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A Study of the Effects of External Pressure on the Electrical Performance of a Lithium-ion Pouch Cell

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***Abstract*—The introduction of lithium-ion batteries for vehicle powertrain electrification has increased in recent years. They feature high energy density, high power density, long cycle life, and is also environmentally friendly compared with other types of batteries. A large number of Li-ion cells are usually required to meet the demand in capacity and power for automotive applications. Pouch cells have been favored by many manufacturers because of the high packaging efficiency and, therefore, a higher pack energy density. However, robust packaging is required for performance and safety criteria due to their low mechanical stability, which results in them being compressed in the module/pack. This paper describes research into the effects of external pressure on the electrical performance of lithium-ion pouch cells. The authors have adopted pulse power test, capacity test and electrical impedance spectroscopy test to characterize the effects and the test result indicates lithium-ion pouch cell's performance changes under varying external pressures. Conclusions are drawn on how to make use of the results presented to influence and improve the design of automotive battery modules and packs to meet the challenges in the automotive industry.**

Keywords — lithium-ion battery; electrical performance; external pressure; impedance

I. INTRODUCTION

Over the past decades, lithium-ion (Li-ion) batteries have attracted wide attention in a number of applications from small cells in consumer electronics to full size battery packs in large scale applications. The Li-ion battery has high energy density, high power density, long cycle life, and is also environmentally friendly compared with other types of batteries. Hence, the adoption of Li-ion batteries for vehicle powertrain electrification has increased in recent years. A large number of Li-ion cells are usually required to meet the demand in capacity and power for automotive applications. A Li-ion cell typically has a characteristic voltage of ~3.7V and vehicle applications typically require voltages of ~300V. Pouch cells have been a popular choice for manufacturers because of the high packaging efficiency and, therefore, a higher pack energy density. However, pouch cells have low mechanical stability and robust packaging is required for performance and safety criteria, which results in them being compressed in the module/pack. In consideration of how the pouch cells are packaged, this study investigates the effects of external pressure on the electrical performance of lithium-ion pouch cells.

A study of the major testing standards, i.e., IEC 62660-1, ISO 12405-2, FreedomCAR Battery Test Manual, [1-3] gave no guidance regarding how a cell's performance shall be characterised against different external pressures although other external factors have been specified, e.g., temperature, orientation. According to the literature reviewed, link between the stack level mechanical stress and capacity fade of Li-ion cells has been considered [4, 5]. However, there is no widely accepted theory of how large scale Li-ion pouch cell's performance changes under varying external pressures. Previously researchers investigated effect of pressure on individual component such as anode, cathode, and separator. Gnanaraj et al reported compressing graphite electrodes has an adverse effect on the contact between the active mass and ions in solution [6]. They also testified to a slight improvement of performance of $LiCoO_2$ electrode under compression. The theory of performance increase of $LiCoO_2$ electrode is also investigated on individual electrode in a later study by Mao-Sung et al [7]. An explanation was given on the root cause of performance increase, as applying 0.08 bar pressure increases the electrode-electrolyte interface. Previous work has also indicated that ion transport properties of porous separator reduce when compressed [4, 5]. The author of this paper has developed a bespoke test rig and carried out experiments to investigate the effects of external pressure on the electrical performance of commercially available lithium-ion pouch cells. This paper also discusses how the performance of a vehicle pack is affected according to the experimental results observed on the individual cell. Finally, this paper draws conclusions about how to make use of the results presented to influence and improve the design of automotive battery modules and packs to meet the challenges in the automotive industry.

II. EXPERIMENTAL METHOD

A. Test matrix

Experimental studies were performed on commercially available nickel manganese cobalt oxide (NMC) based 25Ah lithium-ion pouch cell. The minimum discharge voltage of the cell is 2.7V and the maximum charge voltage is 4.2V, with a nominal voltage of 3.7V. Two cells were selected for this study. Prior to the experiment, the cells were inspected and passed the cell selection procedure adopted by the battery characterization laboratory, WMG center HVM Catapult to make sure cells had consistent manufacturing process and they were not damaged during delivery. Electrical impedance spectroscopy (EIS), capacity and pulse power tests were performed on these cells at three different temperatures i.e. 45^0C, 25^0C and 0^0C, and with four different external pressures applied, i.e. ambient air pressure, 0.2 bar, 0.4 bar and 0.8 bar. Hence, there were 12 different test conditions. These characterization tests give insight of the cell performance in-terms of capacity, maximum power and internal impedance. Three temperature points are selected to represent room temperature and representation of high and low temperatures that an automotive battery can experience. The pressure cells will experience within a pack/module depends on the design of the pack/module. The pressure range selected here is influenced by the cell manufacturer recommendations and current automotive battery pack designs. The experiments were conducted using a bespoke pressure control rig, temperature controlled chamber, a potentiostat outfitted with a 20A booster module and a battery cell cycler.

