Supplementary information

Data from other sources

The bulk of the data and results used are dependent on data from other sources. Thus, it is important to provide information on which data has been used, and how they have been digitised and converted to be used in the parameter fits. That data are also uploaded on the github page, see .dat and .xls files.

Figure 1

Li et al, 2019 [30]: The relevant data have been taken from Figure 8 of the respective work, which presents the reversible deformation with respect to the percentile state-of-health. Firstly, the percentile SOH [%] is transformed to 0≤SOH≤1. Then, damage is defined as $\omega = 1 - \mathrm{SOH}$. The reversible deformation is divided by the battery thickness reported in Table 2, to return the reversible strains.

Cannarella and Arnold, 2014 [31]: This work presents a constant thickness mechanical constraint. This translates into stress changes that would correspond to thickness changes that would occur without the rigid constraint. In Figure 4 of the respective work, the authors present levels of cell expansion normalized by charge $\mathrm{d}\varepsilon/\mathrm{d}Q$ [%/Ah], for each stress magnitude classification. In the next step, normalized capacity is transformed into damage as: $\omega = 1 - \mathrm{SOH}$ and $\mathrm{SOH} = Q(N)/Q_0$ as current capacity divided by the nominal capacity. Using the equations above, one can calculate the reversible strain amplitude $\Delta\varepsilon_{\mathrm{rev}} = \frac{d\varepsilon}{dQ}Q(N) = \frac{d\varepsilon}{dQ}Q_0(1-\omega)$. Nominal capacity can be found in the experimental section of the paper.

Aufschläger et al, 2024 [32]: In Figure 5 of the respective work, the authors present reversible and irreversible thickness during cycling. The lower limit of each coloured area is the discharged state, whereas the upper limit is the charged state. This means that from the difference of the two limits one can get the reversible thickness change with respect to the number of equivalent full cycles. Similarly, the lower limit gives the irreversible thickness which is used as well. The digitisation process consists of getting the two limits, computing their difference $\Delta H_{\mathrm{rev}}(N)$ and then divide by the nominal thickness presented in Table 1 to get the reversible strain amplitude. Capacity transformed to damage ω as described above.

Material parameter for the irreversible strain

Li et al, 2019 [30]: Same procedure as shown above for Fig.1 but now using Figure 10 of the respective work.

Li et al, 2021 [2]: Figure 5a of the respective work shows the linear increase of irreversible strain with SOH. The slope return q_{irrev}.

Aufschläger et al, 2024 [32]: Digitisation process was described above. Lower limit of the coloured area was used as mentioned.

Hahn et al, 2021 [8]: The expression for irreversible swelling presented in this manuscript is directly related to the expression from Hahn et al. for the unconstrained cell thickness, $l_{C,0,aged} = l_{C,0,BOL} + G(1 - SOH)$. Since their model is in thickness and this work in strain, one can define $\varepsilon_{irrev} = \frac{l_{C,0,aged} - l_{C,0,BOL}}{l_{C,0,BOL}}$ and then $q_{irrev} = \frac{G}{l_{C,0,BOL}}$, and, finally, use the fit they performed for G.

Mohtat et al, 2021 [36]: In Figure 2a of the respective work the irreversible thickness change with respect to capacity retention are reported. Capacity retention is first transformed into SOH and then damage, whereas irreversible strain is calculated by dividing the irreversible thickness change with the cell thickness from Table 1. Again, the slope gives $q_{\text{irrev.}}$

Figure 2

Deich et al, 2021 [7]: In Figure 4a) of Deich et al, the graph reports SOH%(Maximum Pressure in MPa). The data is grouped and coloured differently for each Cycle Number starting from BOL and reaching 1000 cycles with 200 cycles intervals. This gives rise to a total of 6 datasets, but here BOL is excluded for this expression, resulting in 5. For each pressure, five SOH are reported which are transformed into damage ω. Then, since $p_{\text{min}} \approx 0$ as described in the main text, $\Delta P = p_{\text{max}}. \frac{d\omega}{dn}$ is calculated for each pressure and damage value using a central differences scheme based on the values from the plot. Curve fit gives the presented results for the material parameters.

Validation figures

Model validation is based on datasets presented in the respective works. In the following paragraphs, the figures used, and the digitisation process are summarised.

Cannarella and Arnold [31]: Their work presents three different cases a) high stack pressure, b) medium stack pressure, and c) low stack pressure. Stress evolution with respect to time and number of cycles is presented in figures 2 and 3. The dotted lines in each of these figures represent the maximum and minimum pressure for this case, whereas the bold line is the average evolution. For the pressure evolution comparison, since in this manuscript, the maximum pressure is presented, the maximum yellow line (from the medium stack pressure case of Fig. 3) is digitised using the MATLAB tool digitize2.m (https://matlab.mathworks.com/open/fileexchange/v1?id=928, accessed on 1 June 2024). Similarly, the SOH evolution is digitised by the Normalized Capacity vs Cycle Number Fig. 5 of the respective work.

Deich et al [7].: Details of their work are reported in the main text. The SOH evolution is digitised by the Relative C/10 Discharge Capacity (%) vs Cycle Number Fig. 2 a) of the respective work, again using the MATLAB tool digitize2.m (https://matlab.mathworks.com/open/fileexchange/v1?id=928, accessed on 1 June 2024). In their Fig. 2 c) both minimum and maximum pressure are presented for all the cases considered. Since $P_{\text{min}} \approx 0$, the digitisation is targeting P_{max} (full lines).

