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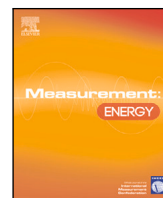
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Regular paper

Accelerated lithium-ion battery cycle lifetime testing by condition-based reference performance tests

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ABSTRACT

Lifetime testing of lithium-ion batteries is time-consuming and costly. To reduce the time-to-market, application-specific accelerated lifetime tests are conducted. The test conditions must be carefully designed and controlled, both the test environment and load profile. During the lifetime test, measurable properties of the cell, most commonly the capacity and internal resistance, are tracked by a reference performance test (RPT). The frequency of RPTs is a variable of the number of test cycles or time. Setting the wrong frequency for the RPT results in either too many RPTs or, in the worst case, too few. To mitigate this issue, a test object capacity-driven approach has been developed. This new method is described and demonstrated in this article conducting the RPTs based on the cycling capacity of the cell. The method ensured the desired numbers of RPTs during the test period at the selected intervals corresponding to steps of 1% capacity loss. When compared to the most used traditional test method, using a fixed number of 200 cycles between RPTs, the method generated 44% more cycles over the initial 100 days.

1. Introduction

Testing of lithium-ion batteries (LIBs) is crucial for evaluating their applicability and durability in various applications. These tests provide a foundation for designing a battery management system (BMS) that accurately estimates the state of charge (SOC), state of power (SOP) and state of health (SOH) during usage. However, conducting these tests on full-scale battery packs is time-consuming, costly and complex, requiring special equipment for connections to the cooling system and communication software to the BMS. As a result, tests are often conducted on single cells in a controlled environment to reduce costs and complexity.

High-quality commercial LIBs have a long cycle life expectancy of several thousand cycles. Lifetime testing under realistic conditions would thus take several years. Hence, traditional methods are designed to carefully accelerate the lifetime testing. The most common way is to separate the calendar ageing and cycle ageing. In doing so, the cycle ageing can be accelerated by removing or reducing pauses between test cycles [1]. The tests can also be accelerated by the SOC interval used [2–4], increasing or decreasing the ambient temperature [2,5] or a combination of these, further combinations are described in [6,7]. This puts a high demand on the characterisation test [8–10], test

environment [7,9,11] and the design of the test cycle so that it is representative of the expected load cycle in the application [1,11,12].

Even when accelerated, the lifetime tests can take several years and thus be very costly [1,2]. Therefore, other methods to reduce time and cost have been developed such as testing of cells on-board plug-in hybrid trucks in real field time [13], and lifetime estimations based on accurate measurements of coulombic efficiency [14]. However, these methods require specific equipment which is not standard equipment in a battery test facility [13,15].

Another trend is to utilise data-driven models for lifetime estimation [16,17]. These models require access to large amounts of data that has to be collected from usage in applications or generated from laboratory tests. It is not easy to get access to the large amount of data required and the effectiveness of the models is heavily dependent on the quality and size of the available dataset [16]. In addition, these models will be based on previous generations of LIBs. Any major change in material or cell design will likely reduce the model's accuracy.

Despite these developments, the so far best practice is to use the carefully accelerated laboratory lifetime tests, separating the calendar ageing and cycle ageing. For the cycling ageing, continuously run cycles and only stop to conduct the reference performance test (RPT).

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The RPT is designed to measure the current SOH of the battery cell [18]. By conducting the RPT throughout the cycle lifetime, the degradation history of the battery cell can be recorded [11]. The by far most common method is to use fixed periodic intervals between the RPTs, a predetermined number of cycles or time [1,7,8,19].

How to set the number of cycles between RPTs is though not always clear. The most common is to base it on the expected cycle life under normal operation, as stated by the battery manufacturers in the specification sheet, and the number of desired RPTs during the test period. Typically, a period of 50–200 cycles is used [2,11]. However, if the load cycle or conditions to be tested significantly differ from the stated normal operation it can be quite difficult to determine the periodic interval.

A consequence of using a poorly set interval between RPTs, which is not correlated to how the battery cell ages, is not reaching the desired number of RPTs. In the case where the chosen test procedure results in faster ageing than expected, fewer RPTs will be conducted over the lifetime until the End of Life (EOL) criteria is reached, and in the worst case no RPT at all. If the test procedure ages the cell slower than expected, then more than the desired number of RPTs will be performed. This will increase the overall test time needed and result in higher costs for the test [2].

In the latter case, an additional issue can arise. Depending on how the RPT is designed, it can instead be the RPTs that strongly influence the cell's ageing instead of the test cycle itself [7,8,20]. However, when conducting a carefully designed RPT with the proper frequency in relation to cell ageing, the impact from the RPTs can be kept to an acceptable level [1,7–9,11,20]. Still, using a predetermined periodic cycle interval is the dominating method used to determine how batteries age during cycle lifetime tests.

In this article, the purpose is to present an alternative method for when to conduct the RPTs. The method uses a during-cycle measurable cell property to determine when to conduct the RPT. The specific contributions of this article are:

- the demonstration of a test method for experimentally determining the cycle life of LIBs using the measured cycle capacity of the cell to determine when to conduct the RPTs.
- removing the need to estimate the expected cycle life of the LIB until the EOL criterion.
- successfully ensuring that the predetermined number of RPTs is conducted over the test period.
- adding an additional safety feature that any rapid change in the measured property (in this case cycle capacity) during the test will result in the test being stopped in order to inspect the cell or conduct an RPT.

The method was successfully applied for lifetime testing of a power-optimised cell. For the demonstration of the method, the frequency of the RPTs is desired at every 1% loss of 1C discharge cell capacity. The discharge capacity of the test cycle is used to estimate when the cell has lost 1% 1C capacity. As a redundancy, a maximum of 501 cycles between RPT is added to the program. For cases with mild ageing, the cycling test will continue until the predetermined maximum number of 501 cycles is reached before an RPT is triggered.

The outline of this article is as follows: In Section 2, the test object, test equipment, traditional test method and the proposed test method are presented. The results of the traditional test method and the proposed test method are presented in Section 3 and discussed in Section 4. Finally, the conclusions are listed in Section 5.