At the beginning of the experiment, capacity, pulse power tests [1] and EIS tests were performed at three temperatures without applying external pressure. Next, the same tests were repeated by applying external pressure at 0.2 bar, 0.4 bar and 0.8 bar respectively, using the test rig shown in Fig. 1.

Within the test rig the cell sits between a flexible plate and a fixed plate. The pressure is provided by an inflatable air bag which is positioned between two flexible plates to ensure the pressure applied on the two cells surface is equalized and unified. Moreover, the air bag is connected with air supply and regulated in a manner to maintain the set pressure constant. Pressure was increased gradually to avoid any irreversible change caused by higher external pressure.

For the temperature sequence, 25^0C was performed first; then the temperature was increased to 45^0C and finally decreased to 0^0C. The temperature sequence was selected to minimize any irreversible ageing caused by high and low temperatures. For each change in temperature the cells were allowed a minimum 3 hours to soak at the set temperature.

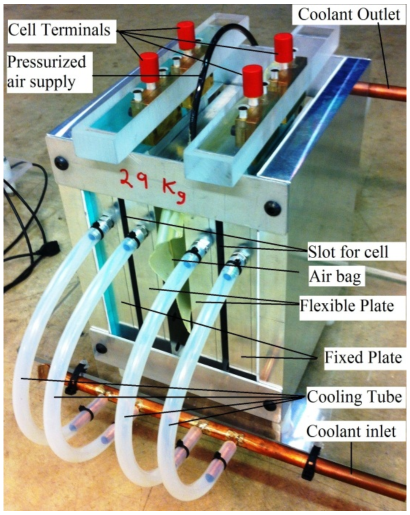


Fig. 1. Test rig

B. Characterization test details

The pulse power test was performed only at 50% SoC to maintain a balance between charge and discharge pulses. The SoC adjustment was carried out in accordance to IEC 62660-1 standard [1]. Specifically, 50% SoC was reached by discharging a fully charged cell for 30min at 1C rate. The pulse profile is consisted of 1.6C, 3.2C, 4.8C, 6.4C and 8C charge and discharge pulses of 10s duration each. This pulse profile is charge neutral, but not energy neutral. So, SoC was re-adjusted before repeating it at different temperatures and pressures.

EIS measurements were performed in the frequency range of 10 mHz to 10 kHz with a 1.25 Amp RMS sinusoidal current. Impedance spectra were measured only at 50% SoC. SoC was adjusted at 25^0C and EIS were performed at different temperatures.

For the capacity test, first the cells were fully charged at 25^0C using CC-CV method at 1C rate. Next, the test temperature was adjusted and the cell soaked for 3 hours before being discharged to the lower voltage limit at 1C rate.

III. RESULTS AND DISCUSSION

A. Pulse power test

Fig. 2 shows the pulse power test results of the cell under four different pressure conditions for both discharging and charging.

A1. – Discharging Behavior

In fig. 2(a), the cell voltage after the 10s discharge pulses at 45^0C are plotted. As pressure increases the cell voltage decreases. The effect increases with increase in discharge current which is clearly visible from the gradient of linear fits shown in the figure. This indicates an increase of internal impedance of the cell with increasing external pressure, resulting in lower power and capacity. Fig. 2(b) shows similar results obtained at 25^0C. It is also follows the result shown in fig. 2(a) except that the cell voltage was increased when 0.2 bar external pressure was applied compared to the no-pressure condition. That means applying 0.2 bar external pressure at 25^0C actually improves the performance of the cell. Above this, the voltage continues to drop with increase in external pressure. At 0^0C it was not possible to continue discharging the cell at or above 4.8C as the cell voltage drops below the minimum discharge voltage. Because of this reason the data at 0^0C is not analyzed here.

A2. – Charging Behavior

In figure 2(c), the cell voltage after the 10s charge pulse at 45^0C is plotted. As pressure increases, cell voltage increases, which continues to increase with increasing charge current as can be seen from the gradients of linear fits. Figure 2(d) shows similar result at 25^0C. High current charge pulses were not completed as the cell voltage reached maximum charge voltage early in this condition. Similar to the behavior of the cell during the discharge pulses at 25^0C here also cell has better performance at high current when 0.2 bar external pressure applied compare to the no-pressure condition.

