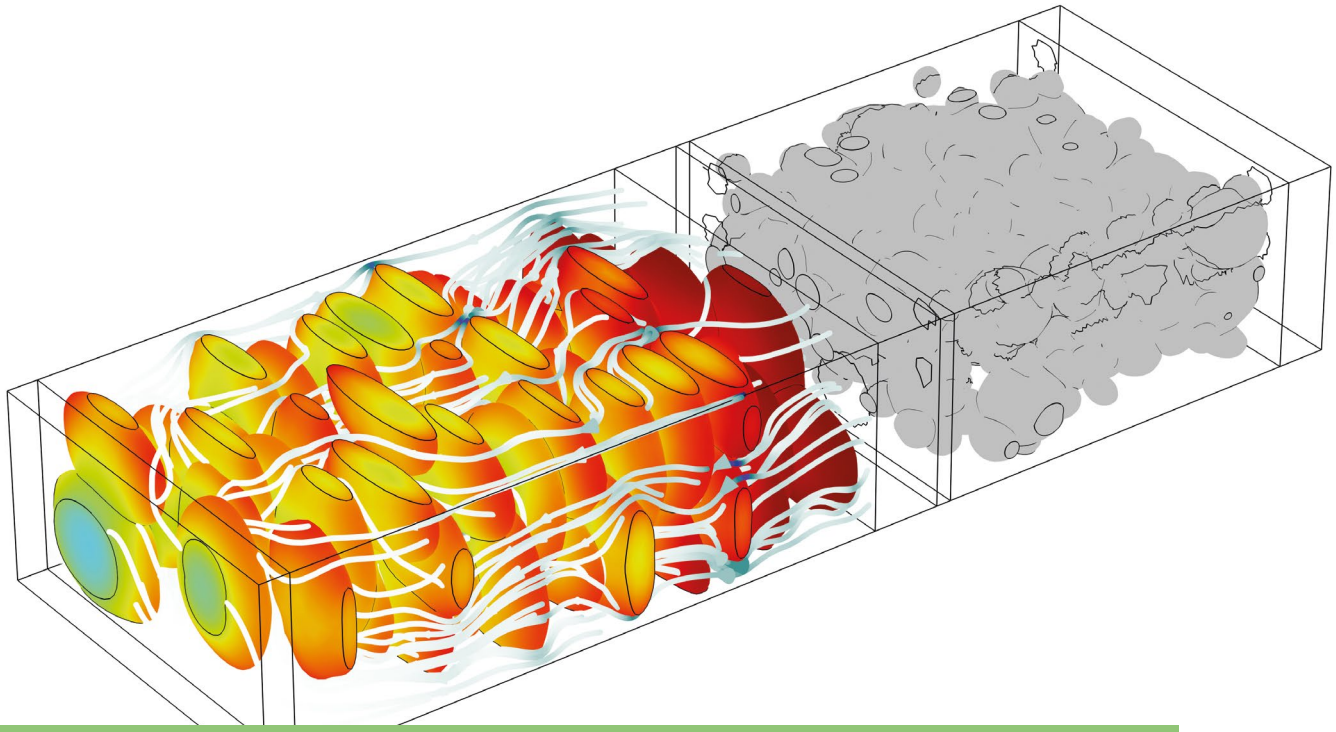

Lifetime Simulation for Aging Processes of Lithium-ion Batteries

In addition to many advantageous characteristics of lithium-ion batteries, one of their disadvantages lies in the degradation mechanism. By analyzing this aging behavior more precisely during development, improved performance and a greater range for electric vehicles can be achieved. FEV has developed several mathematical-physical approaches with which the lifetime of the batteries can be modeled and thus extended.

