LAPONITE® as an additive in lithium-ion battery electrodes

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Abstract

The synthetic clay LAPONITE-RD, manufactured by BYK, was examined as a functional additive for lithium-ion battery electrodes. LAPONITE acts as a rheology additive in the electrode coating slurry and as a lithium-ion conducting additive in the electrode. The synergistic effect of LAPONITE in combination with sodium carboxymethyl cellulose (CMC) was used to reduce the necessary amount of thickening agent to formulate stable aqueous electrode coating slurries. By using mixtures of LAPONITE/CMC instead of only CMC, a distinct reduction of thickening agent could be accomplished without sacrificing adhesion strength or electrical conductivity of the coating. Reducing the portion of thickening agent, which also acts as a binder in the electrode coating, could be used to increase the relative amount of active material in electrodes and consequently increase the electrode energy density. The electrochemical stability of LAPONITE-RD was analyzed by cyclic voltammetry (CV). The absence of additional signals in CV measurements indicated that the LAPONITE/CMC nanocomposite binder behaves identically to conventional CMC binders, and no electrochemical redox reactivity was observed. The beneficial effects of LAPONITE-RD as an additive for electrodes of lithium-ion batteries were analyzed by full-cell measurements. Electrodes containing mixtures of LAPONITE/CMC as a binder showed improved cycle life and rate capability compared to conventional electrodes.

1. Introduction

The increase in energy density and rate capability, and the improvement of the cycle life of lithium-ion batteries are major challenges that need to be solved to meet the requirements of advanced applications such as in stationary electrical energy storage, electric vehicles, or certain industrial applications. Apart from innovative active electrode materials, the use of additives – substances that are added in only small amounts to the electrodes or the electrolyte – has been proven to be an effective way to meet the aforementioned requirements of advanced lithium-ion batteries. In this study, we present the positive effect of LAPONITE, a synthetic clay mineral of high purity manufactured by BYK, as an innovative additive in the electrodes of lithium-ion cells.

LAPONITE has a layered structure and forms nanosized platelets with a diameter in the range of 25 nm and a thickness of 0.92 nm. The typical empirical formula of the synthetic hectorite clay can be given with Na+0.7[(Si8Mg5.5Li0.3)O20(OH)4]-0.7. In the form of dry powder, the negatively charged crystals are arranged in stacks which are held together electrostatically by the sharing of sodium counter ions in the interlayer region between adjacent crystals. The crystals can be separated by dispersion process in a polar solvent like water and, depending on additional polar components (e.g. particles, binders, simple salts, etc.), the LAPONITE crystals can interact with these to form complex 3D gel structures. This ability to modify the rheological properties of complex dispersion systems in combination with its high purity makes LAPONITE a prominent rheology additive in many applications like coatings or cosmetics. Furthermore, the synergistic effect between combinations of clay minerals and several organic thickening agents, such as sodium carboxymethyl cellulose (CMC), to develop a high level of gel structure is well known. LAPONITE therefore can be used to stabilize dispersions and prevent sagging of particles. It furthermore can stabilize particles or fluids in the form of Pickering emulsions.

It is known that clay minerals, such as smectite, bentonite, hectorite, montmorillonite, etc., are effective ion conductors, and they already have been described as additives to improve battery characteristics such as cycle life, rate capability, and temperature stability. The disc-shaped crystals, e.g. of LAPONITE, are either used in the electrodes or in the electrolyte to improve their characteristics, such as ion mobility. Ryu et al. reported that the electrode wettabillity (especially of the oleophilic anodes) with the polar electrolyte solvent as well as the mechanical strength of the electrode coating and cycle
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stability could be improved by addition of clay minerals to the electrodes of lithium-ion cells. Additionally, the thermal stability of these inorganic materials has been used to improve the stability and ionic conductivity of lithium-ion battery separators. However, the use of clay minerals in combination with CMC as thickening agent in aqueous electrode slurries and as a composite binder in electrodes has not been described before.

