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Mohammed Alabdali, Franco Zanotto, Virginie Viallet, Vincent Seznec, Alejandro Franco. Microstructurally resolved modeling of all solid-state batteries: Latest progresses, opportunities, and challenges. *Current Opinion in Electrochemistry*, 2022, 36, pp.101127. 10.1016/j.coelec.2022.101127. hal-03880347

HAL Id: hal-03880347

<https://u-picardie.hal.science/hal-03880347>

Submitted on 29 Jun 2023

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Review Article

Microstructurally resolved modeling of all solid-state batteries: Latest progresses, opportunities, and challenges

Mohammed Alabdali¹, Franco M. Zanotto^{1,2}, Virginie Viallet^{1,2}, Vincent Seznec^{1,2,3} and Alejandro A. Franco^{1,2,3,4}

Abstract

All solid-state batteries are promising high-energy-density storage devices. To optimize their performance without costly trial and error approaches, microstructure-resolved continuum models have been proposed to understand the influence of the electrode architecture on their capabilities. We discuss the recent advances in the microstructure-resolved modeling of solid-state batteries. While not all of the experimentally observed phenomena can be accurately represented, these models generally agree that the low ionic conductivity of the solid electrolyte is a limiting factor. We conclude by highlighting the need for microstructure-resolved models of degradation mechanisms, manufacturing effects and artificial intelligence approaches speeding up the optimization of interfaces in all solid-state-battery electrodes.

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Current Opinion in Electrochemistry 2022, 36:101127

This review comes from a themed issue on **Fundamental & Theoretical Electrochemistry (2022)**

Edited by Rama Kant and M.V. Sangaranarayanan

For complete overview about the section, refer [Fundamental & Theoretical Electrochemistry \(2022\)](#)

Available online 5 September 2022

<https://doi.org/10.1016/j.coelec.2022.101127>

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Keywords

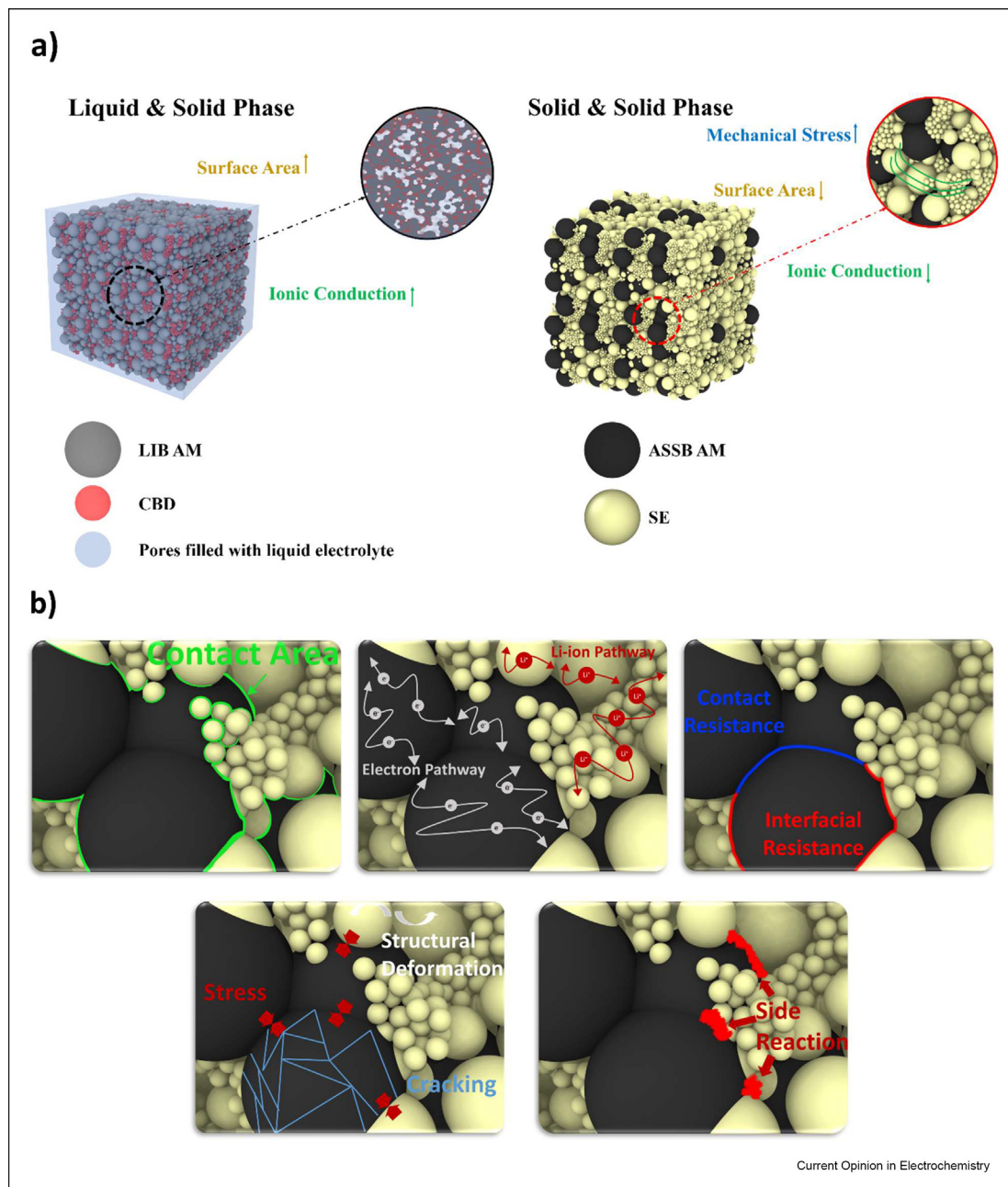
All solid-state batteries, Microstructurally-resolved modeling, Composite electrodes, Machine learning.