2. Materials and methods

2.1. Test object

The Lithium Werks ANR26650M1B 2.6 Ah (min 2.5 Ah) cell, with lithium iron phosphate (LFP) and graphite electrodes, was selected as

a test object. The cell has a cycle life expectancy of > 4000 cycles when cycled using 1C/1C (2.6 A) current and 100% depth of discharge (DOD). This is a popular cylindrical cell, commonly used in power-requiring applications and has been on the market for several years. The cell-to-cell deviation of commercial cells are expected to be low [7, 12,21]. However, even with low initial deviations the deviation will increase with ageing [22]. Due to this, two samples for each test are used. The measured initial capacity and resistance at 50%SOC for all six cells are shown in Table 1. For the tested cells, the standard deviation in initial capacity and resistance is 0.5% and 1.7%, respectively.

The selected test cycle is a constant current constant voltage (CCCV) charging and discharging cycle using the full SOC window. The CC charge rate of 6 A (or 2.3C rate) is used with CV at 3.6 V with a cut-off current of 0.26 A. The CC discharging is conducted with 6 A (or 2.3C rate) until 2.0 V. Three different frequencies of the RPTs were used and two test cells for each test variant. The different test regimes are summarised in Table 1. The EOL criteria for this study is set to when the cell's capacities have reached 80% of their initial values.

2.2. Test equipment

A Neware BTS4000 system 5V6 A tester with the BTS7.6 data acquisition software and inbuilt cell holders were used and tests were conducted at room temperature (RT). The CA-4008 temperature thermistor auxiliary equipment was used to measure the surface temperature of the cells during the tests.

The BTS7.6 software has the main functionalities needed for a battery tester and the possibility to store and compare variables during an ongoing test. However, it does not support sequencing of tests, which is an available option for many other testers. This means that it requires a manual start of the next test.

The Neware BTS4000-5V6 A series tester has an accuracy of $\pm 0.05\%$ of full scale for the voltage and current sensor. The current sensing has a triple range setup of 0.5–100 mA, 0.1–3 A and 3–6 A. The tester is thus capable of recording the expected 1% capacity loss with a maximum error of 5.2% at BOL and 6.5% at EOL. The maximum error in the Ah-counting will be 1.3 mAh ($\pm 3 \text{ mA}\Delta t$, where Δt for discharging with 2.3C maximum will be 26 min). However, as the same tester is used for all measurements, the error is expected to be much smaller.

2.3. Method

Traditional lifetime test protocols are initiated with a Beginning of Life (BOL) characterisation of the cell. The design of this BOL test depends on the cell characteristics that are to be studied over the lifetime test period and can be more or less extensive, including tests at different temperatures, C-rates, electrochemical impedance spectroscopy, etc. Typically, the BOL and EOL characterisation tests are more extensive than the RPT used throughout the test period [2,7,11]. The extensive test can also be run with a selected frequency throughout the test period if additional information during the lifetime test is desired. However, the RPT always contains capacity and resistance measurements.

For this project, the BOL and EOL characterisation has been the same as for the RPT used during cycling. The RPT is designed so that it can be conducted on the same channel as the cycling ageing. This removes a potential error when moving cells between channels, reducing the risk of introducing poor connections. The channel has a max capacity of ± 6 A. The RPT is presented in Fig. 1, containing a 1C capacity measurement, resistance measurements with 2.3C (6 A) current for 10 s at three SOC levels (30, 50 and 70% SOC), and a slow C/10 charge and discharge. The 1C discharge capacity and the 10 s pulse resistance at 50% SOC are selected for tracking the ageing trends in this article unless otherwise stated. All measurements are conducted at RT.

An example of a traditional test process is shown in Fig. 2. The procedure starts with the BOL characterisation (RPT) before the lifetime

Table 1

The test cells planned RPT frequency, initial capacity and resistance at BOL. The capacity was measured at 1C and C/10. The 10 s 6 A pulse resistance was measured at 50% SOC.

Cell id	RPT frequency	$Q_{1C,BOL}$ [Ah]	$Q_{C/10,BOL}$ [Ah]	$R_{50\%SOC,BOL}$ [mΩ]
Cell 1	50 cycle	2.487	2.518	14.59
Cell 2	50 cycle	2.493	2.520	14.37
Cell 3	200 cycle	2.505	2.540	14.22
Cell 4	200 cycle	2.514	2.542	14.12
Cell 5	1% discharge capacity loss	2.516	2.550	14.27
Cell 6	1% discharge capacity loss	2.490	2.525	14.78

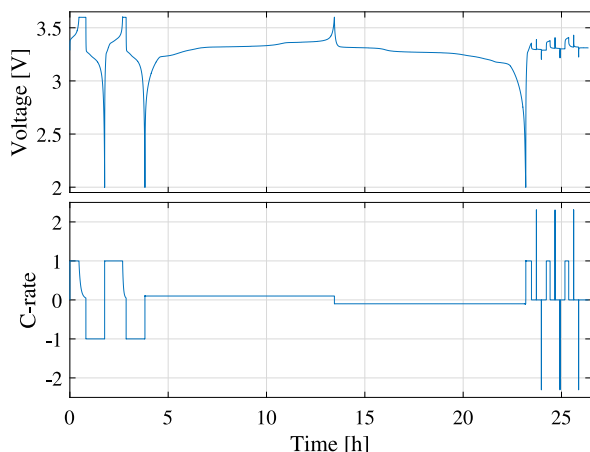


Fig. 1. The RPT used for characterisation of the M1B cells. Starting with an activation cycle and one to measure the capacity, followed by a slow charge and discharge with C/10 and finally a pulse test at three different SOC levels, 30, 50 and 70% SOC.

test of the cell is initiated. The lifetime test regime can be very different for various applications but is typically counted in the number of test cycles or equivalent full cycles (EFCs). After reaching a predetermined number of cycles, an RPT is conducted to track the SOH of the cell. The lifetime cycle regime is then restarted and the process is repeated until the EOL criteria is reached.

The method described in this article is very similar to the traditional test procedure, with the only difference being that the usable discharge capacity for the test cycle is used as a measure for when to conduct the RPT. The test procedure is shown in Fig. 2 and uses the test equipment's ability to perform coulomb counting, which most battery test equipment used today is capable of. The accuracy of the coulomb counting is proportional to the accuracy of the current sensor in the tester. Hence, test equipment with high-accuracy current sensors will have higher accuracy in the coulomb counting.

The cycling protocol, programmed in the BTS7.6 software, uses the first cycle as a reference. It records the discharge capacity and saves it in a variable in the test protocol. After the reference cycle, the program goes into the cycling phase. The discharge capacity for the cycle is recorded in a second variable. Before each new cycle is started, the recorded cycle capacity is compared to the reference cycle through an *if* condition, if $Q_{cycle}/Q_{init} < 0.99$ go to finish. If the recorded capacity is above the condition, a new cycle is started and the process is repeated until the condition is met, to record an RPT at every 1% discharge capacity loss.

