SoftwareX 20 (2022) 101234

Contents lists available at ScienceDirect

SoftwareX

journal homepage: www.elsevier.com/locate/softx

Original software publication

ELC: Software and tutorial for finite-element modeling of electrochemical desalination

Johan Nordstrand *, Joydeep Dutta

Functional & NanoMaterials Group, Applied Physics Department, School of Engineering Sciences, KTH Royal Institute of Technology, Hannes Alfvéns väg 12, 114 19 Stockholm, Sweden

ARTICLE INFO

Article history:
Received 21 July 2022
Received in revised form 30 September 2022
Accepted 12 October 2022

Keywords:
Capacitive deionization
Desalination
Finite element
Modeling

ABSTRACT

Many works in capacitive deionization (CDI) use finite-element (FEM) simulations to investigate process behavior. Here, we present ELC, comprehensive software that integrates these methods with COMSOL Multiphysics. It can save significant time for common research questions in CDI operations and is well-suited for new research questions in complex and upscaled device designs. The ELC software has already been used for the simulation of time-varying desalination output, charge leakages, bipolar electrode devices, and stacks of over 100 CDI cells. Finally, we provide a video tutorial on how to use the software. In conclusion, ELC could be a strong software for aiding current and future research in electrochemical desalination.

Code metadata

Current code version	*V1*
Permanent link to code/repository used for this code version	https://data.mendeley.com/datasets/fy7c4z4vbn
Permanent link to reproducible capsule	*none*
Legal code license	*GNU*
Code versioning system used	*none*
Software code languages, tools and services used	*COMSOL Multiphysics*
Compilation requirements, operating environments and dependencies	*COMSOL Multiphysics 5.6*
If available, link to developer documentation/manual	*Nordstrand, Johan (2022), "ELC Software and Tutorial", Mendeley Data, V1, doi: 10.17632/fy7c4z4vbn.1*
Support email for questions	johanno3@kth.se

1. Motivation and significance

Capacitive deionization (CDI) uses electrochemical methods for the desalination of water [1,2]. The complex physics behind this process means that spatially resolved modeling traditionally requires large equation sets, intricate fitting processes, technical tricks, and extensive efforts to set up a model [3]. Recently, we developed a theory for CDI modeling starting from the analogy of battery simulations. The compiled theory was named the electrolytic-capacitor model (ELC). The approach meant that we could develop a model that blends with the existing simulation support for finite-element modeling (FEM) found in COMSOL Multiphysics. That makes the model generalizable, and we now present software based on it,

A key application of the program is simulating different device structures and configurations with a given electrode material. This could be different upscaled stacks, connection types, or geometrical sizes such as electrode thickness. It could also aid research questions related to operations and internal dynamics. As an example, one study [4] used the software to investigate 2D dynamics, leakages, and multi-ion solutions.

An advantage of the software is that the physics stays the same across applications, and new users can implement the software by simply entering the system parameters corresponding to their device. Having the parameters, it is possible to simulate the charging and adsorption process over time, including the internal state of the device.

The ELC software has several points of significance. First, ELC is user friendly. It requires limited knowledge about the detailed theory to use and technical tricks required to implement a model are already included in the program. Thus, it saves time when pursuing existing research questions. Secondly, it is

* Corresponding author.
E-mail address: johanno3@kth.se (Johan Nordstrand).

https://doi.org/10.1016/j.softx.2022.101234

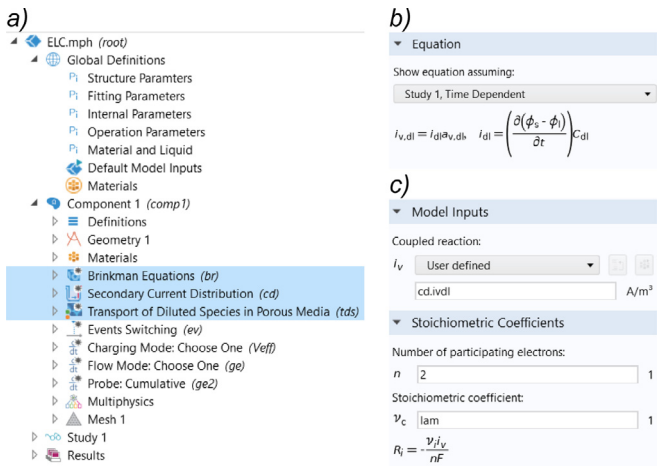

Fig. 1. An illustration of the software architecture. (a) This panel shows the name of the interface components. The three main ones are marked and correspond to the water flow, currents, and adsorption. (b) Each interface shows the equations they implement. This one shows that the volumetric current $i_{v,dl}$ depends on the area current i_{dl} and the area-to-volume ratio $a_{v,dl}$. Also, the local current i_{dl} depends on the capacitance C_{dl} and how quickly the relative voltage changes between the solid ϕ_s and liquid ϕ_l parts of the electrode. (c) The adsorption rate R_i of species i follows from the volumetric current i_v, the Faraday constant F, the stoichiometric coefficient (= charge efficiency "lam"). Also, n = 2 means two charges are required to remove one salt molecule.

