

Mechanics of Lithium-Ion Batteries: Aging and Diagnostics

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Executive Summary

This work investigates the mechanics of lithium-ion batteries both from the aging point of view, and as a tool to diagnose the battery health and degradation mechanisms in real-world scenarios.

This work presents POLIDEMO, the first battery aging model explicitly addressing mechanical degradation phenomena. The model provides a physics-based description of the coupled electrochemical and mechanical aging processes in lithium-ion batteries. It enables the prediction of key degradation indicators, including capacity fade—capturing the knee point behavior—and the irreversible thickness increase associated with long-term aging. .

Mechanics is not only an issue in lithium-ion batteries: the close correlation between the lithiation of the electrodes and the deformation of the battery allows to define algorithms for battery diagnostic and state of charge estimation based on the measurement of the battery deformation. The strength of mechanical-based algorithms with respect to traditional voltage-based algorithms is the absence of current-dependency, making mechanical-based algorithms working excellently even with high currents, typical of real-world scenarios.

Keywords: Batteries, Battery Management System, Modelling & Simulation, Health and Safety Considerations

1 Introduction

During operation, lithium-ion batteries experience a macroscopic deformation due to the deformation induced by the electrochemical processes, involving the interaction of lithium ions with the crystal structure of the active material of electrodes. Such deformation, originating at the atomic scale, has an impact on the macroscopic structural deformation of the battery (Fig. 1a-b), which can be both mathematically computed [1, 2] and measured experimentally with dedicated sensors [3, 4, 5, 6, 7, 8]. In particular, batteries swell during charge, and contract during discharge, following the deformation the negative electrode being lithiated (expansion) and delithiated (contraction), respectively. The deformation of the positive electrodes used in commercial batteries is lower than graphite (or silicon), then it counterbalance the deformation of the negative electrode but it does not overcome it. This makes the macroscopic deformation of the battery to have the same trend of the negative electrode deformation. This deformation occurring during charge and discharge is referred to as reversible deformation, as the the same amount of expansion in a full charge cycle 0%-100% state of charge (SOC), is completely recovered during the discharge back to 0% SOC.

An irreversible deformation is observed during aging on the top of the reversible deformation, meaning that the thickness of the battery continuously increases during aging [9, 8, 6, 5, 10] because of degradation phenomena, such as the SEI growth and the associated gas generation.

The interaction of lithium ions with the electrode microstructure does not only cause the macroscopic deformation of the battery, but also the mechanical degradation of the electrodes at microscale. Indeed, the insertion of lithium ions in the crystalline structure of the active material causes the so called diffusion induced stress [11, 12, 13, 14] in the active material particles of the electrode. This stress leads to crack propagation in the particles [15, 16, 17], resulting in the disconnection of portion of active materials from the rest of the electrode (loss of active material) and in the accelerated solid electrolyte interface (SEI) growth over the cracks surfaces because of the contact with the electrolyte. These phenomena leads to the performance decay of the battery during aging, such as capacity fade and resistance increase.

For this purpose, a mechanical-electrochemical degradation model has been studied to model the coupling between the mechanical and electrochemical degradation processes. This study resulted in the implementation of POLIDEMO [18], an open-access software for lithium-ion battery degradation modeling. The key features of the model are three: (a) The innovative approach to model mechanical degradation and its relationship with loss of active material, the increasing tortuosity and the subsequent resistance increase ultimately; (b) The ability to predict the occurrence of the knee point in the capacity loss curve; (c) The ability to calculate the irreversible swelling of the battery during aging.

Mechanics is not just an issue in lithium-ion batteries. Indeed, the reversible deformation of the battery provides useful insight in the battery internal states that can be leveraged for innovative battery diagnostic methodologies [19], with some advantages over traditional voltage-based methods.

The reversible expansion of the battery is proportional to its SOC, as visible in Figure 1b, and it is slightly affected by the applied current, differently from voltage. These observations led to the development of POLISOC, a mechanical based estimation algorithm that leverages the reversible deformation of the battery to estimate SOC. It is shown that the SOC can be estimated by inverting the thickness-SOC relationship even in dynamic condition with rmse in the order of 2%. Furthermore, a hybrid Kalman filter is implemented, running both with deformation and voltage measurements, besides the current measurements for coulomb counting. The application of the algorithm for SOC estimation in LFP, LCO and NMC batteries shows promising results. The key advantages are two: (a) The estimation of SOC in LFP batteries that suffer the flat SOC-open circuit voltage (OCV) relationship and the OCV hysteresis; (b) The SOC estimation in aged batteries, where the high resistance increase occurring during aging negatively affects the performance of the electrical equivalent circuit model in the kalman filter for the voltage calculation.

Mechanical measurements can be used to diagnose the battery health as well, through the estimation of degradation mechanisms. The phase transition occurring during the electrode lithiation leave a trace in the deformation characteristic (deformation vs state of lithiation) of the electrode, visible on the macroscopic battery deformation measurement. Then, these traces appear as peaks when computing the derivative of deformation with respect to SOC - differential expansion - in analogy to the differential voltage methodology. The main advantage with respect to the traditional voltage based methodology is that deformation does not suffer polarization, then mechanical-based method are applied also at high current in industrial relevant applications, and not just at very low current in lab applications as voltage based methods.

