



HAL
open science

NEOLAB: A Scilab tool to simulate the Negative Electrode of Lead-Acid Batteries

Mikael Cugnet, Florian Gallois, Angel Kirchev, Denys Dutykh

► To cite this version:

Mikael Cugnet, Florian Gallois, Angel Kirchev, Denys Dutykh. NEOLAB: A Scilab tool to simulate the Negative Electrode of Lead-Acid Batteries. *SoftwareX*, 2023, 22, pp.101394. 10.1016/j.softx.2023.101394 . cea-04119659

HAL Id: cea-04119659

<https://cea.hal.science/cea-04119659>

Submitted on 6 Jun 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Original software publication

NEOLAB: A Scilab tool to simulate the Negative Electrode of Lead-Acid Batteries

Mikaël Cugnet^{a,*}, Florian Gallois^{a,c,d}, Angel Kirchev^a, Denys Dutykh^{b,d}^a Univ. Grenoble Alpes, CEA, Liten, Campus Ines, F-73375 Le Bourget-du-Lac, France^b Mathematics Department, Khalifa Univ. of Science and Technology, PO Box 127788 Abu Dhabi, United Arab Emirates^c Haulotte, R&D, F-42420 Lorette, France^d Univ. Grenoble Alpes, Univ. Savoie Mont Blanc, CNRS, LAMA, F-73375 Le Bourget-du-Lac, France

ARTICLE INFO

Article history:

Received 12 April 2021

Received in revised form 5 January 2023

Accepted 20 April 2023

Dataset link: <https://github.com/FlorianGallois/NEOLAB>

Keywords:

Lead-acid battery

Electrode

Scilab

Mathematical model

Nondimensionalization

ABSTRACT

Responding to the need of a growing community of students and researchers who want to get involved in the field of electrochemical storage systems, NEOLAB offers a new tool dedicated to a modeling domain where almost no open-source solutions exist. Physics-based models of batteries require extensive knowledge in thermodynamics, electro-chemistry, mathematics, material and computer sciences. Based on the idea that a minimum working example is the best way to learn gradually how to model a battery, NEOLAB provides a solution to simulate the behavior of the negative electrode of lead-acid batteries and a framework to investigate other primary and secondary technologies.

© 2023 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Code metadata

Current code version	1.0
Permanent link to code/repository used for this code version	https://github.com/ElsevierSoftwareX/SOFTX-D-21-00064
Code Ocean compute capsule	
Legal Code License	GNU Lesser General Public License v2.1
Code versioning system used	git
Software code languages, tools, and services used	Scilab 6.1.0
Compilation requirements, operating environments & dependencies	Windows, Linux, and Mac OS X
If available Link to developer documentation/manual	https://github.com/FlorianGallois/NEOLAB/blob/main/README.md
Support email for questions	mikael.cugnet@cea.fr

Software metadata

Current software version	1.0
Permanent link to executables of this version	https://github.com/FlorianGallois/NEOLAB
Legal Software License	GNU Lesser General Public License v2.1
Computing platforms/Operating Systems	Windows, Linux, and Mac OS X
Installation requirements & dependencies	Scilab 6.1.0 or earlier
If available, link to user manual – if formally published include a reference to the publication in the reference list	https://github.com/FlorianGallois/NEOLAB/blob/main/README.md
Support email for questions	mikael.cugnet@cea.fr

* Correspondence to: 50 Avenue, du Lac Léman, 73375 Le Bourget-du-Lac, France.

E-mail address: mikael.cugnet@cea.fr (Mikaël Cugnet).

<https://doi.org/10.1016/j.softx.2023.101394>

2352-7110/© 2023 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Motivation and significance

Energy storage is definitely a major ambition of our modern society to stay on track with the Sustainable Development Scenario pathway [1]. Electro-chemical storage solutions are necessary to accommodate a larger share of renewable energy sources

into our electricity grid in the short term. Batteries are indeed considered as one of the most important and efficient ways of stabilizing electricity networks. Many battery chemistries have been developed over the past century. Lead-acid batteries represent the second largest installed capacity in the rechargeable battery market, behind lithium-ion. Recent lead batteries exhibit long cycle and calendar lifetimes and are by far the most efficiently recycled commodity, with over 99% of lead batteries being collected and recycled in Europe and USA [2].

Mathematical modeling of battery porous electrodes has a history of half a century [3]. Making predictions, interpolate and extrapolate data points, spare you considerable effort, time, and money, as long as you select the modeling approach that suits user needs [4]. From quantum chemical computation of material properties, through the continuum performance models of electrodes, all the way up to the life cycle analysis of a complete battery solution, there is a wide range of models for all spatial and temporal scales [5]. To understand the phenomena limiting battery performances, physics-based models have been continuously improved [6–10].

The authors believe that in the twenty-first century, commercial solutions providing battery models will sooner or later be replaced by open-source ones, accessible not only by people who can afford them, but also constantly improved by a growing worldwide community who cannot pay for them, but still invest its time and intelligence, as evidenced by Python success stories [11]. More and more students and researchers are willing to get their hands on battery modeling in some universities where commercial solutions are not always easily accessible. We believe that SoftwareX open-access papers can give them the opportunity to play their role in battery innovation [12]. Since there is not yet any battery model available in SoftwareX, we want to bring our contribution to the community with a code developed on Scilab[®], which is the free alternative to Matlab[®] since 1980 [13]. This project started over a decade ago, while developing from scratch a lead-acid battery model that ultimately ended up in the *Batteries and Fuel Cells Module* of COMSOL Multiphysics[®] 4.2a [14].

2. Software description

The idea behind NEOLAB is to provide a simple tool able to simulate the behavior of the negative electrode of a lead-acid battery. It is actually a code that anyone can use and modify to adapt it to any kind of electrode chemistry. The model is based on a minimal set of ordinary and partial differential equations describing the physics behind the lead electrode used as the negative plate in lead-acid batteries.

2.1. Software architecture

NEOLAB runs with the help of only two Scilab[®] routines. The main routine is called `main_dassl_*.sce`, the star symbol being either replaced by `dim` or `adim`, depending on whether you want to work with a dimensional or dimensionless set of equations. The second routine called `res_dassl_*.sci` is the function computing the residue of the differential algebraic equations (DAEs).