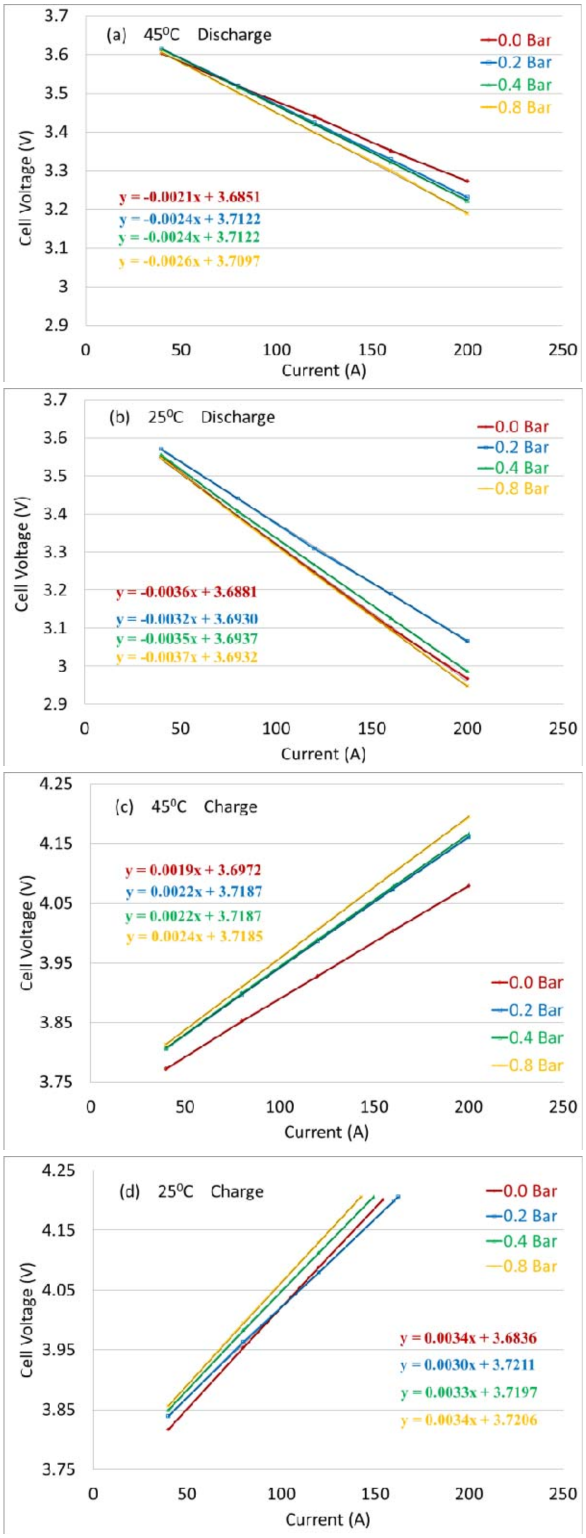
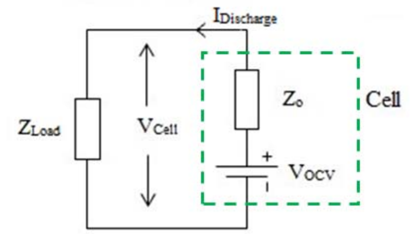


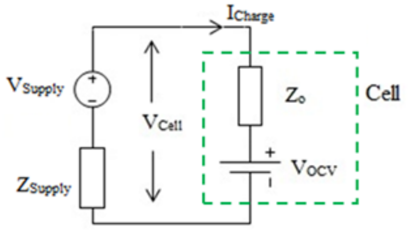
Fig. 2 Cell voltage at the end of 10s charge-discharge pulse, under four different pressure conditions (a) 45^0C, (b) 25^0C, (c) 45^0C and (d) 25^0C.

(a) Discharge Circuit



$$V_{cell} = V_{OCV} - IZ_0$$

(b) Charge Circuit



$$V_{cell} = V_{OCV} + IZ_0$$

Fig. 3 Charge and discharge equivalent circuit of a battery cell (a) during discharge (b) during charge.

These results also indicate an increase in internal impedance with increase in external pressure despite the gradients having opposite signs. This can be explained by a consideration of the equivalent circuit of a cell during discharge and during charge and is shown here in figure 3 (a) and (b). The equations are developed using Kirchhoff's circuit law. Here, Z_0 is the internal impedance of the cell. If Z_0 increases, the cell voltage, V_{Cell}, goes up during charge and down during discharge. Also, when the cell voltage increases more quickly, maximum charge voltage will be reached sooner, which leads to temporary capacity loss.

