_2 coating method for aqueous processing of cathodes. This method avoids the use of organic solvent and sophisticated instrumentation and can be easily scaled up. Because no solvent is used, the method also mitigates Li-ion leaching from the cathode materials during coating and thus helps retain the initial capacity of cathodes. Additionally, we tested these coated cathode materials by fabricating cathodes using aqueous processing and compared their electrochemical performance to that of uncoated pristine cathodes. The results indicate the developed SiO_2 coating improves the electrochemical performance by synergistically combining several individual effects.

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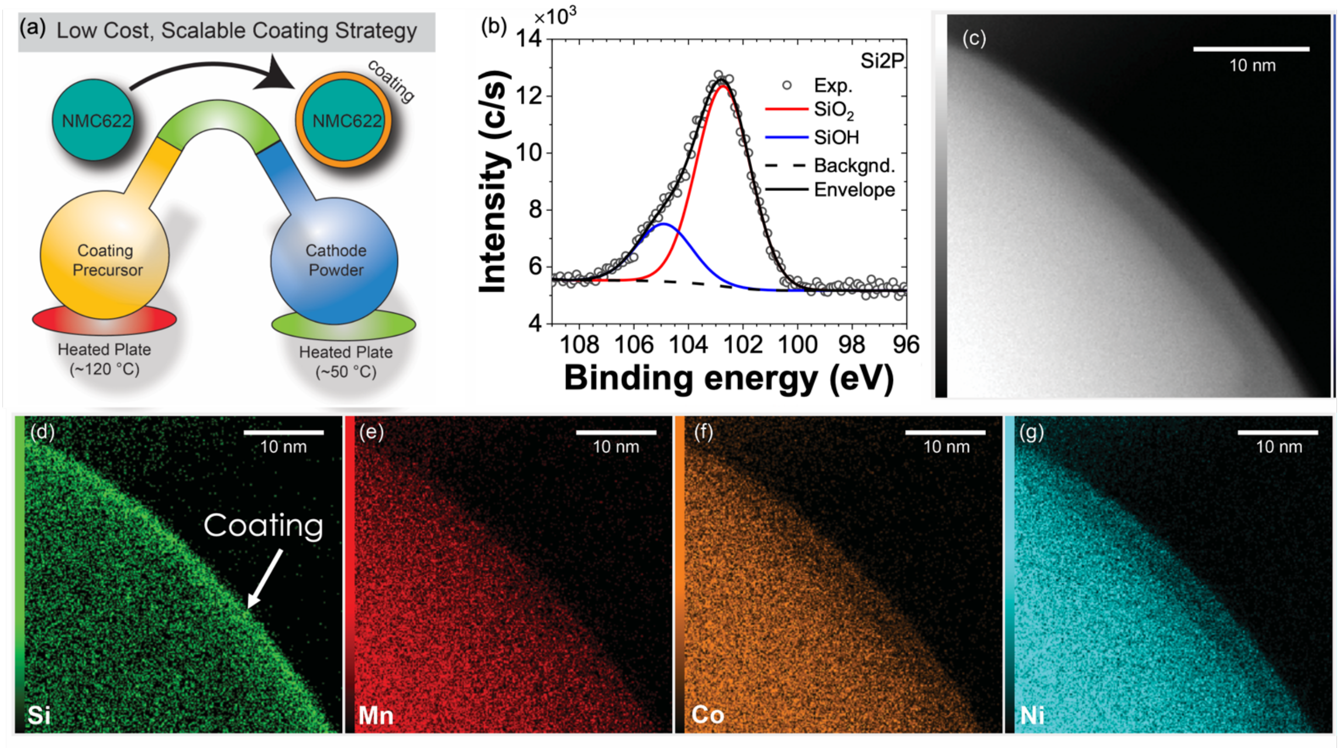

Figure 1. (a) Schematic diagram of the coating process, (b) plot of x-ray photoelectron spectroscopy analysis of the SiO_2-coated NMC622 particles, (c) TEM image of the SiO_2-coated NMC622 particle, and (d)-(g) EDX maps of the NMC622 particle showing presence of (d) Si, (e) Mn, (f) Co, and (g) Ni. Note. More TEM and EDX images are provided in supporting information.

