Modification of Degradation Mechanism Identification Technique for Cell Grading

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Abstract— **The service life of the retired automotive lithium-ion battery (LIB) can be extended through reusing without major changes and reusing through remanufacturing. Reusing through remanufacturing at the module level likely outweigh remanufacturing at the cell level and reusing without major changes. To remanufacture a battery pack, cells/modules are required to be graded and cells/modules in a remanufactured pack needs to be at identical state. The current approach utilises cell/module capacity i.e. State of Health (SoH), to grade the cells/modules at the end of first life. However, the only capacity may not be enough to match the cells/modules; how the cells/modules have been degraded (degradation mechanism, DM) may play a vital role in defining their performance in second life. However, the existing DM detection techniques are not designed to grade the cells/modules at the end of the first life. In this paper, a modified DM detection process is proposed. Here, the proposed modified DM detection process is verified by applying them to the degradation experimental results. Employing the proposed modified DM detection method, cells at same SoH at the end of first life can be graded based on their DM.**

Keywords— Lithium-ion battery, Incremental Capacity, Degradation Mechanism, End of Life, Cell grading.

I. INTRODUCTION

The demand for Electric Vehicles (EV) is increasing rapidly. The number of EVs in Europe has risen to around 300 thousand in 2019 from 97 thousand in 2017 [1]. It is forecasted that 20 million EVs will be on the road in Europe by 2030 and more than 100 million by 2040 [2]. A small number of EVs start to reach the End-of-Life (EoL) recently, which is expected to rise to a significant number in the early 2030s. Since the number of EoL EVs is increasing, many EV batteries will retire from their first life in automotive application. Hence, well-planned management of these retired batteries is required. Giving a 2nd life to the EoL EV batteries is getting progressively attractive as well-known original equipment manufacturers (OEMs) such as Tesla, Nissan-Renault, and BMW are patronising the idea [3, 4].

The EoL LIB market size is growing; however, there are no well-established protocols and strategies for the EoL LIBs [5]. There are several EoL options, such as recycling [6], reusing without major changes [7] and reusing through remanufacturing [8].

Recycling is a process where batteries are safely disassembled to extract precious metals, materials and other bi-products [9]. These extracted elements can be used to manufacture new cells and in other applications. In fact, according to EU legislation, 70% of LIB needs to be recycled by 2030, and 70% (lithium) to 95% (copper/cobalt/nickle) metal needs to be recovered [10]. Also this legislation state, by 2030 all new LIB need to have 4% recycled lithium, 4% recycled nickle and 12% recycled cobalt [10]. However, to reduce carbon footprint and maximise revenue, before the EoL automotive batteries are recycled, they should be reused in a second life application as long as it is efficient to do so.

Reusing an EoL automotive battery pack without major changes to the battery pack requires testing for degradation and failure measurement, the addition of electrical hardware, control systems, and safety systems [11, 12]. Moreover, to allow reusing without major changes, till EoL from automotive application, the battery pack needs to be well maintained and all cells/modules degraded in a similar fashion. Nevertheless, for automotive applications, the temperature gradient within a battery pack exists [13], and the pack capacity is defined by the weakest cell/module [14]. Therefore, it is likely that there will be a gradient in module/cell degradation. This degradation gradient will remain and widen in second life application. Hence, when reusing without major change, battery-pack may prematurely reach the end of second life (EoL2), therefore, not an ideal way to extend service life of modules/cells.

Reusing EoL automotive battery packs through remanufacturing is widely accepted due to its economic [9] and environmental benefits. In general, "reusing through remanufacturing" can be performed in two ways 1) dismantle down to the cell and remanufacture (Option 1) and 2) dismantle down to module and remanufacture (Option 2) [15].

Option 1 can produce most efficient battery packs with the longest possible 2nd life. However, dismantling down to cell

and remanufacture from the cell requires too much time, effort, and previously highlighted not viable commercially [9]. Moreover, current automotive battery modules are not designed for remanufacturing at the cell level; welding [16] and gules [9] are commonly used for assembling the cells in a module. If modules are dismantled into the cell, there is a high chance of disfigurement of the cells [9]. Thus, it is not recommended as a feasible way of remanufacturing.