In this study, the influence of the synergistic effect between LAPONITE and CMC on the stability of electrode coating slurries is investigated. The physical properties (e.g. adhesion strength and resistance) of electrode coatings containing a LAPONITE/CMC composite binder compared to a CMC binder are analyzed. Furthermore, the electrochemical properties of LAPONITE as an additive in electrodes of lithium-ion cells are investigated by means of potentiostatic and galvanostatic measurements.

2. Results and discussion

2.1. LAPONITE/CMC in DI water

The synergistic effect on viscosity by combining clay minerals with organic co-thickeners like CMC, other cellulose ethers, xanthan, guar and other natural gums, polyurethanes, ASE and HASE polyacrylic acid polymers, etc., is well known. The effect is often used to reduce the thickener concentration, stabilize particles, and prevent sedimentation in dispersions, or to fine-tune the rheological profile of formulations.

In this study, LAPONITE-RD – which in the following will be depicted as LAPONITE – and a high purity CMC for lithium-ion battery application were used. The synergistic effect is caused by an electrostatic interaction between the carboxylic groups of the CMC, the negatively charged clay platelets and the sodium cations, which leads to the formation of a complex 3D network in water and results in a strong thickening effect, especially at low shear.

Figure 1 shows the viscosity values of pure CMC solution (100 CMC) and LAPONITE (100 LAPONITE) dispersion, as well as mixtures of those thickening agents at different CMC:LAPONITE ratios. In all systems, the overall thickening agent concentration was set to 2%. With 9739 cP, the pure CMC solution had the lowest viscosity value of all systems. A partial substitution of only 10% of the CMC by LAPONITE already gave a more than fivefold increase in viscosity to 55160 cP. The mixtures with CMC:LAPONITE ratios between 80:20 and 50:50 had comparable viscosity values in the range of 142827 to 177182 cP. The viscosity values then increased to a range of 235011 to 254122 cP for the 40:60 to 20:80 ratios, before a stepwise decrease of 173730 cP for the 10:90 ratio and 86132 cP for...
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the 2% LAPONITE dispersion was observed. With 254122 cP, the system with a 30:70 ratio showed the highest viscosity value.

![Figure 2: Viscosity values of the 2% CMC solution (green) and different thickening agent concentrations of the CMC/LAPONITE mixtures with 80:20 ratio.](image)

To further investigate the relationship between thickening agent concentration and viscosity value, an active content ladder study was carried out with the CMC:LAPONITE ratio fixed at 80:20. The green bar in Figure 2 represents the viscosity value of the 2% CMC solution with 9739 cP, and the blue bars represent the viscosity values of the CMC/LAPONITE mixtures at concentrations of 2, 1.75, 1.5, 1.25, 1, and 0.5%. A rather linear decrease of the viscosity over concentration can be observed, decreasing from 160624 cP for the 2% system to 9949 cP for the 0.5% system. This means that 0.5% of an 80:20 mixture of CMC/LAPONITE has a comparable thickening effect in DI water to a 2% CMC solution at low shear.

2.2. Aqueous graphite electrode coating slurries

To investigate the influence of the thickening agent concentration and composition (CMC vs. CMC/LAPONITE) on the rheological properties, graphite slurries with a varying amount of thickening agent and composition were prepared (see Table 4). As CMC and likewise CMC/LAPONITE mixtures act as a thickening agent in slurry formulations and further function as a binding component in the electrode coating, the nomenclature of the samples in this paper, e.g. CMC 2%, reflects the weight fraction of CMC and CMC/LAPONITE mixtures in the electrode coating.