In a typical experiment, we took 1 mL of the SiO_2 precursor tetramethyl orthosilicate (TMOS) in one flask and 5 g of $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ cathode (NMC622) in another flask. We connected the flasks using a glass connector and heated the TMOS at ~120°C to generate the silane vapors. Figure 1a shows the schematic of the experimental setup (More experimental details are available in supporting information). We performed x-ray photoelectron spectroscopy (XPS) analysis to test for the presence of SiO_2 coating on the NMC622 particle surface and found different molecular species, with a Si2p spectrum indicating two peaks at binding energies 102.7 eV and 104.9 eV, indicating SiO_2 and -SiOH, respectively (see Figure 1b).[12] The presence of the smaller -SiOH peak indicates the surface may still have retained some incompletely reacted silane groups or that the coating may have reacted with H_2O vapors resulting in SiOH groups. The presence of incompletely reacted silane groups, is expected because the coated NMC622 particles were not sintered at high temperature, but only dried at 100°C. Additionally, Si is clearly present in the transmission electron microscopy energy-dispersive x-ray (TEM-EDX) images of the SiO_2-coated NMC622 powder shown in Figure 1c–g.

For TMOS, a minimum temperature of 120°C is required to boil the silane solution and generate large amount of silane vapors. We used TMOS because of its high vapor pressure and low boiling point. Additionally, TMOS reacts quickly with the exposed −OH groups and thus easily creates the coating on cathode particles. TEM-EDX maps show the coating is approximately 2–3 nm thick (see Figure 1d and the Figure S2 in the supporting information). Coating thickness can be controlled by manipulating the exposure time of NMC622 powder to the silane vapors. Generally, exposure of 5 g of NMC622 powder to vapors generated from 1 mL of TMOS for 4–5 h resulted in a coating approximately 2 nm thick. We observed that coatings were formed on easily accessible areas on the particle surface, but joints between particle aggregates may not be sufficiently coated. Therefore, the particles should be fully dispersed to achieve highly homogeneous coating.

To test if SiO_2 coating alters the lattice structure of NMC622 powders, X-Ray diffraction (XRD) analysis was performed. XRD analysis showed that the both pristine and SiO_2 coated NMC622 powder show similar XRD patterns and no cation (Ni^{+2} and Li^{+}) mixing and destruction of layered structure occurred after SiO_2 coating (more details in supporting information).[13]

To confirm whether the SiO_2 coating reduces the leaching of Li-ions from NMC622 powder when the NMC622 powder is added to the water, the pH values of aqueous suspensions of coated and uncoated NMC622 were measured after 10 min of suspension preparation. 10 min time was selected to allow sufficient leaching of Li-ions out of the NMC622. The suspension of coated NMC622 (1 g NMC622 in 10 mL H_2O) has a pH of 9.45, whereas the suspension of uncoated NMC622 (1g NMC622 in 10 mL H_2O) has a pH of 9.95. The lower pH of the SiO_2-coated NMC622 suspension indicates that fewer Li-ions leached from the NMC622 particles (i.e., fewer Li-ions exchanged with the H-ions in the water). The reaction for pH increase can be written as [11a,14]

$$Li^{+} + H_2O \rightarrow LiOH + H^{+} \quad (1)$$

The Li-ion leaching was mitigated by the presence of SiO_2-coating, which reduces the interaction of water with the NMC particle surfaces. Additionally, the inductively coupled plasma-optical emission spectroscopy (ICP-OES) analysis revealed that approximately 10% fewer Li-ions leached from the aqueous suspension of coated NMC622 compared with the aqueous suspension of uncoated NMC622 (see details in supporting information). Table 1 lists the amount of Li metal detected by the ICP-OES in filtrates obtained from aqueous suspensions of coated and uncoated NMC622 particles.

Table 1. Amount of Li present in filtrates obtained from aqueous suspensions of coated and uncoated NMC622.