flexible. It applies to a variety of designs and could enable new research in complex and upscaled devices. Thirdly, it has a tractable implementation, requiring a minimal set of relatively uncomplicated experimental data for fitting its reduced parameter set. This makes simulation-based studies more accessible.

2. Software description

A detailed description of how to use the ELC software can be found in the attached video.

2.1. Software architecture

The default geometry in the ELC software is a typical flow-between CDI structure (fbCDI). This includes an inlet pipe, a spacer channel, two electrodes, and an output channel. The user can change the geometry and flow modes freely.

The ELC software has three main interfaces that work "under the hood" (Fig. 1a). In CDI, the process mechanism is that a current drives adsorption and the purified water follows the water stream out of the device. Thus, the first main interface is the Brinkman Equations, which solves the water flow inside the channels and porous materials. Notably, it automatically handles internal transitions between internal boundaries. The second main interface is the Secondary Current distribution (Fig. 1a). It calculates the charging rate based on the effective voltage distribution and the resistive components in the solution and electrodes. Here, it is also possible for the user to enable the interface for electrochemical reactions on the porous electrodes (leakage currents). The third main interface is the Transport of Diluted Species in Porous Media (Fig. 1a). It calculates the transport of ions based on migration, advection, and diffusion. It also simulates adsorption (Fig. 1c). Charge efficiency, the fraction of current that leads to adsorption, is taken care of through a stoichiometric coefficient based on the dynamic Langmuir (DL) model, as explained elsewhere [5,6]. Normally, the user will not have to change any settings in these interfaces.

Several auxiliary interfaces in the ELC software are critical to getting a stable implementation (Fig. 1a). Firstly, the Events interface handles the switching between desalination and regeneration modes. A second ODE interface calculates the effective potential based on the external constant voltage (CV). As an alternative, the same interface can define the voltage implicitly through a specified constant current (CC). Finally, the Definitions node includes features for stabilizing the model, including a step function for smooth transitions.

The parameters (Fig. 1a) are classified as structure, operation, material, internal, and fitting parameters. The structure parameters concern the geometrical conditions, the material parameters correspond to the electrode material and electrolyte composition, and operational parameters correspond to the operational conditions. The internal parameters are derived automatically from other parameters. There are three fitting parameters. The first is the total capacitance of the device and the second is the external resistance. The third is the baseline voltage, as explained in detail in Ref. [5]. With a single fitting experiment (CV), it is $V_0 = V_{fit}\left(1 - \Lambda_{fit}\right)$. Here, V_{fit} is the fitting voltage and Λ_{fit} is the fitting charge efficiency.

The ELC software has some limitations though. The charge efficiency is considered as a constant with time and also varies linearly with the applied voltage, instead of time-varying and non-linear conditions. This can raise the simulation errors somewhat at the very beginning of any desalination process and for very low concentrations or below 0.4 V applied voltages (in practice voltages at or above 1.2 V are used). Also, the double layer capacitance is considered independent of the ion concentration, which raises the errors if the simulation covers a wide concentration range. More sophisticated models that exist such as the modified Donnan (mD) model can handle such ranges but we are not aware of an open-source program with that model but can share our implementation upon a reasonable request. We have chosen to communicate the more practical and less computationally intensive ELC model since it is a useful tool under normal simulation conditions.

Another limitation with that the Secondary Current Distribution (with Transport of Diluted species) is built for solutions with two species. NaCl, for instance. More complicated solutions require a Tertiary Current Distribution. We will share such software upon a reasonable request. However, that model is slower, less computationally stable, and sometimes less accurate. Therefore, we recommend using the standard ELC software whenever possible.

2.2. Software functionalities

The ELC software focuses on simulating desalination by CDI in multiple dimensions. The default required time of a simulation is 22 s, as tested on a laptop computer. In the outputs, the ELC software generates time-varying trends for the effluent concentration, overall current cumulative current and adsorption, and cell voltage. It also produces the internal time-varying performance including current, adsorption rate, flow velocity, and ion concentration. In the input, it automatically handles varying cycle times and switching rates, as well as variations in operational parameters such as ion concentration, flowrate, and external voltage.