The main routine is composed of a header where the user must specify the path to the residue function and the experimental data to be simulated by the model. The experimental data set is optional. It can contain the model inputs for simulation purpose (current and temperature for instance) or outputs for validation purposes (voltage or power for instance). The model initialization requires a quite long list of constant parameters (more than 30 for a single electrode). There are also a dozen of physical

relations characterizing parameters which evolve in time with the electrode state (see Supplementary Material). The choice of implementing these parameters in the form of analytical expressions instead of data tables is dictated by code compactness and computation time, the latter being significantly shorter when you do not have to interpolate the data for each time step. Most of the thermodynamic data come from the seminal work of Hans Bode [15]. To illustrate the accuracy of our analytical functions compared to the experimental data available, we present some of the newly developed expressions of the relative molar Gibbs free energies of water (Fig. 1a)

$$\bar{G}_{\text{H}_2\text{O}} = \Delta G_{\text{H}_2\text{O}}^0 - m_{\text{A}} (16.9 m_{\text{A}} + 90.7) \quad (1)$$

and sulfuric acid (Fig. 1b)

$$\bar{G}_{\text{H}_2\text{SO}_4} = \Delta G_{\text{H}_2\text{SO}_4}^0 + 12\,900 \sqrt{m_{\text{A}}} - 79\,200, \quad (2)$$

as well as the partial molar entropies of water (Fig. 1c)

$$\bar{S}_{\text{H}_2\text{O}} = 69.3 + 1.22 \exp \left[-\frac{(m_{\text{A}} - 3.13)^2}{13.1} \right] \quad (3)$$

and sulfuric acid (Fig. 1d)

$$\bar{S}_{\text{H}_2\text{SO}_4} = 139 \exp \left\{ -0.374 \left[\frac{\sqrt{\exp(1.41 - 0.828 m_{\text{A}})}}{1 + \sqrt{\exp(1.41 - 0.828 m_{\text{A}})}} \right] - 0.315 \exp(1.41 - 0.828 m_{\text{A}}) \right\}, \quad (4)$$

with respect to the molality of sulfuric acid in the electrolyte soaking the electrode. We can clearly see that these quite simple analytical relations actually provide a good fit of the experimental data available in the literature.

There are three DAEs solved in the `res_dassl_*.sci` routine, representing respectively charge, volume, and species conservation. Three unknown variables are involved in the equations, varying over time and space: the interfacial voltage, electrode porosity, and acid concentration (see Supplementary Material). By interfacial voltage, we mean the difference between the negative electrode potential and a reference electrode with constant electrochemical potential. The model is developed by discretizing the space (electrode thickness), and then by solving the equations as a function of time, for each space discretization step.

The initial values of the interfacial voltage and electrode porosity are computed based on the model assumptions concerning the electrolyte molality at the beginning of the simulation for the former and some other assumptions concerning the electrode design for the latter. The electrode consists of a current collector equipped with a meshgrid, through which the active material is dispersed (Fig. 2a). The separator, made of polyethylene and a layer of fiberglass, is placed against the electrode. This geometry being complex, the current collector and active material are considered as two distinct layers in the model (Fig. 2b). Furthermore, only a single spatial dimension, normal to the electrode plate, is taken into account. The meshing is quite simple with a number of nodes equally spaced to be set by the user (Fig. 2c).

The simulation part of the main routine gives the possibility to discharge the electrode in galvanostatic conditions (constant current) and let it rest for a given period of time. The possibility of discharging the electrode and leaving it to relax for some time is an example of what is possible to do with the equations, but there is absolutely no limit to the simulation possibilities. The user may want to move from galvanostatic charging or discharging conditions, defined as Neumann boundary conditions setting the current density, to potentiostatic conditions, defined as a Dirichlet boundary condition setting the voltage [16]. In current-controlled

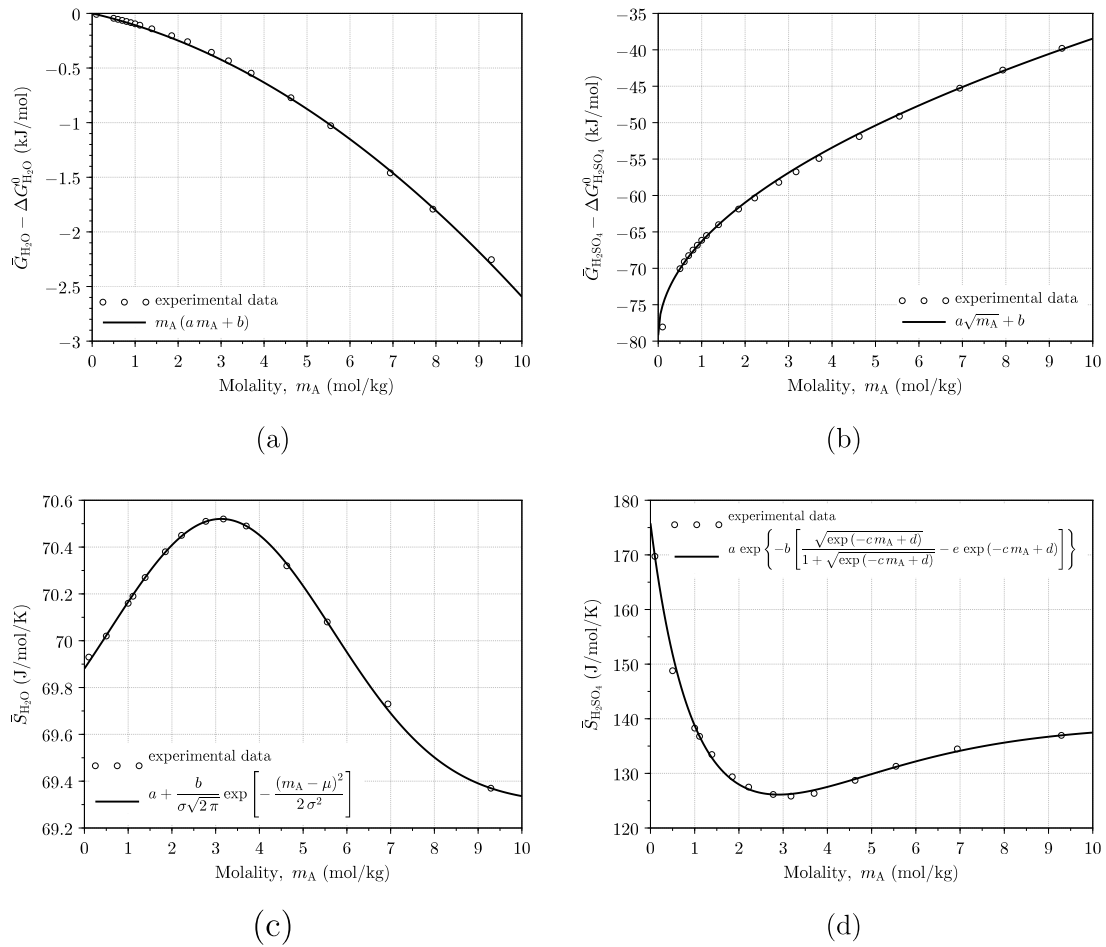


Fig. 1. Experimental data (circles) and analytical expressions (curves) of relative molar Gibbs free energies from [15, p. 57] (a, b) and partial molar entropies from [15, p. 65] (c, d) of water (a, c) and sulfuric acid (b, d) with respect to molality. In each legend appears the analytical expression used to draw the black curve passing through all data points.

