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A market definition paradigm equilibrium voltage analysis of ageing and temperature in lithium-ion cells

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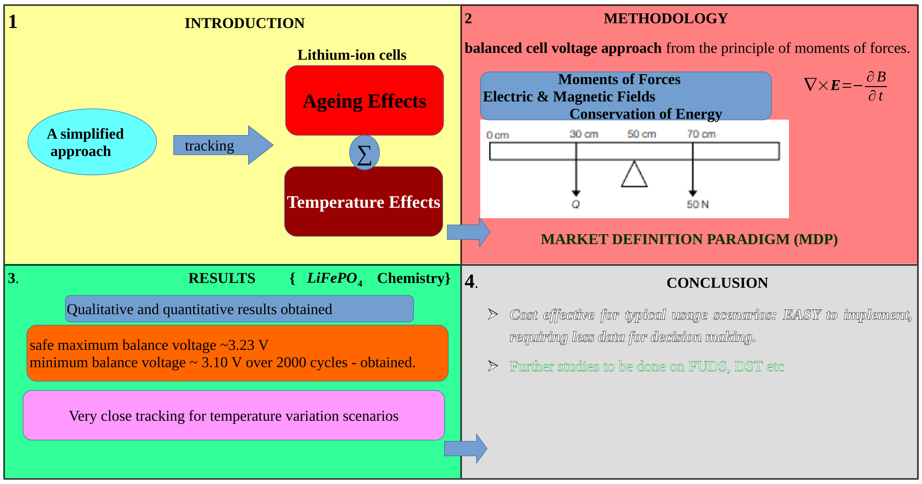
Abstract

This study was on the use of the market definition paradigm (MDP) to track ageing and temperature effects in lithium–ion cells. This was with a view to using the technique to obtain a sequence of equilibrium voltages from readily available datasets in order to profile the effects of ageing and temperature on cells and batteries. The method employed involved using the MDP with its capability to obtain a sequence of equilibrium voltages for lithium-ion cells. This approach integrated radio incidence with radio geometry, transmission and emergence in a simplified form of the cell's equilibrium voltage (and amperage). A standard dataset was obtained from the centre for advanced life cycle engineering repository. The data were processed and analysed using Coulomb counting, charging and discharging energy comparison methods on Python 3.8 programming tool and LibreOffice spreadsheet software. Results obtained show a close tracking of ageing and temperature phenomena in the cells studied. A respective maximum and minimum equilibrium voltages of 3.23 V and 3.10 V over two thousand (2000) cycles were similarly obtained for ageing and temperature investigations. The equilibrium voltage shows a downward trend as the battery ages and is more reliable for studies on these cells than the open circuit voltage traditionally used to track phenomena in such cells. In conclusion, typical lithium-ion cells can be classified at begin-of-life using the equilibrium voltage and useful predictions made with respect to end-of-life. This approach is relatively inexpensive, requiring fewer data points and low-cost hardware and extensible to online applications.

Keywords: Lithium-ion, Cell, Battery, State, Age, Temperature, Charge, Discharge

Graphical Abstract



Introduction

Batteries stand out among electronic components and or materials, which are crucial for ensuring the existence, application, development, characterization, reliability and safety of systems. The profiling of the ageing and temperature of a battery, which is a great resource like the atmosphere, via roundabout online techniques instead of its traditionally expensive laboratory-type characterization, is definitely as good as getting ready to further explore its usefulness and compatibility with other media and systems [1–4].

The need to understand and consequently effectively manage temperature and ageing effects on batteries in use is a huge and multifaceted challenge. This is evident at the micro- or even nanoscale levels such as single material layers up to large battery systems such as those employed in electric vehicles [5]. This is as a result of the nonlinearities and complexities associated with batteries [6]. Consequently, it is imperative that effective but also cost-friendly approaches be employed in tracking the ageing and temperature effects in lithium-ion batteries (LiBs). A functional analog to this is the current trend whereby traditional machine learning (ML) techniques and tools are replaced with those of tinyML (tiny machine learning) due to the huge data, hardware ,and energy requirements of the former [5, 7, 8].