There are various approaches to modeling the lifetime of Lithium-ion Batteries (LIBs) for Battery Electric Vehicles (BEVs). These approaches are used in different applications of monitoring and controlling battery cell design and battery system design. On the one hand, the aim is to optimize battery operation in real time and, on the other, to predict battery performance from the cell to the system. Accordingly, the choice of a suitable lifetime modeling approach depends on the special application context. **FIGURE 1** illustrates a classification of approaches for lifetime modeling. The four models are based on the level of physical interpretation incorporated into the model, as well as the resulting complexity and computational effort.

DATA-DRIVEN MODELING APPROACH

The data-driven approach is based exclusively on testing or fleet feedback requiring specific boundary conditions. By using Bayesian optimization according to Frazier [1], it is possible to design a meaningful test plan, derived from the specified test range. Subsequently, the conduction of the lifetime tests based on the defined test matrix is followed by training the lifetime model on the resulting test data. The accuracy of the model is further enhanced through an iterative closed-loop process where the results are utilized to define a new set of tests or parameters from the fleet. The iterative process can be realized by automation or in terms of machine learning.

The developed model enables the optimization of the cell operating conditions in order to expand the lifetime. Since the model does not incorporate chemical or

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physical influences and relies exclusively on the range of input data, the model prediction might differ from the measured aging rate if the LIB is operated beyond the tested range.

SEMI-PHYSICAL MODELING APPROACH

With the semi-physical approach, the physical-chemical interactions are realized by the embedded mathematical formula to reflect the effect of degradation factors such as temperature, State of Charge (SOC), Depth of Discharge (DOD), and charge/discharge rate (C-rate). The mathematical correlations account for the effect of physical phenomena such as a growth of the passivation layer (Solid Electrolyte Interphase, SEI), lithium plating and other reactions that take place in the battery during operation. Furthermore, contrary to fully physical model, there is no need for interior characteristics of the battery cell to parameterize the semi-physical model. Accordingly, the electrothermal behavior is initially calibrated based on Beginning of Life (BOL) tests such as Hybrid Pulse Power Characterization (HPPC). Hereafter the model is further characterized by calendric and cyclic aging tests for the different aging factors to account for the battery cell performance in various State of Health (SOH) conditions. These lifetime tests should be done for at least two cells with a reasonable time frame respectively cycle frame to cover all the relevant effects.

The described data processing of the tests is done model-based. Here, the results are analyzed, and unsuitable measuring points are removed. This enables the best possible model accuracy. In order to cover different applications, a difference is made between cyclical ageing (charging and discharging) and calendar ageing during calibration. The resulting ageing factors are implemented in the semi-physical model shown in **FIGURE 2**. This model is composed of two different sub-models. While the electrothermal sub-model serves to predict the electrothermal performance of the battery, the aging sub-model provides the capacity fade and resistance increase over lifetime.

For the electrothermal behavior of the cell, the Thevenin equivalent circuit diagram model is used with up to three Resistor-Capacitor branches (RC-elements) and combined with a differential equation for evaluating the SOC. For thermal behavior, lumped model approach has been adopted in which thermal point masses are connected via thermal resistances. Therefore, the cell is separated into thermal masses and resistances and, depending on the use case, connected to a cooling plate or directly to the coolant (direct cooling). The second sub-model describes the aging behavior of the battery based on the driving cycles over the lifetime. The input cycles are decomposed into charge, discharge, and storage (idle) periods and analyzed corresponding to the aging effects. Performing statistical analysis (histograms), the capacity fade and impedance rise with the aging factors are calculated and then integrated over time for the total aging.

This model can be used to predict life and to analyze the influence of parame-

Empirical data requirement, real-time capability

Full physical	**Reduced physical**	**Semi-physical**	**Data-driven**
Modelling actual processes and using physical models	*Phenomenological and physical processes*	*Phenomenological and physically motivated dependencies*	*Phenomenological and mathematical fitting of dependencies*
- Concentration - Plating and dendrite - SEI growth - Gas evolution - Volumetric change - Heat generation and exchange - Cell geometry: 1-D (particle) to 3-D (micro- and/or macroscopic) - Concentration gradient - Layer thickness	- Shift of potentials - Reduction of conductivity - Expansion of the cell - Macroscopic - Loss of lithium - Layer growth and change - Loss of basal and surface active area	- Loss of (real) capacity - Increase in resistance - Arrhenius behavior - Temperature - Electrode potential - Currents and overpotentials - Cycle depth	- Loss of (real) capacity - Increase in resistance - Correlation-focused - Simple physical functions (hybrid approach) - Efficient handling of large amounts of data, for example for automation and root cause analysis