![Figure 3: Viscosity vs. shear rate of graphite anode slurries with varying amount of CMC.](image)

In Figure 3 the viscosity vs. shear rate of the slurries with concentrations of 2, 1.5, and 1% CMC are shown. The samples demonstrated a shear thinning behavior and the expected stepwise decrease of viscosity with a decreasing amount of CMC. In Figure 4 the rheological properties of graphite slurries...
containing different amounts of CMC/LAPONITE mixtures are compared to the samples containing only CMC as a thickening agent. Based on the results of viscosity measurements of CMC/LAPONITE mixtures in the previous section, a CMC:LAPONITE ratio of 80:20 has been chosen (see Table 4). Comparing the flow behavior of the sample CMC 2% with CMC/LAPONITE 1.5% shows that, due to the synergy effect between LAPONITE and CMC at low shear (< 1 s⁻¹), the viscosity of CMC/LAPONITE 1.5% is higher than that of CMC 2%. This means that the combination of LAPONITE and CMC leads to a stronger thickening effect, even although the amount of thickening agent is lowered from 2% to 1.5%.

However, over the shear rate, the slope of the curve of CMC/LAPONITE 1.5% is more negative, and at a certain shear rate (2-3 s⁻¹), the viscosity of CMC/LAPONITE 1.5% is lower compared to CMC 2%. At higher shear (> 100s⁻¹), the viscosity of CMC/LAPONITE 1.5% is comparable to that of the sample CMC 1.5%. A similar rheology profile of higher viscosity at low shear and lower viscosity at high shear is observed for the samples CMC/LAPONITE 1% and CMC 1%. This means that the storage stability (viscosity at low shear) is being improved due to the synergistic effect between LAPONITE and CMC, although the concentration of thickening agent is lowered by 0.5%. Furthermore, no negative effects on pumpability and flowability are observed, as the high shear viscosity of the samples is comparable to the corresponding sample containing CMC at the same weight fraction.

The dependency of low shear viscosity and slurry stability becomes obvious when imaging the sedimentation of graphite in the slurries over time. Figure 5 shows a photo of the slurries after four days of storage at room temperature. The slurry CMC 1.5% showed a slight tendency towards graphite sedimentation. The behavior is reflected by the two-phase appearance of the slurry. The larger graphite particles (gray) sag over time, whereas the carbon black stays in dispersion, coloring the supernatant phase dark black. This behavior is even more pronounced in the case of slurry CMC 1%, which had the lowest low shear viscosity of all samples. A rather negligible tendency of sedimentation is observed for the slurries CMC/LAPONITE 1.5% and CMC/LAPONITE 1% respectively.
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Figure 5: Photographic image of the graphite slurries after four days storage at room temperature.

In case of slurries containing only CMC, the formation of a hard cake over time was observed, whereas the slurries containing CMC/LAPONITE could be easily stirred after several weeks of storage. Although electrode coating slurries are usually used directly, this long-term tendency indicates the high degree of stability of the slurries containing LAPONITE.

Furthermore, the physical properties of the electrode coating, such as peeling strength and specific resistance, were measured. The proper adhesion of the electrode coating to the current collector, reflected by a high peeling strength, is known to have a positive effect on the cycle life of lithium-ion cells. CMC fulfills the function of a binder in electrode coatings, as the carboxyl groups of the polymer can adhere to the metallic current collector. Lowering the amount of CMC in the electrode coating slurries therefore results in a decreasing peeling strength. The coating slurries of this study showed a decrease ranging from 32.5 N m⁻¹ for CMC 2% and 22.5 N m⁻¹ for CMC 1.5% to 10.1 N m⁻¹ for CMC 1% (see Figure 6).

Figure 6: Peeling strength of the coatings of the different samples.

Applying the composite of CMC and LAPONITE at a CMC:LAPONITE ratio of 80:20 seems to have a positive impact on the peeling strength. With 30 N m⁻¹ for CMC/LAPONITE 1.5%, the value is almost as high as for CMC 2%, although the sample contained 0.5% less of the binding / thickening component. With 11.7 N m⁻¹, the CMC/LAPONITE 1% also outperformed the comparison specimen CMC 1% with only 10.1 N m⁻¹.

