



An Invitation to Engage with Computational Modeling: User-Friendly Tool for In Silico Battery Component Generation and Meshing

Mehdi Chouchane^[a, b] and Alejandro A. Franco^{*[a, b, c, d]}

With the growing interest in battery computational modeling to work hand-in-hand with experiments, the lack of user-friendly software, in particular accessible for experimentalists, is an impediment to its development. In the battery field, time dependent 3D-resolved computational modeling is a more promising approach compared to trial and error to capture the impact of the mesostructure of components (e.g. electrode,

separator) on the electrochemical and transport properties of the cell. In this Concept, we introduce and describe an application, freely accessible and usable through an internet browser, that allows to import, generate and mesh battery components suited for time dependent 3D-resolved electrochemical and transport process simulations.

1. Context

When compared with the experimental community, the battery modeling community is significantly smaller. Indeed, during the last thirty years less than 20% of published articles in the battery field were related to modeling (query on the 19th of April, 2021, on Web of Science with the keywords "battery" and "battery modeling"). This trend has been persistent despite improvements and promising computational modeling achievements in recent years (e.g. higher resolution, coupled electrochemical-mechanical model).^[1–5] Computational models can be very useful, especially time dependent ones resolving processes in three Cartesian spatial dimensions (3D), for assisting in the design of components (e.g. electrodes) thanks to their capabilities to assess heterogeneities upon battery cell operation.^[6,7] However, there are numerous obstacles to the development of the modeling field, and most of them are related to the need of expertise outside of the electrochemist's conventional skillset. Firstly, the acquisition of a component (e.g. electrode, separator) mesostructure spatially resolved in 3D can be challenging if access to a tomogram is not available. The mesostructure will have to be computationally generated,

typically using a stochastic algorithm.^[8–10] "Stochastic" means here that the electrode generation will be performed through a random process that can be more or less controlled. For instance, spherical particles can be placed randomly in a given volume representing the electrode, and these random positions can be chosen in a way to reach a given surface area between the particles and the electrolyte or a given amount of overlap between the particles. It means that launching the algorithm several times with the same input parameters will result in different outputs.

Then, a mesh that connects the component mesostructure to the computational model will be needed before proceeding with the numerical simulation through the solving of partial differential equations describing electrochemical and transport processes. Meshing is the way to divide the component mesostructure into finite elements (usually tetrahedrons) without deforming the original geometry. This step might be the most obscure to unexperienced users but must be well understood to avoid introducing errors resulting from complications in the computational representation of the mesostructure used in battery cell simulations. In a first effort to ease this step, we reported in 2019 an efficient meshing algorithm called "INdefinite Number Of phases meshing through Voxelization" (INNOV) capable of importing both the active material (AM), pores and the carbon and binder domain (CBD) present in Lithium Ion Battery (LIB) electrodes into computational modeling software.^[11] Since then, this algorithm has been deeply improved with several new features that have been implemented into a graphic user interface (GUI) to make it more accessible, which are presented in the present Concept. GUIs allow users to control the software with interactive elements (e.g. push button, slider, etc.) rather than editing the code directly, which significantly reduces the required expertise.

With INNOV, the objective is to enable more participation to the battery modeling field thanks to a free and user-friendly tool that significantly accelerates the computational modeling

[a] M. Chouchane, Prof. Dr. A. A. Franco
Laboratoire de Réactivité et Chimie des Solides (LRCS),
UMR CNRS 7314, Université de Picardie Jules Verne,
Hub de l'Energie, 15 rue Baudelocque, 80039 Amiens Cedex, France
E-mail: alejandro.franco@u-picardie.fr

[b] M. Chouchane, Prof. Dr. A. A. Franco
Réseau sur le Stockage Electrochimique de l'Energie (RS2E),
FR CNRS 3459, Hub de l'Energie, 15 rue Baudelocque, 80039 Amiens Cedex, France

[c] Prof. Dr. A. A. Franco
ALISTORE-European Research Institute,
FR CNRS 3104, Hub de l'Energie, 15 rue Baudelocque, 80039 Amiens Cedex, France

[d] Prof. Dr. A. A. Franco
Institut Universitaire de France, 103 boulevard Saint Michel, 75005 Paris, France

 Supporting information for this article is available on the WWW under
<https://doi.org/10.1002/batt.202100096>

