INTRODUCTION

Lithium-ion (Li-ion) technology has supported mankind for almost 30 years achieving remarkable advancements since its first commercialization. It is the most preferred choice now in the automotive sector especially in electric and hybrid-electric vehicles (EV/HEV). However, the propelled development of several different Li-ion chemistries has also faced numerous challenges while accomplishing today's excellence. Battery aging is one of the crucial ongoing challenges irrespective of the technologies addressed and constantly improved by research-driven work. The...
battery degradation can simply resemble human aging behavior where a commercial product can be compared to a fully grown body. Whereas different biological factors contribute to the decline of the lifetime, multiple environmental and operating conditions similarly degrade the battery performance. Thus, prognostics and diagnostics of battery cells draw special attention which can prolong the longevity just like medical care improves the health of the human body. This is why accurate prediction models are requisite tools yet challenging to explain the highly nonlinear and complex systems like batteries.4

The aging of a Li-ion battery is twofold often expressed in terms of capacity fade and internal resistance growth. Multiple interconnected operating factors such as depth of discharge (DoD), state of charge (SoC), temperature, current rate (C-rate), time, and the number of cycles contribute to battery degradation during cycling and/or calendar life which occurs in parallel.5 All these stressful parameters affect battery life in different magnitudes and result in performance loss. A battery cell is considered unfit for the automotive application reaching the end of life (EoL) when the capacity fade and the resistance growth exceed 20% and 100% of the beginning of life (BoL) value, respectively.6 Both these aging outputs are crucial for automotive applications, thus require precise estimations. The prediction of these degradation results is not straightforward as multiple contributing factors are involved. That is why a laboratory-level battery aging study is performed before modeling which reduces the number of aging characterization significantly during operation.7

The capacity and power fade of lithium batteries are often modeled individually considering different degradation factors.8,9 This is mainly because of the extensive research involved in covering a wide range of operating conditions. Moreover, choosing the right modeling methodology can also impact the prediction performance.10 Researchers have investigated techniques that include electrochemical and performance-based methodologies for battery lifetime predictions.9,11 In this research, a semi-empirical methodology is selected that is a tradeoff between complexity and accuracy. Researchers have followed a semi-empirical methodology that can also be used to represent electrochemical phenomena12,13 or for thermal and control strategies.14,15 Typically, the laboratory-level and long-term battery characterization data are used to formulate algorithms to fit the nonlinear battery aging behavior. In this way, the calendar life and the cycle life capacity fade factors can be modeled separately and then merged to have total degradation.16,17 The performance-based modeling methodology may also support data-driven techniques that can be used for early life predictions18,19 or to find degradation paths.20 However, the prediction of the aging trend or life estimation often mismatches to the reality due to the nonlinear nature of the battery capacity fade. In contrast, the same exercise with the internal resistance growth has been done not so often considering either cycle life or calendar life aspects as it is a challenging task.21-24 Nevertheless, only a few researchers have performed a detailed analysis of all the crucial aging factors before selecting model parameters and for both the aspects of cycle and calendar life investigations.25,26 On the other hand, a handful of modeling works have predicted the pair of degradation outputs (capacity fade and resistance growth) considering both the aging aspects.27,28 However, they still lack either a detailed analysis of the influential parameters within regular operating conditions or dynamic validation of the developed model which is crucial to proving the model feasibility in real-life application. A robust and accurate model could be verified under realistic scenarios where a dynamic current pattern drives the degradation scenario.