The digitised data used for the validations are published online.

Useful figures:

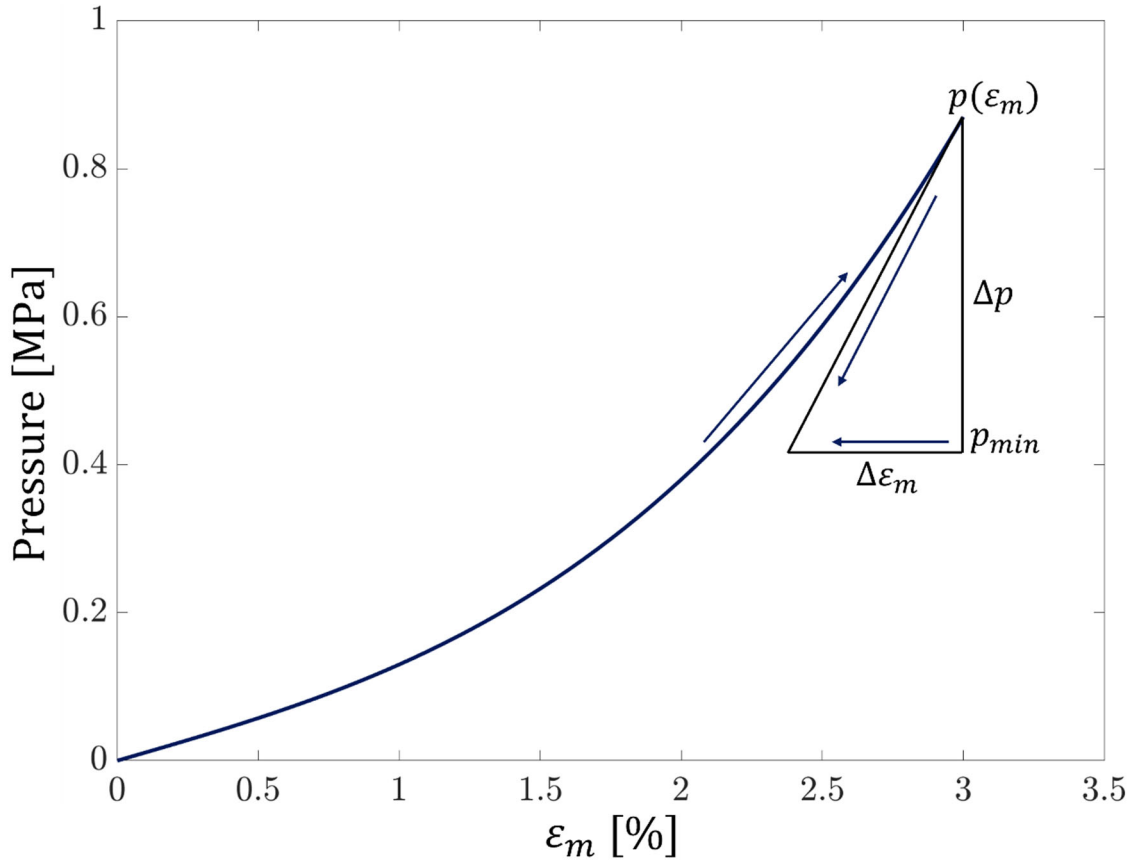


Figure S1. Pressure evolution illustration using our constitutive law with the fitted values from Deich et al. and illustration of the unloading path.

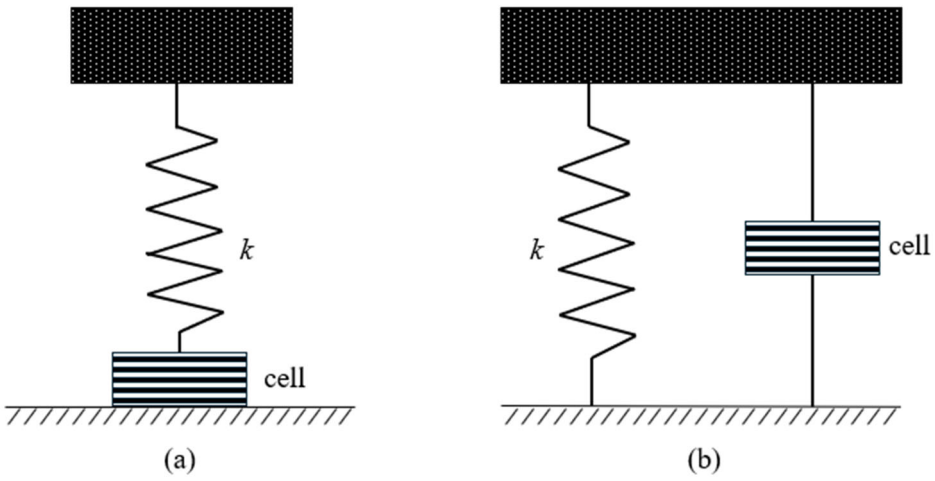


Figure S2. Illustration of different experimental boundary constraint equivalents: a) spring in series with the cell, b) spring in parallel with the cell.