The condition for when to conduct an RPT can be adjusted to any level of capacity loss, only limited by the accuracy of the test machine's Coulomb counting. Other parameters than coulomb counting can also be used to track the battery SOH. Any measurable property of the cell, such as discharge time or temperature, can be utilised. However, caution must be taken so that the chosen parameter reliably tracks the SOH or condition for which the RPT should be conducted.

The software for the tester does not support the sequencing of tests, thus each test needs to be manually started after the previous test

Table 2

Initial estimation of the number of RPTs and time required for the three test procedures. The estimated time assumes no stop time between RPTs and cycle tests. The test period is based on the time taken during the 100 test days for the cases with fixed cycles and the actual time for the proposed method.

RPT frequency	Estimated nr RPTs	Estimated time [days]	Test period [days]
50 cycle	81	240	400 (estimated)
200 cycle	21	174	228 (estimated)
1% discharge capacity loss	23	176	215

is finished. This is a typically occurring situation within laboratory testing, where a test operator needs to manually restart tests. This is sometimes desired, there might be a need to evaluate the previous test before starting the next. It can also be that the RPT demands a higher power than installed for the channel where the cycling is conducted. Then the cells need to be moved between channels manually. Moving cells between channels is not only time-consuming, but it also introduces a potential error factor when changing the connection to the cell. The new connection will not be exactly the same. Additionally moving the cells will cause wear to the fastening of the connector, potentially damaging the cell tabs. To save time and reduce this risk, the RPT was designed to be able to be run on the same channel as the cycling.

For all lifetime tests, a test duration estimation should be conducted before starting tests. The test period required for the three different procedures was estimated by assuming that the cells will have a maximum cycle life of 4000 cycles with the chosen test cycle. The test cycle is a CCCV charge and CC discharge using 2.3C and is estimated to take approximately 0.9 h and the RPT to take 26.5 h. The actual time needed for each cycle and RPT is linked to the cell capacity and impedance, both will change with the cell's ageing. Thus, assuming that the cycle and RPT time is constant throughout the test period simplifies the estimation. The cycling of 4000 cycles would take the same amount of time for all cases, 150 days, thus the difference in time required is based on the frequency of RPTs.

The number of RPTs wanted during the test period differs and depends on how many checkup points one requires during the test period. For this work a rather high resolution is wanted, around 20 RPTs. As the chosen load cycle can be considered more aggressive than the cycle used in the datasheet it can be difficult to set the interval between the RPTs, as we do not know how much more aggressive it is. Hence, two fixed intervals are selected. The first one is based on the assumption the selected load cycle will have a similar ageing as to the cycle stated in the datasheet. The second one instead assumes that the load cycle is much more aggressive. This gives intervals of 200 and 50 cycles, respectively. An estimated test duration for the lifetime test was calculated and summarised for the three test procedures in Table 2. The RPT frequency of 50 cycles was calculated to take the longest time while the proposed method and 200 cycles frequency only differed by two days, the time for the two additional RPTs needed for the proposed method.

The actual test duration will however be longer, as the BTS software does not support sequencing of tests and the laboratory only has an operator at hand during office hours in the work week. Thus, if a test is finished outside office hours, the cell would be at rest until a

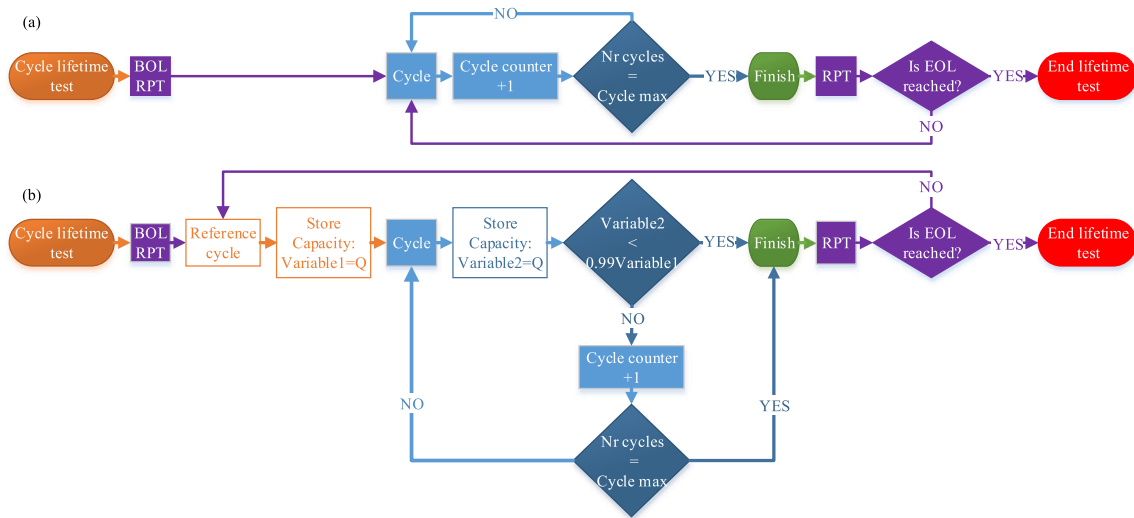


Fig. 2. Schematic overview of (a) the traditional and (b) suggested dynamic test procedure for reducing overall test time.

new test is started the following workday. Additionally, all long-term tests are likely to be subject to planned or unplanned pauses due to calibration of equipment, faulty equipment, power outage, sickness or other unforeseen incidents.

3. Results

Six cells, two for each test, were used to test the three different test procedures. The initial capacity and resistance for the cells are included in Table 1. All cells were cycled for around 100 days and the three procedures resulted in different amounts of RPTs, stand still time, operator time and different amounts of cycles per day (Fig. 3).

The repeatability between the cells is very good, which can be seen in the ageing trends over test cycles for the first 100 test days (Fig. 3(a)). All cells follow the same ageing trend as a function of cycles, with one difference. The four cells tested with a predetermined number of cycles between RPTs show a small gain in the capacity for the 200 first cycles. This is an often seen effect, especially for calendar ageing of cells at low SOC or for cells cycled with a load profile that can be considered as moderate to mild load for the cell [2,4,23,24]. This capacity increase has been attributed to a slow flow of active lithium from the passive (excess negative electrode) to the active part of the negative electrode [24]. As the proposed method is targeted to conduct an RPT after a 1% capacity loss, this behaviour is not recorded in the RPT degradation curve, however still present in the cell. The increase in capacity over the initial cycles can be seen in the 100% DOD 2.3C cycle discharge capacity in the top graph in Fig. 6(b).

