DEFACTO - battery DEsign and manuFACTuring Optimisation through multiphysic modelling

D.7.4

Date: 28/02/2023

This document is the Deliverable D7.4 “Economic potential of using models for Li-ion cell manufacturers”. This document contains an overview of the DEFACTO models and their benefits from a cell manufacture point of view. It provides the economic potential by using the DEFACTO models to reduce time and costs during the electrode and cell development.

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Content

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1 Executive Summary

This deliverable will report how the use of DEFACTO models in Li-ion cell manufacturing activities by cell manufacturers could bring economic benefits. The process chain of a Li-ion battery production is distinguished into slurry mixing, coating and drying, calendering, and electrolyte filling, which are covered separately by specific DEFACTO models. Electrochemical models were developed to connect the electrode and cell properties with cell performance such as cycle stability and c-rate capability. Moreover, an optimization tool was developed which can predict optimized electrode and cell properties such as electrode thickness, porosity and number of mono stacks.

Using the DEFACTO models and the DEFACTO workflow will significantly reduce the costs and time involved in R&D activities. In addition to a shorter and cheaper development time, this leads to a deeper insight into the process-property relationship.

2 Acronyms and abbreviations

NMC622	Li ($Ni_{0.6}$ $Mn_{0.6}$ $Co_{0.6}$) O_2
NMC811	Li ($Ni_{0.8}$ $Mn_{0.1}$ $Co_{0.1}$) O_2
LNMO	Li ($Ni_{0.5}$ $Mn_{0.5}$) O_2
G	Graphite
SI	Silicon
CB	Carbon black

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3 Introduction

The main objective of the DEFACTO project is to develop a multiphysics and multiscale modelling integrated tool to better understand the material, cell and manufacturing process behaviour. This allows to accelerate cell development and the R&I process and facilitates the development of new high capacity and high voltage Li-ion cell generation 3b batteries. This will increase the understanding of multiscale mechanisms and their interactions, reducing the R&D cell development resources, therefore unlocking an innovation-led cell manufacturing industry in Europe. [1]

This work is based on an iterative exchange process for model development, validation and optimisation using two cell technologies for the automotive market: a commercial NMC622/G cell taken from the product portfolio from one of the DEFACTO partners and last generation prototypes (NMC811/G-Si). Characterisation tests provide data for model development and validation, and for gaining understanding on ageing mechanisms. The optimization algorithms enhance cell performance and durability through optimised designs and manufacturing processes. The novel fast-track cell development procedure achieved will be further extended to LMNO/G-Si prototypes.[1]

In parallel, the set of individual multiscale and multiphysics models will be correlated through a workflow that connects outputs from a model with inputs that are needed to feed other models.

This includes mechanical and electrochemical ageing with outstanding accuracy at reasonable computational cost. The project consortium, which covers the whole cell manufacturing value chain, has the required experience to ensure a smooth and high-quality delivery of the outcomes of the project. The order and connections of the models are given in Figure 1.

