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Effects of external pressure on the performance and ageing of single-layer lithium-ion pouch cells

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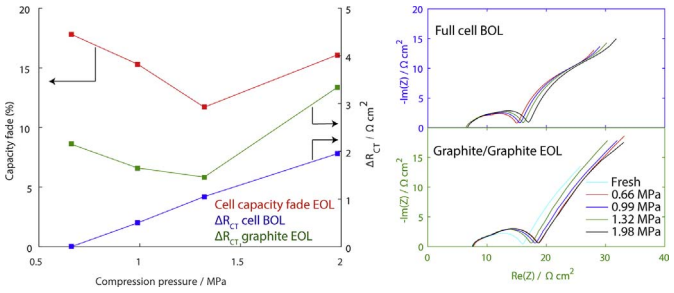
HIGHLIGHTS

- An optimum compressive pressure exists that extend the battery life.
- Cyclable lithium loss is reduced at the optimum pressure.
- Pressure-induced current distribution does not explain ageing in parallel connection.
- High compressive pressure impedes the electrochemical kinetics and mass transport.
- Results indicate coupling between electrochemistry and mechanics.

GRAPHICAL ABSTRACT



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ABSTRACT

The effects of external compression on the performance and ageing of NMC(1/3)/Graphite single-layer Li-ion pouch cells are investigated using a spring-loaded fixture. The influence of pressure (0.66, 0.99, 1.32, and 1.98 MPa) on impedance is characterized in fresh cells that are subsequently cycled at the given pressure levels. The aged cells are analyzed for capacity fade and impedance rise at the cell and electrode level. The effect of pressure distribution that may occur in large-format cells or in a battery pack is simulated using parallel connected cells. The results show that the kinetic and mass transport resistance increases with pressure in a fresh cell. An optimum pressure around 1.3 MPa is shown to be beneficial to reduce cyclable-lithium loss during cycling. The minor active mass losses observed in the electrodes are independent of the ageing pressure, whereas ageing pressure affects the charge transfer resistance of both NMC and graphite electrodes and the ohmic resistance of the cell. Pressure distribution induces current distribution but the enhanced current throughput at lower pressures cell does not accelerate its ageing. Conclusions from this work can explain some of the discrepancies in non-uniform ageing reported in the literature and indicate coupling between electrochemistry and mechanics.

1. Introduction

Lithium-ion batteries are the dominant sources of power in portable electronics such as laptops and smartphones owing to the higher energy and power density they offer compared to other types of batteries [1,2]. They are now increasingly used to fully or partially power vehicles where a longer lifetime of 10–15 years is required [3]. With the aim to prolong the battery lifetime for electrified vehicles, the effects of several ageing conditions such as temperature [4–7], charge/discharge rate [8,9], depth of discharge [10,11], and external compressive stress [12,13] on the battery degradation have been studied. Lithium-ion batteries can be subjected to stack pressure from different sources: from the rigid cans of cylindrical and prismatic cells, externally applied stack pressure in pouch cells, jelly-roll winding, material expansion and gas

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evolution in mechanically constrained cells. Besides the less attention given to the effect of external compressive stress as compared to the other ageing conditions, its effect on non-uniform lithium-ion battery ageing has not been studied in a controlled manner so far. Hence, it becomes necessary to study the effect of external compressive pressure in model lithium-ion cells in order to understand how it affects the battery degradation and optimize it for longer battery lifetime.

Previous studies have shown that external pressure can affect the cycle life of lithium-ion batteries [12] and cause non-uniform ageing when it is unevenly distributed [14]. It has been reported that prismatic cells age faster than cylindrical cells made from identical electrodes [15]. The difference was attributed to the lower stack pressure in the prismatic cell configuration resulting in increased anode porosity with isolated or less accessible particles during cycling. On the contrary, the work by Arnold et al. [12] on commercial pouch cells shows that high stack pressure causes higher capacity fade, and that a small stack pressure (~0.1MPa) is beneficial to extend the lifetime. Furthermore, inhomogeneous stack pressure has been discussed in relation to non-uniform ageing in lithium-ion batterers [14,16,17]. Klett et al. [16] reported thicker SEI layer while Bach et al. [14] reported thinner SEI layer as going along the electrode tape towards the core of a cylindrical cell jelly roll where stack pressure is expected to be higher. Additionally, pressure has been shown to affect lithium plating in opposing manner; the work by Ptzel et al. [17] reported less lithium plating towards the core of a cylindrical cell than the surface while that of Bach et al. [14] show more lithium plating at characteristic areas of high pressure. Hence, the above discrepancies call for further investigation on ageing of model systems with single layer cells (anode/separator/cathode) subjected to different levels of constant stack pressure. Such model system has advantages of excluding additional and non-uniform pressure caused by the jelly roll winding, and uses a constant pressure for better control unlike the cyclic pressure that fluctuates with the state of charge (SOC) as experienced by a cell constrained in a constant thickness scenario [18].

