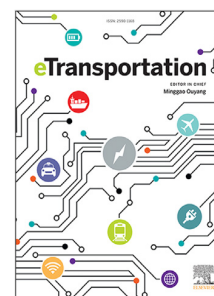


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Efficient physics-based modeling and experimental validation of parallel-connected battery cells enabled by the transmission line model

Pablo Rodríguez-Iturriaga, Salvador Rodríguez-Bolívar, Simona Onori, Juan Antonio López-Villanueva



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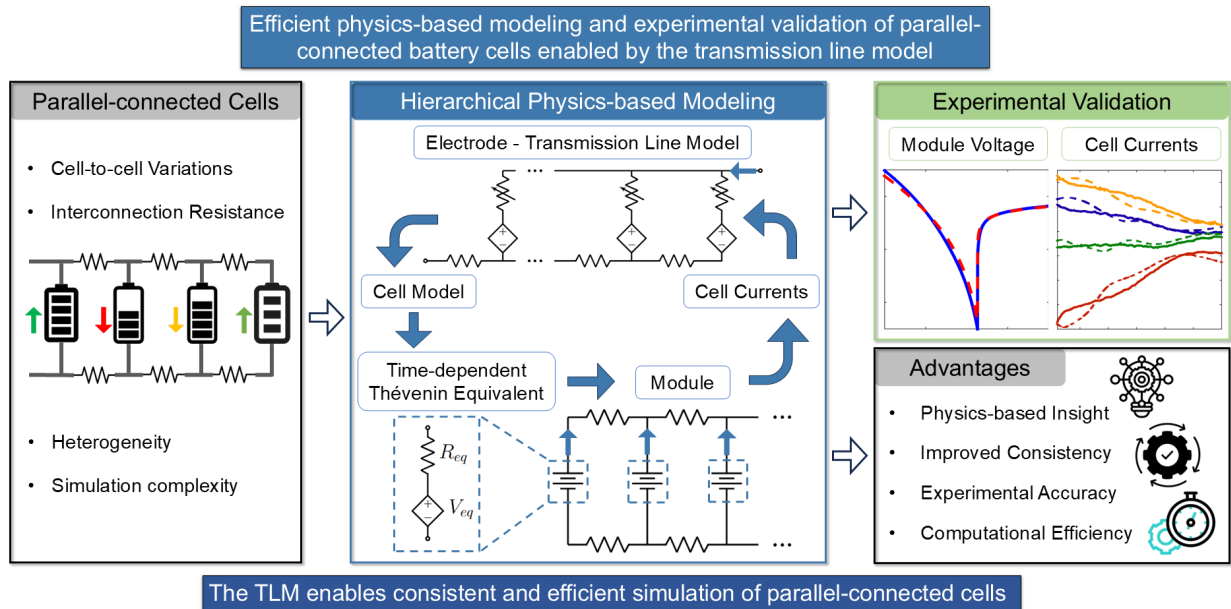
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## Graphical Abstract

## Efficient physics-based modeling and experimental validation of parallel-connected battery cells enabled by the transmission line model

Pablo Rodríguez-Iturriaga <sup>1</sup>, Salvador Rodríguez-Bolívar <sup>2</sup>, Simona Onori,  
Juan Antonio López-Villanueva



<sup>1</sup>Corresponding Author: priturriaga@ugr.es

<sup>2</sup>Corresponding Author: rbolivar@ugr.es

## Highlights

**Efficient physics-based modeling and experimental validation of parallel-connected battery cells enabled by the transmission line model**

Pablo Rodríguez-Iturriaga <sup>3</sup>, Salvador Rodríguez-Bolívar <sup>4</sup>, Simona Onori,  
Juan Antonio López-Villanueva

- Physically motivated efficient simulation framework for parallel-connected cells
- Determination of a cell's electrically consistent equivalent voltage and resistance
- Accurate results for module voltage ( $\leq 20\text{mV RMS}$ ) and current distribution ( $\leq 4.4\% \text{ RMS}$ )
- Experimental validation covering a wide range of operating scenarios
- Improved consistency and computational cost with respect to established approaches

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<sup>3</sup>Corresponding Author: priturriaga@ugr.es

<sup>4</sup>Corresponding Author: rbolivar@ugr.es

# Efficient physics-based modeling and experimental validation of parallel-connected battery cells enabled by the transmission line model

Pablo Rodríguez-Iturriaga<sup>1 a</sup>, Salvador Rodríguez-Bolívar<sup>2 a</sup>, Simona Onori<sup>b</sup>, Juan Antonio López-Villanueva<sup>a</sup>

<sup>a</sup>*Department of Electronics and Computer Technology, University of Granada, Granada, 18071, Andalusia, Spain*

<sup>b</sup>*Department of Energy Science and Engineering, Stanford University, Stanford, 94305, California, USA*

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## Abstract

Battery modules composed of parallel-connected cells are commonly used as building blocks of battery packs, but their behavior is complex due to cell dynamics, as well as cell-to-cell heterogeneities and interactions. Furthermore, their simulation by means of empirical equivalent circuit models poses limitations because of lack of generalization, whereas electrochemical models lead to a challenging calculation of the current distribution. In this article, an electrically consistent method for the calculation of the equivalent voltage and resistance of a cell is presented according to the physically motivated discrete transmission line model. This enables the efficient computation of output voltage and current distribution for parallel-connected cells while providing interpretable physical information about the operation at each level. The presented approach is validated experimentally against a dataset of a 4P

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<sup>1</sup>Corresponding Author: priturriaga@ugr.es

<sup>2</sup>Corresponding Author: rbolivar@ugr.es

module in which interconnection resistance, ambient temperature, and the presence of an aged cell are considered as input parameters, with accurate and consistent results for module voltage ( $\leq 20$  mV RMS) and current distribution ( $\leq 4.4\%$  RMS). Moreover, the proposed framework exhibits higher computational efficiency and comparable scalability in relation to established approaches, while providing improved consistency between module-level behavior and cell-level dynamics. Therefore, the proposed method based on the transmission line model and hierarchical simplification is a suitable alternative for the physically motivated simulation and analysis of battery modules.

*Keywords:* Lithium-ion battery, Parallel connection, Battery module, Transmission line model, Silicon-Graphite

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## 1. Introduction

Lithium-ion batteries (LIBs) play a central role in transportation electrification [1]. As a result of energy and power requirements, large battery packs comprising hundreds to thousands of individual cells are utilized [2, 3]. Battery strings composed of parallel-connected cells are commonly employed as the building blocks of large battery packs due to advantages in available capacity, reliability [4, 5] and robustness [6]. For this reason, the study of parallel-connected cells constitutes an essential line of research for the adequate management of battery modules and packs [7, 8]. Nevertheless, the modeling and analysis of the behavior of parallel-connected cells is more complex than that of individual cells due to non-uniform real-world operation caused by cell dynamics, as well as cell-to-cell heterogeneities and interactions [9].

14 A detailed overview of the origins of cell-level parameter variations, their  
15 cross-interaction, and their resulting effects is provided in [10]. At the cell  
16 level, tolerances and inconsistencies in the manufacturing processes lead to  
17 fluctuations in capacity, AC impedance, DC resistance, and rate capability  
18 among cells in the same batch [11, 12], whose experimental characterization  
19 has been the subject of study in previous works [13, 14]. At the module  
20 level, assembly choices are also a source of non-uniform cell behavior: non-  
21 negligible interconnection [15] and contact resistances [16] influence the cur-  
22 rent distribution and rate capability of the module [17], therefore making  
23 the connection topology a relevant factor as well [18, 19]. Furthermore, the  
24 design of the cooling and thermal management systems [20, 21] can result  
25 in heterogeneous heat transfer characteristics between cells [22, 23]. As a  
26 result, both cell-level and module-level effects, as well as their interrelation,  
27 cause imbalances in stress factors such as C-rate, depth of discharge and  
28 temperature increase [24, 25, 26, 27], which in turn may lead to uneven cell  
29 degradation [28] and decrease in module performance [29]. Therefore, the  
30 development of high-fidelity models for the efficient simulation of battery  
31 modules constitutes a crucial line of research for an improved and quanti-  
32 tative understanding of non-uniform behavior at the cell level as well as its  
33 impact on module performance.

34 Since a module composed of the interconnection of several battery cells  
35 can be represented by a higher-level circuit network, most works that at-  
36 tempt to recreate the operation of battery modules make use of equivalent  
37 circuit models (ECMs) to emulate cell behavior due to simpler integration  
38 into the former. In particular, ECMs composed of an Open Circuit Volt-

39 age (OCV) source and a series resistance, and optionally several parallel  
40 resistor-capacitor (RC) networks, have been widely employed for the analyt-  
41 ical calculation of the current distribution in ideal parallel-connected mod-  
42 ules by means of suitable mathematical transformations [30, 31], as well as  
43 its experimental validation in some instances for a 2-parallel (2P) system  
44 [32, 33, 34]. This approach may be extended for the development of state  
45 observers for the estimation of individual cell variables, such as state of charge  
46 (SOC) [35, 36, 37]; it is also the basis for the simulation of larger battery  
47 packs to analyze the influence of pack topology [38, 39], current imbalances  
48 [29, 40] or statistical parameter distribution [41, 42] on pack performance,  
49 as well as experimental validation at a larger scale [3, 16]. The modeling  
50 of temperature distribution and the effect of cooling system design are also  
51 considered in some instances [22]. Additionally, the effect of interconnection  
52 and contact resistances may easily be incorporated within this approach,  
53 whose impact on the current distribution [19] as well as the state of available  
54 power [43] have recently been emphasized. However, standard ECMs, despite  
55 acceptable accuracy within certain operating regimes, lack a connection to  
56 the underlying physical processes [44] and require empirical dependencies of  
57 electrical parameters with SOC, which are major factors influencing the cur-  
58 rent distribution along with the nonlinearities in the shape of the OCV curve  
59 [24, 33]. As a result, their generalization ability is limited by the span of the  
60 experimental data used to calibrate them [3], and thus, a more general and  
61 physically motivated modeling approach for the simulation and validation of  
62 battery modules is desirable.

63 In contrast, fewer approaches in the literature employ electrochemical

64 models for the analysis of the behavior of parallel-connected cells because of  
65 the challenge of interfacing them with the circuit network representing the  
66 module [45]. In this regard, a thermally coupled Single Particle Model (SPM)  
67 was embedded into MATLAB-Simulink for the simulation of a 6P module  
68 in [46], including the effect of the contact and interconnection resistances.  
69 In [47], Padé approximations were employed to simplify the transcendental  
70 transfer functions obtained from the SPM with electrolyte (SPMe), which  
71 were then integrated into a Simulink submodel for module simulation. In the  
72 two previous cases, the coupling between behaviors at cell and module lev-  
73 els was not implemented explicitly and was handled by the Simulink solver.  
74 Some authors have also suggested employing an electrochemical model for  
75 internal cell dynamics, but replacing it by an equivalent circuit for an ef-  
76 ficient calculation of module voltage and current distribution. In [48], a  
77 thermally coupled Doyle-Fuller-Newman (DFN) model is employed, but the  
78 current distribution is solved by substituting each cell by an equivalent circuit  
79 composed of an OCV voltage source in series with a polarization resistance  
80 calculated from simulated galvanostatic discharges. However, it was pointed  
81 out that this approach is not fully electrically consistent and is only adequate  
82 in steady state. The authors in [49] also employ a thermally coupled DFN  
83 model, solved in COMSOL, for the simulation of a 5P module including in-  
84 terconnection and contact resistances, with the coupling being implemented  
85 via MATLAB. In this case, the equivalent resistance is calculated from the  
86 difference between the terminal voltage given by the electrochemical model  
87 and the OCV corresponding to the SOC of each cell. A similar approach  
88 is implemented in `liionpack` [50], a software package for the simulation of

89 battery modules employing the cell models available in PyBaMM [51] within  
90 a circuit network symbolizing the module. Nevertheless, if the equivalent  
91 resistance is calculated as the ratio between cell overpotential and current,  
92 it is implicitly assumed that the former is fully ohmic in nature, which is  
93 not completely consistent with the internal dynamics of the electrochemical  
94 model and may overestimate instantaneous cell resistance. Furthermore, this  
95 approach leads to numerical instability during relaxation, since the resulting  
96 equivalent resistance may take unphysical values as currents approach zero  
97 or change direction during module rebalancing. For this reason, the determi-  
98 nation of an electrically consistent equivalent circuit from an electrochemical  
99 model remains an open question.

100 In summary, the following gaps are identified in the literature regarding  
101 the simulation and experimental validation of parallel-connected cells:

- 102 • Most of the literature on the modeling and analysis of parallel-connected  
103 cells has employed empirical ECMs, with interconnection and contact  
104 resistances often being omitted.
- 105 • Electrochemical models lead to a complex computation of the current  
106 distribution, and the method for calculating their equivalent circuit in  
107 an electrically consistent manner has not been adequately addressed.
- 108 • The experimental validation of module voltage has received substan-  
109 tially less attention than that of the current distribution.

110 For the reasons above, an accurate physics-based cell model is required,  
111 which allows for a consistent computation of its equivalent voltage and resis-  
112 tance for an efficient simulation of the parallel connection of several cells.

113 In a previous work [52], we developed a physics-based ECM for a com-  
114 mercial Nickel-Maganese-Cobalt/Silicon-Graphite (NMC/Si-Gr) cell based  
115 on the transmission line model (TLM), with the key difference of a discrete  
116 instead of continuous spatial distribution of active material particles within  
117 the electrode. We showed that an optimal number of particles, along with a  
118 macroscale ohmic resistance and a particle size distribution for graphite, were  
119 required to reproduce cell voltage at increasing C-rates as well as the subse-  
120 quent relaxation with a single set of parameters. In this article, we introduce  
121 the hierarchical procedure to integrate this model within a parallel module  
122 for an efficient simulation and experimental validation of module behavior in  
123 a variety of operating scenarios. To the best of our knowledge, this is the  
124 first time the circuit formulation of the TLM has been leveraged for the elec-  
125 trically consistent calculation of equivalent voltage and resistances, leading  
126 to an efficient computation of module voltage and current distribution.

127 The main contributions of this article are summarized as follows:

- 128 • A novel hierarchical simulation framework for parallel-connected cells  
129 is presented based on the physically motivated discrete transmission  
130 line cell model and the calculation of electrically consistent values of  
131 equivalent voltages and resistances.
- 132 • Module voltage and current distribution are simultaneously validated  
133 with high accuracy against an experimental dataset in which the inter-  
134 connection resistance, ambient temperature, and the presence or ab-  
135 sence of an aged cell are considered as input parameters.
- 136 • The presented methodology is assessed in relation to established ap-

137       proaches for the physically motivated simulation of battery modules,  
138       as well as the usage of traditional ECMs for this purpose.

139       The outline of this paper is the following: the employed cell model is  
140       described in Section 2, and the procedure for module simulation is presented  
141       in Section 3. The proposed approach is validated and discussed in Section  
142       5 with the aid of the experimental dataset described in 4. Some concluding  
143       remarks are provided in Section 6.

## 144   2. Transmission line cell model

145       In a previous work [52], we developed and validated a physics-based equiv-  
146       alent circuit model ECM for a NMC/Si-Gr cell following the TLM approach  
147       (see Fig. 1). The application of TLMs to porous electrode modeling was first  
148       presented by De Levie as a method for the description of their impedance  
149       response [53], on which several authors have subsequently elaborated [54, 55].  
150       The TLM approach may also be employed for time-domain simulations as a  
151       physically motivated equivalent circuit of the standard DFN model [56, 57],  
152       as demonstrated in [58, 44]. As the authors in [44] provide a detailed deriva-  
153       tion of the electrical equivalence of each physical process, herein only a brief  
154       overview is included, along with the key differences with their approach.

155       • **Solid diffusion.** The auxiliary subcircuits in Fig. 1-b) model the solid  
156       diffusion process within active material particles by approximating the  
157       transfer function associated with this process (see Eq. 1) by electrical  
158       elements and providing both average and surface concentrations, thus  
159       reducing the number of states required. The surface concentrations are

160 used to evaluate the Open Circuit Potential (OCP) function, repre-  
 161 sented by controlled voltage sources in Fig. 1-a). It is noted that there  
 162 is one subcircuit per spatial node, so intercalation currents are local.  
 163 This process is determined by electrode capacities  $Q_P, Q_N$  and time  
 164 constants  $\tau_{d,NMC}, \tau_{d,Si}, \tau_{d,Gr}$ . In the case of graphite,  $\tau_{d,Gr,ref}$  is multi-  
 165 plied by the ratio of particle sizes to obtain individual time constants.

