Conventional Electrolyte and Inactive Electrode Materials in Lithium-Ion Batteries: Determining Cumulative Impact of Oxidative Decomposition at High Voltage

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High-voltage electrodes based on, for example, LiNi_{0.5}Mn_{1.5}O_{4} (LNMO) active material require oxidative stability of inactive materials up to 4.95 V vs. Li | Li^+. Referring to literature, they are frequently supposed to be unstable, though conclusions are still controversial and clearly depend on the used investigation method. For example, the galvanostatic method, as a common method in battery research, points to the opposite, thus to a stability of the inactive materials, which can be derived from, for example, the high decomposition plateau at 5.56 V vs. Li | Li^+ and stable performance of the LNMO charge/discharge cycling. This work aims to unravel this apparent contradiction of the galvanostatic method with the literature by a thorough investigation of possible trace oxidation reactions in cumulative manner, that is, over many charge/discharge cycles. Indeed, the cumulated irreversive specific capacity amounts to \( \approx 10 \text{ mAh g}^{-1} \) during the initial 50 charge/discharge cycles, which is determined by imitating extreme LNMO high-voltage conditions using electrodes solely consisting of inactive materials. This can explain the ambiguities in stability interpretations of the galvanostatic method and the literature, as the respective irreversible specific capacity is obviously too low for distinct detection in conventional galvanostatic approaches and can be only detected at extreme high-voltage conditions. In this regard, the technique of chronoamperometry is shown to be an effective and proper complementary tool for electrochemical stability research in a qualitative and quantitative manner.

Introduction

Apart from using electrode materials with higher capacity and rate performance, an increase of the specific energy and power of lithium ion batteries (LIBs) can be realized by further increase of the cell voltage.\(^{[1–3]}\) The accompanied raise in redox potential at the positive electrode requires the electrochemical (anodic) stability of the state-of-the-art (SOTA) inactive materials, that is, separator,\(^{[4,5]}\) electrolyte,\(^{[6]}\) conductive carbon,\(^{[7]}\) Al current collector\(^{[8,9]}\) and binder.\(^{[10–13]}\)

The inactive materials, in particular the electrolyte, is believed to be unstable for high voltage applications, for example, when LiNi_{0.5}Mn_{1.5}O_{4} (LNMO) is used, which requires charge cut-off potentials up to 4.95 V vs. Li | Li^+.\(^{[14,15]}\) Depending on used method, different decomposition onsets are reported.

For example, according infrared spectroscopy the decomposition onset is reported to be already at 4.0 V\(^{[16]}\) while gas detection techniques, for example, differential electrochemical mass spectroscopy point to an onset of 4.2–4.3 V\(^{[17–19]}\) Quartz micro balance technique points to an onset of 4.4 V while surface analysis techniques, for example, X-ray photoemission spectroscopy to of 4.5 V.\(^{[20]}\) Even though these methods can be sensitive, it is worth noting that the results based on these methods can be complex and ambiguous, rendering distinct interpretations not always possible. For example, alterations of the cathode electrolyte interphase (CEI) can also originate from chemical, that is, aging reaction, and must not be necessarily the result of electrolyte oxidation.\(^{[21–23]}\) Also, gaseous products cannot always be distinctly assigned to electrolyte oxidation as they can result from chemical reactions, for example, CO\(_2\) from Li\(_2\)CO\(_3\) decomposition, which is a common native layer on active materials of positive electrodes and can lead to misinterpretations.\(^{[24,25]}\)

Interestingly, results from galvanostatic methods claim to the opposite, thus to the stability of inactive materials. Proper overcharge experiments on composite electrodes point towards sufficient electrochemical stability of the SOTA inactive materials up to \( \approx 5.5 \text{ V vs. Li | Li}^+ \).\(^{[26]}\) This conclusion is in line with the behavior of positive electrodes during high voltage operation at room temperature, where obvious oxidative decomposition reactions are missing.\(^{[27]}\) For instance, parasitic capacity reasoned potential plateaus as indication for electrolyte decomposition, known for, for example, graphite-based negative electrodes, are absent on positive electrodes.\(^{[28]}\) Also, the
specific capacity loss, particularly in the initial charge/discharge cycle, is only a valid indication for irreversible electrolyte decomposition for, for example, graphite-based negative electrodes.\cite{29-31} For positive electrodes, the specific capacity loss is predominantly the result of an incomplete discharge (= re-lithiation) process, thus is of a kinetic and reversible nature.\cite{2,27,32}

Clearly, SOTA inactive materials in LIB cells are more electrochemically stable than commonly believed according to galvanostatic methods, at least for the “mild” conditions typically used in lab research experiments. In this work, the apparent stability is questioned by applying more extreme conditions, that is, in cumulative manner during long-term charge/discharge cycling or during longer duration of the cell in charged state. Detection of possible trace decompositions in galvanostatic manner can explain and finally unravel the ambiguities with literature reports, which claim anodic instability of inactive materials by the use of different methods.

