Mass-Scalable Molecular Monolayer for Ni-Rich Cathode Powder: Solution for Microcrack Failure in Lithium-Ion Batteries

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1. INTRODUCTION

Global warming issues make lithium-ion batteries (LIBs) become more important for building credible ESSs and EVs.1–5 However, the current energy level of LIBs is not sufficient for large applications. Various types of high-capacity anode materials have been suggested.6–9 However, candidates for high energy density positive electrodes are highly limited because of their complex crystalline structure.

For classical LiCoO$_2$ having theoretical capacity of 276 mAh·g$^{-1}$, its irreversible phase transition during high delithiation limits practical capacity to half at approximately 140 mAh·g$^{-1}$.10,11 Ni ions have been attempted as substitutes for the expensive cobalt ions in LiCoO$_2$ and stabilizers of a highly delithiated state for high capacity (>200 mAh·g$^{-1}$).12–15 Therefore, Ni-rich layered oxide cathode materials are the most proper candidates for actual LIB systems.

However, in a nickel-rich cathode, Ni ions should have a 3+ oxidation state, but they are prone to be Ni$^{2+}$, causing a phase transition to a NiO-like phase.15–17 This failure mechanism is severe at the electrolyte/electrode interface because of the contact of the chemically fragile electrolyte.18 These Ni-rich cathode materials usually have a secondary particle morphology that is aggregated by primary particles owing to the synthesis method of coprecipitation for homogeneous transition metal distribution. In many papers about electrolyte additives for Ni-rich cathode electrodes, Yang and co-workers demonstrated that the boundary between primary particles had unfavorable phase transitions which can be mitigated by an optimized electrolyte system with various kinds of lithium salts.1,12,13,19

Surface modifications that increase surface firmness by covering the fragile Ni$^{3+}$ surface with inorganic materials such as ZrO$_2$ and Al$_2$O$_3$ have been studied.17,21–24 These inactive coatings are beneficial for reducing the side reaction of the electrolyte by inhibiting the direct contact between the Ni-rich cathode surface and the electrolyte. Nevertheless, most coating materials, which are commonly prepared by precursor precipitation and a subsequent heat treatment at temperatures higher than 400 °C, are usually thick (from 100 nm to several micrometers).17 Moreover, achieving homogeneity on the entire surface of the particles is a difficult challenge because the
inorganic coating resulting from precipitation is easily aggregated. Inefficient thick inactive coatings, which are usually incorporated under 10 wt %, reduce the energy density of LIBs and cause high polarization.\textsuperscript{17} In addition, the cation mixing of Ni and Li ions and the highly fragile surface of Ni-rich cathodes limit the post-heat-treatment temperature to low values.

On the other hand, many recent papers have noted the microcrack problem in secondary particles of Ni-rich cathodes as a unique critical failure mode, which has not been seriously considered as a problem for other layered oxide cathodes.\textsuperscript{25} Many studies have demonstrated that the microcracks are due to special large anisotropic volume changes of the Ni-rich cathode during the charging process.\textsuperscript{5,27} Once unfavorable microcracks take place, the electron-conducting pathway in a particle is severely hindered, causing high polarization. Electrolyte can then penetrate into the secondary particles,\textsuperscript{28} which is the characteristic morphology of aggregations of the primary particles from the coprecipitation method.\textsuperscript{29,30} Therefore, the electrolyte side reaction on the boundary of the primary particles possibly triggers microcracking. Zhang and co-workers reported the coating on the primary particles from the coprecipitation method.\textsuperscript{29,30} In this study, a self-assembled monolayer (SAM) is first used to deliver an electrolyte-phobic surface modification on a Ni-rich cathode, LiNi\textsubscript{0.82}Mn\textsubscript{0.09}Co\textsubscript{0.09}O\textsubscript{2} (NCM82). For our electrolyte-phobic concept, decreasing wettability tends to increase resistance, and the electrolyte-phobic coatings must be made very thin. A SAM is a very versatile molecular-level thin coating that is homogeneously prepared via spontaneous chemical reactions and self-terminates after the formation of a single layer.\textsuperscript{31–34} The above-mentioned nature of the SAM is highly affordable because they are a few nanometers thick as well as homogeneous. Additionally, the characteristics of the SAM can be finely tuned by controlling the molecular structure of the coupling agent.\textsuperscript{32,33} Among the various SAMs examined, octyltrichlorosilane (OTS), one of the most credible SAMs, has been frequently used for SAM approaches in the other applications.\textsuperscript{35–37} Indeed, OTS is treated on the surface of the Ni-rich cathode material by vapor deposition to form a uniform nanolayer. With the OTS SAM, it is possible to suppress the penetration of the electrolyte into the particles and to prevent internal failures due to electrolyte decomposition inside the particles; the above mechanism suppresses the formation of microcracks.

