Polyurethane Binder for Aqueous Processing of Li-Ion Battery Electrodes

Nicholas Loeflter,a,b,c Thomas Kopel,c Guk-Tae Kim,a,b,c,∗ and Stefano Passerini,a,b,∗

aHelmholtz Institute Ulm (HIU), Electrochemistry 1, 89081 Ulm, Germany
bKarlsruhe Institute of Technology (KIT), 76021 Karlsruhe, Germany
cInstitute of Physical Chemistry, University of Muenster, 48149 Muenster, Germany

In this manuscript the investigation of the heat-curable polyurethane (PU)/carboxymethyl cellulose (CMC) mixture as binder for Li-ion battery electrodes is reported. The experimental results demonstrate the outstanding thermal and electrochemical stability of PU, as well as the good electrochemical performance of cathode (NMC) and anode (graphite) electrodes in which PU is used as binder. The most important finding is the ability of PU to prevent current collector corrosion usually occurring when an aqueous dispersion (slurry) of NMC is coated on aluminum foils. SEM investigation shows that PU encapsulates the positive active material particles preventing a pH raise in the slurry. Finally, the cycling performance of PU/CMC based anodes and cathodes are tested in half as well as full lithium-ion cell setups.

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The awareness of the finite nature of our earth’s natural resources as well as growing concerns for increased greenhouse gas emissions are leading to a strong demand for a more environmentally-friendly generation of energy. Considering the importance of sustainable energy solutions, strong efforts are made to develop improved systems for large and small scale energy storage.1–3 Currently, Li-ion batteries seem to be suitable energy storage devices to fulfill the requirements of small energy storage due to their high energy density and long lifetime.4 Additionally, their lightweight fulfills very well the rising demand for use in smartphones, computers and other portable devices, as well as for electromobility solutions, facilitating the development toward a modern mobile society.

Lithium-ion batteries are composed of several materials including metals and metal oxides, graphite and other carbonaceous materials, organic solvents, lithium salts and polymers. These latter are present in the porous separator, laying in between the electrodes (usually a polyolefin), and as binding components in the composite electrodes. Binders are counted among the so-called inactive components since they do not directly contribute to the capacity of the cells. However, their key role in the electrode processing and their dramatic influence on the electrochemical performance of electrodes has been extensively outlined.4–5 Relevant physical and chemical properties for binder materials are (i) thermal stability, chemical and electrochemical stabilities, (ii) tensile strength (strong adhesion and cohesion), (iii) flexibility, and (iv) viscosity (of the slurries).6–9 The main purpose of using a binder is to form stable networks of the solid electrode components, i.e., the active materials and conducting agents (cohesion). Moreover, the binder has to ensure a close contact of the composite electrode toward the current collector (adhesion).

In state-of-the-art commercial Li-ion batteries polyvinylidene-difluoride (PVdF) is the binder material of choice due to its superior adhesion properties and electrochemical stability.10 Important points of criticism for this polymer, however, are the requirement of volatile and toxic solvents, such as N-methyl-pyrrolidone (NMP) as well as high costs for both, the binder and the solvent. Furthermore, the recycling of the battery components at the end of life involves firing or dissolution of PVdF (in NMP), which are posing further environmental problems. All of these concerns might be strongly reduced, if not eliminated, by using water soluble binders.11–14 This, thus, with the aim for cheaper, safer and environmentally friendlier batteries, water-soluble binders have gained great significance.11,13,15–18 Aqueous electrode processing is easier and cheaper compared to the solvent-based approach (i.e., PVdF), because there is no need for, e.g., controlling of humidity during electrode production.19,20 However, while polymers dissolved or dispersed in water (and their mixtures) are nowadays the state-of-the-art binder materials for commercial Li-ion anode electrodes, the making of cathode electrodes with water-based binders is still under ongoing investigation because of several problems arising when the metal oxide-based active materials are processed with water. The two main problems are, actually, associated to the reactivity of the above-mentioned oxides with water leading to leaching of Li+ ions as well as metal cations, and strong pH increase, in turns causing the corrosion of the aluminum current collector.21–24

In this work we introduce a water-processable polyurethane/carboxymethyl cellulose mixture as binder for oxide-based cathode electrodes, strongly reducing the above mentioned inconveniences. The binder is found to offer outstanding performance also for the manufacturing of anode (graphite-based) electrodes.

