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Enabling Quality-oriented Process Development for sulfidic All-Solid-State Battery Cathodes

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Abstract

After major advances in material research throughout recent years, the industrialization of all-solid-state batteries now depends on the development of cost-effective production technology for novel materials and components. To enable a fast production scale-up and complex process interdependency handling, production engineering needs a quantitative evaluation and comparison approach for manufacturing strategies and process parameter settings. To address this challenge, we derive microstructural quality criteria from specifications at the product-level such as driving range and charging speed of battery electric vehicles. These range from porosity and agglomerate density on a macroscopic level to microscopic properties such as pore size distribution and particle contacts. By listing comprehensive characterization methods, the work enables engineers to efficiently evaluate these criteria. Experimentally applying the proposed approach, the influence of different mixing process parameters is analyzed. Thereby, sulfidic composite cathodes manufactured in a scalable procedure are used as samples.

Keywords

All-solid-state battery; Battery production; Process development; Quality criteria; Electrode characterization

1. Introduction

A central issue in present-day society is the search for sustainable energy storage solutions to counteract climate change and accelerate the electrification of a wide range of products [1–4]. Due to their beneficial storage capabilities, batteries gained significant importance in the process, and forecasts predict a rapid increase in global demand in the coming years [5–7]. The strongest driver of this development is the electrification of vehicles, which requires a particularly large amount of high-quality battery cells [8, 9]. Today, the majority of these cells are conventional lithium-ion batteries (LIB) since they possess advantageous storage properties resulting from their rapid development in recent years [10–12]. Despite multiple approaches to further improve the LIB capacity by advancing the components' materials, the unsurpassable physiochemical LIB storage potential will be reached soon [12, 13]. Therefore, the only way to meet the globally growing demand is the development of novel battery generations [12, 14–19]. Among various technologies being pursued, the all-solid-state battery (ASSB) stands out in particular [12, 20–23]. ASSBs possess a dense separator made from a solid-state electrolyte featuring high mechanical stability counteracting dendrite growth at its anode interface [13, 23, 24]. This enables the use of lithium metal anodes, significantly reducing the volume and weight of the battery cell [13, 23, 25]. As a result, an increase

in energy density of up to 70 % regarding the volume and up to 40 % regarding weight is predicted [12, 20, 23]. Furthermore, the solid electrolyte possesses slower calendrical aging, a higher intrinsic safety, as it cannot ignite, and in addition, is superior to the liquid electrolyte in terms of ionic conductivity [20, 23, 25, 26].