mode, the current I going through the electrode is the model input, which implies the following boundary conditions:

$$\mathbf{n} \cdot \mathbf{i}|_{x=0} = \mathbf{n} \cdot \left\{ -\sigma_1 \left[-\nabla(\Delta\phi) + \frac{1}{F} \left(t_{\text{H}^+}^0 - \frac{1}{2} \right) \nabla\mu_A \right] \right\} \Big|_{x=0} = 0, \quad (5)$$

$$\mathbf{n} \cdot \mathbf{i}|_{x=L} = \mathbf{n} \cdot \left\{ -\sigma_1 \left[-\nabla(\Delta\phi) + \frac{1}{F} \left(t_{\text{H}^+}^0 - \frac{1}{2} \right) \nabla\mu_A \right] \right\} \Big|_{x=L} = -\frac{I}{S}, \quad (6)$$

at the current collector and separator interfaces, respectively, while the electrode voltage U is the model output obtained at the current collector interface:

$$U = \Delta\phi|_{x=0}. \quad (7)$$

In voltage-controlled mode, Eq. (7) acts as a Dirichlet boundary condition applied to the same interface, while the current is computed at the separator interface:

$$I = S \sigma_1 \left[-\nabla(\Delta\phi) + \frac{1}{F} \left(t_{\text{H}^+}^0 - \frac{1}{2} \right) \nabla\mu_A \right] \Big|_{x=L}. \quad (8)$$

2.2. Software functionalities

NEOLAB offers the possibility to study the impact of discharge rate on the voltage curve through a robust and optimized model. For instance, the solver used and its tolerance parameters have

been meticulously selected based on their performance (execution time and reliability of the solution). Two standard Scilab solvers are applicable for our model: DAE (implicit method) and ODE (explicit method). Because of the stiffness of the problem, DASSL (DAE sub-solver) and STIFF (ODE sub-solver) offer the best compromise accuracy/speed. Both of them approximate the derivative using the BDF (Backward differentiation formula), but differ on the computation of the Jacobian matrix [17]. Applying the quasi-Newton's method, DAE and specially DASSL are much faster than ODE and STIFF for a similar solution accuracy (Fig. 3a). Then, optimal absolute and relative error tolerances of solution have been determined so as to minimize the error while keeping a reasonable execution time. A preparatory study revealed that setting the absolute tolerance ten times lower than the relative tolerance was a good practice. Because we do not have access to the exact solution of our problem, we estimate the error we make on each of the model variables based on their values obtained with a reference tolerance of 10^{-11} . However, a relative tolerance of 10^{-5} provides a voltage accuracy below the millivolt, which is acceptable enough for this model (Fig. 3b).

A rigorous study has been conducted to determine the impact of the equations non-dimensionalization, which is a mathematical method particularly adapted to complex equations such as partial differential equations, having the capacity in some cases to improve the performance of the model [see Supplementary material]. However, the use of dimensionless expressions does not allow to reduce the execution time (Fig. 3c).

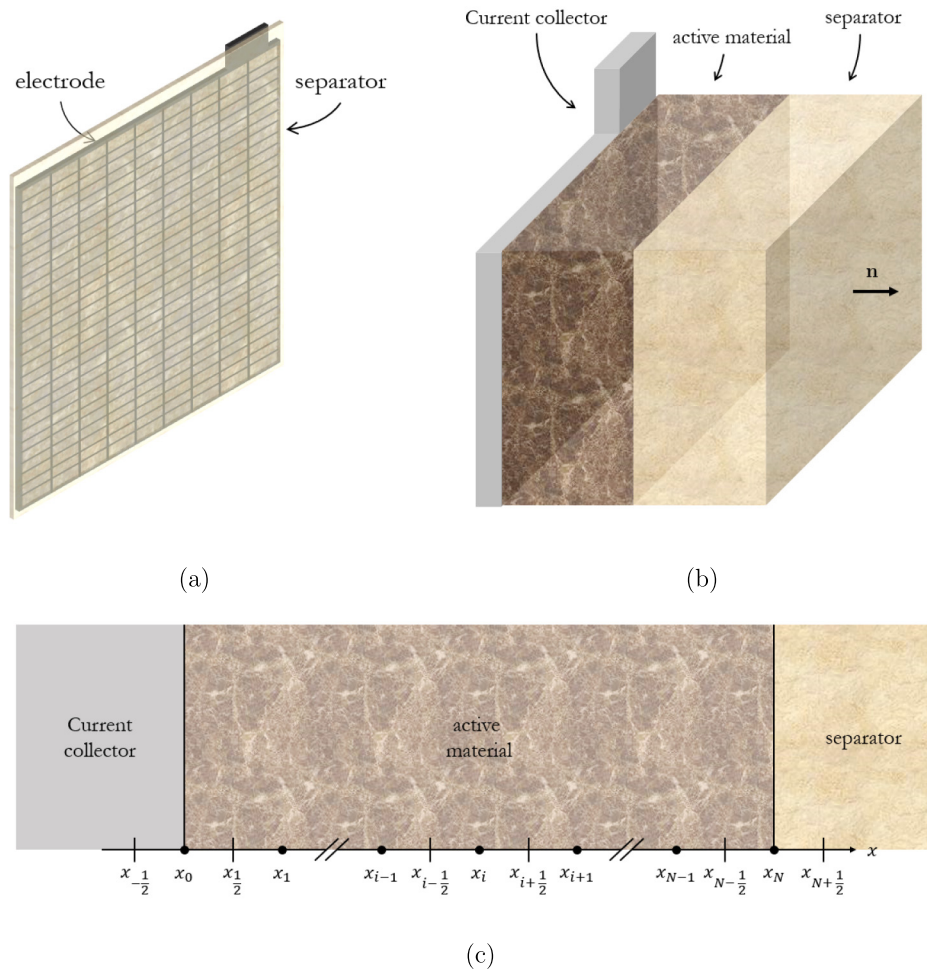


Fig. 2. Schematic drawing of a lead electrode along with a separator extracted from the cell, in real dimension (a), illustration of the model geometry simplicity in which the active material is considered pasted on a foil-like current collector, instead of a pasted grid (b), and one-dimensional numerical scheme used to solve the three differential algebraic equations, representing respectively charge, volume, and species conservation (c).