2. EXPERIMENTAL SECTION

2.1. Preparation of OTS Coating. A Ni-rich layered oxide cathode, NCM82, was prepared by a previously reported method. First, transition metal precursors were prepared by coprecipitation with NiSO\textsubscript{4}·6H\textsubscript{2}O, CoSO\textsubscript{4}·7H\textsubscript{2}O, and MnSO\textsubscript{4}·H\textsubscript{2}O acting as the Ni, Co, and Mn sources, respectively. Second, the mixture with the prepared transition metal precursors and an excess amount of LiOH·H\textsubscript{2}O was calcined at 750 °C in an O\textsubscript{2} atmosphere for 20 h. Prior to treating the NCM82 surface, the NCM82 particles were washed by exposure to UV/O\textsubscript{3}. The surface of the NCM82 particles was modified by a vapor deposition method using octyltrichlorosilane (OTS, Tokyo Chemical Industry Co.). In the vapor deposition process, NCM82 powder (5 g) and an aluminum reservoir with OTS (0.5 mL) were loaded into a glass Petri dish for self-assembly. The glass Petri dish was mounted inside a vacuum chamber and left overnight. The temperature in the chamber was kept at 30 °C using a heating mantle. After the OTS vapor deposition process, nitrogen gas was used to purge the vacuum chamber to remove excess unreacted OTS vapor. Following this purge step, the NCM82 particle was thermally annealed at 130 °C, and then, the NCM82 particle was stored in a vacuum oven until characterization.

2.2. Electrochemical Measurements. The pristine and OTS-treated cathode powders were homogeneously mixed with a conducting agent of Denka Black and a binder of polyvinyl difluoride (PVdF, KF1100, Kureha) at a weight ratio of 96:2:2 by adding N-methyliptylridone (Aldrich) to prepare an electrode slurry by using PDM-300 automatic mixing equipment (KM tech). This slurry was cast on Al foil and dried in a convection oven at 120 °C for 10 min. The loading level was controlled at 10 mg cm\textsuperscript{−2}. The electrodes were pressed to a density of 3 g cm\textsuperscript{−2} using a roll-press machine (MP300, Roh tec). After the roll pressing, the electrodes were thoroughly dried in a vacuum oven overnight at 120 °C. 2032-type coin cells were assembled using a prepared composite electrode and lithium foil as the cathode and anode. To block direct contact between the cathode and anode, a polypropylene separator (Celgard) was used. The electrolyte, which consisted of 1 M LiPF\textsubscript{6} in a mixture of ethylene carbonate and diethylcarbonate (1:1 ratio by volume), was obtained from Panaxetec (South Korea). All 2032-type coin cells were
3. RESULTS AND DISCUSSION

3.1. Fabrication and Characterization of Electrolyte-Phobic Monolayer Coating. Figure 1a displays the molecular structure of the gas-phase OTS precursor. This long aliphatic precursor was selected because a long-saturated olefin group has nonpolar characteristics and is relatively repulsive to the polar electrolyte. The electrolyte-phobic monolayer surface modification on the active material was tried by this OTS precursor. In detail, Figure 1b exhibits a schematic illustration of the atomic-scale surface modification by a facile gas-phase precursor of OTS. First, Si–Cl, the reacting group in the OTS precursor, is readily hydrolyzed to Si–OH. Then, it is coupled with the native hydroxides on the surface of the Ni-rich cathode active material by forming a Si–O–Ni bond. This sequence repeats at the next site on the active material; therefore, the surface is homogeneously covered by OTS. Because of the alkyl-terminated end group of OTS, which is not reactive with additional OTS, the surface modification is just a single layer with the OTS chemical deposition, as shown in Figure 1b. The van der Waals force between adjacent OTS molecules produces several grains composed of ordered alkyl chain grains tilted 10°–15° to the active material surface. Finally, through the baking process at a moderate temperature of 130 °C, the remaining Si–OH bond that does not react with the active material is coupled with the Si–OH bond of the neighboring OTS so that O–Si–O bonds homogeneously cover the surface. H₂O is subsequently extracted by thermal annealing at 130 °C, resulting in the formation of a strong chemical Si–O–Si bond between neighboring OTS molecules. The moderate heat treatment temperature of 130 °C is also beneficial for Ni-rich layered oxide because cation mixing and NiO are inhibited at this low temperature.