Introduction

To keep up with the increasing energy storage demand, high-performance batteries with low cost and long-life cycles are required. Lithium-ion batteries (LIBs) have remained the choice for portable devices because of their high gravimetric energy densities, but safety concerns and limited energy density in LIBs have increased interest in batteries based in alternative chemistries [1]. In this context, all solid-state battery (ASSB) technology shows the promise to partially meet the demand for energy storage. Safety stands out among the advantages of ASSBs, since the use of an inorganic solid electrolyte (SE) reduces the risk of fire in comparison with LIBs, which contain flammable organic electrolytes. Additionally, modern ASSBs with composite electrodes made of blends of active material (AM) and SE particles present high energy density [2]. However, there remain significant performance limitations because of the limited ionic and electronic percolation networks within the composite electrodes [3]. The insufficient surface contact area between the AM and the electrolyte particles plays a role in the low performance of ASSBs, in contrast to classical LIB electrodes where the surface area of the AM particles is well wetted with the liquid electrolyte (Figure 1a) [4,5].

Research in ASSBs is currently focused in three main issues: i) Understanding and minimizing the degradation mechanisms (Figure 1b) that occur at the interfaces among the different components of the composite electrode, that is, AM/SE/carbon additives, as well as between the different compartments of the cell, that is, positive composite electrode/SE layer/negative electrode. In some cases, there is a low chemical compatibility among the different compartments, which may promote chemical reactions and even dendrite growth (in cases when lithium metal is used as an anode), leading to the formation of new undesirable phases with high resistivity. This induces fast capacity fading in the ASSBs or even short circuits [3,5]; ii) The microstructure of the composite electrode hinders performance because of crack formation [7]. This calls for improvement in the electrodes architecture to optimize ionic and electronic conductive paths; iii) Eventual (de)intercalation-

Figure 1



(a) Schematic comparison of interfaces in all solid-state battery and lithium-ion battery electrodes; (b) possible degradation mechanisms at the contact area between the active material and solid electrolyte particles in all solid-state battery (inspired from [4,6]).

induced AM volume changes induce mechanical stresses between particles, damaging the SE particles or causing contact loss between the AM and electrolyte. This causes mechanical aging, which is expected to decrease power density, as fractures in the electrolyte represent a barrier for lithium-ion transport inducing rate performance decay. This calls for strategies to minimize contact loss due to mechanical stress, of which the most common is

the inclusion of carbon additives to improve the electronic pathways within the electrode. However, this increases the degradation rate of the SE, resulting in limited ionic percolation of lithium [8].

These challenges could be solved by means of the appropriate design of electrode microstructures. Experimentally, this optimization involves costly and time-

consuming trial and error approaches. However, computational modeling facilitates and speeds up the process [5] by allowing the exploration of the effects of different microstructural parameters in the cell performance. There are many possible approaches to model the relevant physical processes taking place in an ASSB cell, which include electrochemical, transport and thermo-mechanical phenomena involving the active, inactive, and electrolyte materials constituting the cell. These range from the nanoscale, such as molecular dynamics-based approaches, to the millimetric scale in the case of continuum approaches. The latter ones typically have the form of partial differential equations which are numerically solved in terms of state variables such as concentrations and electrostatic potentials as a function of spatial coordinates and time. This makes them very convenient to obtain macroscopic information, which can be directly compared with experimental results, such as discharge-charge curves or other electrochemical performance indicators. In this short review, we focus exclusively on the continuum modeling of ASSBs based on SEs (excluding polymer-based electrolytes) aiming to account for the electrodes' microstructures, by highlighting the most important works of the last years from our viewpoint. We have also summarized some of the characteristics of the covered models in Table 1 to facilitate comparison between them.

Models of all solid-state batteries with composite electrodes

The first continuum models describing ASSB operation principles were devoted to micro-device applications (e.g. wireless sensors and medical implants). In 2011, Danilov *et al.* proposed a one-dimensional continuum model of ASSBs featuring a metallic lithium anode, a thin LiCoO_2 intercalation electrode, and a Li_3PO_4 SE [7]. The model describes Butler-Volmer-like charge transfer kinetics at the electrolyte|electrode interface, lithium diffusion in the electrode, and transport of lithium ions in the electrolyte. The latter is assumed to be governed by both diffusion and electromigration. Because of the fitting procedure of the implicated parameters in the model (e.g. kinetic constants), the calculated galvanostatic voltage profiles are in agreement with the experiments. The model allows calculating diffusion and electromigration fluxes, concentration profiles and overpotentials. Interestingly, this early model already shows that, especially at high discharge rates, the transport limitation in the electrolyte determines at least half of the overall overpotential. It is an early electrochemical model that paved the way for other studies, yet it does not involve any of the microstructural issues faced by modern ASSBs. A similar model was reported a year later by Fabre *et al.* [8].

In principle, continuum models can be used to gain insights into the interfaces and to propose ways to

maximize percolation and surface contact area while minimizing mechanical stresses. It is important to note that the one-dimensional (1D) continuum models (e.g. the above-mentioned Danilov's model) [7] consider electrodes as bulk AMs; thus, they are not able to capture electronic percolation aspects in ASSB composite electrodes. Significant conceptual work on electronic percolation in composite electrode of solid oxide fuel cells has been reported for more than 20 years [9–12], which may have been a source of inspiration for several models reported in the following discussions. It should be approached by describing the physical phenomena taking place at each of the AM and SE particles within the electrode volume, by explicitly accounting for their spatial location in three dimensions. A typical composite electrode ranges from ten to hundreds of thousands of such particles, and with the current computational power it is impossible to account for every phenomenon. Nevertheless, there are examples of realistic composite electrode structures in reduced volumes, arising from tomography images or stochastic algorithms generators, thanks to the spectacular rise in computational power [4,6].

