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The ARTISTIC Online Calculator: Exploring the Impact of Lithium-Ion Battery Electrode Manufacturing Parameters Interactively Through Your Browser

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This article presents the ARTISTIC online calculator, a web platform that enables both experimental and computational researchers to access the ARTISTIC project three-dimensional (3D) manufacturing models. This platform is free of charge and utilizes a user-friendly interface to guide users among the different manufacturing steps and their parameters. The current version of the online calculator accounts for the slurry phase, its drying, and electrode calendering; it gives access to a variety of relevant parameters, as slurry solid content, electrode formulation, particle size distribution, and drying/calendering conditions. To utilize this platform, the user should simply register freely to the ARTISTIC computational portal, select the manufacturing parameters of interest, and launch the simulations through the user-friendly interface. As soon as the simulation ends, the user who launched it receives an email with the links to visualize and recover the resulting electrode/slurry microstructure. Furthermore, all the results obtained through this platform are shared among all the users, i.e., everyone can visualize and recover all the microstructures available. Therefore, this platform also constitutes an open-access database linking manufacturing conditions and simulated electrode microstructures. In addition, these microstructures can be embedded straightforwardly in electrochemical models. In brief, we hope that the battery community will see this online platform as a tool to explore the vast manufacturing parameter space accessible through our 3D models and to establish a deeper knowledge of the manufacturing-microstructure-electrochemistry relationships in a collaborative and fully transparent way.

Introduction

The interest in battery research is raising among governments, international institutions, and industries due to the expected growth of the electric vehicle (EV) market and their possible usage for stationary applications.[1–3] Among the different battery technologies, lithium-ion batteries (LIBs) are the state-of-the-art for commercial applications, but their performance and cost should be further improved for making their market expansion economically convenient. In this context, the improvements and upscaling of LIB manufacturing were the main drivers of the drop in LIB cost during the last decade, and there is still space for further improvements.[4]

The production of LIB electrodes constitutes a highly convoluted process, comprised of a series of subsequent steps, each affected by a large number of parameters.[5] Disentangle the effects of each parameter on the arising electrode microstructure (referred to as mesostructure in previous publications

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by us) and associated electrochemical performance is a complex task. Three-dimensional (3D) models can be used as an advantageous approach to study the effect of each parameter, and their combinations, on the electrode microstructure and electrochemical performance.[6]

In particular, 3D physics-based models simulating LIB electrode manufacturing represent a good compromise between accuracy and throughput.[7,8] Furthermore, they allow accurate control of each manufacturing parameter accounted in the model, unlocking a more detailed study of the relationships between manufacturing and 3D electrode microstructure. In addition, if combined with heterogeneous 4D (3D + time) electrochemical models, a direct link between manufacturing, microstructure, and performance can be established. In this context, and within the ERC-funded ARTISTIC project,[9] our group recently demonstrated to have developed a series of manufacturing models accounting for the slurry phase,[10] its drying,[11,12] electrode calendering,[12,13] and electrolyte filling,[14] which can be directly coupled to 4D electrochemical models simulating galvanostatic discharge[15] and electrochemical impedance spectroscopy.[16] Our group has also presented these tools in the ARTISTIC project Webinar Series organized since 2020 by us.[17]

In this concept article, we introduce the ARTISTIC online calculator, an interactive and free web application that allows simulating LIB electrode manufacturing in 3D through a user-friendly interface. The user can directly control a number of relevant LIB electrode manufacturing parameters, from the slurry solid content (SC) and electrode formulation to the active material (AM) particle size distribution (PSD) and drying/calendering conditions. Afterwards, the user can launch the associated simulation and visualize/download the arising 3D microstructure. The simulations launched through this web interface are run in devoted computational resources, and the results are shared among all the users. This makes the online calculator both a user-friendly tool to simulate LIB electrode manufacturing *and* an open access and FAIR[18] database of simulated 3D electrode microstructures linked to specific manufacturing conditions.

Currently, the ARTISTIC online calculator allows its users to simulate three subsequent steps of LIB electrode manufacturing: the slurry phase, its drying, and electrode calendering. These three steps are directly connected in the platform, meaning that the slurry microstructure obtained is used as input of the drying model, and the dried electrode microstructure is used as input of the calendering model. In addition, in the near future we plan to embed also our electrolyte filling[14] and galvanostatic discharge/charge[15] models.

