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Data required for modelling the material, cell and manufacturing process for cells for the automotive market

CCMC will prepare and attach the official title page.

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European foreword

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Introduction

Modelling the material, cell and manufacturing process behaviour allows to accelerate cell development and the R&D process. The work can be done on an iterative exchange process for model development, validation and optimisation using two cell technologies for the automotive market: an industrial scale state of the art Layered Oxide LiNi0.6Mn0.2Co0.2O2 NMC622/Graphite cell (NMC622/G) and a competitive Nickel Rich Layered Oxide LiNi0.8Mn0.1Co0.1O2 NMC811/silicon-carbon composite prototype (NMC811/G-Si). Additionally, High-Voltage Spinel Oxide LiNi0.5Mn1.5O4/silicon-carbon composite (LMNO/G-Si) can be studied to explore the versatility of the built models.

Modelling work requires input parameters and data for validation. Before starting the experimental work, it is necessary to define precisely the nature, the sensitivity requirements for input parameters and the appropriate experiments and characterisation techniques for a list of physical and chemical characteristics.

This CWA is based on some of the results of the European Union’s Horizon 2020 research and innovation programme DEFACTO (funded under grant agreement No 875247).

1 Scope

This document specifies the data required for modelling the material, cell and manufacturing process for cells for the automotive market, based on physical and chemical characteristics of cells of NMC622/G, NMC811/G-Si, LMNO/G-Si chemistry types.

This document shall be read in conjunction with the document prCWA XXXX-2 Experiments and characterisation techniques for data required for modelling cells".

2 Normative references

prCWA XXXX-2, *Experiments and characterisation techniques for data required for modelling cells*

3 Acronyms and abbreviations

NMC	$LINI_XMN_YCO_ZO_2$ WITH $x + y + z = 1$
NMC622	$LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$
NMC811	$LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$
LMNO	$LiMn_{1.5}Ni_{0.5}O_4$
G	Graphite
Si	Silicon
PNM	Pore Network Model
P4D	Pseudo 4D
DEM	Discrete Elements Method
CFD	Computational Fluid Dynamics
LBM	Lattice Boltzmann Method

4 List of Models

The multiscale models follow a bottom-up approach: atomistic models' parameters will be scaled up to homogenised parameters to be used in the continuum model. The scale range is extended from atomistic to continuum and the physical domains covered (adding mechanical ageing mechanisms to the electrochemical ones) of the cell performance and ageing model built. To do so, different models, simulation methods, and software tools are used (see Table 1).