Suspension type	Amount of Li (ppm)
NMC622 (1 g in 10 mL H_2O)	117
SiO_2-NMC622 (1 g in 10 mL H_2O)	108
NMC622 (2 g in 10 mL H_2O)	217
SiO_2-NMC622 (2 g in 10 mL H_2O)	195

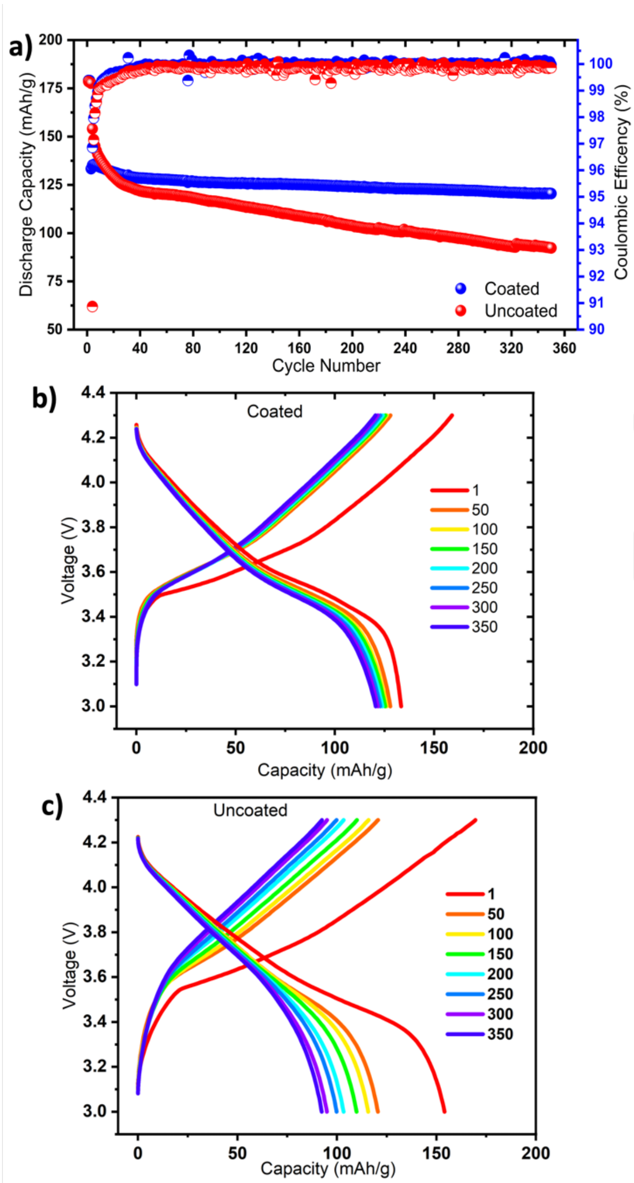

Figure 2. Electrochemical performance testing. a) Plot showing the cyclic stability (Right-Y axis) and coulombic efficiency (Left- Y axis). Charge and discharge profiles after different number of cycles at 1C/1C charge/discharge rates for full cells having SiO_2-coated (b) and uncoated NMC622 (c) cathodes.

To understand the effect of SiO_2 coating on the electrochemical performance of battery cells, cathodes of coated and uncoated NMC622 were fabricated using an aqueous processing technique (see details in supporting information). Full-coin cells were assembled using these cathodes and graphite anodes. The cells were cycled at 1C/1C charge/discharge rates for 350 cycles after 3 formation cycles at 0.05C/0.05C charge/discharge rates to test cyclic stability and coulombic efficiency (see details in supporting information). The cathodes had an aerial capacity of 2 mAh/cm^2, active material loading 11.12 mg/cm^2, thickness of 75 µm, and N/P ratio of 1.2. The cycling results shown in Figure 2 indicate that the cyclic stability of coin cells with SiO_2-coated NMC622 cathodes is significantly higher than those with uncoated NMC622 cathodes. The enhanced cyclic stability of SiO_2-coated NMC622 is expected because fewer Li-ions leach from these cathode materials during cathode fabrication, and the coated cathode particles react less with the electrolytes. Similarly, the cells with SiO_2-coated NMC622 cathodes showed slightly higher coulombic efficiency compared with the cells with uncoated NMC622 cathodes (see Figure 2a). It was observed that initial coulombic efficiency of both SiO_2-coated and uncoated NMC622 based full cells was lower and gradually increased with cycling, and reached maximum and stabilized after 15-20 cycles. The lower initial coulombic efficiency can be due to (1) some trapping of Li-ions in graphite anode and (2) in the beginning electrolyte may not have wetted the electrodes properly. Similar types of low initial coulombic efficiency have been observed in literature.[15] In case of SiO_2-coated NMC622 cathode based cells small amount of Li-ions can also get trapped in the SiO_2 coating during the first couple of cycles.[15]

