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Accelerated multiscale and multiphysics modelling tools for battery cell manufacturing improvement

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Abstract

The recent launch of battery factories in Europe, motivates intense efforts to achieve cost-effective, scalable and sustainable battery manufacturing processes. Within DEFACTO project, multiscale multiphysics modelling tools are developed to increase lithium-ion battery (LIB) cell manufacturing process productivity and performance. A novel workflow framework that mimics the main cell manufacturing steps such as the electrode processing and electrolyte filling and later predicts cell performance and ageing is presented to turbocharge the development of next-generation LIBs. In addition, taking advantage of the characterization and manufacturing data to feed and validate the computational tools, the resulting workflow aims at providing deep understanding and therefore guidance to reduce the production process time and cost while increasing the overall efficiency of battery cells.

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Keywords: battery; cell manufacturing; cell ageing; workflow; multiscale models; multiphysics models.

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1. Overview and motivation

In recent years, the acceleration of green and digital transitions is required to build a more sustainable and resilient society and economy. Sustainable mobility can only be achieved through a radical change in the transport system, far-reaching innovation and a coherent implementation of greener, more reliable and smarter transport solutions. The surge in e-mobility has been accompanied by a significant demand for EV (electric vehicle) batteries. The basic components of batteries, electrochemical cells, represent an important market for the EU industry, estimated at 625b€/year by 2030 as pointed out by European Commission (2022). Taking advantage of the presence in Europe of many EV manufacturers, the establishment of a complete battery value chain is imperative for a clean transition. Furthermore, it will increase the regional competitiveness to a level where it would be possible to succeed against the current market leaders, historically dominated by Asian countries due to proximity to raw materials and low labour costs.

To do so, new design and manufacture routes for achieving high-performance LIB cells need to be defined to allow cell manufacturers to be more competitive. Thus, multiscale and multiphysics modelling tools applied to battery cells are seen as enablers to increase battery cell manufacturing efficiency and achieve high-performance cells, as they provide in-depth understanding of the impact of manufacturing parameters on the final LIB cell performance and ageing, as stressed by Saldaña et al. (2019). In addition, LIB cell characterization and testing can be substantially accelerated by efficient and accurate simulation models as indicated by Ramadesigan et al. (2012) and Franco (2013).

In computational science, the workflow approach is a long-established tradition to understand the physical nature of complex systems while accounting for dependencies and concurrency of tasks, as Schaarschmidt et al. (2022) and Deelman et al. (2009) stated. Here, a novel “weak” workflow describing the main steps in the current LIBs cell manufacturing routes (i.e. roll-to roll processing) through a multiscale-multiphysics simulation framework is presented. This involves computational tools that mimic the behaviour of the electrode production from wet dispersing and drying, through to the final calendering, fixing the electrode structure, as well as the electrolyte filling step and cell finishing. As stated by Safari et al. (2009) and Reniers et al. (2019), a multiscale multiphysics workflow is employed to gain in-depth insight on the mechanical aspects of the cell’s components and their corresponding degradation mechanisms, based on a methodology that leverages recent advances in reduced order modelling (ROM). Fig. 1 shows the methodological framework proposed in this work.