In order to precisely investigate the possible traces of the electrochemical decomposition of the inactive materials, electrodes without active material are used (Scheme 1) in this work. The exclusion of reversible redox reactions improves the detection accuracy and distinct assignment of the irreversible oxidation reactions. In addition, the minimized current flow in this experimental setup diminishes the growth of high surface area lithium (HSAL) on the Li-based opposite electrode through decreased amount of Li plating and diminishes the risk of interfering reactions and capacities because of (micro) short circuits through, for example, penetrating Li dendrites (Scheme 1).\cite{33-37} In this manner, this work aims to precisely determine the irreversible capacity in a cumulative way by simulating extreme application conditions of high voltage spinel LiNi_{0.5}Mn_{1.5}O_4 (LNMO) using a charge cut-off potential of 4.95 V vs. Li|Li^+ at an electrode solely consisting of inactive materials.

### Experimental Section

#### Used materials and sample preparation

The conductive carbon electrodes consisted of 90% (SuperC65, Imerys) and 10% polyvinylidene difluoride (PVdF) binder (Kynar 761, Arkema). For the electrode paste preparation, the binder was dissolved in N-methyl-pyrrolidone (NMP, Sigma-Aldrich) and mixed with conductive agent. Homogenization was achieved by dispersing at 17500 rpm for 30 min with a Dissolver Dispermat LC-2 (VMA-GETZMANN GmbH). Aluminum foil (Evonik) was selected as current collector. The electrodes had an average active mass loading of (1.01 ± 0.07) mg cm^{-2}.

The binder electrodes consisted of 100% PVdF on Al. PVdF dissolved in NMP was casted on roughened aluminum foil via a doctor blading technique. The complete procedure was repeated five times until the formation of a visible, thick PVdF film on Al. The mass loading after drying was (0.96 ± 0.42) mg cm^{-2}.

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electrodes were punched into discs (Ø = 12 mm) after cleaning with ethanol and a microfiber cloth.

Graphite electrodes consisted of 95 % graphite (KS6 L, Imerys) and 5 % sodium-carboxymethyl cellulose binder (Wolcell CRT, Dow Wolff Cellulosics) dissolved in water. The components were mixed together and then homogenized using a T25 Ultra Ultra-Turrax (IKA) at 5000 rpm for 1 h. Afterwards the electrode paste was coated on aluminum foil and dried at 70 °C for 2 h in a laboratory oven (Binder GmbH). The active mass loading was (1.04 ± 0.12) mg cm⁻².

As electrolyte, 1 M lithium hexafluorophosphate (LiPF₆) in ethylene carbonate (EC)/ethyl methyl carbonate (EMC), (1 : 1, by wt.; LP50 Selectlyte, battery grade) from BASF was used in all experiments. The cell assembly was performed in a glove box (UNILab, MBRAUN) under argon protective atmosphere with a water and oxygen content under 1 ppm.

**Galvanostatic experiments**

All experiments were carried out in a three-electrode setup (Swagelok) at 20 °C on a Maccor 4000 series battery test system (MACCOR, Inc.). LiNi₀.₅Mn₁.O₂O₂ (LNMO), LiNi₀.₅Mn₁.₅O₄O₂ (NMC622) or conductive carbon was used as working electrode (WE), whereas Li metal served as counter electrode (CE) and reference electrode (RE). The lower cut-off potential was set to 3.0 V vs. Li | Li⁺ and the upper cut-off potential is 4.30, 4.60, 4.95 and 6.00 V vs. Li | Li⁺, respectively. The initial three charge/discharge cycles were performed with a 28 mA cm⁻² and thereafter with 140 mA cm⁻². In addition, in all experiments with conductive carbon electrodes, a 5 h constant potential (CP) step at the upper cut-off potential for the formation and a 1 h long CP step for the remaining cycles was added. Both, the C-rate and the CP step were chosen to simulate the electrochemical conditions of conductive carbon in a high voltage spinel electrode (LiNi₀.₅Mn₁.₅O₂O₂).[26]

**Chronoamperometric measurements**

Chronoamperometric measurements were conducted on a VMP3 Multi-channel potentiostat (Bio-Logic Science Instruments). After a rest step at open circuit potential (OCP) conditions for 30 min, the potential was increased to the considered cut-off potential using a scan rate of 1 mV s⁻¹ and was held for 24 h.