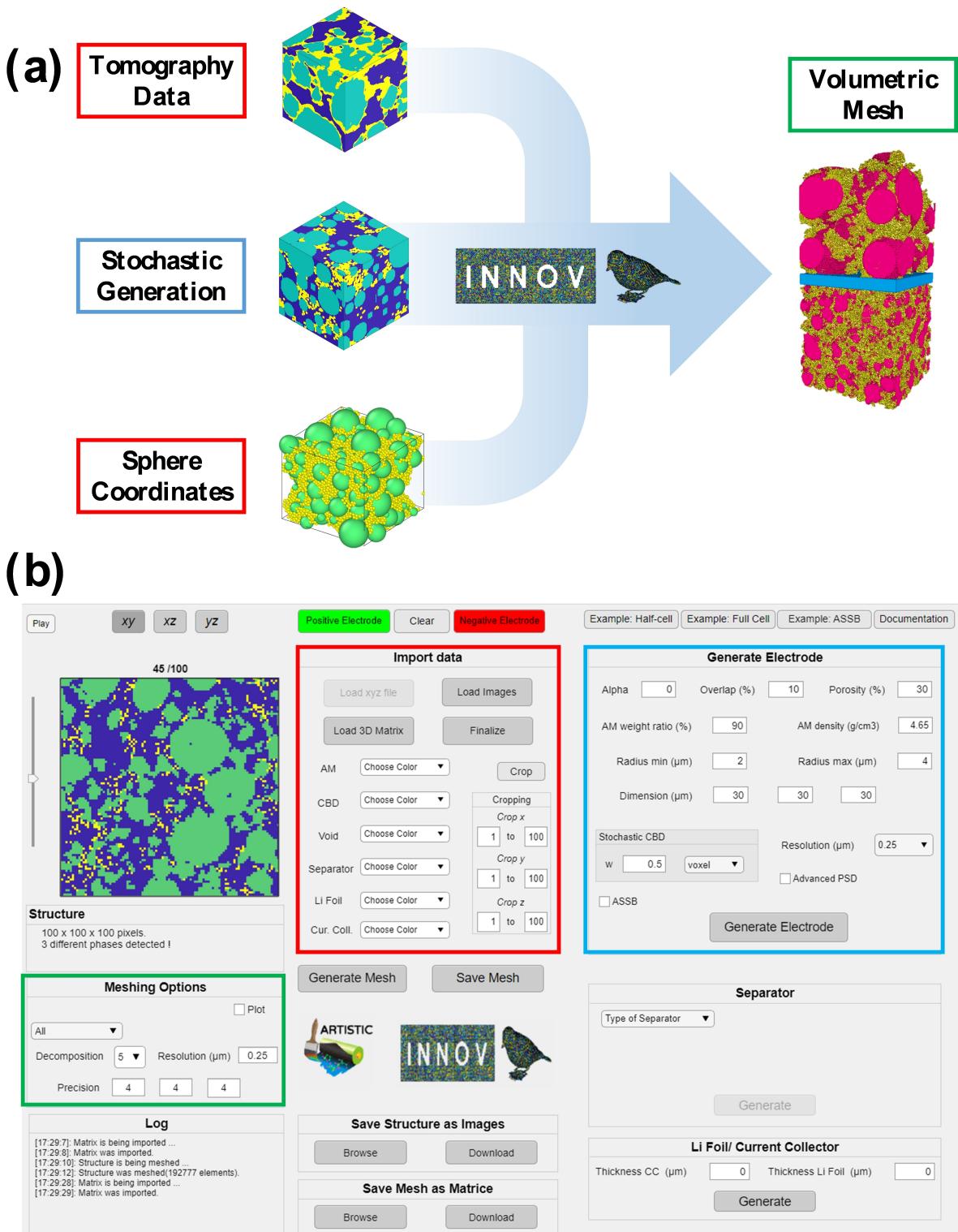


Figure 1. a) Workflow of INNOV with the different options to import or generate 3D structures to perform the mesh. b) Graphic user interface of INNOV with the highlighted panels related to the importation (red) or generation of electrode (blue) and meshing (green).

process involving 3D-resolved components. It is noteworthy that very recently, efforts in a similar direction start to emerge ("Matbox").^[12] Still, such efforts are scarce due to the complexity

for scientists unfamiliar with both programming and modeling to undertake such initiatives.