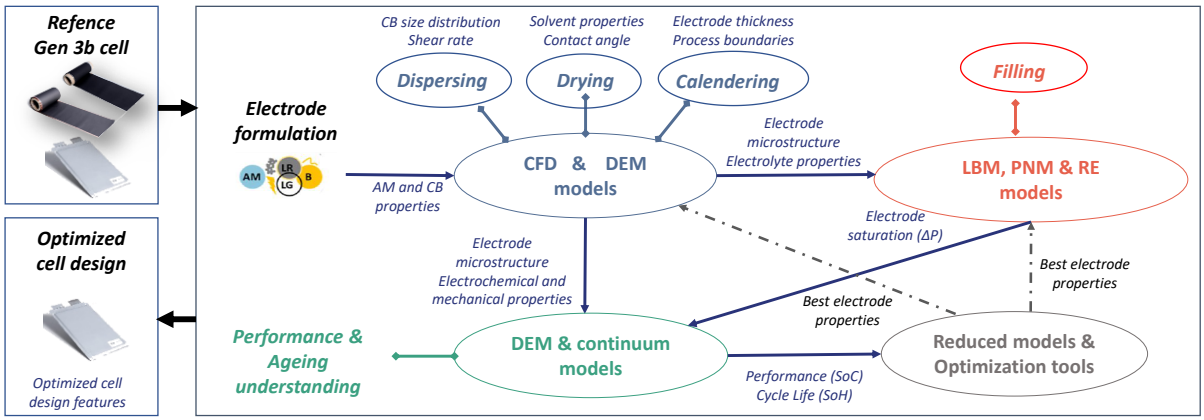

Figure 1: Connections of the DEFACTO models along the process chain.

[1] General Agreement DEFACTO

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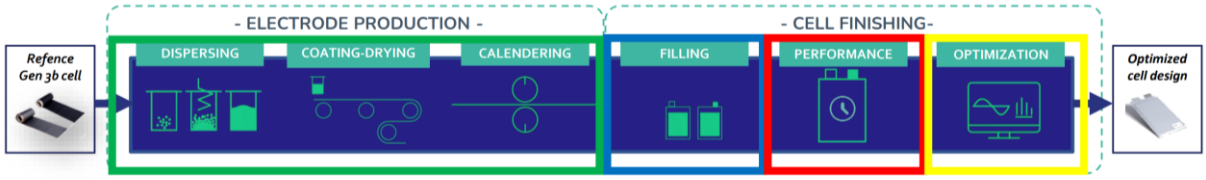
4 DEFACTO Models

4.1 Overview of the models

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 particle simulation 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, the electrolyte filling process is modelled as it is a critical step that impacts 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 are 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 Figure 2.[2]



Model codename	Method/Software	Scale	Outputs from models
CFD/DEM	Discrete Elements Method (LIGGGTHS) / CFD (OpenFOAM)	Electrode/Slurry	Slurry composition after dispersing. Microstructure after drying and calendering
LBM	Lattice Boltzmann Method with Palabos library	Electrode	Dynamic wetting behaviour, permeabilities, and electrolyte distribution
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
DFT	Density Functional Theory (DFT) method	Material	Mechanical input parameters
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)
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

Figure 2: Overview of the models linked to the process steps inside the electrode and cell manufacturing.

In Table 1 the models are listed with their outputs and benefits for cell manufacturers.

[2] María Yáñez et al. Accelerated multiscale and multiphysics modelling tools for battery cell manufacturing improvement. Transport Research Arena (TRA) Conference. 2022

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Table 1: Output and benefit for electrode and cell manufactures of each DEFACTO model in the specific process step.

Model	*Process*	*Output*	*Benefit*
CFD/DEM	Mixing	• Number, size and morphology of the CB agglomerate fragments • Effective critical breakage force	Basic research and understanding of the CB fragmentation during dispersion.
CFD/DEM	Coating - Drying	• Development of film thickness and particle volume fraction dependent on height over time • Final electrode thickness/porosity • Porosity over height • particle sizes over height	Prediction of structural electrode characteristics without the need of time and resource consuming experiments.
CFD/DEM	Calendering	• Final electrode thickness/porosity as a function of the calendering stress	Reduced experimental effort.
LBM	Filling	• Capillary pressure-saturation curve • Permeability • Highly resolved (regarding time and space) local distribution of electrolyte and gas phase in pore space	High temporal and spatial resolution; considers all relevant physical phenomena on the pore scale; efficient parallelization (for usage on many CPU-cores), open-source software.
PNM	Filling	• Capillary pressure-saturation and permeability in dependence on the structure and electrolyte-specific input parameters	High computational efficiency; free and open-source software; optimized for usability.
RE	Filling	• Time- and space-dependent distribution of electrolyte saturation on the cell scale	Better insight into wetting process on cell level; estimates of required wetting time to optimize process by choosing wetting time sufficiently long to ensure complete wetting but not longer than necessary.
DFT	Atomistic Model – Performance	In general, there can be various outcome of atomistic simulations,	