Physical-chemical knowledge, computational effort

FIGURE 1 Classification of battery lifetime modeling approaches (© FEV Europe GmbH)

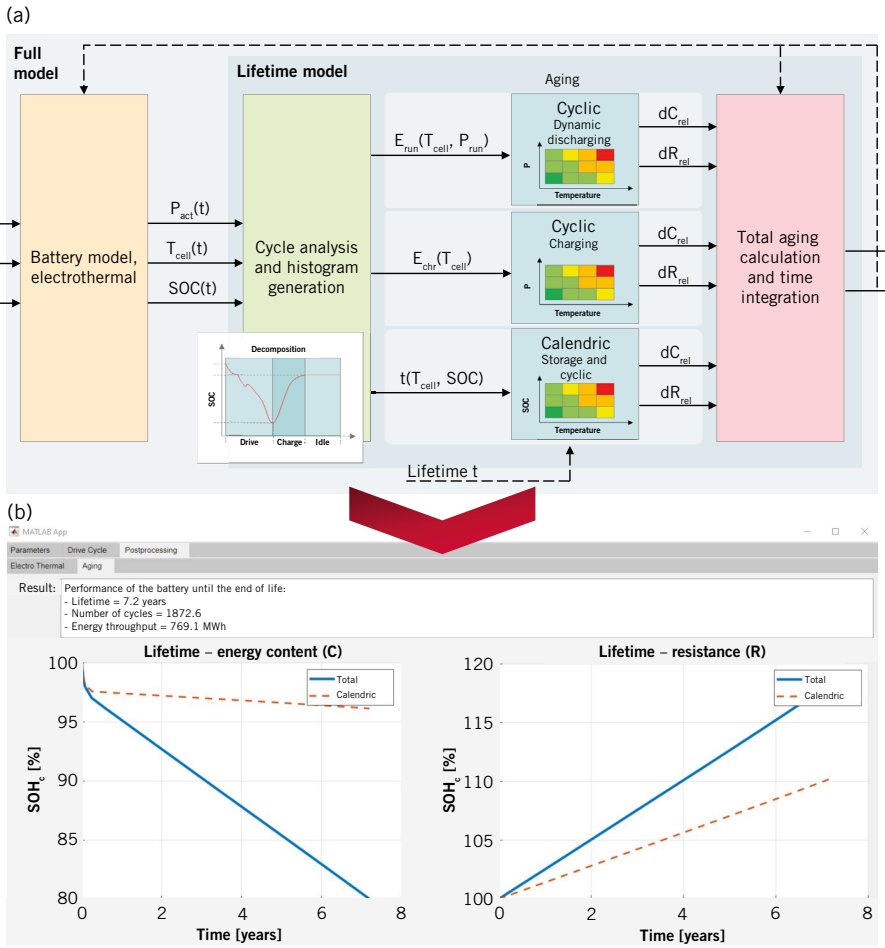

FIGURE 2 Description of the semi-physical model approach (a) and representation of the developed model graphical user interface (b) (© FEV Europe GmbH)