- 166 • **Charge transfer.** Charge transfer overpotentials are modeled by non-  
 167 linear charge transfer resistances (see Eq. 3) depending on the local  
 168 intercalation current and the local exchange current (see Eq. 4). This  
 169 process is determined by the exchange currents  $K_{0,NMC}, K_{0,Si}, K_{0,Gr}$ .
- 170 • **Electronic transport in the solid.** This process is accounted for by  
 171 means of distributed resistors on the outer rails that model ohmic losses  
 172 in the solid [58, 44]. However, since electronic conductivity is usually  
 173 several orders of magnitude larger than ionic conductivity [59], its effect  
 174 may be safely neglected without compromising accuracy. As a result,  
 175 solid potential does not depend on spatial position, which facilitates  
 176 simulation as well as the application of circuit network simplifications.
- 177 • **Ionic transport in the electrolyte.** This process is commonly  
 178 split into two components: ohmic losses are modeled by distributed  
 179 resistances on the inner rails  $R_{ohm,PE}$  and  $R_{ohm,NE}$ , whereas several  
 180 approaches exist to account for ionic diffusion within this modeling  
 181 framework. Some authors include additional controlled voltage sources  
 182 to account for the overpotential due to concentration gradients [58, 44],  
 183 while others include a second transmission line on the electrolyte rail

184 [55]. In this work, all the electrolyte effects are combined in the ohmic  
185 resistances  $R_{ohm,PE}$  and  $R_{ohm,NE}$  due to the challenge posed by the sep-  
186 arate identification of these two mechanisms from experimental data.

187 The main distinction of the model in Fig. 1 from a standard TLM lies in  
188 a discrete spatial distribution described by a finite number of particles within  
189 each electrode  $N_{PE}, N_{NE}$  and a macroscale ohmic resistance  $R_{ohm}$ , instead  
190 of a continuous spatial structure with a large number of nodes  $N$  and a dis-  
191 tributed resistance  $R_{ohm}/N$  [55]. This assumption is based on the fact that a  
192 continuous spatial geometry may not be the most suitable description of elec-  
193 trode spatial structure [60], since active material particles are not microscale  
194 with respect to electrode thickness for this chemistry as reported in [59]. We  
195 have shown in previous works [61, 62] that a discrete spatial structure is also  
196 more consistent with EIS spectra, as the ohmic resistance is interpreted as  
197 the resistance between particles instead of a lumped element.

198 This modeling framework allows for the simplified inclusion of silicon  
199 particles, as well as a particle size distribution for graphite (3 sizes shown  
200 in Fig. 1), by defining additional branches in each spatial node. As a result  
201 of these considerations, the model is able to provide highly accurate results  
202 upon validation against experimental data at different C-rates with a single  
203 set of constant parameters. In particular, we showed that  $N_{PE} = 6, N_{NE} = 6$   
204 allows reproducing the slope of cell voltage during galvanostatic discharges  
205 due to increasingly sequential operation [52], while a particle size distribu-  
206 tion for graphite contributed decisively to matching relaxation profiles after  
207 discharge. This approach presents two key advantages: first, only electrical  
208 parameters are required instead of all the physical parameters of an electro-

209 chemical model, and second, the TLM formulation enables the application  
210 of circuit network simplifications, which enable an efficient hierarchical sim-  
211 ulation of parallel-connected cells.

212 Given that the profiles employed for experimental validation in this work  
213 are discharges followed by relaxation, we have employed delithiation OCPs  
214 for both silicon and graphite. The presented modeling framework also enables  
215 the separate simulation of module charging by making use of lithiation OCPs,  
216 as illustrated in Fig. S1. Nevertheless, the implementation of the cell-level  
217 switching between lithiation and delithiation OCPs in dynamic operating  
218 conditions at the module level poses a significant challenge due to inherent  
219 numerical instability and potentially inconsistent results. We have verified  
220 that a sigmoid function of instantaneous cell current, as in [63], results in nu-  
221 merical divergence at the module level in several common operating scenarios.  
222 We believe that the current understanding of hysteresis and present modeling  
223 alternatives are not sufficient to properly address dynamic operation at the  
224 module level while taking into account the transient component of cell-level  
225 hysteresis. However, we understand that this limitation is not exclusive to  
226 this modeling approach, and we identify this issue as the key challenge for a  
227 comprehensive understanding of the behavior of parallel-connected cells.

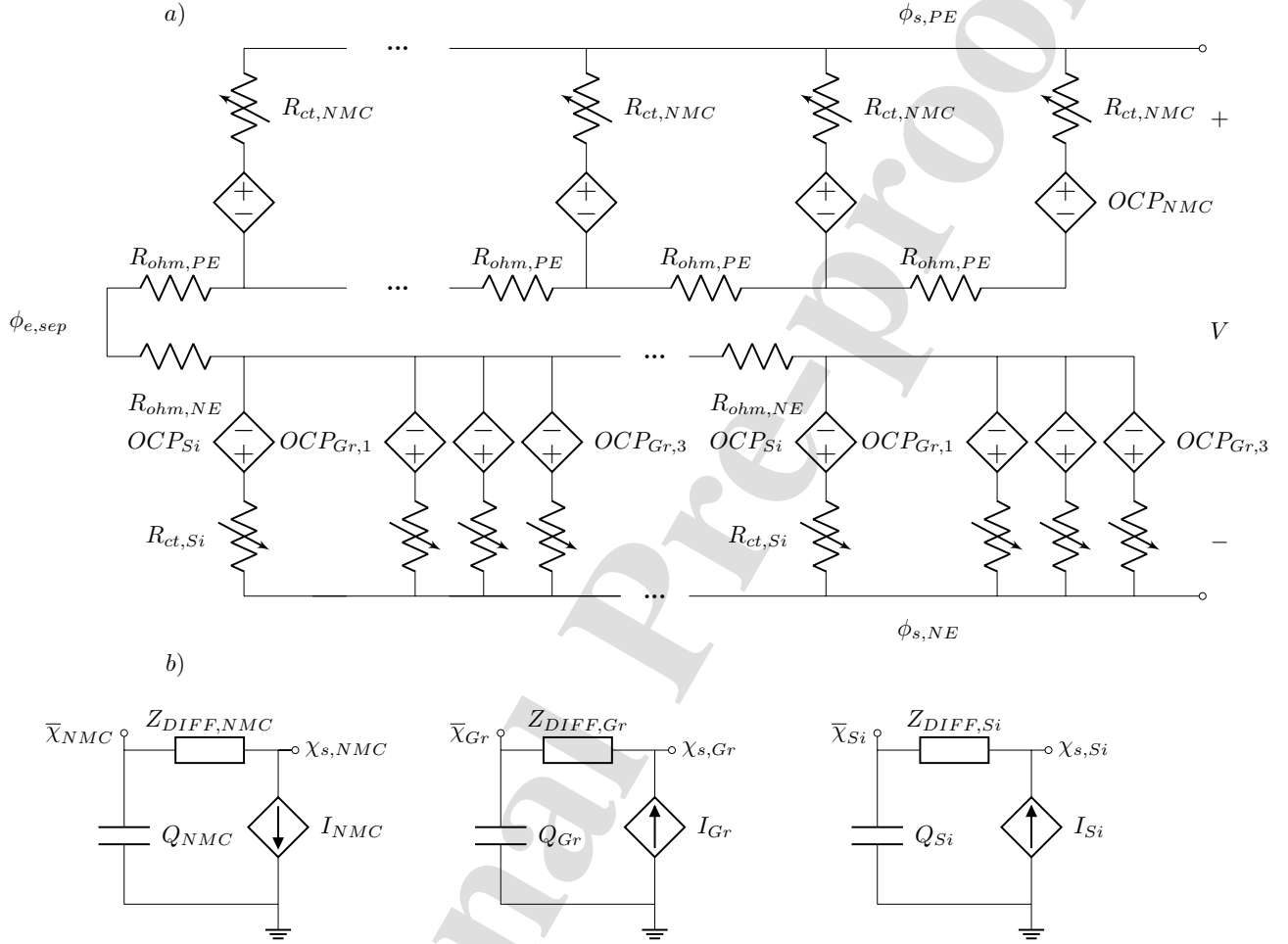


Figure 1: Discrete transmission line model for a NMC/Si-Gr cell. Subcircuit a) represents macroscale electrode structure, whereas subcircuits in b) symbolize solid diffusion in active material particles. It is noted that each controlled voltage source  $OCP$  in a) has an associated solid diffusion subcircuit like those in b), corresponding to each particle type.  $Z_{DIFF}$  stands for the impedance associated with solid diffusion after subtracting integrator dynamics.  $\phi_s$  and  $\phi_e$  stand for solid and electrolyte potential, whereas  $N_{PE}$  and  $N_{NE}$  indicate the number of spatial nodes in the positive and negative electrode, respectively.

### 228 3. Module simulation framework

229 In this section, the method for hierarchical module simulation is outlined.  
230 The 4P module in ladder configuration analyzed in this article is depicted  
231 in the schematic in Fig. 2, where  $R_{int}$  and  $R_{cont}$  denote the interconnection  
232 and contact resistances, respectively.

233 The simulation procedure for the ECM in Fig. 1 was outlined in our  
234 previous work and is based on the separation of timescales [64], i.e., the  
235 assumption that solid diffusion is sufficiently slower and charge transfer is  
236 sufficiently faster than the selected timestep, also made in [44]. The assump-  
237 tion that the charge transfer process takes place instantaneously is also made  
238 in standard formulations of the DFN model and the SPM<sub>e</sub> [65]. Ultimately,  
239 the validity of this assumption relies on the reaction kinetics of electrode  
240 materials as well as ambient temperature.

241 In the case of parallel-connected cells, individual cell currents are not  
242 known initially, so the circuit formulation of the model in Fig. 1 is exploited  
243 in order to derive a simplified equivalent circuit  $V_{eq}, R_{eq}$  of each cell that  
244 is substituted into the ladder structure in Fig. 2 at each timestep for the  
245 computation of the current distribution in the module. Once the current  
246 through each cell has been computed for one timestep, it is treated as the  
247 input to the cell model in Fig. 1 for the calculation of the internal current  
248 distribution within each electrode. It is noted that the values of intermediate  
249 variables  $V_{eq}, R_{eq}$  are updated at the beginning of each timestep for each cell  
250 and evolve over time with cell and module operation. As a result of the  
251 separation of timescales, the full-cell Thévenin equivalent of the TLM does  
252 not contain a transient component associated with charge transfer as it does

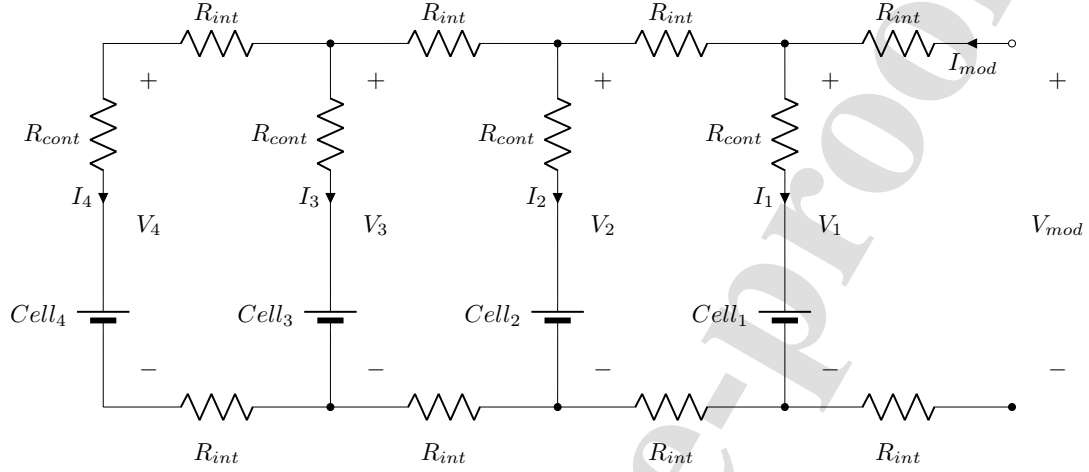


Figure 2: Schematic of the 4P module in ladder configuration, where  $R_{int}$  and  $R_{cont}$  denote the interconnection and contact resistances, respectively, and each battery symbol represents the full cell model in Fig. 1.  $V_{mod}$  and  $I_{mod}$  stand for module voltage and current, whereas  $V_{1:4}$  and  $I_{1:4}$  symbolize branch voltage and current, respectively.

253 with solid diffusion, which may lead the model to slightly overestimate the  
 254 dynamic voltage response to sufficiently quick changes in current, both at cell  
 255 and module levels. However, the experimental sampling time would have to  
 256 be small enough to capture this transient behavior.

257 A detailed outline of each simulation step is provided below.

258 1) **Updating of solid diffusion states.** The impedance describing the  
 259 dynamics of the difference between surface and average normalized con-  
 260 centrations in a spherical particle is defined in general as  $Z_{DIFF}$ , which  
 261 is particularized for each material in Fig. 1-b). It is approximated by  
 262 a series connection of three RC elements, as shown in Eq. (1).

$$Z_{DIFF}(s) = \frac{K_d}{\sqrt{\tau_d s} \coth(\sqrt{\tau_d s}) - 1} - \frac{3K_d}{\tau_d s} \approx \frac{K_d}{5} \left( \frac{a_1}{1 + b_1 \tau_d s} + \frac{a_2}{1 + b_2 \tau_d s} + \frac{a_3}{1 + b_3 \tau_d s} \right) \quad (1)$$

263 where the time constant  $\tau_d$  is defined as  $\tau_d = \frac{R^2}{D}$ ,  $R$  being particle radius  
 264 and  $D$  solid diffusivity. The proportionality constant  $K_d$  is related  
 265 to  $\tau_d$  by the integrator constant  $Q$  which represents particle capacity  
 266  $Q = \frac{\tau_d}{3K_d}$ . Coefficient values  $a_i, b_i$  for the approximation are shown in  
 267 Table 1 and have been determined according to the distribution of time  
 268 constants associated with  $Z_{DIFF}(s)$  [61].

Table 1: Coefficient values for the approximation in Eq. 1

$i$	1	2	3
$a_i$	0.5344	0.2724	0.1932
$b_i$	0.0479	0.0101	0.0020

269 Thus, the average and surface concentrations are updated according to  
 270 the individual particle currents computed in the previous timestep. In  
 271 order to account for the effect of solid diffusion dynamics on the current  
 272 timestep, the following equivalent diffusion resistance  $R_{diff}$  is derived  
 273 based on the 3RC approximation in Eq. 1 and Table 1:

$$R_{diff} = \frac{\partial OCP}{\partial x} \Big|_{x_s} \frac{\Delta x_s}{I} = \frac{\partial OCP}{\partial x} \Big|_{x_s} \cdot \left[ \frac{\Delta t}{Q_p} + \sum_{i=1}^3 \frac{K_d a_i}{5} \left( 1 - \exp\left(\frac{\Delta t}{b_i \tau_d}\right) \right) \right] \quad (2)$$

274 where  $Q_p$  and  $x_s$  stand for particle capacity and surface concentra-  
 275 tion, respectively. This is an auxiliary resistance that is not physically

276 present in the model, but it is connected in series with the OCP voltage  
 277 source to account for the linearized dynamic change in diffusion volt-  
 278 age during the current timestep. This implementation is made possible  
 279 by the 3RC approximation to the solid diffusion transfer function, and  
 280 provides improved numerical stability in the regions where the slope of  
 281 any OCP function  $\frac{\partial OCP}{\partial x}$  is large.

282 2) **Calculation of charge transfer resistances.** Charge transfer re-  
 283 sistance are defined as a function of the exchange current  $I_0$  with a  
 284 nonlinear dependency on particle current  $I$  according to the Butler-  
 285 Volmer equation, as shown in Eq. (3):

$$\eta_{ct} = \frac{2RT}{F} \sinh^{-1} \left( \frac{I}{2I_0} \right) \quad R_{ct} = \frac{\eta_{ct}}{I} = \frac{RT}{FI_0} \frac{\sinh^{-1} \left( \frac{I}{2I_0} \right)}{\frac{I}{2I_0}} \quad (3)$$

286 The exchange current  $I_0$  is expressed as a function of parameter  $K_0$ ,  
 287 which contains the temperature dependency, and its dependence on the  
 288 normalized surface concentration  $\chi_s$ :

$$I_0 = K_0 \sqrt{\chi_s(1 - \chi_s)} \quad (4)$$

289 The exact expression for the charge transfer resistance in Eq. (3)  
 290 presents a nonlinear dependency on the ratio between particle current  
 291  $I$  and exchange current  $I_0$ . For this reason, herein the charge transfer  
 292 resistances in timestep  $k$  are calculated as a function of the particle  
 293 current in the previous timestep  $k - 1$ , as shown in Eq. (5):

$$R_{ct,k} = \frac{RT}{FI_{0,k}} \frac{\sinh^{-1} \left( \frac{I_{k-1}}{2I_{0,k}} \right)}{\frac{I_{k-1}}{2I_{0,k}}} \quad (5)$$

294 This approach allows for an approximation to the exact value of charge  
 295 transfer resistances that is more accurate than a simple linearization  
 296 providing that the timestep  $\Delta t$  is small, and enables the computation  
 297 of the equivalent resistance of each cell.

298 The next step for a simpler solution of the current distribution in the  
 299 negative electrode is collapsing all branches  $n$  at a single node in the  $x$   
 300 dimension corresponding to silicon and different graphite particle sizes,  
 301 following the approach in [31]:

$$R_{n,eq} = \frac{1}{\sum_{n=1}^M \frac{1}{R_{ct,n} + R_{diff,n}}}, \quad OCP_{eq} = \frac{\sum_{n=1}^M \frac{OCP_n}{R_{ct,n} + R_{diff,n}}}{\sum_{n=1}^M \frac{1}{R_{ct,n} + R_{diff,n}}} \quad (6)$$

302 This is an efficient approach to the mathematical treatment of a blend  
 303 of active materials as well as a particle size distribution.