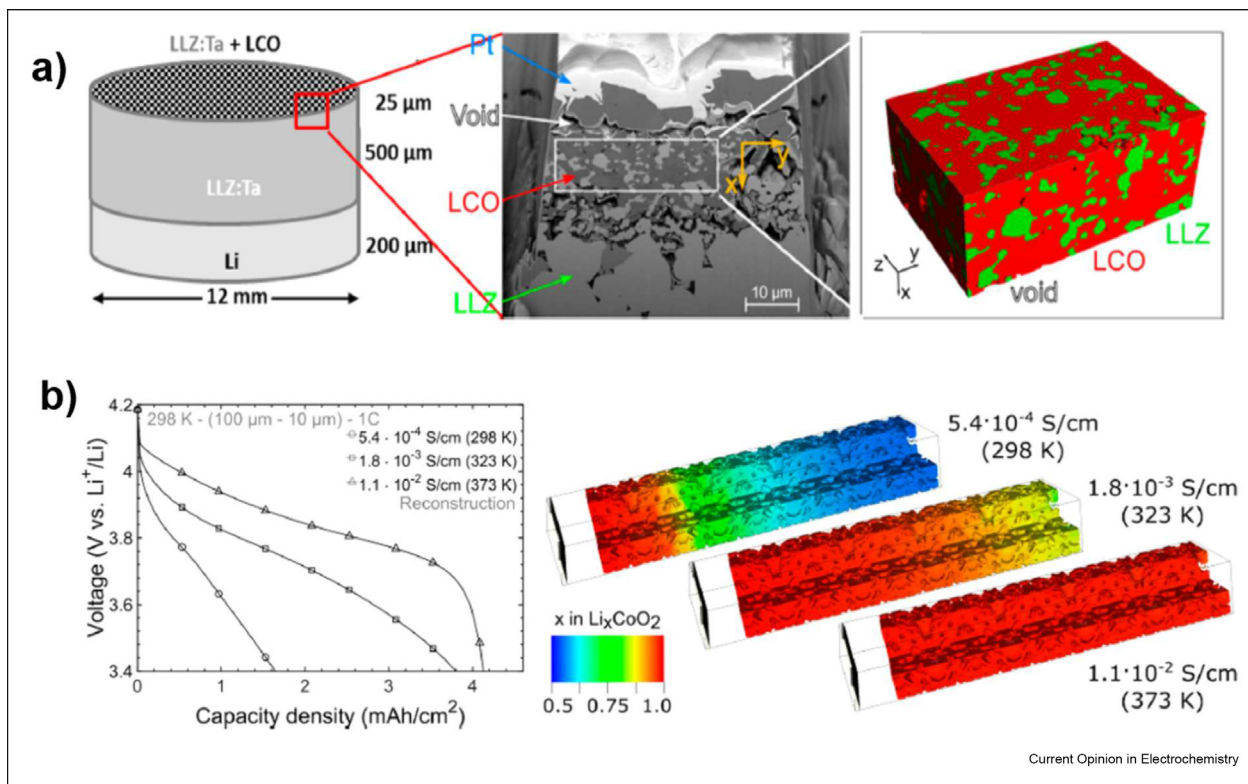
Bistri and Di Leo reported a chemo-mechanical model able to simulate in two dimensions the effect of particle size distributions (PSDs) on the electrochemical response and the resulting mechanical stresses in ASSB composite electrodes [13]. As far as we know, the first three-dimensional (3D) resolved electrochemical model of an ASSB cell was reported in 2018 by Finsterbusch *et al.* [14]. The modeled cell arises from computer tomography (CT) of a real cell made of a Li metal negative electrode, a Ta-substituted $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZ:Ta) garnet electrolyte, and a LLZ:Ta/ LiCoO_2 positive composite electrode (Figure 2a). The physics implemented in this model is similar to the Danilov *et al.* models previously mentioned [7], where the model can inherently capture the effects of materials heterogeneities and tortuosity factor on performance manifested by the ionic and electronic percolation paths in the composite electrode. Calculations demonstrate the ability of the model to reproduce the electrochemical behavior at elevated temperatures, but the model fails in simulating the performance at room temperature. Cathode AM/SE interfaces were found to induce low performance at room temperature. The authors provide recommendations to improve the power performance. For instance, the model results show the importance of considering several elements in optimizing SE|AM interfaces to reduce the SE layer thickness. Reducing the SE layer thickness improves the cell rate performance and the energy density by decreasing its porosity to increase the compactness of the cathode electrode. However, the tradeoff remains between the amount of the AM content and the thickness of composite electrodes to reach the optimal capacity and charge rate. Although it

Table 1

Comparison between the features of studies presented in this review.

