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(Article begins on next page)

Thermal behaviour assessment and electrical characterization of a cylindrical Lithium-ion battery using infrared thermography

Luca Giammichele ^{a,*}, Valerio D'Alessandro ^a Matteo Falone ^a,
Renato Ricci ^a

^a*Dipartimento di Ingegneria Industriale e Scienze Matematiche
Università Politecnica delle Marche
Via Brecce Bianche 12, 60131 Ancona (AN), Italy*

1 Abstract

2 This paper presents an experimental evaluation of the thermal and electrical per-
3 formance of a commercial 26650 cylindrical LiFePO₄ battery cell. The thermal
4 management of lithium-ion batteries is a key problem for electric mobility appli-
5 cations, where batteries are subjected to severe operating conditions. Accordingly,
6 this study aims to demonstrate the reliability of infrared thermography in the
7 quantitative analysis of heat generation in battery cells. In our opinion, infrared
8 thermography can be very attractive owing to: (i) the non-contact nature of this
9 technique and (ii) its capability to quickly scan measurement areas. Consequently,
10 infrared thermography and thermocouple probe results were compared, providing
11 evidence of similar behaviour. Moreover, we present an electrical characterisation of
12 our lithium-ion battery. In particular, the cell potential, open circuit potential, and
13 entropic heat coefficient vis-à-vis the state of charge were experimentally measured.
14 The obtained experimental data were used to evaluate a simplified heat generation
15 term that is widely employed in numerical approaches. The different contributions
16 to heat generation were carefully analysed. The results show that the reversible
17 term considerably influences the total thermal power. Moreover, infrared-based heat
18 generation estimation can be considered reliable.

19 *Key words:* IR thermography, Lithium-ion batteries, Thermal management,
20 Electrochemical heat generation

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* Corresponding author.

Email addresses: l.giammichele@staff.univpm.it (Luca Giammichele),
v.dalessandro@univpm.it (Valerio D'Alessandro), m.falone@pm.univpm.it
(Matteo Falone), ricci@univpm.it (Renato Ricci).

22 Nomenclature

23 Acronyms

24 *BTMS* Battery Thermal Management System

25 *CCCV* Constant Current Constant Voltage

26 *EHC* Entropic Heat Coefficient

27 *EV* Electric Vehicles

28 *IR* Infrared

29 *SOC* State of Charge

30 Subscript

31 *e* Environment

32 *eod* End of discharge

33 *in* Initial value

34 *IR* Infrared thermography

35 *s* Surface

36 *tc* Thermocouple

37 Symbols

38 $\dot{Q}$ Thermal power [W]

39 $\dot{Q}_{irr}$ Irreversible thermal power [W]

40 $\dot{Q}_{rev}$ Reversible thermal power [W]

41 ϵ Emissivity

42 σ Stefan-Boltzmann constant [$5.67 \cdot 10^{-8} W/m^2 K^4$]

43 A Area of battery surface [m^2]

44 C Nominal capacity [Ah]

45 C_p Mean heat capacity at constant pressure [J/gK]

46 E_{acc} Total dispersed energy [J]

47 E_d Dispersed energy [J]

48 E_{in} Initial energy [J]

49 h Convection heat transfer coefficient [$W/m^2 K$]

50 I Charge/Discharge current [A]

51 m Cell mass [g]

52 T Temperature [K]

53 t Time [s]

54 T^* Dimensionless temperature

55 U_{OC} Open circuit potential [V]

56 V Cell potential [V]

57 V_{max} Maximum cell potential [V]

58 1 Introduction

59 Currently, sustainable mobility is a crucial topic in a wide range of energy-
60 saving and emission reduction scenarios. In this context, the development

61 and commercialisation of electric vehicles (EVs) is essential in minimising the
 62 environmental impact in our daily life [1, 2]. EVs commonly adopt lithium-ion
 63 (Li-ion) batteries as a power supply because of their higher energy density and
 64 specific power, lighter weight, lower self-discharge rates, higher recyclability,
 65 and longer cycle life than other rechargeable batteries [3–6].
 66 This technology has been widely improved during the years from an electro-
 67 chemical point of view; however, thermal management of Li-ion batteries is still
 68 an open challenge. They are inherently subject to ageing not only over time but
 69 also due to operating conditions, including their state of charge (SOC), deliv-
 70 ered/received current, and extreme operating temperatures. Temperature also
 71 has a certain influence on the performance degradation and lifetime of nearly
 72 all Li-ion cells when they are not operating in an appropriate temperature
 73 range ($20^{\circ}\text{C} - 40^{\circ}\text{C}$) [7, 8]. This is a crucial topic because several studies are
 74 devoted to the battery thermal management system (BTMS). In the existing
 75 studies, different cooling strategies have been proposed. The simplest method
 76 consists of air cooling systems, both in natural and forced convection, and is
 77 often supported by heat sinks/pin-fins [9, 10]. Liquid cooling strategies are more
 78 effective for EV applications because of their high thermal capacities. This
 79 technology exploits a single-phase or two-phase refrigerant [11–13], even with
 80 nano-fluid usage [14] that flows into a heat exchanger rounding the cells of the
 81 power module. In this configuration, despite its effectiveness, further thermal
 82 resistance is added owing to the presence of the heat exchanger. Moreover, in
 83 recent studies, the capability of innovative BTMS in controlling the thermal
 84 conditions of the power module, such as phase change materials [15, 16], heat
 85 pipes [17, 18], and pulsating heat pipes [19], have been investigated.
 86 In this scenario, it is very important to evaluate battery heat generation. An
 87 appropriate determination of single-cell heat generation improves the BTMS
 88 design method. In a battery cell, heat generation is related to the following
 89 mechanisms: activation of interfacial kinetics, concentration species transport,
 90 and ohmic Joule heating from the movement of charged particles, which be-
 91 comes significant for large cell sizes [20]. In the existing studies, battery heat
 92 generation has been separated into two main contributions: irreversible heat
 93 and reversible heat generation. The first term results from ohmic losses over
 94 the cell internal resistance, charge transfer overpotentials at the interface, and
 95 mass transfer limitations in the cell components. The second term is related
 96 to the entropic heat coefficient (EHC) which depends on the nature of the
 97 chemical reaction in the cell and the electrodes composition. In some previous
 98 studies, this reversible heat was neglected [21, 22] or considered constant with
 99 the SOC of the battery [23–25]. However, the entropic term can have a sub-
 100 stantial influence on heat generation. Moreover, it could have a value of the
 101 same magnitude of irreversible heat [26, 27]. Consequently, the battery surface
 102 temperature simulation is also significantly influenced by the correct evaluation
 103 of the reversible heat [28]. The most common method of measuring the EHC is
 104 the potentiometric test [26, 27, 29–31]. Thomas et al. [30] emphasised that it is
 105 more effective to vary the temperature at a specific SOC. Nieto et al. [26] stated

