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Performance of 26650 Li-ion cells at elevated temperature under simulated PHEV drive cycles

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ABSTRACT

Cylindrical (type: 26650) Li-ion cells (LiFePO₄ cathodes) currently used in the electric vehicles (EVs), plug-in hybrid electric vehicles etc. were subjected to simulated federal urban driving schedule at 25 and 50 °C for performance evaluation. Drive profiles (current versus time) for charge sustaining and charge depleting modes were derived from the federal urban driving schedule velocity profiles considering acceleration, regenerative braking, rolling resistance, drag force etc. for typical plug-in hybrid electric vehicles. In particular, the batteries were cycled extensively at 50 °C under charge sustaining as well as charge depleting modes to monitor capacity values, followed by analyzing the LiFePO₄ cathode material by X-ray diffraction analysis. The capacity degradation was found to be very significant in both the modes with 13 and 19% under charge sustaining and charge depleting modes after 337 and 1007 cycles, respectively at elevated temperature. High frequency resistance values measured by electrochemical impedance spectroscopy were found to increase significantly under high temperature cycling, leading to power fading. As evident from Rietveld analysis, phase change in LiFePO₄ is observed beyond 1000 cycles at elevated temperature under charge depleting mode, with the observation of FePO₄ from the powder diffraction data of the cathodes from the cycled cells. In addition, there was also significant change in crystallite size of the cathode active materials after charge/discharge cycling under charge depleting mode.

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Introduction

With increasing number of automobiles around the world, it is imperative that the greenhouse gas emissions can be reduced through minimizing fossil fuel consumption [1]. In this

context, automotive industry is shifting towards a sustainable mode of transport by hybridizing powertrains with battery and/or fuel cell systems. In the recent past, several automakers have commercialized plug-in hybrid vehicles (PHEVs) and hybrid electric vehicles (HEVs) [2,3] with various batteries. The PHEV technology is more advanced than HEV due to its

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ability to drive longer ranges solely with electric power using smarter energy management algorithms and convenience of recharging battery pack with household wall outlets [4].

The PHEVs could be series and parallel powertrains with parallel architecture showing higher efficiencies as detailed in the literature [5,6]. However, the efficiency also the PHEVs depends on driving patterns, energy management algorithms etc.. In general, the PHEVs operate on both charge depleting (CD) and charge sustaining (CS) modes based on torque requirement and driving conditions, making the control algorithm for energy management complex. Equivalent consumption minimization strategy (ECMS) is the most widely used algorithm in PHEVs leading to improved fuel economy as detailed by Tulpule et al. [7].

In recent years, electronic and automotive industries are extensively investing in developing Li-ion batteries (LIBs) due to higher energy and power densities with low self-discharge rates compared to other batteries [8]. Cycle life of the LIBs is largely influenced by type of cathode/anode materials and type of discharge profiles. State of the art cathode materials for PHEVs/EVs applications are LiCoO_2 (LCO), LiFePO_4 (LFP), $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$ (NMC) and LiMn_2O_4 (LMO), while anode materials are carbon or $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (LTO) in the LIBs.

LIB with LFP cathode is one of the major battery systems projected for automotive applications, due to its low cost, long cycle life (~5000 cycles) compared to LCO (~1500 cycles) with much better safety in comparison with NMC [9–13]. Even though LTO is a potential anode with longer cycle life compared to that with graphite, they suffer from lower energy density [14,15].

The LFP shows excellent flat discharge voltage of 3.5 V vs Li [16]. As LFP is not highly conducting (electronic conductivity: $10^{-9} \text{ S cm}^{-1}$), the rate capability is being improved by carbon coating [17,18]. Battery life and degradation mechanism is strongly dependent on battery test profiles, discharge/charge methods, and ambient conditions. Han et al. [12] reported the capacity fading for commercial LIBs with NCM/LTO, LFP/C and LMO/C under EV loads using hybrid pulse power characterization. Among various systems evaluated, the LFP cathode showed capacity fade of 15% after 1020 cycles before end of life, while NCM/LTO showed no capacity fade. Fading mechanism in LFP/C was reported mainly due to loss of usable Li^+ ions, no substantial change in internal resistance values was reported after end of 1020 cycles.