Figure S1 displays X-ray diffraction (XRD) patterns. The XRD pattern of pristine sample confirms the R3m space group (JCPDS: 01-085-1968). After the SAM treatment, the XRD pattern from OTS-NCM82 has the same crystalline structure as well. Moreover, additional impurity phases are not found. With these XRD patterns between pristine and OTS-NCM82, it is confirmed that the OTS SAM precursor reacts only with the cathode material surface while preserving the original crystalline structure at the low temperature of 130 °C for the OTS coating.

Scanning electron microscopy (FE-SEM) images of both samples are depicted in Figure 2a–d. Pristine NCM82 consists of secondary particles with diameters of approximately 10 μm in which the primary particles are aggregated because they are synthesized by a coprecipitation method. The morphology of OTS-NCM82 in Figure 2b is also almost the same, consistent with the result of the XRD patterns. At the high magnification in Figure 2c, the pristine NCM82 has a clean surface with sharp edges, representing high crystallinity. However, the surface of the OTS-Ni-rich sample in Figure 2d has a different surface morphology with a round surface and merged edges. Generally, because the surface film by OTS is only a monolayer with a thickness of ~1 nm, the single molecular layer formed by the OTS-SAM treatment is not observed by SEM. The OTS treatment alters the surface to an organic material that does not have high electrical conductivity.
As a result, the secondary electrons for SEM analyses tend to charge on the insulating surface, which is thought to be the reason for the blurred SEM image of the OTS-Ni-rich sample, as shown in Figure 2d. Additionally, transmission electron microscopy (TEM) images are displayed in Figure 2e,f. The pristine material has a flat surface. It has been confirmed that a 1.2 nm thin layer with a different contrast in the image is found on the surface of the OTS-treated material. When the OTS precursor delivers a deposited molecular layer on the surface of the material, the thickness of the OTS layer is reported as 1.2 nm.99,40 Therefore, the presence of a 1.2 nm layer on the surface strongly supports that OTS reacts with the Ni-rich cathode material and that the OTS is polymerized with same adjacent molecules. To date, in a conventional coating method, it is difficult to control the coating at the nanoscale because they are usually prepared by precipitation methods, which deliver thick coatings from hundreds of nanometers to several micrometers. Therefore, the conventional coating harms the energy density of the LIB because of the excess of inactive coating material on the electrode. Occasionally, a micrometer-scale thick coating has a negative effect hindering surface lithium diffusion. In this new SAM method coating approach at the molecular scale, the coating can be reduced to a nanometer-scale thickness. Therefore, the SAM coating fundamentally solves the above-mentioned problem by reducing the coating layer as well as the surface lithium diffusivity in practical applications.

X-ray photoelectron spectroscopy (XPS) was performed to understand the OTS coating on the cathode material. Figure 3a shows the C 1s spectra of pristine and OTS-NCM82 before and after the 130 °C heat treatment. In the C 1s spectra of the pristine sample in the bottom spectra of Figure 3a, the curve is deconvoluted into three peaks of C−C/C=H at 285, C−O bonding at 288.5, and C=O at 290 eV.10,41 The additional peaks of C=O/C−O are considered to originate from the inevitable lithium impurities on the surface, such as Li2CO3, from the initial excess of the lithium precursor during the synthesis.5 In the top spectra of Figure 3a representing the surface of the OTS-Ni-rich sample, the intensity of the C−H/C−C peak at 285 eV is increased with or without heat treatment of 130 °C. The peak growths represent the presence of a long aliphatic chain from the octyl group of OTS anchored on the surface. Additionally, the C 1s at 290 eV is also highly reduced because of the surface coverage of OTS-SAM. In addition, the strong Si 2p peak in the top and middle spectra of Figure 3b for the OTS-coated sample, which is not found in the results from the pristine sample, is distinct evidence to support the presence of the OTS layer because the OTS has Si in the anchoring group. In O 1s spectra, the OTS-Ni-rich sample before the heat treatment exhibits a strong peak at 533 eV corresponding to Si−OH. This peak is highly reduced in the top result from the final OTS-Ni-rich sample after the heat-treatment. The most investigated SAM systems are alkyltrichlorosilanes, which are usually believed to irreversibly graft both to the surface and to the neighboring SAM molecules by a siloxane Si−O−Si bonding network. Organo-monolayers are formed by condensation on substrates exposing hydroxyl groups of substrates because of the extensive binding of the head groups to the surface via strong and localized bonds.32−46