When it comes to Li-ion battery research, so far, functionalized polyurethanes have mostly been investigated as component for gel polymer electrolytes (GPEs) as reported by Wen et al.,25 Kim et al.26 and Cao et al.27 to name but a few. Sato et al.28 used a caprolactone segmented polyurethane as electrode constituent (to delay the onset of a thermal runaway at the cathode electrode) in combination with a GPE to investigate a polymer battery with improved safety characteristics. Only very few authors reported details on the use of water-dispersible polyurethanes as binders for Li-ion battery electrodes. Hwang et al.29 synthesized a waterborne polyurethane binder, comprising double bonds and a (chemically bonded) dispersant, which facilitates its dispersibility in water. The double bonds may be additionally crosslinked by addition of a water-soluble initiator, resulting in enhanced elastic properties. While the authors researched the impact of different polyurethane binder formulations on the volume change during charge and discharge of only graphite anodes, however, no electrochemical data was reported. Very recently Zhu et al.30 investigated self-synthesized nonionic waterborne polyurethane as binder for cathode electrodes, utilizing lithium iron phosphate (LFP) as active material.

To shed more light on the opportunities of polyurethanes as binder in aqueous-processed Li-ion batteries, we examined the electrochemical stability of a one component heat-curable polyurethane dispersion, the influence of different polyurethane concentrations on the performance of anode and cathode electrodes and their electrochemical properties in a Li-ion battery. CMC was used in small percentages as electrode thickener to maintain a favorable viscosity of the electrode slurries.
Table I. Formulation of prepared electrodes.

|                | Anode | Cathode |
|----------------|-------|---------|
| Active Material| 93    | 90      |
| Conductive Agent| 4    | 7       |
| Binder         | 3     | 3       |

**Experimental**

Electrode preparation.— The electrode slurries used in this work contained the active material, conductive agent, binder and water, resulting in the dry electrode compositions given in Table I. For the investigation of the waterborne polyurethane dispersion (EVONIK, blocked isophoronediisocyanate (IPDI) emulsified in water, one component heat curing system) as binder in water-processed electrodes, different electrode formulations were prepared (as given in Table II). The slurries containing only the polyurethane binder (no acronym) showed very low viscosities, making the coating of the electrode on the current collector metal foil impractical. Therefore, those electrode formulations were discarded from this study. Electrodes made using only CMC compositions (RefC and RefA) were used as reference to assess the effect of the polyurethane binder on cathode and anode electrodes’ performance. For the electrode formulations C1, C2 and A1, the first step consisted in dispersing the polyurethane binder in deionized water (magnetic stirring for nearly 30 minutes). In the next step (first step for the binder formulations RefC and RefA) sodium carboxymethyl cellulose (CMC, DOW WOLFF CELLULOSICS, degree of substitution of 1.2) was admixed into the aqueous solution and stirred at 300 rpm overnight. Then the required amount of conductive agent C-NERGY Super C45 (TIMCAL, primary average particle size: 30 nm) was added and stirred for 3 hours. Afterwards the active material (cathode or anode) was admixed into the slurry. As anode and cathode active materials, commercially available SLP 30 graphite (TIMCAL, average particle size: d_{90} = 30 μm) was added and stirred for 3 hours. Afterwards the active material (cathode or anode) was admixed into the slurry. As anode and cathode active materials, commercially available SLP 30 graphite (TIMCAL, average particle size: d_{90} = 30 μm) was added and stirred for 3 hours. Afterwards the active material (cathode or anode) was admixed into the slurry. As anode and cathode active materials, commercially available SLP 30 graphite (TIMCAL, average particle size: d_{90} = 30 μm) was added and stirred for 3 hours.

After stirring for 3 hrs, the cathode slurry was dispersed with a T 25 Ultra Turrax at lowest stirring speed for 30 minutes to get a more homogeneous dispersion. The same procedure was not used for the anode to avoid damaging the soft graphite particles.

Copper (Schlenk, thickness: 10 μm) and aluminum (thickness: 20 μm) foils were used as current collector, respectively, for the anode and cathode. The metal foils were washed with ethanol to remove impurities, such as dust. The mixtures were coated on the appropriate metallic foil using a doctor blade technique and, then, immediately dried at 80 °C in an atmospheric oven for approximately 30 minutes.