304 3) **Calculation of full-cell Thévenin equivalents.** The distributed cir-  
 305 cuit network can be simplified by calculating its instantaneous Thévenin  
 306 equivalent. For each electrode, the instantaneous Thévenin resistance  
 307  $R_{eq}$  can be computed by solving the linear system of equations in Eq.  
 308 7 with the definitions in Eq. 8. From here on,  $N$  stands for the number  
 309 of spatial nodes in each electrode ( $N_{PE}$ ,  $N_{NE}$ ).

$$\begin{bmatrix} 1 & -1 & 0 & 0 & \dots & 0 \\ -\alpha_1 & 1 + 2\alpha_1 & -\alpha_1 & 0 & \dots & 0 \\ 0 & -\alpha_2 & 1 + 2\alpha_2 & -\alpha_2 & \ddots & \vdots \\ \vdots & 0 & \ddots & \ddots & \ddots & 0 \\ 0 & 0 & \ddots & -\alpha_{N-1} & 1 + 2\alpha_{N-1} & -\alpha_{N-1} \\ 0 & 0 & \dots & 0 & -\alpha_N & 1 + \alpha_N \end{bmatrix} \begin{bmatrix} Z_0 \\ Z_1 \\ Z_2 \\ \vdots \\ Z_{N-1} \\ Z_N \end{bmatrix} = \begin{bmatrix} R_{ohm} \\ 0 \\ 0 \\ \vdots \\ 0 \\ 0 \end{bmatrix} \quad (7)$$

$$\alpha_{1:N} = \frac{R_{n,eq,1:N}}{R_{ohm}}, \quad R_{eq} = Z_0 \quad (8)$$

310 The Thévenin voltage is calculated by solving the electrode ladder  
311 structure when the current is set to zero:

$$\begin{bmatrix} 1 + \alpha_1 & -\alpha_1 & 0 & \dots & 0 \\ -\alpha_2 & 1 + 2\alpha_2 & -\alpha_2 & \ddots & \vdots \\ \vdots & \ddots & \ddots & \ddots & 0 \\ 0 & \ddots & -\alpha_{N-1} & 1 + 2\alpha_{N-1} & -\alpha_{N-1} \\ 0 & \dots & 0 & -\alpha_N & 1 + \alpha_N \end{bmatrix} \begin{bmatrix} \phi_1 \\ \phi_2 \\ \vdots \\ \phi_{N-1} \\ \phi_N \end{bmatrix} = \begin{bmatrix} OCP_{eq,1} \\ OCP_{eq,2} \\ \vdots \\ OCP_{eq,N-1} \\ OCP_{eq,N} \end{bmatrix} \quad (9)$$

312 where  $V_{eq} = \phi_1$ . Finally, the full cell Thévenin equivalent is computed  
313 as the series connection of both electrodes:  $R_{eq,FC} = R_{eq,PE} + R_{eq,NE}$ ,  
314  $V_{eq,FC} = V_{eq,PE} - V_{eq,NE}$ , which is substituted into the module ladder  
315 structure for an efficient computation of the current distribution. It is  
316 emphasized that this method provides values of intermediate variables  
317  $V_{eq}$  and  $R_{eq}$  that are electrically consistent with the dynamics of the  
318 cell model in Fig. 1 and do not depend on module-level current.

319 **4) Calculation of module voltage and current distribution.** For a  
320 4P module in ladder configuration, as indicated in Fig. 2, the module  
321 voltage  $V_{mod}$  as well as the branch voltages  $V_{1:N}$  can be calculated as the  
322 solution to the linear system of equations in Eq. 10 and the definition  
323 in 11.

$$\begin{bmatrix} 1 & -1 & 0 & 0 & 0 \\ -\beta_1 & 1+2\beta_1 & -\beta_1 & 0 & 0 \\ 0 & -\beta_2 & 1+2\beta_2 & -\beta_2 & 0 \\ 0 & 0 & -\beta_3 & 1+2\beta_3 & -\beta_3 \\ 0 & 0 & 0 & -\beta_4 & 1+\beta_4 \end{bmatrix} \begin{bmatrix} V_{mod} \\ V_1 \\ V_2 \\ V_3 \\ V_4 \end{bmatrix} = \begin{bmatrix} 2R_{int}I_{mod} \\ V_{eq,1} \\ V_{eq,2} \\ V_{eq,3} \\ V_{eq,4} \end{bmatrix} \quad (10)$$

$$\beta_{1:N} = \frac{R_{eq,1:N} + R_{cont}}{2R_{int}} \quad (11)$$

324 where  $V_{eq,1:N}$  and  $R_{eq,1:N}$  are the full-cell Thévenin voltages and resis-  
325 tances computed in the previous step,  $I_{mod}$  is the module current,  $R_{int}$   
326 is the interconnection resistance and  $R_{cont}$  is the contact resistance.  
327 The current through each branch may be calculated from the branch  
328 voltages according to Eq. (12).

$$I_{1:N} = \frac{V_{1:N} - V_{eq,1:N}}{R_{eq,1:N} + R_{cont}} \quad (12)$$

329 It is possible to reiterate the procedure outlined in Step 3 to compute  
330 the instantaneous Thévenin equivalent of the module, since it is akin to  
331 the ladder structure of each electrode in the employed cell model. This  
332 enables interfacing the module with loads other than a current source.  
333 A simulation of the discharge of a module through a switched shunt  
334 resistor is provided in Fig. S4 in order to emulate the most common  
335 alternative for passive balancing.

### 336 5) Calculation of cell electrode potential.

337 Once cell currents have been computed, they are treated as the input to  
338 the ECM in Fig. 1 to calculate the potential and current distribution

339 within each electrode in each cell. Electrode potential is calculated as  
 340 the solution to the linear system of equations in Eq. 13, characterized  
 341 by cell input current, and the variables calculated in step 2.

$$\begin{bmatrix} 1 & -1 & 0 & 0 & \dots & 0 \\ -\alpha_1 & 1 + 2\alpha_1 & -\alpha_1 & 0 & \dots & 0 \\ 0 & -\alpha_2 & 1 + 2\alpha_2 & -\alpha_2 & \ddots & \vdots \\ \vdots & 0 & \ddots & \ddots & \ddots & 0 \\ 0 & 0 & \ddots & -\alpha_{N-1} & 1 + 2\alpha_{N-1} & -\alpha_{N-1} \\ 0 & 0 & \dots & 0 & -\alpha_N & 1 + \alpha_N \end{bmatrix} \begin{bmatrix} \phi_0 \\ \phi_1 \\ \phi_2 \\ \vdots \\ \phi_{N-1} \\ \phi_N \end{bmatrix} = \begin{bmatrix} R_{ohm} I_{cell} \\ OCP_{eq,1} \\ OCP_{eq,2} \\ \vdots \\ OCP_{eq,N-1} \\ OCP_{eq,N} \end{bmatrix} \quad (13)$$

342  $\phi_{1:N}$  stands for electrode potential at nodes 1 through N, defined as  
 343  $\phi = \phi_s - \phi_e$ , and  $\phi_0$  is the electrolyte potential at the separator interface,  
 344 measured with respect to  $\phi_s$ . Subsequently, the current through each  
 345 particle at each node may be calculated from the potential distribution  
 346 in the electrode according to Eq. (14).

$$I_{1:N} = \frac{\phi_{1:N} - OCP_{eq,1:N}}{R_{n,k,1:N}} \quad (14)$$

347 This current will be the input to the state equation for solid diffusion  
 348 for the next timestep and will also be employed to update the charge-  
 349 transfer resistance for the next timestep.

350 The presented framework based on hierarchical model solution (electrode  
 351  $\rightarrow$  cell  $\rightarrow$  module  $\rightarrow$  cell  $\rightarrow$  electrode) proves computationally efficient, since  
 352 no equations must be solved by iterative methods, while also providing in-  
 353 sightful and interpretable information about the operation at each level. This

354 framework mainly consists of matrix-vector operations and tridiagonal sys-  
355 tems of equations, which makes it suitable for its implementation in embed-  
356 ded systems as demonstrated for a simpler case in [66].

357 The equivalent circuit described by  $(V_{eq}, R_{eq})$ , with values computed ac-  
358 cording to the procedure above, is electrically consistent with the dynamics  
359 of a battery cell since it constitutes the best linear approximation to its in-  
360 stantaneous current-voltage relationship in the neighborhood of its current  
361 operating point. Fig. 3 qualitatively exemplifies the instantaneous current-  
362 voltage relationship of a battery cell; the curvature is due to the nonlinearity  
363 of the Butler-Volmer equation as well as the dynamics of transient processes  
364 which produce a delayed voltage response to input current.

365 The static resistance approach approximates the behavior at an operating  
366 point  $(I_0, V_0)$  by the static component of cell voltage  $OCV$  and the chordal  
367 resistance. However, this approach implies that the full-cell overpotential is  
368 ohmic in nature, which is not fully in accordance with the dynamics of an  
369 electrochemical model. In contrast,  $V_{eq}$  is the value of cell voltage in the  
370 theoretical low current limit for said operating point, but factoring in the  
371 current state of dynamic processes, which makes it different from the equi-  
372 librium voltage  $OCV$ . In consequence,  $R_{eq}$  enables a closer approximation  
373 to instantaneous cell dynamics around the operating point  $(I_0, V_0)$ , which is  
374 required for the adequate simulation of module operation. In the case of  
375 the TLM, the Thévenin equivalent gives well-defined values of  $(V_{eq}, R_{eq})$  re-  
376 gardless of the operating conditions, whereas an electrochemical model only  
377 enables the computation of the static resistance since it is not formulated in  
378 terms of circuit elements. Fig. 3 also anticipates why numerical instability

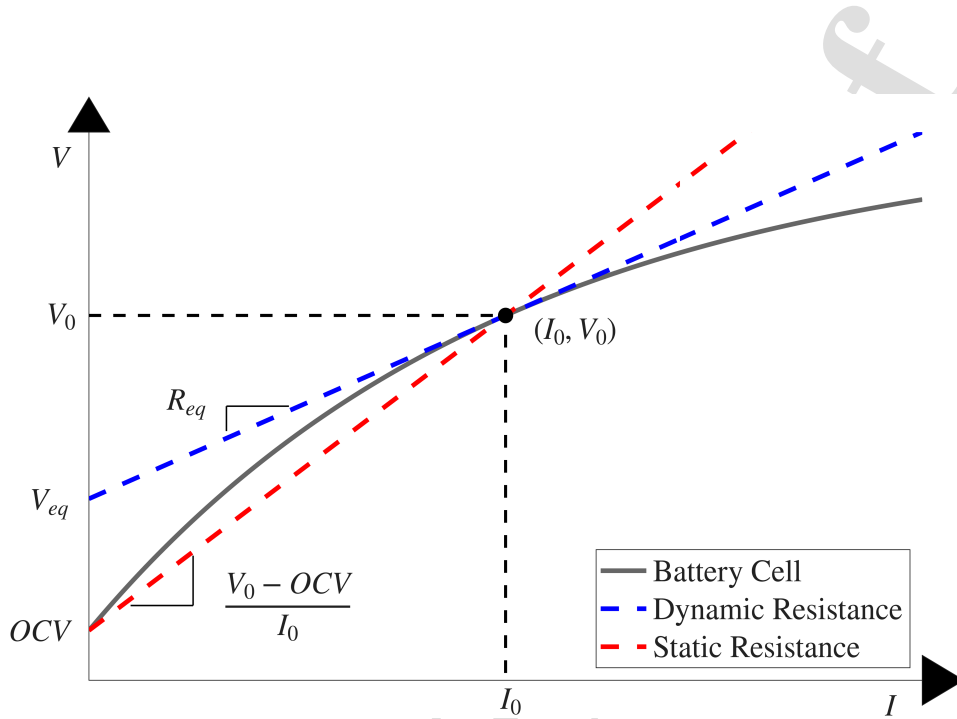


Figure 3: Schematic illustration of the difference between the static and dynamic resistance approaches for approximating the instantaneous current-voltage relationship of a battery cell at an operating point  $(I_0, V_0)$ .

379 may arise in the calculation of the static resistance when cell current is close  
 380 to zero. These aspects will be illustrated quantitatively in Section 5.3.

#### 381 4. Experimental setup and testing

382 In this article, the dataset presented by Piombo et al. [67, 68] is em-  
 383 ployed for experimental validation. Although readers may find an extensive  
 384 description of the experimental setup and equipment in the source publi-  
 385 cations [67, 68], critical details about the experimental setup and module  
 386 configuration that are relevant to the research work conducted herein are re-

387 produced for the sake of clarity. The aforementioned experimental campaign  
388 was aimed at quantifying cell-to-cell variations in capacity and internal re-  
389 sistance in commercial batches, as well as analyzing the performance of a 4P  
390 module under a variety of operating scenarios considering the following in-  
391 put parameters: interconnection resistance, ambient temperature, chemistry,  
392 and the presence of an aged cell. In particular, the subset of experimental  
393 data corresponding to NMC/Si-Gr cells is studied in this work.

394 A batch of 19 commercial NMC/Si-Gr cells (INR21700-M50T manufac-  
395 tured by LG Chem) were analyzed. The investigated cells have a nominal  
396 voltage of 3.63 V and exhibit a nominal discharge capacity of 4.85 Ah be-  
397 tween the voltage limits of 4.2 V and 2.5 V. Their standard constant charging  
398 current is 0.3C (1455 mA), with a maximum limit of 0.7C (3395 mA) from  
399 25°C to 45°C. Conversely, their standard constant discharging current is 0.2C  
400 (970 mA), with maximum values of 3C (14550 mA) from 10°C to 25°C, and  
401 1.5C (7275 mA) from 25°C to 55°C. Additionally, one identical cell from an  
402 earlier batch, stored at 50% SOC for 12 months under calendar aging condi-  
403 tions, was added to the pool in order to assess the effect of the presence of a  
404 slightly aged cell on module performance.

405 The physical configuration of the 4P module is illustrated in Fig. 4, while  
406 the magnification shows cell connections in greater detail. Hall-effect sensors  
407 (Honeywell SS495A) are employed to measure individual cell currents, while  
408 T-type thermocouples, taped to the cells' surface at mid height (not depicted  
409 in Fig. 4), are used to monitor individual cell temperatures. One additional  
410 thermocouple is employed to measure the ambient temperature inside the en-  
411 vironmental chamber. Hall sensors are mechanically fixed to the cell holders,

412 measuring the current through the bolts that enable electrical contact be-  
413 tween the cells and the busbars. The spacing between the cells is 5 mm, and  
414 both front and back sides of the cells shown in Fig. 4 are available for heat  
415 dissipation through natural convection. Two types of busbar are designed:  
416 a solid copper busbar to emulate an ideal interconnection (mounted on the  
417 module in Fig. 4), and a copper busbar composed of different segments be-  
418 tween which precision shunt resistors are soldered (shown on the right-hand  
419 side of Fig. 4). The nominal values of the employed shunt resistors are 1  
420 m $\Omega$  and 3 m $\Omega$ , whereas their respective average measured values are 1.05  
421 m $\Omega$  and 3.02 m $\Omega$ . The maximum value of  $R_{int}$  (3 m $\Omega$ ) was selected to be  
422 approximately 10% of the internal resistance of the investigated cell, and 1  
423 m $\Omega$  was chosen as an intermediate value between 0 m $\Omega$  and 3 m $\Omega$ . The  
424 interconnection resistance associated with the copper busbars is calculated  
425 based on its dimensions, giving a value of 5.4  $\mu\Omega$ . The value of the contact  
426 resistance (1.21 m $\Omega$ ) is determined experimentally by electrically character-  
427 izing identical bolt junctions at similar levels of pressure. Module terminals  
428 are connected to the battery cycler, which provides measurements of module  
429 voltage and current.

430 In order to calibrate the Hall sensors, a staircase current profile with  
431 known values from -2C to 2C is tested, and the resulting voltage response  
432 is recorded, as shown in Fig. 5-a). The calibration curve for one particular  
433 sensor is shown in Fig. 5-b), including the linear fit, which enables retrieving  
434 current values from the sensor's voltage response. It is noted that Hall sensor  
435 calibration data is also available in the source dataset.

436 The experimental campaign comprises two separate sections: single-cell

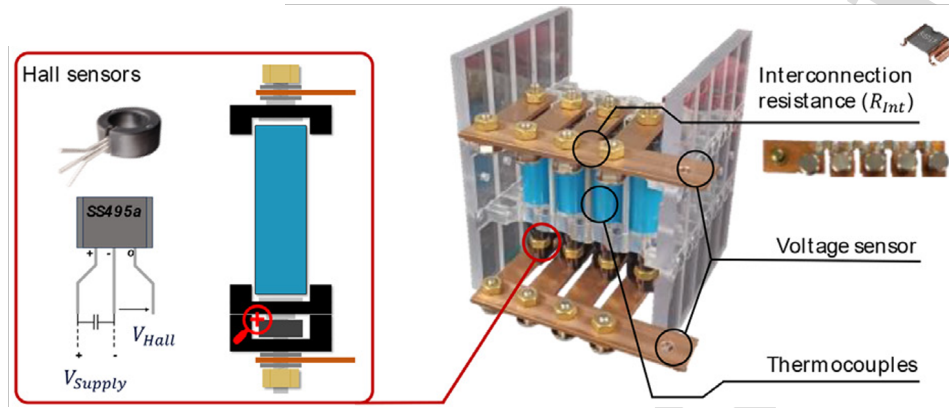


Figure 4: Physical configuration of the 4P module [67, 68].