Option 2 was first proposed by Cready *et al.* [12]. At present, it is expected that this is the most feasible way of remanufacturing. In current automotive battery packs, modules are connected to busbar using nuts and bolts or similar devices, rather than welding or adhesives; favourable for remanufacturing activity [17]. There are several advantages of working with modules in comparison with packs. If faulty modules are found by dismantling down the pack to the module level, only faulty modules can be sent for repair or recycling. This process will reduce the relocation transportation cost [18]. Also, working with lower voltage modules requires lesser safety precautions in comparison with high voltage packs [18].

In a remanufactured battery pack, the electrical performance of the cell/module needs to be precisely at the same state to avoid the pack imbalance and degradation gradient in 2ndlife. According to current practice, capacity estimation or State of Health (SoH) ensures that all cells/modules electrical performance is in the same state. As batteries are being used, unintended, irreversible internal chemical reaction occurs, which causes battery capacity drop and resistance increase, which defines the SoH. Equation 1 and 2 shows the most common way of defining SoH.

$$SoH_c = C_{actual}/C_{nominal} \qquad (1)$$

$$SoH_r = R_{actual}/R_{nominal} \qquad (2)$$

In an application, after reaching under the predefined threshold SoH value, the battery becomes unable to store and deliver energy efficiently, requiring replacement. In automotive, under warranty, battery-pack retired and replaced when reached to 80-66% SoH, depending on manufacturer [19]; this is also known as EoL point of LIB. Yet, outside warranty, the owner decides when to retire and replace the battery pack.

However, the current approach of using SoH may partly valid to match the cell/module precisely. The reason is, it was previously highlighted in [20] and [21] that only SoH is not enough to get a conclusive result on the present state of the battery. The service life (i.e. degradation) in 2nd life application could be related to how modules/cells are degraded in 1st life application, known as the Degradation Mechanism (DM); however, it is not proven yet. Previously Barai *et al.* [20] and Dubarry *et al.* [21] raise the concern whether DM identification is required while grading the cells/modules for remanufacturing. To get this answer, DM of the cells/modules should be identified first. In tandem with SoH, DM should be utilised to determine the similar state of the cell/module for remanufacturing.

Degradation of the Li-ion cell happens due to different electrochemical processes which are dependent and accelerated by many factors, e.g., temperature, pressure, state of charge, discharge current, etc. [22] [23]. It is challenging to relate all the causes and determine a specific reason for degradation due to close coupling and interdependency between degradation modes. Thus, to simplify the understanding of the degradation mechanism, Dubarry *et al.* [24], Birkl *et al.* [25], Marongiu *et al.* [26] proposed to categorise them into three different groups: loss of active material (LAM), loss of lithium inventory (LLI) and conductivity loss (CL). The degradation that occurred for the decomposition of the binder or corrosion of the current collector is related to the CL [23]. Within the cell, electrical charge is stored and delivered by lithium-ions, which intercalate and de-intercalate from electrodes during the charge/discharge process. LLI is defined by the loss of these lithium-ions [23]. Structural transformations in the positive electrode and negative electrode elements allow lithium intercalation/de-intercalation, i.e., active materials. Loss of the active material and electrolyte elements mainly defined by LAM [23]. Therefore the DM can be defined by the LAM, LLI and CL. The relationship between degradation causes and the most pertinent DM was developed by Uddin *et al.* [22] and Vetter *et al.* [23].

For DM identification (c.f Section III.B), Incremental Capacity (IC) and Differential Voltage (DV) curves are widely used ([20] and [27]). The information which will be obtained from the IC curve should not be different from the DV curve as both are derived from a typical voltage (V) vs. capacity (Q) curve. From a typical IC curve, DM is identified from the peak changes, while from the DV curve, it is determined from the plateaus. It's easier to get the information from a peak area instead of a flat region. Thus IC curves offer more traceability than DV curves. More details of the IC-DV curve can be found in [20],[28],[29], [21] and [30].