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		depending on the question to be answered. (See 4.2.5 DFT in Performance)	
DEM	Performance	• Non-linear mechanical effects at microstructure scale related to granular material	Insight into irreversible swelling of electrodes related to applied pressure on cell

BEST	Performance	• Transport parameters for electrodes and electrolyte:
		  ○ Li diffusion constant,
		  ○ Electrode conductivity, ionic conductivity,
		  ○ Transference number;
		  ○ SOC-dependent Open-circuit potential of electrodes;
		  ○ Reaction rates (Butler-Volmer).
		• Possibly more parameters for degradation, mechanical and thermal model extensions
			Insight into the influence of microstructure on cell properties; Justification of homogenized models like p4d.

p4D_ageing	Performance and Ageing	• Cell performance based on effective transport parameters • Voltage-time curve during charge/discharge	Cell performance can be easily linked to electrode properties and cell geometry, which is important for the electrode/cell manufactures.
p4D_ROM	Optimization	• Optimal selection of electrode/cell design parameters	Cell optimization can be accelerated very significantly. Typically, this tool can complete the whole cell optimization process using less computational resources than a single cell performance evaluation using the underlying p4d full order model.

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4.2 Model description

4.2.1 CFD/DEM in Mixing

The CFD/DEM model for the mixing process covers the impact of CB agglomerate size and shape inside the slurry during dispersing. Depending on the slurry viscosity, shear rates and slurry recipe the CB agglomerates experience different forces. These affect the size and morphology of the CB agglomerates, which in turn affects the slurry viscosity. The slurry viscosity is an important parameter for the coating step and the CB agglomerate size and morphology is important for the electrical conductivity of the electrode. Therefore, this model is relevant for the processing route. Its main focus is the interrelationship between CB agglomerate and the electrochemical cell performance, although it can also be used to obtain slurry characteristics for slurry preparation and electrode manufacture since it provides a basic understanding of the CB fragmentation during dispersion and the corresponding viscosity.

4.2.2 CFD/DEM in Coating – Drying

The CFD/DEM model for the coating and drying process covers the particle distribution as a function of electrode thickness and drying time. For a given wet film thickness, the dry film thickness highly depends on the electrode porosity after drying. The active material particle size distribution as well as the slurry formulation and the process conditions affect the electrode porosity. The development of the electrode and pore structure is covered by this model. Additionally, it gives insights in the distribution of the particles and pores over the electrode thickness, which is important for the electrochemical performances of the cell. This model provides microstructure information of the 3D electrode, which usually has to be obtained by complex measurement techniques like synchrotron tomography or scanning electron microscopy combined with focused ion beam which is very time and resource- intensive. Especially the predictability of the dry electrode thickness, density and porosity is a big advantage for electrode manufactures and reduces the experimental effort significantly.

4.2.3 CFD/DEM in Calendering

The CFD/DEM model for the calendering process calculates the behavior of the electrode thickness and porosity during calendering. The final electrode thickness is extremely important for stacking the electrodes for cells because the cell thickness has to stay within tolerances defined for production. The final electrode porosity, especially for the cathode, is one of the most important parameters for the rate capability of the cell. This model connects the electrode thickness and porosity with the process parameters of the calender, like the calendering stress, the temperature during calendering and the rotational speed of the calender rolls. Especially, with the knowledge of the electrode thickness and porosity as a function of the calendaring stress, the aimed porosity and thickness values can be adjusted very fast. Therefore, throughout the R&D process, expensive and time-consuming experiments can be reduced drastically. Additionally, the electrode structure is simulated and optical impressions or statistical values like tortuosity and particle- as well as, pore size distributions can be obtained from this model.