Authors	Reference	Chemistry	Aim of the study	Structure	Simulation technique
Danilov <i>et al.</i>	7	LiCoO ₂ /Li ₃ PO ₄ /Li metal full cell	Analysis of fluxes, concentration profiles, and potentials	1D	mathematical modeling
Bistri, Di Leo	13	LiCoO ₂ /Li ₁₀ GeP ₂ S ₁₂ composite electrode	Effect of PSD and mechanical stresses on the electrochemical response	2D	surface element framework
Finsterbusch <i>et al.</i>	14	LLZ:Ta/LiCoO ₂ composite electrode/LLZ:Ta/Li metal	Improvement of power performance	3D	microstructure-resolved simulations
Neumann <i>et al.</i>	15	NMC622/thiophosphate composite electrode	- Assess lithiation distribution along length - Assess the impact of a reduced contact between the electrode and the current collector - Investigate the impact of delamination of the solid electrolyte from the active particle surface	3D	microstructure-resolved simulations
Bielefeld <i>et al.</i>	19	Chemistry neutral	Analyze the effect of contact surface area	3D	Ionic and electronic conduction clusters calculated according to percolation theory
Bielefeld <i>et al.</i>	20	NMC/thiophosphate/binder composite electrode	Assess the impact of binder content, AM particle size, and porosity on the effective ionic conductivity and tortuosity factor	3D	Flux-based simulation approach
Bielefeld <i>et al.</i>	22	NMC811/Li ₆ PS ₅ Cl composite electrode	Study the influence of residual voids within the electrode, different PSDs, and arrangement of AM and SE particles	3D	Electrochemical simulation (FEM)
Yamakawa <i>et al.</i>	23	LiCoO ₂ /SE	Unravel correlations between the power density, the volume fraction and the particle size of AM and SE particles. Effect of sintering.	3D	Phase-field modeling approach (FEM) and ML
So <i>et al.</i>	25	Si AM/sulfide SE	Study SE plastic deformation and mechanical interactions within the microstructure	3D	DEM

Figure 2



(a) Schematic of the all solid-state battery cell modeled by Fintersbuch *et al.* [14] along with its SEM micrograph and 3D reconstruction; (b) calculated impact of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ conductivity on the performance of the composite electrode and distribution of lithium in the active material at the lowest cutoff voltages (reproduced with permission from [14]).

is a joint experimental and modeling approach to obtain a 3D reconstruction that couples the structure of the cell with the electrochemical behavior to accurately predict the discharge curves at elevated temperatures, there is still a room for improvement to cover a broader range of operating conditions since the model remains unfitted and all parameters were kept as obtained experimentally from CT.

As a result of a collaboration between the groups of Latz and Janek, a study was reported combining experimental characterization techniques and computational simulations of $\text{Li}(\text{Ni}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2})\text{O}_2$ (NMC622)/thiophosphate-based ASSBs [15]. Simulations of the discharge curves and electrochemical impedance spectroscopy were carried out on cathode electrode microstructures arising from CT and compared with experimental data. The model identifies the decreasing electronic conductivity of the NMC622 on lithiation causes capacity losses especially at high rates where there is a low utilization of the region nearby the current collector. Furthermore, it was found that the low ionic conductivity of the SE favors intercalation

reactions to happen near the separator. These two competing aspects eventually lead to a sandwich-like lithiation of the AM (i.e. a higher lithiation takes place on the sides where it is close to the current collector and the separator than in the center of the electrode) particularly in the case of thick electrodes. The model is also able to assess the impact of a reduced contact between the electrode and the current collector, by artificially modifying the electrode microstructure. This reduces the specific capacity at high currents, causing enhanced local currents and results in heterogeneous lithiation near the current collector, accelerating the local reduction of the AM electronic conductivity, destroying the electronic percolation network within the composite electrode. Thus, the current collector coating and the addition of carbon additives are recommended as possible strategies to avoid this phenomenon. Finally, by artificially modifying the electrode microstructure, the model is used to investigate the impact of delamination of the SE from the active particle surface on the cell performance eventually caused by AM volume changes during cycling. However, the model is not able to capture the

specific capacity loss at high currents observed experimentally, indicating that there are other phenomena involved.

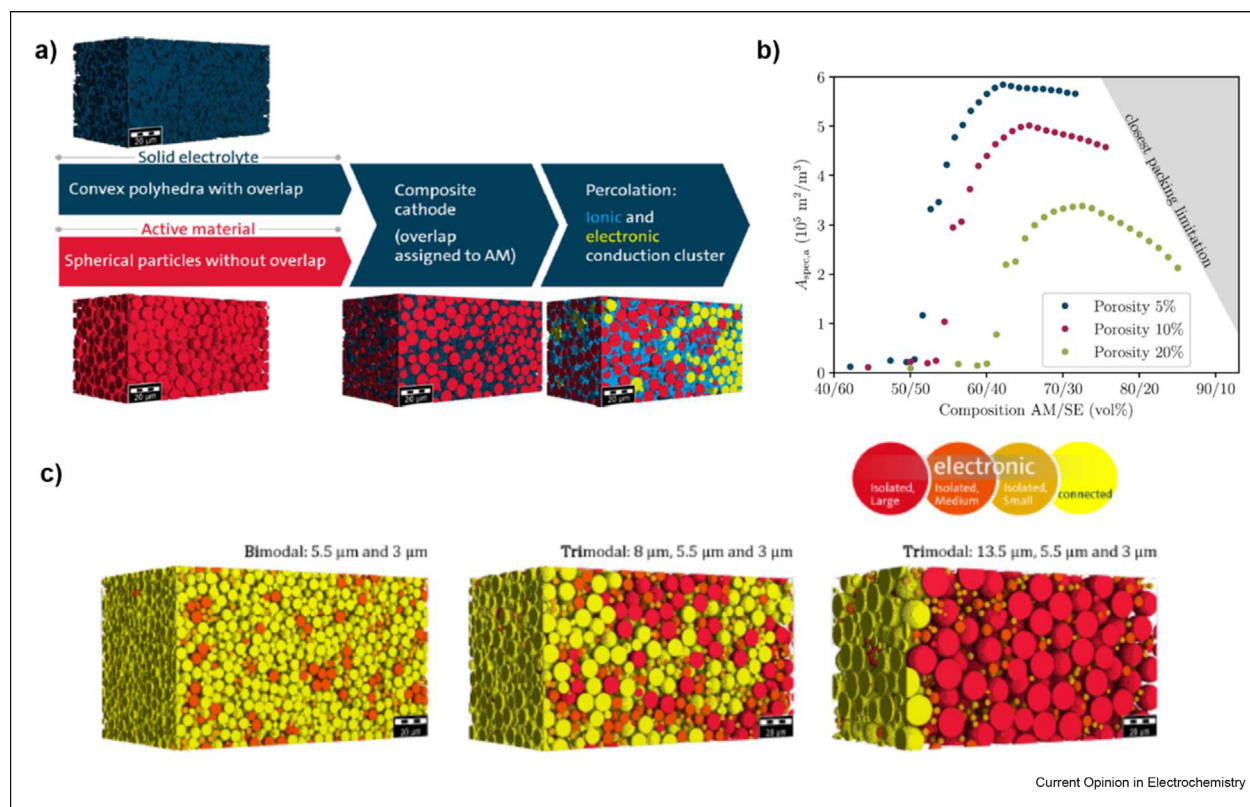
Fathiannasab et al. used tomography to demonstrate the limitations of the 1D modeling approach to model the electrochemical behavior of an ASSB cell [16]. The authors compare the discharge curves calculated by using a homogeneous 1D model with a heterogeneous 3D model developed using CT, demonstrating that although the predicted voltage profiles are almost identical. The 3D model predicts higher ohmic losses because of heterogeneities caused by the lithiation process at high rates particularly. Furthermore, the compression of the cathode electrode is found to decrease the void volume fraction and improve the capacity since it facilitates lithium-ion transport through the SE.