The cathode electrodes were cut into stripes (width: 6 cm), which were roll-pressed with a calendering machine (HOHSEN CORP, Roll Press Machine HSRP-2025) loaded with a line force of about 50 μm as counter electrode, a single layer of polyethylene separator (ASAHI KASEI, Hipore SV718), and the cathode disc electrode. In all pouch bag cells, 120 μL of electrolytic solution (1M LiPF₆ in EC:DMC (1:1 w/w), LP30, BASE) were added.

**Table II. Composition of the binder (mixtures) for the prepared electrodes.**

|                | CMC (%) | Polyurethane (%) | Total binder amount (%) | Acronym | Cathode/Anode |
|----------------|---------|------------------|------------------------|---------|---------------|
| a)             | 3       | 0                | 3                      | RefC/RefA |               |
| b)             | 2       | 1                | 3                      | C1/A1   |               |
| c)             | 1       | 2                | 3                      | C2/−    |               |
| d)             | 0       | 3                | 3                      | −/−     |               |

Round cathode and anode electrodes were punched out of the coated foils and dried for 12 hours in a glass oven at 120 °C under vacuum.

Electrochemical tests.— The electrochemical tests of cathodes were performed on half-cells, which were vacuum-sealed in pouch bags. The half-cells were assembled in the dry room (R.H. < 0.1%) using metallic lithium (ROCKWOOD LITHIUM, battery grade, thickness: 50 μm) as counter electrode, a single layer of polyethylene separator (ASAHI KASEI, Hipore SV718), and the cathode disc electrode. In all pouch bag cells, 120 μL of electrolytic solution (1M LiPF₆ in EC:DMC (1:1 w/w), LP30, BASE) were added.

The electrochemical investigation of the anodes was performed in three-electrode Swagelok cells. The cells were assembled in the glove-box under inert argon atmosphere (H₂O and O₂ < 0.1 ppm).

The graphite electrodes were tested in half-cell setup using metallic lithium (ROCKWOOD LITHIUM, battery grade) for the counter and reference electrodes. A glass fibre (Whatman, GF/D) separator soaked with 120 μL of LP 30 was used as the electrolyte layer.

Full lithium-ion (Graphite/NMC), vacuum-sealed, pouch-bag cells were assembled in the dry-room by piling up anode, separator and cathode. Copper (anode) and aluminum (cathode) tabs were used as cell terminals while the Asahi membrane soaked with LP30 electrolyte was used as separator.

Thermogravimetric analysis.— Thermal measurements of polyurethane were performed using a Q 5000 IR TGA instrument (TA INSTRUMENTS). Platinum pans containing 8 mg of sample were heated up from 30 °C to 1000 °C with a heating rate of 5 °C min⁻¹. As purge gas nitrogen was used with a gas flow of 25 mL min⁻¹.

Scanning electron microscopy.— Scanning electron microscopy images were prepared by a ZEISS EVO MA 10 microscope. Images were taken from different electrodes. To investigate corrosion issues at the surface of aluminum foils, the composite coating was removed from some electrodes and images of the underneath bare metal foils were taken.

Cyclic voltammetry.— Measurements of the electrochemical stability window (ESW) of polyurethane binder were carried out on bare and polyurethane-coated aluminum and copper foils using a VMP3 (BIOLOGIC). Polyurethane was casted on aluminum and copper foils with an approximate coating thickness of 10 μm. After pre-drying the foils in an atmospheric oven at 80 °C for 30 minutes, round electrodes with a diameter of 12 mm were punched and dried for 12 hours at 120 °C in a glass oven under vacuum. All electrochemical tests were performed in three-electrode Swagelok cells using metallic lithium for the counter and reference electrodes (see Electrochemical tests). For polyurethane-coated aluminum foils the potential was ramped from OCV to 5.0 V vs Li/Li⁺ and then cycled between 1.5 V to 5.0 V vs Li/Li⁺ (anodic sweep). For the polyurethane coated copper foils the potential was ramped from OCV to 0.02 V vs Li/Li⁺ and then cycled between 0.02 V to 3.0 V vs Li/Li⁺ (cathodic sweep). All tests were performed at a scan rate of 0.5 mV s⁻¹.