In general, to identify the DM's growth, it is necessary to track the changes of DM from a new cell to aged cells. This variation of DM can be determined from the IC curve, observing the shifting and decreasing peaks (c.f. section III.B.1)). The shifting and decreasing of the IC curves can be monitored by plotting the IC curve of the new and aged cells in a single graph. In the experimental assessment, cells are aged by cycling, and the characterisation test tracks their performance. The data from the characterisation test are used to plot the IC curve. A number of IC curves are plotted in a single graph from several characterisation test data. When several IC curves are plotted in a single graph, it is challenging to detect the shifting or decreasing of the peaks only by visual observation. As DM identification is dependent on the shifting and decreasing of the peaks, if shifting and decreasing the peaks cannot be identified precisely, it will not be possible to correctly identify the DM of the cells. In addition, DM identification at the module level currently does not exist. Therefore, issues

regarding DM identification at the cell level are discussed and analysed here.

For this study, IC peaks for several cells are plotted from the degradation experiment results produced by a team of researchers at WMG, the University of Warwick. It is noticed all the peaks are shifting by a few millivolts (c.f. Section III.B). There are rarely any peaks that can be found, which are precisely at the same point. Employing the current DM detection technique, it is not possible to detect whether the cells are at the same state or a different state. Therefore, it may require selecting a threshold point (currently not mentioned in the existing technique) to detect the shifting and decreasing among the peaks, which can eventually detect whether DM's are at the same state or a different state.

This paper aims to propose a modified DM detection technique (by indicating a threshold point) for cell grading. Here, the proposed process is applied to degradation experiment results produced by a team of researchers at WMG, the University of Warwick.

II. EXPERIMENTAL DETAILS

In this experiment, 5Ah 21700 Li-ion cells were used. The safe operating voltage of the cell is 4.2 – 2.5 V, and the maximum constant current charge-discharge rates are 0.7C and 1.5C consecutively. The overall ageing experiment process is divided into two phases: the constant cycling ageing and reference performance test (RPT). In constant current cycling, cells were repeatedly charged-discharged using different charge-discharge current values (details in Section II. A). In a regular interval of constant current cycling, cells performance were measured using a set of test, referred as reference performance test (RPT), detailed in Section II. B. During both cycling and RPT tests, cells were immersed in a liquid cooling bath, and a constant temperature was maintained. All charge-discharge were performed using a commercial Digatron battery cycler.

A. Constant Cycling Ageing:

In this step, cells were charged and discharged using the constant current - constant voltage (CC-CV) protocol at 10°C with five different charge-discharge C-rates as outlined in Table 1. At the CC phase, the cells are charged with the respective C-rate (i.e., 0.5C/0.7C/0.3C) for each group until 4.2 V and then at CV phase until charge current drops below C/20 (0.25A). The discharging process consists of discharging the cell with respective C-rate (i.e. 0.3C/1C/2C) for each group until cell voltage drop to or below 2.5 V. 1 hour rest period is used here between charge and discharge step. For each test conditions, a group of 4 cells were used to achieve statistical variance.

B. Reference Performance Test:

In this experiment, RPT test consisted of a capacity test, a pseudo-OCV (p-OCV) test and a pulse power test. The tests were performed at 25°C. At the beginning of the RPT test, the temperature of the liquid cooling bath was changed from cycling temperature to 25°C and soaked for a couple of hours. In the capacity test, cells were charged first

following CC-CV protocol, with C/3 current for CC phase to 4.2 V and C/20 cut-off current at CV phase, then discharged using C/3 to 2.5 V. This process was repeated with C/3 discharge being replaced by C/10. Therefore discharge capacity was measured at C/3 and C/10.