To study the composite electrodes in 3D, the easiest approach is to model their microstructure assuming particles as spheres and artificially construct the electrode by randomly packing the spheres in a given volume until reaching the experimental porosity or the volume

fraction allowing different degrees of overlapping [17]. The process is repeated several times for different random seeds and the average microstructure is retained. Inputs for such approaches are then conducted again for the PSD for the different materials present in the electrode corresponding to the volume fraction and porosity. One of the most popular tools in constructing these electrode structures *in silico* is the commercial software GeoDict®. INNOV is a free-of-charge alternative developed by our group within the ARTISTIC project, which only requires a standard Matlab® license to be used [18].

Following the GeoDict®-based approaches, and based on percolation theory, Bielefeld et al. studied the impact of carbon-free composite electrode formulation (AM/SE ratio), porosity, particle size and electrode thickness on the formation of ionic and electronic percolation networks (Figure 3) [19]. The model is built from small spherical AM particles and polyhedral SE particles, where the conduction clusters are calculated according to the percolation theory. It was found that the small AM particles enhance the effective electronic conductivity,

Figure 3



(a) Bielefeld's et al. [19] modeling approach of all solid-state battery composite electrodes (active material [AM] particles in red, convex polyhedra as solid electrolyte particles in dark blue, ionic conduction clusters (light blue) and electronic conduction clusters (yellow)); (b) calculated specific active area of interface between the clusters of ionic and electronic conduction at different formulations and porosities for 5- μm AM; (c) multimodal AM with different particle size distributions and the resulting electronic conduction clusters at 55 vol.% of AM fraction (reproduced with permission from [19]).

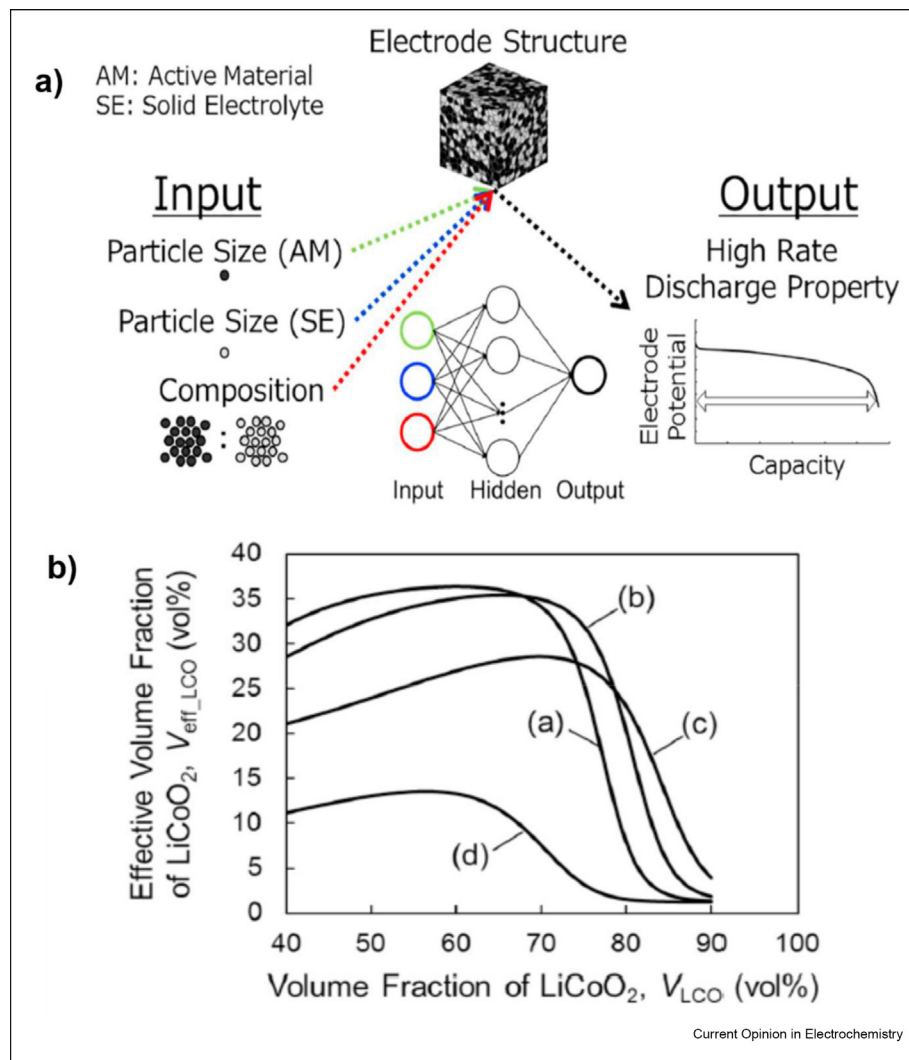
offering a high surface area and a higher number of possibilities to connect particles with each other. However, the authors underline that a high surface area may also enhance aging mechanisms at the AM/SE interface and consequently negatively influence the overall cell performance. On the other hand, porosity is found to strongly impact both ionic and electronic conduction. Based on this, the authors encourage experimentalists to systematically measure the porosity for the sake of comparability with experimental studies. The model is able to detect the impact of the electrode thickness on the effective electronic conductivity only for thin electrodes. Furthermore, ideal compositions that ensure good ionic and electronic conduction at given porosities are identified. The approach is chemistry-neutral, but the authors discuss possible guidelines to optimize $\text{Li}(\text{Ni}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1})\text{O}_2$ (NMC811) with lithium thiophosphate electrolytes. The model still represents a generic approach for microstructure optimization in ASSB. However, it was upgraded to involve NMC811 and sulfide SE to inspect main microstructural challenges in later studies.