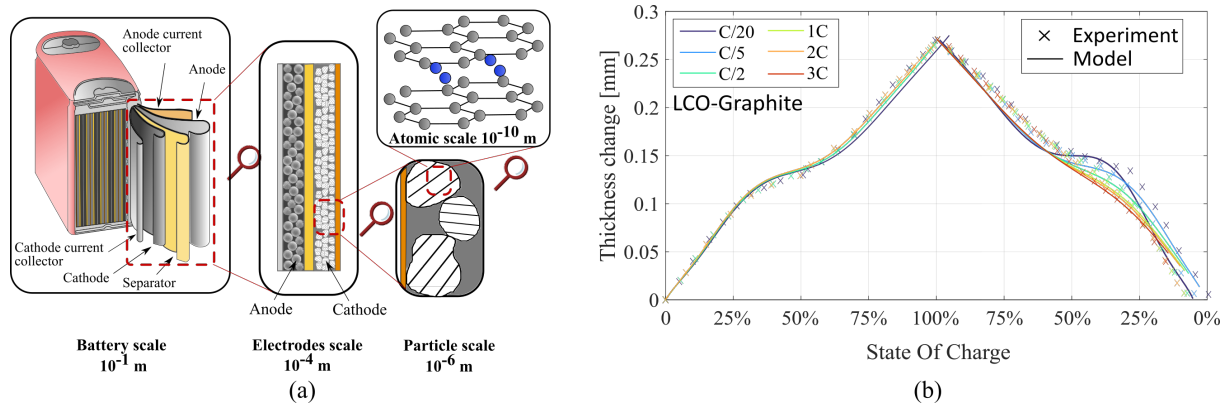


Figure 1: (a) Multi-scale structure of lithium-ion batteries; (b) Reversible deformation of an LCO battery in a single charge/discharge cycles as a function of current rates.

2 Experiment

Two kind of experimental tests have been carried out: 1) Mechanical characterization tests to study the reversible deformation of the battery as a function of state of charge and different charging/discharging currents; 2) Aging tests to track how the electrical performances (capacity and resistance) as well as mechanical performances (reversible and irreversible deformation) change with aging.

The most common battery chemistries (LFP, NMC, LCO) have been mechanically characterized, whereas aging tests have been carried out on LCO batteries.

2.1 Mechanical characterization

The reversible deformation of LFP, NMC and LCO batteries (the first two prismatic and the latter pouch) has been measured with the experimental setup explained in detail in the authors' previous works [3, 4]. Briefly, batteries undergone constant current full depth of discharge cycles followed by constant current/constant voltage charge cycles. Current rate spans from C/20 up to 3C in discharge and from C/20 up to C/2 during charge. The battery deformation is measured with laser displacement sensors measuring the thickness change of the battery. In particular, the couple of laser sensors measure the displacement of the central point of the two larger battery surfaces, so that the sum of the two displacement measured is the thickness change of the central point of the battery. The reversible deformation measured on LFP, NMC and LCO batteries during charge and discharge is reported in Figure 2.

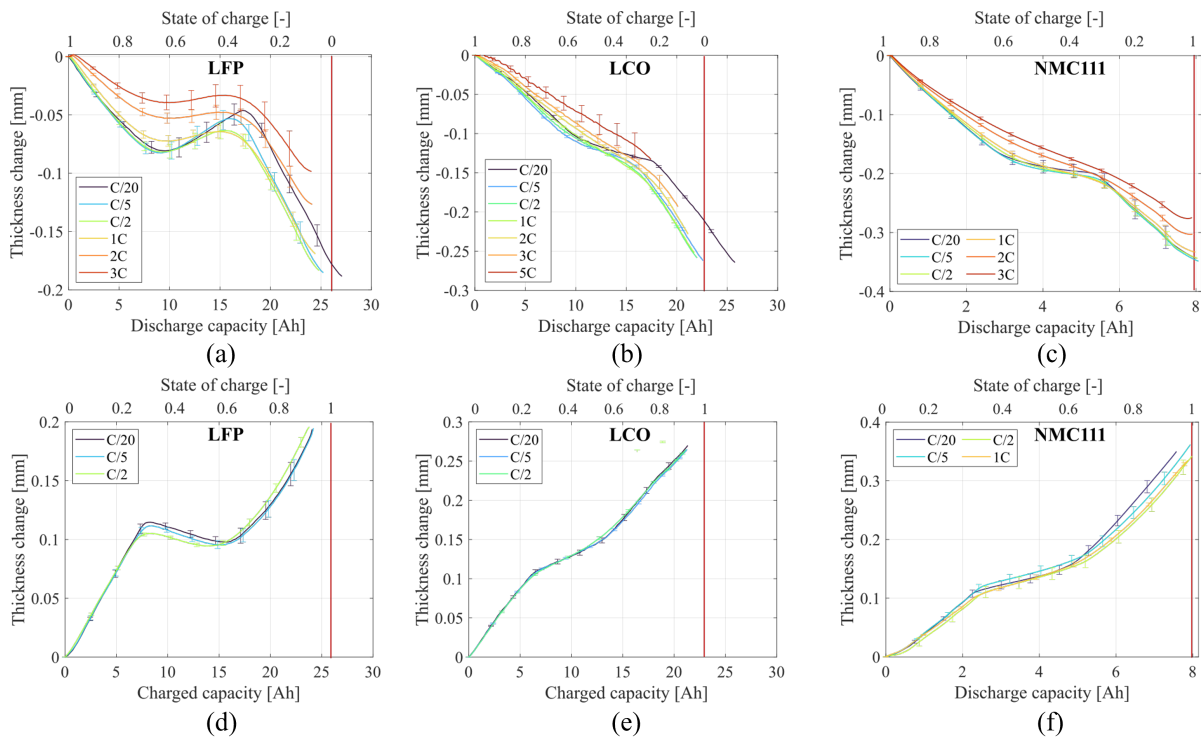


Figure 2: Reversible deformation measurements at different current rates during discharge (a-c) and charge (d-f) cycles in LFP, LCO and NMC batteries.