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4.2.4 Electrolyte Filling LBM, PNM and RE[3]

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.[4], it is used to simulate electrolyte flow in 3D electrodes with and without binder at sub-micrometer resolution. To also consider relevant effects at the electrode- and cell level, pore network models (PNM) and continuum methods (CM) are used, respectively. All in all, the three approaches are coupled to a framework which is schematically shown in Figure 3. The arrows show the flow path of external and internal input data needed for the different simulation approaches. They are depicted in blue, orange, and grey for microscopic input data (e.g. material properties), macroscopic input data (e.g. process parameters), and internal input data, respectively. The workflow within the framework is the following. For each battery component, the capillary pressure-saturation behavior and electrolyte permeability are determined once as references using an LBM simulation. These data are used to calibrate the geometrical correction factor of the PNM which is then applied to efficiently conduct large parameter studies. The resulting capillary pressure-saturation curves and permeability correlations are used as input to the cell-scale simulations, where finally also geometrical properties of the battery cells as well as process conditions are considered. All in all, the results are useful to optimize the filling process and serve as a baseline for further electrode and electrolyte design. The optimized process parameters are further defined in Deliverable D4.3.

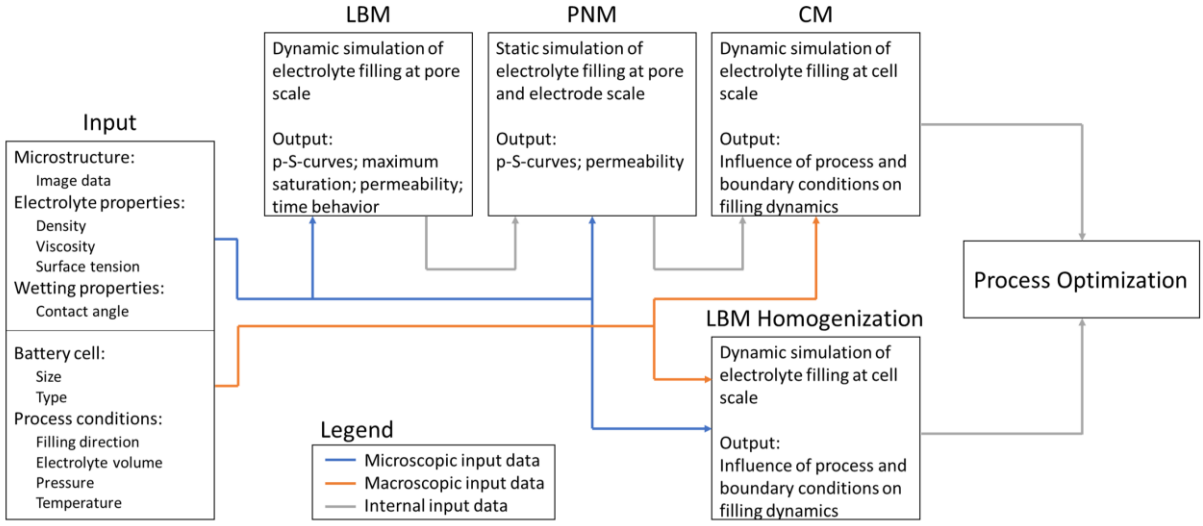

Figure 3: Network of input and output parameters for the filling models LBM, PNM and CM.[5]

[3] María Yáñez et al. Accelerated multiscale and multiphysics modelling tools for battery cell manufacturing improvement. Transport Research Arena (TRA) Conference. 2022

[4] Lautenschlaeger, M. P., Prifling, B., Kellers, B., Weinmiller, J., Danner, T., Schmidt, V., Latz, A., 2022. Understanding Electrolyte Filling of Lithium-Ion Battery Electrodes on the Pore Scale Using the Lattice Boltzmann Method. Batteries & Supercaps 5.7, 1-14.

[5] DEFACTO - Deliverable D4.2

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4.2.5 DFT in Performance

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. The DFT model for the active material aims the points below:

- Chemical and electrochemical reaction paths and energies
- Volume changes;
- Ionic diffusivity, and conductivity;
- Electronic conductivity;
- (Electro)chemical potentials;
- Surface and interface energies;
- Mechanical properties: Bulk modulus, shear modulus, Young's modulus, Poisson ratio

DFT modeling allows numerous possibilities to investigate. An additional, very important advantage of atomistic modeling is the possibility to calculate material parameters needed as input for models acting on higher scales (such as the Finite elements method (FEM)). The approach in the DEFACTO project is not to use one model on its own, but to combine it with other models. Therefore, it is not feasible to assess the benefit for only one process step along the production line, since all process steps are interlinked with one another.