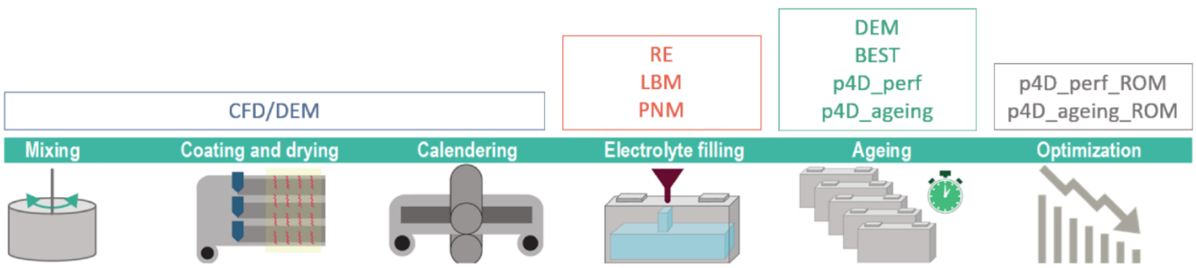

Figure 1 — Models in the process workflow

Table 1 — List of models

Model codename	Method/Software	Scale	Output from model and characterization	Output for battery optimisation
CFD/DEM	Discrete Elements Method with LIGGGHTS coupled with Computational Fluid Dynamics (OpenFOAM)	Electrode/ Slurry	Slurry composition after dispersing and microstructure after drying and calendering	Optimisation of carbon black structure for maximum electric conductivity. Optimisation of the mixing process. Efficient electrode structure prediction, optimisation structure regarding ionic and electric conductivity. Mechanical integrity. Systematic electrode design.
LBM	Lattice Boltzmann Models using in-Palabos	Electrode	Wetting behaviour and electrolyte distribution for a selected range of influencing factors at high level of detail	Optimised electrode materials and structures. Optimal electrolyte filling process conditions
PNM	Pore Network Models using OpenPNM and PoreSpy	Electrode/ Cell	Wetting behaviour and electrolyte distribution for a broad range of influencing factors at medium level of detail	
RE	Richards-Equation using in-house code FLUID	Cell	Simulation of filling process	
DEM	Discrete Elements Method with LIGGGTHS	Electrode	Mechanical behaviour of electrode (performances and ageing)	Deformation (evolution of microstructure) and change of material properties due to mechanical ageing. Efficient continuum model (p4D cell model) including mechanical and electrochemical ageing (addressing inhomogeneous and homogeneous ageing). Optimum conditions for using and battery pack design.
p4D_perf	Finite Element platform FENICS	Cell	Electrochemical and mechanical behaviour of the cell	
p4D_ageing	Finite Element platform FENICS	Cell	Electrochemical and mechanical degradation of the cell	
p4D_perf_ROM	Finite Element platform FENICS and Model Order Reduction tool EchROM	Cell	Electrochemical and mechanical degradation of the cell	

Model codename	Method/Software	Scale	Output from model and characterization	Output for battery optimisation
p4D_ageing_ROM	Finite Element platform FENICS and Model Order Reduction tool EchROM	Cell	Electrochemical and mechanical degradation of the cell	
BEST	Continuum model	Electrode	Mechanical behaviour of electrode (performances and ageing)	

5 List of Model requests

The following tables list all the data and detail the data usage (input parameters, model calibration[1]) or model validation[2]). Some methods for data determination and experimental techniques have been proposed. The descriptions of the experiments and the data types are contained in the prCWA XXXX-2.

Table 2 — List of requested data for models' construction and validation on electrode processing

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
CFD/DEM	model input parameter	Solvent viscosity (solvent for slurry preparation)	Rheological Measurements	**Rheology electrode**
CFD/DEM	model input parameter	Solvent surface tension	Tensiometer	**Tensiometer**
CFD/DEM	model input parameter	Solvent density	Mass/Volume; literature for water and NMP solvent	**Solvent density**
CFD/DEM	model input parameter	Particle Shape	µCT (T3.5)	**µCT**
CFD/DEM	model input parameter	Particle size distribution (Active Material + Additive)	Laser diffraction analysis	**Laser diffraction**
CFD/DEM	model input parameter	AM and Additive density	Literature pycnometer	**Pycnometry**
CFD/DEM	model input parameter	Poisson's ratio of AM and additive particles	Literature	**Literature**
CFD/DEM	model input parameter	Coefficient of restitution of particles	Literature	**Microcompression/ Nanoindentation**
CFD/DEM	model input parameter	Young's modulus of AM and additives	Nanoindentation Literature	**Microcompression/ Nanoindentation**

1) Model calibration also called analytical validation, refers to the identification of individual parameter.

2) Model validation refers to the comparison and quantification of the results obtain by numerical simulation and experimental data.