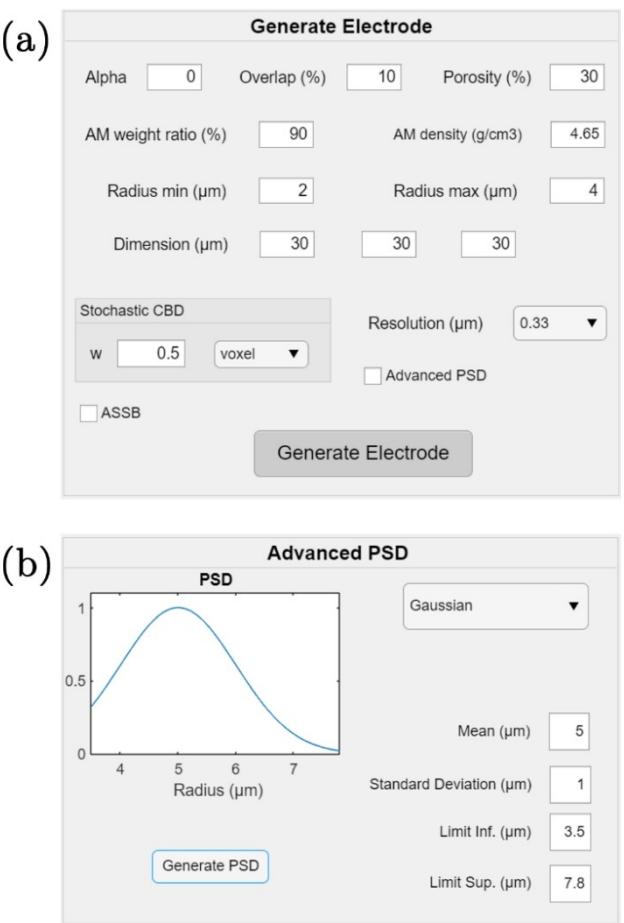


Figure 2. Panels in INNOV GUI for a) the generation of the electrode and b) the advanced particle size distribution settings.

2. INNOV's Purpose

INNOV is a Web App freely accessible through our ARTISTIC project website^[13] that offers a user-friendly tool for both new and advanced modeling researchers. All the computation will be performed in the project servers, which means that only a proper internet connection will be required from the user.

The application embeds options to import or to generate the geometry of the investigated component or system as highlighted in Figure 1a and with the GUI in Figure 1b. In the battery field, the system can be composed of positive and negative electrodes, a separator and current collectors. Nonetheless, INNOV can be used in a variety of scientific fields thanks to its meshing tool that allows importing the generated or meshed geometry into modeling software to solve mathematical partial differential equations describing electrochemical and transport properties, and perform simulations. Initially, INNOV was designed to ease the importation of volumetric multiphase meshes into COMSOL Multiphysics® (each phase being a different material or porous media) by its native format and embedded domain selections but can now be used for any computational modeling software thanks to several options to export the mesh. It provides a tool to support the current

modeling field and to encourage a wider audience to couple their experimental investigations with computer simulations. A user guide is also accessible through the GUI to access more technical information related to the algorithms and the role of each parameter.

3. 3D Structures

Several options exist to obtain 3D structures in INNOV, the associated data being either imported or stochastically generated.

For the importation, the user can have the data as a stack of images, as a matrix with the native MATLAB format, or, for the case of electrodes for instance, as a list of the coordinates and radii of AM and CBD spheres, with the CBD having a constant radius. For the latter, an additional slicing step will be required (see Supporting Information).

For the stochastic generation of electrodes, a built-in tool allows tailoring the 3D structures to the need of the users' thanks to a variety of settings. The workflow of the stochastic generation in INNOV takes place in the "Generate Electrode" panel in Figure 2a and will be described in the following paragraph. A flowchart of the workflow is available in the Supporting Information.