Furthermore, a simplification of the system of DAEs is proposed to the user who has the choice of deleting some terms that have very little influence on the model's performance. Therefore, NEOLAB is the promising result of a simple, fast, and efficient software allowing the simulation of the negative electrode behavior. Simulation data of the interfacial voltage, electrode porosity, and acid concentration are saved and plotted so it is possible to observe their evolution in space and time. Comparisons between simulation and experimental data can also be made to validate the model adjustable parameters.

3. Illustrative examples

To showcase the possibilities offered by NEOLAB, we can simulate the electrode performance over a broad range of discharging rates ($C/20$, $C/5$, $C/2$, $1C$, $2C$, $4C$, $8C$, and $12C$), which means from the very low rate of a 20-h discharge all the way up to the very high rate of a 5-min discharge. The initial conditions represent a fully charged lead electrode. The first simulation step consists of completely discharging the electrode and the second to leave it relax during 30 min. The physical quantities provided in the code are related to a 50 mm wide, 55 mm high, and 0.28 mm thick lead electrode, that is to say a lab-scale thin electrode able to deliver the power usually required to start an internal combustion engine.

The electrode voltage allows the user to know what to expect from the physics, according to the model parameter values. Alternatively, it can also be used to identify model parameters based

on the consistency between experimental data and simulation results, represented by dashed and solid lines in Fig. 4 respectively. The discharge current is applied to the electrode as a homogeneous current density at the electrode/separator interface. During the first seconds of discharge, the electrode behavior is dictated by the value of its double-layer capacitance [18,19]. Then, the electrode polarization evolves according to the Butler-Volmer equation [20], as a non-linear function of the discharge current. Finally, the end of discharge is characterized by the coverage of the porous electrode with lead sulfate crystals [21], which induces pore-clogging and consequently a porosity reduction. The voltage peak comes from the rapid decrease of the electrode active surface. The latter is linked to the porosity by means of a local state of charge of the electrode ensuring that the porosity stays inside its domain of definition ($\epsilon \in [\epsilon_{\min}, \epsilon_{\max}]$) [22]:

$$A_{\text{pb}} = A_{\text{pb},0} \frac{\epsilon - \epsilon_{\min}}{\epsilon_{\max} - \epsilon_{\min}}. \quad (9)$$

However, the model simplicity does not allow to take into account the impact of the discharging rate on the size distribution of lead sulfate crystals [23]. As can be seen from experimental data (Fig. 4), the curvature radius of the electrode voltage increases roughly in correspondence to the increasing current, demonstrating that Eq. (9) is way too simple. Other alternatives provided in the literature have been tried without much success [14,24]. Make the active surface dependent on the applied current in a physics-based framework has not been devised yet [25], but

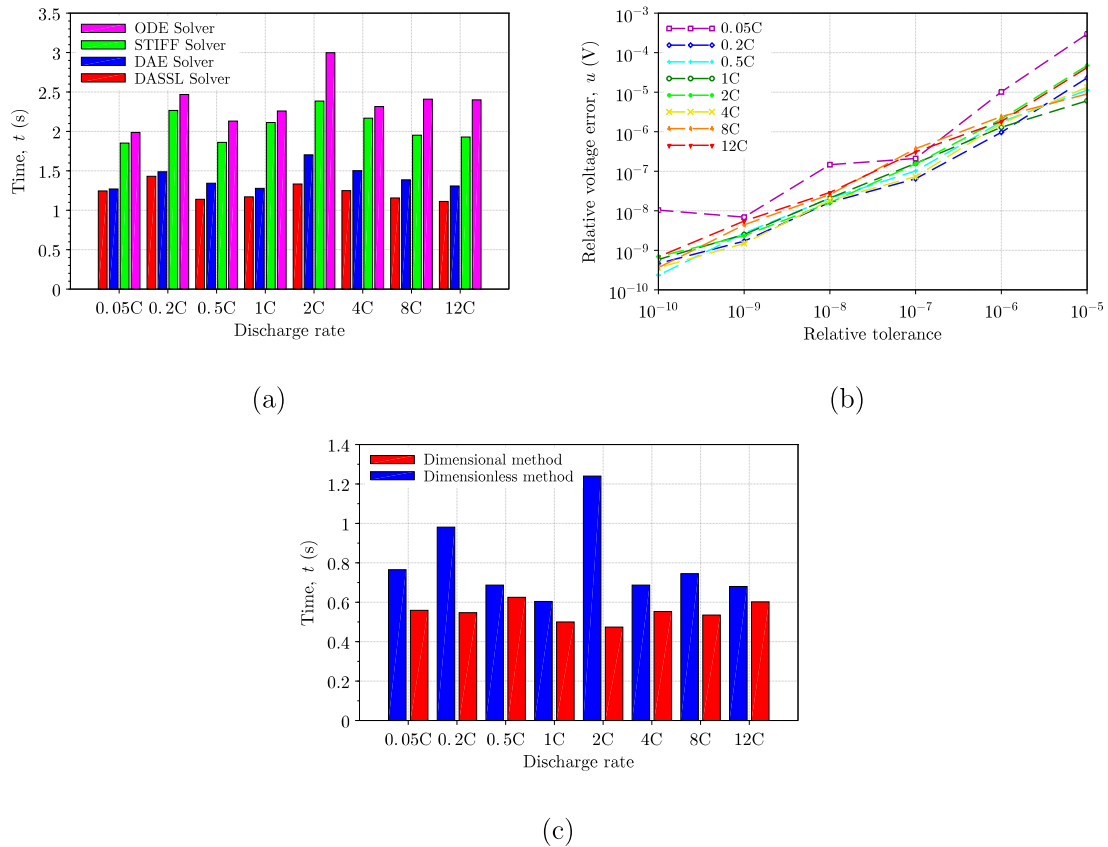


Fig. 3. Comparative analysis of the computation time for each solver with respect to the discharging rate from 0.05C to 12C followed by a systematic 30-minute rest period (a), relative voltage error for each rate with respect to the relative tolerance value of the solver, based on the simulation results obtained for a relative tolerance of 10^{-11} (b), and comparative analysis of dimensional and dimensionless models on the computation time with respect to the rate of discharge with the DASSL solver (c).

nucleation and growth of lead sulfate crystals [26] are the mechanisms that actively contribute to the electrode active surface evolution, on which the authors intend to focus in the next future.