106 that the EHC is not influenced by the temperature chosen for the thermal
 107 cycle and if it is evaluated during charge or discharge. The low influence of
 108 different thermal cycles was also observed by Madani et al. [31].
 109 This study aims to apply IR thermography to quantitatively evaluate the
 110 heat generation of a cylindrical LiFePO_4 battery. Presently, some papers re-
 111 lated to IR thermography of Li-ion cells have been published. In particular,
 112 pouch or prismatic cells have been mainly studied [32–38]. Kim et al. [32]
 113 used IR thermography to evaluate the surface temperature of a pouch cell to
 114 validate the numerical model presented. Menale et al. [33] also applied this
 115 technique to evaluate the temperature distribution on the battery surface for
 116 different battery types and discharge conditions. They also used these data
 117 to qualitatively study the effects of battery ageing. Goutam et al. [34] used
 118 infrared thermography to detect the location of the hottest region for the
 119 large pouch and prismatic cells of different geometries and materials. Robinson
 120 et al. [35] applied lock-in thermography to investigate the effect of periodic
 121 electro-thermal stimulation on both new and aged Li-ion pouch cells. Rani
 122 et al. [36] measured the surface temperature of the battery using thermocou-
 123 ples and IR thermography. The two methods were compared considering the
 124 temperature behaviour during discharge and the maximum value. Bazinski et
 125 al. [37] applied IR thermography to quantitatively evaluate the thermal power
 126 generation of a large pouch cell. Wang et al. [38] studied the thermal behaviour
 127 of a Li-ion through IR thermography. The IR images were used to evaluate
 128 the temperature fluctuation and distribution by analysing the variance and a
 129 local overheating index proposed by the authors.
 130 Other studies have applied this technique to cylindrical batteries [29, 39–41].
 131 Forgez et al. [29] developed a lumped parameter thermal model to estimate
 132 the internal temperature by measuring the surface temperature using ther-
 133 mocouples. In this study, IR thermography was used to verify the uniformity
 134 of the surface temperature. Robinson et al. [39] combined thermal imaging,
 135 X-ray tomography, and electrochemical impedance spectroscopy to study the
 136 thermal characteristics of a commercial cylindrical battery. IR thermogra-
 137 phy was used to characterise the heterogeneous temperature distributions
 138 during discharge. Chiew et al. [40] presented a pseudo-three-dimensional
 139 electrochemical-thermal model of a Li-ion cylindrical battery. Experimental
 140 measurements were performed with both thermocouples and IR thermography
 141 to validate the numerical model considering the battery surface temperature
 142 distribution. Wang et al. [41] proposed a thermal inertia prediction model for
 143 a cylindrical battery validated using IR thermography measurements.
 144 Most of the reviewed papers applied infrared thermography to measure the
 145 surface temperature distribution. In this study, we used the experimental data
 146 in a simplified thermal model to estimate the thermal power generated. In our
 147 opinion, IR thermography can be considered a powerful method to evaluate
 148 battery heat generation with the advantage of being a fast and a non-intrusive
 149 measurement technique. To evaluate the reliability of IR thermography, our
 150 results were compared with thermocouple probe measurements. Furthermore,

151 electrical characterisation of the Li-ion battery was performed under a wide
 152 range of operating currents, and the cell potential during discharge, open
 153 circuit potential, and EHC were measured. These experimental data were used
 154 to calculate the battery heat generation using a simplified equation. Both
 155 irreversible and reversible heat terms were measured to carefully evaluate their
 156 contributions to the total heat generation.
 157 The remainder of this paper is organised as follows. Section 2 introduces the
 158 heat generation term equation. The experimental setup is described in Section
 159 3. Section 4 presents the results. Section 5 is devoted to the evaluation of the
 160 battery heat generation. Finally, the conclusions are presented in Section 6.

161 2 Heat generation model

162 Bernardi et al. [42] proposed a complete thermodynamic energy balance for
 163 Li-ion batteries. The energy balance can be expressed using Eq. (1). The
 164 energy accumulation in the cell is equal to the heat generated minus the heat
 165 loss through convection and radiation.

$$166 \quad mC_p \frac{dT}{dt} = \dot{Q} - hA(T_s - T_e) - \epsilon\sigma A(T_s^4 - T_e^4) \quad (1)$$

167 In present studies, a simplified equation, derived from Bernardi et al., is com-
 168 monly adopted to evaluate heat generation in Li-ion batteries [20, 22, 26, 27, 29,
 169 30, 43]:

$$170 \quad \dot{Q} = \underbrace{I \cdot (V - U_{OC})}_{\dot{Q}_{irr}} + \underbrace{I \cdot T \cdot \frac{\partial U_{OC}}{\partial T}}_{\dot{Q}_{rev}} \quad (2)$$

172 where I is the discharge current, V is the battery potential, T is the battery
 173 temperature (expressed in K), U_{OC} is the open circuit potential, and $\partial U_{OC}/\partial T$
 174 is the EHC.

175 The irreversible term of the equation depends on the cell overpotential, which
 176 is equal to the difference between V and U_{OC} and the operating current. This is
 177 indicative of all irreversible processes such as Joule heating within the battery
 178 owing to internal resistance and the energy dissipated in electrode overpo-
 179 tentials. In this study, the operating current was fixed to be negative during
 180 discharge. This implies that the irreversible contribution is always positive.

181 The reversible term in Eq. (2) is related to the entropy change in the electro-
 182 chemical reaction and is strongly influenced by the EHC. This coefficient varies
 183 significantly with the SOC and chemistry of the battery. It can be negative or
 184 positive; a negative value means that reversible heat is exothermic, whereas a
 185 positive value of EHC signifies that reversible heat is endothermic during the
 186 discharge process.

Eq. (2) allows evaluates the thermal power generated during the battery discharge. We can also determine the instantaneous energy dispersed in the form of heat generation:

$$E_d = \dot{Q} \cdot \Delta t \quad (3)$$

where Δt is the time interval at which the heat was generated. The total amount of dispersed energy can be expressed using Eq. (4), where τ is the time required to fully discharge the battery at the operating current.

$$E_{acc} = \int_0^\tau E_d dt \quad (4)$$

Finally, the battery efficiency could be defined as follows:

$$\eta = \frac{E_{in} - E_{acc}}{E_{in}} \quad (5)$$

where E_{in} is the initial battery energy assumed to be equal to the maximum electric energy that the battery can supply without any losses. Therefore, the numerator in Eq. (5) is the electric energy supplied by the battery. The ideal electric energy is defined using Eq. (6), where V_{max} is the battery voltage when fully charged, and C is the nominal capacity of the cell.

$$E_{in} = V_{max} \cdot C \quad (6)$$

In the above equations, the variables E_{in} , E_{acc} , and E_d are expressed in Joule.

