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Application note

Introducing:
True Constant Volume Pouch Cell Cycling

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Introduction

Controlling the pressure applied to pouch cell batteries is crucial to improving their performance and cycle life [1, 2, 3, 4, 5]. One of two modes of pressure application is typically used: **Constant volume (CV)** or **constant pressure (CP)**. Keeping a constant volume is usually attempted by fixing the pouch cell in a stiff jig to inhibit cell expansion (**Figure 1b**). Nevertheless, in practice any cell fixture will expand under the high forces developed under cell cycling, in accordance with the spring constant of the jig. To level out these high forces, springs are often included in the cell fixture, accommodating the cell expansion while keeping the pressure as constant as possible (**Figure 1a**). However, when the cell expands during charging, the springs will be compressed, increasing the pressure. Because of these short-comings, in this application note we will refer to these two modes of applying pressure are pseudo-CV and pseudo-CP, respectively.

One way to realize true CP is to actively regulate the pressure applied to the cell in closed-loop control with a force sensor, as in the CompreDrive [6] (**Figure 1c**). A servo motor is then controlled based on the actual measured force, allowing precise regulation of the real pressure applied to the pouch cell.

The possibility of true CV control has been added in the latest software update to CompreDriveControl (1.16). Using a distance sensor to measure the cell thickness during the experiment, the so-called "Direct drive" mode can be employed to use the distance value as the process parameter to control (**Figure 1d**). In a true CV charge/discharge experiment, the cell thickness can be kept constant even when the cell would normally expand as a result of charging, by increasing the pressure accordingly.

In this application note, the four different pressure application modes shown in **Figure 1** are compared by cycling a $LiCoO_2$/graphite pouch cell and monitoring the resulting cell thickness and pressure.

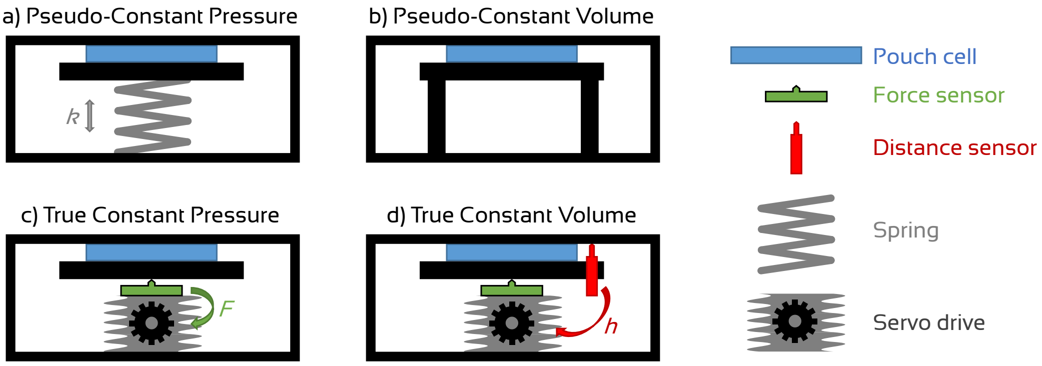
a) Pseudo-Constant Pressure
b) Pseudo-Constant Volume
c) True Constant Pressure
d) True Constant Volume

Figure 1*. Schematic illustration of the different modes of applying pressure to a pouch cell discussed here: a) With a spring, b) in a stiff cell fixture, c) by active pressure control using a force sensor, and d) by active thickness control using a distance sensor.*

Experimental

A lithium ion battery pouch cell (ICP606168PRT, Renata AG) with a nominal capacity of 2.8 Ah was used for these tests [7]. It had a $LiCoO_2$ cathode and a graphite anode, with $LiPF_6$ in ethylene carbonate / ethyl methyl carbonate / diethyl carbonate (1 : 1 : 1) electrolyte and a polyethylene / polypropylene separator. The size of the pouch cell stack was 63.1 mm × 58.3 mm × 5.10 mm. According to the data sheet, the cell should withstand being pressed between two flat plates at a force of 13 kN (3.5 MPa) at 100% state of charge (SOC) without explosion or fire.