The galvanostatic charge/discharge profiles for the same full cells at specific current of 1 C (180 mA/g) in voltage range of 3.0-4.3 V are presented in Figure 2b and 2c. There are no significant differences in the initial charge/discharge voltage profiles, indicating the coating of SiO_2 does not jeopardize the intrinsic electrochemical performance of the active cathode materials. Although the 1st discharge capacity of the cells having SiO_2- coated NMC622 is lower than that of the baseline one, SiO_2-coating greatly improves the cycling stability of NMC622. After 350 cycles at 1C/1C charge/discharge rates, the cells having SiO_2-coated cathodes showed a discharge capacity of 120 mAh/g, while the cells having uncoated cathodes exhibited a much lower capacity of 92 mAh/g. This could be attributed to the inert SiO_2 that protects the active cathode material from reacting with the electrolyte and reduces the interfacial resistance between the cathode and the electrolyte.[4i] Electrochemical impedance spectroscopy (EIS) measurements showed that the interfacial resistance (197.2 Ohm) was higher for cells containing SiO_2-coated NMC622 based cathodes in comparison to the resistance (166.5 Ohm) of cells containing pristine NMC622 based cathodes. The higher resistance for cells containing SiO_2-coated cathodes was expected as silica coating initially results in increased resistance to the Li-ion transfer at the cathode interface (more details in supporting information).[16]

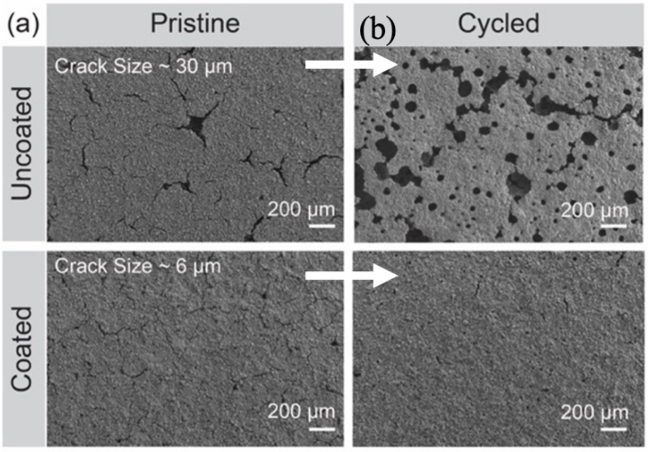

Figure 3. SEM images of uncoated and SiO_2-coated cathodes (a) before and (b) after cycling.

Aqueously processed, SiO_2-coated NMC622 cathodes have smaller cracks compared with the uncoated NMC622 cathodes, as shown in Figure 3a. Postmortem analysis of the cycled (350 cycles at 1C/1C rate) full cells revealed that the cathodes fabricated from SiO_2-coated NMC622 have smaller and fewer cracks than the cathodes fabricated from uncoated NMC622, as shown in Figure 3. Scanning electron microscopy (SEM) imaging shown in Figure 3a and b revealed that aqueously processed, SiO_2-coated NMC622 cathodes have better structural integrity than the uncoated NMC622 cathodes. The structural integrity is likely better (i.e., the cracks are small) because of the improved cohesion among the SiO_2-coated NMC622 particles and between the SiO_2-coated NMC622 and carboxymethyl cellulose (CMC) binder. Postmortem analysis of silica coated cathodes after cycling showed that in some cells, the cathodes have even less cracking than the cathodes before cycling (Figure 3). This phenomenon was not consistent in all cells. We assume that crack size may have decreased due to pressing during coin cell fabrication.

We also measured the adhesion strengths of the cathodes prepared from SiO_2-coated NMC622 and the cathodes prepared from uncoated NMC622 with the Al current collector (experimental details in supporting information). The adhesion strength of the coated cathodes (25 ± 3 N/m) was better than that of the uncoated cathodes (12 ± 2 N/m) (Figure 5 e), which indicates that SiO_2 coating also improves the adhesion of the cathode layer with the current collector.

The results of this work indicate that NMC 622 cathodes with SiO_2 coating improve the electrochemical performance (i.e., cyclic stability) of cells by reducing the leaching of Li-ions, improving structural integrity, and strengthening adhesion with the current collector. To our knowledge, this is the first time these types of effects have been observed for any coating, especially in aqueously processed cathodes.