3 Experimental apparatus

In this study, the thermal behaviour of a commercial LiFePO_4 battery was determined during the discharge process. The battery used in the experiments was A123 26650 (series name ANR26650M1-B). It is a cylindrical cell of diameter 26 mm, height 65 mm, and weight 76 g. The nominal battery voltage was 3.3 V and the maximum voltage when fully charged was 3.6 V. The cell capacity was 2.5 Ah with a maximum continuous discharge current of 70 A. The active materials of the positive and negative electrodes were LiFePO_4 and LiC_6 , respectively, and the electrolyte was LiPF_6 dissolved in a mixture of 2:1 ethylene carbonate/dimethyl carbonate. The current collectors were made of aluminium and copper.

The cell charge and discharge processes are managed by a programmable power supply (RMX-4125) and DC electronic load (RMX-4005) controlled through a LabVIEW routine. The current setting accuracy of the electronic load was $\pm 0.1\%$ F.S. ranging from 0 to 70 A with a resolution of 2 mA. The current and voltage of the battery were constantly measured during the entire process. The

221 voltage was directly measured using an NI 6289 data acquisition device with a
222 resolution of 0.076 mV ranging from 0 to 10 V and uncertainty of $\pm 0.25\text{ mV}$.
223 The operating current was measured using a current transducer with an uncer-
224 tainty of $\pm 0.02\text{ A}$ in the range $\pm 85\text{ A}$. Ambient parameters (temperature and
225 relative humidity) were measured with an uncertainty of $\pm 0.6^\circ\text{C}$ and $\pm 2.5\%$
226 for temperature and relative humidity, respectively. Transducer signals of the
227 current and ambient parameters were then acquired using the NI 6289 data
228 acquisition device.

229 Battery surface temperature measurements were performed using thermocou-
230 ples and IR thermography. Four T-type thermocouples were placed along the
231 height of the battery with a high-conductivity adhesive. The thermocouples
232 were attached to an ice point reference before being acquired by the data
233 logger. They were previously calibrated by submerging the battery into a high-
234 precision calibration bath. This procedure allows us to obtain an uncertainty of
235 $\pm 0.05^\circ\text{C}$. Thermographic measurements were performed using a FLIR SC3000
236 IR camera equipped with a 320×240 QWIP sensor array and a 20° lens with
237 a $20^\circ \times 15^\circ$ field of view with a minimum focus distance of 0.3 m . The thermal
238 sensitivity was 20 mK at 30°C , with an uncertainty of $\pm 1\%$ of the measured
239 value. The battery was covered with black matte paint with an emissivity of
240 0.94 , measured through a black body.

241 Fig. 1 shows the measurement setup described above and the thermocouple
242 arrangement on the battery surface.

243 During the tests, we performed the following measures: battery voltage, oper-
244 ating current, and temperature (using thermocouples) which were continuously
245 recorded at 10 Hz . Meanwhile, IR images were also acquired once per second.
246 The thermal camera was placed at a distance of 0.3 m from the battery, result-
247 ing in a spatial resolution of 0.3 mm per pixel. Each image obtained during
248 discharge was then subtracted from the reference thermal image related to the
249 initial condition. In addition, thermocouple measurements are referred to as
250 the initial value to avoid the influence of environmental temperature variation.
251 The ambient temperature and relative humidity were measured during the
252 tests. These values were used in the thermographic image acquisition soft-
253 ware as input parameters to perform temperature measurements. The ambient
254 temperature for all tests was between 21°C and 23°C . Thermal images were
255 processed using MATLAB. Four areas of 10×10 pixels were selected in the
256 same position of the four thermocouples along the battery height and in the
257 middle line of the battery width. The extension of these areas was chosen by
258 analysing the ratio between the view factor of the cylindrical surface and that
259 of a flat plate. In the reference areas, the ratio assumes a minimum value of
260 0.987 ; consequently, the error introduced by the view factor was considered
261 negligible in these areas. The mean temperature was calculated in the reference
262 areas to compare the two measurement methods.

263 Uncertainty analysis was performed according to the reference regulation.
264 In this study, we considered the instrument uncertainty for the parameters
265 directly acquired (i.e. thermocouples, cell potential, and current). Type A

uncertainty was considered in the case of mean values (i.e. U_{OC} and EHC). For correlated quantities (i.e. thermal power generated and total dispersed energy), the combined uncertainties were evaluated. Finally, a coverage factor of 2 was considered in the calculation of the expanded uncertainties. The obtained uncertainties are ± 0.25 mV for U_{OC} measurements and 0.3% for the SOC calculation. This last value is due to the setting current accuracy of the electronic load and the measurement uncertainty of the current sensor. In the evaluation of the EHC, we obtained an uncertainty of ± 0.001 mV/K. The measurement uncertainty of the correlated quantities is summarised in Table 1, where the largest percentage uncertainty for each C-rate is reported. The C-rate is the rate at which a battery is charged or discharged. It is defined as the ratio between the discharge current applied and the discharge current under which the battery delivers its nominal capacity per hour.

4 Experimental results

Eq. (2) requires several parameters to be determined to evaluate the battery heat generation. In this study, three types of tests were performed to experimentally determine these quantities. U_{OC} and EHC were evaluated every 10% of the SOC from full charge to complete discharge. The first with subsequent cycles of discharge and relaxation, and the second through a potentiometric test. Finally, a constant current discharge was applied to the battery to measure the cell potential, operating current, and surface temperature.

4.1 Open circuit potential

The open circuit potential was evaluated at every 10% of the SOC using the method described below. First, the battery was discharged at a specific SOC by a low operating current of 2.5 A. It was then allowed to relax for 1 h. At the end of the relaxation, the voltage value reached by the battery is the U_{OC} at a particular SOC. This test was repeated 5 times, and the U_{OC} value used in Eq. (2) is the mean value of the 5 tests; it is also shown in the plots in Fig. (2). The maximum percentage difference between each test and the mean value was very low (0.07%). This ensured the excellent repeatability of the measurements.