2.2 Aging tests

Aging tests are carried out on LCO batteries to track how electrical performances (voltage profile, capacity and resistance), thermal performance (temperature profile), and mechanical performances (reversible and irreversible deformation) change with aging.

The aging test protocol consists of alternating aging cycles and reference performance tests. In particular, reference performance tests consisting in 3 full DOD charge-discharge cycles at high rate (C/2 charge - 1C discharge) are performed each 25 aging cycles. Voltage, temperature and deformation responses at high rate cycling are obtained from this reference performance tests. Irreversible deformation is obtained measuring the battery thickness each 25 aging cycles. Low rate charge-discharge cycles and hybrid pulse power characterization tests (HPPC) are performed each 50 aging cycles to obtain the voltage and deformation responses at low rate and the Ohmic and diffusion resistances.

Figure 3a reports the capacity loss measured from the high and low rate reference performance tests, as well as the calendar aging measured on other battery samples not subjected to aging. Figure 3b reports the trend of the Ohmic resistance during aging. Figure 3c reports the reversible deformation measured from the high and low rate reference performance tests. Finally, Figure 3d reports the irreversible deformation of the battery through aging.

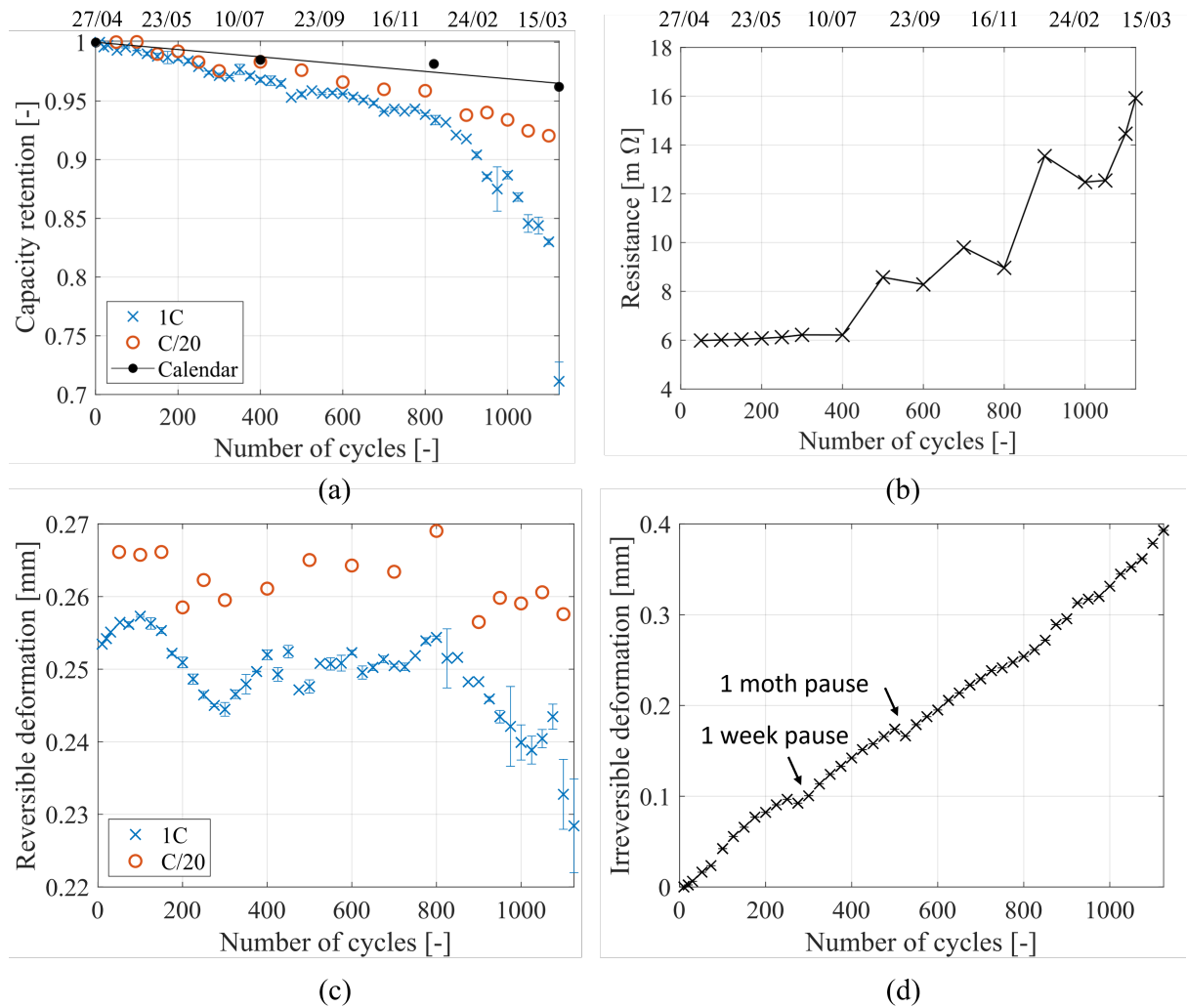


Figure 3: Electrical and mechanical performance change through battery life. (a) Capacity loss, (b) Ohmic resistance, (c) Reversible deformation, (d) Irreversible deformation.