We attribute the improved structural integrity of cathodes to the better cohesion among the SiO_2-coated NMC622 particles and improved binding of SiO_2-coated NMC622 with the CMC binder. Improved cohesion among SiO_2-coated NMC622 particles results from the conjugation reaction shown in Figure 4a between −OH groups on the SiO_2-coated NMC622 particles. This conjugation reaction is a common mechanism among the silanes to make SiO_2.[17] Similarly, the higher density of −OH groups on the SiO_2-coated NMC622 particle surface can increase its binding to the CMC binder (see Figure 4b) because there are also several −OH groups on the CMC binder surface. Similarly, increased density of −OH groups on the NMC622 particle surface can also increase the binding of SiO_2-coated NMC622 particles with the Al_2O_3/AlOH layer on the Al current collector (see Figure 4c) and strengthen adhesion of the cathode layer with the Al current collector.

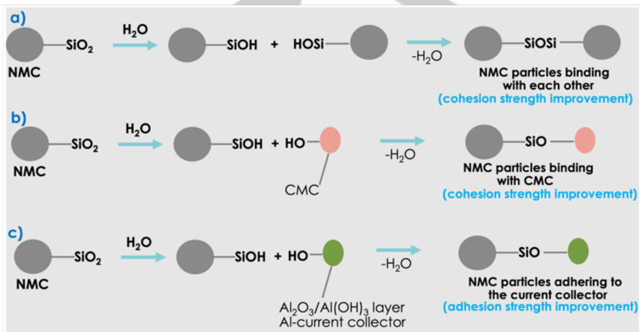

Figure 4. Expected binding mechanisms (a) among the SiO_2-coated NMC622 particles, (b) between the SiO_2-coated NMC622 particles and the CMC binder, and (c) between the SiO_2-coated NMC622 particles and the Al current collector.

We performed simple experiments to investigate our assumed mechanisms of enhanced particle–particle, particle–CMC binder, and particle–current collector interactions. In one experiment, we mixed equal amounts of SiO_2-coated and uncoated NMC622 particles (0.3 g each) in equal amounts of water (2 mL). The coated particles settled on the glass vial bottom within 5 min, whereas the uncoated particles remained suspended for longer (see Figure 5a&b). This experiment indicates that because of the higher number of −OH groups on the SiO_2-coated NMC622 particle surface, coated particles crosslinked with one another and settled quickly because of the large effective weights of the aggregates. In contrast, the uncoated particle surfaces did not have enough −OH groups to crosslink with one another. Thus, they stayed afloat as individual particles or small aggregates of particles. Aggregation of particles was further determined by SEM images, which showed large aggregates for coated particles and small aggregates or individual particles for uncoated particles (see Figure 5c). Additionally, zeta potential measurements provided lower absolute value of zeta potential for the coated particles (-41 mV) and higher for the uncoated particles (-54 mV). However, we expected that the coated particles would have higher absolute value of zeta potential because of more negatively charged −OH groups on their surface. This effect was nullified due to crosslinking (i.e., aggregation), which increased the effective particle size. A decrease in absolute zeta potential with particle aggregation resulting in an increased particle size is a known phenomenon.[18]

To test our hypothesis that SiO_2 coating also improves adhesion with the Al current collector in aqueously processed cathodes, we dipped equally sized Al foils in aqueous suspensions of SiO_2-coated and uncoated NMC622 particles, each with the same concentration (see additional details in the supporting information). The Al foil that was dipped in the SiO_2-coated NMC622 particle suspension adsorbed more particles than the Al foil that was dipped in the uncoated NMC622 particle suspension (see Figure 5d). Similarly, we coated the Al foil with a thin (2 µm) layer of CMC binder and dipped it in aqueous suspensions of SiO_2-coated and uncoated NMC622 particles.