For quantitative analyses of Si in the bulk structure, ICP-OES was performed (Table S1). The transition metals were highly correspondent with the designed stoichiometry for NCM82. The Si content from the OTS coating was not available for ICP analyses, indicating the coating amount was almost negligible. Considering that the surface-sensitive XPS analyses exhibit a measurable Si 2p peak in OTS-NCM82, Si is concentrated only on the surface as a surface SAM treatment. This opposite behavior between XPS and ICP-OES analyses proves that only a thin monolayer of the surface layer containing Si successfully forms during the OTS-SAM coating without consuming a significant mass of the active material for use in battery applications.

To check whether the surface characteristics are changed through OTS treatment, we prepared pellets with only active materials. The contact angle was measured by dropping the electrolyte on these pellets. For the pristine NCM82 pellet, the electrolyte directly penetrates into the pellet, and the contact angle is measured as low as 3.3° (Figure 4a). On the other hand, after the OTS treatment, the contact angle is greatly increased to 42.5° (Figure 4b) by hindering wettability. In addition, the prepared powders are dispersed in a water solvent. Figure 4c shows that these samples have completely different surface characteristics. After pouring the pristine NCM82 powder in the electrolyte, it instantly begins to disperse and fall to the bottom, whereas the OTS powder floats on the surface of the electrolyte without dispersing (Figure 4c). The different dispersions are constant after 4 h, as shown in the right-most panels in Figure 4c. Similar to the case with the electrolyte shown in Figure 4a,b, the pristine sample is also easily dispersed in a water solvent, but the OTS-treated powder is not highly dispersed in the solvent. Furthermore, it not only aggregates but also separates from the electrolyte even though the density of the particles is much greater than that of the water.

The cross-sectional area was observed by energy dispersive X-ray spectroscopy (EDS) mapping with fluorine after stirring in the electrolyte for 24 h to provide enough time for electrolyte wetting. After the wetting process, cross-sectional images of the particles were obtained to determine whether the electrolyte was soaked inside the secondary particles via gaps among the primary particles (Figure 5a−d). The cross-sectional SEM images of the pristine and OTS-Ni-rich samples have a clear pattern. In the case of the pristine sample shown in
results in poor wettability and low surface energy. The results support that the OTS treatment leads to the electrolyte-phobic layer. High wettability is commonly pursued to reduce the resistance by increasing the active interface. The pivotal role of the reduced wettability by thin coating of the Ni-rich cathode will be discussed in detail in a later section.

3.2. Electrochemical Performances of OTS-NCM82.

The initial voltage curves from both samples are depicted in Figure S2a. The OTS treatment causes slightly lower charge and discharge specific capacities of 225.8 and 186.0 mAh g⁻¹ compared to the pristine samples with values of 233.6 and 199.6 mAh g⁻¹, respectively. In dQ dV⁻¹ curves in Figure S2b, three pairs of redox peaks related to the phase transition are observed in both curves. During delithiation, the first peak at 3.7 V vs Li/Li⁺ represents the initial hexagonal structure changing to a monoclinic structure. The next peak at 4.0 V reflects the phase changes from a monoclinic to a hexagonal structure. The final peak at 4.15 V represents the formation of the third hexagonal structure.20,38 As shown in the EDS mapping image in Figure 5, the electrolyte in the pristine NCM82 secondary particle increases the active interface between the primary particle and electrolyte for initial delithiation, whereas the OTS sample has only the electrode/electrolyte interface of the secondary particle outer phase. For the comparison of ion diffusion behaviors, galvanostatic intermittent titration technique (GITT) voltage curves were analyzed in Figure S2c,d. The measured lithium ion diffusivities were comparable to each other, despite the diffusivity of the OTS-NCM82 being slightly low. In terms of wettability between active material particle and electrolyte, the electrolyte-phobic surface treatment limits this interface area, and therefore, the OTS-treated sample has initial polarization slightly higher than that of the pristine electrode; however, this is not significant.