The specific resistance of the coatings was analyzed by four-point probe measurements. As shown in Table 1 no difference between samples containing the same concentration of CMC and CMC/LAPONITE mixtures was observed.
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Sample | Specific resistance / Ω cm
---|---
CMC 2% | 0.15
CMC 1.5% | 0.15
CMC/LAPONITE 1.5% | 0.15
CMC 1% | 0.16
CMC/LAPONITE 1% | 0.16

Table 1: Specific resistance of electrode coatings.

The results of this section show that the synergistic effect between LAPONITE and CMC can be used to reduce the overall amount of thickening agent in the electrode slurries with a parallel increase of the active material content. No negative effect on slurry stability and physical properties of the electrode coatings was observed.

2.3. Electrochemical measurements

To evaluate the electrochemical properties of LAPONITE and analyze if there is a difference between conventional CMC binder and CMC/LAPONITE composite binder, electrochemical measurements were performed. Cyclic voltammetric (CV) measurements in half-cells against Li-foil were performed to analyze the electrochemical redox reactivity of LAPONITE. Due to the fact that CMC and LAPONITE are electrical insulators, CB was added to the electrode coating as a conductive additive.

As LAPONITE potentially can be used as an additive for anodes as well as cathodes, the potential window of 0.01 to 4.8 V (vs. Li/Li⁺) was scanned. Figure 7 shows the voltammograms recorded in the range 0.01 to 2.5 V (vs. Li/Li⁺) and Figure 8 the voltammograms recorded in the range 2 to 4.8 V (vs. Li/Li⁺). Three cycles at a scan rate of 0.05 mV s⁻¹ were measured. In both graphs, the red curves belong to the CMC and the blue curves to the CMC/LAPONITE containing electrode. The identical shape of the CV curves of CMC and CMC/LAPONITE containing electrodes means that LAPONITE is electrochemically as inert as CMC, and no redox reaction was observed. The current that was measured can be attributed to the reduction and oxidation of the organic electrolyte, also known as surface electrolyte interface (SEI) formation.
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To further evaluate the influence of LAPONITE as an additive on the electrochemical performance of electrodes, the graphite anodes with and without LAPONITE (see Table 4 Section 4.2.2) were used for electrochemical evaluation in full cells. At the selected dosage, this corresponds to a weight fraction of 0.2 to 0.3% of LAPONITE in the electrode. The anodes were cycled against commercially available NCM-111 cathodes (Custom Cells Itzehoe, Germany).

Figure 9 shows the rate performance of the cells with the different anodes (see also Table 2 in Section 3). The highest discharge capacity retention over all discharge rates (0.1, 0.2, 1, 2, and 3C) was observed for CMC/LAPONITE 1.5%, and the lowest for CMC 1%. The addition of the ion-conductive additive LAPONITE to the anodes clearly results in a distinct improvement of the electrode rate performance. This positive effect of clay mineral as an additive in lithium-ion battery electrodes was previously reported by Ryu et al. The effect is even more pronounced when comparing the discharge capacity retention values of the anodes containing the same overall amount of thickening agent at 3C.

CMC 1% without LAPONITE only showed capacity retention of 43%, and a 26% improvement is observed for CMC/LAPONITE 1% with a capacity retention of 69%. Comparing the capacity retention of CMC 1.5% without LAPONITE with CMC/LAPONITE 1.5%, an improvement of 14% from a capacity retention value of 64% to 78% is observed.

Cycling tests of the graphite anodes against commercially available NCM-111 cathodes were performed in a potential window of 3 to 4.3 V (room temperature, CCCV charge, CC discharge). Figure 10 a) (see also Table 2 in Section 3) compares the cycle stability of the cells with anodes.
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CMC 1.5%, CMC 1%, and CMC/LAPONITE 1%. The cell with anode CMC/LAPONITE 1% that contained 0.2% of LAPONITE as an additive in the anode showed the highest discharge capacity retention. It reached the end of life (EOL) of 80% relative discharge capacity retention after 280 cycles at 1C. The strongest capacity fade was observed for the cell with the anode CMC 1% reaching the EOL already after 200 cycles at 1C. For this electrode, the lowest peeling strength was observed (see Figure 6), which is known to have a strong influence on cycle performance. However, the peeling strength cannot be the only determining factor.