ASSB technology can be subdivided by the materials used as electrolytes [27–30]. Current research focuses on the application of polymer, oxide, and sulfide materials [31–35]. This contribution focuses on the latter, as it possesses various advantages over polymers and oxides [21, 36]. These include the electrolytes' high ionic conductivity [31, 37, 38], as well as room temperature processing in routes already established in LIB production [20, 36–38]. However, due to the low maturity of the technology and especially its currently unavailable production capabilities, sulfidic ASSBs have not been utilized in products today [39, 40]. The industrialization and application of the sulfidic all-solid-state battery now depend on the development of industrial-scale production [12, 24, 36, 38]. To support this aim regarding the sulfidic composite cathode, we derive product-specific quality criteria to systematically guide process studies toward high-quality components.
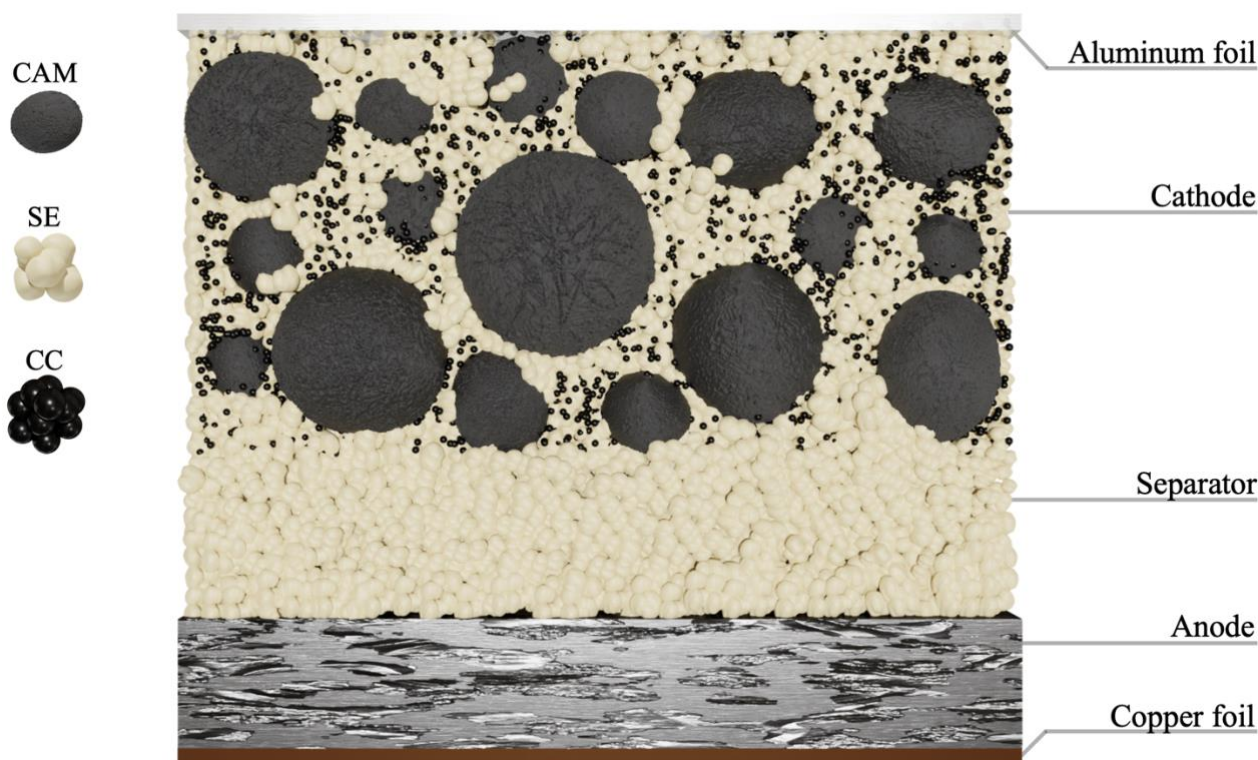

Figure 1: Schematic top-view of an ASSB galvanic cell containing cathode active material (CAM), solid electrolyte (SE), conductive carbon black (CC), SE separator and lithium metal anode.

2. Sulfidic all-solid-state battery and composite cathode

A sulfidic ASSB can be divided into multiple galvanic cells which represent the smallest electrochemically operational unit. Such a galvanic cell is depicted in figure 1. It consists of an anode, a composite cathode, and a separator in between [24, 36, 41]. While the electrodes provide the host structure for lithium-ions enabling electrical energy storage, the separator serves both as electrical insulation and an ionic connection between them. During battery charging, ions move from the composite cathode through the separator and deposit onto the anode. Simultaneously, electrons flow from the composite cathode to the anode, driven by an electrical charger outside the cell. During discharging, respective directions are reversed and a product consuming electrical energy is powered.

The sulfidic all-solid-state composite cathode is composed of an aluminum current collector for electrically contacting the electrode, lithium-ion storing cathode active material (CAM), conductive carbon (CC) improving electrical conduction within the cathode, a polymeric binder setting mechanical properties, and sulfide solid electrolyte (SE) [36, 38]. The latter is new to battery technology and specific for this type of ASSB. The SE is an ion conductor and enables lithium ions to move between the electrodes during the charging and discharging of the cell. Due to the fact, that the composite cathode is the largest and heaviest component of a galvanic cell, it possesses high influence on an ASSBs' energy density. The same correlation exists in terms of power density, as ion-conducting pathways are most critical inside the cathode. It can thus be concluded, that harnessing the full potential of sulfidic ASSB cells is only possible by managing the establishment of quality-oriented production technology for its composite cathode.

The identification of suitable manufacturing routes, stabilization of individual processes, and fine-tuning of their parameters are the focus of the composite cathode production scale-up [42–44]. According to the current state of research, slurry-based production processes already in use for manufacturing conventional electrodes may be applicable [36–38]. These include mixing, coating, drying, and calendaring. The major difference and novelty to production technique compared to LIB cathode production is the addition of the electrolyte as one of the cathodes' integral constituents at the beginning of the production process [12, 13, 45, 46]. As its production-related properties are unknown, the transfer of expertise from manufacturing LIB cathodes is limited. Accordingly, comprehensive studies are necessary for establishing suitable processes.