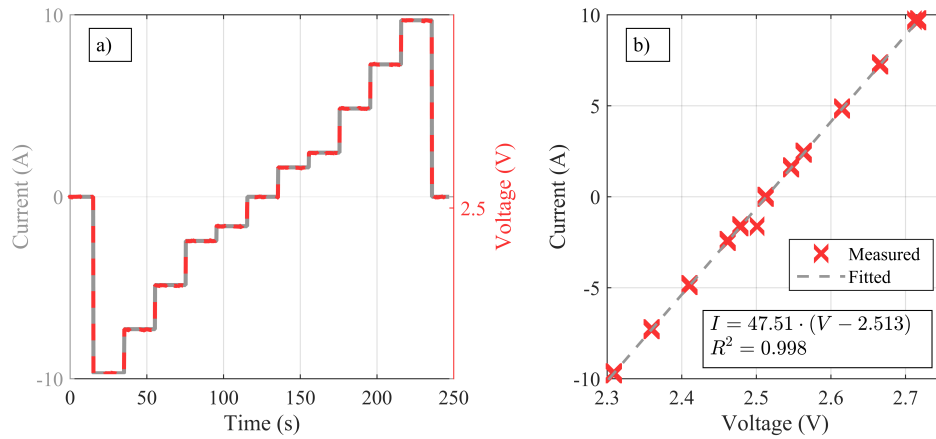


Figure 5: a) Input current (solid grey line) and Hall sensor voltage response (dashed red line). b) Linear calibration curve (dashed grey line) and the measured voltage-current data (red crosses) for one particular Hall sensor.

437 characterization and module-level operation. Single-cell characterization was  
 438 performed at a constant temperature of 23°C, and consisted of a pseudo-

439 Open Circuit Voltage (OCV) discharge at  $C/20$  for full-cell reconstruction,  
440 and a Hybrid Pulse Power Characterization (HPPC) schedule for dynamic  
441 characterization. HPPC current pulses are applied at 10% SOC intervals  
442 from 90% SOC down to 10% SOC with a charge/discharge ratio of 0.75  
443 ( $2.66\text{ A}/2\text{ A}$ ) and a duration of 10 s. The charging procedures in this case  
444 were Constant Current-Constant Voltage (CC-CV) at  $C/3$  up to 4.2 V, and  
445 then until a 50 mA cutoff was reached.

446 Module-level operation comprises a matrix of tests including 3 values of  
447 interconnection resistance ( $0\text{ m}\Omega$ ,  $1\text{ m}\Omega$  and  $3\text{ m}\Omega$ ) and 3 ambient temper-  
448 atures ( $10^\circ\text{C}$ ,  $25^\circ\text{C}$  and  $40^\circ\text{C}$ ), which were then repeated for the case where  
449 the aged cell is located furthest from the module terminals (position 4 in Fig.  
450 2), yielding a total of 18 different cases. The module-level testing protocol  
451 consisted of a CC-CV charge as described above, followed by a discharge  
452 at a nominal module rate of  $0.8C$  ( $15.5\text{ A}$ ), set so that individual cell cur-  
453 rents are guaranteed to remain below the discharge limits specified above  
454 at all temperatures. All the experimental schedules were performed under  
455 temperature-controlled conditions, ensured by an Amerex IC500R environ-  
456 mental chamber, with the aid of an Arbin LBT22013 battery cycler, which is  
457 employed in conjunction with a Data Acquisition System (DAQ) in module  
458 operation for the collection of cell-level current and temperature measure-  
459 ments with a constant sampling time of 1 s.

## 460 5. Results and discussion

### 461 5.1. Single-cell characterization

462 In this section, the parameterization methodology for the TLM of a single  
 463 cell in Fig. 1 is described and performed for all the cells in the batch. Based  
 464 on the experimental campaigns outlined in 4, single-cell characterization is  
 465 composed of two steps: thermodynamic and dynamic characterization.

466 1) **Thermodynamic characterization.** Pseudo-OCV discharges at C/20  
 467 are employed for the full-cell reconstruction of all 20 cells in the batch  
 468 as explained below. Experimental thermodynamic profiles are shown in  
 469 Fig. 6-a), whereas the full-cell reconstruction of a fresh cell is shown in  
 470 Fig. 6-b). In order to ensure consistent reconstruction results, the same  
 471 percentage of silicon contribution to anode capacity (11%) is assumed  
 472 for all cells, as well as the initial stoichiometry values corresponding to  
 473 SOC=100% ( $\chi_{NMC} = 0.025$ ,  $\chi_{Gr} = 0.93$ ,  $\chi_{Si} = 0.99$ ), leaving electrode  
 474 capacities  $Q_{PE}$  y  $Q_{NE}$  as the fitting parameters. Furthermore, the ana-  
 475 lytical expressions for OCPs provided in Appendix A are employed for  
 476 electrode balancing, whose experimental data were originally presented  
 477 in [69]. Full-cell reconstruction results are summarized by the following  
 478 electrode capacities:  $Q_{PE} = 5.386 \pm 0.019$  Ah y  $Q_{NE} = 5.203 \pm 0.039$   
 479 Ah, with a larger variation in anode capacity and an average voltage  
 480 error of 12.8 mV RMS. These results are analogous to those presented  
 481 in [70] for the INR21700-M50T cell, with a minor decrease in  $Q_{PE}$  as-  
 482 cribed to the fact that the original half-cell data for NMC811 in [69]  
 483 does not include the abrupt voltage drop at high degrees of lithiation.  
 484 As a result, the estimated electrode capacity is not an absolute value,

485 but it is computed in relation to the available half-cell data. However,  
 486 this region falls outside the full-cell operating window, and thus, it is  
 487 not expected to affect full-cell simulation results. It is also noted that  
 488 a slightly lower silicon contribution is obtained herein in comparison  
 489 to [70] (11% vs. 15%), which is likely due to minor differences in the  
 490 employed silicon OCP profile.

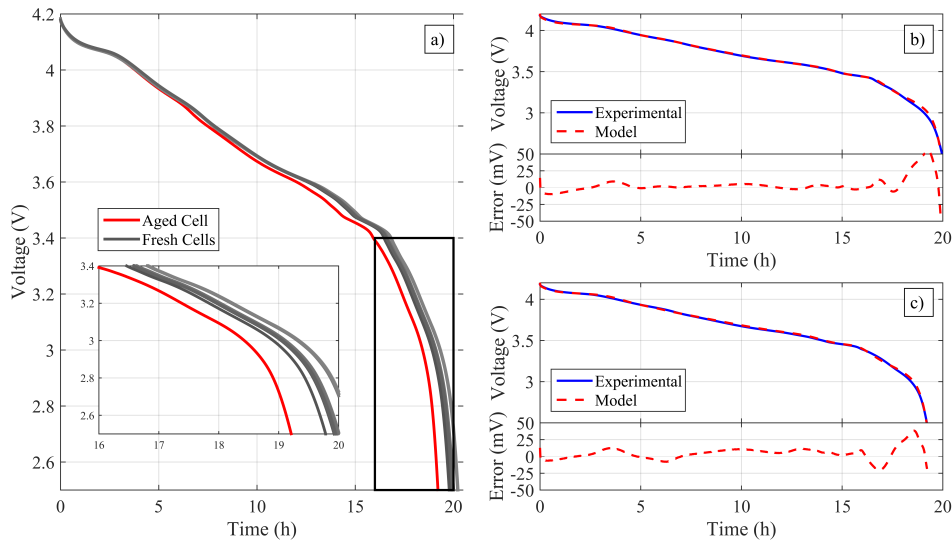


Figure 6: a) Pseudo-OCV discharge profiles at C/20 for all 20 cells in the batch. Fresh cells are plotted in grayscale, while the aged cell is plotted in red. b) Full-cell reconstruction for one fresh cell, along with voltage error between experimental and reconstructed profiles. c) Full-cell reconstruction for the aged cell, along with voltage error between experimental and reconstructed profiles.

491 With respect to the aged cell, full-cell reconstruction results are shown  
 492 in Fig. 6-c), with a voltage error of 10.4 mV RMS. It is pointed out  
 493 that, in both cases, the voltage error between experimental and recon-

494 structured thermodynamic profiles remains below 10 mV for SOCs above  
495 15%, i.e., before 17 h into the C/20 discharge. A higher error is ob-  
496 tained in the low SOC region, where the contribution of silicon to anode  
497 capacity is the greatest, ascribed to possible differences in the behavior  
498 of silicon in full-cell and half-cell settings [69, 71]. Given that this cell  
499 was subjected to calendar aging, which is primarily associated with loss  
500 of lithium inventory, its full-cell reconstruction is performed by assum-  
501 ing a negative electrode shift to lower stoichiometries. However, since  
502 this cell is in only the early stages of aging (i.e., 96% of nominal capac-  
503 ity), there are other possibilities that are able to yield accurate full-cell  
504 reconstruction results. In any case, the goal of this analysis is to obtain  
505 an accurate OCV profile for module simulation, which is achieved, and  
506 not a comprehensive study of aging behavior. Although the TLM does  
507 not include real-time degradation submodels, it can account for the ef-  
508 fects of degradation modes by adjusting model parameters similarly to  
509 an electrochemical model. For instance, the initial stoichiometries can  
510 be adjusted to reproduce the shift in electrode balancing due to loss of  
511 lithium inventory, whereas the capacity associated with each material  
512 can be decreased to account for loss of active material. In consequence,  
513 the TLM can effectively be reparameterized for the current state of an  
514 aged cell in order to introduce it into module simulations.

515 2) **Dynamic characterization.** Once the static characteristics have  
516 been determined for all cells, HPPC data is employed to determine  
517 the necessary dynamic parameters for the model in Fig. 1, i.e., solid  
518 diffusion time constants and charge transfer parameter  $K_0$  for each

519 material, as well as ohmic resistances  $R_{ohm}$  for each electrode. A key  
 520 advantage of the model in Fig. 1 with respect to a full electrochemical  
 521 model lies in the fact that only electrical parameters are required  
 522 instead of physical parameters.

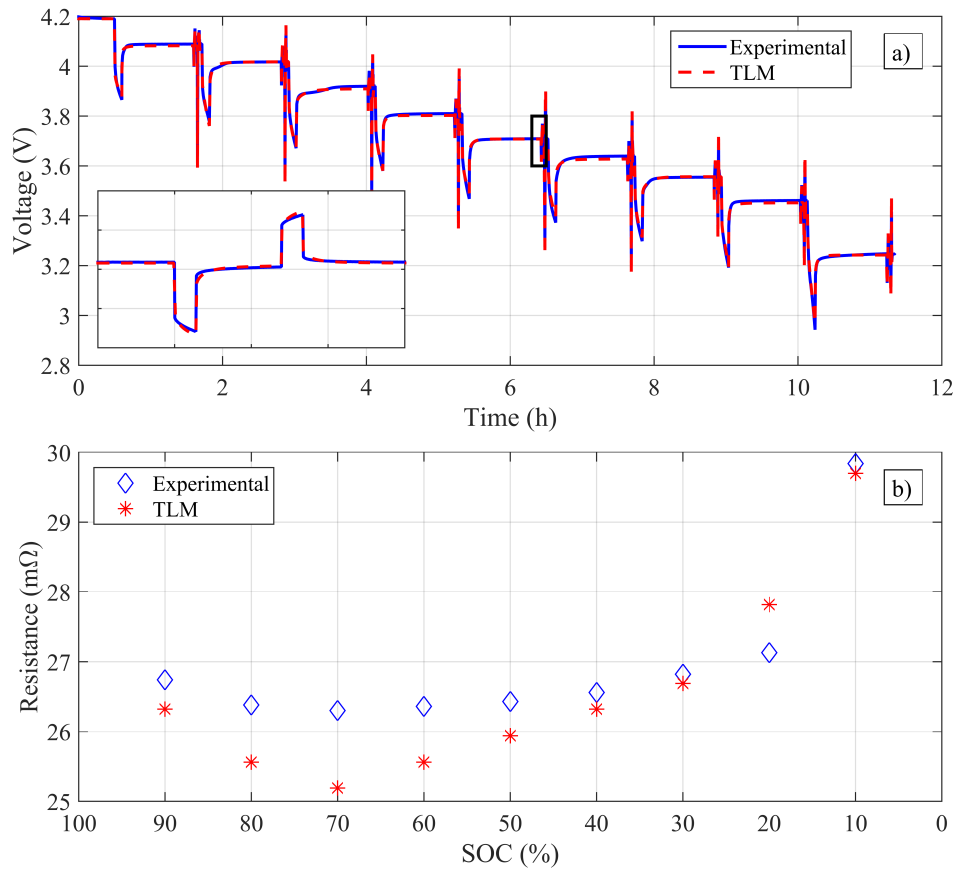


Figure 7: a) Comparison between experimental and simulated responses to the HPPC protocol for the identified model parameters. Inset depicts a detailed view of the short current pulses at 50% SOC. b) Comparison between the average HPPC resistance as a function of SOC determined experimentally and obtained from the TLM.

523 In our previous work [52], we detailed the parameterization methodol-  
524 ogy for the TLM for a commercial 18650 NMC811/Si-Gr cell (Samsung-  
525 SDI INR18650-35E, 3.4 Ah) also considering EIS and rate capability  
526 tests. However, the cell under study in this work is a commercial 21700  
527 NMC811/Si-Gr cell (LG Chem INR21700-M50T 4.85 Ah). If the in-  
528 crease in capacity is assumed to be due to an increase in electrode area,  
529 this leads to a proportional decrease in impedance, which provides an  
530 initial parameter estimation. Although the dynamic parameters of the  
531 TLM are not completely identifiable from an HPPC profile, it is used  
532 to refine said initial parameter values with the aid of Particle Swarm  
533 Optimization. The resulting parameters are manually adjusted in order  
534 to obtain a single set of dynamic parameters valid for all cells.

535 A comparison between experimental and simulated responses for a sam-  
536 ple cell is shown in Fig. 7-a); an average error of 8.98 mV RMS is ob-  
537 tained for the fresh cells with the parameter values reported in Table  
538 2. A close agreement with experimental data is observed through-  
539 out the profile, including relaxation periods and equilibrium voltages,  
540 which indicate an accurate determination of diffusion time constants  
541 as well as OCV reconstruction. As shown in the inset, dynamic be-  
542 havior as a response to quick current pulses is also captured, which is  
543 attributed to an appropriate description of electrode spatial structure.  
544 Fig. 7-b) shows the comparison between the average HPPC resistance  
545 determined experimentally and that obtained with the TLM for 90%  
546 SOC down to 10% SOC. It is noted that the TLM is capable of provid-  
547 ing an accurate approximation to experimental results, with an RMS

548 error of 0.63 m $\Omega$  RMS (2.3%) and a maximum error of 1.11 m $\Omega$  (4.2%),  
549 which implies an appropriate description of electrode spatial structure  
550 as well as an accurate parameterization. Finally, it is noted that an  
551 accurate result (11.2 mV RMS) is also obtained for the aged cell with  
552 the same parameter set, which we ascribe to the fact that the aging  
553 on this cell is not severe enough to cause a substantial variation in  
554 dynamic behavior. It is observed that the TLM does not appear to  
555 overestimate the experimental response to the current pulses, sampled  
556 with a timestep of 0.1 s. In consequence, we hypothesize that current  
557 fluctuations down to this timescale can be simulated accurately within  
558 the range of operating conditions considered.

559 An additional advantage of the TLM approach in relation to conven-  
560 tional ECMs is the direct connection established between circuit states  
561 and internal state variables of electrochemical models beyond a current-  
562 voltage correspondence. In order to illustrate the internal behavior of  
563 the TLM, Fig. 8 shows the time evolution of internal state variables  
564 of interest that have a direct equivalence to those of electrochemical  
565 models, i.e., the normalized lithium surface concentration in NMC, sil-  
566 icon, and graphite active material particles and the anode potential,  
567  $\phi_{s,NE} - \phi_{e,NE}$ , during the HPPC protocol shown above.