In the following sections, we discuss the specificities of the ARTISTIC manufacturing models, we present the online calculator, and we illustrate all the parameters that can be tuned by the users. Lastly, we focus on which are the benefits that we expect and hope this platform will bring to the battery community, and we conclude by discussing the perspectives of this web application.

The ARTISTIC Manufacturing Models

During the last years, one of the main goals of our ERC-funded ARTISTIC project[9] has been the production of accurate 3D physics-based models simulating different LIB electrode manufacturing processes. Three of these models, the slurry phase, its drying, and electrode calendering, are already implemented in the ARTISTIC online calculator and are briefly discussed below. All these models rely on a coarse-grained particle dynamics approach, describing explicitly the secondary AM particles, while accounting for the carbon-binder domain (CBD) phase through CBD particles. Each CBD particle describes here one CBD agglomerate, accounting for carbon, binder, and the nanoporosity of these agglomerates.[19]

The first model accounts for the slurry phase. In this context, the CBD particles are treated as effective particles accounting not only for carbon and binder, but also for the solvent. For this, the CBD particles at the slurry phase are expanded and their density decreased, which allows reproducing the rheological properties of their experimental counterpart, as we recently demonstrated.[10] During this step, many of the characteristics of the final electrode are defined, as the AM PSD and the electrode formulation, i.e., the weight percentage (wt.%) of AM and CBD. The calculation of the number of AM and CBD particles accounts for the AM PSD, the CBD particle size and nanoporosity, AM and CBD weight percentages, and the mass of the electrode fraction to be simulated. Both AM and CBD particles are approximated to be spherical. The CBD nanoporosity found experimentally is equal to 50%,[19] but, to the best of our knowledge, no study analyzed the effect of manufacturing conditions, nor the effect of carbon to binder weight ratio or chemistry, on the CBD nanoporosity. Therefore, in the online calculator we have decided to leave the freedom of controlling this parameter, and in particular the nanopores' volume fraction in the CBD phase can range between 30% and 70%. In addition, the slurry model allows the explicit control of the slurry SC, i.e., the mass of the solid components divided by the mass of the slurry (solid components + solvent). This is accounted for by defining the size of the expanded CBD particle as a function of the solvent volume.

The slurry model considers periodic boundary conditions (PBCs) for all the direction (x, y, z) to enhance its representativeness.

Two different drying models were developed and implemented in the platform, here referred to as homogeneous and heterogeneous drying approaches. The first simulates drying by shrinking the CBD particles, to remove completely the solvent, at the very beginning of the simulation, which leads to a structure re-organization and shrinkage, and finally to the dried electrode microstructure. This method was tested on multiple conditions, and it is the reference drying approach employed in our group. However, this approach cannot capture heterogeneities developed during drying in terms of additive migration.[20,21] Therefore, if the user is interested in capturing this phenomenon, s/he should employ the heterogeneous drying approach. This methodology was recently reported by us,[12] and it demonstrated to allow capturing additive migra-

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tions as a function of the drying rate (DR), i.e., the higher the DR, the higher the carbon migration and the arising electrode heterogeneities. Nonetheless, it should be accounted that this model is very recent, and it was not possible to test it as extensively as the homogeneous approach. The choice of releasing this model into the platform is also a way to test it more deeply in a collaborative manner.

The homogeneous drying approach considers the boundaries in all the directions (*x*, *y*, *z*) as PBCs, while the heterogeneous drying approach considers *x* and *y* as periodic and *z* (electrode thickness) as non-periodic.

The third model included in the ARTISTIC online calculator simulates electrode calendering, which is performed by applying a first plane at the electrode bottom (current collector), a second plane at its top (calendering roll), and moving downward the top plane to mimic electrode compression. This model was utilized in a recent work by us,[12] and is an upgraded version of our previous calendering model.[13]. In addition to the degree of electrode compression, this model accounts for other two parameters: the consideration (or not) of the elastic recovery, and the CBD nanoporosity decrease. The elastic recovery is a phenomenon observed experimentally[22–24] and refers to the tendency of calendered electrodes to recover part of their initial thickness after a few hours/days of the calendering step. Concerning the CBD nanoporosity, to the best of our knowledge no evidence of its decrease during calendering was previously reported. However, it is reasonable to believe that calendering reduces nanopores, which is the reason why this parameter was implemented. Briefly, the CBD nanoporosity decrease is accounted by shrinking the CBD size iteratively during electrode compression.