During the 100 days, Cell 1 and 2 were cycled 1000 cycles in sets of 50 cycles at a time, 20 cycling and 21 RPT programs. Thus resulting in several stops and starts of test programs for the operator. The cells had an average of 10 cycles per day and had only reached 97% remaining capacity. Accordingly, this RPT frequency results in too many RPTs, i.e. an unnecessary amount of operator and test time. In addition, this amount of RPTs over the test period will start to influence ageing, resulting in less reliable test data [7,8,20].

Cells 3 and 4 ran 1600 cycles (200 cycles/program) over 97 days, with an average of 16.5 cycles per day, reaching 95% remaining capacity. During these days, the cells ran 8 cycling and 9 RPT programs, less than half the amount of test cells 1 and 2 experienced. This frequency of RPTs will, if the cells manage 4000 cycles, result in 21 RPTs and can be considered a reference case for the traditional test procedure.

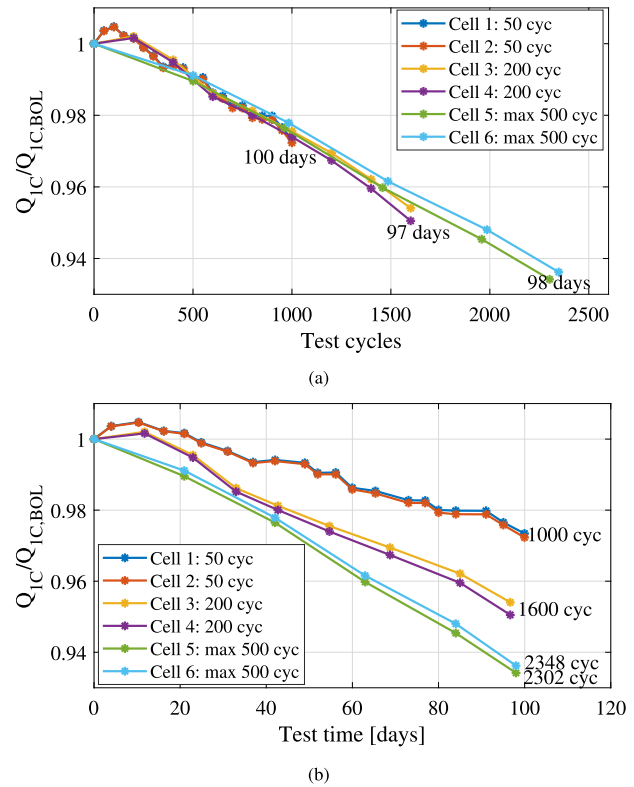


Fig. 3. Capacity degradation for the tested cells as a function of the (a) test cycles and (b) test days.

With the same assumption, cells 1 and 2 would require 81 RPTs over the test period. 21 RPTs is a more reasonable number compared to 81 RPTs, which is far too many.

The two cells tested with the proposed method, using 1% loss of cycling discharge capacity as a criterion for conducting the RPT, ran more than 2300 cycles over 99 days. The cells reached 93.5% remaining capacity, with an average of 23.5 cycles per day. They only required 5 cycling and 6 RPT programs to be started. With this method,

the planned number of RPTs is 23, which is in the same range as the estimate for cells 3 and 4.

Cells 1 and 2 had a total of 42 days of rest time, approximately a 1-day standstill between each test. Cells 3 and 4 had a total of 31 days of rest time, 6 of which were due to an unforeseen stoppage of the tester. Excluding these 6 days resulted in 1.5 days of standstill between each test. Cells 5 and 6 only had 11 days of rest time and approximately a 1-day standstill between tests. All tests had similar standstill times between tests, though with different amounts of tests conducted over the period.

The initial test time estimation was based on the possibility of sequencing tests (minimum stand-still time between tests) and thus only includes the expected cycle and RPT time. However, all tests had to be manually started when the previous test was finished, resulting in several short pauses in the testing. This is a common problem for laboratories where 24 h operator monitoring is not possible, and sequencing of tests is not supported by the software.

The cost for a lifetime test can be separated into three sections. The purchase of test objects, laboratory cost (static and dynamic), and operator cost. The largest cost for lifetime testing is, in most cases, the laboratory cost. The static cost would include the purchase of equipment and the dynamic cost is the everyday run cost and maintenance. The second largest cost is the operator time for connecting cells, programming test procedures, and collecting and processing test data. Having and running a test facility is not an option for all, thus, renting test capacity, equipment and operator time is common. For renting test capacity; the channels in a laboratory are typically rented per day (price based on channel capacity in ampere and accuracy). Hence, every extra day a cell occupies a channel is an additional cost, and the rest periods between tests can thus be costly. In addition, having a test operator manually restart or move cells between channels for RPTs also adds to the overall cost of the test. Having an optimal number of RPTs over the test period is important not only from a data analysis and reliability point of view.

However, selecting the appropriate number of cycles between RPTs can be difficult. For the tested cell type, using 50 cycles between the RPTs is clearly too frequent. From a data analysis perspective, around 20–25 RPTs to 80–70% remaining capacity is desired. Thus, using 200 cycles between the RPTs is a more optimal RPT frequency.

Based on the 100-day test period, the total required test time to reach 4000 cycles, including the stop time between tests, for cells 1–4 was calculated to take 400 and 228 days respectively. Based on the 100 first days of testing, cells 5 and 6 would have a rather short test time. However, as the number of cycles between RPTs will change with the ageing of the cell, the initial cycles per day cannot be used to estimate the total test time. Cells 5 and 6 were instead tested with the proposed test method until they reached EOL (80% remaining capacity). This took 215 days. The proposed method and using 200 cycles between RPTs have similar overall test times.

The capacity decrease and resistance change for all cells during the entire test period are included in Fig. 4. The change in resistance during the first 100 test days is rather small for all the cells and the duplicates for the three procedures follow the same pattern. Hence, the cell-to-cell variations are still very small. However, the cells do not follow the same resistance trend and further investigation into how the different test methods impact the cell ageing was made.