Charge/discharge testing.— Electrochemical charge/discharge tests were performed with a MACCOR Battery tester 4300 at room temperature (20 ± 2 °C). Cathode half-cell tests started with a rest step of eight hours to ensure the homogeneous wetting of the electrodes by the electrolyte. Initially, cathode half-cells were cycled with a charge/discharge rate of 0.1 C for 5 cycles. Thereafter, the C-rate was increased to 1 C for 50 cycles. The lower and upper cutoff potentials were set to 3.0 and 4.3 V (vs Li/Li⁺), respectively.

Analog to the cathode half-cells, anode half-cells started with a rest step of eight hours. The cycling tests were carried out between 0.02 V and 1.5 V (vs Li/Li⁺) cutoff limits and started with two cycles at the C-rate of 0.1 C, to favor the SEI formation, followed by 50 cycles at 1 C rate. The lithiation step in the constant current (CC) mode was followed by a further constant voltage (CV) mode (constant
voltage: 0.02 V, cut-off current: 0.02 C) to emulate the standard charge protocol used for lithium-ion batteries (LIBs). In fact, the electrochemical investigation of full lithium-ion cells was also carried out using the CCCV test protocol within the cell voltage range from 3.0 V to 4.2 V (cathode limited). After three initial formation cycles at 0.1 C the current density was increased to 1 C for 200 cycles.

Results and Discussion

Fig. 1 shows the weight loss profile of the polyurethane dispersion, which was dried at 80 °C prior to conducting TGA measurements. The result indicates that the dried polyurethane dispersion contained approximately 5 wt% water, which is released below 140 °C. The thermal decomposition of the polyurethane polymer starts above 170 °C with a weight loss of nearly 92 wt% up to 455 °C. Above 455 °C no considerable residue was left, meaning that the polymer was completely decomposed. The TGA results grant for the polyurethane thermal stability up to 170 °C. Therefore, the drying temperature for anode and cathode electrodes was set to 120 °C under vacuum.

The results of cyclic voltammetry measurements of bare and polyurethane coated aluminum and copper foils are presented in Fig 2. The dashed arrows in the panels point in the direction of increasing cycle numbers. The consecutive CV curves of bare aluminum foil (Fig. 2a) show a quite stable trend with a rather low current flow. The observed decrease of current flowing at potentials above 4.0 V vs Li/Li⁺ is caused by the well-known formation and growth of the passivation layer protecting aluminum from anodic dissolution. This layer mostly consists of aluminum fluoride (AlF₃) originating from the decomposition of the electrolyte salt anion (PF₆⁻). This effect is also noticeable, although at a lower extent, in Fig. 2b, indicating that the aluminum surface protection mechanism takes place also in the presence of the polyurethane coating. Neither for the bare aluminum nor for the polyurethane coated aluminum foils remarkable faradaic current responses are detectable, which would indicate major decomposition or dissolution reactions. Thus, it can be assumed that the polyurethane is stable at the potentials occurring at the cathode electrode.

The CVs in Fig 2 (panels c and d) refer to bare and polyurethane-coated copper foils. In the first cathodic sweep of both electrodes, remarkable faradaic current responses are observed. Thus, these peaks cannot be attributed to polyurethane decomposition. Instead, as already reported in literature, they originate from the reduction of electrolyte solvent molecules (between 0.02 V to 1.00 V vs Li/Li⁺), interaction of Li⁺ ions with the native copper oxide layer (0.5 V/0.6 V vs Li/Li⁺) and, finally, the reduction of residual oxygen and water in the electrolyte (1.1 V to 1.7 V and 1.6 V vs Li/Li⁺, respectively).

Figure 1. TGA weight loss profile of polyurethane.

Figure 2. Cyclic voltammogram of a) bare Al-foil, b) polyurethane coated Al-foil (depicted cycles: 1st, 2nd, 3rd, 5th, and 10th), c) bare Cu-foil and d) polyurethane coated Cu-foil (depicted cycles: 1st, 2nd, 3rd, 5th, and 10th). For cell details see the Experimental section.
Therefore, it can be concluded that polyurethane is electrochemically stable at the potentials occurring at the anode electrode.