The calendering model considers *x* and *y* directions as periodic and *z* (electrode thickness) as non-periodic.

All these models were developed using the open-source software LAMMPS and utilizing a combination of two force fields (FFs): the Lennard-Jones[25] (LJ), mimicking the adhesive forces between particles, and the granular Hertz[26] (GH), accounting for the mechanics of the system. The FF parameter values used in the online calculator are kept constant and were optimized to allow an accurate description of the slurry and electrode macroscopic characteristics, as slurry density and electrode porosity, for a large spectrum of different conditions. However, considering the vast parameter space accessible through this platform and the dependence of these properties on the operator and the machinery used, a single set of FF parameter values cannot reproduce exactly the features of any possible slurry/electrode manufactured experimentally. This means that the slurry and electrode characteristics obtained through the platform should follow the main trends observed experimentally and output similar values compared to experiments. However, for a more accurate fitting, a devoted FF parametrization should be performed. For this, the interested users can utilize the open-source code of these models on the Github page of the ARTISTIC project[27] and optimization algorithms previously reported by us to accelerate the FF fitting process.[10]

Figure 1 reports an example of a series of thin structures obtained through this platform for two arbitrary sets of AM PSD, formulation, SC, drying and calendering conditions. Thicker electrodes can be simulated as well through our computational procedure. Here, ρ stands for slurry density, and ε_{bulk} and ε_{nano} stand for the bulk (in between the particles) and nano (accounted implicitly into the solid CBD particles) porosities.

Despite that the ARTISTIC website contains already in house experimental data, the main limitation of this modeling approach is the difficulty of systematic 3D-resolved validations with respect to experimental microstructures. This would require large datasets constituted of series of electrode microstructures obtained by imaging techniques, such as X-ray computed tomography or FIB-SEM,[28–32] associated with specific manufacturing conditions and distinguishing between AM, CBD, and pore phases through appropriate segmentation approaches. Building such a dataset and sharing it with the battery community could be a major help for the development of procedures to consistently validate the electrode microstructures obtained by the models presented here, as well as other computational approaches that start to emerge in the literature.[33–38] When and if such data will become available, it would be of interest to establish automatic procedures to link it to the electrode microstructures generated in silico through the calculator presented here. Nevertheless, today the ARTISTIC models, and the associated online platform, should be seen as useful tools to study qualitatively or semi-quantitively the effect of several manufacturing parameters on the 3D electrode microstructure, which we believe can be of particular interest for a deeper understanding of the main trends linking manufacturing, 3D microstructure, and performance.

The ARTISTIC Online Calculator

The ARTISTIC online calculator is a web application allowing both expert and non-expert users to access our 3D manufacturing models through a user-friendly interface and without the need of owning any computational resources. The concept of this online calculator is schematized in Figure 2. To access it, the users should first register, for free, in the ARTISTIC online calculator,[39] and, once obtained her/his credentials, access to it and select the "Online calculator" section from the menu at the top of the web page.

The online calculator enables an interactive exploration of the LIB electrode manufacturing process by offering its users the possibility to select a number of parameters concerning the slurry, drying, and calendering steps. While moving through these steps, the users can visualize and download the slurry/electrode microstructures generated after the execution of the models in a computer cluster.

The computational infrastructure behind the ARTISTIC online calculator is comprised of two main elements: the access server, in charge of handling the user connections and storing the results, and the MatriCS platform,[40] a computational cluster