568 With respect to particle concentrations, it is observed that a discrete  
569 spatial distribution causes heterogeneity, with the particle located clos-  
570 est to the separator (Particle 1) experiencing greater fluctuations in its  
571 concentration. Regarding NMC, the greater slope of its OCP prevents  
572 large variations in concentration between particles at different spatial

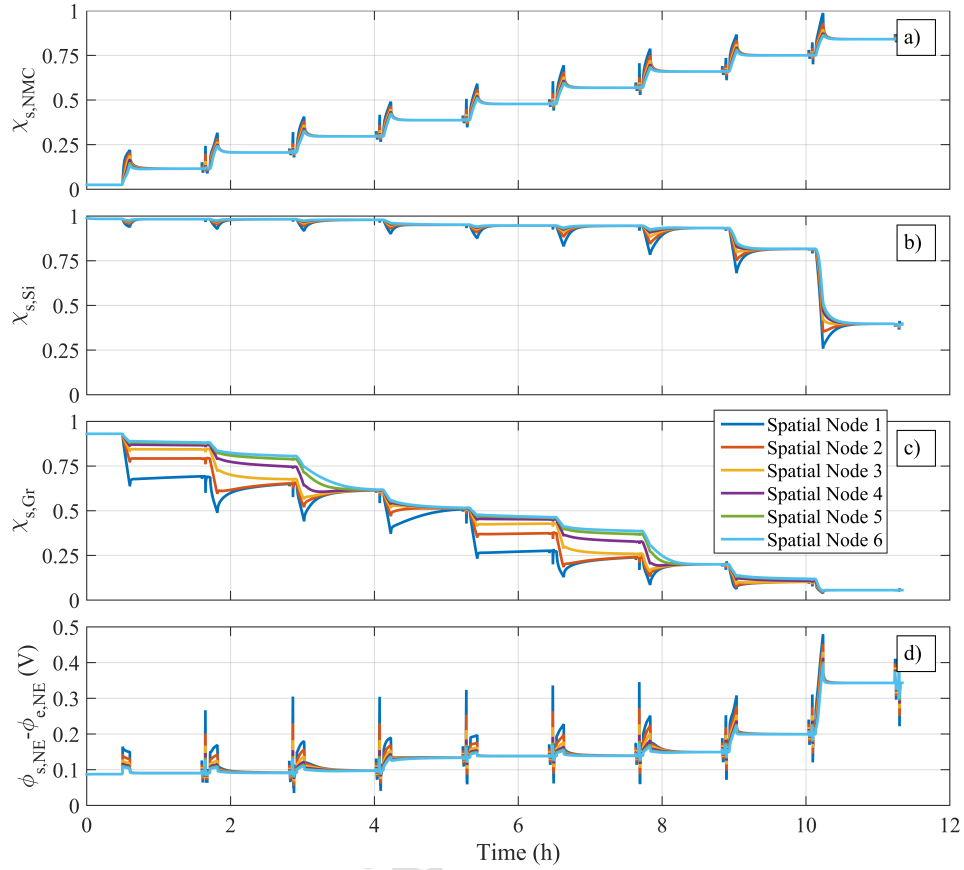


Figure 8: Time evolution of the normalized surface concentration of a) NMC, b) silicon, c) graphite active material particles, where each line corresponds to each spatial node. d) Time evolution of negative electrode potential  $\phi_{s,NE} - \phi_{e,NE}$ , where each line corresponds to each spatial node.

573 nodes, which only occur during discharge pulses. In contrast, the step-  
 574 wise flat OCP profile of graphite leads to considerable heterogeneity  
 575 in concentration that persists during relaxation periods, since small

576 voltage differences between spatial nodes translate into large variations  
 577 in concentration in regions where the OCP profile is approximately  
 578 flat. Another key feature of this particular cell is the blended Si-Gr  
 579 anode, whose sequential operation is characterized by a predominant  
 580 graphite utilization for SOC's above 20%, with silicon utilization being  
 581 primarily restricted below said threshold. The spatial distribution of  
 582 intercalation current within each electrode also constitutes a variable  
 583 of interest; however, since it is directly correlated to concentration, it  
 584 is not depicted in Fig. 8 to avoid clutter. Another relevant internal  
 585 variable that may be monitored is anode potential, since a high risk  
 586 of lithium plating is assumed to appear when this variable crosses the  
 587 0 V threshold. Although the behavior shown in Fig. 8 corresponds  
 588 to operation under an HPPC profile, which is primarily a discharge,  
 589 it is noted that the particle located closest to the separator (Particle  
 590 1) undergoes greater overpotentials both during charge and discharge  
 591 pulses, therefore being at a higher risk of lithium plating as a result.  
 592 For a more comprehensive discussion on behavior during charging and  
 593 the onset of lithium plating for this chemistry, readers are referred to  
 594 our previous work on the application of the TLM at the cell level [52].  
 595 With regard to the particle size distribution for graphite, we follow a  
 596 similar approach as in our previous work [52] by defining three particle  
 597 radii  $[R_1 \ R_2 \ R_3] = [1 \ 3 \ 10]$ , which leads to subsequent diffusion  
 598 time constants  $\tau_{d,Gr} = \tau_{d,Gr,ref} [1 \ 9 \ 100]$ . The consideration of large  
 599 particle sizes is because outlier particles, while not significantly increas-  
 600 ing electrode capacity, can decisively influence the relaxation process

601 [72]. Additionally, although particles are generally considered to be  
 602 spherical and evenly distributed, their shapes may actually be irreg-  
 603 ular, causing slower lithium redistribution [73]. Lastly, the relative  
 604 capacity associated with each particle size  $[\epsilon_1 \ \epsilon_2 \ \dots \ \epsilon_m]$  is defined  
 605 and adjusted from the values in [52] to match relaxation profiles after a  
 606 full module discharge. Temperature-dependent parameters  $\tau$  for solid  
 607 diffusion and  $K_0$  for charge transfer are modified according to an Ar-  
 608 rhenius equation with parameters  $E_{a,dif} = 45$  kJ/mol and  $E_{a,ct} = 65$   
 609 kJ/mol, in agreement with previous identification and validation results  
 610 for the discrete TLM modeling approach and chemistry [52, 62]. It is  
 611 noted that the Arrhenius parameters employed herein may differ from  
 612 those in [59], due to the different interpretation of EIS resulting from  
 613 the discrete TLM modeling approach as opposed to the traditional ap-  
 614 proach to EIS fitting based on the series connection of circuit elements  
 615 [74]. A summary of parameter values is reported in Table 2.

Table 2: Dynamic model parameters

Positive electrode		Negative electrode			
$R_{ohm,PE}$	6 m $\Omega$	$R_{ohm,NE}$	7 m $\Omega$	$[\epsilon_1 \ \epsilon_2 \ \epsilon_3]$	$[0.813 \ 0.163 \ 0.024]$
$\tau_{d,NMC}$	4000 s	$\tau_{d,Si}$	160 s	$\tau_{d,Gr,ref}$	840 s
$K_{0,NMC}$	Very Large	$K_{0,Si}$	0.2 A	$K_{0,Gr,ref}$	1.5 A

616 In summary, all 20 cells in the batch, including the aged cell, are charac-  
 617 terized thermodynamically and dynamically, resulting in specific values for  
 618 electrode capacities, but uniform values for dynamic parameters. Next, each

619 individual cell model is introduced into the module structure for each re-  
620 spective case, and module operation is simulated according to the procedure  
621 described in Section 3.

## 622 5.2. Module validation

623 In this section, module behavior, characterized by terminal voltage and  
624 current distribution, is validated against the experimental dataset described  
625 in 4; it is worth noting that only several representative cases in the matrix of  
626 tests are shown. Although the current distribution among parallel-connected  
627 cells has been the subject of numerous previous works, herein we also em-  
628 phasize the importance of simultaneously reproducing module voltage, as it  
629 determines the power and energy delivered to the load.

630 Simulation and experimental results are shown in Fig. 9 for the case of  
631 fresh cells at 25°C, with an interconnection resistance  $R_{int} = 3 \text{ m}\Omega$ . Regard-  
632 ing module voltage, it is worth noting that its magnitude and slope during  
633 the constant-current discharge are accurately matched, which we mainly as-  
634 cribe to an appropriate spatial structure of the single-cell model in Fig. 1.  
635 We demonstrated in our previous work [52] that this consideration is a key  
636 aspect to reproduce cell voltage at increasing C-rates. Although the average  
637 C-rate is below 1C, individual cell currents reach peaks of 1.25C, which ne-  
638 cessitates a cell model with a broader range of applicability. Additionally,  
639 module voltage during the relaxation phase is replicated with a high degree  
640 of accuracy, which is attributed to the incorporation of a particle size dis-  
641 tribution for graphite. A slight voltage discrepancy is observed from  $t=0.8$   
642 h, reaching its maximum deviation at  $t=0.96$  h, which is attributed to the  
643 presence of silicon; nevertheless, the overall error in module voltage is 10.0

644 mV RMS, which is indicative of the accuracy of the proposed method.

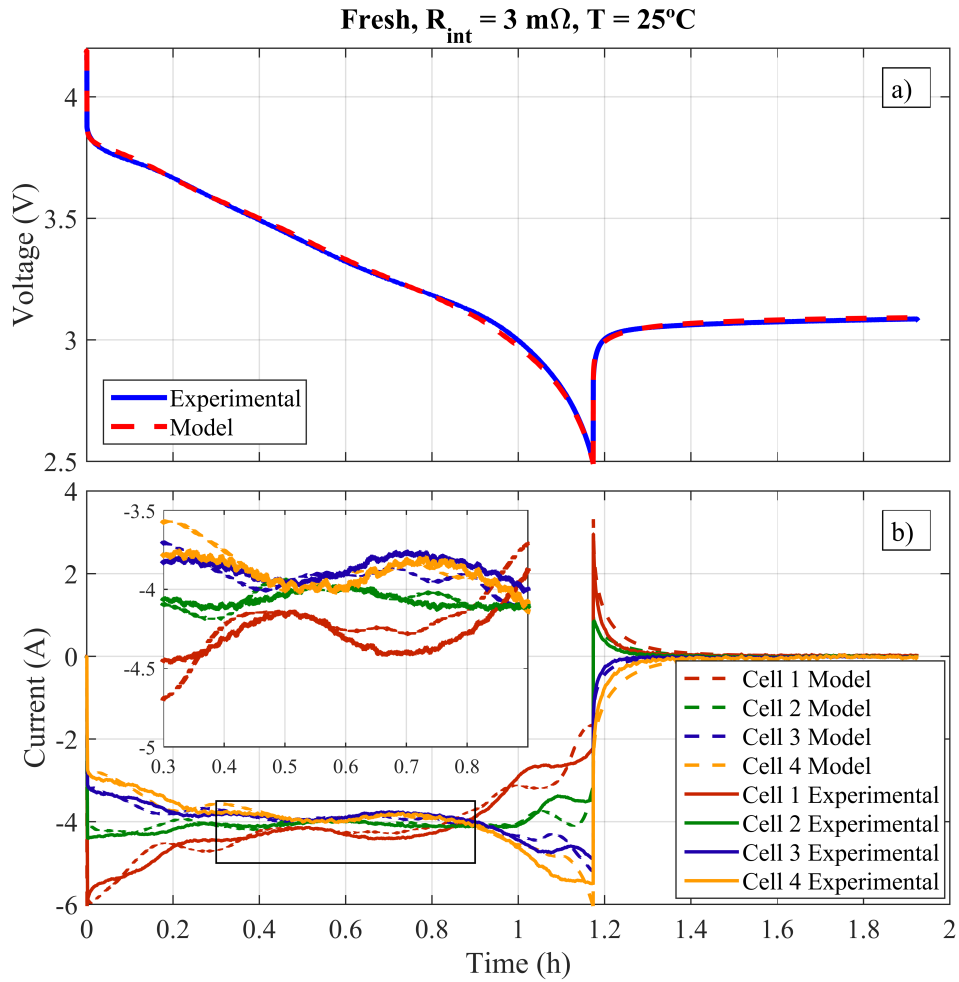


Figure 9: a) Comparison between experimental (solid) and simulated (dashed) module voltage for  $R_{int} = 3 \text{ m}\Omega$  and  $25^\circ\text{C}$ . b) Comparison between experimental (solid) and simulated (dashed) current distributions. Inset depicts a magnification on the current distributions from 0.3 h to 0.9 h.

645 As for the current distribution, accurate predictions are provided through-  
646 out the operating window. It is worth noting that the initial current distri-  
647 bution is accurately reproduced, which provides an estimation of the maxi-  
648 mum individual cell current for this module configuration. The fluctuations  
649 in the current distribution during the middle segment of the discharge are  
650 also quantitatively replicated, as detailed in the inset of Fig. 9. Although  
651 the qualitative behavior is also similar in the final phase of the discharge,  
652 simulation results appear to be ahead of experimental results, which could  
653 be justified by the fact that the presence of silicon and the large slope of  
654 graphite in this region result in a more unstable current distribution that  
655 is more challenging to predict. In fact, the greatest current error occurs at  
656  $t=1.16$  h, towards the very end of the discharge. Lastly, the current distribu-  
657 tion during relaxation is also accurately predicted, including the peak reverse  
658 balancing current, most notably on the Cell 1, which is located closest to the  
659 module terminals.

660 The case of an interconnection resistance  $R_{int} = 1$  m $\Omega$  at 40°C is discussed  
661 next, whose results are shown in Fig. 10. With respect to module voltage, a  
662 direct consequence of a decrease in interconnection resistance is an increase  
663 in voltage throughout the constant-current discharge, also amplified by the  
664 decrease in impedance due to the higher temperature. These behaviors are  
665 captured by the model, which yields accurate predictions during both the  
666 constant-current discharge and the relaxation periods with a voltage error of  
667 15.0 mV RMS. Regarding the current distribution, the effect of reducing the  
668 interconnection resistance is a noticeable decrease in spread, which is mostly  
669 restricted to the initial and final segments of the discharge. The model is able

670 to reproduce the current distribution within these regions to a high degree  
671 of accuracy (0.093 A RMS on average), as detailed in the inset of Fig. 10,  
672 as well as anticipate a faster module self-balancing as a result of the lower  
673 interconnection resistance. In this case, the largest error in both voltage and  
674 current occurs at  $t=1.15$  h, close to the end of the discharge.

675 Other relevant cases where the model is validated experimentally are those  
676 in which the aged cell replaces a fresh cell in position 4 in the module. Results  
677 at  $40^{\circ}\text{C}$  with an interconnection resistance  $R_{int} = 3$  m $\Omega$  are shown in Fig.  
678 11. Similarly, the model is also capable of producing highly accurate results,  
679 matching the magnitude and slope of module voltage during discharge, as well  
680 as the profile during relaxation, with an overall voltage error of 11.1 mV RMS.  
681 Analogously, an acceptable prediction of the current distribution is provided  
682 throughout the complete process (0.137 A RMS on average), including the  
683 initial spread and the fluctuations in the middle segment, detailed in the  
684 inset. For this value of the interconnection resistance, the key difference  
685 with the case analyzed previously lies in the current distribution towards the  
686 end of the discharge: it is observed that cell 4 provides a similar current to  
687 cell 3 in this segment, which is effectively captured by the model, as with the  
688 current distribution during relaxation. In this case, the largest error in both  
689 voltage and current occurs at  $t=1.33$  h, towards the end of the discharge.

690 The effect of the presence of an aged cell is also evaluated for an intercon-  
691 nection resistance  $R_{int} = 1$  m $\Omega$  at  $25^{\circ}\text{C}$ , and results are shown in Fig. 12. In  
692 this case, a smaller value of the interconnection resistance renders cell-to-cell  
693 variations in capacity more impactful. Whereas in the previous case with  
694  $R_{int} = 1$  m $\Omega$  the current distribution was virtually uniform in the middle

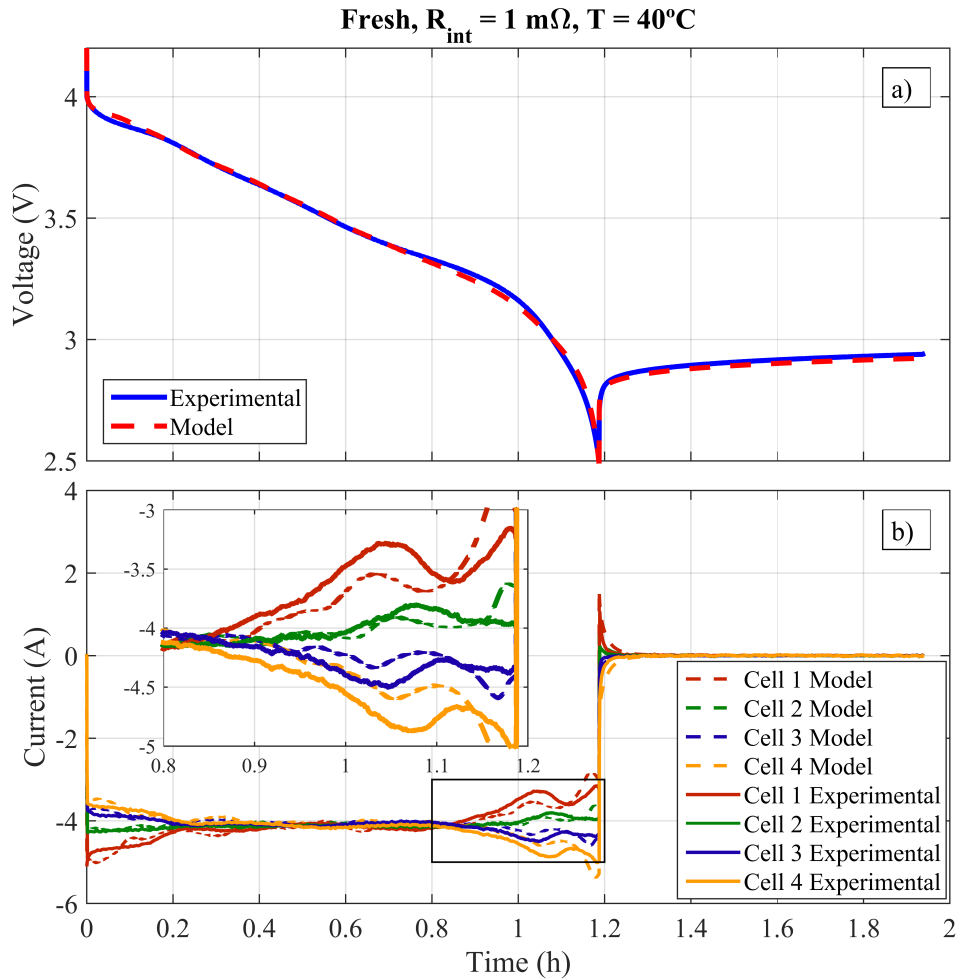


Figure 10: a) Comparison between experimental (solid) and simulated (dashed) module voltage for  $R_{int} = 1 \text{ m}\Omega$  and  $40^\circ\text{C}$ . b) Comparison between experimental (solid) and simulated (dashed) current distributions. Inset depicts a magnification on the current distributions from 0.8 h to 1.2 h.