The voltage simulation over the 30 min of rest following the full discharge of the electrode converges to the right asymptotic values for all applied rates, showing that the model is perfectly able to move from one equilibrium state (fully charged electrode) to another (fully discharged electrode) while staying consistent with thermodynamics. Applying a constant Dirichlet boundary condition, such as $c_{A|x=L} = c_{A,sep}$ to the partial differential equation of species conservation would not have led to such a result [27]. Since this boundary concentration value changes with the generation of lead-sulfate, one mole of electrons leading to the consumption of one mole of sulfuric acid, one can estimate it by integrating the current applied to the electrode over the discharge duration:

$$c_{A|x=L} = c_{A,0} + \frac{1}{V_1 F} \int_0^t I(\tau) d\tau. \quad (10)$$

The effective volume of electrolyte V_1 is a key parameter to adjust in order to ensure that only the volume of electrolyte contained inside the electrode porous structure and in the separator can supply the electrochemical reaction. Indeed, the diffusion coefficient of sulfuric acid inside the electrolyte is too low to exploit the species that are not located in the immediate vicinity of the electrode, even at small rates such as C/20. The simulated voltage stabilizes much faster than the real one, mostly because of the lack of knowledge of what is exactly going on inside the separator due to the influence of the counter electrode on this reserve of electrolyte.

Table 1

Maximum voltage drop within the thickness of the electrode simulated by the model at the end of discharge, with the corresponding current density applied as a boundary condition for each rate.

Discharge rate	Current density ($A\ m^{-2}$)	Voltage drop (mV)
C/20	5.82×10^0	0.3
C/5	2.32×10^1	1.0
C/2	5.82×10^1	2.0
1C	1.16×10^2	3.7
2C	2.33×10^2	6.2
4C	4.65×10^2	10.4
8C	9.31×10^2	16.2
12C	1.40×10^3	20.7

We could have also plotted the evolution of the electrode voltage within the thickness of the electrode at any given period of time during the discharge or the consecutive rest period, since our model gives access to this knowledge. However, to demonstrate that the electrical percolation in the porous electrode does not impede the current flow, we simply provide the maximum voltage drop within the thickness of the electrode observed at the end of the discharge, for each rate, in Table 1.

The evolution of the electrode porosity along its thickness during the discharge process is presented in Fig. 5. Starting from the same uniform value of ϵ_{max} , which is an initial condition common for all rates, one can see that the higher the rate, the higher the porosity gradient. The porosity is lower at the separator interface because more sulfuric acid is available, coming from the separator to fuel the electrochemical reaction. Fig. 5 allows to quantify the heterogeneity of active material utilization, that is to say, the local

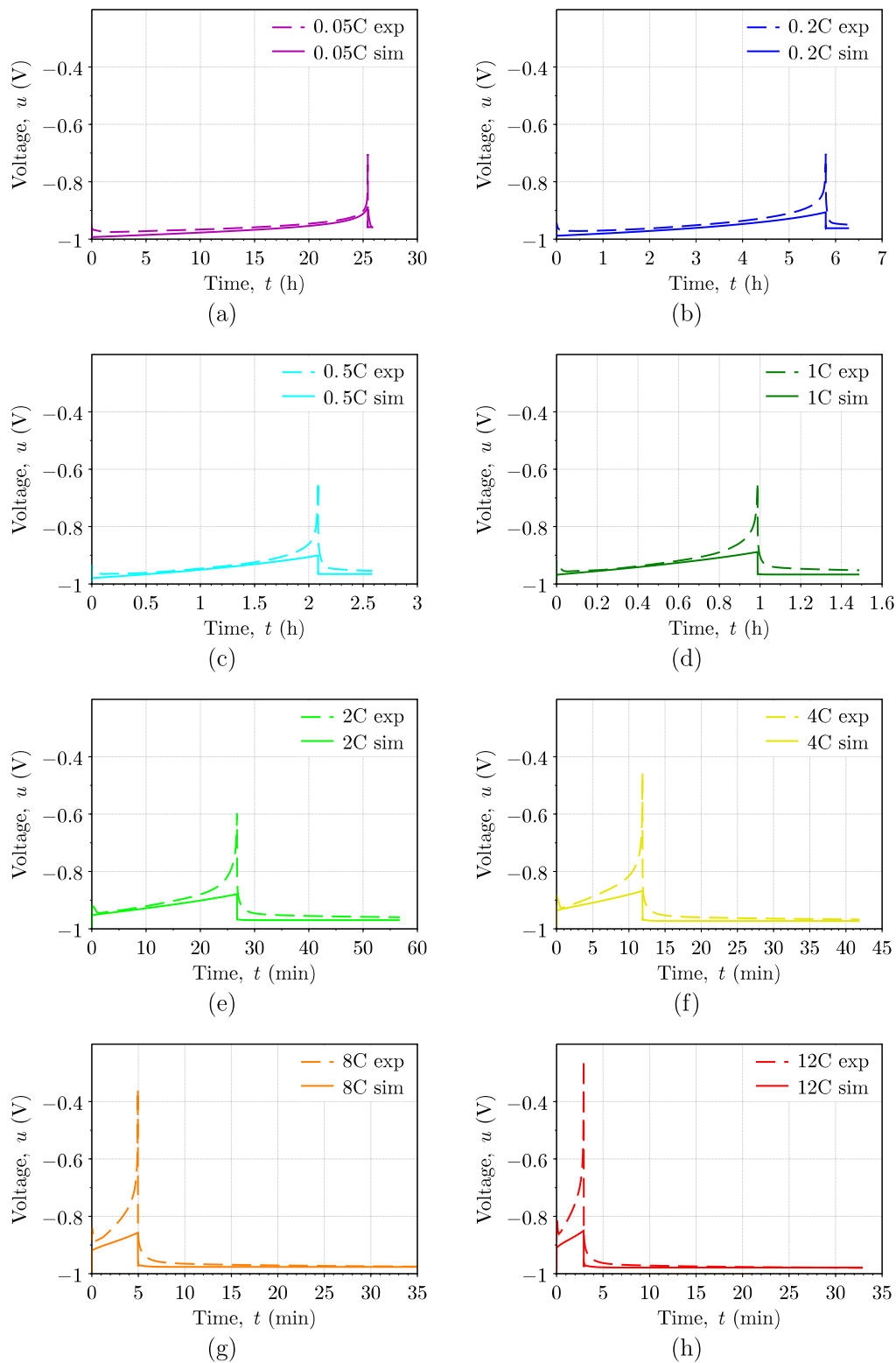


Fig. 4. Measured (dashed lines) and simulated (solid lines) electrode voltages with respect to time during a constant current discharge (from 0.05C (a) to 12C (h)) and a 30-minute rest period.

state of charge imbalance, for each rate. This may potentially help in designing new electrode architectures offering higher available capacity at very high rates through an optimal combination of porosity, thickness, and conductive additives.