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
CFD/DEM	model calibration	Friction coefficients	Angle of repose	**Angle of repose**
CFD/DEM	model calibration/ validation	CB agglomerate strength	Microcompression	**Microcompression/ Nanoindentation**
CFD/DEM	model calibration	VdW-Attraction/ Electrostatic repulsion	Zeta Potential Measurement, Agglomeration Kinetics	**Zeta potential**
CFD/DEM	model input parameter	Slurry viscosity	Rheological Measurement	**Rheology electrode**
CFD/DEM	model input parameter	Shear rate during dispersing	Simulation/Calcul. based on stirrer tip speed/planetary motion, viscosity	**Simulation**
CFD/DEM	model calibration/ validation	Electrode 3D structure before and after calendering	SEM, µCT in T2.2 and T3.5	**FIB-SEM and µ-CT**
CFD/DEM	model calibration/ validation	Pore size distribution of electrodes	Mercury intrusion	**Mercury intrusion**
CFD/DEM	model calibration/ validation	Binder distribution	SEM, EDX on cathode, Later Induced Breakdown Spectroscopy on anode with CMC	**SEM-EDX**
CFD/DEM	model input parameter	Mass related slurry component concentrations	simple calculations based on formulations used in WP7	**Simulation & Slurry density**
CFD/DEM	model input parameter	Coating thickness right before drying	In-line measurement	**Coating thickness**
CFD/DEM	model input parameter	Coating temperature during drying	Infrared, T° sensor	**Coating_T_drying**
CFD/DEM	model input parameter	Convection	Volumetric flow measurement	**Flow_T_drying**
CFD/DEM	model input parameter	Air temperature	T° sensor	**Flow_T_drying**
CFD/DEM	model input parameter	Drying rate	Solvent content during drying/coating mass during drying/ coating thickness during drying	**Electrode thickness drying**
CFD/DEM	model calibration/ validation	Adhesion strength	Material Testing Machine	**Electrode adhesion strength**

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
CFD/DEM	model calibration/ validation	Micromechanical properties of electrode	Nanoindentation	**Microcompression/ Nanoindentation**
CFD/DEM	model calibration/ validation	Electrode thickness before and after calendering	laser triangulation for contact-free measurement (or tactile measurement if not available)	**Electrode thickness**
CFD/DEM	model calibration/ validation	Electrode elastic recovery thickness during calendering (depending on gap between rolls)	Force-Sensors, Displacement Sensors, in line measurement	**Electrode thickness calendering**
CFD/DEM	model calibration/ validation	Temperature of calender rolls		**T-calendering**
CFD/DEM	model calibration/ validation	Lab scale drying behavior for model development	Tabletop coater + heating + laser triangulation	**Electrode thickness drying**

Table 3 — List of requested data for models construction and validation on electrolyte filling process

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
LBM/PNM	model input parameter	Electrode and separator morphology	3D-FIB-SEM in T2.3 and DEM Simulations in WP3	**FIB-SEM and µ-CT and tortuosity separator**
LBM/PNM	model input parameter	Electrolyte surface tension	Tensiometer	**Tensiometer**
LBM/PNM	model input parameter	Electrolyte contact angles with active materials, separator	Optical measurements on model materials	**Angle of repose**
LBM/PNM/RE	model input parameter	Electrolyte density	Balance	**Electrolyte density**
LBM/PNM/RE	model input parameter	Electrolyte viscosity	Rheometer	**Rheology electrolyte**
LBM/PNM/RE	model calibration/ validation	Pressure Saturation curves of electrodes and separator	Measurements in special setup	**p-s-curves**
RE	model calibration/ validation	Relative permeabilities of electrodes and separator	calculation; empirical approximation	**Angle of repose**
LBM/PNM/RE	model calibration/ validation	Electrolyte distribution pore scale	Ultrasonic acoustic wave or operando tomography/radiography with extra-collaboration with Large Instruments?	**Acoustic and Chronoamperometry**

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
RE	model validation	Electrolyte distribution cell scale	Impedance measurements on segmented cell	**Segmented cell**
RE	model validation	Pressure profiles of filling process over time	Pressure sensors in production process	**Pressure electrolyte**

Table 4 — List of requested data for models construction and validation on cell performances and ageing