Ageing and temperature variations are two crucial factors in lithium-ion battery operations [9, 10]. Challenges in effective tracking of these factors and their effects in LiBs systems have been previously reported [2, 11]. In studying ageing phenomena, there are two key aspects identified in LiBs: calendar and cyclic ageing, respectively [2, 12]. Calendar ageing is mainly concerned with ageing that takes place in a LiB due to the passage of time whether it is operational or not. Cyclic ageing on the other hand characterizes ageing effects due to charging and discharging of such batteries. The ageing- and temperature-associated complexities make tracking their causes and effects complicated [10]. These complexities arise from nonlinearities inherent in these systems, as well as operating environmental conditions which includes temperature [13].

[14] reported the use of a particle filtering (PF) algorithm to track battery ageing with nonlinear and non-Gaussian characteristics. They furthermore reported that the dynamic characteristics of battery ageing are difficult to predict for a non-rigorous monotonic decrease. However, [14] added that for a developed ensemble model to actually track the ageing trend, there was a need for measuring more capacity data and adjustment of the model parameters online. They also noted that the issue of the interaction between the random cut-off discharge and fully discharged voltage was a necessity in quantifying the actual maximum capacity in batteries. 2000 cycles (charge and discharge) can be the entire usage life for typical consumer electronic devices such as laptops and mobile phone batteries many of which use the $LiFePO_4$ chemistry [15].

According to [16], ambient temperature is a significant factor that influences the accuracy of battery state of charge (SoC) estimation. If not adequately taken care of, this can lead to disastrous consequences; for instance, electric vehicles running out of charge, satellites in orbit running out of power, and so forth. Xing et al. [14] also reported that temperature effects on battery degradation were to be considered in model modification and validation. Subsequently, the same battery (or cell) having different open-circuit voltage (OCV)–SoC curves as the temperature varies. SoC estimation improvement was also reported when using a temperature modified resistance–capacitance (RC) model by [16].

$$U_{\mathrm{term},k} = U_{\mathrm{OCV}}(\mathrm{SoC}_k, T) - I_k \times R(T) + C(T) \tag{1}$$

where $U_{\mathrm{term},k}$: terminal voltage measured across the battery terminals while in use; U_{ocv}: open-circuit voltage as a function of state of charge at time, k and ambient temperature, T; I_k:the dynamic current measured at time, k; $R(T)$: simplified total internal resistance of the battery at ambient temperature, T; $C(T)$:$C(T)$ is a function of temperature that facilitates the reduction of the offset due to model inaccuracy and environmental conditions.

[14] also showed higher SoC estimates for 0 °C than at 20 °C and 30 °C. This was for a range of 3.28–3.32 V with an interval of 0.01 V. Subsequently, the temperature increased from 0 °C to 40 °C, and the OCV–SoC curve was displaced in direction of the positive vertical axis.

Basically, the market definition paradigm (MDP) has proven to be a roundabout method for carrying out EIS [1, 4]. This is because it weaves the principles of material informatics in agreement with established theories of radiation, electromagnetism and materials in order to reconstruct the performance of energy storage systems. In line with these, the MDP aligns with the combined phenomena of ageing, radiation, convection and conduction as a generalized system for studying ageing and temperature effects. Subsequently, Einstein's mass–energy equation follows Maxwell's equation and Planck's theory of radiation, in terms of the temperature-dependent equilibrium voltage (related to the system's work function). Tracking of temperature effects, therefore, requires a tracking of the equilibrium voltage, which is the linkage between the principles of equality in radio incidence from a storage system's work function or the MDP's activation–concentration equation (ACE), polarity of radio emergence in the clustering coefficient or motivation (M), linearity of radio transmission in the path difference/length or appreciation (A) and modularity of radio geometry in the degree distribution or inertia (I), which collectively mirrors the I-A-M set of metrics of a system and obtainable using the market definition paradigm [3, 17]. The effects of ageing and temperature on the performance of cells and/or batteries have consistently shown that they depend on a set of highly correlated variables that define the cell's equilibrium voltage/amperage from the activation–concentration equation (ACE).