Concerning the evolution of acid concentration along the electrode thickness (Fig. 6), one can see the uniformly distributed

initial value of $c_{A,0}$ at the beginning of discharge, common for all rates. As in the case of porosity, the higher the rate, the higher the concentration gradient. Unlike porosity, the minimum concentration value is observed at the current collector interface. However, this is the consequence of the same cause, the acid concentration inside the separator being higher than the one

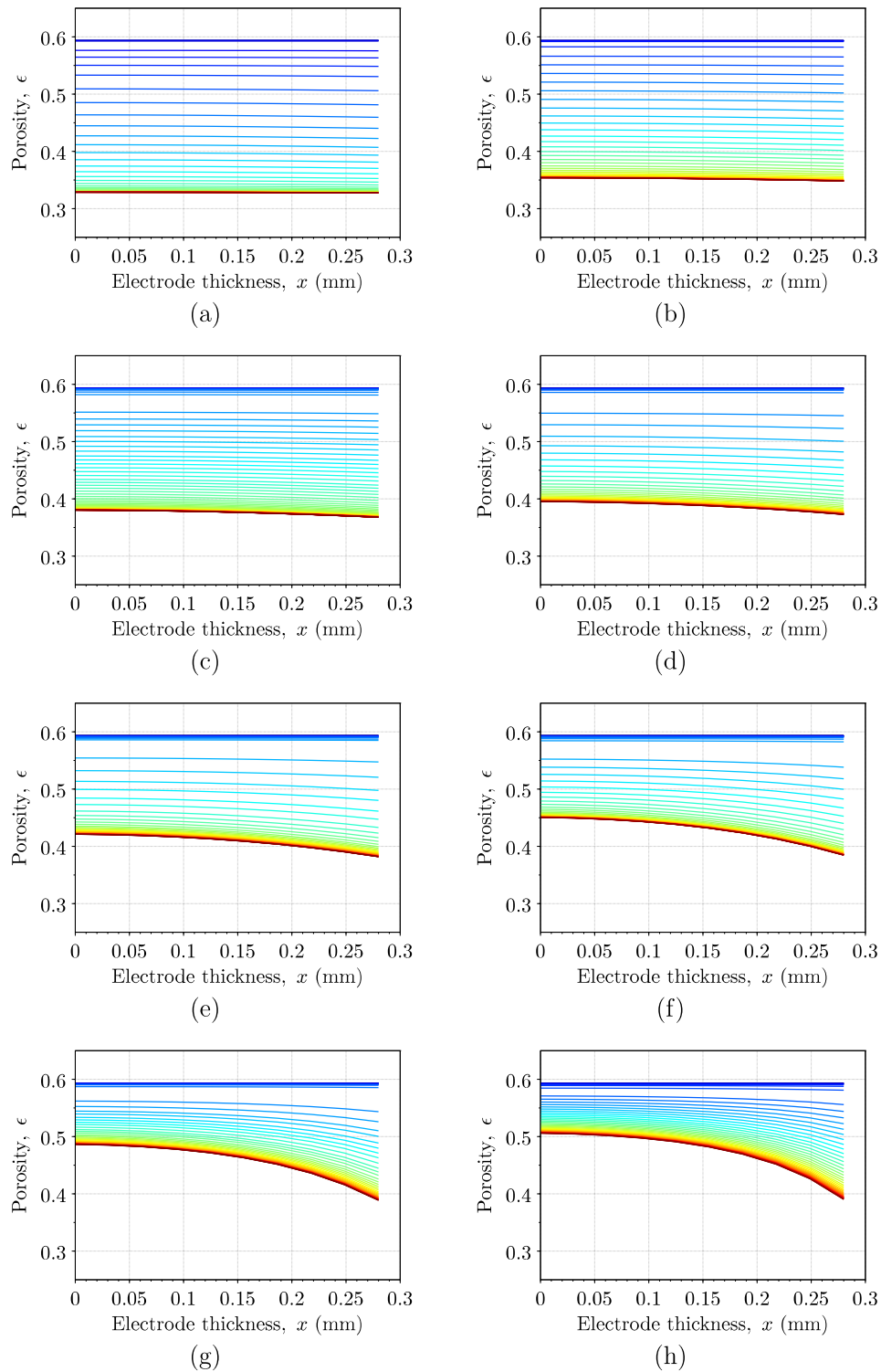
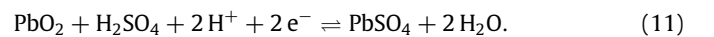


Fig. 5. Simulated electrode porosity (volume fraction of the electrode filled with electrolyte) with respect to the electrode thickness (expressed in its real dimension and starting from the current collector), during a constant current discharge ranging from 0.05C (a) to 12C (h).

inside the pores of the electrode due to acid consumption to generate lead sulfate crystals.

Internal physical quantities, such as porosity and acid concentration, are directly affected by the assumption made regarding the acid concentration of the separator. We have chosen in this example to take into account the consumption of species by the counter electrode (lead-dioxide), as described in Eq. (10). However, another phenomenon neglected with this assumption is

the fact that the lead-dioxide electrode also produces water while being discharged:



The conversion of lead dioxide into lead sulfate contributes to the dilution of sulfuric acid inside the electrolyte contained in the separator, and consequently lowers the concentration value for the same amount of charge exchanged.