To provide a basic understanding of the composite cathode design choices, table 1 contains the correlation between the cathode constituents and an ASSB cells' energy and power density. The former correlates product characteristics such as the driving range of an electric vehicle, the latter influences properties such as its acceleration and charging speed.

Table 1: The influence of component-level sulfidic composite cathode design choices on product-level vehicle performance, like charging speed or acceleration and driving range is shown. Therefore, the influence of the design parameter's increase to the respective component-level quantity is indicated by ↑ (increase) or ↓ (decrease).

Criterion ↑	Function	Energy density	Power Density
CAM|SE ratio	Higher lithium-ion storage capacity	↑	↓
Loading	Shorter lithium-ion pathways	↓	↑
CC share	Higher electric conductivity	↓	↑
Binder share	Lower weight and volume of inactive material	↓	↓

The CAM|SE ratio describes the relative amount of cathode active material to solid electrolyte in the composite cathode. Adding more lithium-ion storing CAM and simultaneously reducing the solid electrolyte share will lead to an increase in energy density [45]. For an increase in power density, the ratio should be reduced to provide more ion-conducting electrolyte and a lower flux density in the composite cathode [22, 56]. This enables faster ion transport and therefore reduces charging and discharging times [38]. However, varying the volume fractions beyond certain thresholds may lead to either an excess of unused CAM or stagnation in power density [22, 45]. The additive shares relate to conductive carbon which provides electrical conductivity and binder enabling mechanical stability [36, 38]. Since both materials do not contribute to lithium-ion storage, their share should be as low as possible in terms of energy density increase. However, to boost power density, the proportion of CC should be increased to ensure sufficient electrical conductivity, correlating to simultaneously increasing ionic conductivity [47]. Through a cathodes' loading, the storage capacity available per unit of area is influenced. The higher the loading, the thicker the

component. This leads to an increase in energy density due to the fact, that fewer separators and current collectors are incorporated given a certain cell geometry. However, since thicker electrodes result in longer ion pathways, loading should be reduced to increase power density [23, 45].

3. Microstructural quality criteria for composite cathodes

To support the development of production technology for sulfidic composite cathodes, we provided orientation for process engineers understanding of sulfidic composite cathodes regarding the influence of various constituents in the previous chapter. For a certain composite cathode composition, both energy and power density are highly influenced by production processes as they set details of the microstructure determining its performance in an ASSB [22, 45]. Consequently, the task is to optimize both criteria by finding suitable production processes and their parameters.
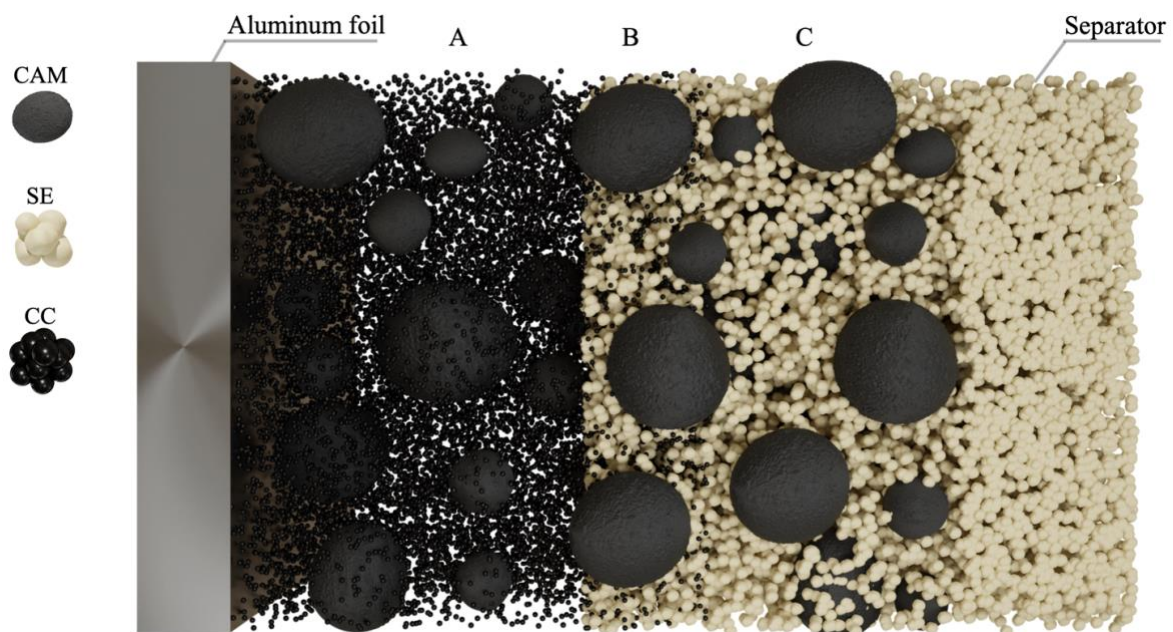