Research gap and motivation

Techniques for tracking ageing and temperature effects are essential for battery performance prediction and/or reconstruction. Standard methods for tracking these phenomena such as electrochemical impedance spectroscopy (EIS) which are relatively accurate are expensive and not suitable for real-time applications, which are ubiquitous [18–21]. Consequently, there is a need for methods that are much easier to implement and deliver acceptable levels of accuracy for appropriate decision-making [11]. Such techniques are urgently required to advance the currently available characterizations, understandings and applications of energy exchange schemes among materials, devices, circuits and/or systems and, hence, this paper [2, 9–12]. Furthermore, the requirement for more data especially capacity data as earlier reported by [14] is a challenge especially for critical systems that are already in operation. Hence, there is a need to find inexpensive, effective techniques to track ageing and temperature effects in these cells.

Contributions and paper organization

This paper will bring about the tracking of ageing and temperature effects in lithium-ion batteries by reconstructing lithium-ion cell performances from suitable online characterizations and significantly contribute to improving reliability and longevity of systems and products, and decision-making and/or deployment of robust battery management systems, which are crucial in various industries, including electronics, aerospace, automotive and energy systems. Following this, the paper is structured to present a seamless flow between quantitative and qualitative principles in lithium-ion battery ageing and temperature investigations via sections on materials and methods, results and discussion and conclusion.

Materials and methods

This section presents the data requirements, collection and processing for the respective ageing and temperature investigations using equilibrium voltage analysis.

Data requirements

According to the market definition paradigm (MDP) [1], for every increasing field, there will be a corresponding decreasing counterpart field at equilibrium. Typical battery (or cell) parameters that relate to these fields are the terminal voltage and current (charging and discharging). Fortunately, these parameters can be readily measured in real-time using either simple (like hand-held multimeter) or sophisticated (like the ARBIN BT2000) instruments. Table 1 shows a data structure requirement.

Table 1 Structural requirements for relevant datasets

Step time/(Seconds/3600)	Terminal voltage (V)	Current (charge/discharge) (Amperes/A)
t_1	V_1	I_1
t_2	V_2	I_2
…	…	…
…	….	…
t_{n-1}	V_{n-1}	I_{n-1}
t_n	V_n	I_n

Table 2 Current integration for charging and discharging

Charge (Ah)	Discharge (Ah)
$I_1 t_1$	$I_{D1}\, t_1$
$I_2 t_2$	$I_{D2}\, t_2$
…	…
…	…
$I_{n-1} t_{n-1}$	…
$I_n t_n$	$I_{Dn} t_n$

Thus, it is expected that a suitable dataset should contain these two parameters: current (which tracks magnetic fields) and voltage (V) (which tracks electric fields). Furthermore, the data are expected to show the step that generated it. Such a step would be one of charge, discharge or rest. Consequently, as shown in Table 1, terminal voltages are V_1, V_2 to V_n which are the voltages corresponding to step times (t_1 to t_n). Integrating the step time enables cyclic ageing tracking. A second structure requirement was temperature at which measurements were made.

This required tables with same structure as Table 1, but with the temperature condition added, leading to Table 1_θ, where θ represents the temperature (in degrees Celsius) at which such measurements were made.

Data collection, processing and analysis

To carry out this study, the $LiFePO_4$ chemistry was chosen. Lithium-ion battery data matching the data requirements earlier described were collected from the open data repository of CALCE (Center for Advanced Life Cycle Engineering) of the University of Maryland, USA. The first dataset comprised of data from two (2) cells, which underwent several cycles of charge, discharge and rest periods. The second dataset comprised of another two (2) cells, which were subjected to a low open-circuit voltage (low OCV) characterization at various temperatures. The details are of the Low-OCV characterization were reported by [16]. These data were processed and analysed using Coulomb counting, charging and discharging energy comparison methods on the Python 3.8 and Libre Office 7.3.7.2 Spreadsheet applications.