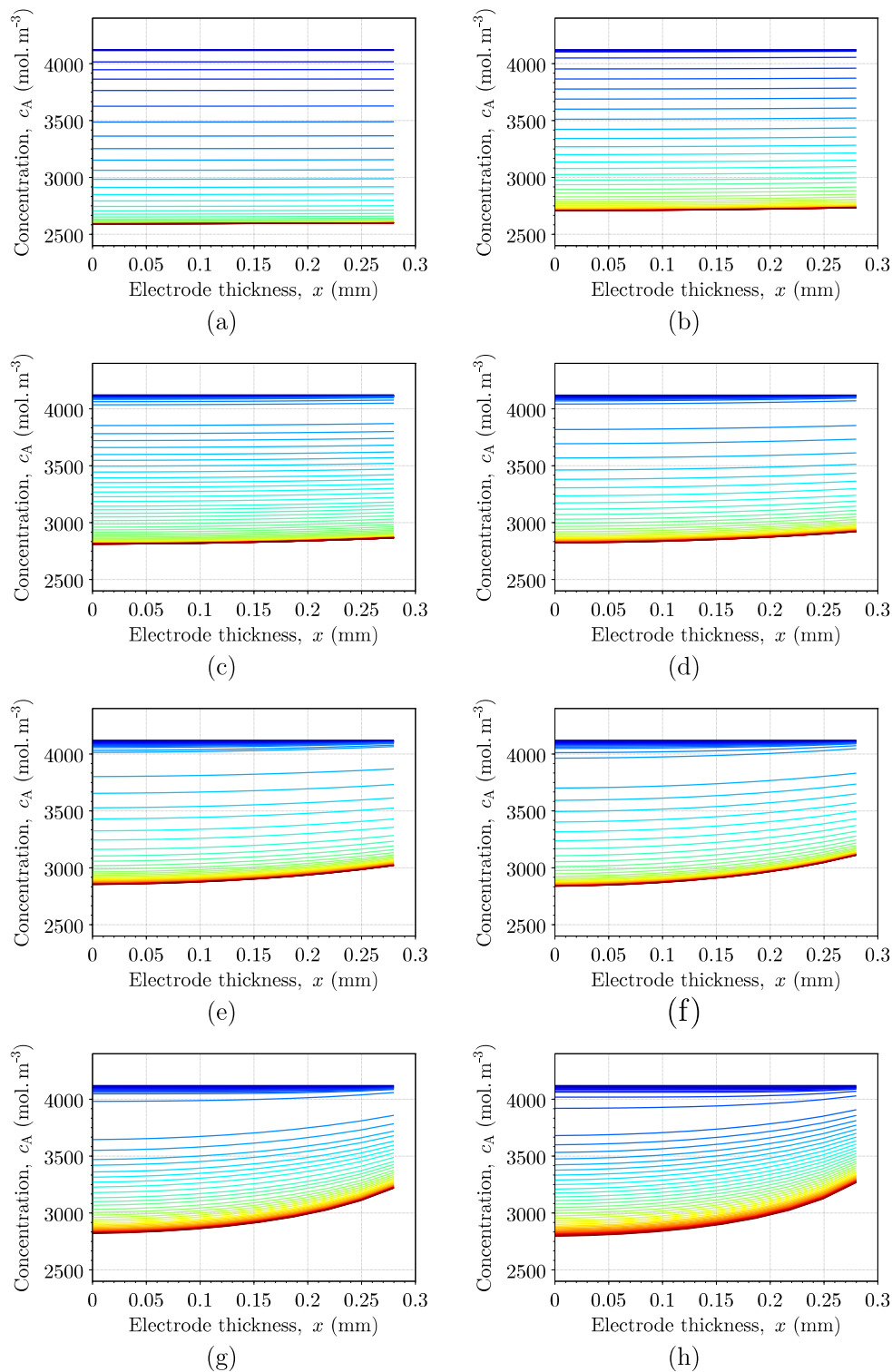


Fig. 6. Simulated acid concentration (electrolyte molarity) with respect to the electrode thickness (expressed in its real dimension and starting from the current collector), during a constant current discharge ranging from 0.05C (a) to 12C (h).

4. Impact

NEOLAB is more than just a physics-based model of the negative electrode of lead-acid batteries. It should be thought as a minimum working example intended to grasp the complexity of the modeling work required to simulate any electrode behavior with first principles. NEOLAB is simple enough to help proceeding step by step with this demanding modeling work:

- how to proceed with the multiple parameters needed;
- how to handle the physical relations that are not the main model equations;
- how to deal with the model initial conditions;
- how to solve the partial differential equations;
- how to manage multi-step simulations including discharge, rest, etc.

Based on an open-source platform, NEOLAB gives anybody the possibility to learn how to master all steps of multiphysics modeling of electrochemical systems for free. Battery modeling is a very lucrative job for those who know how to do it the proper way and this is particularly true when it involves physics and thus the knowledge of the battery internal state. Most of the people who want to achieve such an understanding of how physics explains the battery behavior are required to pay expensive licenses for software that are very useful when the geometry is complex or when one has no time to invest in learning how it works, but simply wants the things done. For sure, NEOLAB deals with a single spatial dimension, which makes the meshing simple whatever the number of domains to consider. However, this is not too much of a constraint, since the direction normal to the electrode plane is the direction of the moving species and charges and as such the most influential on the battery behavior. It gives also the advantage of fast computation time due to an optimal balance between complexity and accuracy.

We sincerely believe from our experience in this field that students and researchers all over the world will benefit from this tool and will make it able to simulate a broad range of electrochemical storage systems our modern societies demand.

5. Conclusions

Responding to the need of a growing community of students and researchers who want to get involved in the field of electrochemical storage systems, NEOLAB offers a new tool dedicated to a modeling domain where almost no open-source solutions exist. Physics-based models require extensive knowledge in thermodynamics, electrochemistry, mathematics, material and computer sciences. Based on the idea that a minimum working example is the best way to learn step by step how to model a battery, NEOLAB provides not only an immediate solution to simulate the behavior of the negative electrode of the well-known lead-acid battery, but also the framework to investigate plenty other primary and secondary battery technologies. Its simplicity and fast computation speed will help anyone to dig into the fundamental physics behind the battery behavior without limits, since any kind of equations can be tested, as long as the stiffness of the problem stays compatible with the selected solver. The authors intend to continue the development of this tool by adding more functionalities such as the simulation of the complete cell and the possibility to simulate discharges or charges based on either a current or voltage control.

CRedit authorship contribution statement

Mikaël Cugnet: Conceptualization, Methodology, Software, Validation, Resources, Data curation, Writing – original draft, Visualization, Project administration, Funding acquisition. **Florian Gallois:** Software, Validation, Writing – review & editing, Visualization. **Angel Kirchev:** Investigation, Resources, Data curation, Writing – review & editing. **Denys Dutykh:** Conceptualization, Software, Formal analysis, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data and code can be accessed here: <https://github.com/FlorianGallois/NEOLAB>.