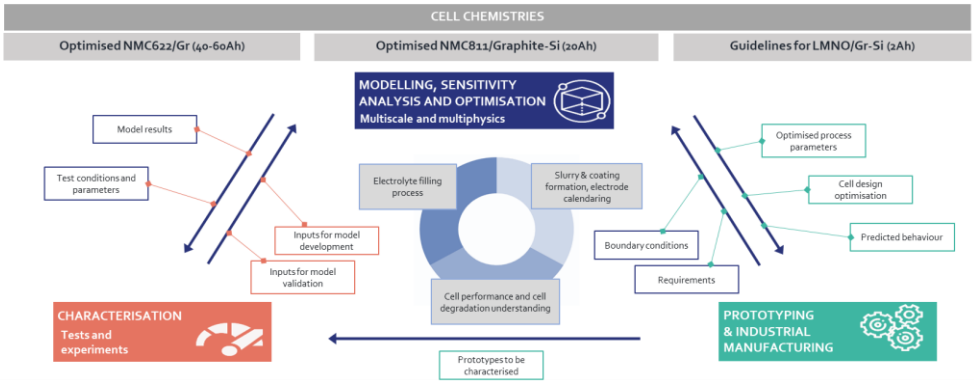

Fig. 1. Schematic overview of the DEFACTO workflow

The resulting simulation tools predict optimized cell design and cell manufacturing parameters which have been validated by prototyping and manufacturing data from current G/NMC622 industrial cells to latest generation 3b prototype cells (G-Si/NMC811 and G-Si/LMNO). The goal of these modelling tools is to significantly reduce the need for sophisticated parametrisation experiments and, therefore, to accelerate the advanced LIB cell development by 30% and to reduce the overall R&I costs of a cell in the manufacturing-line-to-market process by 20%. Finally, the innovative simulation framework also contributes to the generation of new standards in the battery sector that can facilitate the acceptance and utilisation of the developed solutions.

2. DEFACTO's multiscale multiphysics tools

The DEFACTO modelling tools aim at reducing the time to market of the next generation battery cells by providing more understanding on the impact of process parameters on battery cell performance and ageing. In this regards, three main steps of the battery cells manufacturing routes have been considered in the project: electrode processing, electrolyte filling and cell performance and ageing. Firstly, electrode structure formation on the electrode processing step is modelled through mesoscopic contact model's approach. Thus, the structure formation process as a function of the formulation of the electrode slurry and the process parameters during mixing, drying and calendering is properly and efficiently addressed. Then, electrolyte filling process is modelled as it is a critical step that impact the battery cell performance and ageing. The multiscale approach followed in DEFACTO will provide valuable information on the best filling process parameters to maximize cells lifespan. Finally, battery cell performance and ageing is studied through multiscale approach, where parameters resulting from atomistic and mesoscopic microstructure-resolving models are scaled up to homogenized parameters to be used in the continuum scale models. This modelling approach extends the scale range from the atomistic to the continuum and the physical domains covered in the cell performance and ageing model, paying special attention to the mechanical degradation. The latter continuum scale model is then reduced to build a more efficient tool and then coupled to optimization algorithms to further improve battery cell properties. The different tools, scale and their corresponding output are listed in Table 1. Details regarding the approaches for each process step are given below.

Table 1. List of models developed for the design-dependent properties of the cell and manufacturing process simulations

Model codename	Method/Software	Scale	Outputs from models	Output for battery optimization
CFD/DEM	Discrete Elements Method (LIGGGTHS) / Computational Fluid Dynamics (OpenFOAM)	Electrode /Slurry	Slurry composition after dispersing and microstructure after drying and calendering	Optimization of carbon black structure and the mixing process. Efficient electrode structure prediction. Mechanical integrity.
LBM	Lattice Boltzmann Method with Palabos library	Electrode	Dynamic wetting behaviour, permeabilities, and electrolyte distribution	Optimized filling process parameters, electrode and electrolyte materials, and microstructures.
PNM	Pore Network Models with OpenPNM	Electrode /Cell	Wetting behaviour and permeabilities	
RE	Richard's equation using in-house code	Cell	Simulation of filling process	Optimal electrolyte filling process conditions.
DFT	Density Functional Theory (DFT) method	Material	Mechanical input parameters	Deformation and change of material properties due to mechanical ageing.
DEM	Discrete Elements Method with LIGGGTHS	Electrode	Mechanical behaviour of electrode (performances and ageing)	
BEST	Continuum model with BESTmicro and BESTmeso	Electrode	Mechanical behaviour of electrode (performances and ageing)	Efficient continuum pseudo four-dimensional (p4D) battery models including mechanical and electrochemical ageing.
p4D_ageing	Continuum model with FEniCS library	Cell	Electrochemical and mechanical of the cell	
p4D_ROM	p4D simulations with cideMOD / model order reduction (ECHROM)	Cell	Electrochemical and mechanical of the cell	Optimum conditions for using and battery pack design. Reduce the computational effort needed to solve p4D battery models.
ROM	sPGD non-intrusive method	Cell	Battery Design Space	