The cell was mounted in a CompreCell Pouch 10S HC cell fixture [8] fitted with a confocal distance sensor (rhd instruments GmbH & Co. KG) [9]. The pressure and temperature were controlled with a CompreDrive [6] equipped with a 100 kN force sensor and a Huber Unistat 405 circulator (Peter Huber Kältemaschinenbau AG), controlled with CompreDriveControl 1.16 (rhd instruments GmbH & Co. KG).

A Biologic SP-240 potentiostat/ galvanostat controlled by EC-Lab 11.61 (Bio-Logic SAS) was used for cycling at 25 °C. The cell was charged by a C/2 (1.4 A) constant current to 4.2 V, followed by a constant voltage stage to C/5, and then C/2 discharge to 3.0 V. For each pressure mode, three cycles were performed, followed by a 3-hour rest step.

At the start of each pressure mode, the pressure was set to 100 kPa. True CV was achieved with the new “Direct drive” mode using a relative distance setpoint of 0 mm and force limits of 0.03 – 16 kN. This keeps the cell thickness constant throughout the experiment as long as the force stays within those limits. True CP mode was achieved simply through the normal CompreDrive force regulation [6]. For the pseudo-CV mode, the force regulation was switched off, which causes the servo motor to stay in the current position, in effect turning the CompreDrive into a stiff cell fixture.

In order to simulate the operation of a spring-loaded pseudo-CP setup, the cell thickness as measured by the distance sensor was used to control the applied pressure: The spring constant (k) determines the increase in force (ΔF) exerted by a spring when it is compressed as a result of the cell thickness increasing (Δh), according to Hooke’s law:

$$k = \frac{\Delta F}{\Delta h} = \frac{F - F_0}{h - h_0}$$

where F and h are the force and cell thickness at any point during cycling, and F_0 and h_0 are the initial values. Hence, the force applied can be calculated as

$$F = F_0 + k \cdot (h - h_0)$$

and the pressure (P) is

$$P = \frac{F}{A}$$

where A is the pouch cell area. During the pseudo-CP step, the pressure setpoint was