Figure 6a shows the cycle performances of the pristine and OTS-treated electrodes. Even though the initial capacity of the OTS sample cycle is recorded as 186.0 mAh g⁻¹, which is less than that of the pristine sample (199.2 mAh g⁻¹) owing to its high kinetic hindrance from the small active interfacial area contacting the electrolyte, the capacity of the pristine sample decreases drastically after the 70th cycle because of high polarization (42.6 mAh g⁻¹ at the 100th cycle).36–49 On the other hand, this rapid cyclability failure mode is highly mitigated by the OTS coating, showing a value of 136.5 mAh g⁻¹ after the 100th cycle. In detail, the obtained voltage curves and the derived dQ dV⁻¹ curves over cycles are described in Figure 6b–e. First, highly increasing polarization is clearly observed in Figure 6b for the pristine sample. In addition to the high polarization growth in the cell having the pristine sample after the 60th cycle, dQ dV⁻¹ results show that the polarization is steadily increasing even in the results before the 60th cycle. The delithiation peak at 3.6 V for the first cycle is gradually polarized to 4.1 V for the 100th cycle in Figure 6d. The behavior of the reverse peaks on the lithiation process displays a similar pattern over increasing cycle numbers. However, the OTS-NCM82 sample provides highly repeating dQ dV⁻¹ curves in Figure 6e as well as highly preserved voltage curves in Figure 6c. The electrochemical failure of the pristine sample is highly mitigated by the OTS-SAM coating. The OTS coating is highly beneficial for improving the cyclability by solving the problem of high polarization.

Electrochemical impedance spectroscopy (EIS) analyses were performed to analyze the polarization after the first and
70th cycles. Two semicircles are clearly observed in all results, and the related equivalent circuit is shown as an inset in Figure 7a. The fitted line is also described by solid red lines. In this regard, the high-frequency semicircle on the left side and the medium-frequency semicircle on the right side are assigned as the resistances from the surface film and charge transfer, respectively. Especially for the results for the first cycle, the sum of the film and charge transfer resistance from the OTS-NCM82 sample is larger than that from the pristine sample. This result is highly consistent with the initial high polarization of the OTS-NCM82. Interestingly, the intercept on the Z’ axis where the high-frequency semicircles begin reflects the internal series resistance, including the contact resistance. In the EIS results of the pristine sample after the 70th cycle, the contact resistance is dramatically increased compared to that for the first cycle. The fitted contact resistance suddenly increased from 2.5 to 13.4 Ω. This resistance growth is not observed in the OTS-coating sample, as evidenced by the preservation of the Z’ axis intercepts from 3.6 to 5.6 Ω in Figure 7b.

3.3. Suppression of Microcracks. To understand this failure mode, we disassembled the cells after 50 cycles, and cross-sectional SEM analysis was performed again. The SEM images of both electrodes after 50 cycles are indicated in Figure 8. In Figure 8a, the secondary particles are totally broken and segregated because of microcracks in the pristine electrode. Interestingly, the microcrack problem is not highly
significant in the OTS electrode, as shown in Figure 8b. It is known that the Ni-rich layered oxide cathode has a critical problem that is estimated by large lattice changes during the delithiation process. However, the microcrack issues are highly relieved by surface coating. We believe that the OTS coating blocks electrolyte penetration, thus avoiding the internal electrolyte decomposition and internal gas generation from electrolyte decomposition. On the other hand, panels c and d of Figure 8 show the cross-sectional images and the F-EDS mappings of the particles of the pristine and OTS-NCM82 electrodes, respectively, after 50 cycles. A cross-sectional image of the microcrack in the white rectangle in Figure 8a is enlarged as an inset in the same panel. The inner rough surface also reflects the formation of a passivation film, the so-called cathode electrolyte interphase (CEI), which is thought to be derived by electrolyte decomposition.10,50–52 This CEI on the surface accelerates the phase transitions. The relieved electrochemically fragile electrolyte on the surface accelerates the phase transitions. The relieved electrolyte–electrode surface was thought to be effective to preserve the initial crystalline structure, which is essential for long cycle life.