However, for the dataset described in Table 1 to be useful, there is a requirement for them to be processed into actual energy conversion which the charging and discharging processes actually do. To do this, the current is integrated over time (using Coulomb counting technique) based on Table 2. This processing is partly achieved in the ARBIN BT2000 battery system as shown in the online datasets obtained from the CALCE repository [14].

However, within each cycle, there was a separation of charging currents (and corresponding step times) and discharge currents (and corresponding step times). The absolute values of the currents were thereafter obtained using a step in the application software as shown in Eq. (1).

$$\begin{aligned} &I_n = |I_n| \text{ for Charge} \\ &\text{and} \\ &I_{\mathrm{Dn}} = |I_{\mathrm{Dn}}| \text{ for discharge} \end{aligned} \tag{2}$$

Consequently, all the negative current values (which represent) the discharging process were converted to positive values. Cumulative charge and discharge ampere-hour (Ah) was then computed using Eqs. (3) and (4), respectively.

$$\sum\nolimits_1^n I_n t_n - \text{forcharge} \tag{3}$$

$$\sum\nolimits_1^n I_{Dn} t_n - \text{fordischarge} \tag{4}$$

Thereafter, the equilibrium voltages (the voltage values for which the charge and discharge curves intersected) were obtained for each cycle using Python 3.8. The equilibrium point which was the key metric used is the point of zero loss, i.e. the point at which the (input) charge energy equals the output (discharge) energy. At this point, the system is exhibiting optimized behaviour. This was achieved using the structure illustrated in Table 3.

Results and discussion

The application of the equilibrium voltage in line with the MDP is utilized for the ageing and temperature effect analysis in line with established principles, which support the section on methods and materials. The following subsections discuss ageing and temperature investigations.

Table 3 Structure for cycle number and equilibrium voltage

Cycle number (i = 1,2,3….n)	Equilibrium charge/discharge ampere-hour (Ah)	Equilibrium voltage/V_{eq} (V)
1	$I_1 t_1$	V_1
2	$I_2 t_2$	V_2
3	…	…
…	…	…
n-1	$I_{n-1} t_{n-1}$	V_{n-1}
N	$I_n t_n$	V_n

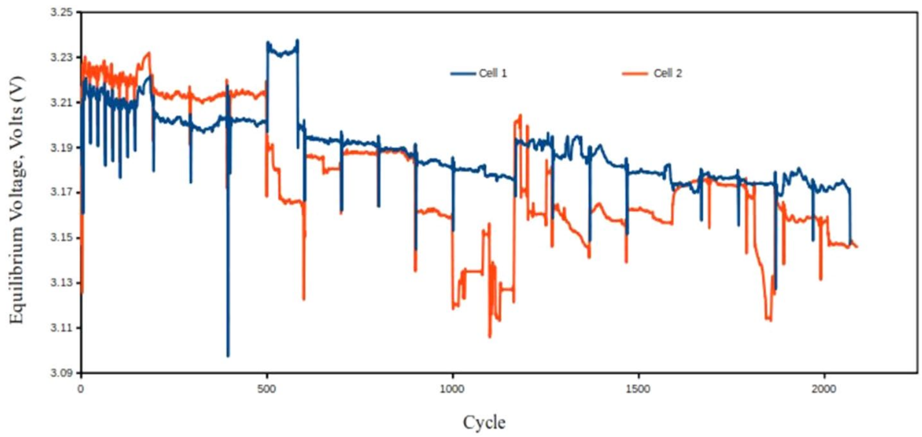


Fig. 1 Equilibrium voltage (vertical axis) against cycle number (horizontal axis) for $LiFePO_4$ Cells

Ageing investigation

The equilibrium voltages for 2069 and 2190 cycles for sample cell 1 and cell 2, respectively, are shown in Fig. 1. For cell 1, one obvious qualitative trend is the reduction in the equilibrium voltage as the number of cycles increased. Over 2069 cycles a maximum equilibrium voltage of 3.235644 V and a minimum of 3.09749 V were observed, giving a range of 0.13817 V. For cell 2, a maximum voltage of 3.23164 V and a minimum voltage of 3.10985 V were observed over 2190 cycles, yielding a range of approximately 0.12179 V. It is noteworthy that the range in both cases was quite close.

