Electrochemical energy storage: Theory meets industry

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Organizers: Marie-Liesse Doublet (Institut Charles Gerhardt) and Mathieu Salanne (Sorbonne Université)

1 State of the art

How much energy can a device store? How fast can it be charged? These two questions are at the heart of the research on electrochemical energy storage (EES). Two main families of devices coexist: supercapacitors which accumulate the charge at the surface of the electrodes through ion adsorption, and batteries in which the storage mechanism is based on redox reactions occurring in the bulk electrodes. Li-ion batteries have a high specific energy, keeping cellular phones, laptop and even cars working throughout several hours. For rapid power delivery and recharging, i.e. for high specific power applications, supercapacitors are then used.

Due to the recent advances in the field of materials science, the range of applications of EES devices has tremendously increased over the past two decades. The development of systems with improved performances and lower costs, as well as their large-scale production are now considered as vital issues for many countries. This can be seen from the recent creation of networks or institutes that gather academics and industrials, both at the national and European levels.

Most of the recent breakthroughs have however implied complex materials, often at the nanoscale. It is thus necessary to control the chemistry at the molecular level in all the active components of the devices, i.e. the two electrodes, the electrolytes. The various interfaces also have to be characterized and understood which implies considering potential dependent mechanistic approaches. Over the years, atomistic and molecular simulations have therefore appeared as one of the main keys to success in designing tomorrow's high-energy and high-power EES devices, in complement with in situ and/or in operando spectroscopy techniques. This is now well established in academic laboratories, which are now routinely building consortiums with synthesis, electrochemical and spectroscopic characterizations, together with modeling for developing new materials. However, this habit does not seem to be adopted yet by the industrial companies in the field. The objective of this workshop was therefore to bring together some of the worldwide experts in the field of EES simulations (and in particular the researchers who are developing the corresponding simulation tools) with the interested industrial partners. We hoped that such a workshop could help bridging the gap between needs and supply, which would put simulation at the centre of the future industrial developments of EES devices.

The state-of-the-art can be considered at two levels: 1/ Simulation methods which are routinely used to simulate EES devices. 2/ Initiatives which are currently undertaken to bring simulation tools and/or results within the reach of non-specialist users.

From the methodological point of view, many different methods are used or developed depending on the nature of the material, the targeted properties and the necessary time/length scales :

- the workhorse for studying the redox activity of bulk electrode materials is standard Density Functional Theory (DFT) since it is necessary to have access to the electronic structure ;
- for electrolytes, determining the transport properties involves the use of molecular dynamics. Depending on the availability of correct force fields, classical or DFT-based MD are generally used ;
- then further statistics or larger systems are generally studied by using lattice-based methods, such as kinetic Monte Carlo or Lattice Boltzmann.

Generally, standard DFT or MD packages can be used to study bulk materials. However in the case of interfaces, additional difficulties need to be overcome so that several groups are developing specific methodologies and/or simulation packages.

Despite the large growth in the simulation communities (especially DFT and MD) over the past decades, using these tools often requires lots of efforts for experimentalists and/or engineers in industry. For this reason, several groups are currently developing user-friendly interfaces, either in specific programs or directly accessible from website. For efficiency reasons, it is necessary to develop high-throughput frameworks and to link these tools with accurate databases. This implies that a common language is established between the communities of theoreticians and experimentalists, in order to build appropriate databases that will be helpful for material designers.

Finally, we should mention that several research groups are developing tools that aim to simulate systems at much larger scales. The objective is to provide a direct link with experiments, by directly computing macroscale properties similar to the ones obtained in electrochemistry experiments. Such multi-scale methods, most often based on the Butler-Volmer equation, are typically top-down approaches that aim to account for the material or electrolyte specificity in an effective manner through appropriate parameterizations. Huge efforts are being devoted to the development of bottom-up approaches, with however major issues due to transferability between different scales.

2 Major outcomes

Two major scientific points were discussed in the meeting:

1/ Virtual screening

The workshop was opened with an introductory lecture by Woomin Kyoung from Hyundai. He provided his company's view on how modeling should help the development of new materials. Although he discussed more specifically the case of electrolytes, he underlined that the process should be general for all the components of the battery. In short, the current approach is that new materials are discovered at the laboratory scale, then validated by using them in setups that are closer and closer to the commercial device. Overall, this discovery process takes up to 20 years. Hyundai believes that this time could be significantly reduced by involving modeling from the start, but it seems from the presentation that no procedure was clearly defined yet.
This issue was then discussed in several presentations. In particular, G. Hautier (Univ. Catholique de Louvain) showed results from a computational screening procedure that led to

the discovery of a new Li-ion battery material, LiTi2(PS4)3, which shows promising performance. G. Hautier nevertheless pointed to the fact that there is still a need for a close interplay between simulations and experiments, the latter being very useful to control the screening procedure at various stages of developments. The difficulty when performing virtual screening is the choice of the descriptors to select the best materials, a problem which has been rather extensively tackled by groups working on electrocatalysis. Several presentations were dedicated to this topic, in particular A. Gross (Ulm University) showed the correlation between the height of self-diffusion barriers, derived from density functional theory calculations, and the occurrence of dendrite growth in batteries. T. Vegge (DTU energy) provided a number of recent examples of how DFT simulations supported by machine learning could be used efficiently to identify the limiting thermodynamic, ionic and electronic transport mechanisms in novel Li-ion electrode materials and Li-S batteries.