Acknowledgments

This work has been realized with the participation from members of INES.2S and received funding from the French State under its investment for the Future Programme with the reference ANR–10–IEED–0014–01. FG acknowledges the French National Research and Technology Association for supporting his PhD thesis under the contract CIFRE No. 2021/1034. The work of DD has been supported by the French National Research Agency, through the Investments for Future Program (ref. ANR–18–EURE–0016 – Solar Academy), and by the Khalifa University of Science and Technology under Award No. FSU-2023-014.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.softx.2023.101394>.

References

- [1] IEA. Energy storage. 2020, <https://www.iea.org/reports/energy-storage>. Accessed: 2021-02-02.
- [2] May GJ, Davidson A, Monahov B. Lead batteries for utility energy storage : A review. *J Energy Storage* 2018;15:145–57. <http://dx.doi.org/10.1016/j.est.2017.11.008>.
- [3] Newman J, Tiedemann W. Porous-electrode theory with battery applications. *AIChE J* 1975;21(1):25–41. <http://dx.doi.org/10.1002/aic.690210103>.
- [4] Cugnet M, Liaw BY. Mathematical modeling of lead-acid batteries. In: Jung J, Zhang L, Zhang J, editors. *Lead-acid battery technologies*. Boca Raton, FL: CRC Press; 2016, p. 229–318, Ch. 9.
- [5] Howey DA, Roberts SA, Viswanathan V, Mistry A, Beuse M, Khoo E, DeCaluwe SC, Sulzer V. Free radicals: Making a case for battery modeling. *Electrochem Soc Interfaces* 2020;29:30–4.
- [6] Tiedemann WH, Newman JS. Mathematical modeling of the lead-acid cell. In: Gross S, editor. *Battery design and optimization PV 79-1*. Pennington, NJ: The Electrochemical Society Softbound Proceedings; 1979, p. 23–38.
- [7] Ekdunge P, Simonsson D. The discharge behavior of the porous lead electrode in the lead-acid battery. II. Mathematical model. *J Appl Electrochem* 1989;19:136–41.
- [8] Harb JN, Johnson VH, Rausen D. Use of a fundamentally based lead-acid battery model in hybrid vehicle simulations. In: *Proc. electrochem. soc. conf.*. Seattle, Washington; Spring 1999.
- [9] Boovaragavan V, Methakar RN, Ramadesigan V, Subramanian VR. A mathematical model of the lead-acid battery to address the effect of corrosion. *J Electrochem Soc* 2009;156(11):A854–62. <http://dx.doi.org/10.1149/1.3190510>.
- [10] Sulzer V, Chapman SJ, Monroe CW, Please CP, Howey DA. Faster lead-acid battery simulations from porous-electrode theory : Part I . Physical model. *J Electrochem Soc* 2019;166(12):A2363–71. <http://dx.doi.org/10.1149/2.0301910jes>.
- [11] Python Software Foundation. Python success stories. 2021, <https://www.python.org/about/success/>. Accessed: 2021-02-05.
- [12] Herring P, Balaji C, Aykol M, Montoya JH, Anapolsky A, Attia PM, Gent W, Hummelshøj JS, Hung L, Kwon H-k, Moore P, Schweigert D, Severson KA, Suram S, Yang Z, Braatz RD, Storey BD. BEEP : A python library for battery evaluation and early prediction. *SoftwareX* 2020;11:100506. <http://dx.doi.org/10.1016/j.softx.2020.100506>.
- [13] Scilab. About the company. 2021, <https://www.scilab.org/about/company>. Accessed: 2021-02-05.
- [14] Cugnet M, Laruelle S, Grugeon S, Sahut B, Sabatier J, Tarascon J-M, Oustaloup A. A mathematical model for the simulation of new and aged automotive lead-acid batteries. *J Electrochem Soc* 2009;156(12):A974. <http://dx.doi.org/10.1149/1.3224868>.
- [15] Bode H. *Lead-acid batteries*. New York, NY: John Wiley & Sons, Inc.; 1977, p. 387.
- [16] Jost J. *Partial differential equations*. 3rd ed.. New York, NY: Springer-Verlag; 1977, p. 356. <http://dx.doi.org/10.1007/978-0-387-49319-0>.
- [17] Petzold LR. Description of DASSL: a differential/algebraic system solver. 1982.
- [18] Tiedemann WH, Newman JS. Double-layer capacity determination of porous electrodes. *J Electrochem Soc* 1975;122(1):70–4. <http://dx.doi.org/10.1149/1.2134161>.

- [19] Srinivasan V, Wang GQ, Wang CY. Analysis of electrochemical and thermal behavior of li-ion cells. *J Electrochem Soc* 2003;150(1):A98. <http://dx.doi.org/10.1149/1.1526512>.
- [20] Bazant MZ. Theory of chemical kinetics and charge transfer based on nonequilibrium thermodynamics. *Acc Chem Res* 2013;46(5):1144–60. <http://dx.doi.org/10.1021/ar300145c>, [arXiv:arXiv:1208.1587v5](https://arxiv.org/abs/1208.1587v5).
- [21] Ekdunge P, Simonsson D. The discharge behavior of the porous lead electrode in the lead-acid battery. I. Experimental investigations. *J Appl Electrochem* 1989;19:127–35.
- [22] Gu H, Nguyen TV, White RE. A mathematical model of a lead-acid cell. *J Electrochem Soc* 1987;134(12):2953–60.
- [23] Wales CP. Effect of discharge current density on structure of the lead negative plate. *J Electrochem Soc* 1981;128(2):236. <http://dx.doi.org/10.1149/1.2127398>.
- [24] Tenno A, Tenno R, Suntio T. Charge-discharge behaviour of VRLA batteries: Model calibration and application for state estimation and failure detection. *J Power Sources* 2001;103(1):42–53. [http://dx.doi.org/10.1016/S0378-7753\(01\)00830-8](http://dx.doi.org/10.1016/S0378-7753(01)00830-8).
- [25] Cugnet M, Dubarry M, Liaw B. Peukert's law of a lead-acid battery simulated by a mathematical model. In: *ECS trans.*, Vol. 25. 2010, p. 223–33. <http://dx.doi.org/10.1149/1.3414021>.
- [26] Bernardi DM. Nucleation of lead sulfate in porous lead-dioxide electrodes. *J Electrochem Soc* 1990;137(6):1670. <http://dx.doi.org/10.1149/1.2086769>.
- [27] Dimpault-Darcy EC, Nguyen TV, White RE. A two-dimensional mathematical model of a porous lead dioxide electrode in a lead-acid cell. *J Electrochem Soc* 1988;135(2):278–85. <http://dx.doi.org/10.1149/1.2095601>.