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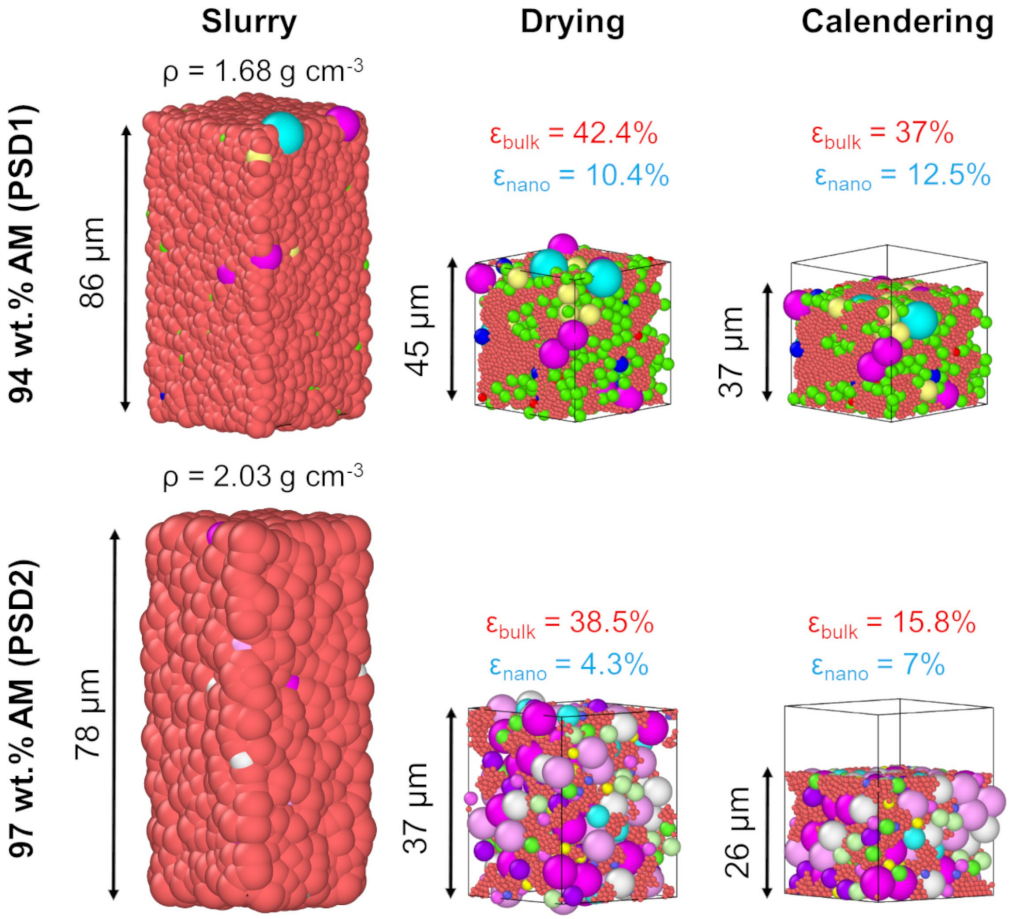

Figure 1. Examples of slurries, dried and calendered electrodes using two arbitrary AM PSDs, named PSD1 and PSD2. Red pale particles stand for CBD, while different colors stand for AM of different sizes (the same color in the top and bottom structures does not necessarily represent the same size). The SC of the top slurry is 58%, and the SC of the bottom one is 64%. The mass loading of the electrodes are 7.6 mg cm^{-2} (top) and 9.9 mg cm^{-2} (bottom). The homogeneous drying was used for both, and the parameters for the calendering are 25% and 35% of compression, 5% and 0% nanoporosity decrease, consideration and no consideration of the elastic recovery for the top and bottom cases, respectively.

located at the Université de Picardie Jules Verne (UPJV), where the ARTISTIC project has dedicated computational nodes.

The user-friendly online interface is one of the crucial components of the ARTISTIC online calculator, since it enables a simple and direct interaction between users and our LIB electrode manufacturing workflow. This interface was designed to guide the users across the three manufacturing processes currently implemented in the calculator. To launch the simulation corresponding to a given process, the users should provide values for its inputs and click the "calculate" bottom. If the associated results are already available, they will be shown immediately. Otherwise, the associated simulation is launched, and the user who launched it receives an email with the links to visualize and recover the resulting electrode/slurry microstructure. The electrode/slurry structure obtained at the end of a manufacturing step is used as input for the next one, together with a new set of manufacturing parameters provided by the user through the interface. A schematic of the overall working principle of the online calculator is depicted in Figure 3 for an arbitrary set of parameters.

The results obtained by all the users are stored in a database together with the corresponding input parameters. This makes possible to avoid launching new simulations when another user requests the same, or similar, simulation in the future.

Furthermore, this data storage approach allows the users of the web application to access all the results previously generated, constituting a collaborative and open access microstructure-manufacturing database.

Lastly, each section (slurry, dying, and calendering) accounts for a comprehensive read-me and a list of available results. The read-me presents the model and the approximations used, explains the meaning of each parameter, offers information on the FFs employed, presents the outputs obtained at the end of the simulation, and gives an estimation of the associated computational cost. The lists of available results allow to have a panorama of the results available and to recover them easily.