2.1. Electrode processing modelling

Mesoscopic contact models are developed and implemented into the simulation software to capture the influence of the salient parameters during electrode production on the final microstructure. For the wet mixing process, single carbon black aggregates and agglomerates in the suspension are represented, coupling the discrete element method (DEM) and computational fluid dynamics (CFD) for simulating particle-fluid interactions. During the drying step, the

effect of the fluid-fluid interface acting on particles are studied and the interaction between the individual particles is modelled using CFD-DEM simulations to investigate the structure formation. Then, the particle structures generated by the drying simulation are transferred to a calendering DEM simulation. The state-of-the-art calendering model is extended to include the adhesive current collector interaction and simulate the influence of heated calender rolls. Finally, computationally efficient process-mimetic digital microstructure reconstruction and in-depth analysis of 2-D microscopic material sections are used to validate as well as to extend the 3-D virtual electrode structures from the full-physics simulations.

2.2. Electrolyte filling modelling

Electrolyte filling of porous battery structures such as electrodes and separators is a crucial step that also affects the battery cell performance, cycle life and safety. Most of the physical phenomena during the filling occur on the pore scale and are hard to study experimentally. Therefore, often computational approaches are used instead. The lattice Boltzmann method (LBM) is such an approach that can predict the filling process and corresponding pore-scale phenomena. Within the workflow indicated by Lautenschlaeger et al. (2022), it is used to simulate electrolyte flow in 3D electrodes with and without binder at sub-micrometer resolution. To consider also relevant effects at the electrode- and cell level, pore network models (PNM) and continuum methods based on the Richards equation (RE) are used, respectively. All in all, the three approaches are coupled to a framework and applied to study the influence of process parameters, structural, and physicochemical properties on process dynamics, electrolyte saturation, and battery performance. The results are useful to optimize the filling process and serve as a baseline for further electrode and electrolyte design.

2.3. Cell performance and degradation modelling

A multiscale multiphysics approach ranging from the atomistic to cell scale is pursued for predicting cell properties and its ageing mechanisms by means of modelling. On the atomistic side, density-functional theory (DFT) is used to determine several crucial properties that are needed for the electrochemical-mechanical simulation of composite electrodes on larger scale continuum cell models. For the mechanics of the electrode microstructure, Discrete Element Method (DEM) and continuum models such as BEST tool, are developed to properly address the mechanical response to external pressure on the battery cell and even understand the structural changes and the macroscopic response during cycling. In addition, a homogenized continuum scale p4D model of Newman type is extended to incorporate all relevant cell performance and ageing mechanisms but is also technically suited to integrate advanced model order reduction methods.

2.4. Reduced simulation for cell optimization

The optimization of the battery cell design is performed using homogenized continuum scale p4D models accelerated by Model Order Reduction (MOR) techniques. Sensitivity analysis of the cell parameters, such as dimensions, electrode thickness and porosity, are carried out to identify those parameters that have the most influence on the model's results. Two ROM software tools are built to allow both fast explorations of the design space and cell optimization processes. The first one is based on the sparse Proper Generalized Decomposition (sPGD) method giving immediate results while the second one is an open-source software tool based on time- and parameter-adaptive reduced order models developed within the frame of the DEFACTO project (2022).

3. Experimental Approach and characterization

The experimental characterization and cell prototyping activities allow to build, validate and optimize multiscale and multiphysics models to better understand all the steps of the electrode manufacturing, the electrolyte filling process and the electrochemical ageing mechanisms. All the experiments which are needed to determine the model parameters defined are included in the public report D2.1 from DEFACTO project (2020). A short description and the data type defined for each experiment is also given.