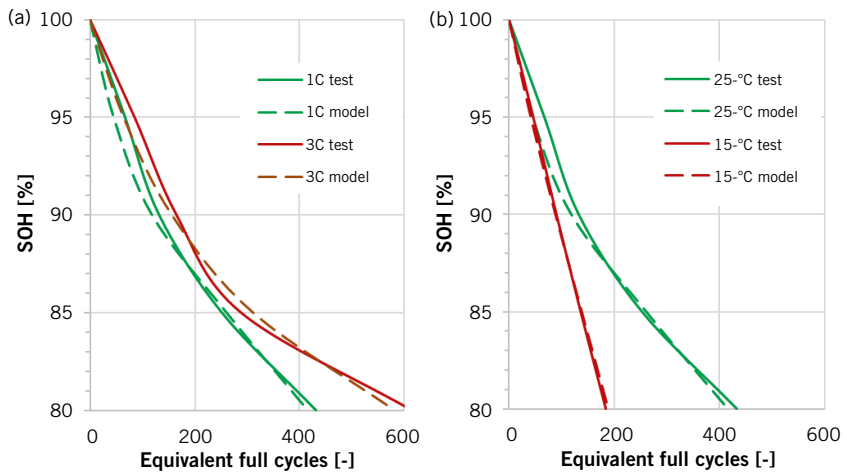

FIGURE 3 Validation of the semi-physical model with testing data [2] for different C-rates (a) and temperature dependencies (b) (© FEV Europe GmbH)

ters by comparing the frequency of different temperatures, C-rate and SOC levels. To make the model easy to use, a Graphical User Interface (GUI) was developed, **FIGURE 2**. With the described input an accuracy of ±2 % can be archived for an aging of up to 80 % SOH. **FIGURE 3** shows a comparison for C-rate and temperature dependences of the model results compared to countering real test values based on Preger et al. [2]. As observed, the model results exhibit an accuracy of 2 % in end of life. Out of these results it is visible that the initial exponential drop in capacity (due to SEI formation) is captured and predicted by the model. After this drop, a linear behavior is modelled. This model enables the most efficient analysis of the driving profiles with the following optimization.

PHYSICAL APPROACHES

The physical model consists of mass conservation and charge conservation equations within distinct region borders of the battery. Consequently, this approach provides the highest predictability with the drawback of a slower computational speed compared to other approaches. The models developed can be divided into three categories: Single Particle Model (SPM), continuum model and heterogeneous model. While the latter two are denoted as physical models, the former one is labeled as a reduced physical modeling approach. **FIGURE 4** illustrates the schematics of the physical modeling approaches for batteries.

REDUCED PHYSICAL MODELING APPROACH

The fundamental assumption in the SPM is that active material particles within each electrode exhibit similar behavior. Therefore, each electrode is approximated by a single representative particle in which lithiation and delithiation occur. In the SPM a uniform current across the electrodes is assumed while the dynamics within the electrolyte are ignored. Therefore, in contrast to the other physical models, there are no partial differential equations for electrolytes, which results in less computational effort. However, such a model reduction restricts the applicability of the conventional SPM to low C-rates. To address this limitation the model introduced in this work incorporates electrolyte effects into the SPM framework referred to as SPMe. Such a reduced model enables to resolve battery's kinetics more accurately upon cycling at high rates as compared to the conventional SPM. In addition, the thermal behavior is introduced into the model (SPMeT) to consider temperature-dependent aging effects.

Aging of LIBs is caused by complex reactions occurring concurrently at different locations in the battery. The rate of degradation varies across different stages of a load cycle, depending on the potential, local concentration, temperature, and the direction of the current. To allow a prediction of the impedance rise and the capacity fade followed by the degradation the kinetics of the reactions are mathematically defined and coupled to the dynamics of the battery. Finally, the model i s parameterized in terms of kinetic variables, enabling a prediction of the lifetime under different operating conditions, like for example fast charging.