Parameters Included in the User Interface

The most important aspect of the ARTISTIC online calculator is that it allows access to a vast manufacturing parameter space

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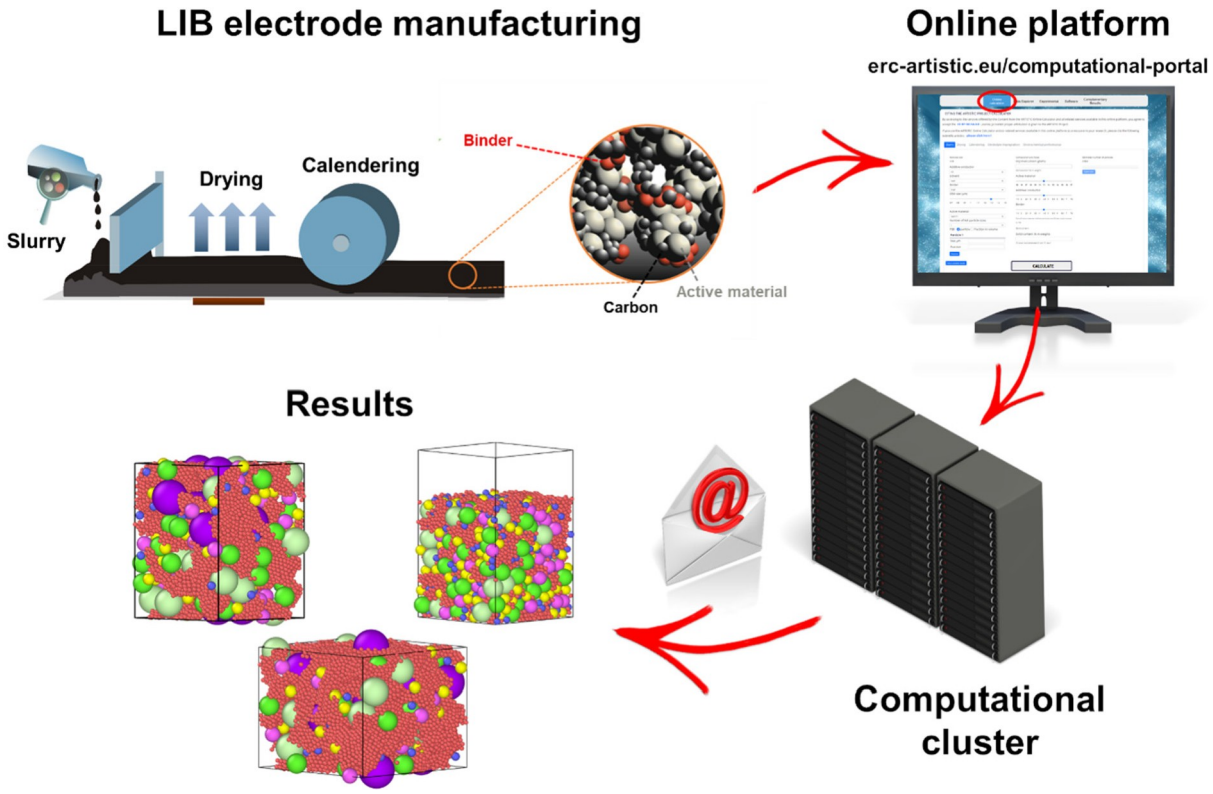

Figure 2. A schematic of the working principle behind the ARTISTIC online calculator, allowing to reproduce LIB electrode manufacturing, from the slurry to the calendered electrode, through a user-friendly web interface. In particular, users can select the manufacturing parameters of interest, as electrode formulation, SC, drying and calendering conditions, which are sent to the computational cluster MatriCS for running the associated simulation. When the simulation ends, the results are stored and reported on the online platform, making them available to anyone, and the user that launched the simulation is informed through an e-mail containing the links for visualizing and recovering the results.

in a user-friendly fashion, enabling any researcher possibly interested into it to investigate the relationships between manufacturing and electrode microstructure. All these parameters have certain pre-defined limits to guide the user, and discretized values to avoid overcharging the calculator with similar simulations. If expert users want to have more precise control on these parameters, they can utilize the associated open-source codes.[27] Table 1 presents all the parameters currently controllable in the calculator.