4.2.6 Cell performance and optimization

4.2.6.1 DEM in Performance

The DEM for the cell performance investigates specifically the influence of cycling and external mechanical load on the electrode microstructure and thickness change. DEM can more easily take into account non linear phenomena like bond breaking between particles and microstructure changes due to the breathing of silicon rich agglomerates.

4.2.6.2 BEST in Performance

In the three-dimensionally resolved electrode microstructure the BESTmicro and BESTmeso models can describe ion and charge transport. Derived from that, values like SOC, cell potential, limiting current, sources of overpotentials, internal resistances, heat sources, etc. can be calculated.

In contrast to the DEM, the BEST models are focused on continuum mechanics. They also include mechanical modelling, but some hysteretic effects are hard to reproduce. Mechanical information gained from DEM modelling can be injected into continuum models to correct some quantities like effective young modulus of the medium.

The BEST models can deliver important parameters like effective properties of the porous medium for the p4D model.

4.2.6.3 P4D_aging in Performance

A pseudo-X-Dimensional (pXD) model that extends the original pseudo-2-Dimensional (p2D) model, proposed by Newman and co-workers for the modelling of lithium-ion battery cells to work with 1D, 2D or 3D cell geometries is developed within this project. In the DEFACTO project, the focus is put on the 3D geometry and therefore on the p4D model. Therefore, charge balance, mass balance and reaction kinetics, as well as energy balance, are spatially resolved for the entire cell geometry, considering the inhomogeneity of cell state properties. For parametrization, among others GITT, EIS, rate capability test are required. Alternatively, non-invasive techniques such as numerical fitting

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against electrochemical rate capability test can be used for parameter estimation purposes. Furthermore, a thermal model as well as SEI growth and the loss of active material due to concentration gradients in the active material particles can be coupled to the electrochemical model.

4.2.6.4 Summary

DEM models are mechanical only models that are specifically designed to address the problem of electrode swelling, in particular when silicon is used. They provide validation and calibration information to the more general BEST micro/meso models that primarily focus on the electrochemistry but also include the geometry change due to breathing of active materials. Microscale and mesoscale model results are then interpreted to extract effective properties of the electrodes that are needed by the p4D homogenized methods.

The latter are the fastest and the currently the best compromise between speed and accuracy to address cell design optimization problems.

4.2.7 P4D_ROM in Optimization

This tool represents the implementation of a ROM-based algorithm. At the core, the tool incorporates time- and parameter-adaptive model order reduction techniques in order to speed-up cell optimization tasks (based on p4D cell models). Inexpensive model sensitivities are also computed in order to use fast, gradient-like optimization techniques. Based on a p4D model, the optimization tool solves a minimization problem. It aims to identify the best electrode parameters like thickness, porosity and the best cell parameters like height and width of the cell and the number of individual cathode-anode pairs for the stacking process.

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5 Example for a development process

This example covers the application of DEFACTO models in comparison of an experimental approach. Assumption: There is a well-known electrode and cell production. The process chain is modeled by the DEFACTO models and thus the DEFACTO models are validated on this system. The R&D department of a company is changing the active material for the cathode, which also brings with it a different particle size distribution and electrochemical behavior. Such an example is already shown in the DEFACTO project by substituting the cathode active material $Li(Ni_6Mn_2Co_2)O_2$ by $Li(Ni_8Mn_1Co_1)O_2$. The electrode structure is highly affected by the active material particle size. The most optimal electrode porosity and pore size distribution regarding the electrochemical cell properties is unknown. After entering all relevant data (like particle size distribution, and specific surface area of the new active material) in the DEFACTO p4D model for optimization, the optimal electrode parameters like electrode thickness and porosity is the output of the simulation. The next step is to get the relationship between the electrode porosity and the calendering stress. The DEFACTO CFD/DEM model has to be calibrated by one or two calendering experiments with the new active material. The results are important values for calibrating the binder stiffness and damping in the model. After the calibration of the new electrode system, the electrode thickness/porosity can be calculated as a function of the calendaring stress. After testing a prototype with these parameters obtained by the DEFACTO models, the developing process is successfully completed.