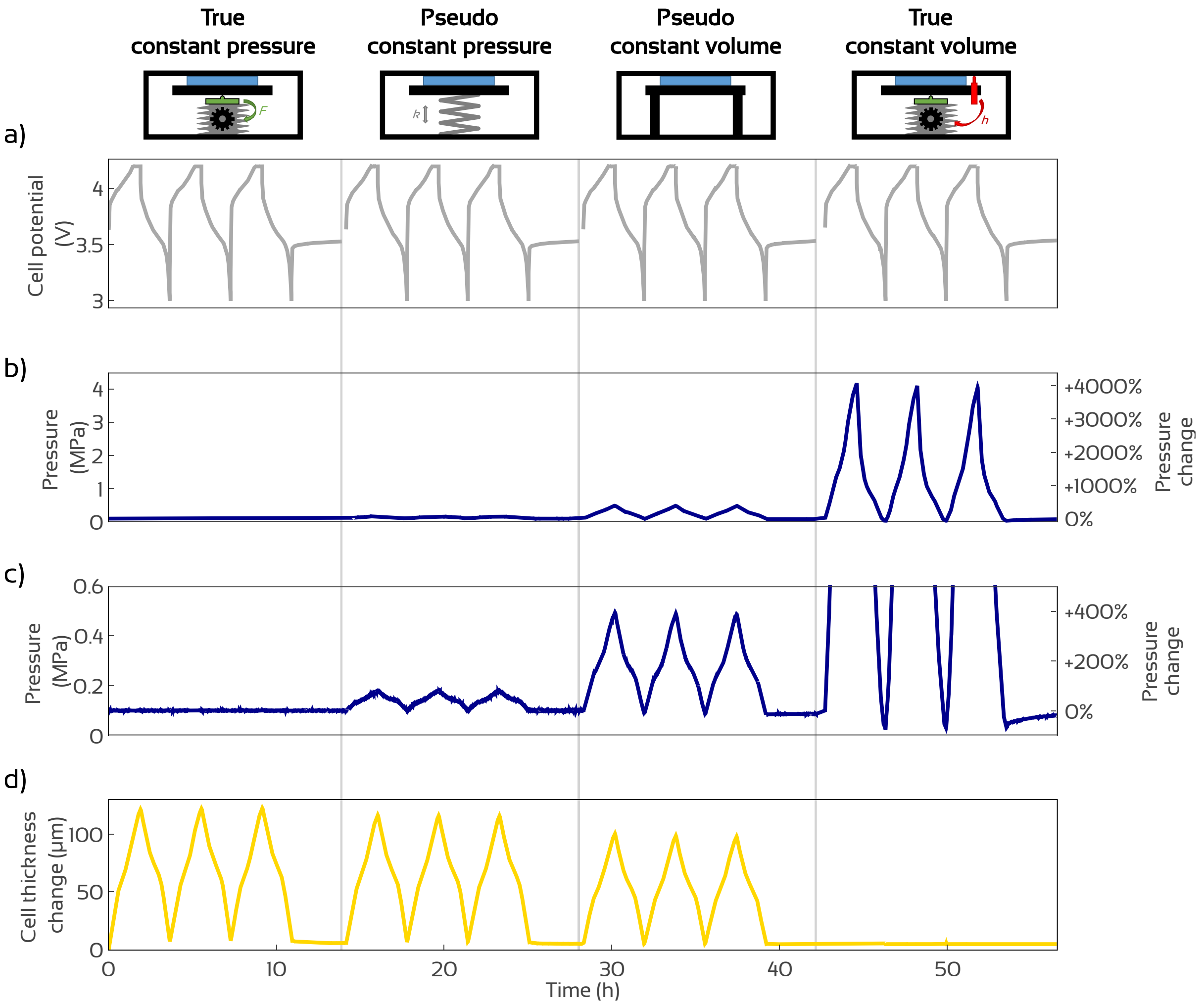

Figure 2. *Pouch cell cycling under different pressure application modes: a) Cell potential, b) pressure, c) pressure zoomed in, and d) cell thickness change vs time, as measured in operando.*

calculated in this way and applied in the CompreDrive procedure every 10 seconds. A spring constant of $k = 2.575$ kN/mm was chosen, which is the value for the standard spring in the CompreFrame, a passive pressure application frame [5, 10].

The graphs in this application note were created with Edelweiss 0.1.1 (rhd instruments GmbH & Co. KG).

Results and Discussion

The potential, pressure, and change in cell thickness are shown in **Figure 2**. During true CP mode, the measured pressure was completely constant, as expected, while the cell thickness increased by 115 µm as a result of charging. In pseudo-CP mode, however, the pressure was in fact not constant, but increased from 100 kPa to 179 kPa during charging. As a result of the higher pressure, the cell thickness increased slightly less than in true CP mode (110 µm). A more detailed investigation of cell cycling at various constant pressures was published previously [7].

Perhaps surprisingly, cycling the cell in a stiff cell fixture (pseudo-CV mode) resulted in a cell expansion of 95 µm during charging (**Figure 2**), almost the same as for the spring-loaded system (pseudo-CP mode). This is due to the high pressure exerted by the pouch cell as it is charged (493 kPa at the top of charge), which is enough to elongate even the very stiff cell fixture of the CompreDrive, constructed from two 45 mm Ø steel rods and a 50 mm thick steel top plate. It is clear that a practically sized cell fixture is not enough to suppress the volume changes of a cycling pouch cell through passive means alone.

As seen in **Figure 2**, the true CV mode in the CompreDrive was able to keep the cell thickness completely constant throughout the charge and discharge procedure. The standard deviation of the measured cell thickness during this step was only 32 nm. The pressure required to keep the cell constrained at 100% SOC was 4.2 MPa, *i.e.* 42 times the initial pressure! This slightly exceeds the maximum safe pressure stated for the pouch cell. The fact that the pressure goes below the initial 100 kPa at 0% SOC (**Figure 2b**) indicates that the cell experienced some plastic deformation, as the pressure required to keep the initial cell thickness decreased after the first cycle.

Irrespective of pressure application mode, the pressure and thickness change were fastest at low and high SOC, with a lower slope around 50% SOC (**Figure 2** & **3**). This behaviour was caused by the two-stage intercalation of lithium into graphite, pushing the graphene layers apart as lithium enters new layers [11].