B. Capacity Test

Fig. 4 (a), (b) and (c) show the discharge curves of the cell under four different pressure conditions at 45^0C, 25^0C and 0^0C, at 1C rate. As the external pressure increases, cell capacity decreases steadily. This experiment shows an external pressure of 0.8 bar leads to a capacity reduction of 2% at 25^0C and 4% at 0^0C. The shape of the discharge voltage remains unchanged except at the beginning and end of discharge period; it is similar for all three temperatures. From these results it can be said that there is no change of voltage plateau of cathode and graphite anode due to external pressure [8]. However, external pressure does change the capacity of the cell which happens at the last portion of the discharge period. At this portion the cell voltage drops quickly, in other words, impedance rises faster which is affected by external pressure. It is a common practice in automotive applications to operate cells within a voltage window narrower than battery manufacturers' recommendations and typically on the voltage plateau.

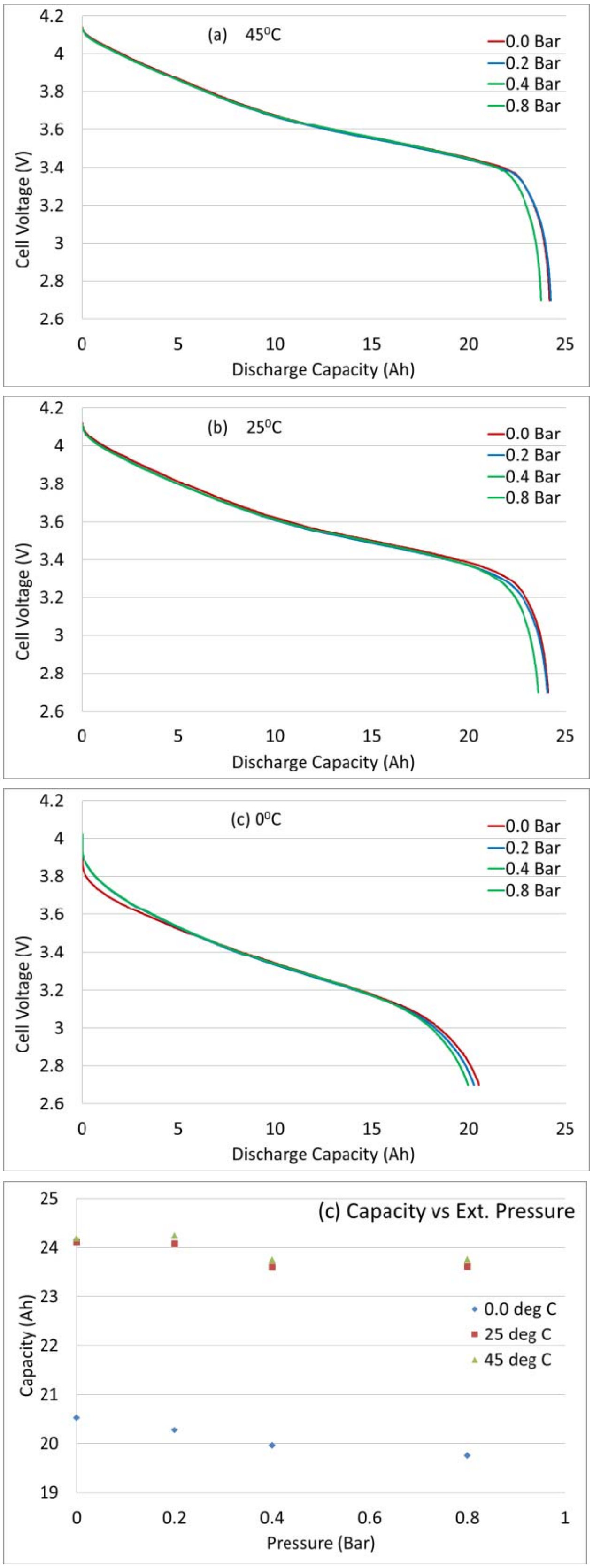


Fig. 4 Discharge curves of the cell at three different temperatures: (a) 0^0C, (b) 25^0C and (c) 45^0C.