298 The battery was discharged to a specific SOC with a discharge current of
 299 2.5 A and was allowed to relax. The relaxation was followed by a thermal cycle
 300 in a thermostatic bath with a setting accuracy of $\pm 0.5^\circ\text{C}$. The open circuit
 301 potential of the battery was constantly measured during the bath temperature
 302 variation. The thermal cycle used in this study consisted of 3 h at 25°C , 3 h
 303 at 10°C , 3 h at 35°C , 3 h at 45°C , and 3 h at 25°C . Temperature values were
 304 chosen in the range where the battery operates safely (as recommended in the
 305 datasheet). This procedure was repeated every 10% of SOC until complete
 306 discharge. Fig. 3 shows an example of the open circuit potential variation
 307 during the thermal cycle. The voltage values measured in the last 15 min of
 308 each temperature step were considered U_{OC} steady-state values. The mean
 309 values of this temporal window are plotted as a function of temperature, as
 310 shown in Fig. 4. The gradient of the curve determines the EHC for each SOC.
 311 Finally, Fig. 5 shows the EHC measured as a function of SOC. The EHC
 312 assumes positive values until an SOC of 30%; subsequently, it becomes negative.
 313 Similar behaviour was also observed in previous studies [29, 43]. Therefore, the
 314 reversible heat would be negative; thus, its effect is endothermic during the
 315 discharge process until 30% of SOC. Successively, as discharge proceeds, the
 316 reversible heat change its behaviour and it exhibits an exothermic effect, and
 317 it starts to assume positive values.

318 4.3 Constant current discharge

319 In this test, the battery was fully discharged with a constant operating current.
 320 The discharge was stopped when the cell potential reached a minimum voltage
 321 of 2 V. The test was performed at ambient temperature and under a wide
 322 range of C-rates (Table 2). After the complete discharge, the battery is fully
 323 re-charged with a constant current constant voltage procedure: first, a constant
 324 current of 1 C was applied until the battery reaches the maximum voltage
 325 of 3.6 V (CC phase); subsequently, the voltage was kept constant until the
 326 operating current decreases to 25 mA (CV phase).
 327 The cell potential at different discharge rates is reported as a function of the
 328 SOC in Fig. 6. When a current is required from the battery, the cell potential
 329 has a sudden fall owing to its internal resistance, and it flattens until a very
 330 low SOC when the voltage falls to its minimum value. For a higher discharge
 331 current, the voltage value of the flattened zone always decreases, and the
 332 difference with the open circuit potential rapidly increases. When the cell
 333 potential decreases, the surface temperature of the battery increases, especially
 334 at low SOC. The temperature rise results from the loss of the initial energy as
 335 heat generated by the battery. Fig. 7 shows the thermal images acquired for

the discharge rates tested. For each discharge rate, 4 SOC levels were chosen as representative points for the discharge: 75%, 50%, 25%, and 0%. The images show the difference between the temperature value measured during discharge and its initial value (before the test starts). The ambient temperature, relative humidity, and distance from the battery surface were considered in the infrared image acquisitions. The colour scale applied to the images was always the same for the tests to appreciate the temperature increase with the C-rate. Thermal images underline the warm-up of the battery during discharge. It is clear that a higher current produces a higher temperature rise. The temperature reached by the battery at the end of discharge is moderate for lower C-rates (approximately 10°C greater ambient temperature) but becomes very high when higher C-rates occur. In these cases, the battery far exceeds its operating temperature range. For a discharge current of 10 C, the final temperature was 45°C higher than the initial temperature. Furthermore, for all C-rates, the battery surface temperature was not uniform across its height; however, there was a greater temperature in the upper zone at the negative electrode. This trend is also confirmed using thermocouple measurements, as shown in Fig. 8, where only a case of 2 C is reported for compactness. This effect can be attributed to natural convection on the battery surface. On the bottom side, there is a higher heat transfer, while proceeding upwards, the surrounding air is influenced by the surface temperature and the heat transfer decreases. Finally, Fig. 9 shows the temperature trend measured by the thermocouples and IR thermography for all the discharge currents tested. The IR thermography results, illustrated in Fig. 9, were obtained from an averaging procedure involving the four reference areas described in Section 3. Fig. 9 shows that the temperature rises with greater intensity at a low SOC when the voltage sharply decreases. Furthermore, IR thermography perfectly copies the thermocouple data and the temperature trend is in good agreement. We also introduced a percentage difference between the mean temperatures measured by thermocouples and IR thermography, expressed using Eq. (7):

$$\delta = 100 \cdot \left| \frac{T_{tc} - T_{IR}}{(T_{eod} - T_{in})_{tc}} \right| \quad (7)$$

where the subscript "tc" refers to thermocouple data and "IR" to thermography. T_{eod} is the temperature at the end of discharge, and T_{in} is the initial value. This parameter was calculated for each acquisition during the discharge; however, in Table 3, only the maximum value of δ for each C-rate was reported for compactness. The maximum absolute temperature difference recorded between the two measurement methods is also reported in Table 3. This analysis confirmed that the IR thermography temperature values were close to the thermocouple measurements. The absolute temperature difference is always less than 1°C. The parameter δ shows the error of IR thermography compared to the range of temperature measured during discharge. The higher values correspond to a low C-rate, where the increase in surface temperature

378 is small.

379 5 Determination of the heat generated

380 Once the open circuit potential and EHC were measured, the thermal power
381 generated can be determined using Eq. (2) for all constant-current discharges.
382 In Eq. (2), we use the surface temperature measured using thermocouples and
383 infrared thermography. We considered a uniform temperature inside the battery
384 because the Biot number assumes a value lower than 0.1; this could be a good
385 approximation for a low C-rate, whereas, for a high discharge current, the error
386 in the absolute thermal power value can be larger. This measurement error was
387 estimated by considering the temperature difference between the surface and
388 the internal part of the battery at 15°C experimentally measured by Forgez
389 et al. for a discharge current of 20 A [29]. Thermal power was also evaluated
390 with this increased temperature, and a maximum percentage difference of 1%
391 was found.

392 Fig. 10 shows the instantaneous thermal power at each SOC and the accu-
393 cumulated heat through the discharge process for all C-rates tested. Because
394 the overpotential increases with the discharge current, the thermal power and
395 accumulated heat are also greater at higher C-rates. Furthermore, thermal
396 power has a faster increase at low SOC, where the battery voltage has an
397 abrupt decay and the EHC becomes negative. In this zone, the reversible
398 thermal power is positive, and it increases the generated total thermal power.
399 Otherwise, when EHC reaches its maximum positive value (approximately
400 SOC equal to 60%), the thermal power shows a minimum for all discharge
401 currents. From the above considerations, it can be understood that the thermal
402 power generated by the battery is significantly influenced by the reversible
403 part. The heat accumulated during all discharges was compared to the initial
404 energy available to obtain the battery efficiency, as expressed in Eq. (5). Table 4
405 shows the efficiency for all discharge currents and the electric power supply
406 by the battery. Similarly, it can be noticed that at higher discharge currents,
407 the efficiency decreases owing to the greater overpotential and increase in the
408 battery temperature. The battery supplies a greater electric power at higher
409 C-rates; however, the efficiency is low, owing to the increase in thermal power
410 generated.