For p-OCV test, cells were discharged initially to 2.5 V, then charged using CC to 4.2 V (no CV phase) and finally discharged to 2.5 V. All charge-discharge were done using C/20 current. Here, data from the C/20 discharge step is used to derive the incremental capacity (IC) curve. Pulse power test (PPT) were performed using 10 sec charge-discharge pulses at 95 %, 80 %, 50 %, 20 % and 5 % SoC. However, PPT data will not be analysed in this article.

III. RESULTS AND DISCUSSION

In this section, modified DM detection process is described. The current DM detection technique is not designed to grade the cells after their 1st EoL. Therefore, current DM detection technique is modified here to grade the cells for remanufacturing.C-rate. Thus for this modification process, authors employed cells with similar SoH (±1%). The SoH measurement and DM detection method are discussed here in the next two subsections.

A. SoH Measurement

Equation (1) is used here to estimate the SoH of the cells. The measured SoHs of the cells are provided in Table 1.

Table 1: SoH of the Cells used in this study.

Cell No.	SoH%	Charge_Discharge rates	Cycle
Cell33	90.67	0.3C_0.3C	713
Cell34	90.08	,,	,,
Cell35	90.28	,,	,,
Cell36	90.59	,,	,,
Cell 37	**85.97**	**0.5C_0.3C**	**235**
Cell 38	85.11	,,	,,
Cell 39	73.8	,,	,,
Cell 40	72.87	,,	,,
Cell 41	64.40	0.7C_0.3C	190
Cell 42	64.42	,,	,,
Cell 43	error	,,	,,
Cell 44	64.78	,,	,,
Cell 45	**85.91**	**0.3C_1C**	**600**
Cell 46	86.78	,,	,,
Cell 47	87.10	,,	,,
Cell 48	87.08	,,	,,
Cell 49	**86.28**	**0.3C_2C**	**2600**
Cell 50	**85.99**	**,,**	
Cell 51	**85.54**	**,,**	
Cell 52	**86.16**	**,,**	

Table 1 provides the information regarding SoH of the cell at different C-rate and cycles. As explained in Section I and III, cells with similar SoH is required for this analysis. Table 1 depicts that, SoH of the Cell37,45,49,50,51 and 52 is nearly 85.54% (i.e. SoH of these cells are identical), and variation among them is ±0.5%. Thus the mentioned cells are employed here for further analysis.

B. DM Identification

In a typical voltage (V) vs capacity (Q) curve, the voltage variation is approximately 1700 mV. Therefore, it is

challenging to identify the few mV voltage changes due to ageing and the electrodes' electrochemical reactions. A solution to this problem is to differentiate the Q with respect to V. It is known as incremental capacity (IC) or dQ/dV [20]. Another derivative method was introduced by Bloom *et al.* ([31], [32] and [33]), where V is differentiated by Q and is commonly known as Differential voltage analysis (DV) or dV/dQ. Here IC peaks are used for DM identification. At first, IC curves are produced for selected cells; an example is given in Figure *1* (a).

As discussed in Section I, DM is categorised into three groups: CL, LLI and LAM. However, in [30], negligible growth of CL is observed while analysing the IC curve; thus, only LLI and LAM are considered here. Summary of existing DM identification technique and further steps are described in the next two subsections.