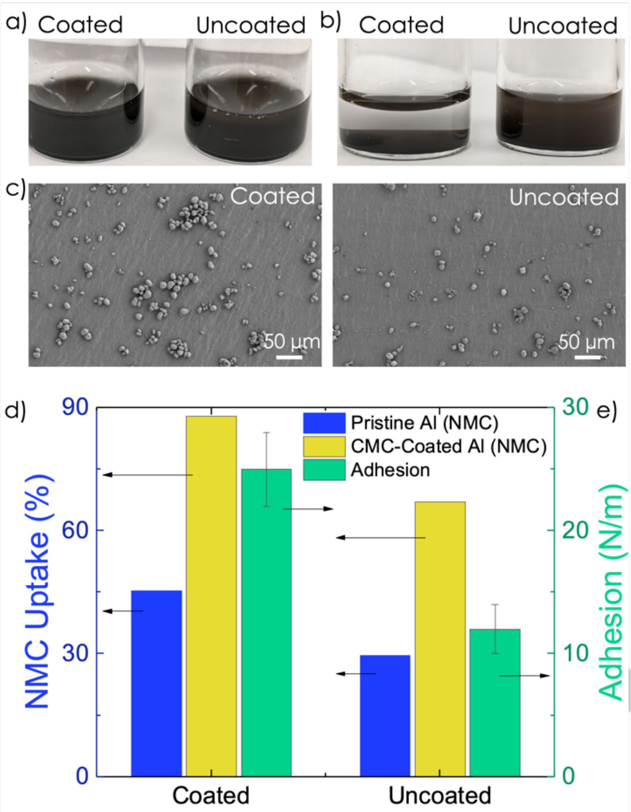

Figure 5. Photos of SiO_2-coated and uncoated aqueous suspensions of NMC622 (a) before and (b) after 5 min, (c) SEM images of coated and uncoated aqueous suspensions of NMC622, and plot showing (d) NMC622 particle uptake by pristine and CMC-coated Al foil and (e) adhesion strength of SiO_2-coated and uncoated NMC622 cathodes with an Al current collector. Note. NMC uptake % is calculated by normalizing the absorbed NMC particle weight with the original weight of Al-foil or CMC-coated Al-foil weight.

The CMC-coated Al foil that was dipped in the SiO_2-coated NMC622 suspension adsorbed more NMC622 particles than the CMC-coated Al foil that was dipped in the uncoated NMC622 particle suspension (see Figure 5d). Detailed information can be found in Table S1.This result supports our hypothesis that SiO_2-coated NMC622 particles have better binding capability with the CMC binder than uncoated NMC622 particles.

Conclusions

We demonstrated a low-cost and scalable strategy to make SiO_2 coatings on cathode materials (NMC622). The coated cathode material showed less leaching of Li-ions in aqueous suspensions. Additionally, we demonstrated for the first time that SiO_2 coating can affect the adhesion properties of cathodes with a current collector and the cohesion of cathode particles with one another and with binders in aqueously processed cathodes. Cyclic stability of full-coin cells with SiO_2-coated NMC622 cathodes was improved considerably compared with the coin cells with uncoated NMC622 cathodes. This coating strategy can also be used for other cathode materials, such as lithium nickel manganese oxide to improve their properties. Because previous coating studies have focused only on reducing parasitic reactions of cathodes particles with the electrolyte and did not examine possibilities of improved cathode structural integrity, we expect that this work will open new opportunities to design new coatings that can both reduce parasitic reactions and improve structural properties of aqueously processed cathodes based on other types of materials also

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Keywords: coating • cathode • battery • NMC • electrochemical

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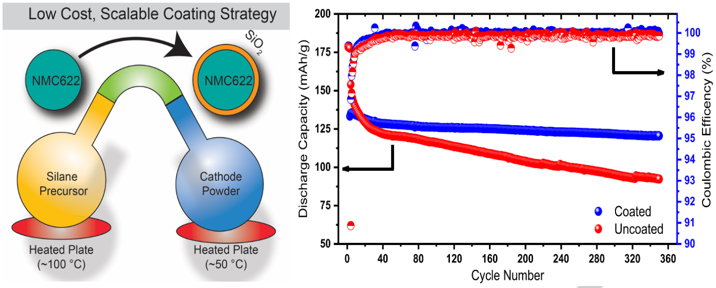

A low-cost and scalable strategy for coating the battery cathode materials with SiO_2 is demonstrated. Improved cohesion and adhesion strength and lesser cracks were observed in SiO_2-coated cathode materials, indicating increased cathode structural integrity. Electrochemical performance was also improved compared with the uncoated cathode materials in aqueously processed cathodes.