2/ Multi-scale modeling of Li-ion batteries

Although the discovery of new materials is a major challenge, it is difficult to assess whether they can be commercialized based on a few intrinsic properties computed at the microscopic level. Indeed, most of the performance of the device depends on how the electrodes can be processed, put in shape, etc. These problems can be tackled by using a multi-scale modeling approach, which principles were detailed in the lecture of A. Franco (Univ. of Picardie Jules Verne), who presented results from a collaboration with Renault. His approach combines discrete modeling techniques (kinetic Monte Carlo, Coarse Grained Molecular Dynamics, Lattice Boltzmann,…) with continuum models at multiple spatio-temporal scales and deep learning algorithms. He showd how this approach could be connected to experimental characterizations by focusing on the simulation of the manufacturing of lithium ion batteries. A second illustration of the importance of multi-scale simulations was provided by C. Delacourt (Univ. of Picardie Jules Verne), who showed that it was possible to predict the occurrence of degradation phenomena, such as Li plating during charge, in order to better control (or avoid) it.

In addition to these two points, all the presentations provided a good panorama of the simulation techniques available to simulate Li-ion batteries and supercapacitors, ranging from DFT to continuum methods. The feedback was very positive from the various industrial representatives, since some of them knew very little about modeling methods before the workshops.

3 Community needs

The various presentations showed that there is a huge number of codes to simulate Li-ion batteries at all the scales. Nevertheless, several challenges still exist, in particular for the understanding of the electrode/electrolyte interface.

Not surprisingly, most of the presentations underlined the need for large-scale HPC resources. In particular, virtual screening requires to perform simulations on thousands of materials, which cannot be made on local computers.

It seems that the most important lack is currently the database. Even if there are several initiatives such as the Materials Genome, which was introduced by A. Van der Ven (University of California Santa Barbara), the various presentations showed how the simultaneous utilization of data from multiple domains (and not only simulation) plays a critical role in accelerating the industrial discovery process. More precisely, the accelerated discovery and inverse design of future batteries using generative deep learning models trained on all sources

of available data, i.e., large scale multi-fidelity data sets, multi-scale computer simulations and databases, operando characterization from large-scale research facilities, high-throughput synthesis and laboratory testing will be needed very quickly. It would be interesting to discuss whether the CECAM could play a role on the development of such a database, since it is one of the only European organization which includes multi-scale modeling as an objective and not only a subpart of it (several centers of excellence were dedicated to materials but they were mostly interested in the use of DFT to study them). Anyway such an initiative should strongly include the participation of experimentalists.

4 Funding

During the meeting we mainly focused on the European project Battery 2030+, which has just been launched in 2019. This flagship program includes most of the main European universities and research centers, and it is supported by many organisations (see https://battery2030.eu). T. Vegge, who is implied in the proposal writing, has devoted a large part of his talk to present the objectives and how they should be tackled by the computational community. In particular, we should be implied in BIG-MAP: Battery Interface Genome - Materials Acceleration Platform. Several actions will be funded in the project, among which the most interesting for our community are:

- The built of databases
- The inverse computational design of battery materials and interface
- Machine learning modules for automated analysis
- Multiscale simulations and physical models

All the results will be put together in a central Artificial Intelligence. It is clear that this project will be a very important opportunity of funding for European researchers working in the field of energy storage over the next few years.

5 Will these developments bring societal benefits?

There are two main societal benefits of the development of better electrochemical energy storage devices:

1/ A better inclusion of renewable energies in the electricity mix. Indeed, due to their intermittent character, it is very difficult to fully benefit from the production of wind mills or solar panels in highly electrified regions such as Europe. They are very often complemented by fossil fuels-based production, which is highly problematic in terms of the carbon footprint. Nevertheless these applications need the development of low-cost technologies in order to be competitive. Deploying these technologies is very urgent since global warning is already impacting our societies.

2/ Accelerating the electrification of the transport sector. The latter is responsible for a large share of the CO2 emissions in European countries. The rapid growth of the Li-ion batteries has allowed electric vehicles to progressively increase their share on the market. In countries where the electricity is low-carbon (France, Norway, Belgium, Switzerland, etc), the impact on the carbon emissions should be very important on the future.

These two benefits reflect on the list of industrial partners who participated to the workshop: Hyundai, Peugeot (PSA), Umicore, CEA, IFP, Happy electrons & Equinor.

6 Participant list

Organizers

Doublet, Marie-Liesse
Institut Charles GERHARDT, France
Salanne, Mathieu
Sorbonne Université, France

Franco, Alejandro A. – Université de Picardie Jules Verne, Amiens, France
Van der Ven, Anton – University of California Santa Barbara, USA
Gross, Axel – Ulm University, Germany
Kirchner, Barbara – University of Bonn, Germany
Morgan, Ben – University of Bath, UK
Merlet, Celine – CNRS - Université Paul Sabatier, France
Zhang, Chao – Uppsala University, Sweden
Delacourt, Charles – Université de Picardie Jules Verne, Amiens, France
Hautier, Geoffroy – Université catholique de Louvain, Belgium
Honkala, Karoliina – University of Jyväskylä, Finland
Martirez, Mark – Princeton University, USA
Ghadrdan, Maryam – Equinor, Norway
Gosika, Mounika – Indian Institute of Science, India
Cabelguen, Pierre-Etienne – Umicore, Belgium
Clement, Raphaele – University of California Santa Barbara, USA
Vegge, Tejs – Technical University of Denmark, Denmark
Kyoung, Woomin – Hyundai Motor Company, South Korea
Chandesris, Marion – Atomic Energy and Alternative Energies Commission (CEA), France
De Bruin, Theodorus – French Institute of Petroleum (IFP), France
Ercole, Loris – EPFL, Switzerland
Hollóczki, Oldamur – University of Bonn, Germany
Jeanmairet, Guillaume – Sorbonne Université, France
Perez, Miguel – University of Oxford, UK
Prezzi, Deborah – CNR Nano, Modena, Italy
Thorner, Gentien – Groupe PSA, France
Zanetti-Polzi, Laura – Università degli Studi dell'Aquila, Italy