![Figure 10: Relative discharge capacity retention of the cells containing different amounts of thickening agent (RT, CC discharge, 1C).](image)

The cell with anode CMC 1.5%, that had a larger peeling strength than CMC/LAPONITE 1%, also showed a lower discharge capacity retention reaching the EOL after 250 cycles. This observation leads to the assumption that there are additional positive effects when LAPONITE is added to the electrode. Ryu et al. report a similar behavior for electrodes containing clay as an additive.[6] They attributed the improved cycle life to an improved wetting of the electrodes by the electrolyte when a polar clay mineral is added and consequently a more homogeneous SEI formation takes place that protects the structural integrity of the electrode during cycling. Furthermore, the formation of a nanocomposite of CMC/LAPONITE could bring improved thermal conductivity and mechanical strength to the electrode. An additional positive effect is the better ion mobility in the electrode coating (see Figure 9) leading to lower capacity losses due to lower impedance. The addition of 0.2% of LAPONITE to the electrode thereby led to a 40% increase in cycle life.

A similar result can be observed when the discharge capacity retention of the cells with anodes CMC 2%, CMC 1.5%, and CMC/LAPONITE 1.5% is compared (see Figure 10 b). Although in the case of CMC/LAPONITE 1.5% the binding component was reduced by 0.5% compared to CMC 2%, the electrode that contained 0.3% of LAPONITE showed the best cycle stability of all samples tested in this study. CMC/LAPONITE 1.5% reached the EOL after 430 cycles whereas the comparable CMC 1.5% reached it already after 235 cycles. This is an improvement of 82%. The cell with anode CMC 2% reached its EOL after 380 cycles.
3. Conclusion

LAPONITE® is an effective additive to improve electrode slurry stability due to its synergistic effect in combination with several organic co-thickeners. It furthermore shows a distinct improvement of the physical as well as electrochemical electrode properties like rate performance and cycling stability.

The synergistic effect between LAPONITE and CMC could be used to reduce the overall amount of thickening agent in the electrode slurries without a negative effect on slurry stability. Consequently, the active material content could be increased, which resulted in electrodes of higher energy density (see Table 4 in Section 4.2.2).

The addition of 0.2 to 0.3% of electrochemical inert LAPONITE to graphite anodes results in a distinct improvement of the rate performance and cycle life of lithium-ion cells with NCM-111 counter electrodes (see Table 2).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Content of LAPONITE in anode / %</th>
<th>Discharge capacity retention after 250 cycles / %</th>
<th>Discharge capacity retention at 3C / %</th>
</tr>
</thead>
<tbody>
<tr>
<td>CMC 2%</td>
<td>---</td>
<td>88</td>
<td>75</td>
</tr>
<tr>
<td>CMC 1.5%</td>
<td>---</td>
<td>80</td>
<td>64</td>
</tr>
<tr>
<td>CMC/LAPONITE 1.5%</td>
<td>0.3</td>
<td>90</td>
<td>78</td>
</tr>
<tr>
<td>CMC 1%</td>
<td>---</td>
<td>78</td>
<td>43</td>
</tr>
<tr>
<td>CMC/LAPONITE 1%</td>
<td>0.2</td>
<td>82</td>
<td>69</td>
</tr>
</tbody>
</table>

Table 2: Discharge capacity retention of the cells at 3C and after 250 cycles at 1C.

LAPONITE as an additive for electrodes appears as a promising approach to improve the manufacturing process and product performance of lithium-ion cells. The positive product properties and effects are:
- High purity (important for application in lithium-ion cells)
- Improving slurry stability due to a synergistic effect with organic co-thickeners
- Improving electrode mechanical strength
- Improving electrode wettability with electrolyte
- Improving the C-rate performance of electrodes
- Improving the cycle life of electrodes
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4. Experimental

4.1. LAPONITE/CMC in deionized (DI) water
LAPONITE-RD (BYK Additives, U.K.) and WALOCEL™ CRT 2000 PA 07 (Dow Wolff Cellulosics, Germany) were used as received. Dispensions of LAPONITE and CMC were prepared with a concentration of 2% in DI water. Different CMC:LAPONITE ratios were adjusted by combining these two dispersions in the respective ratios. The solids level thereby remained constant at 2%.