Observation of results for the first 100 cycles in Fig. 1 for both cell 1 and cell 2 reveals an almost exact pattern. This shows a definite behavioural relationship between cycle number (age) and the equilibrium voltage. A similar relationship was reported by [22] and [23] where the voltage in relation to amperage within a single cycle was monotonically decreasing for the discharge cycle and monotonically increasing for the charge cycle. The decrease in equilibrium voltage is not monotonic. Consequently, it captures unique phenomena associated with the cells, including the memory effect observed in lithium iron phosphate electrode, as discussed by [24]. [25] also reported a similar non-monotonic relationship between SoC and OCV, as well as between SoC and static stress. They could also be as a result of insufficient rest time in between cycling. For example, after a heavy drive of an electric vehicle, it is quickly recharged and put to use again without adequate rest time.

These dips appear to be regular which may also be indicative of a regular charging–discharging regime, which was the case in the datasets used. Dips have also been observed in similar batteries where voltage decrease in relation to time was attributed to the battery chemistry [26]. [27] also noted the presence of dips in lithium–sulphur batteries, attributed to solid nucleation processes.

Peaking activity for the first (see Fig. 2) and last (see Fig. 3) 100 cycles appears to be more regular, predictable when compared to the over 2000 cycles. This is indicative of early capacitive fading which is related to ageing [28, 29]. The equilibrium plots which reveal peaks have been reported to represent phase equilibria in both the anode and cathode and are indicative of ageing phenomena taking place in such cells [30]. These peaks and also valleys have been demonstrated to appear when estimating

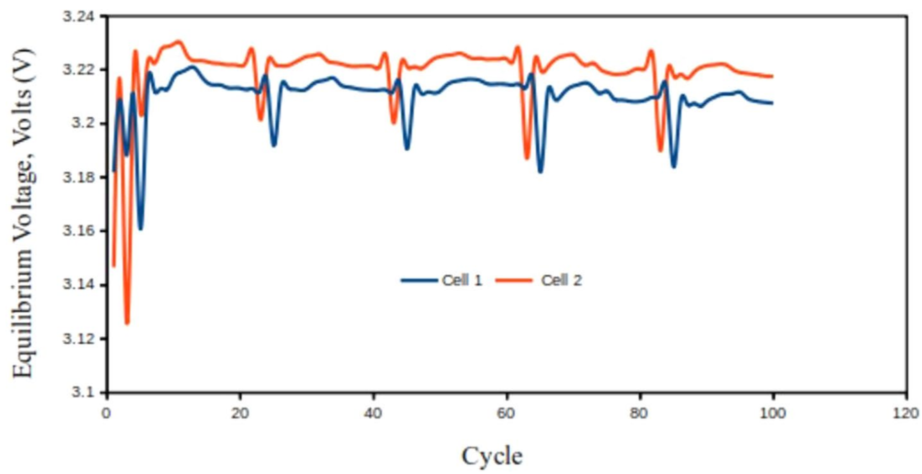


Fig. 2 The equilibrium voltage plotted against cycle number (first 100 cycles) for cells 1 and 2