Following, the performance of polyurethane-based NMC cathodes (C1 and C2) was investigated and compared with that of CMC-based reference electrode composition (RefC). In Fig. 3 the cycling performance of each cathode electrode formulation in half-cells is illustrated. At the first glance, it appears evident that a distinct capacity fading affects the RefC cathode. This capacity loss, already evident in the 10th cycle (121 mAh g⁻¹ vs 123 mAh g⁻¹ at the 6th cycle), continuously increases up to the 100th cycle (104 mAh g⁻¹), resulting in the average capacity fading of 0.19 mAh g⁻¹ per cycle and the capacity retention of only 86% (after 100 cycles). A different trend is observed for C1. After the 10th cycle, the C1 electrode shows a remarkably stable cycling performance with the average capacity loss per cycle being only 0.09 mAh g⁻¹ and the capacity retention after 100 cycles being 93%. The best electrochemical performance is, however, observed for the C2 electrode. While the enhanced capacity might originate from the lower areal loading of C2 electrodes (3.1 mg cm⁻²) compared to 5.1 mg cm⁻² and 5.4 mg cm⁻² for C1 and RefC, respectively, though, C2 does not suffer of any distinct capacity fading. This results is even more surprising considering that a capacity fading has always been observed in water-processed, metal oxide-based electrodes because of the aluminum current collector corrosion.36–38 Thus, it is clear that polyurethane is somehow avoiding such a corrosion. To gain a deeper understanding on this excellent result, SEM images of the various cathode electrodes were taken (Figs. 4a–4c). Moreover, to investigate the surface of the aluminum current collector underneath the composite material coating, the latter was removed and SEM picture of the aluminum surface were also taken (Figs. 4d–4f). Fig. 4a shows the typical morphology of the commercial NMC powder with tiny primary particles (0.5 to 1 μm) agglomerated into larger secondary particles (5 to 10 μm), which are surrounded by the network of conductive carbon nanoparticles. As usual, the CMC binder is not evident in the SEM image. However, the addition of polyurethane (Figs. 4b and 4c) results in the appearance of a polymeric layer onto the surface of the NMC particles. The extent of this polymeric layer strongly depends on the amount of polyurethane added, as a matter of the fact, the primary NMC particles can hardly be recognized in C2 (Fig. 4c). In parallel, a decreasing pitting corrosion (dissolution) is observed on the underneath aluminum current collector. In fact, while pits are large and deep in the RefC current collector (Fig. 4d), their size decrease with increasing amount of polyurethane binder, which seems to inhibit the onset of aluminum corrosion. In sample C2 only small surface features are observed, which did not evolve into pits of the aluminum foil. Aluminium corrosion is known to arise from the high pH achieved when the NMC active material gets in contact with water.35 Thus, it is more than plausible to assume that the observed encapsulation of the active material particles by the polyurethane reduces the interaction between the metal oxide and water, leading to the observed reduced aluminium corrosion. As already demonstrated in earlier work,36 this results in improved electrode performance as shown in Fig. 3.

To further investigate the influence of polyurethane as binder for NMC cathode electrodes, differential capacity plots of RefC and C1 electrodes were elaborated. These two kinds of electrodes were used because of the comparable active material areal loadings. Actually, due to lower areal loading, C2 cathodes were discarded from this comparative analysis and the following full cell investigations (impractical cathode : anode capacity ratios). The differential capacity curves of RefC and C1 computed for the 5th (0.1 C rate) and the 50th (1 C rate) cycles are illustrated in Fig. 5. Surprisingly, no significant changes in the onset potentials of the oxidation and reduction processes are observed. The differential capacity versus voltage profiles of RefC and C1 electrodes are pretty much overlapping, indicating that polyurethane does not considerably hinder the electrochemical reactions, i.e., the Li⁺ ion transfer into and out of the active materials particles, although it coats quite effectively the active material particles. This is in good agreement with existing literature showing that polyurethane-based GPEs exhibit appreciable ionic conductivities.25–27 and with the above reported ESW measurements (see Fig. 2), which demonstrated that the electrolyte ions could very well pass through the rather thick polyurethane layer.