In this work, a novel lifetime modeling twin framework is developed corresponding to the capacity fade and the internal resistance growth responses. The twin model is designed to operate independently generating a single response as well as combined, predicting both the capacity and power fade. Such a flexible approach is rarely found in the literature that would provide a certain advantage of selecting the degradation output as per requirement. To support the model construction, the authors have investigated 48 commercial NMC cells for more than one and a half years in the MOBI research group. The novel dataset and the generated knowledge cover an extensive range of laboratory-level operating conditions providing a good understanding of the studied batteries’ aging behavior. Identifying the crucial degradation factors, they are thoroughly studied and analyzed separately for cycle and calendar life and in terms of degradation outputs. The selected most sensitive parameters are used to construct the robust semi-empirical aging model to predict the lifespan of the investigated cell. The simulated model assumptions of capacity and resistance growth are then validated with a realistic drive profile demonstrating the model robustness. Model validation based on realistic drive profiles is also unique. It is targeted to achieve less than 1.5% root-mean-squared error (RMSE) to the actual measurement. To the best knowledge of the authors, such extensive study and comprehensive modeling framework have not been established before. This refers to the thorough lifetime characterization of a large number of cells generating a database of degradation results including both the capacity fade and the internal resistance increase. Besides, all the crucial impact factors are modeled/scripted separately to have their own contributions on the total aging outputs. The constructed model can be a powerful online tool that is capable of outlining the whole life or
estimating the state of health (SoH) based on any dynamic signal. Further in this article, Section 2 describes the performed experimental methodology and Section 3 presents the aging results with sensitivity analysis identifying the impact factors. Section 4 includes the modeling framework developed in this work using the plentiful database. The model simulation and the validation results are then compared in Section 5 with further model optimization. Finally, concluding remarks are reported in Section 6 stating the model performance and its rationality.

2 | EXPERIMENTAL SETUP

The commercial NMC cells which had been investigated are of a prismatic shape and with NMC and graphite as cathode and anode materials, respectively. The cathode material composition is undisclosed due to confidentiality. The physical and electrical properties of the battery cell are listed in Table 1. All the studied cells were initially preconditioned following standard in-house test procedure and set up which have been explained in detail in this section. The electrical test campaign was performed using PEC manufactured ACT model battery cyclers and the environment test conditions were controlled by CTS made climate chambers.

The lifetime characterization consists of three types of tests, and they are calendar life test, cycling test, and validation test. Each test type is part of the campaign following different test methodology. Checkup tests are also performed at BoL, during intermediate breaks and at EoL to evaluate the cells’ health in terms of capacity fade and resistance growth. Figure 1 presents the test flow charts that have been followed for the aging tests and can be summarized as follows.

- The preconditioning of the cells includes a couple of charge–discharge cycles to activate ions inside the complex electrochemical system and to confirm a robust electrical connection. A constant current and constant voltage (CCCV) procedure is followed for charging and constant current (CC) for discharging the batteries within the specified voltage window.
- Capacity check is performed at room temperature by several charge–discharge cycles with a C/2 rate (C refers to the rated current) as part of the regular checkup procedure. This value is considered as the nominal capacity and the capacity loss (CL) is calculated by dividing the actual discharged capacity by the nominal one for each cell.
- Hybrid pulse power characterization (HPPC) is performed at three different SoC points (80%, 50%, and 20%) and with three different C-rates (0.33C, 0.5C, and 1C) to calculate the internal resistance of the battery cells. Similarly, resistance increase (RI) is calculated from the actual and the nominal value where the calculated resistance with C/2 discharge pulse at 50% SoC is considered for comparison purposes. It is found that the change in the rate of resistance growth at different SoC levels by aging is quite similar.
- Twenty-five different battery cycle life and calendar life aging conditions (distributed to the investigated 48 cells) are performed according to the test matrix presented in Figure 2. The selected combinations from the figure are considered ensuring the use of all the operating parameters at least once. Most of the investigated cells are used to characterize the cycle life by continuous CC charge and discharge cycles at specified conditions. The repeatability of the studied commercial cell response is considered very good with a standard deviation of 0.6 A based on nominal C/2 discharge capacities at room temperature. The full equivalent cycle (FEC) counter based on charge ampere-hour (rated current) throughput is used to compare all the cell characteristics (CL and RI). After every 100 FECs, a regular checkup (capacity and HPPC test) is used to record the degradation impact. The storage test typically follows a 30-day rest at a specific temperature and then a room temperature checkup is performed.
- A standard validation test based on a real-life WLTC is calculated on the cell level and performed on separate cells. A single one-hour-long WLTC test cycle as shown in Figure 3 includes two suburban dynamic currents which are performed at 90% SoC and 25 °C. The