3 Aging Diagnostics and State of Charge Estimation with Mechanical Measurements

Algorithms for the on-line battery diagnostics have been developed and patented [20] aiming to estimate SOC, state of health (SOH) and to identify the degradation mechanisms (loss of active material and lithium inventory) from in-operando mechanical measurements. These deformation-based algorithms leverage the close correlation between the electrode lithiation and the battery deformation, overcoming the difficulties of voltage-based algorithms when dealing with high current because of polarization. The final goal is to provide health and the degradation estimation as well as improved SOC estimation when operating with fast charging profiles, typical of the automotive sector.

3.1 State of Charge estimation - POLISOC

The SOC estimation algorithm based on mechanical measurements relies on the fact that the reversible deformation of the battery during operation is directly linked to the amount of lithium ions in the electrodes, that is the true indication of the SOC. This makes the thickness of the battery to be directly proportional to its charge: higher the charge of the battery, higher its thickness. This behavior is reversible, making the battery to contract during discharge, so that a certain thickness corresponds to a certain SOC. Furthermore, this characteristics remains unchanged also when the battery ages.

The state of charge-thickness (SOC-THK) relationship of LCO and NMC batteries is almost linear (thus monotonic), as shown in Figure 2b,c,e,f, making possible to accurately estimate SOC just inverting the

SOC-THK curve. Then, a SOC value is attributed according to the actual thickness of the battery, even in dynamic condition like DST and drive cycles current profile with currents up to 3C. LFP batteries have a non monotonic SOC-THK relationship, as shown in Figure 2a,d, thus an Extended Kalman Filter (EKF) is developed to estimate SOC on the basis of the deformation measurement.

In this regard, an hybrid SOC estimation algorithm, called POLISOC, is created [21]. POLISOC can estimate SOC in three modes: 1) The traditional EKF approach with current (Coulomb counting) and voltage measurement as explained in reference textbooks [22]; 2) The mechanical EKF approach where current measurement for Coulomb counting are combined with deformation measurement, in analogy with the approach 1; 3) The hybrid EKF approach, where current measurement for Coulomb counting are combined with voltage and deformation measurement.

The logic of the algorithm, as all the EKF algorithm for SOC estimation, is that current measurements give the time evolution of the SOC with Coulomb counting. Coulomb counting alone is error prone because of the noise in the current measurement sums up at each time step making the estimation to drift from the true value. Furthermore, also other terms in the Coulomb counting expression can induce errors, such as the actual battery capacity changing with aging and the unknown initial SOC. For this reason, measurable quantities, in this case voltage and deformation, depending mathematically on the SOC are included in the EKF to correct the estimation. The goal is to model these quantities as a function of the SOC state to be able to correct the estimation with a feedback loop: the estimation is good and does not need correction if the calculated quantity (depending on the SOC state being estimated) is in agreement with the measured quantity; On the other hand, the estimation may benefit of a correction when the calculated quantity is different from the measured quantity.

It is assumed that errors between the calculated and measured quantity are due to the bad estimation of SOC fed into the model for the deformation or voltage calculation. In reality, the estimation error may be caused by the model's poor representation of the quantity being modeled as a function of the state of charge. In this regard, the model used to compute deformation as a function of SOC is relatively simple, as it only involves the SOC-THK relationship. Conversely, the model used to compute voltage from the state of charge is a more complex equivalent electrical circuit model, which requires the identification of several resistance and capacitance parameters. These parameters not only may suffer identification errors, but they also may vary significantly as the battery ages.

As a result, estimation algorithms based on voltage measurements are inherently subjected to model errors—both due to inaccuracies in parameter identification and, more critically, due to the possible lack of parameter updates that reflect the current aging state of the battery. On the other hand, deformation-based algorithms do not suffer from this issue, since the deformation model requires no parameter identification and the SOC-THK characteristic remains stable over aging. In fact, as the battery loses capacity, the amplitude of the reversible deformation also decreases proportionally. Therefore, relying on deformation allows the algorithm to inherently account for the current state of health, without the need to update the actual battery capacity using a dedicated identification algorithm.

POLISOC is applied to LFP batteries subjected to dynamic DST and drive cycles test (Figures 4a-b, respectively), showing that SOC estimation may be improved when including deformation measurement. In the hybrid configuration, the algorithm is influenced by the covariance values assigned to the input signals, which represent the assumed measurement uncertainties. In this case, identical covariance values are attributed to all sensors (current, voltage and deformation). Nonetheless, further work could aim to improve the estimation logic so that, even under equal covariance assumptions, the algorithm naturally converges toward the signal with lower actual error—typically the deformation measurement.

POLISOC is further applied to aged LCO batteries subjected to DST profile, as shown in Figures 4c-d, respectively. In this case it is evident that when relying to the voltage based method without updating the resistance values, measured at the beginning of life (BOL), the estimation gets very bad because the current pulses cause voltage drops higher than expected because of the resistance increase during aging, causing erroneous SOC changes. On the other hand, the deformation based algorithm performs excellently, even with the SOC-THK relationship measured at the BOL.