1)   Existing DM Identification Technique

At present, to identify the growth of LLI and LAM from the IC curve, the following relationship is used, as shown in Table *2* ([24], [28] and [30]) and Figure *2* [30].
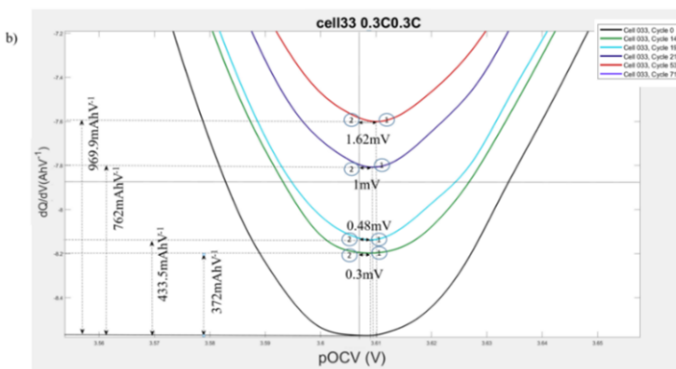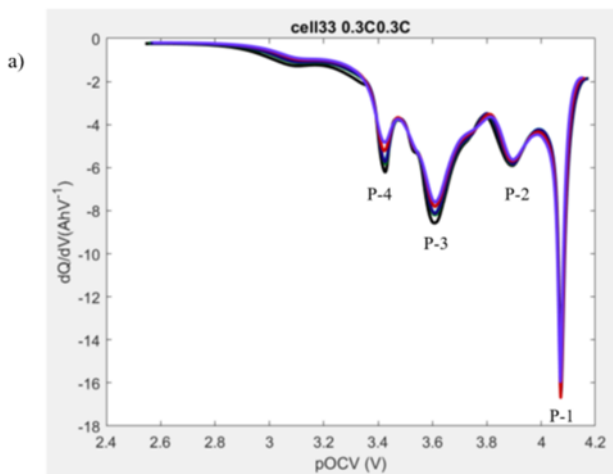

Figure 1: a) IC curve for cell 33 with Peak-1 (P-1), P-2, P-3 and P-4 on 0.3C_0.3C at 10 DegC condition. b) magnified Peak-3 (P-3) of IC curve for cell 33. on 0.3C_0.3C at 10 DegC.
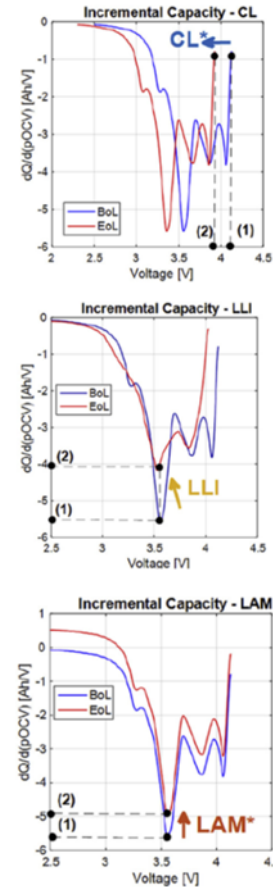
Table 2: Variation of the IC curve with respect to DM.

Change in IC peak	Unit	DM
Decrease of the height of the peaks and shift toward lower or higher voltages	$[AhV^{-1}]$ and V	LLI
Shifting toward lower voltages.	V	CL
Decrease of the height of the peaks at an approximately constant voltage	$[AhV^{-1}]$	LAM

Figure 2: Changes in the IC peaks with respect to DM[30]

Figure 2: Changes in the IC peaks with respect to DM[30] Table *2* and Figure *2* illustrate that to identify the DM from IC peak, it is crucial to identify the shifting and decreasing of the peaks at the EoL with respect to a reference from the BoL

Figure *1* (b) illustrates the IC peaks (P-3) from several cycles for Cell33, and it indicates that all the IC peaks are shifting by less than 2 mV. Similarly, IC curves are drawn for other cells and found all the peaks are shifting on abscissa by a few mV, and the intensity of the peaks are decreasing on Y-axis by a few $mAhV^{-1}$. To overcome these issues and to distinguish the growth of DM it is required to select a threshold point. The function of this threshold point is explained in the following subsection.

Here, 5mV (X-axis) or 5mAhV^{-1} (Y-axis) peak position difference is chosen as threshold point to detect the peak shifting or the decrease of intensity. It is expected that 5mV or 5mAhV^{-1} difference is trivial, may occur from experimental error and can be considered almost negligible, Thus, if there is 5mV difference between two peak positions than it can be said that their position is constant. Instead of 5mV, any other voltage level lower than 5mV or slightly higher than 5mV can be selected as a threshold point. However, while detecting the DM similar threshold point should be followed to maintain the consistency of the DM detection process.