The first step is to define the desired size of the electrode through the "Dimension" parameter (see Figure 2a). Then, the AM particles are considered as spheres and are added to the system with conditions based on the distance between the boundaries of the box and the centers of the spheres (parameter α , see Supporting Information) and based on the overall AM-AM overlap. Regarding the particle size distribution (PSD), the user can either use a uniform distribution between two limit values or used advanced PSD. Advanced PSD allows the user to either import his/her PSD as text file with the list of radii or with the radii and the associated probability, to either create a Gaussian distribution, or to either use built-in PSD for LiNi_{1/3}Mn_{1/3}Co_{1/3}O₂ (NMC-111) and spherical Graphite. For particles that are intersected by the boundaries of the box, periodic boundary conditions are applied to have a more realistic representative volume. Then the CBD phase is added to the structure with a morphology controlled by the parameter w (see Supporting Information) that varies between 0 (film-like CBD) and 1 (cluster-like CBD) as highlighted in Figure 3a,b. An option to generate an additional "void" phase, particularly relevant for all-solid-state batteries, is also available.

The strength of this tool resides in the simplicity to generate stochastic battery component mesostructures while allowing to finely tune the geometrical features of the electrodes. For instance, to capture the structural changes upon a calendering process, with the AM particles being brought closer to each other, one can increase the AM-AM overlap and decrease the porosity to have a more packed structure (see Figure 3c,d).

Finally, users can add a separator that can be homogeneous, based on tomography data (e.g. Celgard® or Targray®^[14,15]), stochastically generated with the same algorithm

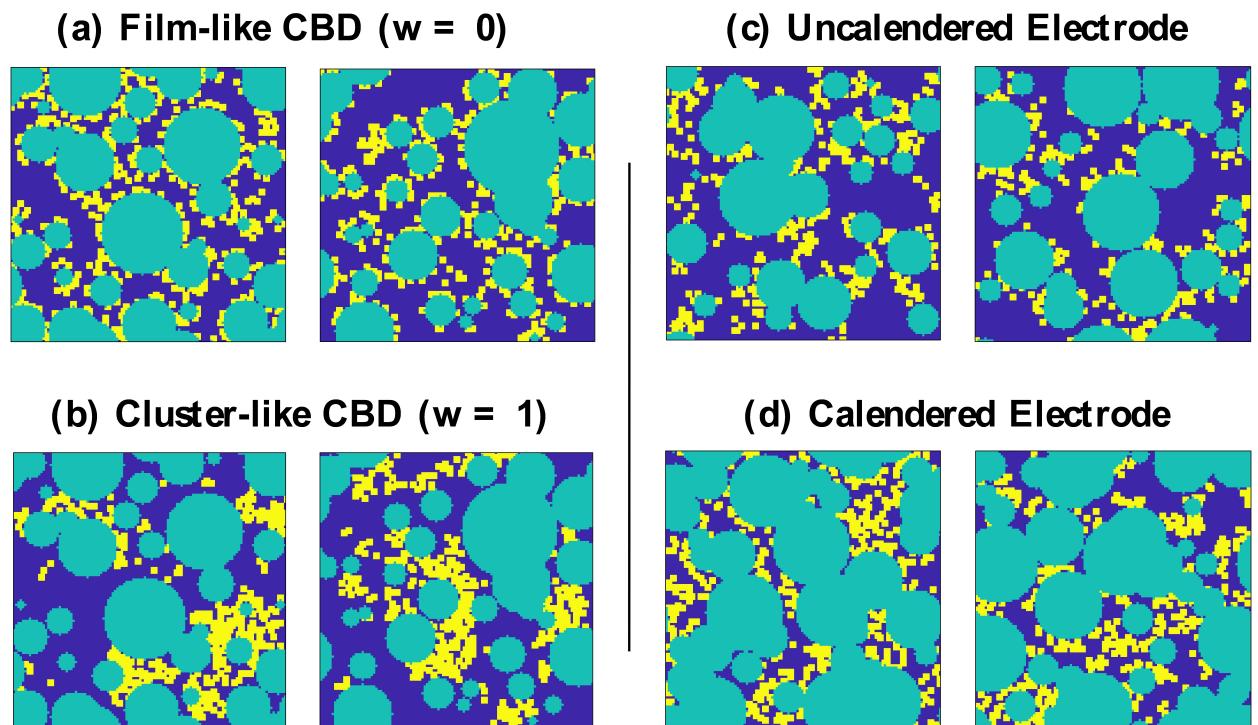


Figure 3. Slice view available in INNOV GUI of stochastically generated NMC electrodes with in green the AM, in yellow the CBD, and in purple the electrolyte. Comparison between a) film-like and b) cluster-like morphology for the CBD and between c) uncalendered and 750*.8 – d) calendered electrodes.