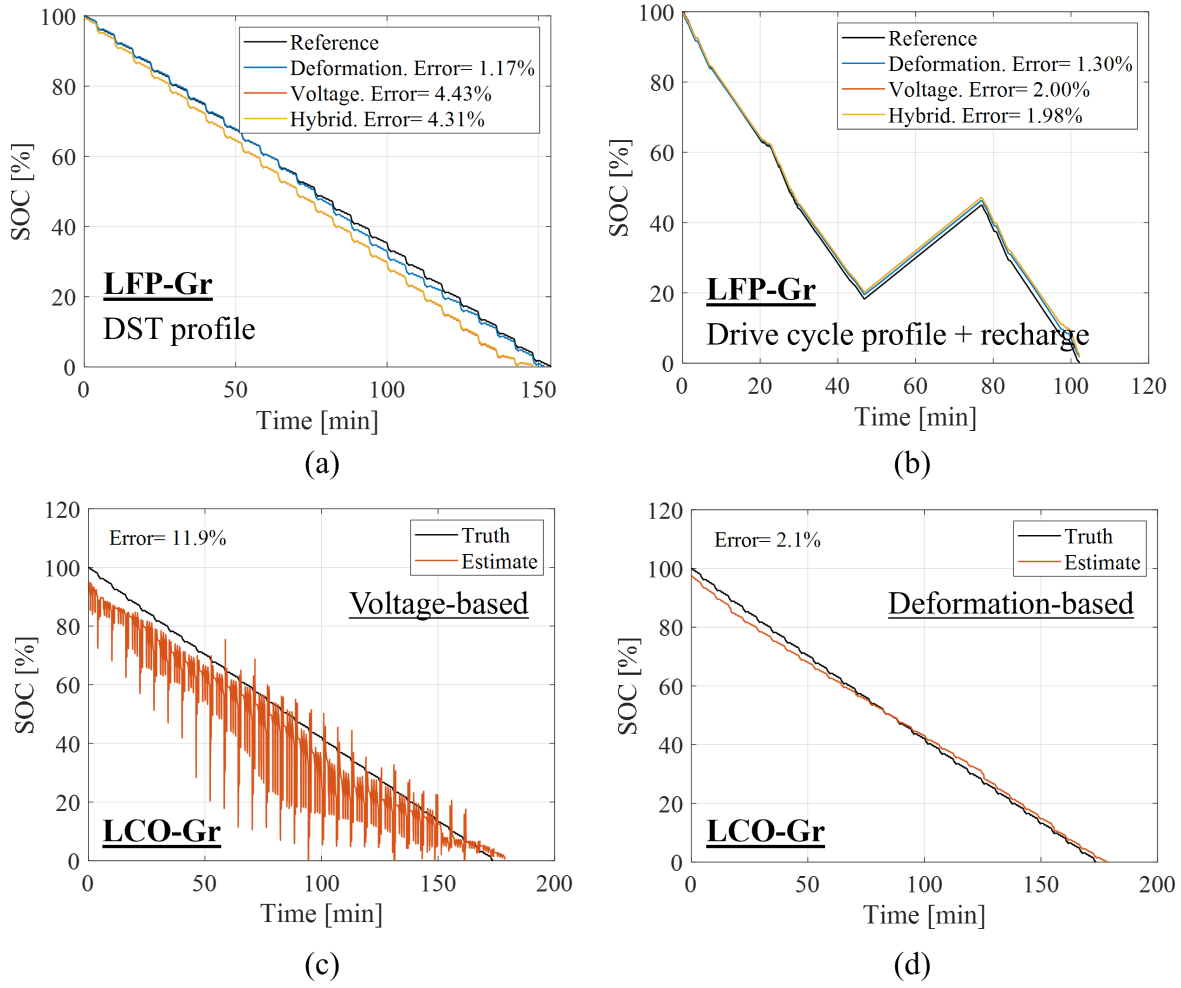


Figure 4: SOC estimation in LFP battery with the POLISOC algorithm (a) DST current profile, (b) Drive cycle current profile. SOC estimation in aged LCO batteries with (c) voltage based and (d) deformation based methodologies.

3.2 Degradation Indicators Estimation

Degradation indicators estimation based on mechanical measurement relies on the fact that the phase transitions occurring in the electrode are accompanied by structural changes detectable by the macroscopic deformation of the battery. The algorithm, called differential expansion (DE), works in analogy with differential voltage, where the phase transitions are detected by sudden voltage drops resulting in peaks in the differential voltage (DV). The significant advantage is that deformation does not suffer from polarization like voltage, so the mechanical-based method is applicable even with high current rate. This is evident comparing the differential voltage and differential expansion curves at low and high rate reported in Figure 5. At low current peaks in the differential voltage are clearly visible, but they become less evident at high current, especially peak c in Figure 5c. On the other hand, peaks remain in the same position both at high and low current with the differential expansion method, as evidenced in Figures 5b,d. This characteristic makes the latter method applicable to real-world charging profile in vehicle applications for degradation indicators estimation.

The differential expansion method used to estimate the loss of lithium inventory (LLI) and the loss of active material (LAM) from the derivative of the reversible deformation at high rate (Figure 5d) is explained in detail in a previous authors' publication [9], interested readers should refer to it for detailed information on the algorithm implementation.