The differential voltage analysis (DVA) was used to evaluate if the different test methods lead to differences in ageing. After 1000 cycles, all the cells have similar capacity, 97.5% remaining capacity, and the largest deviation in the measured resistance. Despite the spread in resistance, the DVA shows only marginal deviations between cells, Fig. 5(a). The deviations between cells at BOL are similar to those seen after 1000 cycles, indicating no change in the ageing of the electrode materials between the three test cases.

For cell 5 and 6, tested with the proposed method, the DVA is included for ~5% capacity loss steps during the entire test period,

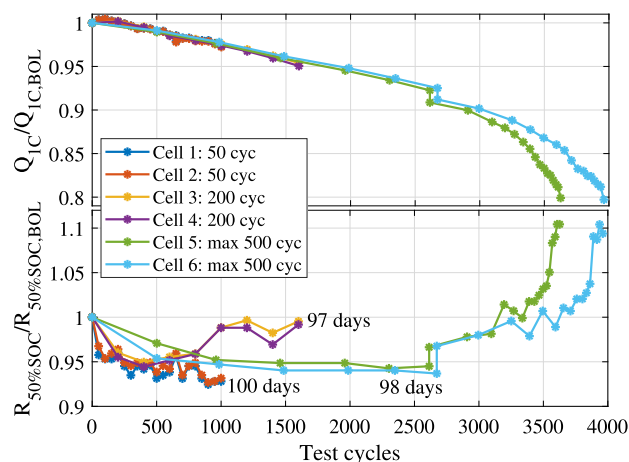


Fig. 4. All cells capacity and resistance as a function of test cycles.

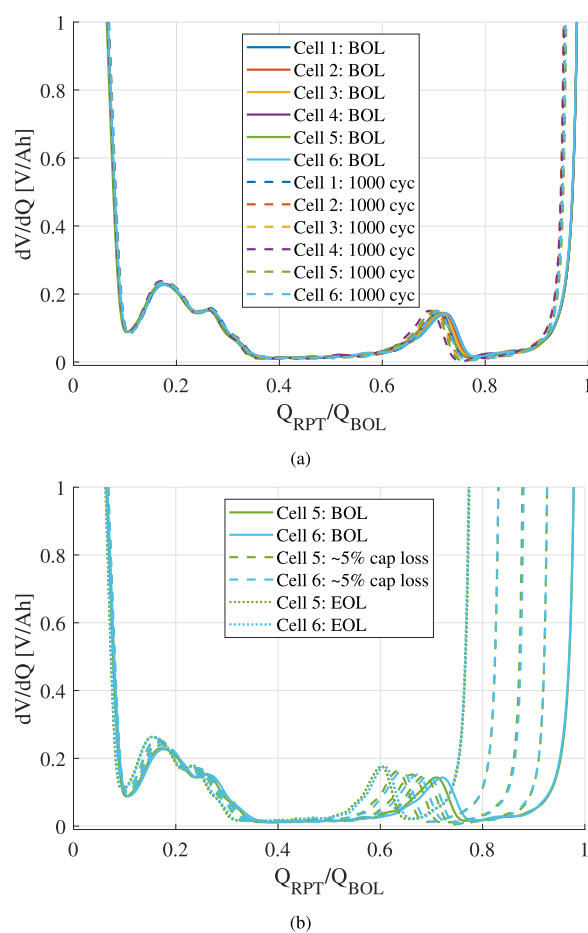


Fig. 5. DVA over the normalised capacity for (a) all cells at BOL and after 1000 cycles and (b) for cell 5 (green) and cell 6 (blue) throughout the test period at ~5% capacity loss steps. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 5(b). For the recorded capacity and resistance in Fig. 4, a vertical drop at around 2600 cycles can be seen. This is due to a rest period of 53 days that took place during the lifetime testing. An RPT was conducted before and after the rest period, i.e. two RPTs measured at the same number of test cycles. The full test history for cells 5 and 6 are visualised in Fig. 6(a) as a function of the number of test cycles. The 2.3C discharge cycle capacity is marked with dots, the measured

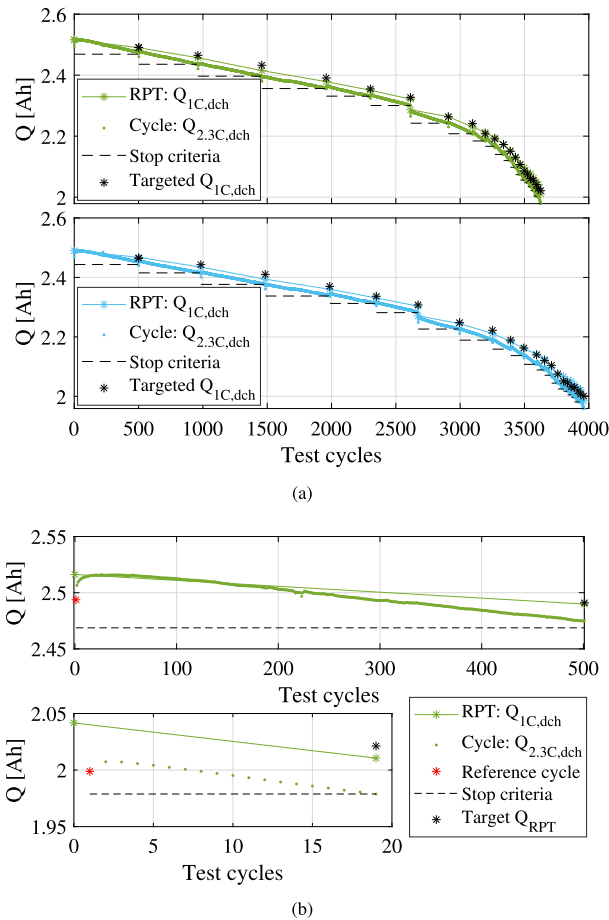


Fig. 6. (a) Capacity for cell 5 (green) and cell 6 (blue) over the cycle lifetime and (b) cycle program 1 (top) and 21 (bottom) for Cell 5. The black asterisk indicates the targeted 1C capacity value based on the previous RPT and the red asterisk is the first cycle in the cycle program used as a reference to calculate the stop criteria (black dashed line). The dots are the 2.3C capacity for each test cycle. In the first cycle batch, the program stops after 500 cycles while in the last cycle batch, the stop criteria is triggered after 19 cycles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

RPT 1C capacity ($Q_{1C,dch}$) with lines and asterisks, and the targeted $Q_{1C,dch}$ are marked with black asterisks and calculated as 99% of the previously measured $Q_{1C,dch}$. The dashed lines indicate the stop criteria for the cycling method. Further details of the full test history for cells 5 and 6 are included in [Appendix](#).