| TABLE 1 | The specifications of the investigated NMC cell |
|---------|---------------------------------|
| Cell technology | NMC/C |
| Rated capacity | 43 Ah |
| Nominal voltage | 3.6 V |
| Operating voltage region | 3–4.2 V |
| Internal resistance | ≤2 mΩ |
| Dimension | 27.5 mm * 148 mm * 91 mm |
| Weight | 840 g ± 10 g |
continuous WLTC cycling rounds were carried out for 12 days before checking the capacity and the internal resistance of the cells through checkup tests at room temperature. The aggressive WLTC is chosen to accelerate the degradation process to some extent. A single WLTC cycle discharged the cell by 33% DoD (90%-57% SoC window) before fully charged to 90% again. The number of WLTC cycles is counted for the degradation calculation.

The core aging study of this work is carefully designed in a combination of multiple stress factors within a broad range of operating conditions. The whole campaign targets to generate a quality dataset by selecting suitable intervals within the safe operating area (SoA). Five different depths of cycling, three intermediate SoC (mid-SoC) points, four temperature conditions, and a mix of charge–discharge C-rates form the cycle life matrix. However, only high and low storage SoCs at three temperatures are used.
3.1 Cycle life stress factors

Battery cycling based on laboratory-level constant current operation is mainly dependent on the operating conditions. These conditions (DoD, mid-SoC, temperature, and C-rates) trigger chemical processes inside the battery and result mostly in loss of cyclable lithium. Other crucial degradation mechanisms for NMC battery may include growth of solid electrolyte interphase, loss of active materials, current collectors, etc. The influential parameters of DoD and the temperature results are partially presented in Figure 4 where clear dependency can be observed on the cells’ cycle life for both the factors. Higher DoD cycling is found to be degraded faster than the lower cycling depth (darker to lighter in a single-color palette). On the contrary, almost the same capacity loss trend is seen for 100% DoD at all the studied temperatures but 35 °C. Thus, 35 °C cycling results seem to have more promising cycle life meaning that this temperature is probably the optimal operating condition for 100% DoD cycling. The slowest capacity loss rate is found to be at room temperature where the cells are cycled at 20% DoD and completed around 2000 FECs on average with only 10% degradation.

The other stress factors of mid-SoC and charge and discharge C-rate results on capacity fade are shown in Figure 5. At the mid-area of the depth of cycling, the variation of the SoC ranges has shown clear sensitivity. The battery cells cycled in the higher SoC region (70%-90%) have significantly degraded than the lower SoC zone (10%-30%). This proves that the operating SoC region or mid-SoC during battery cycling can impact the capacity loss of the studied cell; thus, it should be considered in the model development process. Moreover, when the charge and discharge rates are compared at a similar condition, the C-rate impact is visible except for the 2C discharge condition which has shown better performance than 1C charge–discharge results. When the batteries are charged...
or discharged at a 3C rate against a 1C discharge/charge, the capacity loss is faster, comparatively, and quite similar to each other (see Figure 5B). However, the C-rate effect is very challenging to consider due to the limited test conditions in this work and inconclusive results. Thus, the C-rate impact is excluded from the model parameter list.

### 3.1.2 Cycling resistance growth

The internal resistance growth for the cycling cells is also compared considering the selected parameters from Section 3.1.1 and displayed in Figure 6. The resistance growth is found to be minimal during the lifetime for the accounted SoC and pulse rate and no clear conclusion could be made to evaluate the DoD and temperature influence and the same is observed for mid-SoC influence as well. However, there is an initial resistance increase within the first 500 FECs for a couple of 45 °C cycling cells which can be regarded as outliers because the values got stabilized later over time. To have an asymmetric modeling framework, the DoD, temperature, and the mid-SoC are still utilized in the model construction to align with the capacity fade model parameters.

### 3.2 Calendar life stress factors

The calendar life of a battery is one of the main aspects of degradation as batteries usually spend most of their lifetime without current loads. The capacity loss and the resistance increase can be attributed to the storage SoC, storage temperature, and the duration which may result in unstable SEI, loss of cyclable lithium, and active material in graphite-based cells.