695 segment of the discharge, the current spread in the same region increases

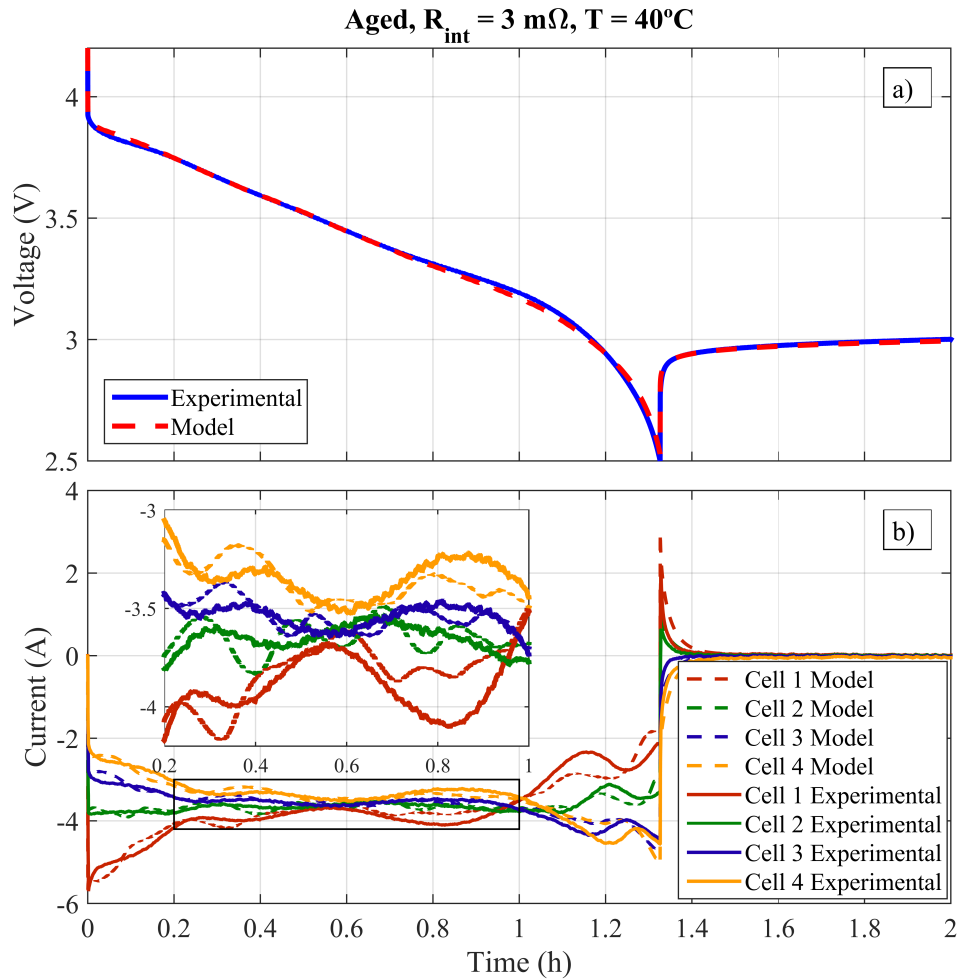


Figure 11: a) Comparison between experimental (solid) and simulated (dashed) module voltage for  $R_{int} = 3 \text{ m}\Omega$  and  $40^\circ\text{C}$ , with cell 4 being the aged cell. b) Comparison between experimental (solid) and simulated (dashed) current distributions. Inset depicts a magnification on the current distributions from 0.2 h to 1 h.

696 due to the presence of the aged cell in this case, with current fluctuations  
697 in cells 1 and 4. Furthermore, the cell providing the largest current towards  
698 the end of the discharge is now cell 3 instead of cell 4, which is detailed in  
699 the inset and correctly predicted by the model. In this case, the largest volt-  
700 age and currents errors occur at  $t=1.19$  h and  $t=1.22$  h, respectively, where  
701 silicon utilization is the greatest.

702 Finally, the effect of the presence of an aged cell can also be studied  
703 separately by analyzing a case where  $R_{int} = 0$  m $\Omega$  (5.4  $\mu\Omega$  in reality), as  
704 shown in Fig. 13 for 40°C. In this case, the current distribution is solely  
705 affected by cell-to-cell variations in capacity. Therefore, negligible differences  
706 are observed for the current split among the fresh cells; however, the aged  
707 cell is only able to provide a slightly lower current. This behavior is sustained  
708 throughout the discharge process, as detailed in the inset, and exacerbated  
709 in the low SOC region. The model is capable of accurately predicting the  
710 current in the aged cell, including its final uptick, as well as the fact that cell  
711 3 counteracts this decrease in current.

712 A summary of model validation is presented in Table 3. Errors in module  
713 voltage and current distribution are broken down into those for just discharge  
714 and relaxation, as well as the combined case, while the value in brackets in-  
715 dicates the maximum error. It is worth noting that consistently accurate  
716 results are obtained for module voltage ( $\leq 20$ mV) when considering just the  
717 discharge as well as when both discharge and the subsequent relaxation are  
718 taken into account, which is ascribed to the appropriate spatial distribution  
719 and parameterization of the employed cell model. Accurate results are also  
720 obtained for the relaxation phase, which we attribute to the consideration

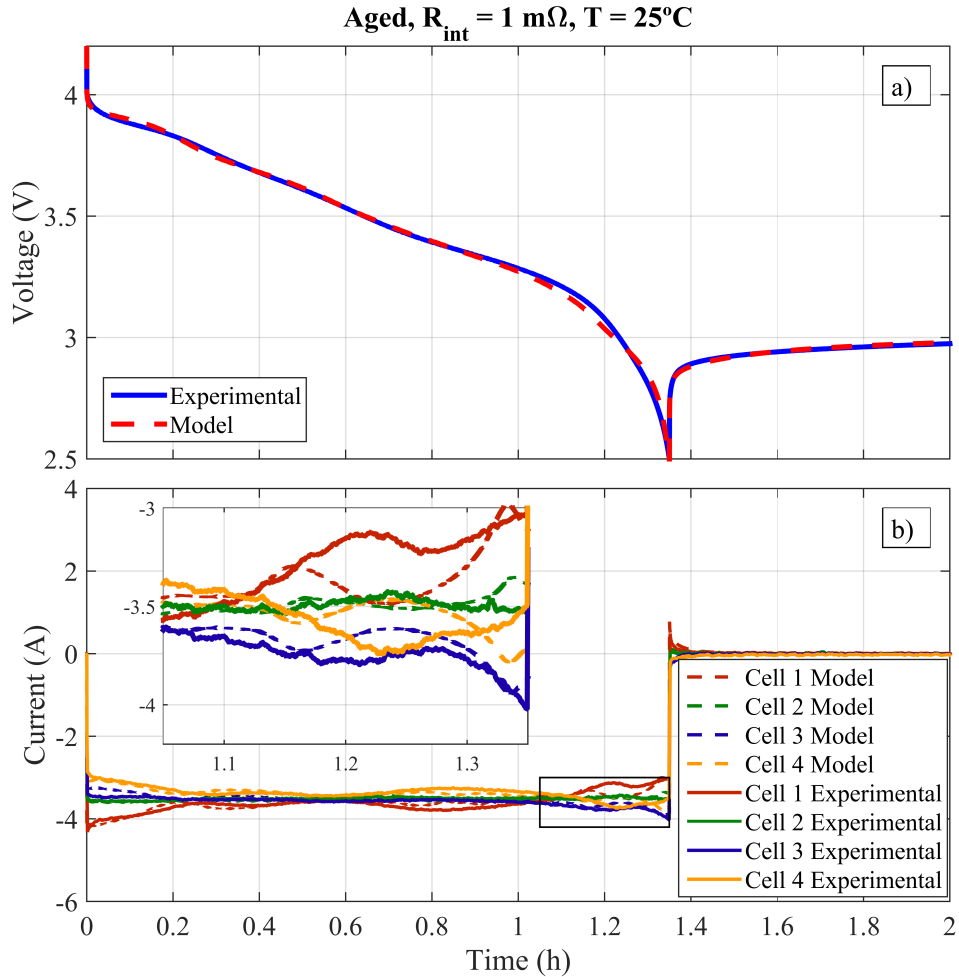


Figure 12: a) Comparison between experimental (solid) and simulated (dashed) module voltage for  $R_{int} = 1 \text{ m}\Omega$  and  $25^\circ\text{C}$ , with cell 4 being the aged cell. b) Comparison between experimental (solid) and simulated (dashed) current distributions. Inset depicts a magnification on the current distributions from 1.05 h to 1.35 h.

721 of a particle size distribution for graphite. Analogously, acceptable results

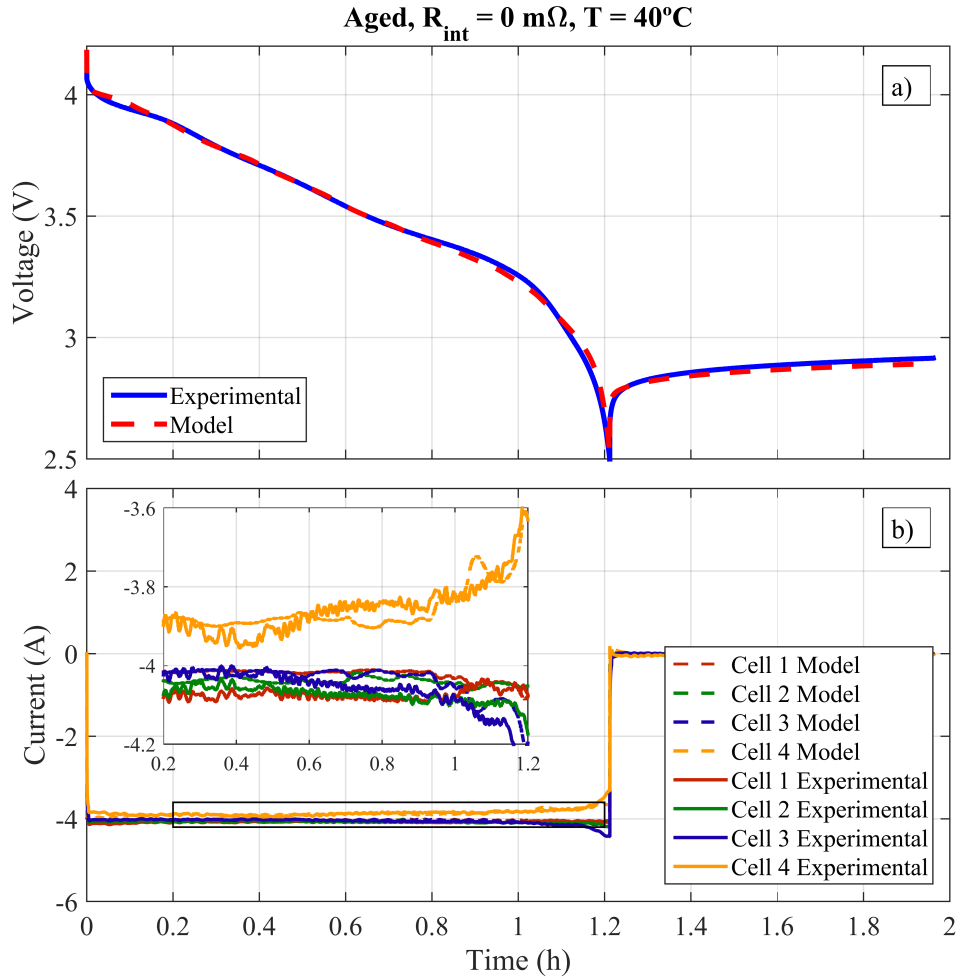


Figure 13: a) Comparison between experimental (solid) and simulated (dashed) module voltage for  $R_{int} = 0 \text{ m}\Omega$  and  $40^\circ\text{C}$ , with cell 4 being the aged cell. b) Comparison between experimental (solid) and simulated (dashed) current distributions. Inset depicts a magnification on the current distributions from 0.2 h to 1.2 h. Experimental currents have been smoothed out in order to improve visualization.

722 are also provided for the current distribution, although in this case, the error  
723 increases with the interconnection resistance. This is an expected outcome,  
724 since a larger interconnection resistance results in greater variability and  
725 complexity in current distribution that make it increasingly challenging to  
726 predict. It is also pointed out that the majority of the error in both voltage  
727 and current is concentrated towards the end of the discharge, due to the chal-  
728 lenge posed by accurately modeling anode behavior in this region because of  
729 silicon utilization, the steep voltage profile of graphite, and the interactions  
730 between both materials. Given that module behavior, characterized by mod-  
731 ule voltage and current distribution, has been validated experimentally with  
732 module current as the only input, the proposed modeling framework enables  
733 obtaining information about individual cell behavior from module-level mea-  
734 surements. For instance, the computed current distribution may be employed  
735 to obtain an estimate of individual cell SOC and module heterogeneity.

736 With respect to computational cost, the presented simulation framework  
737 based on hierarchical network simplification does not require iterative numer-  
738 ical methods, making it computationally efficient. The average simulation  
739 time for the cases considered in Table 3, with a constant timestep  $\Delta t = 0.5$ ,  
740 is 15.3 s per complete simulation run of approximately 2 h of real time, which  
741 equals 1.06 ms per simulation step. Nevertheless, we acknowledge that there  
742 is room for further potential improvements in numerical implementation,  
743 specifically in relation to adaptive timestepping, which lie beyond the scope  
744 of this work. A complementary discussion on numerical stability and scala-  
745 bility is provided in the next section.

Table 3: RMS error results for module voltage and current distribution. The value shown in brackets indicates the maximum error in the same units; the current error value is obtained as the average of the RMS values for all 4 cells. For discharge, the relative current RMS error with respect to the theoretical average current for each case is indicated in brackets. 'Aged' indicates that the aged cell has replaced a fresh cell in position 4.

		25°C			40°C		
		Discharge	Rest	Combined	Discharge	Rest	Combined
Fresh,	Voltage (mV)	17.4	10.7	15.5 (60.1)	19.4	12.6	17.1 (65.8)
$R_{int} = 0 \text{ m}\Omega$	Current (mA)	59.6 (1.8%)	24.0	50.8 (432.1)	75.5 (1.8%)	27.3	61.8 (316.8)
Aged,	Voltage (mV)	18.1	3.0	14.9 (75.1)	18.0	17.9	18.0 (61.2)
$R_{int} = 0 \text{ m}\Omega$	Current (mA)	42.3 (1.4%)	24.1	37.6 (252.3)	55.0 (1.3%)	28.8	47.2 (436.5)
Fresh,	Voltage (mV)	17.5	7.4	14.7 (47.5)	15.3	14.6	15.0 (37.8)
$R_{int} = 1 \text{ m}\Omega$	Current (mA)	92.6 (2.6%)	37.3	77.3 (448.2)	110.5 (2.7%)	50.8	92.2 (484.4)
Aged,	Voltage (mV)	16.4	5.1	13.6 (45.3)	17.0	17.0	17.0 (46.0)
$R_{int} = 1 \text{ m}\Omega$	Current (mA)	83.3 (2.4%)	27.8	69.3 (401.4)	107.4 (2.6%)	45.9	88.9 (620.3)
Fresh,	Voltage (mV)	11.6	6.7	10.0 (27.0)	13.6	10.1	12.5 (49.1)
$R_{int} = 3 \text{ m}\Omega$	Current (mA)	179.8 (4.4%)	93.9	153.1 (765.1)	129.1 (4.0%)	85.3	116.7 (752.2)
Aged,	Voltage (mV)	13.2	3.5	10.5 (32.0)	13.2	5.5	11.1 (38.3)
$R_{int} = 3 \text{ m}\Omega$	Current (mA)	171.6 (4.4%)	94.5	147.1 (712.9)	155.5 (4.3%)	94.6	136.8 (794.7)

746 The module simulation framework presented herein does not include a  
747 thermal submodel; however, in the cases shown above at 25°C and 40°C  
748 ambient temperatures, cell temperature increase is not large enough to con-  
749 siderably alter cell behavior. For instance, the average temperature increase  
750 throughout the discharge is below 2°C, and the maximum temperature in-  
751 crease is below 4°C and only occurs at the very end of the discharge for  
752 some cells. Furthermore, thermal interaction between cell is not expected  
753 to be significant in these cases due to cell spacing. Nevertheless, this as-  
754 sumption leads to inaccuracies in scenarios where cell temperature increase  
755 is significant and effectively modifies cell impedance, such as low ambient  
756 temperatures or high currents, which in turn affects the current distribution.  
757 For example, we have verified that the development of an accurate model  
758 at 10°C ambient temperature requires a coupled electrothermal model; how-  
759 ever, the main challenge in this case lies in an accurate characterization of  
760 thermal boundary conditions, which may be different for each cell, as well  
761 as inter-cell thermal interactions. Therefore, the development of a coupled  
762 electrothermal model of a parallel module, including the computation of heat  
763 generation at the cell level as well as the thermal interaction between cells,  
764 constitutes the subject of further research, for which the proposed framework  
765 may serve as a starting point.