Both cells follow the same ageing trend up until the rest period of 53 days. This can be seen in [Fig. 4](#) as vertical drops in the capacity after approximately 2600 cycles. The ageing trends are still similar, however cell 5 experiences slightly faster ageing and reaches EOL after 3626 cycles. Cell 6 reaches 3961 cycles, close to the expected number of 4000 cycles.

[Fig. 6\(b\)](#) shows the first and last cycling results. The first 2.3C discharge, the red asterisk, is the reference cycle determining the stop criteria of 1% capacity loss during the cycling, the dashed line. During the first cycling test, the cell does not reach the 1% capacity loss. Instead, the cell is cycled the maximum of 501 test cycles. In the last cycling results, the cell is only cycled for 19 cycles before reaching the 1% capacity loss criteria.

As can be seen, the first measured $Q_{1C,dch}$ are slightly lower than the target $Q_{1C,dch}$. The reason for this is attributed to the nonlinear

behaviour of the lithium-ion cells. As mentioned previously the discharge capacity increases in the first cycles. Part of this is attributed to a slow flow of active lithium from the passive (excess negative electrode) and active part of the negative electrode [24]. Another part is due to the temperature increase. During cycling, the temperature increases due to the internal resistance and can also contribute to a small increase in usable capacity, as the internal resistance decreases at higher temperatures. This nonlinear behaviour results in that the stop criteria do not represent a 1% capacity loss in the cell.

However, as can be seen in [Fig. 6\(a\)](#), in most cases the method manages to stop the cycling at the set criteria. Initially, the cell is not stopped by the 1% capacity loss criteria. Instead, it is stopped at the maximum of 501 cycles. As the cell is ageing, it is stopped when reaching the 1% capacity loss stop criteria, running fewer and fewer cycles between the RPTs. This ensures that any drastic change in capacity stops the cycling, and the ageing can be recorded by an RPT.

4. Discussion

The proposed method successfully reached the 23 desired RPTs and required 215 test days for the cells to reach EOL. The initial estimation, not considering any stop time between tests or reduced time of tests due to reduced capacity, was 176 required test days. The actual active test days were 147 days for cell 5 and 156 days for cell 6. Hence, the cells had a total of 68 and 59 days of stop time between tests and an average of 1.5 days rest between the start of a new test.

In this method demonstration, the selected test cycle (2.3C/2.3C at RT) can be assumed to have a similar, or slightly shorter, cycle life as expected for the 1C/1C 100% DOD cycle stated in the specification sheet. Thus, the assumption of a 4000 test cycle life proved to be reasonable and for traditional testing 200 cycles between RPTs is a good frequency of RPTs.

No difference could be seen in the capacity loss or the DVA of the cells after 1000 cycles of testing with the three different approaches. Only in the measured resistance could deviations be seen. This could be a result of the difference in calendar ageing and pauses the methods introduced. It should be noted that the resistance changes were small in the first 100 days of testing.

The demonstrated method ensures that the desired number of RPTs is reached during the test period, as for the case of cell 5 which reached EOL earlier than expected after 3626 cycles. If this had been tested traditionally using 200 cycles between the RPTs, it would have resulted in 19 RPTs instead of the desired 21 RPTs. Hence, the proposed method ensures that RPTs are recorded even when the ageing is expectantly or unexpectedly accelerated. This makes the method especially suitable to be used for aggressive lifetime testing or when the load cycle's aggressiveness is unknown. If the user pattern in an application significantly differs from the cycle profile used in defining the cycle life in the cell specification sheet, it can be difficult to estimate the expected cycle life, and the optimal number of cycles between RPTs.

In this case, there is no major time saved between the proposed method and using 200 cycles between RPTs. Instead, the test would finish in a shorter time than estimated. However, in the case of 50 cycles between RPTs, the required test time is reduced by half using the proposed method. Thus, the proposed method can drastically reduce the required test time in cases with unexpectedly mild ageing, such as described in [25].

Another strength of the proposed method is that results are generated faster at the beginning of the cycling period, with more cycles between RPTs. Under the first 100 test days, the proposed method manages to run 44% more cycles compared to the reference case of 200 cycles between RPTs and more than double compared to the case with 50 cycles between RPTs. However, the method takes more time at the end of the cycling period, due to fewer cycles between RPTs. Similar to the maximum allowed number of cycles between RPTs, it is possible to add a minimum number of cycles between RPTs. This would reduce

the stops in the accelerated ageing phase (passed the knee); however, it would then not record the 1% discharge capacity steps.

The overall test time can also be reduced by increasing the degree of automation in the test process, removing the amount of standstill time where the cell will experience unwanted calendar ageing. Software supporting sequencing of tests is one way to significantly decrease the overall test time and, in combination with the proposed test method, generate results faster.

The full test history is included in Appendix and in Table A.2 are all stop criteria and actual stop capacities included. The tester manages to stop the cycling test at the expected stop criteria, only deviating in the third decimal for the recorded Ah-counting, except when the maximum of 501 cycles has been tested. Indicating that the error in the Ah-counting is much smaller than the specified maximum of ± 1.3 mAh.

Even if the cycling is stopped at the expected capacities, the step size between the recorded 1C capacity by the RPTs show that the steps are not exactly 1% discharge capacity loss, see Table A.2. One cause for this could be due to the nonlinear behaviour of the lithium-ion cells during cycling due to heating, full activation of the electrode materials, and potential flow of active lithium from the excess negative electrode. As can be seen in Fig. 6(b), during the first cycling, the cell gains capacity for several cycles and does not reach the 1% capacity loss criteria. However, when it is stopped after 501 cycles the capacity loss is almost exactly 1%. Thus, due to the non-linearity in the cell discharge capacity, the maximum number of cycles between RPTs was important for ensuring a recording of the cell SOH with a maximum of 501 cycles between RPTs. The maximum number of cycles is a selectable feature that ensures a minimum frequency of RPTs. Depending on the selected property used to monitor the cell during cycling, its linear or non-linear behaviour, it can be removed or adjusted. It does not necessarily require to be cycles, time or current throughput can also be used for this feature.