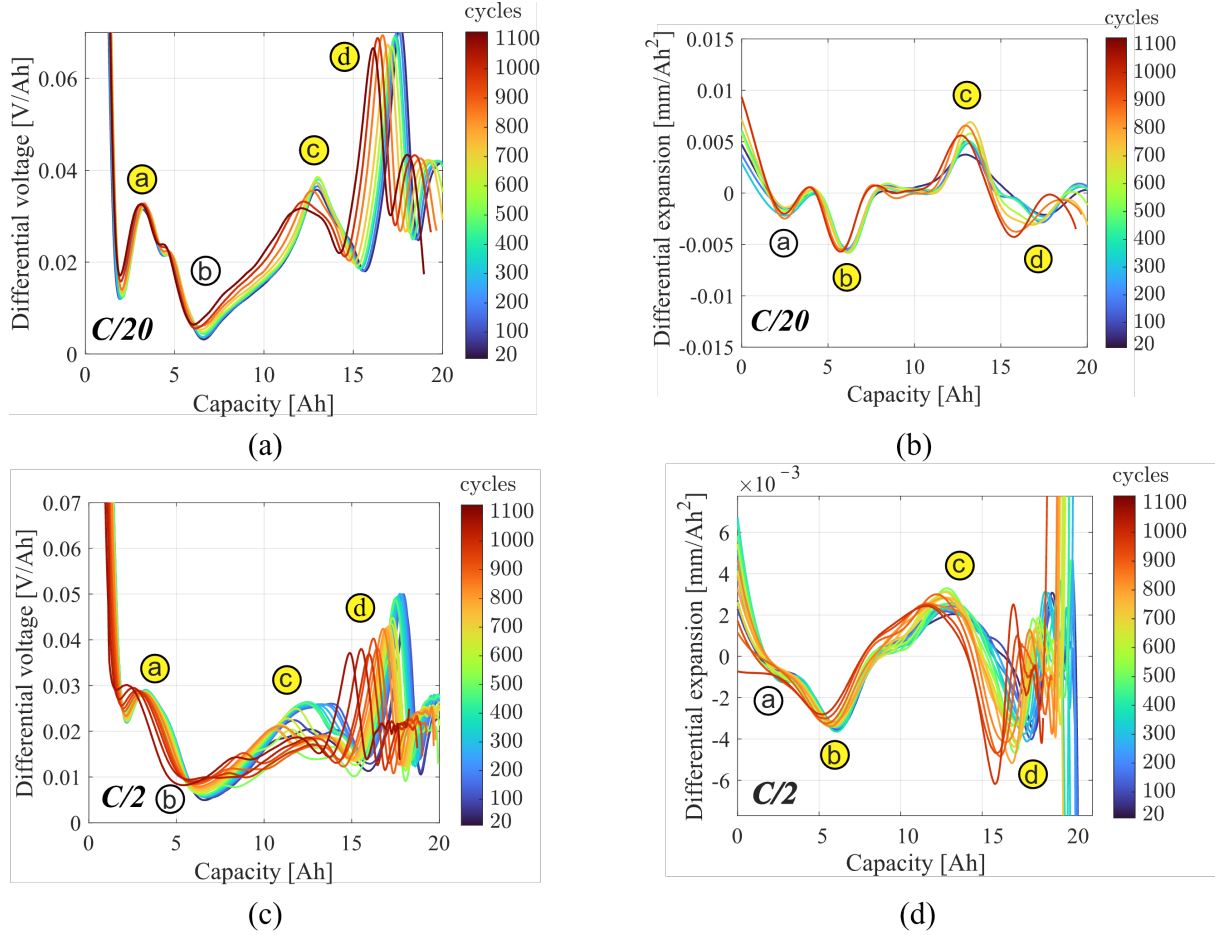


Figure 5: Differential voltage at (a) low- $C/20$ and (c) high- $C/2$. Differential expansion at (b) low- $C/20$ and (d) high- $C/2$.

Degradation indicators computed with the traditional differential voltage method applied to low current ($C/20$) charging cycle are compared with those computed with differential expansion applied to high current ($C/2$) charging cycle, to benchmark the Differential Expansion method against a conventional technique widely recognized as a standard for degradation indicator assessment. The results reported in Figure 6 show that the differential expansion method can adequately compute degradation indicators from high charging cycles, giving results in agreement with those obtained from the traditional differential voltage method performed on low rate charge cycles, validating the correctness of the deformation-based method.

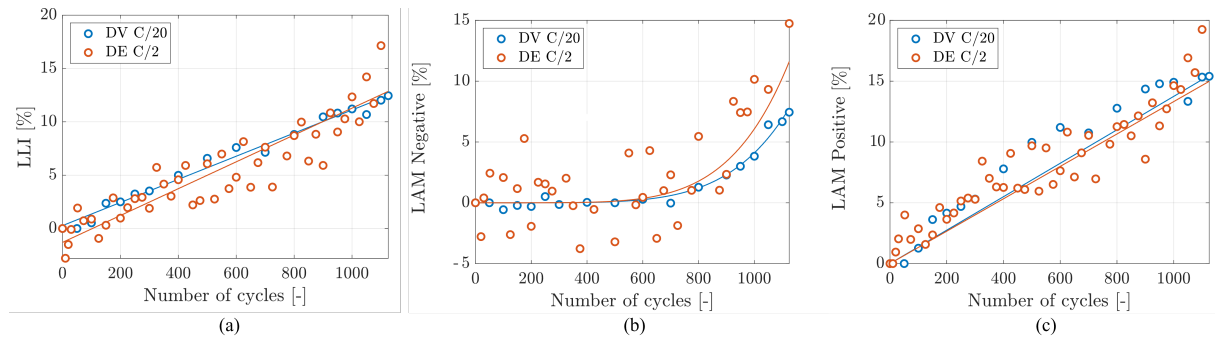


Figure 6: Comparison of degradation indicators (a) Loss of lithium inventory, (b) Loss of active material negative electrode and (c) Loss of active material positive electrode calculated with traditional voltage based methodologies and the innovative differential expansion.