Bielefeld et al. extended their work by incorporating a binder in their composite electrode microstructure analysis [20]. More precisely, the authors studied the impact of binder content, AM particle size, and porosity on the effective ionic conductivity and a tortuosity factor. It was found that the binder, even when added in small amounts, negatively influences the ion transport paths and the active surface area available for lithium insertion. Similar conclusions have been found by us in the context of LIBs by using a 3D resolved model [21]. Moreover, the authors found that the AM particle size can pose a trade-off between ionic and electronic conduction. While small AM particles offer high surface area and good electronic percolation as well as short lithium diffusion paths within the AM, they can also impede ionic conduction as they lead to tortuous ionic transport paths. Increasing their size improves ionic conduction within the electrolyte particles, which decreases the overall electronic percolation, which can be solved by adding carbon particles. Interestingly, they suggest using multimodal particle sizes to control the ionic tortuosity, while maintaining short diffusion paths within the AM. The same group extended their previous work by developing a mathematical model coupled with three other geometrical models to assess the influence of the electrode microstructure on ASSB performance [22]. The mathematical model simulates the charge transport within three different 3D-resolved geometrical domains. It features NMC811 cathode AM and $\text{Li}_6\text{PS}_5\text{Cl}$ SE as two spatial domains having two interfaces (AM/SE and SE/Li metal) in between them. The mathematical model describes a time-dependent electrical potential (vs. Li^+/Li) and concentration distribution of lithium-ion within the simulated 3D geometrical models. The three geometrical models were

created to study the influence of residual voids within the electrode, different PSDs, and input parameters dependent on state of charge (SOC) for the validation of the mathematical model. These geometrical models are as follows: *i*) a cathode AM microstructure model with particles reconstructed using GeoDict® to have an ideal interparticle connectivity. The AM microstructure is used to validate the mathematical model results by comparison with the experimental data resulting from a typical ASSB cathode microstructure. Thereby, lithium diffusion is highly dependent on the SOC of the NMC811 AM. The simulations demonstrate as well that the charge transfer is also dependent on the NMC811/ $\text{Li}_6\text{PS}_5\text{Cl}$ interface and significantly influence the cell power. Thus, proper SOC dependent inputs should be employed in ASSB simulations whereas parameters from literature must not be carelessly relied on, *ii*) a model consisting of one AM particle surrounded by SE particles. This model shows that an increase in the size of AM particle hinders the lithiation/delithiation rate due to longer percolation pathways. *iii*) a cone-type microstructure model where clusters of AM and SE are shaped like cones in contact to each other to limit the effect of the tortuosity and increase the ionic conductivity. These models are considered as an in-depth computational study that links multiple microstructural aspects with the output electrochemical performance. Further improvements can be done by incorporating the influence of manufacturing on the used microstructures.

Yamakawa et al. reported an interesting computational methodology to unravel correlations between power density, the volume fraction and the particle size of AM (LiCoO_2) and SE particles [23]. A wide range of 3D-resolved cathode electrode microstructures is generated using a similar approach to the studies covered previously (random packing of spheres within a simulation box) complemented by a phase-field modeling approach aiming to consider the effects of sintering. The phase-field approach depends on lowering the contact surface energy between materials which translates into a set of Cahn–Hilliard and Allen–Cahn equations already used in the literature to simulate phase changes in LIB materials [24]. Then, the lithium diffusion in the AM and the electronic and ionic electrostatic potential distributions are solved in three dimensions to assess the electrodes' capacity at a high discharge rate, using the finite volume method (herein called *deterministic model*). A machine learning (ML) model (artificial neural network) is used to unravel the link between the microstructure parameters and the high-rate capacity. The results of this study show that the size ratio between AM particles and SE particles impacts the discharge rate, which can be explained by the lithium-ion percolation through the electrolyte particles providing quantified guidelines for AM particle/SE particle size ratio choices for optimal electrochemical performance (Figure 4). The importance of this study is