FULLY PHYSICAL MODELING APPROACH

The fully physical model presented here includes two approaches: the continuum model and the heterogeneous model. The former one describes the underlying electrochemical mechanisms along the electrode's thickness (x-coordinate) and r-coordinate of the active material particle and is therefore also referred to as a Pseudo-Two-Dimensional model (P2D). The continuum model can be further upscaled to include the in-plane directions (y- and z-coordinates) which is then specified as P4D model. While the P4D model allows the inhomogeneities to be resolved across the correct collector

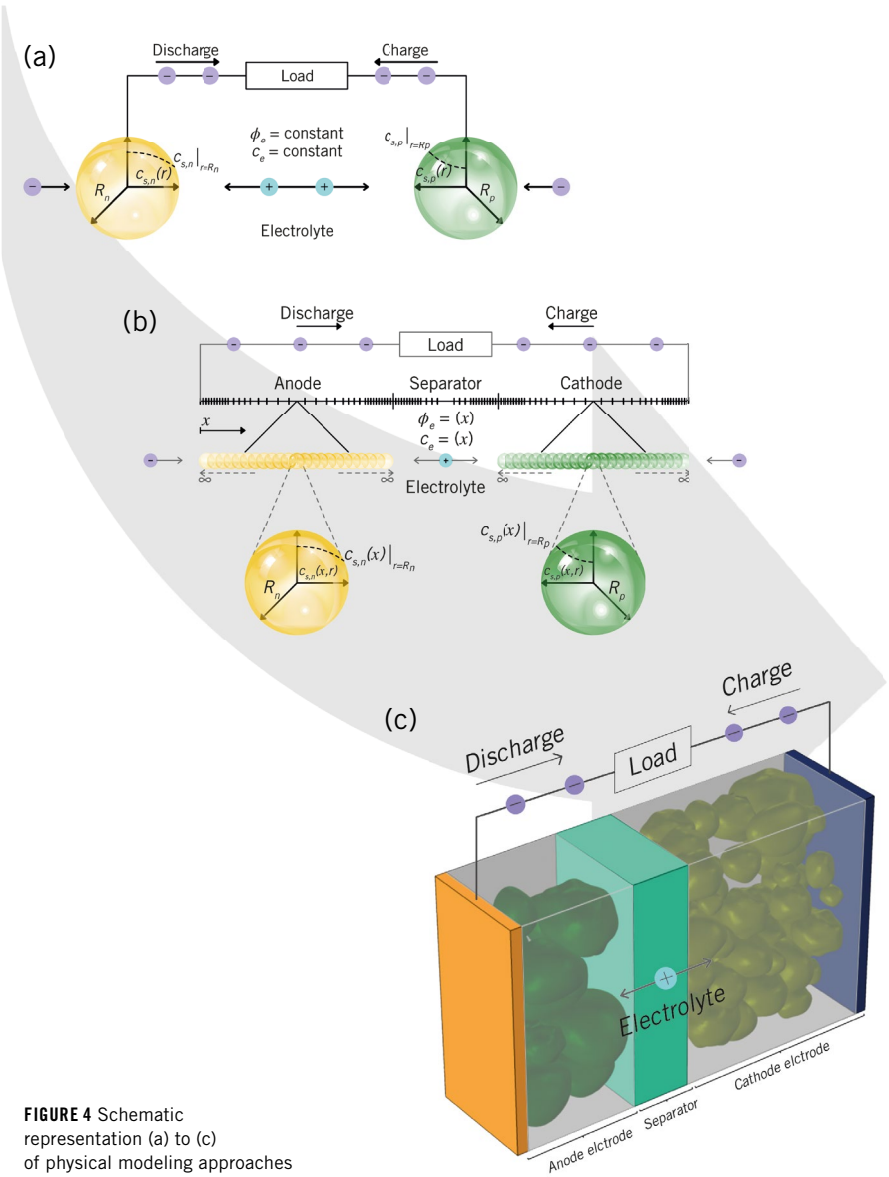

FIGURE 4 Schematic representation (a) to (c) of physical modeling approaches (© FEV Europe GmbH)