Ramadass et al. [19] studied capacity fading of Sony cylindrical 18,650 Li-ion (LCO cathode) cells at 25, 45 and 55 °C using constant current–constant voltage (CC–CV) protocol (1A). LCO on cycling at 45 and 55 °C showed capacity of 36 and 70% respectively after 490 cycles. The higher capacity fade at elevated temperatures was ascertained due to primary active material loss (Li^+) and secondary active material loss (LiCoO_2). In addition, authors also reported analysis on three major modes for capacity fading, (a) loss of secondary active material (LCO/C), (b) primary active material (Li^+) and (c) rate capability loss. Loss of secondary active material was found to dominate regardless of cycling temperature among LCO cathode [20].

Zhang et al. [21] reported capacity and power fading of prismatic Li-ion (LFP cathode) cells at various temperatures under constant charge/discharge protocol and FUDS drive

profile in EV mode. For the first 300 cycles, batteries were cycled at 3 C rate (charge/discharge), later EV current profile was implemented for the next 300 cycles under the operating window of 80–30% SOC. After 600 cycle batteries showed capacity loss of 14.3% at 45 °C. They concluded capacity fading at elevated temperature is due to loss of cyclable lithium during the cycling test and no much power fade was observed at elevated temperature. At lower temperature capacity and power fade becomes severe due large increase in cell impedance.

Capasso et al. [22] evaluated $\text{Li}[\text{NiCoMn}]\text{O}_2$ (NCM) and LFP battery packs using CC–CV protocol and dynamic current using trapezoidal wave form. The LIBs with LFP cathodes showed no significant capacity fading with both stationary and dynamic operating conditions compared to that with NCM cathodes, under CC–CV modes. As reported in the literature, the LIBs reached >45 °C when the ambient temperature is 25 °C, even under active thermal management system in the PHEVs/EVs [23]. Dubarry et al. [24] examined fading mechanism in LFP at 25 and 60 °C by incremental capacity analysis, they suggested capacity degradation at higher temperature is due to loss of lithium inventory followed with loss of active materials. Aging diagnostic analysis using various electrochemical analysis techniques on LFP was carried out at –30, 0, 15, 45, and 60 °C with different CC–CV test protocol by Liu et al. [25] revealed that loss of active lithium is major source of fading at elevated temperature, while no appreciable increase in resistance was observed. Additionally, Safari et al. [26] reported aging mechanism of commercial LFP cells cycled under conventional CC–CV protocol and complex current power profile at 25 and 45 °C along with different storage conditions, their conclusions were in agreement with fading mechanism reported in the literature i.e. aging mechanism is dominated due to loss of lithium inventory, while small increase in cell impedance at 45 °C on cycling was reported due to loss of graphite active material.

In the present study, cylindrical-26650 LFP based cells (5 Ah) were evaluated under a typical PHEV loads subjected plug-in hybrid electric vehicles (FUDS) drive cycle with two different current profiles CS and CD at 25 °C and 50 °C. The present study focuses on the performance and failure mode/capacity fading analysis of the LFP based LIBs at elevated temperatures under CS and CD modes.

Experimental

PHEVs driving profiles

Hybrid powertrains can run in charge sustaining, charge depleting and blended modes, depending on the torque requirement and the state of charge of the batteries. PHEVs are beneficial for urban driving due to their higher electric range compared to HEVs, thereby reducing dependence on ICEs to propel the vehicles. The FUDS protocol from EPA for light duty PHEVs as shown in Fig. 1 provides a total drive distance of 7.45 miles with an average velocity of 19.59 miles per hour [27]. As seen in Fig. 1, the velocity profile displays the typical start/stop, acceleration/braking conditions in an urban traffic situation.

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