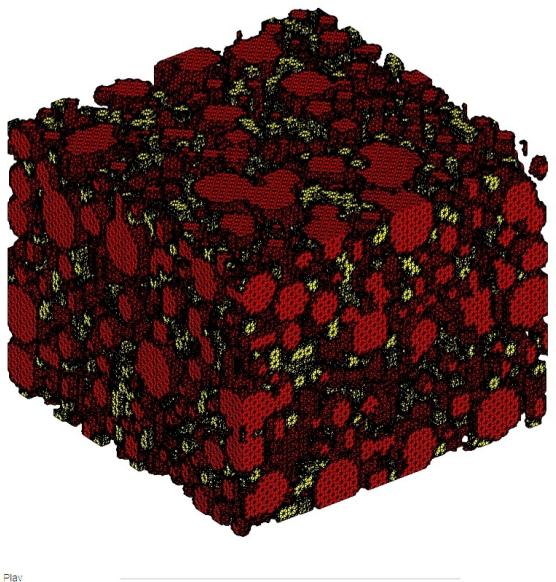


Figure 4. 3D rendering of the mesh of a LIB positive electrode in INNOV GUI with the NMC in red and the CBD in yellow.

used for the CBD generation, or as filaments (see Supporting Information). For models that use explicit current collectors or Li foil as counter-electrode (half-cell configuration), i.e. 3D resolved, homogeneous ones can be added to the final structure. The whole structure can be saved as a .tif file to be

used for analyses in other software (e.g. TauFactor^[16] or GeoDict^[17] to calculate tortuosity factors).

4. Meshing

The meshing algorithm used in INNOV is based on the voxelization approach published by us in a previous article.^[11] This algorithm enables to have an indefinite number of domains in the mesh with elements finer at the interface between materials and coarser in the bulk. The number of elements in the mesh can be controlled through the “Precision” parameter and the output can be visualized in 3D (see Figure 4). The mesh 3D rendering as well as several other functions were reused from Iso2 mesh.^[18] The mesh can then be exported in a variety of formats depending on the users’ needs. The mesh can be exported as a surface mesh (.ply or .stl) or as a volume (.bdf, .mphtxt, .mat). INNOV facilitates the importation of the mesh to COMSOL Multiphysics® by using its native format .mphtxt into which built-in selections are embedded. However, the use is not restricted to COMSOL Multiphysics® since the .bdf (Nastran) format is accepted by most software and the .mat format allows the user to convert it to its desired format.