3.1. Electrode and cell process parameters

Electrodes as well as cells are manufactured at both scales, prototype scale in pilot plant and industrial scale. The key manufacturing parameters and boundary conditions that accurately describe both manufacturing scales are required to develop the modelling and simulation tools described in sections 2.1 and 2.2. In this regard, around 30 parameters are identified to be monitored in the manufacturing process, including online and offline measurement systems. Worth to mention that around 55% of the experiments proposed (i.e., capillary pressure measurement, rheology measurement, ultrasonic acoustic wave, among others) are linked to the process model validation and calibration.

3.2. Electrochemical and Mechanical measurements

Electrochemical and mechanical tests are necessary both for performance and ageing models' (introduced in section 2.3) calibration and validation. Electrode properties are investigated by electrochemical techniques such as GITT and EIS measurements and by physicochemical techniques such as laser diffraction, TGA, XRD among others. Mechanical parameters are also obtained through microindentation technique and mechanical ageing is also investigated trough an in-house set-up.

3.3. Image segmentation and tomography library

Imaging experiments are used for gaining understanding on the final electrode's microstructure and the impact of manufacturing parameters on them. Techniques such as microtomography, SEM coupled with dynamic image analysis, and TEM measurements are used to extract among others the particle size distribution, shape, surface properties of raw powders and electrodes, as well as electrode density profiles.

3.4. Thermal and chemical analyses

Thermal and chemical analyses are mainly used for model calibration to assess the full cell stability behaviour and identify main degradation reactions upon ageing. The thermal behaviour and the related properties of the cells is investigated by ARC test. Electrochemical properties and ageing evolution is revealed by performing properly defined ageing tests and post-mortem analyses. Different physicochemical techniques (i.e., XPS and XRD) are used to characterize the harvested components over the aged cells to further investigate on the degradation mechanisms, such as the SEI growth, Li-Plating, particle cracking, expansion and contraction and transition metal dissolution.

4. DEFACTO's workflow

DEFACTO's workflow is based on a multiplatform tool that has been developed with the purpose of interconnecting battery cell performance and the manufacturing processing parameters. The multiscale-multiphysics-multiplatform tool, named DEFACTO's workflow, refers to the weak coupling among the different models, meaning that the models are interconnected by sharing parameters, where the output of one model will be the input for the subsequent one. Fig. 3 shows the interconnection among different models and link to the characterization and manufacturing activities. In this sense, Gen 3b chemistry cells are manufactured and characterized to provide meaningful data for model validation purposes, such as the ones related to the electrode processing, electrolyte filling as well as performance and ageing. Once the models are calibrated and validated, optimization algorithms are implemented to automatise the battery cell optimization procedure, providing information on the optimum electrode as well as cell properties. With this information and through the DEFACTO's processing tools, the cell manufacturer will gain understanding on the best processing parameters required for achieving the next generation Gen3b battery cells. Worth to mention that the sharing of the model's inputs, output as well as the characterization data will be done through a specific datastore platform.