411 The two contributions of the total thermal power generated were analysed
412 individually. Fig. 11 shows Q_{irr}/Q_{tot} and Q_{rev}/Q_{tot} as functions of SOC at
413 C-rates of 0.5 C, 2 C, 5 C, and 10 C.

414 The reversible heat considerably influences the total heat generated by the
415 battery, especially at low C-rates, where it has the same order of magnitude
416 as the irreversible heat. It can also be seen that when the EHC is positive,
417 the reversible heat is negative, and the discharge process is endothermic. In

418 this case, the reversible part was subtracted from the total heat generation.
 419 Otherwise, from an SOC of 20%, the EHC becomes negative, the discharge
 420 process is exothermic, and the reversible component increases the total thermal
 421 power generation. As shown in Fig. 11, if only the irreversible part of heat
 422 generation is considered, the total heat would be widely overestimated at low
 423 C-rates. The error in the heat evaluation became less significant at higher
 424 C-rates. In these cases, the reversible term can be considered negligible because
 425 its value is much lower than the irreversible thermal power. Furthermore, it
 426 is assumed that a constant value of EHC is not a suitable choice because the
 427 contribution of the reversible term to the total heat is significantly influenced
 428 by the SOC.

429 However, at higher discharge rates, the reversible contribution is always less
 430 significant. At higher C-rates, the overpotential is very large, and the irre-
 431 versible term in Eq. (2) becomes predominant. The increase in overpotential
 432 with the C-rate resulted from the ohmic loss on the internal resistance of the
 433 battery.

434 The reversible term also influenced the surface distribution of the battery
 435 temperature during the discharge process. This is seen in Fig. 12, where a
 436 dimensionless temperature is shown for all C-rates as a function of the SOC.
 437 The dimensionless temperature is defined using Eq. (8):

$$438 \quad T^* = \frac{T - T_{in}}{T_{eod} - T_{in}} \quad (8)$$

439 Fig. 12 shows that T^* assumes an almost linear trend for a current of 10 C
 440 where the overpotential has a very large value. Here, the reversible term in
 441 Eq. (2) assumes a very low value when compared with the irreversible term,
 442 and the thermal power is mostly influenced by these components. When the
 443 discharge current is decreased, this linearity is always less significant because
 444 the EHC becomes relevant with respect to the overpotential. Furthermore, all
 445 the lines have a unique point of intersection (except for 0.5C) at approximately
 446 60% of the SOC when the EHC reaches its maximum value. After this point,
 447 the temperature is higher for higher C-rates, as shown in Fig. 9; conversely,
 448 before the intersection point, the trend is almost the opposite. This is an
 449 interesting result obtained from the dimensionless temperature plot; however,
 450 further tests on different battery models and chemistry compositions should
 451 be performed to confirm this behaviour.

452 Finally, the aforementioned estimations were also applied to the IR thermog-
 453 raphy data. From Table 3, it can be observed that the thermography and
 454 thermocouple data are comparable. This evidence also produces a similar
 455 prediction of the reversible heat and total thermal power. The error made by
 456 thermography on reversible heat calculation is approximately 0.3%, whereas
 457 that of the thermal power is less than 0.1% for all discharge currents, as shown
 458 in Table 5.

459 6 Conclusion

460 In this study, the thermal behaviour of a LiFePO_4 Li-ion battery under
461 various discharge currents was experimentally investigated. Moreover, electrical
462 characterisation of the cell was performed to obtain the open circuit potential
463 and EHC. Heat generation was evaluated using a simplified equation that
464 considers both irreversible and reversible contributions. The thermal power
465 estimation was based on battery surface temperature measurements performed
466 with thermocouples and IR thermography.

467 The results show an increase in the thermal power when the battery is subjected
468 to higher discharge currents. This is due to the increase in overpotential, which
469 is related to a greater battery voltage drop. Consequently, the battery efficiency
470 decreased with higher C-rates. At low SOC, the greatest thermal power was
471 observed because of the abrupt decay of voltage and the negative value of
472 EHC. This also leads to a sharp increase in temperature which reaches very
473 high values over the maximum recommended operating temperature. The
474 contribution of the reversible and irreversible heat to the total thermal power
475 was carefully evaluated. The reversible part cannot be assumed to be constant
476 or not considered at all, because it significantly influences the total thermal
477 power and temperature trend, especially at low C-rates. IR thermography
478 and thermocouple probe measurements showed that the surface temperature
479 was not uniform along the height of the battery. There was a greater warm-
480 up in the upper zone. The temperature measurements performed with IR
481 thermography were in good agreement with the thermocouple results. It has
482 been shown that the temperature trend during discharge is the same for both
483 methods. Furthermore, the maximum deviation of thermography data from
484 the thermocouples is always less than 1°C . These differences lead to an error
485 of less than 0.1% in thermal power evaluation.

486 The obtained results show that IR thermography allows an excellent estimation
487 of thermal power with values very close to the thermocouple data. Moreover,
488 IR approaches can quickly scan the measurement area and are non-intrusive
489 methods of analysis. In contrast, thermocouples must be placed on the battery
490 surface with a special highly conductive adhesive, and they have a more
491 complex acquisition system. Therefore, in our opinion, IR thermography can
492 be considered a powerful method, not only to qualitatively evaluate thermal
493 behaviour but to also perform a quantitative assessment of the heat generated
494 by the Li-ion battery.