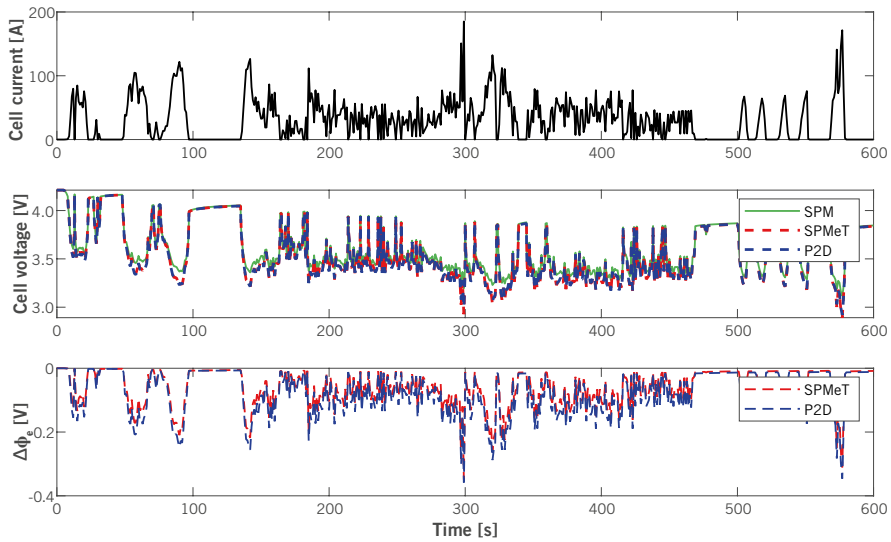

FIGURE 5 Comparison of SPMeT to conventional SPM and P2D upon typical drive cycles with high C-rates (($\Delta\phi_e$: electrolyte potential) (© FEV Europe GmbH)