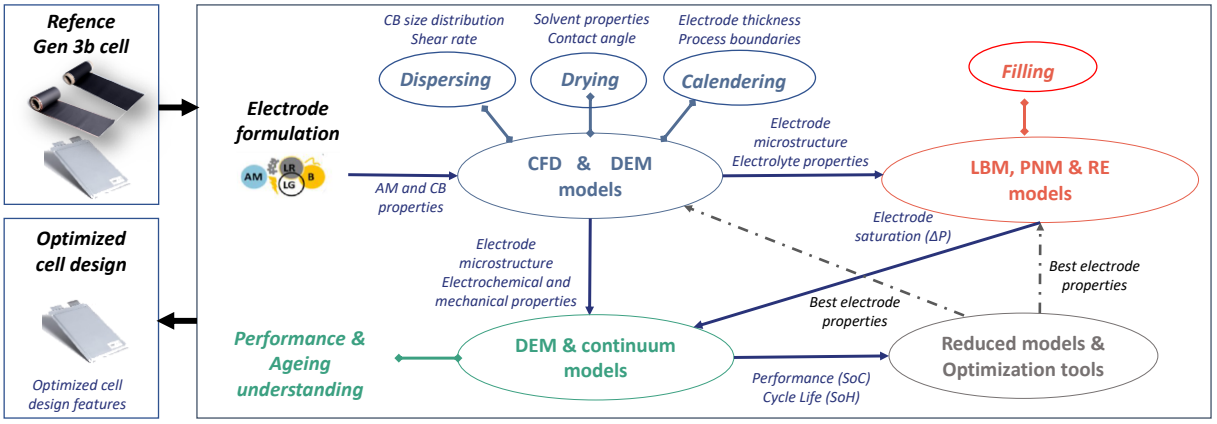

Fig. 3. Schematic work- and information flow of the methodology for cell manufacturing modelling

Several models, concerning different manufacturing stages, have been successfully developed. The models capturing electrode processing setups for the simulation of the electrode manufacturing are refined and parametrized for the utilization in different use cases. Within the slurry step, the ink rheological properties were predicted under different process parameters values. During drying, electrode thickness evolution as well as porosity was obtained playing with its process parameters. At the calendering step, stress layer thickness curves and electrode porosity were virtually predicted under different parameters values. Regarding the electrolyte filling, multi-scale simulation framework is established to capture and to optimize this process step. In particular, at the pore level capillary pressure-saturation curves under the influence of the porosity and the wetting behaviour of the active material are carefully analysed and correlate the gas entrapment. At the cell level, the wetting process was analysed, and cell saturation state was predicted at different case studies. Using this set of models, cell properties resulting from particular manufacturing configurations can be assessed.

When it comes to the cell performance and ageing, multiscale approach is followed to capture the coupling of mechanical and electrochemical ageing phenomena as well as to accurately predict the homogenized effective parameters to introduce at the continuum scale. The resulting continuum model is thus able to provide accurate predictions of the cell performance. Based on this approach, useful computational tools have been built in order to predict the performance of a particular cell design and to optimize a cell design. As previously mentioned, since the computational burden of a p4D can be very large, Model Order Reduction techniques have been used in order to accelerate the numerical simulation. The resulting computational tools have been released under a Free Open-Source Software (FOSS) license through the website of DEFACTO project (2022).

An application example is considered below, where some parameters of a G/NMC622 cell are selected to maximize the released energy under a 1C-rate discharge. For a given set of cell parameters, p, the dimensionless released energy E is defined by:

$$E(p) = \frac{1}{E_{ref}} \int_0^{t_f(p)} i_{app}(p)\, V\,(p,t)dt \tag{1}$$

where $t_f(p)$ represents the discharge final time: 3600 seconds for a 1C discharge unless a cut-off voltage V_c is reached before, in which case a premature end to the cell discharge occurs (as a result, this final time depends on the cell design parameters through an unknown function). In the example anode and cathode porosities will be considered cell design parameters in p, and assumed to take values in some prescribed intervals. Anode and cathode thickness are selected to have a fixed (theoretical) capacity. Remaining cell parameters are kept constant.

Cell optimisation will require to simulate the cell discharge (using a computationally demanding p4D model) at each optimisation iteration. At the same time, since efficient optimisation algorithms need to estimate objective function, E, derivatives with respect to each parameter, the solution sensitivities must be computed. The computational burden associated to these tasks is huge but can be significantly alleviated through the use of adaptive reduced order

models, where numerical solution using the full order model (FOM) is alternated with intervals where reduced order models (ROMs) built on-the-fly are used.