Almost one-year-long (360 days) storage tests are performed to examine the sensitivity of the calendar life influential parameters. Figure 7 includes both the results as the capacity loss and the internal resistance growth of the calendar life cells during the storage days. The capacity fade is found to be directly linked with both the SoC and the temperature as the higher values contributed to bigger capacity loss that is common in literature. However,
The resistance increase does not reflect a clear SoC dependency, but the temperature seems to have an impact on the power capabilities. The maximum resistance growth of 60% at 45 °C storage has only a 3% capacity drop in 1 year proving that the resistance growth is more prominent than the capacity loss at a higher temperature although not significant to trigger the EoL criterion.

4 | MODEL DEVELOPMENT

The combined modeling twin framework is developed and parameterized to investigate both the impacts of cycle and calendar life together in a single representation. The resultant lifetime of capacity fade and internal resistance increase makes it comprehensive covering a wide range of operating conditions. The complete framework is established using the MATLAB coding platform and MATLAB curve fitting toolbox.

First, the sensitive parameters are identified, and the corresponding data are plotted up to a common FEC so that the algorithm fits well. This is achieved by simple extrapolation following the degradation path. Then, the aging outputs from the sensitive parameters are fitted with mathematical equations to represent the individual influences. All the stressful parameters (DoD, temperature, mid-SoC, Storage time, etc) effect is scripted separately for cycle life and calendar life as presented in Figure 8. The following general cycle life and the calendar life semi-empirical equations are used to fit the generated dataset.

\[
\text{Cycling CL or RI (FEC, IP)} = \sum_{i=0,j=0}^{n,m} (A_i(FEC)^i + B_j(IP)^j) \quad (1)
\]

\[
\text{Calendar life CL or RI (time, SoC, Temperature)} = x\times \text{(time)}^y \times \text{(SoC)}^z + \hat{z} \times \text{time} \times \text{SoC} \quad (2)
\]

In Equation (1), IP refers to the influential parameters for cycle and calendar life, respectively, \(A_j\) and \(B_j\) are constant coefficients, \(n\) and \(m\) are surface fitting orders, and \(i\) and \(j\) are polynomial orders to get the best fit. For instance, DoD impact on the cycle life degradation is fitted at every investigated temperature with 4th order polynomials \((i = 4, j = 2)\). A sample of the fitting scenario for the capacity fade in terms of DoD, FEC is presented in Figure 9 which illustrates eminent fitting \((R^2 = 0.9999)\) based on Equation (1). For calendar life, the Arrhenius equation is used together with the Equation (2) where \(x, y, z, \hat{z}\) are fitting coefficients achieved from the Equations (3)-(6) in relation to the temperature. Thus, calendar life fade calculation considers all the respective parameters. The coefficients are listed in Table 2.

\[
x(T) = x_1 \times T_2^x + x_3 \quad (3)
\]

\[
y(T) = y_1 \times T_2^y + y_3 \quad (4)
\]
In this way, the capacity fade model gets completed including the cycle life and calendar life contributions. Similarly, the resistance growth model consists of the same aging aspects following the semi-empirical methodology. Of course, different fitting orders are selected to find the best fit. Both the capacity fade and the resistance growth models are designed to operate separately but the consideration of cycling and calendar life impacts are combined. Finally, the principal model merges both the aging models and simulates the degradation outputs after evaluating the crucial parameters. As the calendar degradation occurs during the cycle life as well, thus, the no-load cycling duration in the driving profile is also considered in the calendar degradation scenario.

The model intakes dynamic or one-dimensional current signal as input together with other initial battery states such as SoC, BoL capacity, and the cycling temperature. The load signal is filtered in the initial section with a rainflow counter that is typically used for fatigue analysis. The rainflow algorithm extracts the crucial information from the dynamic profile and separates the load and no-load conditions to be processed, separately and accordingly by the model scripts. The current profile is analyzed by a simple battery model (coulomb counting) that makes an SoC profiling and the rainflow counter prepares the DoD profile and cycle numbers for the whole simulated duration. The zero currents are identified, and the total duration is forwarded to the calendar life script for consideration. When the comprehensive model is simulated for a realistic profile, all the influential parameters are extracted and analyzed by different scripts, accordingly. As a result, the model predicts the aging of the battery cell for both cycle and calendar life in terms of capacity fade and internal resistance increase for the lifetime. The total degradation represents the summation of the model cycle and calendar life responses.