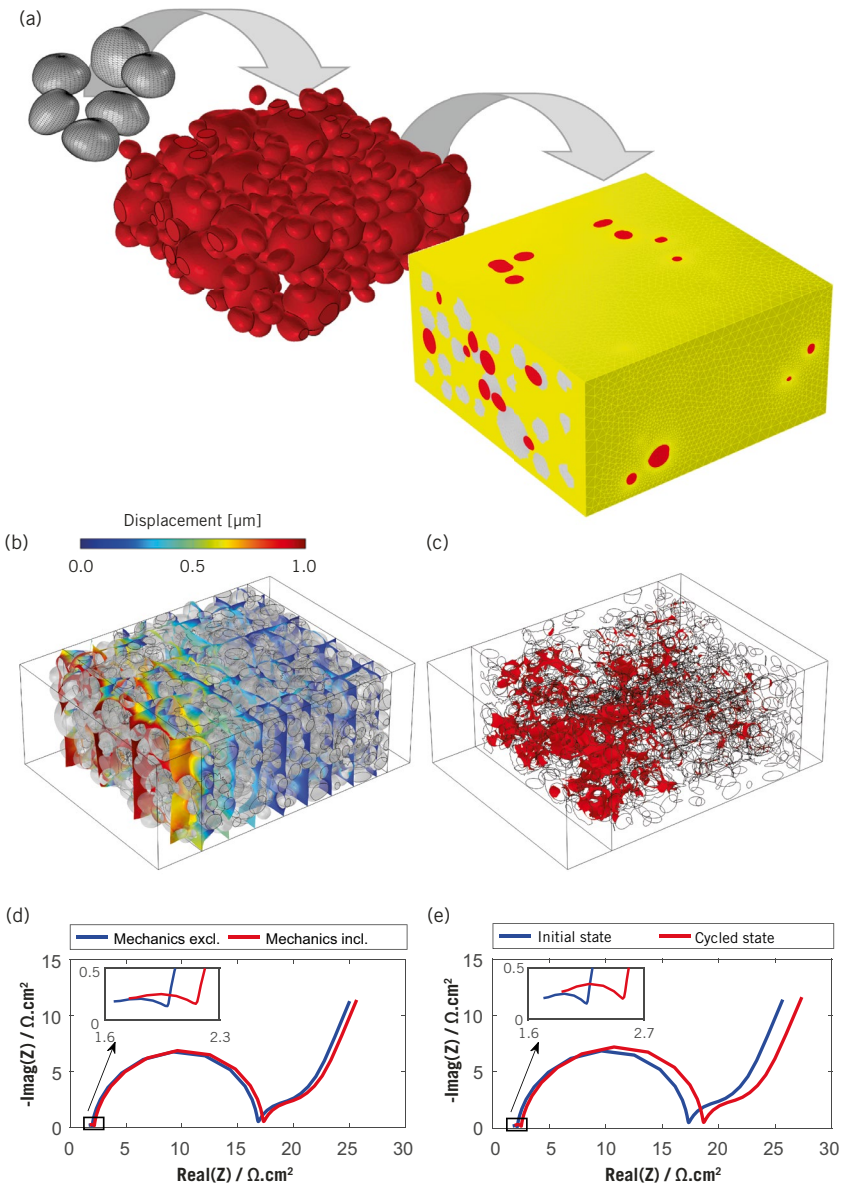

FIGURE 6 Study of the mechanical behavior: development of the digital morphology of an NMC cathode electrode (a); calculated deformation of the NMC electrode microstructure (b); resulting detachment of NMC particles and carbon-binder domain (c); degradation of the electrochemical impedance spectrum impacted by mechanical degradation (d) and through fatigue cycling (e) (© FEV Europe GmbH)

plane in addition to the thickness direction, the choice of the appropriate continuum approach is made according to the existing computational resources in each case study. Referring to the P2D continuum approach as benchmark, the precision of the developed SPMeT and conventional SPM can be compared associated to the US06 drive cycle, representing highway driving comprising of rapid acceleration and high velocity. Accordingly, **FIGURE 5** indicates the cell currents, cell voltages and electrolyte potentials ($\Delta\phi_e$), calculated by P2D approach, SPMeT and SPM. As observed, the cell voltages and electrolyte potentials predicted by SPMeT are sufficiently accurate and match the prediction by the P2D approach.

Despite a fully physical description of the LIB in the continuum approach, it simplifies the complex electrode microstructure and treats it as a continuum using approximation methods, for example the Bruggeman formulation. The simplified representation using the continuum approach has advantages in terms of computational effort but does not fully represent the complex processes.

In contrast, the heterogeneous approach involves a more complex physically based modeling of the LIB. Virtual 3-D microstructural spatial domains are incorporated to address the porous electrode's morphology. While the heterogeneous approach offers the highest accuracy, the computational speed is reduced. To account for the degrada-

tion phenomena, the parasitic reactions are spatially defined and coupled to the electrochemical dynamics of the battery. This provides the ability to resolve the inhomogeneities in degradation rate across the battery, which become more pronounced under critical operating conditions such as fast charging and low temperatures.

FIGURE 6 (a) shows an example of themorphology of a Nickel-Manganese-Cobalt (NMC) cathode electrode for investigating the mechanical deformation of the electrode during lithiation, **FIGURE 6** (b). Following that, the occurrence of mechanical degradation has been visualized by calculation of the detachment between the NMC particles and the carbon-binder domain in cycled state, **FIGURE 6** (c). The mechanical deterioration of the electrode has been also realized in terms of electrochemical impedance spectroscopy to attain a deep understanding on the mechanics-induced effects on electrochemical performance of LIBs, **FIGURE 6** (d) and **FIGURE 6** (e).

SUMMARY

In order to map the battery lifetime for BEVs more accurately, FEV has developed a set of modeling approaches. The introduced models fulfill the existing requirements of the BEV market in terms of various applications, ranging from the on-board design to the battery development process as well as from cell level up to system level. Consequently, significant time and cost benefits are achieved by strongly reducing expensive experimental aging testing. Since all the presented models are able to predict the battery aging accurately enough, the decision on the required model approach and its calibration method is cautiously made by accounting for the existing boundaries in each project to realize driving range extension and fast charging ability in BEVs.

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