Up to date, some of the controllable parameters, as the AM, carbon, binder, and solvent chemistry are not real parameters, as they account for one possibility only, and in particular $LiNi_xMn_yCo_2O_2$ (NMC), carbon black (CB), polyvinylidene difluoride (PVdF), and n-methyl-pyrrolidone (NMP), respectively. However, these parameters were defined to include straightforwardly new chemistries into the calculator in the future, such as $LiFePO_4$ (LFP), graphite, and silicon/graphite.[41,42] In addition, the user can control the carbon and binder wt.% separately, but the model accounts for them through the CBD phase, whose wt.% is the sum of the carbon and binder weight percentages. This way the platform parameters are as similar as possible to the experimental ones, which makes them more user-friendly for experimental researchers. Nonetheless, in the future, different ratios between carbon and binder could be accounted for by, for instance, modulating the adhesive forces of the CBD particles.

Contribution to the Community

The main goal of the ARTISTIC online calculator is to allow expert and non-expert users to explore the vast parameter space accessible through our 3D manufacturing models. We expect that this platform can be a useful tool for the battery community, and it was designed with three groups of researchers in mind: i) experimental researchers willing to approach the modeling field, ii) machine learning-based researchers interested in studying LIB electrode manufacturing, and iii) experts in electrochemical simulations willing to use the 3D electrode microstructures obtained through our computational workflow as input for their electrochemical models.

On the one side, i) can benefit from a user-friendly interface that can introduce them to the field of 3D physics-based modeling, and can visualize what happens to the electrode microstructure while it goes through the different manufacturing steps. On the other side, ii) and iii) can build their own customized datasets by simulating and analyzing diverse electrode microstructures without learning how to use our 3D manufacturing models. Alternatively, the dataset constitued of the results already available in the platform can be utilized. Concerning iii), the electrode microstructures obtained by the online calculator can be either analyzed in terms of their averaged properties, as their tortuosity factor,[43] and use these properties as input of homogenized models,[44,45] or they can be

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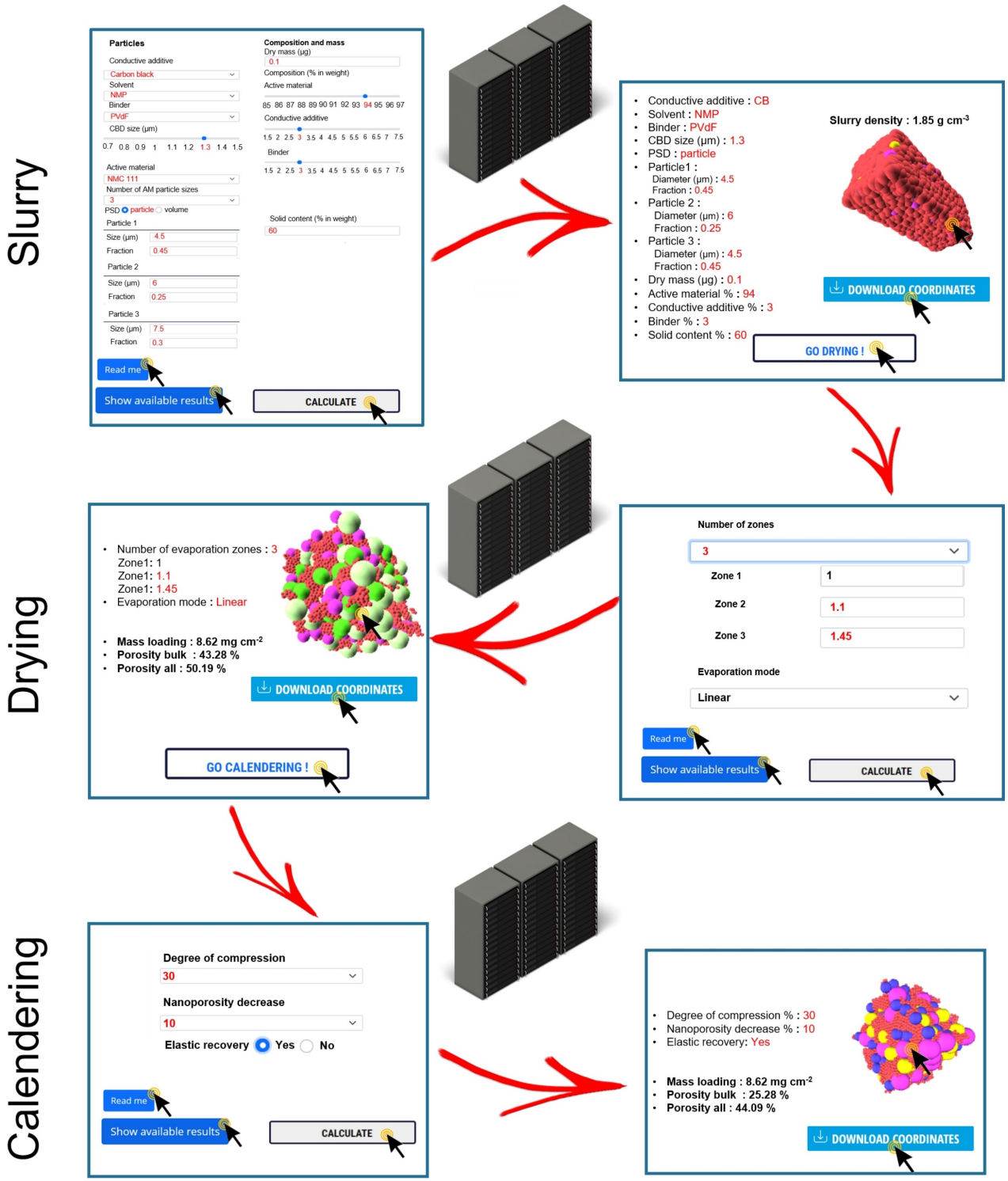