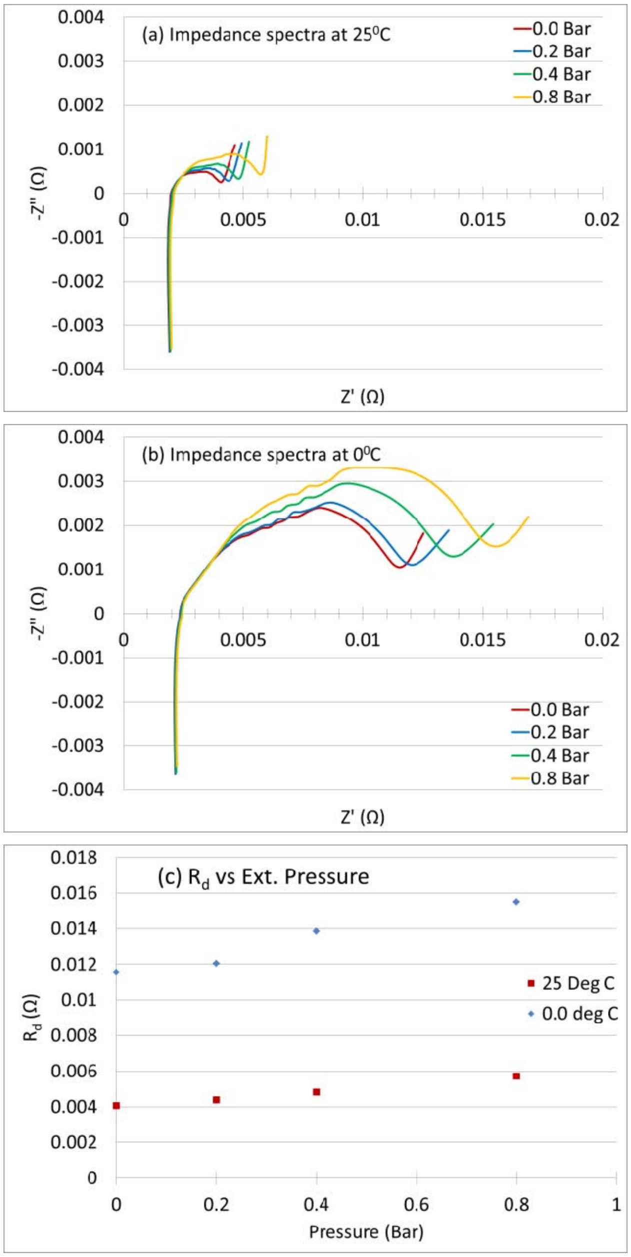


Fig. 5 EIS test results at three different temperatures: (a) 25^0C, (b) 0^0C and (c) shows increase of R_d with external pressure.

C. EIS test

EIS has the potential to give insight into the cause of the impedance rise. Impedance rise originating from different phenomenon are separated by their different time constant [9-11]. By analyzing the impedance spectrum in Nyquist representation the root cause can be identified. However, for simplicity in this study two simple parameters are used to analyze the data. The first parameter is R_0, which is the point on the 'x' axis where the impedance spectrum crosses the axis. It is commonly referred to as the pure ohomic resistance of the cell [9]. The second parameter is R_d, the real part of the battery impedance when negative imaginary part has the local minimum at the higher end of the 'x' axis. R_d is often called the total resistance of the cell as it incorporates pure ohomic resistance and electrochemical impedances.

Fig. 5(a) and (b) shows Nyquist representation of the cell under four different pressure conditions at 25^0C and 0^0C. As, the external pressure increases, R_0 does not change with external pressure at a certain temperature. The change of R_0 between fig. 5 (a) and (b) is due to change in temperature [9, 10]. However, total resistance R_d increases, as external pressure increases at both temperatures tested. This indicates external pressure changes the bulk transport properties of active materials of the cell [9]. R_d is plotted against the external pressure in fig. 5 (c). It has a linear relationship with external pressure. However, 0.2 bar shows less impedance rise compare to linear estimate, which would explain why 0.2 bar pressure perform better in fig. 2.

IV. CONCLUSION

In this study, an experimental test rig has been designed and built that allows the systematic investigation of the cell electrical performance for different applied pressures. The findings have been presented for commercially available NMC based 25Ah lithium-ion cells. External pressure affects the capacity and voltage of a cell in the temperature range 0-45^0C. Increased pressure lowers the cell voltage during discharge and increases it during charge. Increased pressure also lowers the cell capacity.

The impedance rise with applied pressure is most noticeable for the highest external pressure applied. The impedance rise indicates change of bulk transport properties of active materials, as the R_0 remains constant with applied pressure.

From this research it is clear that cells generally have higher capacity and power capability when there is no external pressure applied. However, applying a little pressure is expected to extend the lifetime of the cell; this needs further investigation. In this case there will be a tradeoff between initial performance loss and lifetime of a battery pack.

The research findings presented in this paper focused on the NMC based li-ion batteries although the investigation for other cell chemistries is ongoing. Also, an extensive EIS test is being used for wider SoC and temperature range.

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