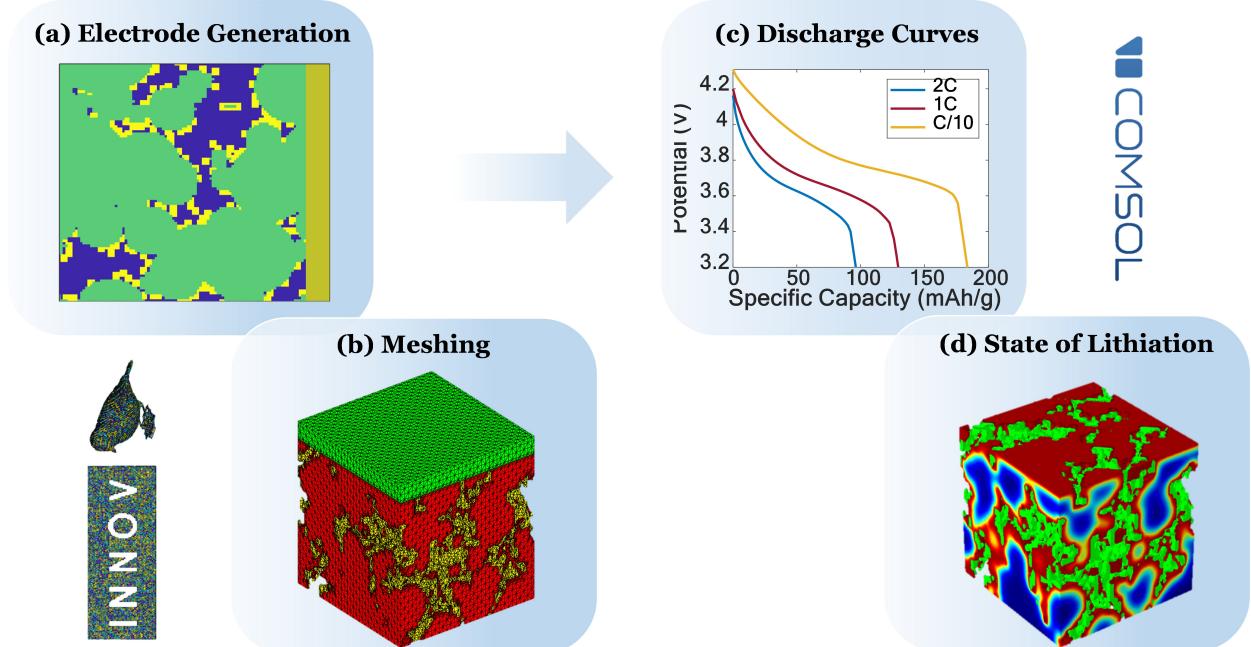


Figure 5. Complete workflow of the NMC half-cell example in INNOV from the a) electrode generation and b) meshing, to the electrochemical simulations in COMSOL Multiphysics® with c) the discharge curves and d) the state of lithiation at the end of the discharge at 2 C with the CBD in green.

5. Application Examples

To explore all the possibilities offered by this Web App, built-in examples are available to allow users to better understand the workflow of INNOV. Upon a single push of a button, the electrodes will be stochastically generated based on fixed parameters and meshed for the users to visualize the 3D rendering. This section focuses on the description of the three built-in examples available.

In the first example, the focus is on the positive electrode so a half-cell configuration with an implicit Li foil (i.e. no thickness considered) as negative electrode was chosen. To simulate a calendered electrode, a porosity of 30% was set with 20% (in volume) of overlap between AM particles. A volume of $35 \times 35 \times 35 \mu\text{m}^3$ was selected with a realistic AM weight percentage (94%) and an experimental PSD for NMC-111. CBD was added as a film covering the NMC particles. To finalize the structure, a 5 μm thick homogeneous separator was added. The pores in the separator were not resolved since the impact of the separator is out of the scope of this example. A slice view of the generated structure and a 3D rendering of the output mesh are shown in Figures 5a and 5b, respectively. The precision of the mesh was slightly refined to reach a mesh composed of ca. 650 000 tetrahedrons. The precision should always be adapted to the size of the system, the characteristic length of the system, and to the computational power available to perform the electrochemistry/transport simulations afterwards. Then the mesh was imported into COMSOL Multiphysics® to perform the electrochemical simulations. Using a half-cell model similar to those already reported by our group,^[6,19,20] three different discharges were simulated at C/10, 1C, and 2C. The discharge curves for each case are represented

in Figure 5c displaying a higher specific capacity at lower discharge rates. The profile of lithiation at the end of discharge at 2C is also reported in Figure 5d where the solid diffusion can be identified as the limiting phenomenon because of the steep Li concentration gradient in the NMC.

A second built-in example provided with the software generates a full cell battery with a uniform PSD for the positive electrode's particles, a homogeneous separator and a custom Gaussian PSD for the AM at the negative electrode. This example illustrates two important features of the App, namely the full cell generation and the use of a custom Gaussian distribution which is more realistic than a uniform distribution.

The last built-in example sheds light on the possibility with INNOV to generate structures suited for all-solid-state batteries modeling. It consists of a generated half-cell with NMC, CBD, solid electrolyte and 10% of void. Then 20 μm in thickness of solid electrolyte is automatically added to act as the separator with 15% of void. With the improvement of simulations accuracy, modeling should assist in the development of this promising new chemistry that all-solid state is.

Thanks to these provided examples, users can understand the overall procedure behind a component structure generation and then interact with the variety of parameters available to study afterwards the desired phenomena in half-cell or full cell computer simulations.

6. Concluding Remarks

INNOV App offers a user-friendly alternative to expensive commercial software. The App allows designing electrodes that can be used to investigate the impact on the battery perform-

ance of a multitude of mesostructural parameters. INNOV provides to both new and experienced modeling scientists the resources to undertake state-of-the-art battery modeling studies. This tool spares the users the considerations related to structure generation and meshing algorithms that can be discouraging and an obstacle to the development of the battery modeling community.