The workflow of an experimental approach takes much more time, cost and effort: To find the most optimal electrode parameters numerous experiments are needed. To identify the optimal electrode porosity a lot of calendering experiments in small porosity steps are necessary. All these electrodes would have to be built into cells. These cells would have to be electrochemically tested at different c-rates and long-term cycling to find out which electrode porosity shows the best cell performance. Especially the long-term cycling takes weeks or months. Only after evaluation of the cycling data and connection with the electrode porosity, a statement can be made which electrode porosity is the most appropriate one.

In reality, the shown example is a multifactorial problem. By using a new active material, not just the optimal electrode porosity is changed. Also, the capacity balancing from cathode to anode is shifting, which means that the mass loadings have to be adjusted. Thus, the electrode thickness is influenced, which results in different electrode stack thicknesses. Therefore, it could be necessary to set a different number of cathode-anode pairs during stacking. This shows how complex a simple substitution of the active material is. This is also true for changing the particle morphology with the same cell chemistry. This leads to comparable influences on the entire process chain and cell performance. By the experimental approach, such developments take a lot of experiments, time, and resources.

By comparing the workflow by using DEFACTO models and the experimental approach, the latter one is extremely time and cost consuming. Doing expensive, time-consuming experiments can be avoided or reduced by using DEFACTO models. Additionally, an acceleration of the R&D process is possible.

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6 Assessment of the DEFACTO workflow

Quantifying the economic potential of the DEFACTO workflow compared to an experimental approach cannot be answered easily and requires many assumptions. An estimation of the economic potential could be possible by focusing on one specific process step and by neglecting the interaction of manufacturing steps and the cell performance. The economic potential is separated into three topics. First come the required experiments, second the costs and third the time. Depending on the process step, the costs for each experiment are different. The costs for one experiment are 20 k€, 2.6 k€, 3 k€ and 0.5 k€ for slurry mixing, coating and drying, calendering, and electrolyte filling, respectively, based on the development process at Leclanché. Derived from the number of experiments, the costs and time for the development process can be calculated, see Figure 5.

	Required experiments		Costs		Time	
	Current status	DEFACTO	Current status	DEFACTO	Current status	DEFACTO
Slurry mixing	3	2	60 k€	40 k€	1 d	1 d
Coating and drying	6	3	15.6 k€	7.8 k€	2 d	1 d
Calendering	10 per electrode, 4 per cell batch	2 per electrode, 2 per cell batch	30 k€	7.2 k€	2 d	1 d
Electrolyte filling	8	4	4 k€	2 k€	1 d	0.5 d

Figure 4: Resource consumption for cell development at Leclanché facilities at the status compared to the estimated DEFACTO approach.

As it is shown in section “5 Example for a development process”, a real optimization process is complex and highly dependent on the previous knowledge of the existing system. Changing one parameter can lead to changes anywhere in the process chain. With the help of the DEFACTO models, these changes can be understood very well and adjusted in a targeted manner.

7 Conclusions and Outlook

DEFACTO set ambitious quantitative targets, i.e. reduction of the cell development time by 30%, decrease the number of experiments needed for cell design and manufacturing optimization by a factor of 3, reduction of battery R&I costs by 20%. Results are so far presented in a qualitative form.[6] For the purpose to get a quantitative assesment the DEFACTO consortium is working at different case studies to get more experience by working with the DEFACTO workflow and to optimize the workflow. The project has the potential to achieve the desired impact and the case studies are expected to further develop this aspect for the final reporting.

From an industry perspective, the biggest advantage comes from networking the models, in particular with the optimization tool. One of the most important point for companies is that the models are easy to use. That is why we suggest an user-friendly interface where the models are running in the background and linked together.

[6] DEFACTO Review Report 2

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