Li-ion batteries life cycle from electric vehicles to energy storage

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Abstract. Vehicle electrification is an emerging solution to reduce fossil fuel dependence and the environmental pollution caused by automobile emissions. Electric vehicles (EVs) are powered by Li-ion batteries which degrade with use and time, and once their state of health (SOH, ratio of current capacity to the initial capacity) reaches 80% they retire from the EVs and need a replacement. In this study, battery degradation behaviour has been investigated and demonstrated under different electrical and thermal loading conditions. A different rate of cell degradation has been observed with different environmental and electrical loading conditions. The rate of degradation of the cells is higher at low temperatures and at high current charging conditions. Additionally, it has been demonstrated that the temperature of the cells within a battery module is different across the 6S2P battery module which would be significantly higher in the case of a bigger battery module. Hence for the potential second-life applications of the retired electric vehicle batteries, knowing the correct cell SOH is highly essential to grouping them which will lead to optimized use of this battery in 2^{nd} life applications.

Introduction

Government and policymakers are continuously promoting vehicle electrification to reduce fossil fuel dependency and minimize carbon emissions [1]. Li-ion batteries are the key contender to power EVs and HEVs (hybrid electric vehicles) due to their high gravimetric and volumetric energy density. However, the bottleneck with LIB is its endurance with time [1-3] which is caused by the capacity degradation due to repeated charge/discharge and storage. This degradation leads to a reduction in the driving range of the vehicle and eventually makes the LIBs incompatible with EVs when they reach 80% SOH (varying with the EV manufacturer and government regulations) [2-5]. At this stage, the battery needs to be replaced which costs significantly to the EV owners and the disposal of the retired batteries is hazardous to the environment [4-5]. The replacement cost of EV batteries could be reduced by selling the batteries to the businesses involved in utilizing the retired batteries in low-power and stationery applications [3,5]. Hence the selection of batteries from the first life and their SOH investigation and finally categorizing and grouping them for the potential second life is an open challenge which needs significant researchers' attention [1-6].

Since the cycle and the shelf life of each cell within a battery pack are dictated by the rate of capacity degradation and operational conditions. In the case of EVs and HEVs, multiple cells are arranged in series, parallel and mixed connections to achieve the desired voltage and current [1,4]. Hence, (i) cells within a battery pack have variations in electrical loading based on the series-parallel architecture of the battery systems and (ii) they would have different temperature conditions based on their location within the pack thermal management system [5-8]. These two variations lead to the uneven degradation of the cells across the battery pack [5-8]. However, the SOH of the pack is specified by the weakest cell [6,7] which has faster degradation due to the

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nonhomogeneous electrical loading or due to the manufacturing defect caused by slower ionic/charge transport [9,10]. Hence, an understanding of the cell behaviour in parallel to the battery pack is highly needed to pinpoint the root causes of the pack failure. Identifying weak cells within the battery pack can facilitate the replacement of that module/cell containing dead cells and can improve the capacity utilization of the battery pack.

Battery End of Life, Reuse and Recycling

End-of-life (EOL) of EVs and HEVs battery systems are considered once they hit 80% SOH, however, these retired batteries have significant leftover energy [11] which must be utilized by some means otherwise will go to waste [9-13]. An EOL of the EV batteries will strongly impact the economic, environmental, and political dynamics of the nation because the battery raw materials are not evenly accessible across the globe [1-3]. Hence, reuse of the batteries for Energy storage systems (ESS) can minimize that impact by offering significant economic and environmental benefits [9-10]. ESS is an essential factor for the smart grid which helps store energy at times of load shifting and low energy demands and supply at times of high energy demand and saves the cost for the consumers and suppliers. For instance, reusing these batteries in utilities can provide energy at the time of the fluctuations since the cost of the retired batteries ranges from \$38-\$147/kWh as compared to new LIBs ~\$209/kWh [14]. However, the application of the retired batteries needs to be encouraged by the policymakers by providing funds to the new businesses [7,9,14]. Major areas of battery 2nd life application are:

- Low-speed EVs
- Mobile power
- Energy storage for home use
- Backup power and energy storage sites

 2^{nd} life application of the batteries can encourage business model innovation which will link transportation with other energy applications [1,4]. These applications could reduce the effective price of EVs and reduce its life cycle impacts on the EV owners. Reuse of the batteries will promote the EVs uses on the road which can also greatly reduce CO₂ emissions and improve air quality using retired batteries in the grid application and UPSs [10]. Since batteries are not simple waste which can be disposed of anywhere, they are electrochemical systems which if not disposed of safely might cause accidents and fire [12]. Therefore, its disposal is also expensive which generates various harmful gases which pollute the environment and atmosphere and this can be minimized by the reuse of the battery systems [2,13-14].