As soon as this Concept will become published, INNOV will be freely accessible through the Computational Portal in the ARTISTIC project webpage. To use it, the interested person needs to register in the ARTISTIC Computational Platform with her/his e-mail address and will receive a password. To extend the App capabilities, several new features such as the generation of non-spherical particles, CBD with PSD, or adding a separate app to perform basic image segmentation will be considered in the future.

To conclude, INNOV gathers expertise developed for several years within our group for a wide battery field audience to raise the interest in 3D-resolved battery computational modeling.

Acknowledgements

The authors acknowledge the European Union's Horizon 2020 research and innovation program for funding support through the European Research Council (grant agreement 772873, "ARTISTIC" project). A.A.F. acknowledges the Institut Universitaire de France for the support.

Conflict of Interest

The authors declare no conflict of interest.

Keywords: 3D modeling • battery • carbon binder domain • mesh • structure generation

- [1] A. Gayon-Lombardo, L. Mosser, N. P. Brandon, S. J. Cooper, *npj Comput. Mater.* **2020**, *6*, 1–11.
- [2] M. E. Ferraro, B. L. Trembacki, V. E. Brunini, D. R. Noble, S. A. Roberts, *J. Electrochem. Soc.* **2020**, *167*, 013543.
- [3] X. Lu, A. Bertei, D. P. Finegan, C. Tan, S. R. Daemi, J. S. Weaving, K. B. O. Regan, T. M. M. Heenan, G. Hinds, E. Kendrick, D. J. L. Brett, P. R. Shearing, *Nat. Commun.* **2020**, *11*, 1–13.
- [4] A. A. Franco, A. Rucci, D. Brandell, C. Frayret, M. Gaberscek, P. Jankowski, P. Johansson, *Chem. Rev.* **2019**, *119*, 4569–4627.
- [5] T. Danner, M. Singh, S. Hein, J. Kaiser, H. Hahn, A. Latz, *J. Power Sources* **2016**, *334*, 191–201.
- [6] M. Chouchane, A. Rucci, T. Lombardo, A. C. Ngandjong, A. A. Franco, *J. Power Sources* **2019**, *444*, 227285.
- [7] X. Lu, S. R. Daemi, A. Bertei, M. D. R. Kok, K. B. O'Regan, L. Rasha, J. Park, G. Hinds, E. Kendrick, D. J. L. Brett, P. R. Shearing, *Joule* **2020**, *4*, 2746–2768.
- [8] A. N. Mistry, K. Smith, P. P. Mukherjee, *J. Phys. Chem. Lett.* **2018**, *10*, 6317–6326.
- [9] M. Duquesnoy, T. Lombardo, M. Chouchane, E. N. Primo, A. A. Franco, *J. Power Sources* **2020**, *480*, 229103.
- [10] J. Joos, A. Buchele, A. Schmidt, A. Weber, E. Ivers-Tiffée, *Energy Technol.* **2021**, n/a 2000891.
- [11] M. Chouchane, A. Rucci, A. A. Franco, *ACS Omega* **2019**, *4*, 11141–11144.
- [12] 2021, https://github.com/NREL/MATBOX_Microstructure_analysis_toolbox.
- [13] <https://www.u-picardie.fr/erc-artistic/computational-portal/>.
- [14] M. F. Lagadec **2016**, <https://doi.org/10.5905/ethz-1007-32>.
- [15] M. F. Lagadec **2018**, <https://doi.org/10.3929/ethz-b-000265085>.
- [16] S. J. Cooper, A. Bertei, P. R. Shearing, J. A. Kilner, N. P. Brandon, *Software X* **2016**, *5*, 203–210.
- [17] <https://www.math2market.com/>.
- [18] A. P. Tran, S. Yan, Q. Fang, *NPh* **2020**, *7*, 015008.
- [19] M. Chouchane, E. N. Primo, A. A. Franco, *J. Phys. Chem. Lett.* **2020**, *11*, 2775–2780.
- [20] A. C. Ngandjong, T. Lombardo, E. N. Primo, M. Chouchane, A. Shodiev, O. Arcelus, A. A. Franco, *J. Power Sources* **2021**, *485*, 229320.

Manuscript received: April 29, 2021

Revised manuscript received: June 2, 2021

Accepted manuscript online: June 2, 2021

Version of record online: June 17, 2021