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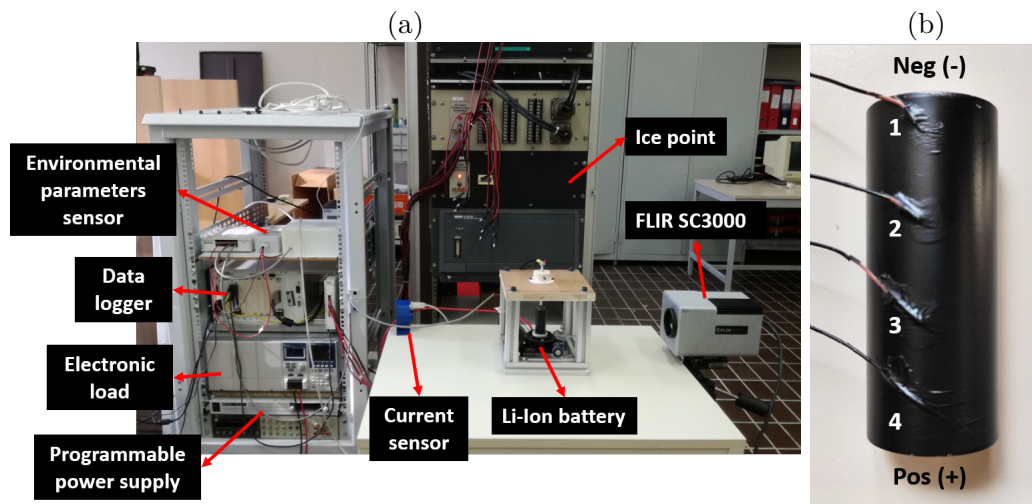


Figure 1. Experimental setup (a) and thermocouples arrangement on battery surface (b).

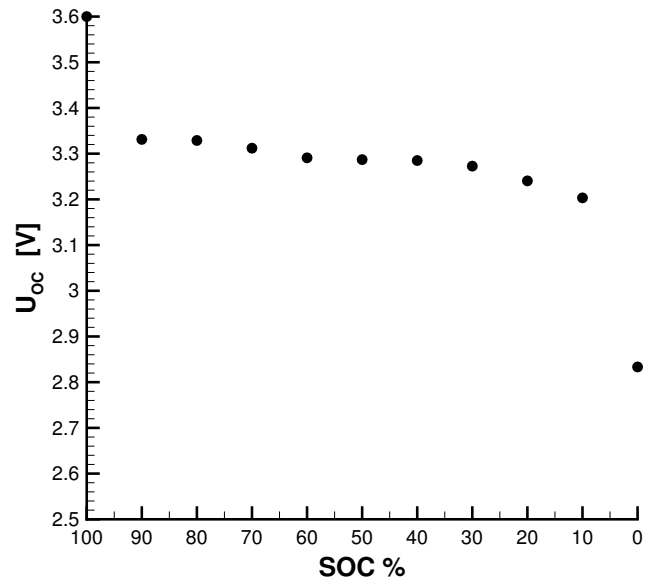


Figure 2. Open circuit potential of the battery as a function of SOC.

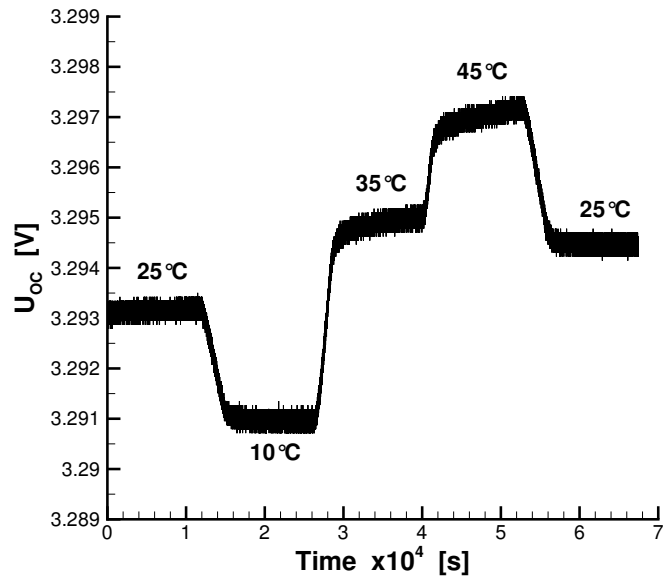


Figure 3. Open circuit potential measured during thermal cycle at SOC 50%.

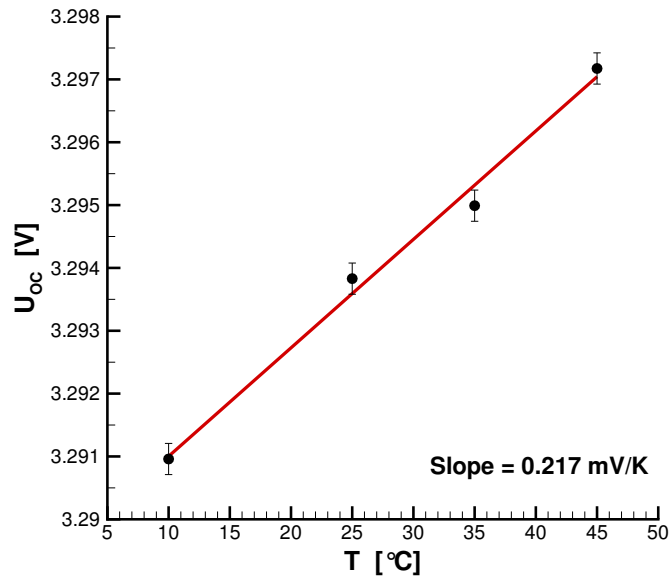


Figure 4. U_{OC} as a function of temperature at SOC 50%.

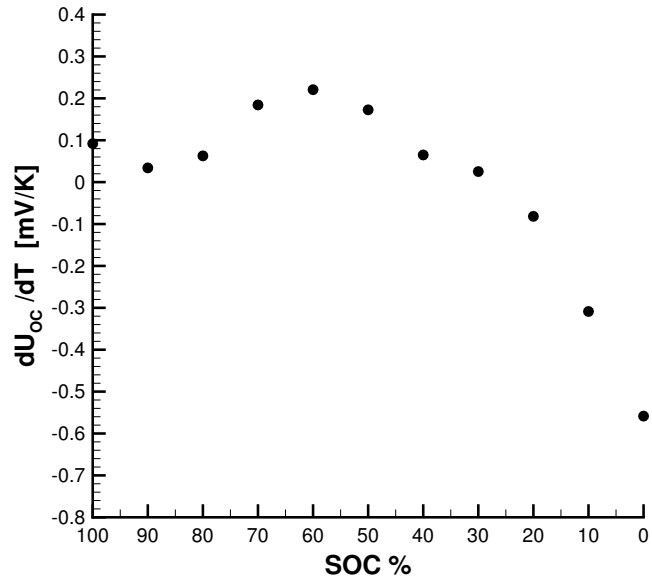


Figure 5. Entropic heat coefficient as a function of SOC.