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
DEM	model input parameter	Young modulus of bulk active materials	DFT calculation in T5.1	**Simulation**
DEM	model input parameter	Binder rheological behaviour	nanoindentation	**Microcompression/ Nanoindentation**
DEM	model calibration/ validation	SEI thickness evolution	2D imaging, XPS, thickness measurement upon aging	**XPS and EQCM**
DEM	model input parameter	Electrode morphology	3D-FIB-SEM in T2.3	**FIB-SEM and μ-CT**
DEM	model input parameter	Volume change of active materials in negative electrode	Literature or μ-CT? Or tomography with extra-collaboration with Large Instruments?	**μCT**
DEM	model input parameter	Positive electrode swelling	Swelling measurement on monolayer pouch cell upon cycling with dedicated test bed	**DFORM**
DEM	model input parameter	OCV profile	Literature and half-cell or 3-electrodes measurements in T2.4	**GITT**
DEM	model validation	Electrode thickness evolution	Swelling measurement on monolayer pouch cell upon cycling with dedicated test bed	**DFORM**
BEST	model input parameter	Young modulus of bulk active materials, binder and separator	calculation/experimental	**DFORM for separator; literature**
BEST	model input parameter	Poisson ratio of bulk active materials, binder and separator	calculation/experimental	**DFORM for separator; literature**
BEST	model input parameter	volumetric expansion coefficient of bulk active materials	calculation/experimental	**XRD?**
BEST	model input parameter	electrode morphologies (3d)	calculation (WP3)	**FIB-SEM and μ-CT**

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
BEST	model input parameter	Which other parameters might be relevant for mechanical aging? e.g. yield strength of active materials	experimental	**NA**
BEST	model input parameter	max. Li Concentration of active materials	experimental/literature	**GITT**
BEST	model input parameter	electronic conductivity of active materials	experimental/literature	**EIS**
BEST	model input parameter	Li Diffusivity of active material	experimental/literature	**GITT**
BEST	model input parameter	Butler-Volmer reaction rate of active materials	experimental/literature	**Electrochemical tests**
BEST	model input parameter	OCV of active materials	experimental/literature	**GITT**
BEST	model input parameter	ionic conductivity of electrolyte, and Carbon Binder	experimental/literature	**EIS**
BEST	model input parameter	Li+ Diffusivity of electrolyte and Carbon Binder	Galvanostatic pulse-relaxation (Li-Li)	**GITT**
BEST	model input parameter	Li+ transference number of electrolyte	experimental EIS (Li-Li)/literature	**EIS**
BEST	model input parameter	effective electronic conductivity of Carbon Binder (optional)	literature	**Literature**
BEST	model input parameter	porosity of Carbon binder (optional)	literature	**Literature**
BEST	model validation	cell voltage	experimental	**Electrochemical tests**
BEST	model validation	Quantification of mechanical aging	experimental	**Electrode adhesion strength**
BEST	model validation	Electrode thickness evolution	experimental	**DFORM**
p4D_perf & p4D_perf_ROM	model input parameter electrolyte	Ionic conductivity	EIS (Electrode - Electrode)	**EIS**
p4D_perf & p4D_perf_ROM	model input parameter electrolyte	Diffusion coefficient in electrolyte	Galvanostatic pulse-relaxation (Li-Li)	**GITT**

Model codename	Data usage	Description of requested data	Method for data determination	Name of experiments
p4D_perf & p4D_perf_ROM	model input parameter electrolyte	Transference number	EIS (Li-Li)	**EIS**
p4D_perf & p4D_perf_ROM	model input parameter - electrolyte	Initial concentration	Material provider	**Input of electrolyte manufacturer**
p4D_perf & p4D_perf_ROM	model input parameter	mechanical parameters/ properties	Model upscaling from DEM and Cont-ITWM	**Simulation**
p4D_perf & p4D_perf_ROM	model input parameter	Specific heat capacity (Cp), thermal conductivity	Thermal characterization	**ARC, TGA-GC**
p4D_perf y p4D_perf_ROM	model validation	Cell performance tests	C-rate tests, cell thermal inhomogeneities, electrode swelling with lithiation procedure	**Electrochemical tests**
p4D_ageing & p4D_ageing_ROM	model input parameter	mechanism identification (SEI growth, Li plating), physicochemical equations, parameters estimation	Ageing tests with post-mortem analyses (NMR, TEM, dQ/dV, reference electrode measurement, …)	**Test campaign**
p4D_ageing & p4D_ageing_ROM	model input parameter	mechanism identification (active material cracking, active material loss), physicochemical equations, parameters estimation	Ageing tests with DFORM at small pouch cell level	**Test campaign**
p4D_ageing & p4D_ageing_ROM	model validation	aging test results of finished cells, characterisation of finished electrodes, etc)	Ageing tests with deformation gauges? With tomography?	**Test campaign**