**Results and Discussion**

State-of-the-art (SOTA) active materials for positive electrodes in lithium ion batteries (LIBs) are layered oxides (LiTMO₂ TM= Transition metal), typically LiNi₀.₅Mn₁.₅O₂ (NMC622). Increase of the charge cut-off potential, for example, from 4.3 to 4.5 V vs. Li | Li⁺, is beneficial for the specific capacity and the overall specific energy but detrimental for capacity retention and cycle life, as exemplarily shown in Figure 1a. Nevertheless, the apparent causation of electrode cut-off potential with capacity retention can be questioned, as for example an LiNi₀.₅Mn₁.₅O₂ (LNMO)-based electrode performs better in capacity retention than an NMC622-based electrode, despite an even higher charge cut-off potential (4.95 V vs. Li | Li⁺).

Clearly, the assumed oxidative decomposition of the inactive materials, for example, electrolyte with increasing electrode potentials seems not to be directly related to the performance of positive electrodes. In fact, a proper overcharge experiment on LNMO electrodes even points to a stability and indicates the onset of oxidative decompositions of inactive materials not below 5.56 V vs. Li | Li⁺, which is beyond the charge cut-off potential of LNMO, shown in Figure 1b. Directly beyond the respective charge cut-off potential of 4.95 V vs. Li | Li⁺, the LNMO characteristic specific capacity of 148 mAh g⁻¹ is not visibly exceeded, which additionally points to the absence of parasitic charge capacity and the apparent stability of the inactive materials at these conditions.

This is in line with previous reports, where the overall performance of the positive electrode primarily depends on the active material itself.[22,23,24] In the case of layered oxide-based active materials, an increased electrode potential leads to a higher Li⁺ extraction ratio (higher specific charge capacity), which progressively distorts the active material structure and decreases the cycle life. Thus, an increased electrode potential rather affects the active materials and less the inactive materials. Though, SOTA electrolyte can influence the electrode performance by affecting the CEI and the structure of the active material, but rather in chemical manner and obviously less by oxidative decomposition products.[21,22]
Although no apparent oxidative decomposition of electrolyte is observed at LNMO in one charge/discharge cycle (Figure 1), it is of practical interest to quantify any traces of irreversible specific capacity, as they can get significant and accumulate during cycling as ‘cumulative irreversible capacity’. For this reason, an electrochemical procedure is selected for conductive carbon electrodes, which is schematically depicted in Figure 2a. In addition to the galvanostatic charge and discharge steps a constant potential (CP) step for 5 h ($\approx 0.2$ C) is added to mimic storage conditions under charged state and thus accelerate possible oxidative decomposition reactions. The difference between the obtained specific charge and discharge capacities can be regarded as irreversible specific capacity. The results are shown in Figure 2c. The cut-off potentials are reached rapidly, which implies negligible electrochemical reactions, for example, anion intercalations. Large specific currents are mainly observed after reaching the cut-off conditions. As shown in Figure 2c, most specific capacity is obtained during the CP charge step yielding a total specific charge capacity of 4.3 mAh g\(^{-1}\) and even a specific discharge capacity is obtained ($\approx 0.3$ mAh g\(^{-1}\)).

The specific discharge capacity is partially reversible and may originate from partially reversible but slight anion intercalation into conductive carbon\(^{[44–46]}\) and/or reversible double layer formation at the conductive carbon.\(^{[47]}\) The irreversible specific capacity can be determined for the following charge/discharge cycles as the difference between specific charge and discharge capacities, which are shown in Figure 3a for varied charge cut-off potentials. For quantitative comparison, the amount of inactive materials in the electrode without active material corresponds to an electrode with 0.011 g active material ($\approx$ as for the experiment displayed in Figure 1).