4 POLIDEMO - Lithium-ion Battery Degradation Model

POLIDEMO is a physics-based battery software implemented in MATLAB, which will be released soon as an open-access software on the following GitHub repository: <https://github.com/Poli-ON/POLIDEMO>. The innovative key features of the model are the computation of the irreversible deformation, namely and the continuous increase in battery thickness during aging and the ability to capture the knee point of the capacity loss curve, thanks to an innovative mechanical degradation law correlating LAM with the increasing tortuosity and thus the increasing internal resistance.

The model framework is based on a set of differential equations explained qualitatively in the following lines. Electrochemistry is computed according to the traditional single particle model (SPM) approach. Mechanics is modeled with an analytical multi-scale sub-model [1] taking in input the lithium concentration in the particle computed by the SPM and computing the lithiation-induced deformation response of the battery during cycling from the atomic scale up to the macroscopic scale, as shown in Fig. 1a. Furthermore, a thermal model at macro-scale computes the temperature and the temperature-induced deformation. Then, the macroscopic reversible deformation of the battery during operation is computed by superimposing the lithiation-induced and thermal-induced deformations. The reversible deformation computed in this way is validated with experimental measurements, as shown in Figure 1b in the case of LCO batteries.

The aging model includes several degradation mechanisms, such as SEI growth, gas generation and crack propagation according to an innovative methodology developed by the authors [23] and its relation with the increasing SEI rate and the loss of active material. The model takes in input the physical and geometrical properties of the battery and computes the capacity loss, the reversible and irreversible deformation of the battery and the degradation mechanisms (LAM and LLI). Full details on the model framework can be found in the relevant authors' previous work [18].

The empirical model parameters are estimated fitting the capacity loss curve, the irreversible deformation and degradation indicators. This procedure allows to get a more reliable parameter set and avoid solutions that are not unique, as more constraints are considered in the error minimization problem solved to get the parameters set.

POLIDEMO is validated with the aging tests carried out by the authors shown in Figure 3 as well as with aging dataset available in the literature [6]. The validation consists in comparing capacity, irreversible deformation and degradation mechanisms, when available. Figure 7 shows the validation of the model with the aging tests carried out by the authors. It evidences that the model can correctly capture the battery capacity when cycling the battery at high and low rate, the irreversible swelling and the degradation indicators. The validation of the capacity curve deserves some additional comments. Experimentally, it is observed that the knee point in the capacity curve coincides with an increase in resistance [9, 24, 8], which in turn is associated with an exponential increase in LAM [9, 24]. It is therefore hypothesized that the increase in LAM—linked to the propagation of fractures in the electrodes—leads to an increase in tortuosity, as also confirmed experimentally through nano-computed tomography [25], ultimately resulting in a rise in battery resistance. As a result, the fact that POLIDEMO models a resistance component that depends on LAM allows capturing the appearance of the knee point when the battery is cycled under high-current RPT conditions. However, the knee point does not appear when the same battery is cycled under low-current RPT conditions. This behavior arises because the increase in resistance causes the voltage limits to be reached earlier during high-current cycling, thus reducing the extractable capacity. At low current, this resistance-related effect is significantly diminished, and therefore the knee point does not manifest under those conditions.

Irreversible swelling is mainly caused by the gas generated by the SEI growth reaction. An electrochemical-mechanical model considers the equilibrium between the expansion of the battery due to gas generation and the constraint of the battery case. A linear trend is in agreement with the SEI growth law under kinetic limited conditions. The accelerated growth towards the end of life is likely correlated with the increasing SEI growing on the cracks surfaces.

Loss of active material due to crack propagation and loss of lithium inventory correlated to SEI growth are correctly captured by the model.

The model is further validated with aging dataset available open access in literature [6]. This dataset consists of three aging conditions with constant current cycles at different current rates (C/5, 1.5C and 2C). In this case, the model parameters are identified from the condition with C/5 cycles and the same set of parameters are used in the other two conditions at higher rates, finding a satisfactory agreement as reported in Figure 8, and proving the generality of the model framework.