Figure 4



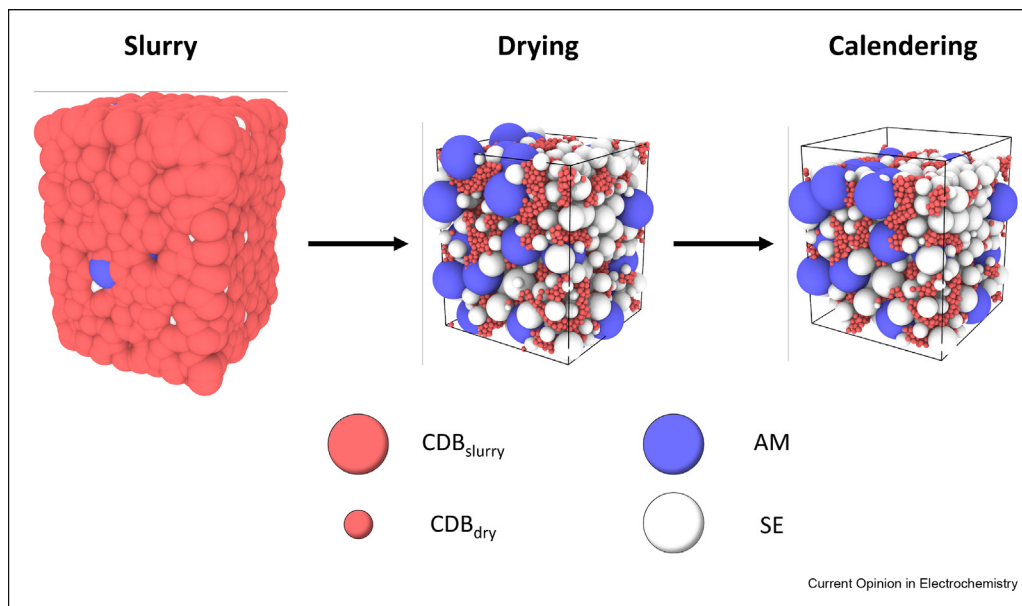
(a) Yamakawa's et al. [23] approach combining deterministic modeling and machine learning; (b) calculated high-rate capacity curves (effective volume fraction in the figure refers to the lithiated volume fraction of LiCoO_2 at the end of discharge) against the volume fraction of LiCoO_2 obtained from the machine learning model. The diameters of particle (a), (b), (c) and (d) are $d_{\text{LiCoO}_2}:d_{\text{solid electrolyte}} = 0.8 \mu\text{m}:0.8 \mu\text{m}$, $d_{\text{LiCoO}_2}:d_{\text{solid electrolyte}} = 2.3 \mu\text{m}:0.8 \mu\text{m}$, $d_{\text{LiCoO}_2}:d_{\text{solid electrolyte}} = 3.8 \mu\text{m}:0.8 \mu\text{m}$, and $d_{\text{LiCoO}_2}:d_{\text{solid electrolyte}} = 0.8 \mu\text{m}:3.8 \mu\text{m}$, respectively (reproduced with permission from [23]).

that it sheds light on a new methodology to investigate the influence of the properties of the AM and the SE on the discharge curves. However, it can be enriched by having electrode microstructures obtained experimentally to understand more the effects of the manufacturing on the microstructure and the resulting power density and conductivity.

Another interesting study is the one of So et al. on plastic deformation of the ASSB composite electrodes resulting from the mold pressure during fabrication [25]. They developed a bottom-up approach performed over three simulations in the following order: annealing, aggregate settling and cold pressing. The annealing step mimics the phenomena taking place during ball milling

to produce aggregates of SE particles. Then, the aggregates settle for the system to be in equilibrium under the effect of gravitational forces and Newton's laws of motion. Finally, the last step of this modeling approach is a 3D discrete element method (DEM) model to study the rearrangement of the particles at room temperature that is due to the fabrication pressure and stress localizations. This model is an upgrade to their previous one in 2D [26] to simulate the mold pressure impact during the fabrication. The model is used to calculate tortuosity factors, and thus the relative ionic conductivity. However, the authors ignore the grain boundary resistance among the SE particles in their model which, in fact, accounts for a high ionic resistance in ASSBs depending on the results of an experimental study based

Figure 5



Plausible approach to adapt the ARTISTIC physics-based models for the simulation of the manufacturing process of all solid-state battery electrodes (indicative relative size of particles considered is provided, CBD stands for carbon binder domain).

on the same assumption [27,28]. They suggest that grain boundaries among SE particles vanish at the particle contact due to the plastic deformation happening during the cold pressing. Thus, their model is accurate enough if the ionic conductivity is only dependent on the porosity and tortuosity factors. Even though it is difficult to distinguish the bulk from grain resistivity due to the high conductivity in sulfide SEs, this assumption cannot be taken for granted since grain boundaries are assumed to be the main reason behind the high resistivity in different types of SE, accounting to several experimental and simulation studies [29,30]. Even though this model is one of the first models to computationally investigate the effect of the mold pressure during fabrication, it lacks the calculations of the electrochemical behavior that should be compared with and validated by experimental studies.

Conclusions, opportunities, and challenges

3D microstructure-resolved continuum models of ASSBs have been emerging recently with the goal of understanding the relationships between electrode formulations and microstructure, effective electronic and ionic conductivities, and electrochemical performance. These are built on electrode microstructures generated either stochastically or (to a lesser extent) from X-ray computer tomography characterizations. While they have proven to be valuable tools to understand the effects of individual microstructural parameters on macroscopic observables, the complex interplay of different physical phenomena in ASSBs remains to be

fully captured. Current models lack a dynamic description of the degradation mechanisms in 3D and the on-the-fly coupling between electrochemistry, transport, thermo-mechanics, and the different aging mechanisms. Degradation mechanisms such as the effect of AM volume changes, cracking, loss of contact between AM and SE particles as well as thermo-mechanical stresses are crucial phenomena that need to be described in the future. Such models will require robust numerical methods to handle complex time-dependent multi-physics couplings in 3D.