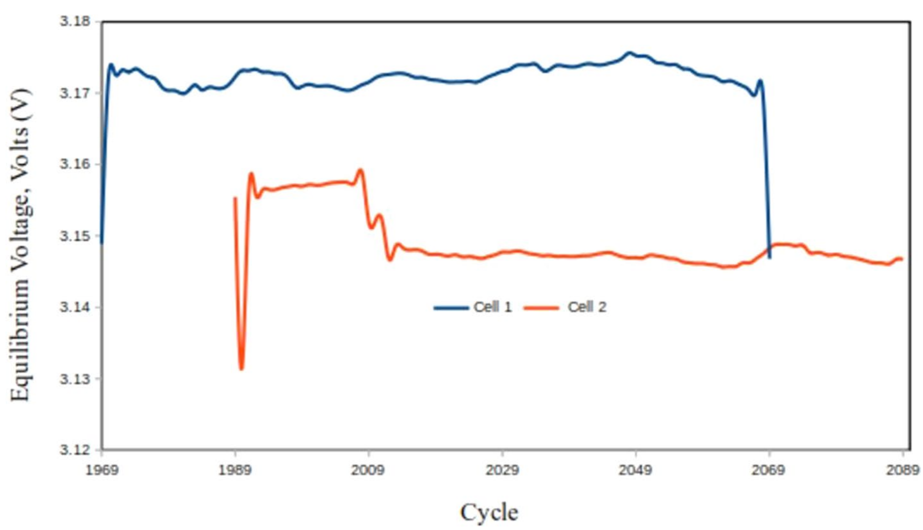


Fig. 3 The equilibrium voltage plotted against cycle number (last 100 cycles) for cells 1 and 2

battery capacity using incremental capacity analysis methods [31]. The low computational demands of the MDP equilibrium voltage approach are comparable to the ICA (incremental capacity analysis) methods as only two parameters are required: terminal voltage and current (see Table 1). The occurrence of dips also reveals a downward trend with a couple of outliers appearing in earlier cycles. Such may be due to sudden loading effects in the case of dips and rest effects in the case of peaks. It is also possible that sudden (but sustained) dips in loading may also result in spikes. From Fig. 1, a peak voltage of approximately 3.23 V and a lowest value of approximately 3.21 V were observed. [32] reported that an organic dual-ion potassium battery could deliver a discharge capacity of 60 mA g^{-1} with the median discharge voltage of 3.23 V. The minimum voltage of 3.21 V is the same value for the median voltage in sodium-doped, Lithium-rich manganese cathodes reported by [33] as indicative of the retention rate of such batteries. This underscores the MDP approach for tracking ageing phenomena

Table 4 Temperature data for cells 1 and 2

Temp. data	Equilibrium voltage, Volts (V)	Equilibrium Ah							
Cell	Temperature (°C)	Mean	std dev σ	Min	max	Mean	std dev σ	min	Max
1	− 25	3.29520	0.00011	3.29515	3.29546	683.86192	1.30568	682.30239	686.30519
1	− 10	3.27344	0.00013	3.27329	3.27360	82.15118	0.48871	81.62038	82.97814
1	0	3.28754	0.00011	3.28745	3.28775	217.12042	0.70888	216.04117	218.11452
1	10	3.29529	0.00016	3.29515	3.29545	469.97681	0.49789	469.64525	470.70775
1	20	3.30492	0.00012	3.30469	3.30500	448.72524	1.37806	446.47095	451.20646
1	30	3.30587	0.00012	3.30561	3.30592	487.77130	1.22137	485.75978	489.13828
1	40	3.30807	0.00000	3.30807	3.30807	554.31402	0.63751	553.42829	555.39232
1	50	3.30749	0.00010	3.30746	3.30777	579.16334	0.53226	578.07696	579.79858
2	− 25	3.30744	0.00008	3.30715	3.30746	754.11852	0.87570	752.98775	756.15357
2	− 10	3.27744	0.00016	3.27730	3.27761	93.05854	0.57996	92.24315	93.95024
2	0	3.29307	0.00012	3.29300	3.29331	249.82449	0.77423	248.72001	251.13931
2	10	3.29577	0.00000	3.29577	3.29577	537.87547	0.65576	536.80368	538.86946
2	20	3.31057	0.00008	3.31054	3.31085	515.50053	1.77543	512.48505	519.18224
2	30	3.31278	0.00012	3.31270	3.31300	560.87593	2.07754	556.80575	564.76037
2	40	3.31574	0.00009	3.31547	3.31577	639.73498	2.26134	634.74191	644.18320
2	50	3.31594	0.00014	3.31577	3.31608	700.40409	1.90252	697.24664	704.21593