Relative efficiency at selected CMC:LAPONITE ratios was measured by accurately weighing the LAPONITE and CMC powders and then dispersing them together in DI water at e.g. 1.5%, 1.0%, and 0.5% total solids content.

The single point viscosity values were measured by using the Brookfield Helipath and T-Bar system (Brookfield Engineering Laboratories, USA) at 1.5 rpm.

4.2. Electrodes
4.2.1. Electrodes for cyclic voltammetric measurements
0.75 g of carbon black (C-NERGY™ Super C65, Imerys, Switzerland) were dispersed in 13 g of a 1.2% dispersion of CMC or CMC/LAPONITE mixture (80:20 ratio). The slurries were coated on Cu- and Al-foils by a doctor blade with a wet film thickness of 250 µm and subsequently pre-dried at 50 °C for 15 min before drying them under vacuum at 120 °C overnight. The coatings had the composition as shown in Table 3.

<table>
<thead>
<tr>
<th>Sample</th>
<th>CB</th>
<th>CMC</th>
<th>LAPONITE</th>
</tr>
</thead>
<tbody>
<tr>
<td>CMC</td>
<td>82.4%</td>
<td>17.6%</td>
<td>---</td>
</tr>
<tr>
<td>CMC/LAPONITE</td>
<td>82.4%</td>
<td>14.3%</td>
<td>3.3%</td>
</tr>
</tbody>
</table>

Table 3: Composition of electrode coating for CV measurements.

Electrode discs of 18 mm diameter were punched out of these electrode tapes, and were assembled in ECC-Std cells (EL-CELL, Germany) in an Ar-filled glovebox (GS GLOVEBOX Systemtechnik, Germany) with Li-foil as counter and reference electrode. Electrolyte Selectilyte™ LP 57 (BASF, Germany) and glass-fiber separator (EL-CELL, Germany) were used.

4.2.2. Graphite anodes
Graphite powder SMGSG1 (Hitachi Chemical, Japan) was used as active material. Carbon black (CB) was used as an electrically conductive additive (C-NERGY™ Super C65, Imerys, Switzerland), and a synthetic clay mineral (LAPONITE-RD, BYK Additives, U.K.) as an ion-conductive additive and in combination with CMC (Walocel™ CRT 2000 PA 07, Dow Wolff Cellulosics, Germany) as a thickening agent. Styrene-butadiene rubber (SBR) was used as a binder (TRD 201A, JSR Micro, Japan). In order to investigate the influence of LAPONITE-RD as an additive for electrodes, different coating slurries with and without LAPONITE-RD, and with different overall thickening agent concentration (2%, 1.5%, and 1%) were prepared. The composition of the electrode coatings of the different samples are summarized in Table 4.
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Table 4: Composition of graphite anode coatings.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Graphite</th>
<th>LAPONITE</th>
<th>CMC</th>
<th>CB</th>
<th>SBR</th>
</tr>
</thead>
<tbody>
<tr>
<td>CMC 2%</td>
<td>94%</td>
<td>---</td>
<td>2%</td>
<td>1.7%</td>
<td>2.3%</td>
</tr>
<tr>
<td>CMC 1.5%</td>
<td>94.5%</td>
<td>---</td>
<td>1.5%</td>
<td>1.7%</td>
<td>2.3%</td>
</tr>
<tr>
<td>CMC/LAPONITE 1.5%</td>
<td>94.5%</td>
<td>0.3%</td>
<td>1.2%</td>
<td>1.7%</td>
<td>2.3%</td>
</tr>
<tr>
<td>CMC 1%</td>
<td>95%</td>
<td>---</td>
<td>1%</td>
<td>1.7%</td>
<td>2.3%</td>
</tr>
<tr>
<td>CMC/LAPONITE 1%</td>
<td>95%</td>
<td>0.2%</td>
<td>0.8%</td>
<td>1.7%</td>
<td>2.3%</td>
</tr>
</tbody>
</table>