Battery SOH and Capacity Estimation

Battery pack SOH and rate of capacity degradation estimation are essential for the appropriate 2nd life applications or subsequent recycling. For 2nd life applications of the batteries, it is highly desirable to have a similar rated capacity and comparable SOH and, also have a capacity greater than 50% of the original capacity [7-8]. A dissimilarity in cell SOH within a pack leads to incomplete utilization of the battery pack since the battery capacity is defined by the capacity of the weakest cell connected in a series [9]. Since the SOH of the battery pack is determined by the SOH of the weakest cell it is inadequate to maximize the energy utilization of the entire pack [4,6]. Therefore, identification of the cell SOH is highly desirable for the best possible uses, because the parallelly connected cells undergo different currents due to varying internal resistance which leads to different degradation rates [10,11]. Even cells in a series connection can have different SOH and degradation rates. Therefore, the SOH and capacity estimation of the pack without knowing the SOH of the cell is misleading information about the battery systems. For the 2nd life application, cells from the same SOH and internal resistance are recommended even in case of parallel connections [12,13]. There are a variety of SOH assessment techniques reported in the literature, however, either they are time-consuming or are not accurate enough [6-8, 15-18]. The SOH

methods must be cheap and fast enough to reduce the SOH testing apparatus time as well as cost which makes it economical and benign [1,8,9]. Battery SOH is widely assessed by a series of charge/discharge cycles which take several hours to conduct and are hence time-consuming and costly [4,8-9]. However, because of 2nd life application, a fast-screening method with high accuracy is highly desirable to make the reuse of EV batteries sustainable. In this study, battery degradation behavior has been investigated and demonstrated under different electrical and thermal loading conditions. Additionally, a simulation of thermal gradients for a 6S2P battery module has been conducted to demonstrate temperature variation within the cells and across the module.

Test Methodology and Data Collection

To conduct the test, fresh batteries are aged under various loading and operational conditions up to the EOL. This process involves initial electrochemical milling/formation of the cells, followed by a reference capacity test (RCT) within a thermal chamber using a battery cycler at 25°C. To accurately assess the cells' performance, we aged them through mild and aggressive electrochemical cycling and various drive cycles, simulating temperature conditions ranging from 0-25°C. To achieve the required thermal environment, the batteries are placed within an incubator to imitate the real-use scenarios. By regularly conducting RCTs, we monitored the SOH and performance of the cells throughout their first life until the cells reached their EOL. With this approach, battery ageing data was collected and analyzed, which are discussed in the results section.

Results and Discussion

In this section, discussion of the cycling behavior of LGM50 commercial cell is presented. Figure 1 shows the variation in cell capacity with respect to cycling at different temperature conditions which is 0, 10 and 25°C. As can be seen, the cycling behavior of the cell is impacted by the environmental temperature. Three different charging conditions have been used which are 0.3C, 0.5C and 0.7C, however the discharge current is 0.3C for all the cases. Observing the discharge behaviour of the cell it is evident that cell degradation is highly dependent on temperature conditions as well as applied current which indicates the loading condition of the cell. Additionally, for similar cycling/environmental conditions there is significant cell-to-cell variation. Even in similar test conditions cell performance and cell degradation would be different, which indicates that even within the same battery module in a real use case scenario the cells within a battery module will degrade differently just because of different loading conditions. Hence talking about Figure 1(a) cells at 0.3C charge condition for all the temperature conditions have shown less degradation even after 800 cycles compared to 1000 cycles. After 800 cycles cell has lost only 15% of its capacity. Furthermore, before the occurrence of the knee point cell-to-cell variation for the capacity degradation is minimal however after the knee point significant variation between cell-to-cell is observed which is highest at 0°C and 10°C.