in these batteries from equilibrium voltage analysis. Figure 3 shows a convergence of the last 100 cycles for both cells to just under 3.13 V reported to correspond to a capacity of 34.5% for a $LiFeO_4$ cathode battery [34]. This value also corresponds to the final Nyquist plot resistance of an EIS measurement for lithium-ion cells [35].

Temperature investigations

Temperature data were obtained for two (2) similar cells, processed and are displayed in Table 4.

The temperatures fell within the acceptable range of − 20 °C to 60 °C [36].The highest equilibrium voltage obtained was 3.3081 V (at 40 °C), while the lowest value was approximately 3.2733 V (at − 10 °C) giving a range of 0.035 V same value for the minimum cut-off charging voltage for galvanostatic electrochemical cycling reported by [37]. This appears to underscore the MDP equilibrium voltage as suitable for battery material characterization. As depicted in Fig. 4, the mean equilibrium voltages tracks the mean equilibrium ampere-hour very closely.

Considering earlier reports (for example [38]) on the temperature dependence of open-circuit voltage (OCV), a metric used to characterize SoC, an inference can be made that equilibrium voltages do track SoC.

This can be seen in Figs. 4 and 5. For Fig. 4, the lowest equilibrium voltage was observed at − 10 °C which corresponded approximately with the least balance Ah as shown in Fig. 4. This corroborates the tracking viability of the equilibrium voltage as system operating temperature varies. A very similar pattern is seen in Fig. 4 and is repeated in Fig. 5. Also it is observed that for Fig. 5, at about 15 °C, the amperage peaks and remains within about 5% till 35 °C. This is comparable to an earlier report by [39] for the cell's optimal temperature range of 15 °C to 35 °C. The cell's performance is very poor from − 15 °C to 8 °C. A very similar pattern is repeated in Fig. 5.