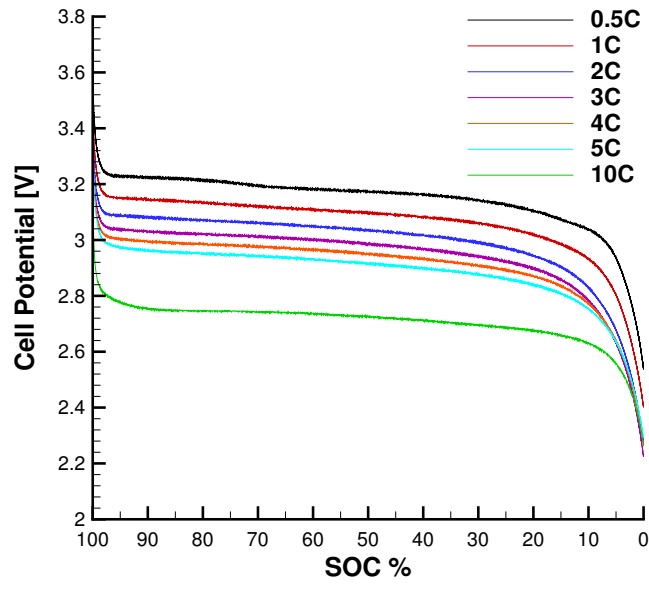


Figure 6. Cell potential as a function of SOC for different C-rate.

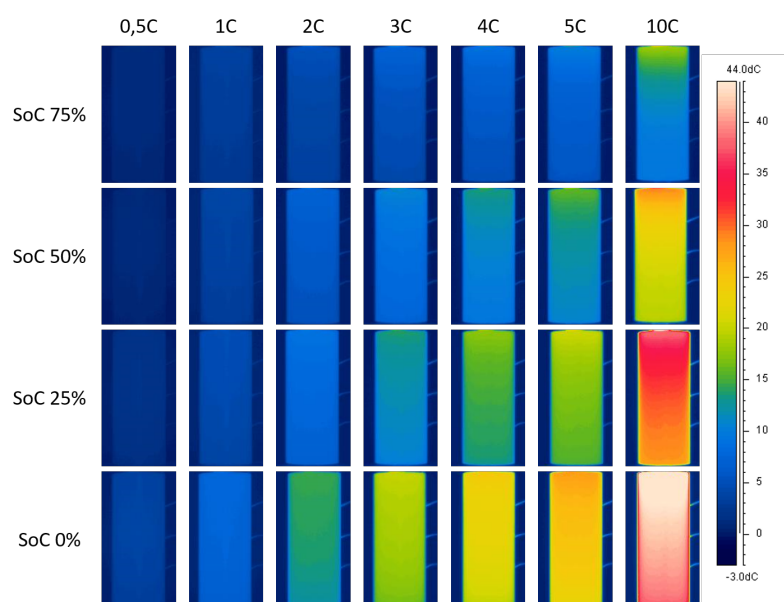


Figure 7. Thermal images of the battery at various discharge rates and SOC levels.

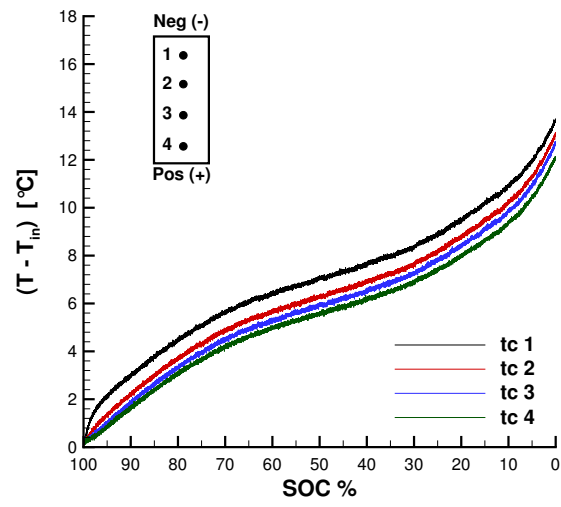


Figure 8. Temperature measured using the four thermocouples for a discharge current of 2 C.

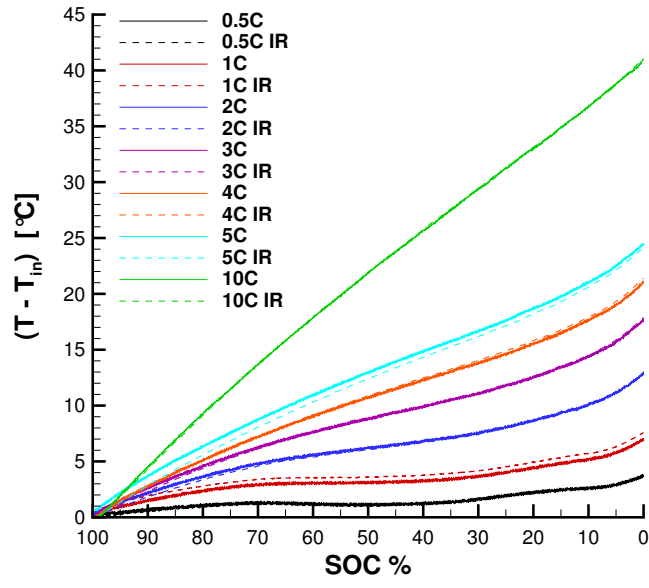


Figure 9. Battery surface temperature for different C-rate measured using thermocouples (solid line) and IR thermography (dashed line).

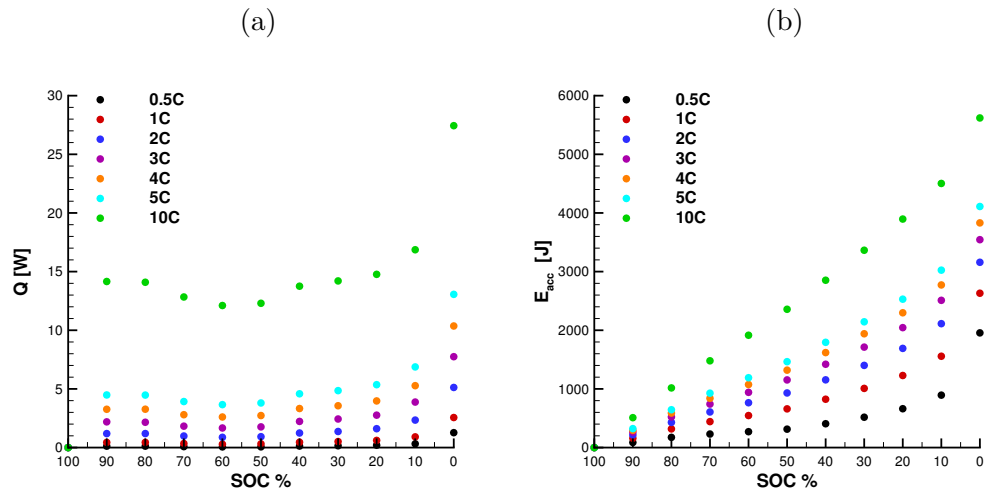


Figure 10. Thermal power generated (a) and accumulated heat (b) at various discharge currents.