2) Further steps of DM identification

Following the relationship in Figure 2 and Table 1, two conditions are assumed:

Assumption X: If the difference between Y-axis peak points (i.e., ③ and ④ from Figure 3(b)) is ≥ 5 mAhV^{-1}, peaks are decreased. If the difference is < 5mAhV^{-1} then it is considered that peaks are not decreased.

Assumption Y: If the difference between X-axis peak points (i.e., ① and ② from Figure 3(b)) is ≥ 5mV, peaks are shifted. If the difference is < 5mV, then it is considered that peaks are not shifted, i.e., peaks are at a constant voltage.

According to Table *2*, if the intensity of the IC peaks decreased and shifted toward lower or higher voltages, then the most pertinent DM is LLI. Thus, if IC curves show the difference between ① and ② is ≥ 5 mV and if the difference between ③ and ④ are ≥ 5 mAhV^{-1} then the peaks are considered as LLI dominated.

According to Table *2*, if the height of the IC peak decreased at an approximately constant voltage, then the most pertinent DM is LAM. Thus, if IC curves provide the difference between ① & ② is < 5 mV, and if the difference between ③ and ④ is ≥ 5 mAhV^{-1}, then the peaks are considered as LAM dominated.

According to Table 2, if IC peaks shifted toward lower voltage, then then the most pertinent DM is CL. Thus, if IC curves provide the difference between ① & ② is ≥ 5 mV, and if the difference between ③ and ④ is < 5 mAhV^{-1}, then the DM of peaks are considered as CL dominated.

According to Table 2, if IC peaks shifted toward lower voltage, then then the most pertinent DM is CL. Thus, if IC curves provide the difference between ① & ② is ≥ 5 mV, and if the difference between ③ and ④ is ≤ 5 mAhV^{-1}, then the DM of peaks are considered as CL.

Moreover, to identify the 5mV and 5mAhV^{-1} peak position difference among the peaks, the following two steps also required:

The difference between the peak position of cycle 0 (at BoL) and the peak position of the remaining cycles on the

X-axis (i.e. p-OCV(V)) reflects the shifting of the peaks. For example, a magnified view of the P-4 for Cell45 is shown in Figure 3(b). Here the black line represents the peak for cycle 0, and the cycle 0 peak position ① is 3.4278 V. While the green line represents the peak for cycle 600, and the peak position ② is 3.4593 V. The difference between the two peak point 31.5 mV. Thus, it is found that P-4 is shifted by 31.5 mV (i.e. P-4 shifted > 5mV) after cycle 600.

Also, the difference between the position of cycle 0 peak and the peak position of the remaining cycles on the Y-axis (i.e. dQ/dV (AhV^{-1})) reflects the intensity of the peaks are decreasing. In Figure 3(b), ③ and ④ represent the peak position on the Y-axis for cycle 0 and 600, respectively, and the difference between these two-position is 1372.5 mAhV^{-1}, reflects that the magnitude of the peak decreased by 1372.5 mAhV^{-1} (i.e. P-4 decreased > 5mAhV^{-1}) after cycle 600.