Table 2 shows the dimensionless delivered energy for the initial cell design and the first two optimisation iterations obtained by a trust region algorithm implemented in SciPy (further optimisation iterations do not significantly improve the cell released energy). Fig. 4 shows discharge voltage curves for the initial cell design and the two iterations. Shaded areas correspond to time intervals where the (expensive) FOM is used. Both as time goes by along the cell discharge or optimisation progresses, more and more information from the cells is collected and the (cheap) ROM is more able to simulate without resource to the FOM. As a consequence, cell optimisation using the developed methodology shown in Table 2 requires significantly less computation time than a single evaluation of the cell performance using (only) the FOM.

Table 2. Results of cell design optimisation

	Initial guess	First iteration	Second iteration
Dimensionless released energy [-]	0.753	0.818	0.835

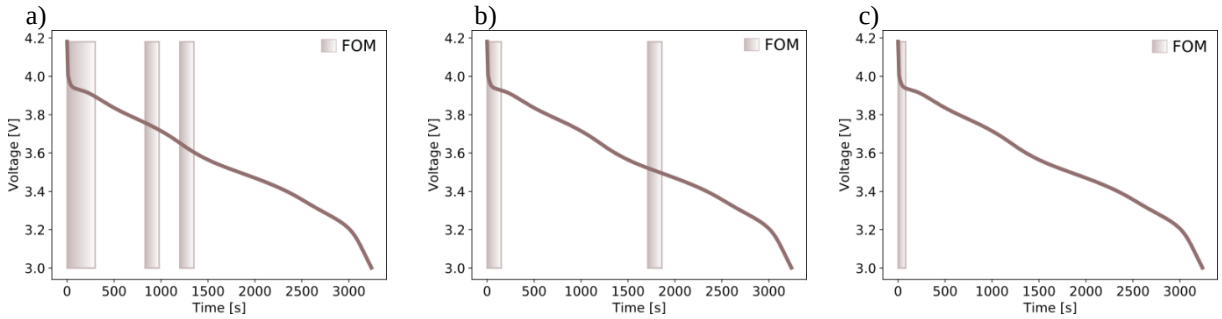

Fig. 4. Cell discharge voltage for the initial cell design and the first two optimisation iterates. Shaded regions correspond to time intervals where the FOM is used. A ROM is used for the remaining time: (a) Initial cell design, (b) First optimization iterate (c) Second optimisation iterate

With the optimized structural electrode parameters, such as the porosities of the electrodes extracted from the optimization tool, suitable process conditions and formulation parameters for electrode manufacturing and electrolyte filling process will be later identified. In the case of the electrode processing, the parameters of the subsequent mixing, drying and calendering step will be identified through the developed models in order to have the best porosity identified in the optimization tool; while in the case of the electrolyte filling, best parameters will be identified to avoid gas entrapment and permit homogeneous wettability of the electrodes.

5. Standardisation activities

Standardisation is an effective way to transfer the results of research projects to the real automotive market, as pointed out by the report on standardisation from European Commission (2022). Within DEFACTO, we aim at contributing with standards and guidelines to boost harmonized experimental and theoretical research on batteries. To do so, two CWAs (CEN Workshop Agreement) has been defined to provide a valuable reference guide for external parties. The first CWA is focused on the data required for all the models to be developed within the project and shown in Fig. 3. while the second one is focused on the experiments and characterization techniques to be carried out in order to extract the data and parameters for model calibration as well as validation.

6. Conclusion and outlook

The multiplatform tool proposed in DEFACTO provides new insights to better understand the effect of the battery cell production parameters and their impact on the cell performance, closing the loop between the manufacturing process and usage scenarios. Thus, the understanding gained through the models and simulation tools will turn into

more efficient LIB manufacturing processes and will serve as a basis for near term development of the digitisation of the LIB manufacturing plants. An application case on the use of time and parameter-adaptive optimisation tool is shown where optimized cell parameters are identified at shorten computational costs in comparison to the full order model simulations, which can facilitate the market uptake of novel LIB cells technologies for a computationally affordable tool and less time-consuming. Furthermore, the innovative and unique DEFACTO workflow for LIB cells manufacturing will respond to new society requirements and demand, making the procedure greener and more sustainable.

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