As can be seen for the last recorded cycling, Fig. 6(b), the aged cells show a much smaller increase during the cycling and only for the first two cycles. When the cell reaches the stop criteria, the cell has lost only 10 mAh more than the targeted capacity for the final RPT. In Table A.2 the expected and recorded capacity at each RPT and stop criteria for the cycling are included. The difference between the expected and measured capacity is more prominent at the beginning of the testing and reduces as the cell ages.

Today, commercial battery cells range from a few ampere-hours to hundreds of ampere-hours. The proposed test method is applicable to all sizes. One shall note that the smaller the cells, the larger the measurement accuracy is needed in order to be able to accurately record the 1% capacity loss during cycling. This must be taken into account when selecting test equipment for the tests. However, the cell capacity and expected user patterns often drive the selection of test equipment.

5. Conclusions

The proposed method of using a measurable property of the cell during cycling has four strengths compared to the traditional lifetime cycling testing of LIBs.

1. It generates results faster at the beginning of the test period with less operator time. Over the 100 first days, 44% more cycles could be generated.
2. It secures that the predetermined number of RPTs is achieved during the lifetime test period, with a selected step size.
3. Using the cell's discharge cycle capacity ensures that any sudden decrease in capacity is recorded and the test stopped for conducting an RPT.
4. The method is flexible to be adjusted to any measurable property or property of the cell, and the step size between RPTs is easily adjustable during the test period.

Table A.1

The initially planned RPTs at the expected remaining capacity values corresponding to 1% discharge capacity loss.

Number of RPTs	Expected $Q_{1C}/Q_{1C,BOL}$
RPT1	1
RPT2	0.990
RPT3	0.980
RPT4	0.970
RPT5	0.961
RPT6	0.951
RPT7	0.941
RPT8	0.932
RPT9	0.923
RPT10	0.914
RPT11	0.904
RPT12	0.895
RPT13	0.886
RPT14	0.878
RPT15	0.869
RPT16	0.860
RPT17	0.851
RPT18	0.843
RPT19	0.835
RPT20	0.826
RPT21	0.818
RPT22	0.810
RPT23	0.802

CRediT authorship contribution statement

Evelina Wikner: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Resources, Project administration, Investigation, Funding acquisition, Formal analysis, Data curation. **Johan Lesser:** Writing – review & editing, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix

This appendix includes selected data for the six tested cells. The C/10 capacity recorded by the RPT at BOL, and after approximately 100 days are included for all cells in Fig. A.1. For cell 5 and Cell 6 also the C/10 recorded by the RPT at EOL are included.

The proposed method had 23 planned RPT at 1% discharge capacity loss that was expected to be conducted at the remaining capacities as noted in Table A.1. Fig. A.2 includes the C/10 cycles from the RPTs during the entire test period, where the blue curve is recorded at BOL and the red EOL.

As always in experimental work unforeseen events happen. Cell 5 and 6, tested with the proposed method, had a rest period of 53 days in the middle of the lifetime testing. RPT7 (measured before the rest period) and RPT8 (measured after the rest period) are measured at the same number of test cycles. This is clearly seen as a vertical drop at around 2600 cycles in Fig. 4.

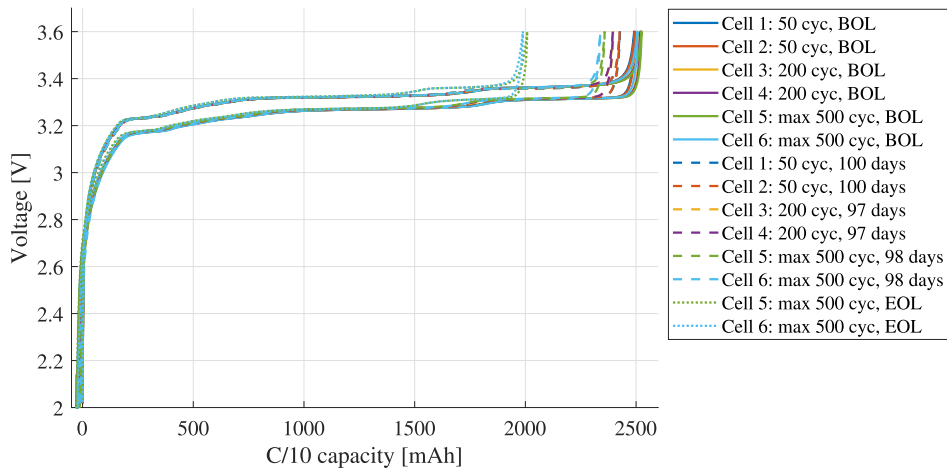


Fig. A.1. C/10 capacity for BOL, after approximately 100 days and EOL for cell 5 and cell 6.

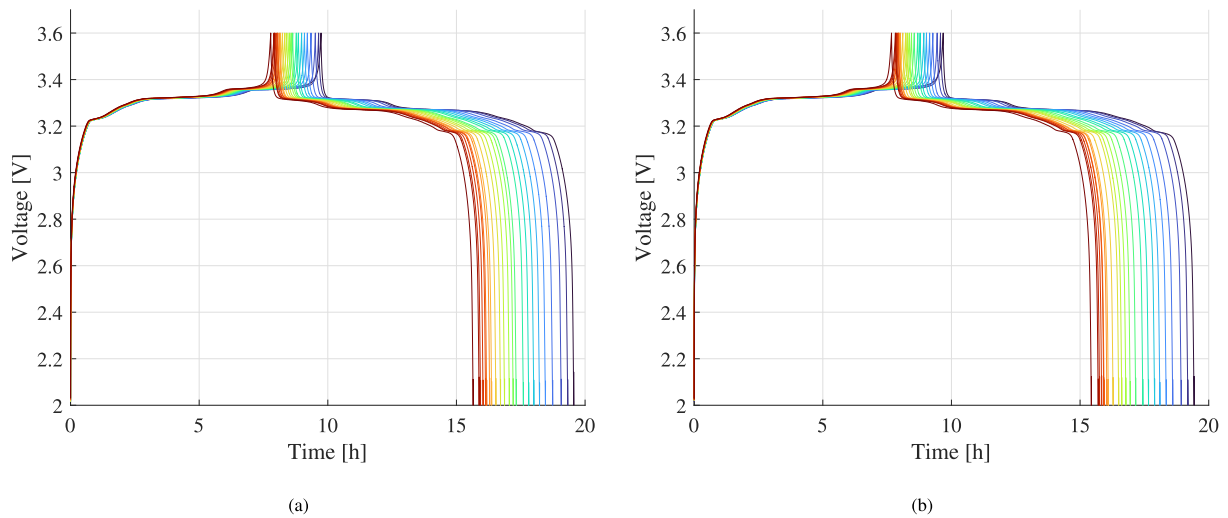


Fig. A.2. Cell C/10 capacity evolution for (a) cell 5 and (b) cell 6 over the cycle lifetime, from BOL (blue) to EOL (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table A.2

Comparison of the planned RPTs and the resulting RPTs during the test period. Including the test program stop criteria, actual stop capacity values, and how this correlates to the cell capacity.