The developed robust model can accurately predict the degradation within the boundary conditions of the generated dataset; however, it is also capable of estimating beyond the considered conditions by extrapolations compromising the accuracy. The semi-empirical framework is also easily adaptable to other fitting algorithms (corresponding to the diversified aging path) for different Li-ion technologies.
5 | MODEL VALIDATION AND OPTIMIZATION

The constructed model based on laboratory-level data with one-dimensional current (charge or discharge) can always be questioned on its feasibility. Thus, dynamic WLTC currents are used to testify to the model’s robustness. The on-road vehicle profile is generated based on the Nissan Leaf 500 battery pack and performed at room temperature for a standard validation. The simulated outputs are evaluated by RMSE and the metric is expressed as Equation (7).

\[
\text{Root-mean-squared error (RMSE)} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (\hat{Y}_i - \bar{Y}_i)^2} \quad (7)
\]

In Equation (7), \( n \) is the number of measurement points, \( \hat{Y} \) is the model response, and \( \bar{Y} \) is the actual discharge capacity. Figure 10 presents the model results vs. the actual measurement for capacity fade and the internal resistance growth, respectively. The simulated results are shown along with the measured values and one can notice that both the simulation outputs have a very good agreement with the real measurement providing RMSE of only 1.31% and 0.56% for the capacity fade and the resistance growth, respectively. The authors have rarely found such comprehensive studies that consider all the relative aspects of aging (cycle life and calendar life, capacity fade and internal resistance growth) constructing models from an extensive amount of experimental conditions. Hence, real-life profile validation of such models also proves the robustness. Similar effort is found in the literature; however, they have only shown the proof of concept without reporting any evaluation on the model performance. If compared errors when the individual models (capacity fade and resistance growth) simulate independently, still the RMSE would give a competitive score as well.

In Figure 10, the simulated capacity degradation curve closely follows the real measurement meaning the model response can make an accurate assumption of the lifetime. However, similar to the presented resistance growth results, the real measurements from the WLTC profile reflect the same path in the beginning but slowly parting away. The model still could predict the result with high accuracy until 4000 cycles. The achieved accuracy can be regarded as acceptable considering extensive laboratory-level study and dynamic validation. The model error can be attributed to the limitation of the semi-empirical methodology, incomplete dataset, and the nonlinear characteristics of the investigated NMC cell.

The constructed model scripts are optimized to utilize simple and built-in sections such as for the battery SoC calculation, rainflow counter, etc to reduce the computational effort. The model simulation takes around 12 seconds to generate 13% capacity fade results which could extend maximum to 16 seconds for the whole first life (till 20% CL). The simulation is done using Intel Core i7-6820HQ CPU (at 2.70 GHz and 16 GB RAM) with the MATLAB 2020a version. The accuracy of the model slightly improves if more complicated SoC and rainflow algorithms are used but it heavily compromises the computational cost. Thus, the simplicity is selected to make the developed model suitable for online applications which can be further tested at different operating temperatures to increase its robustness. Hence, the well-designed test matrix is already minimized to have the optimal investigation time.

6 | CONCLUSION

The comprehensive modeling framework which has been developed in this work can exhibit an extensive amount of information on battery cell aging. The base form of the modeling structure is a laboratory-level aging investigation of NMC cells providing explicit degradation data at multiple operating conditions. The constructed combined model could efficiently predict a real-life WLTC capacity fade and internal resistance growth within the targeted RMSE by error-scoring of 1.31% and 0.56%, respectively. The model could also predict the pure cycling and calendar life degradation contribution separately making it a useful system to identify the fading mechanisms of the aging aspects. The robust tool can perform quick online simulation and can be used as a powerful tool to outline the degradation path by analyzing any dynamic profile.

Further in the research, the authors plan to extend the validity to elevated temperatures and also for other
dynamic profiles. Identifying the challenging physical mechanisms can also add credibility to the model performance by coupling physio-chemical models; however, it would require persistent effort. The developed aging modeling methodology would be implemented for LTO cells in a EU-funded project named GHOST.

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