Figure 3. Schematic of the ARTISTIC online calculator from the slurry (top) to the calendered electrode (bottom). Values in red indicate an example. These parameters are sent to the Matrics computational cluster for running the associated simulation. The results are shown in the online platform as soon as the simulation finishes, and the user requesting the simulation receives an email informing her/him about the end of the simulation and how to recover the associated results. The mouse icons indicate clickable regions of interest, as the read-me and the list of available results for each section, or the possibility to move, (de)zoom, and download the 3D slurry/electrode microstructure.

used as direct input for 3D or 4D electrochemical models. For the latter, these structures should be meshed at first, which can be done through our recently released INNOV App,[46] accessible through the ARTISTIC computational portal as well.[39] The only step needed for using the electrode microstructures outputted from the Online calculator into INNOV is describing

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Table 1. List of parameters controllable through the online calculator, their range of values, and meaning. The manufacturing parameters controlled at the slurry step are reported above the solid content (included), while the ones of calendering are reported below the degree of compression (included). Manufacturing parameters of the drying step stand in between. The components' weight ratios are controlled at the slurry phase only because the possible phenomena that can modify the slurry/electrode composition during manufacturing (for instance particles remaining attached to the mixer border) are not accounted for. Therefore, the composition (AM/CBD weight ratios) of the dried and calendered electrodes are identical to the ones selected during the associated slurry step. More information on these parameters can be found in the "read me" reported in each section (slurry, drying, and calendering) of the online calculator.

Parameter	Range of values	Meaning
Active material	NMC	Self-explanatory
Conductive additive	CB	Self-explanatory
Binder	PVdF	Self-explanatory
Solvent	NMP	Self-explanatory
CBD size	0.7–1.5 μm	Size of the CBD particles
CBD nanoporosity	0.3–0.7	Nanoporosity of the CBD phase
Number of AM particle	1–10	Number of AM particles with different sizes
PSD	%particle; %volume	Defines the unit of the AM PSD
Size particle ***i***	2–25 μm	Diameter of the AM particle *i*
Fraction particle ***i***	0–1	Fraction of the AM particle *i*
Electrode mass	0.1–0.2 μg	Mass of the electrode fraction to be simulated
Thickness	Thinner/thicker	Qualitative control of the slurry/electrode thickness
AM [wt.%]	85 %–97 %	Self-explanatory
Conductive additive [wt.%]	1.5 %–7.5 %	Self-explanatory
Binder [wt.%]	1.5 %–7.5 %	Self-explanatory
Solid content	42 %–70 %	Slurry SC
Number of zones	1–3	Defines the drying model to use: 1 homogeneous, > 1 heterogeneous
Zone1	1	Constant
Zone2 Zone3	1–1.8	Defines the DR for the heterogeneous drying model
Evaporation mode	Square cube – cubic	Defines how different DRs translate in terms of solvent removal speed
Degree of compression	5 %–40 %	Percentage of the initial electrode thickness that is compressed during calendering
Nanoporosity decrease	0–100 %	Relative decrease of the CBD nanoporosity
Elastic recovery	yes; no	Self-explanatory

them using the right text format, for which a simple Python code, released in the ARTISTIC Github page,[27] can be utilized.