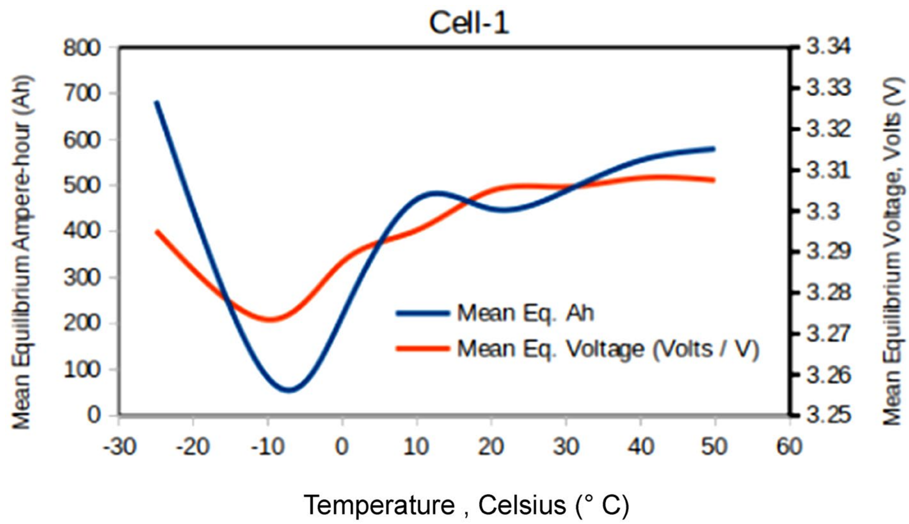


Fig. 4 Mean equilibrium voltage and ampere-hour against temperature for cell-1

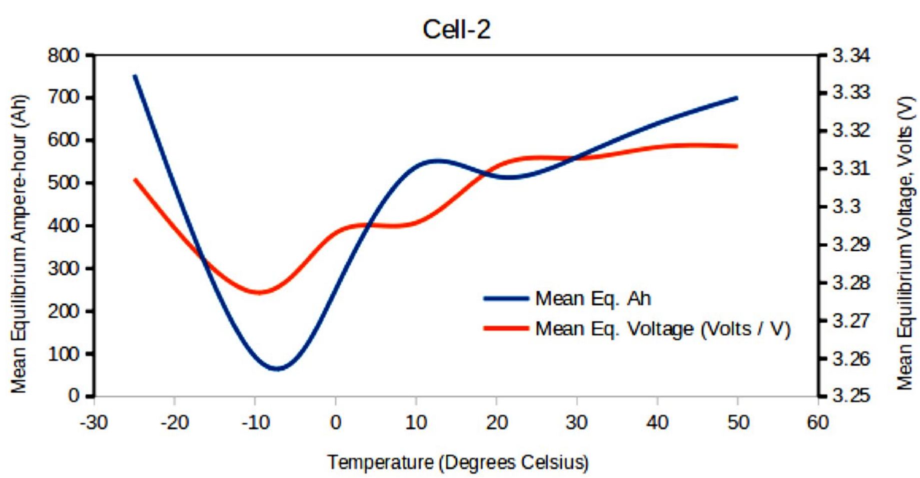


Fig. 5 Mean equilibrium voltage and ampere-hour against temperature for cell-2

Conclusions

A typical energy storage system or cell usually exhibits a recurrence of double transients in the form of charging and discharging cycles. This was suitably represented by the MDP as the activation–concentration equation (ACE), which tracks the resultant sequence of equilibrium voltages per cycle. Accordingly, this has been derived from a system linking concentration of radio incidence to radio geometry of surfaces or spheres of a material by Einstein's mass–energy equation; radio transmission by Maxwell's equations; radio emergence by Planck's theory of radiation; and activation of radio reception by ACE. The ageing and temperature profiles of the atmosphere of any material, device, circuit and/or system are therefore obtained from plotting the obtained sequence. Subsequently, a technique for the roundabout profiling of the effects of ageing and temperature of lithium-ion cells from readily available datasets was presented in this paper.

The usage of equilibrium voltage in the analysis from readily available datasets and consequently tracking ageing and temperature variations and their effects is a very cost-effective approach for battery management. It only takes samples of a few data points to start the trend. Once the equilibrium voltage for a cycle is obtained, further tracking within that cycle may not be required. An almost one-to-one tracking was observed in temperature variations for two identical cells. A highest equilibrium voltage of about 3.23 V was observed, while a minimum of about 3.10 V over 2000 cycles was obtained for ageing investigation. These values were linked to capacity fading, memory effects, etc., and established by the market definition paradigm (MDP) based on the quantitatively exhaustive principles of equality of radio incidence, modularity of radio geometry, linearity of radio transmission and polarity of radio emergence in cells and storage systems. This work is, therefore, essential for improving the performance, safety and durability of batteries, especially in applications like rockets, electric vehicles and renewable energy storage systems.

There is, however, a need for further insight using battery packs of various sizes of same and different chemistries. This will give insight as to how pack level dynamics differ from cell level, as supported in this study. In addition, there is a need to observe temperature variations about equilibrium points for both voltage and amperage adjustments.

Abbreviations

BoL	Begin of life
EoL	End of life
ICA	Incremental capacity analysis
KVL	Kirchhoff's voltage law
LiB	Lithium-ion battery
MDP	Market definition paradigm
OCV	Open-circuit voltage
SoC	State of charge

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Author contributions
Both authors contributed equally to the manuscript.

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Availability of data and materials
The datasets generated and/or analysed during the current study are available in the Center for Advanced Life Cycle Engineering Battery Data repository, https://calce.umd.edu/battery-data (K2 Battery Sample for Dataset 1 and Low-Current OCV (Samples 1 and 2) for Dataset 2).

Declarations

Competing interests
Authors have no conflict of interest relevant to this article.

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