After further analyzing the capacity variation of the cell with cycling at 0.7C charge condition for the similar temperature conditions we can see that cells lose their capacity rapidly for all the temperature conditions. It is also observed that the rate of degradation is highest in case of 0°C as can be seen in the figure 1(b). For this condition the degradation behavior at 25°C is significantly less as compared to 0°C and 10°C. Cell lose 20% of SoH after 200 cycles however in case of 10°C and 0°C cell lose 50% of SoH within 70 and 80 cycles respectively. Aging these cells further even after losing 20% of its SoH, we can see the cells lose around 60% and 70% of SoH within 200 cycles for 0°C and 10°C, however at 25°C cell can go up to 300 cycles before it loses 70% SoH. This degradation at low temperature is attributed to the slower reaction kinetics and transport limitations for ions and electrons and lower diffusivity of lithium ions within the cathode and anode. Further analysing the resistance rise in the cells with cycling condition at 0.3C charge for all the temperature conditions we can see that the rate of rise of cell resistance is quite consistent for all the temperature conditions up to 800 cycles. Additionally, the cell-to-cell variation in internal resistance of the cells is quite minimal up to 800 cycles, however after 800 cycles the rate of rise of the cell resistance is significantly higher, which is also a variation in rate of rise for different temperature conditions as can be seen in the figure 2. For capacity variation with cycling, we can see that the rate of rise in cell resistance at 10°C is significantly higher than the other two temperature conditions including higher cell-to-cell variation. Rapid resistance increases after 800 cycles support the previous discussion and the plots shown in Figure 1(a).

In Figure 1(d) we see variation in cell resistance with cycling at 0.7 C charging current and with 0.3C discharging current at 0, 10 and 25° C temperature conditions, all other conditions are the same for all the cells. The rate of resistance rise in the cell at 25°C is very minimal as compared to 0°C and 10°C up to 170 cycles however after that a significant rise in resistance can be observed even at 25°C. Looking for the resistance rise at 0 and 10° C we can see a significant rise even in less than 50 cycles with 40% and 50% as compared to 1.5% in the case of 25° C. With further cycling the rate of rise of the cell resistance is high in cases of 0 and 10°C and within 180 cycles the cell resistance is 2.6 and 2.8 times the initial resistance of the cell. However, in the case of 25°C cell resistance is up to 2.6 times which shows a rapid increase after 250 cycles. By analyzing the cell resistance, it is observed that a variable rate of cell degradation for different temperature conditions is shown in Figure 1(c).



Figure 1: Discharge capacity vs cycle Number (a) 0.3C and (b) 0.7C charging current, cell resistance with cycle number (c) 0.3C and (d) 0.7C charging current at 0, 10, 25°C. Discharge current for all the cases is 0.3C.

In figure 2 variation in thermal condition across a battery module with 6S2P configuration has been shown. As can be seen, the temperature of the individual cells depends on the location within a battery module. Hence, for a same thermal management system of the battery module we can see a significant variation between cell temperature within the cell as well as across the battery module. Even for a small battery module (figure 2) this much variation in temperature can be seen, hence, in case of a commercial module/pack significant variation in temperature can happen which may lead to different rate of degradation of the cells for similar electrical loading. Further on, cells within a module or a battery pack would have different electrical loading conditions due to different SP architects which may lead to different rate of heat generation within the cells which will degrade cells differently. As discussed in the results shown in Figure 1 that the rate of degradation of the cells are highly dependent of temperature conditions as well as electrical loading. And figure 2 dominates a significant temperature variation which will lead to significantly different rate of degradation of the cells within a battery module. Hence at the rate retired stage of the battery happens when battery reaches 80% SOH because of the weakest cells. These retired modules would have a significant number of cells with higher SOH. With this study, it has been demonstrated the importance of identification of the SOH of the cells for potential 2nd life applications. For an effective utilization of the remaining capacity of the retired cells, cells with capacity need to be grouped and utilized in secondary applications.



Figure 2: Demonstration of thermal gradient for 6S2P module across the cells.

Conclusion

In conclusion, a different rate of cell degradation has been observed with different environmental are electrical loading conditions. The rate of degradation of the cells is higher at low temperature and at high current charging conditions. In case of 0.3C charging conditions the rate of degradation is slower and after that and after the knee point is reached, the rate of degradation is steep. However, in case of 0.7C charging condition the rate half degradation is steep from the early stage of the cycling. Additionally, it has been demonstrated that the temperature of the cells within a

battery module is different across the 6S2P battery module which would be significantly higher in case of a bigger battery module. This temperature condition will lead to a different rate of degradation of the cells within a battery module. Hence for the potential second life applications of the retired electric vehicle batteries, knowing correct cell SOH is highly essential to group them which will lead to optimized us of these battery in 2^{nd} life application. Therefore, future scope of this study is to develop a rapid state of health measurement or estimation technique which can be effectively utilized to grade and group cells with similar SOH for the potential second life applications which can be utilized in energy storage applications or renewable energy storage applications and remote areas. With this method the battery's SOH can be assessed within a short period (<60 seconds).

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