The default is a flow-between CDI device operating CV charging. The geometry can be exchanged for other structures and modes, such as flow-through CDI. Interfaces can be enabled to address CC charging, leakage currents, and batch flow.

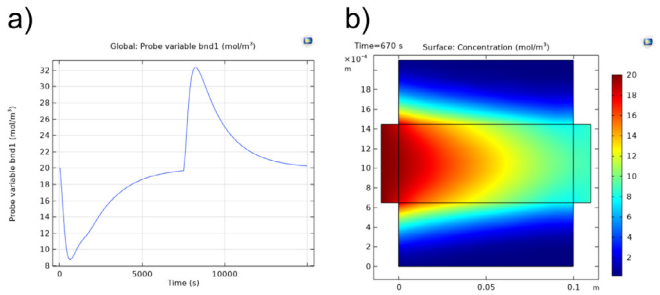

Fig. 2. This is the default output of the ELC software, as shown in COMSOL Multiphysics. The device is based on Ref. [7]. The flowrate is 0.42 mL/min, the voltage is 0.8 V, and the inlet concentration is 20 mM. (a) This is the effluent concentration from the device. The probe "bnd1" calculates the average salt ion concentration over the outlet boundary. (b) The internal concentration at the point of the lowest effluent concentration. The color scale corresponds to the molar concentration as written in the title. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3. Illustrative examples

The example shown here corresponds to the default settings that are shipped with the model.

There are two main outputs. Fig. 2a shows the conductivity profile of the effluent water stream. The concentration decreases at the beginning when the device starts to adsorb salt ions. Later, the accumulation of ions in the electrode reduces the adsorption rate until new ions are coming in faster than the device can remove them. At that time, the outlet concentration starts to increase again. Finally, the salt ions are released again when the voltage is removed, leading to a spike in the effluent ion concentration.

Fig. 2 shows the internal concentration profile at the point of the lowest effluent concentration. From this figure, we can see that the diffusion rate of ions into the electrodes is limiting how quickly the device can charge. The combined diffusion/transport behavior also affects how quickly the internal adsorption relates to a net output.

4. Impact

Several studies in CDI have used FEM methods. Examples of investigations that could be pursued with the current software are the internal concentration distributions [7], pressure drops [8], and flow distributions [8], including how they depend on the structure. More examples include localized ion starvation [7], the impact of flow [9], ftCDI [10] and fbCDI [3], upscaled devices [11], and charging characteristics [12]. The ELC software could save users' time because they do not have to rebuild the underlying program for every new application. Thus, it greatly aids in the pursuit of such research questions and could change the daily practice of the users. Aside from saving time, many critical implementational features presented here are never mentioned in normal papers. By making the methods public, we could thus make it possible for more research groups to pursue these types of research questions.

Unlike common models, the ELC software automatically implements the physics and internal boundaries. It could, therefore, allow users to investigate new and complicated device geometries. Specialized operations, including control systems [13], could also be tractably tested.

As the CDI field evolves, the modeling of upscaled systems will become more relevant. For instance, pilot plants [14] and startup companies [15] are emerging in CDI. Some recent studies have investigated upscaling from the simplified perspective of pressure and flow [8], or with decoupled modeling frameworks [11,16]. However, flows and fields can get complicated inside the device, meaning a comprehensive model requires a general description of internal transitions, as presented here. Another advantage is that the new ELC software is stable enough to handle large and unideal states. Based on this, we expect it to open the door to simulations of upscaled devices with complex geometries.

A good example of a study that has used the ELC software is Ref. [4]. Here, the authors simulate 2D dynamics in CDI. The work also revealed time-dependent simulations of multi-ion solutions. Desalination with various ion types is a core feature of CDI [17,18], but simulating the time-dependent effluent concentration of various ion types can be especially complex. Thus, a core value of the new software in that study is enhanced numerical stability. Finally, the work also shows time-dependent leakage reactions in CDI. This is significant because leakages can be difficult to simulate, especially in a spatially resolved model. We also note that the way of treating such reactions at the electrodes is not limited to leakages in CDI; rather, users could extend this approach to other electrochemical reactions in supercapacitor devices.

The enhanced simulation speed and stability also mean that more complex geometries can be considered. Ref. [19] uses ELC to simulate 5-cell stacks with bipolar electrodes and various flow pathways. Ref. [20] uses ELC to simulate upscaled cell stacks with over 100 CDI cells. The latter thus demonstrates that the model can handle complex systems that cannot be resolved in fewer than two dimensions.

5. Conclusions

FEM methods are common in studies on electrochemical desalination. However, research is hampered by the lack of ready-made tools for simulating process behavior. Specifically, complex physical interactions mean setting up simulations requires extensive theory knowledge and technical tricks. Hence, the new ELC software introduces support for future research. It compiles theory into ready-make interfaces and provides the required implementational tweaks under the hood. The generalized implementation means it can solve many related applications with minor changes in settings and limited demands from the user.