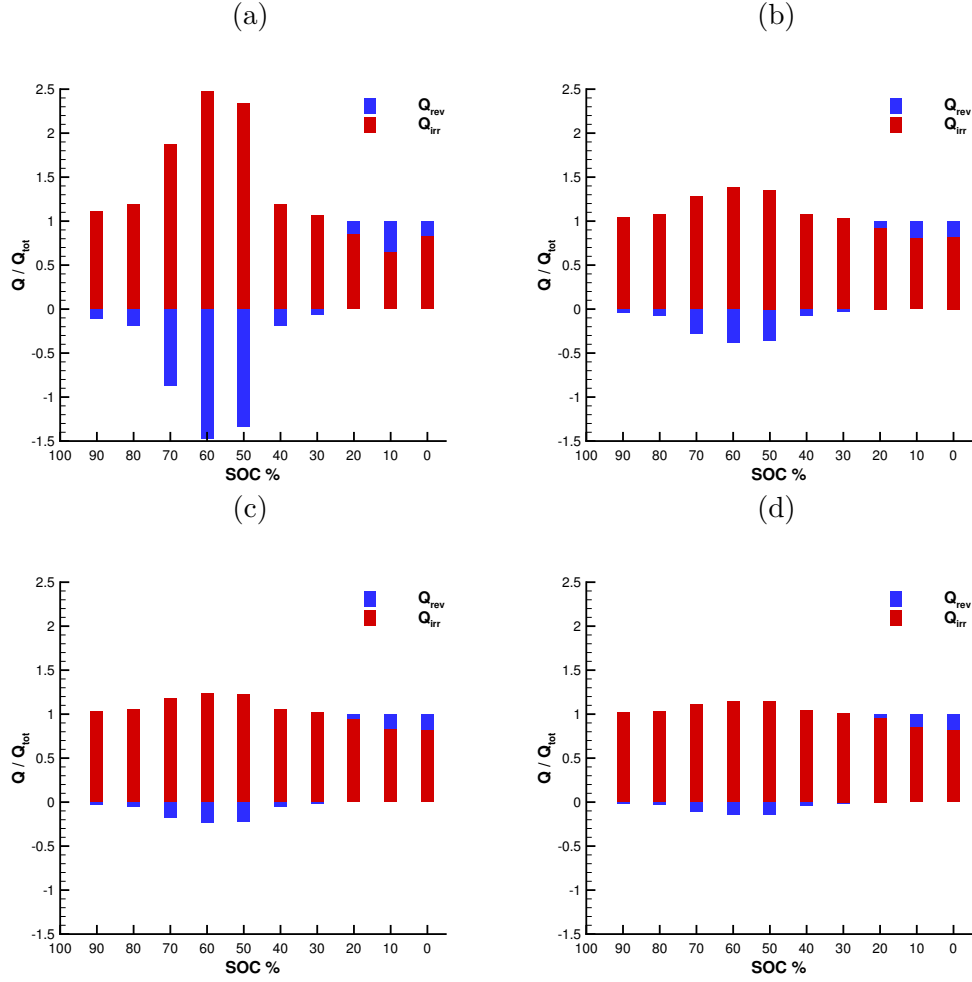


Figure 11. Contributions of the irreversible and reversible parts to the total thermal power generated as a function of SOC for a discharge current of 0.5 C (a), 2 C (b), 5 C (c), and 10 C (d).

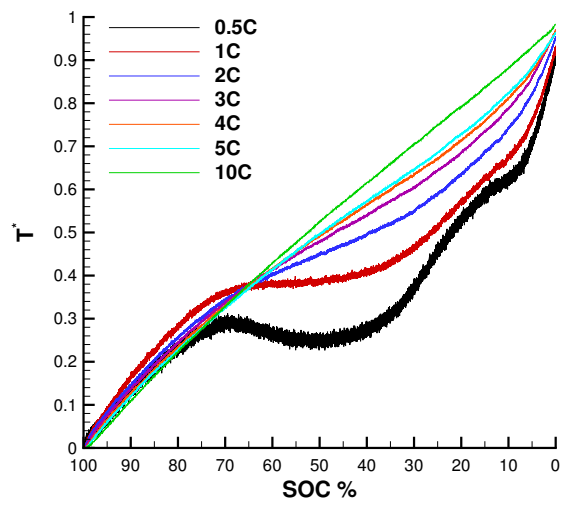


Figure 12. Dimensionless temperature for different C-rates.

Table 1
Measurement uncertainty.

C-rate	0.5	1	2	3	4	5	10
$\dot{Q}_{irr}\%$	± 0.69	± 0.37	± 0.24	± 0.19	± 0.16	± 0.14	± 0.079
$\dot{Q}_{rev}\%$	± 0.55	± 0.39	± 0.35	± 0.35	± 0.38	± 0.41	± 0.51
$\dot{Q}\%$	± 1.84	± 0.6	± 0.34	± 0.24	± 0.2	± 0.17	± 0.09
$E_{acc}\%$	± 1.7	± 0.71	± 0.45	± 0.34	± 0.28	± 0.25	± 0.14

Table 2
Constant current discharge tests performed.

C-rate	0.5	1	2	3	4	5	10
Current [A]	1.25	2.5	5	7.5	10	12.5	25
Time [s]	7200	3600	1800	1200	900	720	360

Table 3

The absolute maximum difference between thermocouples and thermographic temperature measurements.

C-rate	0.5	1	2	3	4	5	10
ΔT [$^{\circ}C$]	0.25	0.74	0.47	0.38	0.46	0.88	0.3
δ %	6.51	10.28	3.57	2.1	2.1	3.58	0.72

Table 4

Battery efficiency.

C-rate	0.5	1	2	3	4	5	10
η	0.94	0.92	0.9	0.89	0.88	0.87	0.82
Q_{el} [W]	4.23	8.27	16.24	20.05	31.74	39.29	74.38

Table 5

Maximum percentage difference between IR thermography and thermocouples on heat calculations.

C-rate	0.5	1	2	3	4	5	10
$\dot{Q}\%$	0.075	0.047	0.062	0.034	0.068	0.071	0.083
$\dot{Q}_{rev}\%$	0.09	0.12	0.15	0.1	0.19	0.23	0.34