For rate capabilities, the voltage curves with various current densities are shown in Figure S5. The discharge specific capacities of the pristine and the OTS-NCM82 were comparable to each other with the discharge current densities lower than 2C. With a further high current density of 5C, the specific discharge capacity of OTS-NCM82 (130.8 mAh g\(^{-1}\)) was not as high as the 172.1 mAh g\(^{-1}\) of the pristine electrode. Whereas the Ni 2p peak of the pristine sample is shifted to negative binding energy because of the formation of Ni\(^{2+}\) being originated from the phase transition from the active material, the Ni 2p peak from the OTS-NCM82 is relatively closed to Ni\(^{3+}\), which is the initial oxidation state. It is thought that the surface irreversible phase transition is also highly relieved by OTS coating via bypassing the electrolyte decomposition. Therefore, this OTS coating is highly effective to reduce the electrolyte decomposition as well.

In Figure S4, the XRD patterns from the cycled electrodes have different peak ratios between (003) and (104) planes from the initial XRD patterns of the electrodes. For the pristine and OTS electrodes in Figure S4a, the peak ratio of (003)/(104) is almost constant. After the 50th cycles, the ratio was decreased to 0.67 and 0.83 for the pristine and OTS electrode, respectively. The reduced peak ratio reflects the phase changes by cation mixing and biphase formation which are considered as negative effects for the electrochemical performance. The high-intensity (104) peak in the pristine XRD pattern indicates that the pristine sample has more cation mixing by NiO formation, which is thought to originate by phase transition. It is also explained by the OTS effect to reduce surface failure of the active materials. The relieved microcracking by reducing electrolyte penetrations was effective to mitigate the phase transition. The electrochemically fragile electrolyte on the surface accelerates the phase transitions. The relieved electrolyte–electrode surface was thought to be effective to preserve the initial crystalline structure, which is essential for long cycle life.
substances are usually resistive by hindering ion diffusion. It is believed that the angstrom-scale thin surface modification by OTS balances the resistive behavior of electrolyte-phobicity with preservation of the coating benefits.

Figure 9 shows a schematic summary of the changes in the pristine and OTS-NCM82 particles after wetting with the electrolyte and cycling. In the case of the pristine NCM82, the electrolyte penetrates inside the secondary particles along the boundaries of the primary particles, as indicated by the red line. As cycling proceeds, electrolyte side reactions occur inside the particles, producing gas-phase byproducts, which severely accelerate the microcrack failure initiated by volume expansion during the charge−discharge process. On the other hand, when the surface of NCM82 is treated with OTS, electrolyte penetration stops on the surface of the secondary particles. Therefore, microcrack formation is finally suppressed by preventing the internal electrolyte decomposition during electrochemical cycling. On the basis of these conclusions, the failure phenomenon is attributed to the microcrack acceleration by the electrolyte side reaction that occurs when the electrolyte penetrates into the secondary particles and the cycle progresses. Therefore, the electrolyte-phobic surface film of the OTS coating prevents electrolyte penetration and inhibits microcrack formation.

4. CONCLUSIONS

The surface of the Ni-rich layered oxide cathode material was modified by using an OTS molecular-layer deposition via a vapor deposition method. A single OTS layer on the surface of the Ni-rich layered oxide cathode effectively controlled the coating and completely changed from hydrophilic to hydrophobic and electrolyte-phobic. Whereas the pristine sample demonstrated that the electrolyte soaked through the primary particle boundaries, the electrolyte-phobic surface of the OTS molecular layer greatly prevented electrolyte penetration into the core position of the particle. The reduced electrolyte wettability caused a slightly higher polarization during the initial cycle; however, the cyclability was highly improved.

After repeated cycles, it was found that the particle pulverization of the pristine Ni-rich layered oxide electrode was greatly accelerated by electrolyte decomposition occurring internally, causing high pressure owing to the production of gas-phase byproducts. As a result, the electrolyte decomposition in the secondary particles and the internal gas generation from the side reactions could be suppressed by using the OTS-surface treatment. Microcracking accelerated by the side reaction inside the particles was highly relieved, and the cycling performance of the OTS-treated cathode material could be highly improved.

■ ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.1c03680.

X-ray diffraction patterns and lattice parameters of pristine NCM82 and OTS-treated NCM82; initial potential and $dQ/dV^{-1}$ curve plots from the coin half-cell with pristine NCM82 and OTS-NCM82; XPS results of the pristine and OTS-Ni-rich electrode after 50th cycle; ICP-OES results of the pristine and OTS-Ni-rich powder (PDF)

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Notes

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