To investigate the electrochemical performance of C1 cathode in full Li-ion cells, graphite anode electrodes were made using the same binder composition (A1) as well as using only CMC (RefA). Fig. 6 illustrates the cycling performance (panel a) and the 50th cycle voltage profiles (panel b) of RefA and A1 (active material mass loadings: 3.5–4.0 mg) in half-cell configuration. After the initial three formation cycles (forming the solid electrolyte interphase (SEI)) at 0.1 C, the RefA electrode (Fig. 6a) showed the expected cycling behavior delivering a capacity of 374 mAh g⁻¹ at the 10th cycle. The slight capacity excess is obviously due to the contribution of the carbon conductive additive, which mass is neglected in the specific capacity calculation. With a charge capacity of 367 mAh g⁻¹ at the 10th cycle, A1 exhibits a

**Figure 3.** Cycling performance of the various cathode electrode formulations. (triangle) RefC, (square) C1, (circle) C2.

**Figure 4.** SEM images of the RefC (a), C1 (b), C2 (c) cathodes and the corresponding aluminum foils after removal of the coating, respectively (d–f).
Figure 5. Differential capacity curves computed from the 5th (a) and 50th (b) cycles of RefC and C1 electrodes (from Fig. 3).

slightly lower capacity (about 7 mAh g\(^{-1}\)) than RefA anode, however, both anode formulations display high average coulombic efficiencies of 99.87%. Additionally, A1 showed a small capacity loss between the 10\(^{th}\) and 50\(^{th}\) cycles (about 1 mAh g\(^{-1}\)) compared to the slight capacity increase of about 0.3 mAh g\(^{-1}\) detected for the reference graphite anode (this latter is due to the ongoing wetting of anode electrode). However, the first cycle irreversible capacities are quite comparable (RefA: 43.6 mAh g\(^{-1}\), A1: 41.8 mAh g\(^{-1}\)), suggesting a similar SEI formation process. Fig. 6b shows the 50\(^{th}\) cycle voltage profiles of the RefA and A1 anode electrodes. A slightly shortened charge plateau at approx. 0.6 V vs Li/Li\(^{+}\) is apparent for the A1 electrode, which is in line with the observations of Fig. 6a. More importantly, however, the voltage profiles of RefA and A1 anodes are largely superimposable, indicating that lithium ion (de)intercalation occurs equally for both anode formulations. Low magnification SEM micrographs of RefA and A1 electrodes are compared in Figs. 7a and 7b. A homogeneous distribution of conductive agent, active material particles and surface pores is observed for the reference anode (Fig. 7a). The A1 anode also displays a homogeneous distribution of particles and pores, but some minor defects at the surface are observable (see circles Fig. 7b), which might be the reason of the slightly lower capacity mentioned above.

Even more important, however, is the absence of the polymeric polyurethane surface layer, confirming that the formation of such a layer is not due to the presence of generic particles. Since anionic waterborne polyurethanes (like the one used in this study) generally exhibit no precipitation or coagulation in alkaline solutions, the basic pH of the cathode slurry is also not responsible for the observed active material encapsulation. As ionic water-soluble polymers (polyelectrolytes) are known to adsorb onto particles of the same and opposite surface charge we propose that the herein used anionic polyurethane strongly interacts with the metal oxide surface of NMC cathode materials and only weakly with the anode’s graphite particle surfaces. This hypothesis is supported by the already reported interaction of anionic polyurethane ionomers with titanium dioxide pigment particles. Furthermore we deduce that upon evaporation of water during the electrode drying step, film formation occurs through the coalescence process of the adsorbed polyurethane resulting in the surface layer evident from Fig. 4.

Full Li-ion cells were realized coupling C1 cathodes with A1 anodes in pouch bag cells. The electrodes’ balance was made with respect to their practical capacities at 0.1 C rate, resulting in cathode...
of electrolyte ions. It is also shown that the polyurethane encapsulates NMC active material particles during the electrode slurry making and coating, therefore greatly reducing the corrosion of the cathodic (aluminum) current collector. Increasing the amount of polyurethane binder improves the cycling stability and discharge capacity of the cathode electrodes.

Finally, the proof of concept for a Li-ion battery comprising rather thick electrodes (about 1.5 mA h cm\(^{-2}\) nominal capacity) using the 1% polyurethane / 2% CMC binder mixture for both the anode and the cathode electrodes, is demonstrated herein. Full cells, having a cathode to anode capacity ratio ranging from 1 to 1.2, were able to deliver capacity close to 120 mA h g\(^{-1}\) (of NMC) at 1 C rate with outstanding coulombic efficiency (ca. 99.9%) and extremely low capacity fading (0.059 mA h g\(^{-1}\) per cycle) upon long-term (CCCV) cycle tests.

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