Artificial intelligence (AI)/ML is blooming for applications in the LIB field [31,32], such as autonomous materials discovery [33], data mining from a large number of publications [34], optimization of recharge strategies [35,36], material characterization [37], battery recyclability optimization [38], electrode manufacturing optimization [39], accelerating parameterization of physical models [40], and battery aging prediction [41]. The observed impact of AI/ML on LIBs points at many opportunities to extend its use to ASSB applications in the near future. Notably, it would be an adequate tool for the unravelling of the influence of numerous formulation and manufacturing parameters on the electrode microstructure and performance of ASSBs. Additionally, the use of surrogate models that allow to greatly accelerate simulations, or smart sampling of the parameter space (both employing AI tools), bring about the potential to find optimal manufacturing parameters for given application requirement.

Nevertheless, to apply these techniques to ASSBs, large sets of high quality and reproducible data are required. We can speculate that the existing scientific publications may already fulfil these requirements. In that sense, Kuniyoshi et al. have reported an automated machine reading system for extracting the synthesis processes of materials for ASSBs available in the scientific literature [42]. The authors defined the representation of the synthesis processes using flow graphs and created a data corpus from the experimental sections of 243 papers. The automated machine-reading system is developed by a deep learning-based sequence tagger. The script automatically creates synthesis graphs that represent the synthesis process of ASSBs in text. However, this preliminary work faces the issue of data incompleteness in literature. This issue has recently been pointed out in a text mining study of 13000 papers about LIBs and sodium-ion batteries, calling for the standardization of data produced both from experiments and simulations [43].

Lastly, there is a remarkable lack of computational tools aimed at predicting the influence of manufacturing parameters on the microstructure and electrochemical performance of ASSB electrodes. The modelling of these processes involves the consideration of the various relevant physical phenomena involved in each step. For these, the complex interactions between the different materials at the micrometer scale has to be accounted for. The challenge then becomes to account for vastly different particle sizes and material deformability in the same mixture. This applies to both dry and wet processes. Each of these presents its unique challenges. In the former case, grain interactions, particle deformation and cracking have to be considered, while in the latter, partial solubility of the components, suspension dynamics and stability, and viscosity dependencies are important. Such kind of models are crucially needed to help overcoming the challenges that ASSBs face regarding large-scale production [44,45], in particular in the view of complex interdependencies between parameters across different processing steps [46]. Furthermore, the fully 3D-resolved electrochemical models of ASSBs reported so far still present two main drawbacks. The first is a lack of systematic comparison between simulation results with experimental measurements under a wide spectrum of formulations (*i.e.* AM/SE weight% ratio within the electrodes). Secondly, to generate electrodes microstructures, most cases either use CT, which require complex data acquisition and treatment procedures, and/or use stochastic generation of microstructures.

Our ongoing ARTISTIC project [47] is the first one of its kind aiming to develop a digital twin of the entire LIB manufacturing process [48]. Such digital twin combines computational physical models at different scales

simulating each of the manufacturing steps: Coarse Grained-Molecular Dynamics (CGMD) to predict in three dimensions electrode slurries as a function of their formulation and solid-to-liquid ratio [40]; CGMD to simulate in three dimensions the coating and the drying [49,50] and to predict the influence of the solvent evaporation rate on the electrode microstructure [51]; DEM to predict in three dimensions the influence of the calendaring pressure and temperature on the resulting electrode microstructure [52,53]; Lattice Boltzmann Method to simulate the electrolyte filling process in three dimensions and to predict the electrode wettability [54,55]; and dynamic 3D-resolved continuum models to predict the electrochemical response of the predicted electrode microstructures [56–59]. Such a digital twin also integrates a set of ML models that accelerates the physical models' parameterization, assesses experimental data and originates surrogate models with lower computational costs [39,60,61] that have been used by us for electrodes optimization and inverse design of manufacturing parameters [62]. Regarding the latter, the ARTISTIC project high-fidelity physics-based models are being used to generate synthetic data that can be used to augment experimental data sets acquired in our LIB pilot line. The use of ML on these synthetic and/or experimental datasets allows deriving surrogate models which are embedded in optimization algorithmic loops to predict which manufacturing parameters (*e.g.* slurry formulation, drying rate, calendaring pressure) must be adopted in order to maximize and minimize multiple electrode properties simultaneously (*e.g.* tortuosity factor, density, conductivity, surface heterogeneity) [62]. Some of our ML models were recently adapted to the design of solid-state separators [63]. Similarly, to our recently published perspective on the manufacturing simulations of lithium-sulfur batteries [64], we believe that the ARTISTIC project digital twin can be adapted for the simulation of the manufacturing (wet and dry) processes of ASSB electrodes. The developed 3D-resolved electrochemical models can also be adapted for the simulation of the mechano-electrochemical behavior of the ASSB electrodes, allowing to capture the link between manufacturing process parameters and performance. This adaptation could follow a workflow such as the one presented in Figure 5. This work is currently being carried out, with results that will be disclosed by us soon [65].

The continuous increase in the need for energy storage solutions is on track to surpass the capabilities of existing technologies. ASSBs are seen as a promising candidate to partially fulfil these high energy and power requirements. While ASSB technology still presents many drawbacks, microstructure-resolved modeling can pave the way toward an accelerated optimization of ASSB electrodes and manufacturing procedures to reach maturity for a faster adoption and integration of the technology in commercial applications.

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

No data were used for the research described in the article.

Acknowledgements

M.A., V.S., and A.A.F. acknowledge the support by Umicore Specialty Powders France within DESTINY COFUND PhD Program, which has received funding from European Union's Horizon 2020 research and innovation program under the Marie Skłodowska-Curie (grant agreement No 945357). A.A.F. acknowledges the European Union's Horizon 2020 research and innovation program for the funding support through the European Research Council (grant agreement 772873, "ARTISTIC" project). A.A.F. and F.M.Z. acknowledge the European Union's Horizon 2020 research and innovation program under grant agreement No 957189 (BIG-MAP). A.A.F. acknowledges Institut Universitaire de France for the support.

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