Cell 5							
Planned RPT $Q_{1C}/Q_{1C,BOL}$	Expected Q_{1C} [Ah]	Measured RPT $Q_{1C}/Q_{1C,BOL}$	Measured RPT Q_{1C} [Ah]	Stop criteria $Q_{2,3C}$ [Ah]	Stopped $Q_{2,3C}$ [Ah]	Cycles between RPT	Test duration [Days]
RPT1 1	2.516	RPT1 1.000	2.516	–	–	–	0
RPT2 0.990	2.491	RPT2 0.990	2.490	2.469	2.475	501	21
RPT3 0.980	2.466	RPT3 0.977	2.457	2.436	2.436	460	42
RPT4 0.970	2.441	RPT4 0.960	2.415	2.397	2.396	497	63
RPT5 0.961	2.417	RPT5 0.945	2.379	2.356	2.358	501	84
RPT6 0.951	2.393	RPT6 0.934	2.351	2.331	2.331	343	98
RPT7 0.941	2.369	RPT7 0.923	2.321	2.301	2.300	312	112
Adjustment of Planned RPT due to pause in testing; Calendar ageing at RT for 53 days							
RPT8 0.909	2.287	RPT8 0.909	2.287	–	–	–	165
RPT9 0.900	2.264	RPT9 0.900	2.264	2.242	2.242	294	179
RPT10 0.891	2.241	RPT10 0.886	2.230	2.208	2.208	188	189
RPT11 0.882	2.219	RPT11 0.880	2.213	2.184	2.184	99	197
RPT12 0.873	2.196	RPT12 0.872	2.195	2.167	2.166	74	203
RPT13 0.864	2.174	RPT13 0.863	2.172	2.140	2.140	68	210
RPT14 0.856	2.153	RPT14 0.855	2.152	2.120	2.119	52	217
RPT15 0.847	2.131	RPT15 0.846	2.128	2.098	2.097	41	221
RPT16 0.839	2.110	RPT16 0.837	2.107	2.074	2.073	36	226
RPT17 0.830	2.089	RPT17 0.833	2.096	2.056	2.056	32	231
RPT18 0.822	2.068	RPT18 0.827	2.082	2.047	2.047	26	234
RPT19 0.814	2.047	RPT19 0.825	2.076	2.034	2.033	23	240
RPT20 0.806	2.027	RPT20 0.819	2.062	2.022	2.021	22	246
RPT21 0.797	2.006	RPT21 0.814	2.049	2.004	2.003	22	254
–	–	RPT22 0.811	2.042	2.002	2.001	16	260
–	–	RPT23 0.799	2.011	1.979	1.979	19	268
Cell 6							
Planned RPT $Q_{1C}/Q_{1C,BOL}$	Expected Q_{1C} [Ah]	Measured RPT $Q_{1C}/Q_{1C,BOL}$	Measured RPT Q_{1C} [Ah]	Stop criteria $Q_{2,3C}$ [Ah]	Stopped $Q_{2,3C}$ [Ah]	Cycles between RPT	Test duration [Days]
RPT1 1	2.490	RPT1 1	2.490	–	–	–	0
RPT2 0.990	2.465	RPT2 0.991	2.468	2.443	2.452	501	21
RPT3 0.980	2.440	RPT3 0.978	2.435	2.415	2.415	483	42
RPT4 0.970	2.416	RPT4 0.962	2.394	2.377	2.377	501	63
RPT5 0.961	2.391	RPT5 0.948	2.360	2.337	2.340	501	84
RPT6 0.951	2.368	RPT6 0.936	2.331	2.312	2.312	362	98
RPT7 0.941	2.344	RPT7 0.925	2.303	2.282	2.281	325	112
Adjustment of Planned RPT due to pause in testing; Calendar ageing at RT for 53 days							
RPT8 0.912	2.299	RPT8 0.912	2.271	–	–	–	165
RPT9 0.903	2.248	RPT9 0.902	2.245	2.226	2.226	322	179
RPT10 0.894	2.226	RPT10 0.888	2.211	2.189	2.189	256	191
RPT11 0.885	2.203	RPT11 0.877	2.185	2.159	2.158	141	198
RPT12 0.876	2.181	RPT12 0.868	2.161	2.137	2.137	103	203
RPT13 0.867	2.159	RPT13 0.860	2.142	2.108	2.108	99	210
RPT14 0.859	2.138	RPT14 0.854	2.125	2.089	2.089	62	217
RPT15 0.850	2.116	RPT15 0.842	2.097	2.071	2.071	54	221
RPT16 0.842	2.095	RPT16 0.832	2.073	2.044	2.044	49	226
RPT17 0.833	2.074	RPT17 0.830	2.066	2.025	2.025	44	231
RPT18 0.825	2.054	RPT18 0.824	2.053	2.017	2.017	31	234
RPT19 0.817	2.033	RPT19 0.824	2.051	2.005	2.005	28	240
RPT20 0.808	2.013	RPT20 0.819	2.038	1.997	1.996	26	246
RPT21 0.802	1.993	RPT21 0.815	2.028	1.981	1.981	27	254
RPT22 0.792	1.973	RPT22 0.812	2.021	1.981	1.980	21	260
–	–	RPT23 0.797	1.985	1.959	1.959	25	268

Due to this rest period, the planned RPTs had to be adjusted in accordance with the cell capacity measured after the rest period, RPT8. As both cells lost more than 1% during the rest period one RPT less was needed during the lifetime testing, though as RPT8 was needed for adjusting the new plan for the RPTs the total number of 23 RPTs ended up being the same.

For cells 5 and 6 additional information from both the cycling tests and RPTs are included in Table A.2. Included are the expected and recorded capacity value during RPT, the stop criteria for the cycling test, the cycle capacity for when the cycling was stopped and the number of cycles run between each RPT.

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