### 766 *5.3. Analysis and discussion*

767 Once the proposed modeling framework has been validated experimen-  
768 tally, the effect of the interconnection resistance  $R_{int}$  on the current distribu-  
769 tion and module voltage is studied through simulation without the impact of  
770 real-world cell-to-cell variations, by employing the average electrode capacity

771 for each cell for increasing values of  $R_{int}$ . A comparison is also established  
 772 with the behavior of the equivalent lumped cell, i.e., a model with electrode  
 773 capacity values four times larger and impedance parameter values four times  
 774 smaller. Simulation results for module voltage and current spread (maximum  
 775 and minimum individual cell current values at each instant) for a 4P module  
 776 are shown in Fig. 14 for  $R_{int} = 0 \text{ m}\Omega$  to  $R_{int} = 5 \text{ m}\Omega$ .

777 Regarding module voltage, it is observed that the lumped cell approxi-  
 778 mation is only accurate in the ideal case where the interconnection resistance  
 779 is precisely  $0 \text{ m}\Omega$ ; a noticeable drop in module voltage is observed even when  
 780  $R_{int} = 1 \text{ m}\Omega$ , which is a realistic value [17]. The effect of the interconnection  
 781 resistance is a more pronounced decrease in module voltage than in discharge  
 782 capacity. In fact, an incremental voltage drop of approximately  $72 \text{ mV}$  on  
 783 average for every  $1 \text{ m}\Omega$  increase in  $R_{int}$  is observed, which translates into  
 784 an incremental  $1.4 \text{ W}$  decrease in delivered power. The current distribution  
 785 shows the characteristic behavior observed in previous experimental results,  
 786 with an initial current spread due to the current step and the nonlinear  
 787 shape of the OCV in the high SOC region. As the OCV-SOC relationship  
 788 becomes approximately linear, the current distribution becomes increasingly  
 789 homogeneous; the steep slope of the OCV at low SOC causes further het-  
 790 erogeneity in the current distribution. The magnitude of the current spread  
 791 is determined by the factor  $\beta$ , with larger values of  $R_{int}$  resulting in greater  
 792 heterogeneity. It is observed that, while the nominal module current is  $1C$ ,  
 793 discharge peaks ranging from  $1.2C$  to  $1.8C$  are reached during the initial seg-  
 794 ment of the discharge due to the effect of  $R_{int}$ . A key consequence of this  
 795 is that module current has to be derated to ensure that no individual cell

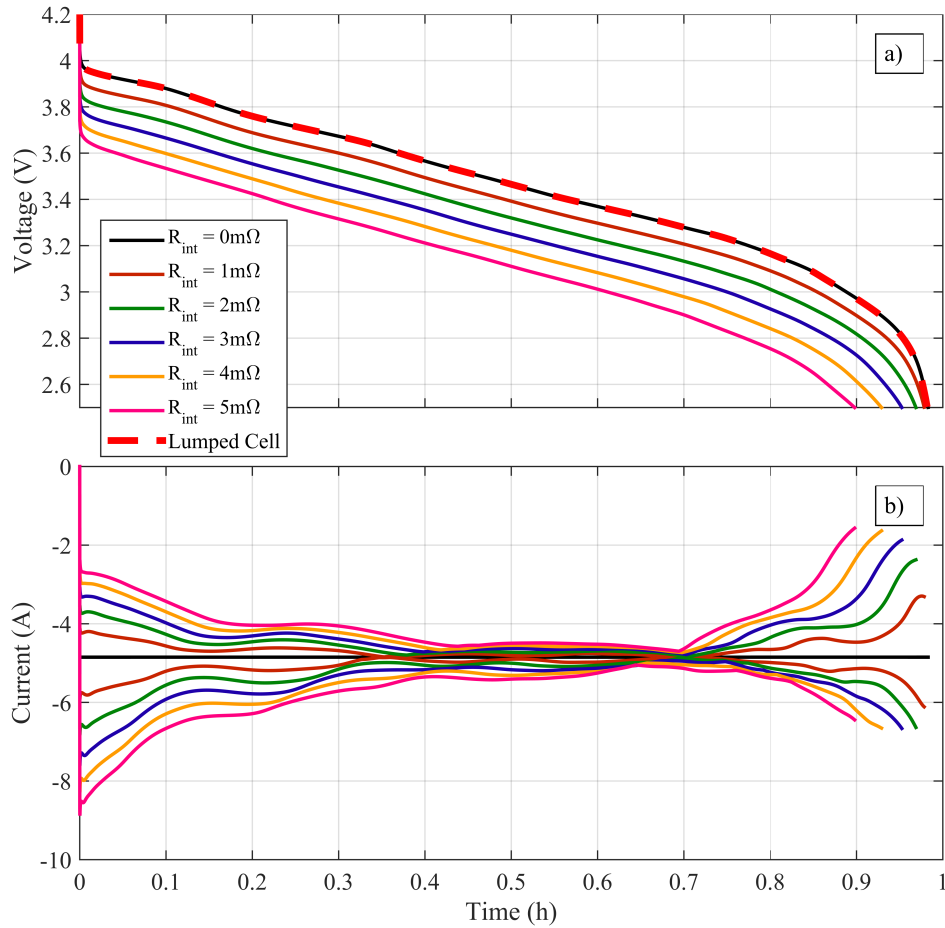


Figure 14: a) Comparison between simulated module voltage for a nominal 1C discharge for  $R_{int} = 0\text{ m}\Omega$  to  $R_{int} = 5\text{ m}\Omega$  for a 4P module. The behavior of the lumped cell is plotted in dashed lines. b) Simulated current spread (maximum and minimum individual cell current values at each instant) for a nominal 1C discharge for  $R_{int} = 0\text{ m}\Omega$  up to  $R_{int} = 5\text{ m}\Omega$  for a 4P module

796 exceeds its respective current limit. Given the complex behavior exhibited  
797 by the current distribution, a further extension of the present work consists  
798 of the development of dynamic derating strategies [75] and the estimation  
799 of power limits [43] as a function of the instantaneous current distribution.  
800 Furthermore, the presence of a nonzero interconnection resistance slightly  
801 increases observability, and makes module behavior sensitive to cell arrange-  
802 ment. For this reason, the observability analysis of the module based on the  
803 presented modeling framework constitutes the basis of further research.

804 In order to highlight the differences between the presented framework  
805 and established alternatives for module simulation based on electrochemical  
806 models, a comparison is made against `liionpack` [50], as well as the integra-  
807 tion of PyBaMM [51] and Simulink. `liionpack` uses PyBaMM models for  
808 the simulation at the cell level, which are replaced by an equivalent circuit,  
809 composed of a voltage source and a series resistance, for the computation of  
810 the current distribution at the module level following the static resistance  
811 approach detailed in Fig. 3. At the time of writing, `liionpack` only allows  
812 simulations with the SPM or SPMe at the cell level, and thus, module simu-  
813 lations employing more comprehensive cell models, such as the DFN or DFN  
814 Si/Gr, are not possible. In order to circumvent this issue, we also follow  
815 the approach outlined in [76] to integrate PyBaMM with Simulink. In this  
816 case, Simulink handles module-level operation, and calls a PyBaMM object  
817 that steps the electrochemical model forward. The interfacing between the  
818 PyBaMM model and Simulink is also done via an equivalent circuit following  
819 the same approach as in `liionpack`.

820 The performance of the proposed framework is compared to that of `liionpack`

821 with the SPM and SPMe, as well as the integration of PyBaMM and Simulink  
822 with the SPM, SPMe, DFN and DFN Si/Gr models for the case of fresh cells  
823 at 25°C with  $R_{int} = 3 \text{ m}\Omega$  and a timestep  $\Delta t = 1 \text{ s}$ , equal to the experimental  
824 sampling time. In order to recalibrate the parameter set in [59] for the partic-  
825 ular cell tested in this work, we follow a similar approach to that employed  
826 by Fasolato et al. [45] for the same purpose, by adjusting active material  
827 volume fractions and leaving dynamic parameters unchanged. The suitability  
828 of this approach is verified by validating the same HPPC protocol, with  
829 results shown in Fig. S2; we understand that a thorough reparameterization  
830 of electrochemical models is beyond the scope of this paper. A more detailed  
831 account of the configuration of PyBaMM models is provided in Appendix B.

832 Experimental data for the case of fresh cells at 25°C with  $R_{int} = 3 \text{ m}\Omega$   
833 are compared to simulation results with the TLM and `liionpack` with  
834 the SPMe in Fig. 15; we have verified that simulations with the DFN give  
835 very similar outcomes with respect to the SPMe for this parameter set, so its  
836 respective results are not plotted to avoid clutter. Regarding module voltage,  
837 it is noted that the SPMe is capable of providing reasonably accurate results  
838 during the CC phase (34.6 mV RMS vs. 11.6 mV RMS for the TLM);  
839 however, the SPMe is not able to match the magnitude and slope of module  
840 voltage from  $t=0.6 \text{ h}$  to  $t=1 \text{ h}$ , which we mainly ascribe to the lack of an  
841 appropriate spatial structure of the electrodes. Furthermore, the module  
842 voltage given by the SPMe shows an excessively abrupt voltage drop towards  
843 the end of the discharge, which is attributed to the absence of a separate  
844 silicon phase in this model. As for the current distribution, the SPMe is also  
845 able to provide reasonably accurate results in terms of RMS current error

846 (5.2% vs. 4.4% for the TLM); however, it is noted that the TLM is capable  
847 of following the experimental profiles more closely, particularly between  $t=0.4$   
848 h and  $t=1$  h, with fewer and less pronounced fluctuations. This is mainly  
849 ascribed to the spatial distribution of the electrodes in the TLM that is not  
850 present in the SPMe, which smooths out cell voltage and results in a more  
851 stable current distribution. We acknowledge that SPMe parameters may  
852 be further optimized for this particular cell; however, we believe that the  
853 differences pointed out are qualitative and not fully explainable by variations  
854 in parameter values.

855 In order to showcase the differences in simulation methodology, the inter-  
856 mediate variables  $V_{eq}$ ,  $R_{eq}$  of each cell are plotted in Fig. 16 for the previous  
857 case. Fig. 16 constitutes a quantitative illustration of the behavior described  
858 in Fig. 3: during discharge, the value of  $OCV$  is higher than that of  $V_{eq}$ ,  
859 which in turn leads to higher resistance values for `liionpack` with respect  
860 to the TLM. This is a direct consequence of how  $V_{eq}$  is computed in both  
861 cases: in `liionpack`,  $V_{eq}$  is just the difference of the OCPs evaluated at  
862 surface concentrations, whereas in the TLM  $V_{eq}$  is determined by the po-  
863 tential distribution along the electrodes, which contains contributions from  
864 ionic transport and charge transfer beyond just the OCP values. Crucially,  
865 the equivalent resistances given by the TLM during discharge are consistent  
866 with the experimental HPPC resistance values in Fig. 7, whereas those given  
867 by `liionpack` slightly overestimate their respective HPPC values shown in  
868 Fig. S2. This highlights the improved consistency between cell and module  
869 behaviors enabled by the TLM.

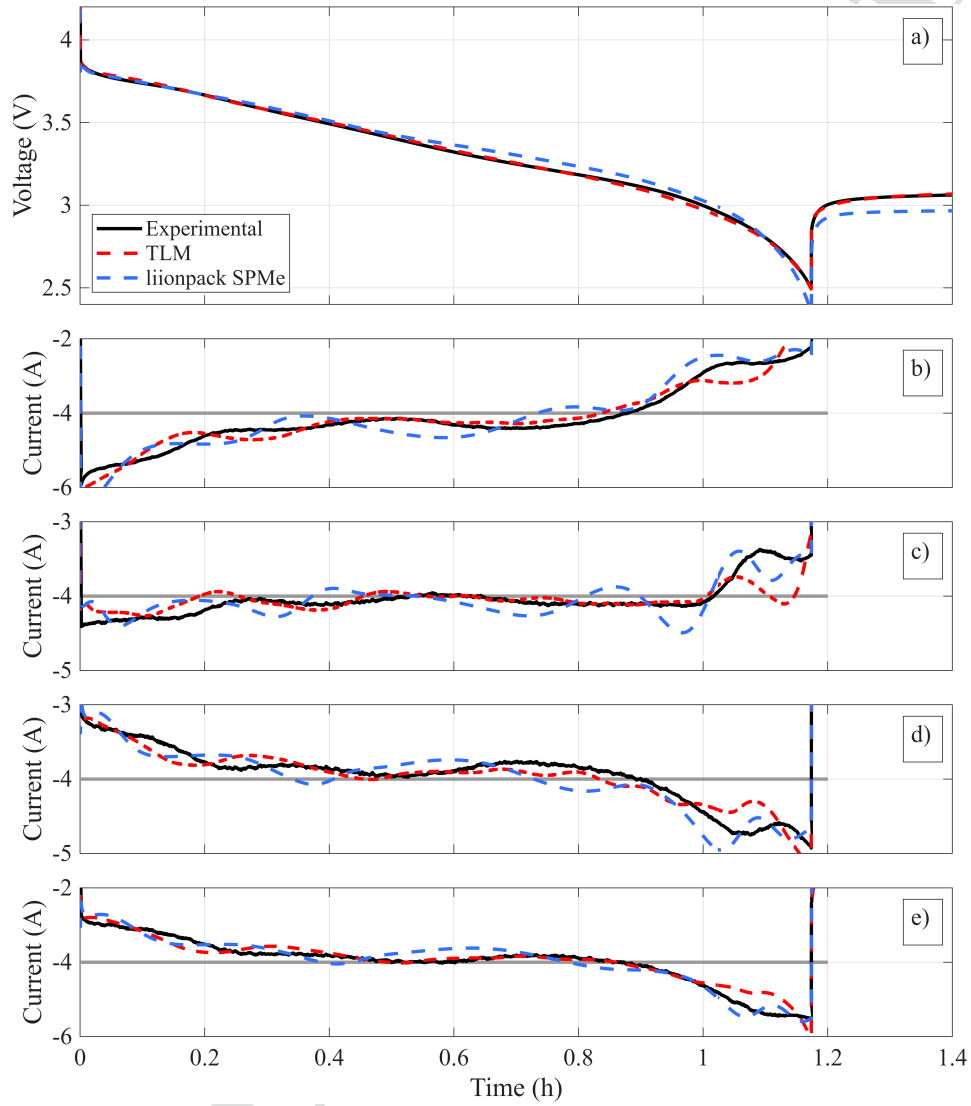


Figure 15: Comparison between experimental results (solid black) and simulation results obtained with the TLM (dashed red) and liionpack with the SPMe (dashed blue) for the case of  $R_{int} = 3 \text{ m}\Omega$  and  $T = 25^\circ\text{C}$ . a) shows module voltage, and b) - e) show individual cell currents during discharge. The horizontal gray line in b) - e) indicates the average cell current during discharge.

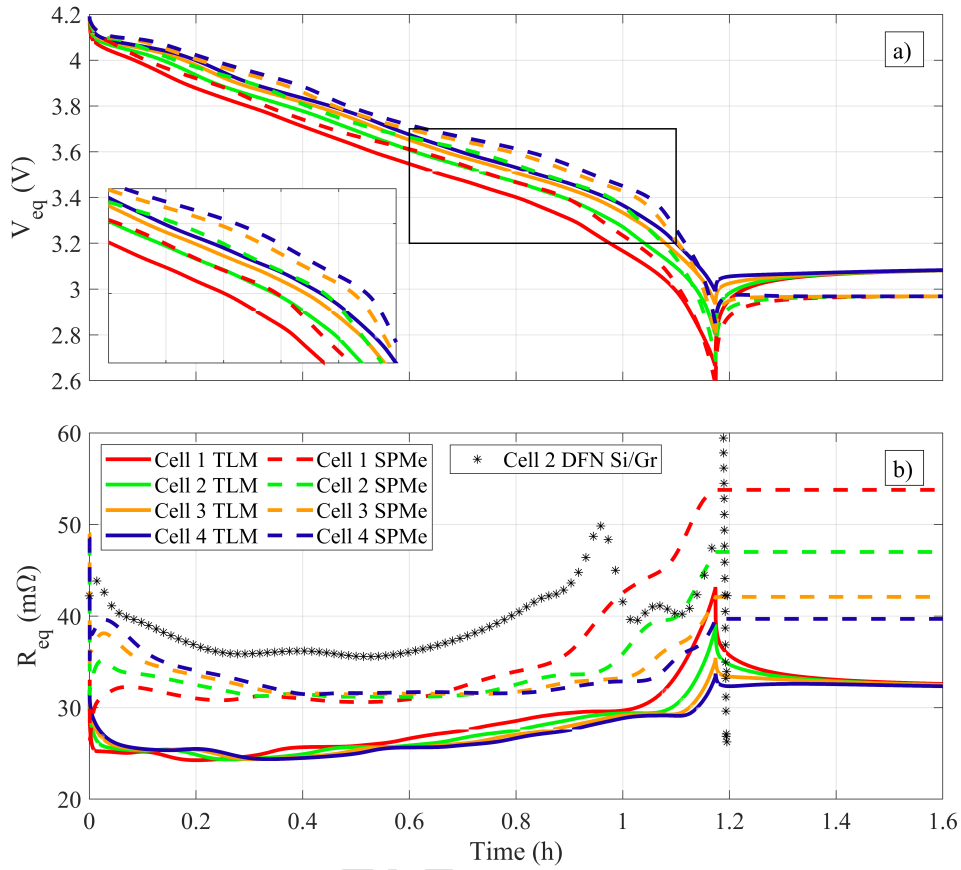


Figure 16: Comparison between the intermediate variables a)  $V_{eq}$  b)  $R_{eq}$  for all 4 cells given by the TLM (solid) and the SPMe with `liionpack` (dashed), with the same color for each cell. The inset in a) shows the equivalent voltages between  $t=0.6$  h and  $t=1.1$  h. Additionally, b) shows the time evolution of  $R_{eq}$  one exemplary cell for the DFN Si/Gr model (black asterisks), where the numerical divergence at  $t=1.2$  h is observed.