The ELC software aids the pursuit of the plentiful ongoing research questions related to structural design and operations. Also, it is especially well-suited for addressing future research efforts in upscaling. The stabilized implementation of the underlying model has proven useful for applications such as multi-ion deionization. Work using the ELC software has further shown that it enables investigations of detailed behavior and charge leakages during the desalination process. The same setup can also address other electrochemical processes in these devices.

Finally, we have deposited video tutorials showing how to use the software.

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

The data has been deposited on Mendeley data.

Acknowledgments

J.N would like to thank the Swedish research council (Diary No. 2018-05387) and J. Gustaf Richert foundation (Diary No. 2020-00584) for funding the work. Finally, J.N sincerely thanks Jonnie Fernlund for editing the tutorial video.

References

[1] Porada S, Zhao R, Van Der Wal A, Presser V, Biesheuvel PM. Prog Mater Sci 2013;58:1388–442. http://dx.doi.org/10.1016/j.pmatsci.2013.03.005.

[2] Suss ME, Porada S, Sun X, Biesheuvel PM, Yoon J, Presser V. Energy Environ Sci 2015;8:2296–319. http://dx.doi.org/10.1039/C5EE00519A.

[3] Nordstrand J, Dutta J. Nano Sel 2022;3:1021–35. http://dx.doi.org/10.1002/nano.202100270.

[4] Nordstrand J, Zuili L, Alejandro E, Carrillo T, Dutta J. Desalination 2022;525:115493. http://dx.doi.org/10.1016/j.desal.2021.115493.

[5] Nordstrand J, Dutta J. J Phys Chem C 2019;123:16479–85. http://dx.doi.org/10.1021/acs.jpcc.9b04198.

[6] Nordstrand J, Laxman K, Myint MTZ, Dutta J. J Phys Chem A 2019;123:6628–34. http://dx.doi.org/10.1021/acs.jpca.9b05503.

[7] Hemmatifar A, Stadermann M, Santiago JG. J Phys Chem C 2015;119:24681–94. http://dx.doi.org/10.1021/acs.jpcc.5b05847.

[8] Nordstrand J, Dutta J. Desalination 2020;500:114842. http://dx.doi.org/10.1016/j.desal.2020.114842.

[9] Mutha HK, Cho HJ, Hashempour M, Wardle BL, Thompson CV, Wang EN. Desalination 2018;437:154–63. http://dx.doi.org/10.1016/j.desal.2018.03.008.

[10] Suss ME, Baumann TF, Bourcier WL, Spadaccini CM, Rose KA, Santiago JG, et al. Energy Environ Sci 2012;5:9511. http://dx.doi.org/10.1039/c2ee21498a.

[11] Nordstrand J, Dutta J. Langmuir 2022;38:3350–9. http://dx.doi.org/10.1021/acs.langmuir.1c02806.

[12] Nordstrand J, Toledo-Carrillo E, Vafakhah S, Guo L, Yang HY, Kloo L, et al. ACS Appl Mater Interfaces 2022;14:1102–13. http://dx.doi.org/10.1021/acsami.1c20910.

[13] Nordstrand J, Dutta J. Npj Clean Water 2021;4:1–7. http://dx.doi.org/10.1038/s41545-020-00094-y.

[14] Tan C, He C, Tang W, Kovalsky P, Fletcher J, Waite TD. Water Res 2018;147:276–86. http://dx.doi.org/10.1016/j.watres.2018.09.056.

[15] Kunjali KL. Stockholm Water https://stockholmwater.com/ (accessed Jan 18, 2022).

[16] Xiaobing W, Jinqiu L, Yang L, Sen L, Dong L, Tingting M, et al. Water Air Soil Pollut 2021;232:1–10. http://dx.doi.org/10.1007/s11270-021-05320-y.

[17] Tang W, Kovalsky P, Cao B, Waite TD. Water Res 2016;99:112–21. http://dx.doi.org/10.1016/j.watres.2016.04.047.

[18] Tang W, He D, Zhang C, Waite TD. Water Res 2017;121:302–10. http://dx.doi.org/10.1016/J.WATRES.2017.05.046.

[19] Nordstrand J, Dutta J. Electrochim Acta 2022;141066. http://dx.doi.org/10.1016/j.electacta.2022.141066.

[20] Nordstrand J, Dutta J. 19th Int. multidiscip. model. simul. multiconference. 2022, p. 1–9. http://dx.doi.org/10.46354/i3m.2022.sesde.004.