In this work, the influence of externally applied pressure on cell performance at the beginning of life (BOL) and on the long-term cycle ageing of both single pouch cells and cells connected in parallel is studied. A custom-built set-up with high-precision spring-loaded blocks is constructed to obtain a controlled compressive pressure over the cells. The performance is measured for stack pressures ranging from 0.66 to 1.98 MPa. Such pressure range is chosen to include pressure values on both sides of the maximum pressure observed between the metal casing and the jelly roll in typical cylindrical lithium-ion cells, little higher than 1 MPa at the end of charging [18], as well as being within the range of external pressures applied to commercial cells in industry, 0–10 MPa, as reported in Ref. [12]. In order to evaluate the source of capacity fade for the different levels of ageing pressure, electrochemical and post-mortem analyses techniques are used. The long-term cycle-induced changes in the impedance of the full cell, the electrodes and the separator are studied by electrochemical impedance spectroscopy (EIS). Scanning electron microscope (SEM) is used to study morphological changes of harvested separators. Possible mechanistic reasons for the difference in ageing as a function of pressure is suggested based on electrochemical and SEM analysis. Finally, fresh cells connected in parallel and subjected to different levels of stack pressure, mimicking pressure distribution in a cylindrical cell or in a battery pack, are cycled to study the influence of pressure distribution on current distribution and the resulting inhomogeneous ageing.

2. Experimental

2.1. Materials and pressure fixture

Experiments are performed on laboratory-scale 10.17 mAh pouch cells (polymer coated aluminum foil) constructed from single layers of Li ($Ni_{1/3}Co_{1/3}Mn_{1/3}$)O_2 and graphite electrodes (ElectrodesAndMore, USA). Specification of the electrodes is given in Table 1. Celgard separator (PP/PE/PP, 2320) of 20 μm thickness and 39% porosity is used. The electrolyte is 1 M $LiPF_6$ in ethylene carbonate (EC): diethylene carbonate (DEC) (1:1 w/w, Merck LP40). The balanced NMC and graphite electrodes used are both 30 mm in diameter. A special 'b' shaped copper and aluminum current tabs are used on each side of the circular coated electrode-separator assembly in order to apply uniform pressure which otherwise may lead to non-uniform ageing [14]. The pouch bags are dried overnight at 60 °C prior to cell assembly in order to remove any residual water. The cells are constructed in an argon-filled glovebox (O_2 and H_2O < 1 ppm).

Table 1
Electrode specification according to the manufacturer.

	Positive electrode	Negative electrode
Active material	$Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$	Graphite
Nominal voltage	3.6 V vs. Li/Li^+	0.1 V vs. Li/Li^+
Loading density	$\sim 1.0 \times 10^{-4}$ g/mm^2	$\sim 0.435 \times 10^{-4}$ g/mm^2
Active material surface area - BET	0.2–0.6 m^2/g	1.5–2.8 m^2/g

The controlled stack pressure on the pouch cell is applied by a high precision spring (Lesjöfors AB, Sweden) loaded fixture developed for this study and shown in Fig. 1. The required amount of stack pressure is set by controlling the compression length of the spring (k = 50) while securely tightening the fixture with the nuts and bolts. The pouch cell with all its components is subjected to a constant pressure because the low force-constant spring predominantly absorbs any cell thickness change during charging and discharging. From Hook's law ($F = k\Delta X$), change in the stack pressure as a function of SOC is negligible due to the small force-constant (k) and change in the thickness of the single layer electrodes (ΔX) during cycling.

2.2. Initial cell performance

The newly constructed and reproducible cells are first subjected to three formation cycles at C/10 rate between 2.7 V and 4.2 V. Cells subjected to only formation are from now on referred as fresh cells. Electrochemical impedance spectroscopy (EIS) measurement of a fresh cell as a function of pressure is performed at 3.665 V (50% SOC based on the C/25 capacity) by successively increasing the pressure in the order 0.66, 0.99, 1.32, and 1.98 MPa, respectively. In order characterize the pressure-impedance relationship in the individual

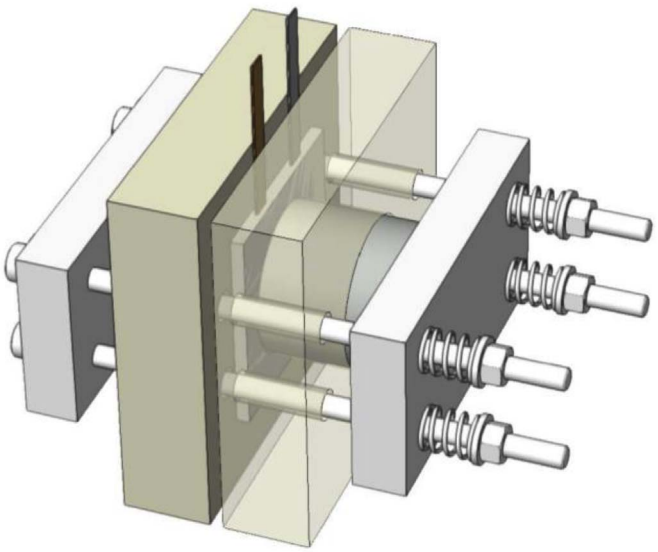

Fig. 1. High precision spring loaded pressure block.