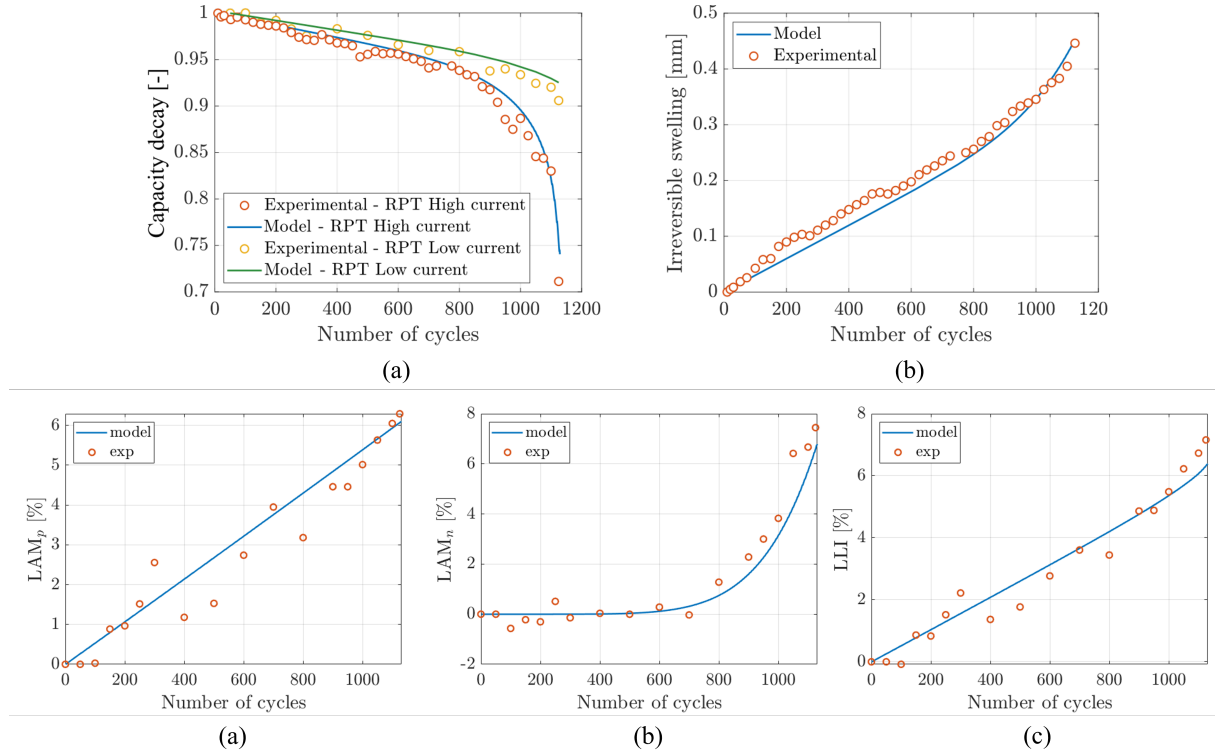


Figure 7: Validation of POLIDEMO with the aging test shown in Section 2. (a) Capacity loss, (b) Irreversible swelling.

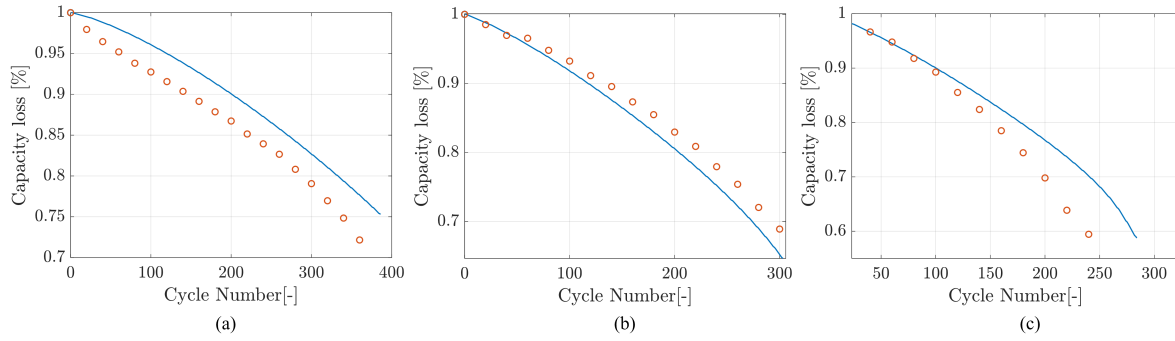


Figure 8: Validation of POLIDEMO with aging test dataset from [6] in different operating conditions: (a) C/5, (b) 1.5C and (c) 2C. Parameters are estimated in condition (a) and then the same set of parameters are used in condition (b) and (c).

5 Conclusions

This work introduced mechanical-based approach for lithium-ion battery diagnostics and degradation modeling, providing both experimental validation and simulation insights. The two key contributions are: (a) The development and application of deformation-based algorithms for battery diagnostics. In particular, a hybrid SOC estimation algorithm (POLISOC) leveraging both battery deformation and (possibly) voltage, and the Differential Expansion method to estimate degradation indicators (LAM and LLI) in real-world charging scenarios. (b) The implementation of POLIDEMO, a multi-physics degradation model capable of capturing key aging phenomena, including irreversible swelling and the knee point in the capacity fade curve.

Mechanical measurements have proven to be a reliable and robust alternative to traditional voltage-based methods, especially under high-current operation where polarization limits voltage-based estimations. SOC estimation using POLISOC shows improved accuracy in dynamic conditions and aged cells, thanks

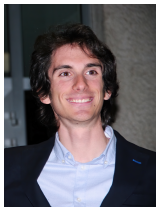
to the stability of the SOC–thickness relationship over time and the absence of model parameters. Moreover, the differential expansion method introduced for degradation diagnostics enables the estimation of Loss of Active Material and Loss of Lithium Inventory even during high-rate charging, making it suitable for automotive applications. On the degradation side, POLIDEMO successfully links mechanical and electrochemical phenomena through a consistent modeling framework. It is able to simulate capacity loss, resistance increase, and irreversible deformation aligning well with experimental results. The model’s capability to capture the onset of the knee point—through a resistance component that evolves with the loss of active material—demonstrates its relevance for battery aging analysis. Overall, this study confirms the potential of mechanics-informed battery diagnostics and modeling as powerful tools to enhance Battery Management Systems, particularly in the context of electric vehicles where real-time, high-current operations are critical.

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Presenter Biography



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