Overall, we hope that this platform can lead to a deeper investigation of the high-dimensional parameter space offered by our 3D manufacturing models, and to a deeper understanding of the highly convoluted and non-linear links between manufacturing, electrode microstructure, and electrochemical performance. This could unlock a better understanding of the effect of each manufacturing parameter and their combinations on the electrode microstructure and performance, potentially supporting the definition of optimized recipes for targeted electrode properties.

Furthermore, allowing every user to access all the electrode microstructures generated through this platform makes any result obtained verifiable and refutable by the entire community, making their utilization fully transparent.

Finally, we believe that providing access to models, and their associated results, through user-friendly interfaces should become a standard in the battery community. The adoption of this approach would greatly facilitate the scientific collaboration in the battery field, by valorizing models developed by different research groups worldwide, facilitating their integration in other projects, and supporting stronger collaborations between experimental and computational researchers.

Conclusions and Perspectives

In this concept article, we have introduced the ARTISTIC online calculator, an online application that allows all types of researchers to explore LIB electrode manufacturing processes through a user-friendly free interface. This interface allows controlling a vast number of manufacturing parameters, which are used as input of 3D manufacturing models, allowing to obtain the simulated electrode microstructure associated to the chosen manufacturing conditions. In particular, up to date, three main steps of the state-of-the-art LIB electrode manufacturing process can be investigated through this platform: the slurry phase, its drying, and the electrode calendering. The results obtained through this platform are shared among all the users, which allows resulting in a fully transparent, and open-access database built collaboratively.

In practical terms, we hope that the online calculator will become a useful and widely adopted tool for the battery community, and we think that it can be of interest for three kinds of researchers: i) experimental researchers willing to approach the modeling field, ii) machine learning-based researchers interested in studying LIB electrode manufacturing, and iii) experts in electrochemical simulations willing to use the 3D electrode microstructures obtained through our computational workflow as input for their electrochemical models. In addition, the electrode microstructures obtained through this platform can be combined with our recently released meshing

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App INNOV, which enables using them straightforwardly as input for 3D or 4D electrochemical models.

In terms of perspectives, the online calculator will be upgraded to account for the electrolyte filling step and galvanostatic discharge and charge, to offer a complete manufacturing-microstructure-electrochemistry link. In addition, the calculator will allow to simulate different active material chemistries (*e.g.* other NMC-based compounds, $LiFePO_4$, graphite, silicon-graphite). Furthermore, an application programming interface (API) could be implemented to offer a higher degree of freedom to expert users. This API could also ease the collaboration with other research groups and projects worldwide, like the European BIG-MAP project.[47] Furthermore, establishing some connections between our online calculator and ontology initiatives like BattINFO[48] could be interesting in the future, even though ontologies for battery manufacturing aspects are still underdeveloped. The integration in digital twins of the infrastructure and workflows behind our calculator could be also foreseen.[49]

From a scientific perspective, we hope that the battery community will see the ARTISTIC online platform as a tool to explore the vast manufacturing parameter space accessible through our 3D models to establish a deeper understanding of the manufacturing-microstructure-electrochemistry relationships in a collaborative way. Up to date, the platform accounts for 220 registered users, *ca.* 75% of which working in academia and *ca.* 25% in industries, and we hope that this newborn community will grow even faster in the near future.

Computational Detail

The ARTISTIC online calculator disposes of six nodes (384 GB of RAM each), each composed of 2 processors (Intel® Xeon® Gold 6148 CPU @ 2.40 GHz, 20 cores). The slurry and drying simulations utilize one full node, while the calendering simulations utilize half of it. The computational resources dedicated to the ARTISTIC online calculator could be increased as a function of the traffic on the platform. If you do not receive the e-mails sent by the online calculator, please indicate the associated e-mail (erc-artistic@u-picardie.fr) as non-spam. If the problem persists, please contact us.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: Electrode · Li-ion battery · manufacturing · modeling · user-friendly

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