Independent of the charge cut-off potential, the irreversible specific capacities are highest in the initial charge/discharge cycles and decrease steadily in the following cycles. This relation is more obvious in the cumulated capacity plot (Figure 3b), where the increase in cumulated irreversible specific capacities over cycling flattens, implying a stepwise passivation reaction. Still, already after 50 charge/discharge cycles, irreversible specific capacities of $\approx 10$ mAh g\(^{-1}\) can be cumulated in this extreme but application relevant conditions, demonstrating that traces of decomposition reaction in individual cycles should not be

![Figure 2](image-url)

**Figure 2.** (a) Electrochemical procedure to estimate the irreversible capacity within a potential range of 4.95–3.00 V vs. Li/Li\(^+\) using conductive-carbon electrodes. The galvanostatic charge/discharge cycle at a specific current of 28 mA g\(^{-1}\) is complemented by a constant potential step for 5 h during charge. The difference between specific charge and specific discharge capacities is regarded as irreversible specific capacity. (b) Cut-off potentials are reached rapidly, implying negligible electrochemical reactions below cut-off. (c) As expected, the major part of irreversible specific capacity is obtained during the 5 h constant potential step at the cut-off potential.
disregarded, as the oxidation reaction can, for example, release gaseous decomposition products that can accumulate in the cell over cycling. Though, the irreversible specific capacities for lower potentials, that is, 4.6 V vs. Li\textsubscript{||}Li\textsuperscript{+} (e.g., high voltage NCM), are already significantly lower, hinting to the stability of electrolyte and inactive materials.

Obviously, traces in decomposition are present but are challenging to detect in conventional galvanostatic methods. A more effective method to determine those traces in oxidative decomposition reactions in qualitative and quantitative manner can be better realized \textit{via} chronoamperometry, that is, monitoring the current flow during initial charge \textit{via} a constant potential at 4.95 V vs. Li\textsubscript{||}Li\textsuperscript{+} for 24 h. (Figure 4a). In order to separate the impacts of each inactive electrode material, also bare Al foil and binder on Al are investigated as working electrodes. The initial current peak is highest for the conductive carbon electrodes, which can be assigned to many processes simultaneously, that is, reversible capacitance build-up and irreversible specific capacity as well as passivation of the aluminum surface with AlF\textsubscript{3}.\textsuperscript{[48–50]} Also, the subsequent current baseline is significantly higher for the conductive carbon electrode (0.0029 mA) than for the binder and the Al electrode (0.0001 mA). Higher surface area of conductive carbon can realize a significantly higher amount of electrolyte decomposition,\textsuperscript{[51–53]} in this case oxidation, thus can be concluded to be the main source of the observed irreversible capacity, while the SOTA binder is, as expected, overwhelmingly stable and show no significant difference to the bare Al electrode.\textsuperscript{[54,55]} The capacity, as integrated current, is shown in Figure 4b as a function of time. In this approach, the steadily increase of capacity points to continuous electrolyte oxidation at this extreme and triggering conditions.

**Conclusions**

In contrast to several literature reports, the galvanostatic method points to an electrochemical stability of the state-of-the-art inactive materials during high voltage applications. This can be derived, for example, from the good capacity retention disregarded, as the oxidation reaction can, for example, release gaseous decomposition products that can accumulate in the cell over cycling. Though, the irreversible specific capacities for lower potentials, that is, 4.6 V vs. Li\textsubscript{||}Li\textsuperscript{+} (e.g., high voltage NCM), are already significantly lower, hinting to the stability of electrolyte and inactive materials.

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of high-voltage spinel LiNi$_2$Mn$_{1/3}$O$_4$ active material during charge/discharge cycling up to electrode potentials of 4.95 V vs. Li|Li$^+$ or by the galvanostatic overcharge experiments on composite electrodes, which demonstrate a stability even up to $\approx 5.56$ V vs. Li|Li$^+$. This apparent contradiction of the galvanostatic method to literature is unraveled in this work by a thorough evaluation of trace decompositions via determining the accumulated irreversible specific capacity over cycling. Therefore, modified electrochemical conditions on electrodes consisting solely of inactive materials are selected for reasons of accuracy. A total irreversible capacity of 10 mAh g$^{-1}$ can be obtained after only 50 charge/discharge cycles demonstrating an instability of inactive materials when extreme (LNMO) conditions are used. The amount of these trace reactions is still too low for the detection via conventional galvanostatic investigations, thus can explain the apparent contradiction to literature reports, which claim instability of inactive materials by using other, more sensitive, methods, for example, gas detection techniques. Therefore, the detection and investigation of the trace decomposition reactions is shown to be more reasonable and effective either at extremes conditions and/or by the use of the proper chronocoulometric technique. This technique demonstrates that electrolyte oxidation contributes most to the irreversible specific capacity while the residual part is overwhelmingly the result of anodic AI dissolution.

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Conflict of Interest

The authors declare no conflict of interest.

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