Batches of 250 g and a solid content of 50% were prepared, following the dispersion procedure of Figure 11 to receive anode coating slurries. For the dispersion process, a DISPERMAT® AE03-C1 (VMA-GETZMANN, Germany) equipped with a 500 ml stainless steel container and a dispersion impeller with tooth profile and 6 cm diameter was used. In a typical batch, the appropriate amount of 2% CMC solution, 2% LAPONITE-RD dispersion, and DI water were homogenized at 600 rpm for 1 min prior to the addition of CB. The CB was dispersed for 90 min at 2000 rpm, before the graphite powder was added and dispersed for an additional 60 min at 1000 rpm. Finally, the SBR binder was added and the slurry was homogenized for 30 min at 1000 rpm.

Electrodes were prepared by casting the slurries on a 15 µm Cu-foil using a doctor blade on an automatic laboratory coater (BYK-Gardner, Germany). The coating had a wet film thickness of 100 µm and was pre-dried at 50 °C for 15 min and subsequently dried under vacuum at 120 °C overnight. After drying, the electrodes were calendared to reach a dry film thickness of 60 µm. Electrode discs of 18 mm diameter were punched out of these electrode tapes and were assembled in ECC-Std cells (EL-CELL, Germany) in an Ar-filled glovebox (GS GLOVEBOX Systemtechnik, Germany) with commercially available NCM-111 electrode coins (Custom Cells Itzehoe, Germany) as counter electrodes. The anodes had a loading of 2.5 mAh cm⁻² at a density of 1.2 g cm⁻³. Electrolyte Selectlyte™ LP 57 (BASF, Germany) and glass-fiber separator (EL-Cell, Germany) were used.
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4.3. Characterization

The rheological properties of the electrode slurries were measured at 23 °C on an Anton Paar MCR 102 Rheometer (Anton Paar, Austria) with cone/plate configuration and a cone diameter of 2.5 cm and cone angle of 1°. The specific resistance of the electrode coatings was evaluated by four-point probe measurement on a Milli-TO 3 ohmmeter (H.-P. FISCHER ELEKTRONIK, Germany). Therefore, the slurries were coated with a doctor blade and a wet film thickness of 125 µm on non-conductive polyethylene terephthalate substrates and dried at 50 °C for 30 min prior to the measurement. The dry film thickness was ~ 70 µm, and specimens of 10 cm length x 2 cm width were cut for measurements.

Peeling strength was measured in 180° configuration similar to the procedure described in DIN EN ISO 8510-2.

Electrochemical measurements were carried out with a Bio-Logic MPG-2 potentiostat and galvanostat (BioLogic, France) at 25 °C. Cyclic voltammetric measurements were performed in half-cells with Li-metal as counter and reference electrode at a scan rate of 0.05 mV s⁻¹. For the potential window of 0.01 to 2.5 V (vs. Li/Li⁺), the coated Cu-foils, and for the potential window of 2 to 4.8 V (vs. Li/Li⁺), the coated Al-foils were used as electrodes. C-rate tests were done at room temperature between 3 and 4.3 V. Charging was performed under CCCV conditions at a constant current (CC) charge rate of 0.2C, followed by a potentiostatic CV step at 4.3 V and 0.02C as cut-off limit. Discharge was performed under CC conditions varying the rate from 0.1C, 0.2C, 1C, and 2C up to 3C. Cycle life analysis was done at room temperature. Charging was performed in CCCV mode with a CC charge rate of 1C followed by a potentiostatic CV step at 4.3 V and 0.1C as cut-off limit. Discharge was performed under CC conditions with 1C.

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