The performance of the pouch cell was mostly unaffected by the pressure application mode during the few cycles shown here. In true CV mode, the cell exhibited a slightly higher overpotential, leading to a lower capacity than for the other modes (2.40 Ah vs. 2.43 Ah, **Figure 3**). The long-term effects of pressure application mode on cell performance may be much larger [1, 2, 3, 4].

The cell expansion during charge and discharge are very similar for true CP, pseudo-CP, and pseudo-CV, although the

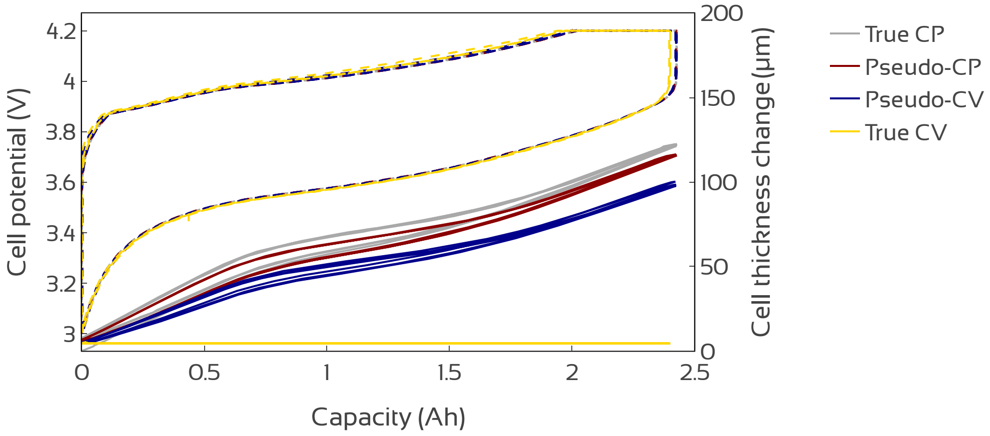

Figure 3*. Cell potential (dashed lines) and cell thickness change (solid lines) vs. capacity.*

pressure differs markedly between the three (**Figure 2** & **3**). The increase in the pressure in both pseudo-CP and pseudo-CV mode is determined by the spring constant of the cell fixture as a whole, including any springs in the setup. In a spring-loaded setup (pseudo-CP mode), this is usually easy to control through the stiffness of the springs: The cell fixture itself is much stiffer than the springs and will only have a very small contribution to the total spring constant. When not employing springs (pseudo-CV mode), it can be much more difficult to control the spring constant of the cell fixture, since it depends on the exact dimensions and materials used to construct it. As shown here (**Figure 3**), even the very stiff cell fixture of the CompreDrive [6] only decreases the cell expansion by a few percent compared to the spring-loaded (pseudo-CP) system, while the pressure increases much more. The exact amount of pressure increase will be very sensitive to the total spring constant of the cell fixture. Inconsistent pouch cell cycle life might be caused by variation in the stiffness of the cell fixture. The spring constant should therefore always be mentioned whenever experimental results on pouch cells are reported, in order for the reader to be able to properly interpret the results, as well as to reproduce the experiments.

Summary

Four different ways of applying pressure to a $LiCoO_2$/graphite pouch cell were compared by monitoring the cell thickness and pressure during cycling (see **Figure 1**). Notably, a true constant volume mode was introduced to the CompreDrive setup by using a distance sensor to measure the cell thickness *in operando*, and actively regulating the servo driven setup based on this value. In this mode the cell thickness was completely constant throughout the charge/discharge test. Meanwhile, the pressure increased from 100 kPa (0% SOC) to 4.2 MPa (100% SOC), demonstrating the massive forces required to keep the cell from expanding.

In contrast, simply relying on a stiff cell fixture to keep a constant volume resulted in a cell expansion that was only a few percent smaller than in constant pressure mode. Conversely, employing springs to keep the pressure constant was not as effective as using active pressure regulation. In these tests, the pressure in the pseudo-constant pressure setup almost doubled when charging the cell. In fact, the two passive pressure application modes are qualitatively similar, and differ quantitatively only by the spring constant of the cell fixture. The actual pressure experienced by the pouch cell depends completely on this spring constant, and it is therefore a key parameter for the setup, regardless if springs are used or not.

Literature

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