870 Furthermore, `liionpack` does not update the calculation of  $R_{eq}$  during  
 871 module relaxation to avoid numerical instability; however, this results in con-  
 872 stant values of  $R_{eq}$  with a substantial spread between cells in a period where

873 they should converge towards the same value due to SOC equalization, as  
874 shown by the TLM in Fig. 16-b). The black asterisks show an example  
875 of the evolution of  $R_{eq}$  obtained with the DFN Si/Gr model with the Py-  
876 BaMM/Simulink integration. In this case,  $R_{eq}$  is always computed as the  
877 static resistance, which leads to numerical instability when cell current is  
878 sufficiently close to zero or opposite sign to cell overpotential as is the case  
879 during module relaxation, and eventually results in convergence failure at  
880  $t=1.2$  h. In summary, the formulation of electrochemical models does not  
881 enable a simple integration into module simulations and requires approxi-  
882 mations for the calculation of  $V_{eq}$  and  $R_{eq}$ , which can lead to inconsistency  
883 and numerical instability. Conversely, these issues are not present in the  
884 TLM-based approach since  $V_{eq}$  and  $R_{eq}$  are intrinsic properties of the model  
885 that only depend on its current state, and can be computed independently  
886 regardless of the operating conditions.

887 Table 4 compares the computational requirements for the considered ap-  
888 proaches for the same simulation case. The TLM enables a more computa-  
889 tionally efficient simulation, on the order of 4 times faster than the SPM and  
890 over an order of magnitude quicker than the DFN and DFN Si/Gr models,  
891 while including a spatial distribution as well as separate silicon and graphite  
892 phases as the latter. The standard PyBaMM discretization consists of 20  
893 points within each particle, and 20 points in each domain, which leads to the  
894 respective number of states for a 4P module indicated Table 4. In contrast,  
895 the approximation to solid diffusion dynamics in Eq. 1 only requires 4 states  
896 per particle, which implies 24 states per electrode and 192 for a 4P module  
897 for the TLM. A separate silicon phase entails 24 additional states per cell,

898 producing a total of 288 states, and two additional particle sizes for graphite  
 899 increase the total count to 480 states. It is noted that the number of states  
 900 for the TLM is the same order of magnitude as the SPM and SPMe while  
 901 providing a physical description closer to that of the DFN Si/Gr.

Table 4: Comparison between computational requirements for the case of a 4P module at 25°C with  $R_{int} = 3 \text{ m}\Omega$ ; all simulations are conducted on an Intel Core i9-10900K processor at 3.70 GHz with 128 GB of RAM. The asterisk (\*) indicates that this simulation failed to converge during relaxation, therefore the corresponding value is only for the simulation time up until that point.

	Time (s)	Number of states
TLM ( $\Delta t = 0.5 \text{ s}$ )	14.3	480 (288/192)
liionpack SPM ( $\Delta t = 1 \text{ s}$ )	56.7	160
PyBaMM/Simulink SPM ( $\Delta t = 1 \text{ s}$ )	57.4	160
liionpack SPMe ( $\Delta t = 1 \text{ s}$ )	60.2	240
PyBaMM/Simulink SPMe ( $\Delta t = 1 \text{ s}$ )	69.5	240
PyBaMM/Simulink DFN ( $\Delta t = 1 \text{ s}$ )	302.7	3440
PyBaMM/Simulink DFN Si/Gr ( $\Delta t = 1 \text{ s}$ )	270.2*	5040

902 The scalability of the proposed approach is assessed in comparison to  
 903 liionpack with the SPMe and a timestep  $\Delta t = 1 \text{ s}$  with an increasing num-  
 904 ber of cells connected in parallel. For the TLM, the timestep and number  
 905 of cells are varied until numerical instability is encountered, and respective  
 906 results are summarized in Table 5. It is pointed out that the TLM-based  
 907 approach outperforms liionpack for smaller modules, and provides com-  
 908 parable performance for larger modules. For instance, a value of 1 ms per

909 simulation step is obtained with the TLM for a 4P module with  $\Delta t = 0.5$   
 910 s, whereas this value is 8 ms per simulation step with `liionpack` for the  
 911 same case. Furthermore, the TLM-based approach scales linearly, with an  
 912 average increase of 2.27 s per additional cell, while `liionpack` scales slightly  
 913 superlinearly, with an average increase of 1.39 s per additional cell. However,  
 914 the larger increase of computation time per cell can be justified by the effects  
 915 included in the TLM that are not present in the SPMe. It is also noted that  
 916 the simulation times in Table 5 are contingent on numerical implementation  
 917 and timestepping; therefore, we leave open the possibility of a more sophisti-  
 918 cated implementation that further increases the computational efficiency of  
 919 the TLM-based approach.

Table 5: Simulation times in seconds for `liionpack` with the SPMe and the TLM-based approach for an increasing number of cells connected in parallel. The dash (-) indicates the cases where numerical instability is encountered for the TLM. All simulations are conducted on an Intel Core i9-10900K processor at 3.70 GHz with 128 GB of RAM.

	4P	8P	12P	16P	20P	24P	28P	32P
<code>liionpack</code> (SPMe, $\Delta t = 1$ s)	28.97	33.54	38.40	43.57	49.00	54.75	60.98	67.72
TLM ( $\Delta t = 0.6$ s)	6.23	12.28	-	-	-	-	-	-
TLM ( $\Delta t = 0.5$ s)	7.48	14.74	22.24	29.46	-	-	-	-
TLM ( $\Delta t = 0.4$ s)	9.32	18.36	27.70	36.73	45.83	55.05	63.90	72.90

920 Lastly, lumped ECMs are a common alternative for the simulation of  
 921 parallel-connected cells. For this reason, we include a final comparison be-  
 922 tween the TLM and the 2RC ECM regarding module voltage and current  
 923 distribution for the case of  $R_{int} = 1$  m $\Omega$  at 25°C, in order to illustrate the  
 924 differences in the module operation given by lumped and distributed ECMs.

925 In order to characterize the 2RC ECM, the OCV-SOC relationship and cell  
926 capacities are taken from respective thermodynamic tests, and dynamic pa-  
927 rameters are adjusted according to the HPPC test carried out at 25°C. A  
928 key drawback of standard ECMs is that they require empirical dependen-  
929 cies of parameters with SOC, shown in Fig S3, which are implemented via  
930 interpolation between values determined experimentally at each SOC.

931 Results are shown in Fig. 17. Regarding module voltage, the 2RC ECM  
932 is able to provide reasonably accurate results for a considerable portion of the  
933 discharge, but yields large errors at low SOCs, resulting in an error of 35.6 mV  
934 RMS for the CC phase vs. 11.6 mV RMS for the TLM. However, the most  
935 interesting difference in behavior is observed in the current distribution. A  
936 spatially distributed cell model results in a smoother voltage response, which  
937 in turn leads to a more stable current distribution. Conversely, in a lumped  
938 cell model, the voltage profile is directly dictated by the shape of the OCV.  
939 Consequently, the 2RC ECM is capable of providing accurate results for the  
940 current distribution in the SOC regions where the OCV is approximately  
941 linear. Nevertheless, when a cell enters a SOC region where the OCV is  
942 nonlinear, fluctuations in the current distribution are induced to balance  
943 module voltage, which are mainly observed in the low SOC region. This  
944 highlights the importance of an appropriate spatial distribution at the cell  
945 level, and indicates that lumped ECMs may give less accurate predictions of  
946 the current distribution for chemistries where the nonlinearities of the OCV  
947 are more pronounced.

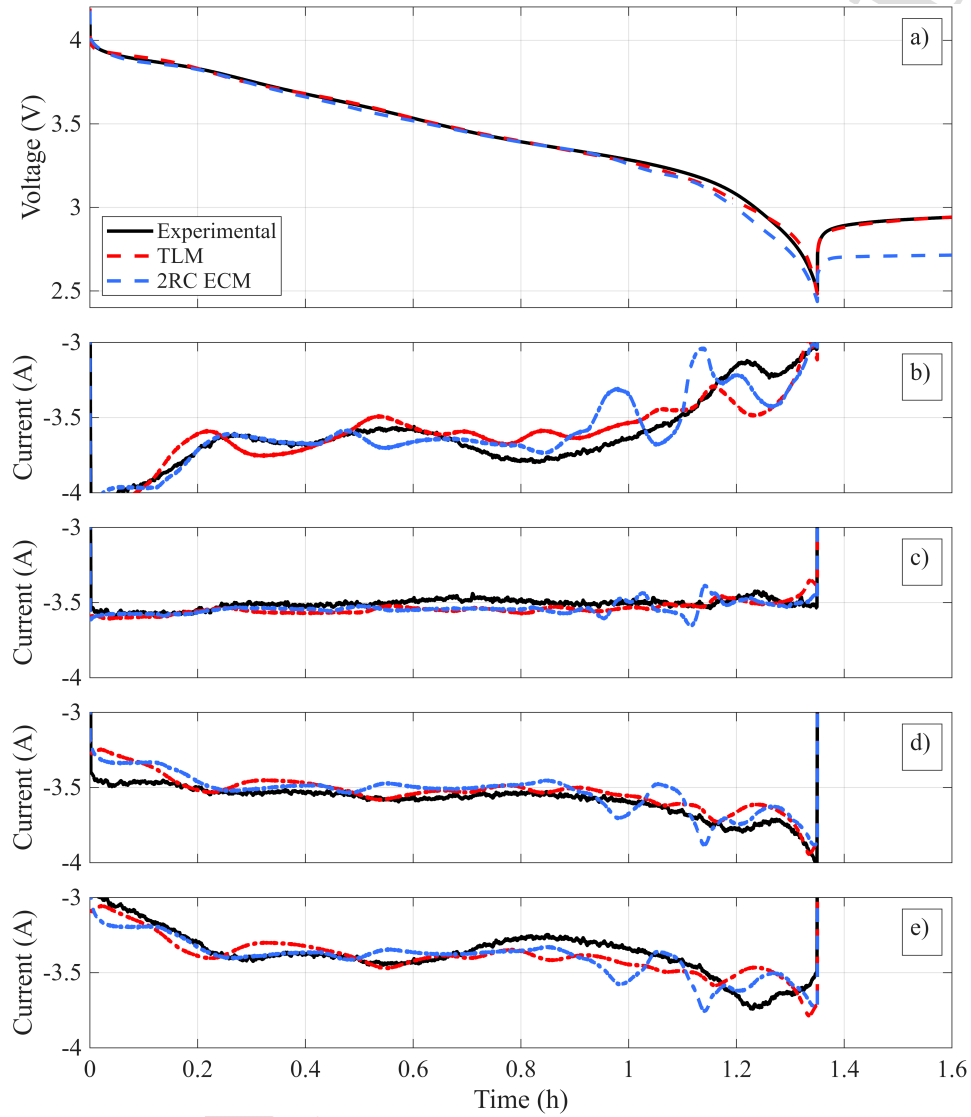


Figure 17: Comparison between experimental results (solid black) and simulation results obtained with the TLM (dashed red) and the 2RC ECM (dashed blue) for the case of  $R_{int} = 1 \text{ m}\Omega$  and  $T = 25^\circ\text{C}$ . a) shows module voltage, and b) - e) show individual cell currents during discharge. The horizontal gray line in b) - e) indicates the average cell current during discharge.

948 In any case, the current distribution obtained with the 2RC during dis-  
949 charge exhibits comparable accuracy (2.5% vs. 2.4% for the TLM), which  
950 underscores the importance of a simultaneous validation of both module volt-  
951 age and current distribution, as the constraint imposed by the total module  
952 current may mask the inaccuracies in the description of cell dynamics. Fi-  
953 nally, although the 2RC ECM is lightweight, its computational efficiency is  
954 dependent on the procedure through which the OCV and dynamic parame-  
955 ters are interpolated for each SOC.

## 956 6. Conclusions

957 In this article, a method for the consistent calculation of the equivalent  
958 voltage and resistance of a battery cell is presented according to the physi-  
959 cally motivated discrete transmission line model, which enables the efficient  
960 simulation of a module composed of cells connected in parallel. The proposed  
961 modeling and simulation methodology is validated experimentally against a  
962 dataset of a 4P module with the interconnection resistance, ambient temper-  
963 ature, and the presence of an aged cell as input parameters, with accurate  
964 results for module voltage ( $\leq 20$  mV RMS) and current distribution ( $\leq 4.4\%$   
965 RMS). The interconnection resistance is identified as a key factor, with even  
966 small values leading to a significant drop in module voltage and an increas-  
967 ingly heterogeneous current distribution.

968 The presented methodology is compared to established approaches for  
969 the physically motivated simulation of battery modules. The consistency be-  
970 tween module-level behavior and cell-level dynamics enabled by the proposed  
971 framework is emphasized, as well as its improved computational efficiency.

972 The key differences between the module behaviors given by lumped and dis-  
 973 tributed ECMs are shown to be a more accurate description of module voltage  
 974 and a more stable current distribution in the latter case.

975 In conclusion, we understand that the proposed approach based on the  
 976 transmission line model and hierarchical simplification constitutes a suitable  
 977 methodology for the physically motivated simulation of parallel-connected  
 978 cells, thus overcoming the challenging integration of electrochemical models  
 979 into module simulations.

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#### 989 Appendix A. Open Circuit Potentials for the TLM

$$\begin{aligned}
 OCP_{NMC}(x) = & 3.501 + 0.36 \cdot (1 + \exp[4.1x] \cdot (0.067948x - 0.0845206)) + \\
 & 0.0208 \cdot (1 - \exp[46x - 46]) + \frac{0.030}{1 + \exp[41x - 34.194]} + \\
 & \frac{0.194}{1 + \exp[14x - 6.062]} + \frac{0.113}{1 + \exp[42x - 9.156]} + \frac{0.43}{1 + \exp[46x + 0.138]} \quad (A.1)
 \end{aligned}$$

$$\begin{aligned}
OCP_{Gr,delith}(x) = & 0.06 + 0.0305 \cdot (1 - \exp [34x - 34]) + \frac{0.048}{1 + \exp [39x - 22.234]} \\
& + \frac{0.075}{1 + \exp [55x - 9.24]} - \frac{0.02}{1 + \exp [55x - 6.6]} \\
& + \frac{0.254}{1 + \exp [102x - 5.916]} + \frac{0.163}{1 + \exp [130x - 3.77]} + \frac{0.83}{1 + \exp [120x - 0.444]} \quad (A.2)
\end{aligned}$$

$$\begin{aligned}
OCP_{Si,delith}(x) = & 0.022 + 0.010 \log [-100x + 100.01] - 0.069 \log [0.996016x + 0.0398406] + \\
& 0.033 \cdot (1 - \exp [1650x - 1650]) \cdot (-4300x + 4301) + 0.088 \cdot (1 - \exp [21x - 21]) + \\
& \frac{0.099}{1 + \exp [12x - 8.1]} + \frac{0.0084}{1 + \exp [25x - 6.25]} + \frac{0.20}{1 + \exp [66x - 0.594]} \quad (A.3)
\end{aligned}$$

## 990 Appendix B. Notes on the configuration of PyBaMM models

991 The PyBaMM models integrated into Simulink are from PyBaMM 22.8.  
992 For the DFN and DFN Si/Gr models, the format of the Simulink object provided  
993 in [76] is modified to match that of the PyBaMM integrator. For the  
994 DFN model,  $V_{eq}$  is computed as the difference between the electrode-averaged  
995 values of the OCPs. For the DFN Si/Gr model, the equivalent voltage of the  
996 negative electrode is computed by means of a weighted average of the OCPs  
997 of both materials according to their relative contribution to anode capacity.  
998 We acknowledge that this is an arbitrary choice; however, there is no  
999 workaround for this issue since there are two distinct values for the electrode-  
1000 averaged surface OCP. Finally, silicon hysteresis has been manually disabled  
1001 for the DFN Si/Gr model by only making use of the silicon delithiation OCP,  
1002 otherwise numerical convergence at the module level is severely hindered.

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Highlights:

- Physically motivated efficient simulation framework for parallel-connected cells
- Determination of a cell's electrically consistent equivalent voltage and resistance
- Accurate results for module voltage ( $<20\text{mV RMS}$ ) and current distribution ( $<4.4\% \text{ RMS}$ )
- Experimental validation covering a wide range of operating scenarios
- Improved consistency and computational cost with respect to established approaches

**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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