Henceforth, detecting the difference between two peak points represents that growth of LLI is detected at P-4 for Cell45.
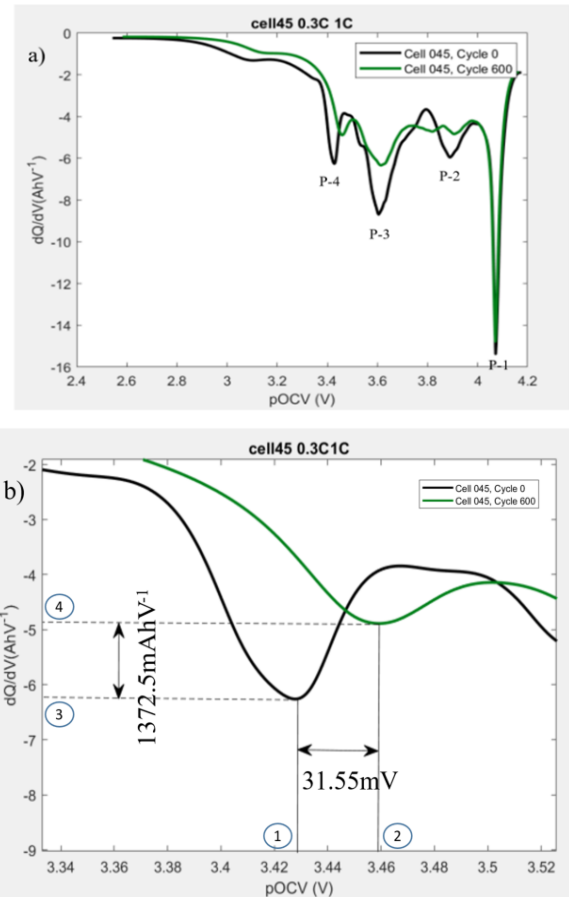

Figure 3: a) IC curve for cell 45 b) magnified Peak-4 (P-4) of IC curve for cell 45 on 0.3C_1C at 10 DegC condition.

Following the process mentioned above, DM of each peak is identified for Cell49-52 with 85.54% SoH(±1%) and identified DMs are mentioned in Table 3.

Table 3: Same DM cells at Same SoH level.

Cell No.		DM	SoH%
Cell49	P-4	LLI	86.28
	P-3	LAM	
	P-2	Peak disappeared	
	P-1	LAM	
Cell50	P-4	LLI	85.99
	P-3	LAM	
	P-2	Peak disappeared	
	P-1	LAM	
Cell51	P-4	LLI	85.54
	P-3	LAM	
	P-2	Peak disappeared	
	P-1	LAM	
Cell52	P-4	LLI	86.16
	P-3	LAM	
	P-2	Peak disappeared	
	P-1	LAM	

Table 3 depicts for all three cells (i.e. Cell49, Cell51 and Cell52), LLI identified at P-4, LAM identified from P-3 & P-1, and P-2 disappeared when the cell reached approximately 85% SoH. For Cell50, P-4, P-2 and P-1 are matched with other cells; the only exception is the P-3. In the case of P-3, the peak position difference is slightly greater than 5mV, i.e., 5.59mV. Thus, DM of cell 50 (P-3) is identified as LLI.

Nonetheless, DM is also identified from Cell 49, 37 and 45 with 85.91% SoH (±1%) and identified DM's are mentioned in Table 4.

Table 4: Different DM cells at Same SoH level.

Cell No		DM	SoH%
Cell49	P-4	LLI	86.28
	P-3	LAM	
	P-2	N/A	
	P-1	LAM	
Cell37	P-4	LAM	85.97
	P-3	LAM	
	P-2	LAM	
	P-1	LAM	
Cell45	P-4	LLI	85.91
	P-3	LLI	
	P-2	LLI	
	P-1	LAM	

Table 4 delineates DM for Cell49, Cell37 and Cell45. For Cell45 and Cell49, LLI identified at P-4 while for Cell37 LAM identified at P-4. Furthermore, for Cell49 and 37 LAM is identified at P-3, while for Cell45 LLI is detected at P-3.

IV. Conclusion

Based on the existing DM identification (Table 2) by IC curve, this study proposed a modified DM detection process to garde the cells. The process is verified by applying them to the degradation experimental results.

The limitation of this work is that due to unavailability of the data, only six cells are analysed here. Among these 6 cells, one cell (cell 50) indicate the DM might be different even cells are cycled with same c-rates and in same temperature. To increase confidence in this work, a similar procedure needs to be performed with large sample size.

The analysis of the results exhibits that the proposed modifed DM detection method can distinguish the cells with similar DM or different DM. This study also suggests that when each peak of each cell of the IC curve is matched, the cells are at the same DM state, and for opposite conditions, cells are at the different DM state.

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