Figure 2: Schematic representation of the particle distribution within the sulfidic composite cathode. For illustration, not all constituents are shown. A) Conductive carbon black (CC) enables electron conduction between the cathode active material (CAM) particles and the aluminium conductor foil. B) CC, CAM and solid electrolyte (SE) are shown. C) SE particles enable ion transport between CAM particles and the separator.

To address this challenge new to production technology, battery cell performance determining cathode microstructure quality criteria are derived (Table 2), providing the baseline for quantitative comparison of their influence. They enable the quality-oriented identification of suitable processes and their parameters along the entire process chain. In the following, the quality criteria are listed, and explanation of their principles of action and application is given.

Porosity

A composite cathodes' porosity is defined as the pore volume in relation to the total volume [48, 49]. As described earlier in LIB technology these pores are infiltrated by the liquid electrolyte, however, for ASSB production, the electrolyte material is already added to the cathode slurry [36, 38, 50–52]. Thus, the remaining porosity represents the share of empty volume not contributing to a battery's functions, reducing energy and power density. Therefore, porosity is associated with the lack of particle contacts, being relevant for most of the derived quality criteria. Via complex to measure but precise mercury intrusion porosimetry (MIP) spectra, the quantification of pore sizes and frequency of appearance is enabled. These can further be interlinked to the pores' causes. In contrast, porosity-determining volume and mass measurement (VMM) is

an easier way but provides no details regarding the pore size distribution. Reducing the porosity of the composite cathode to a minimum, while keeping good particle distributions is one of the main goals in composite cathode production as it significantly influences the ASSB cells' performance [45, 53].

Table 2: Sulfidic composite cathodes quality criteria applied for the evaluation of production processes and their parameters are listed. The feature optimization (Opt.) describes if a criterion is to be maximized (↑) or minimized (↓) to enhance quality. A relatively easy achievable process adaptation for a criterion's fulfillment (ful.) is indicated with +, a difficult one with -. In addition, features are classified about their cell-level functions, improving (↑), decreasing (↓), or not affecting (o) electric or ionic conductivity. The last column lists possible characterization methods of each quality criterion and gives orientation for its measurability (mea.), which is indicated by high (+) and low (-).

Quality criteria				Function		Characterization	
Material	Criterion	Opt.	Ful.	Electric	Ionic	Method	Mea.
-	*Porosity*	↓	-	↓	↓	VMM, MIP	+
CAM	*Homog. of distr. CAM*	↑	+	↑	↑	SEM, EDS	+
SE	*Contact SE|CAM*	↑	-	o	↑	SEM, MIP	+
	Contact SE|SE					SEM, MIP	+
	Homog. of distr. SE					SEM, EDS, MIP	+
CC	*Contact CC|CAM*	↑	+	↑	o	SEM	-
	Contact CC|CC					SEM, MIP	-
	Homog. of distr. CC					SEM, EDS, MIP	+

Cathode active material

The CAM particles are starting point of electron and ion pathways through a cathode's microstructure [56]. That is why their distribution and connection are influential to the electrical and ionic conductivity of composite cathodes. The homogenous distribution of CAM enables SE and CC particles to fill gaps between the larger CAM particles. This counteracts the formation of CAM particles separated from the conduction cluster and therefore ensures high CAM utilization rates. Furthermore, the homogeneous distribution, lowers both, ion and electron pathway length and flux density, ultimately decreasing resistances [45, 53-55]. Process wise the distribution of CAM particles is rather easy to achieve but is linked to SE particle distribution and can therefore be aggravated. To determine the criterion, scanning electron microscopy (SEM) can be used. CAM particles are easily identified on top-view images of cross-sections, the estimation of their distribution can be conducted qualitatively by humans. If needed Energy-dispersive X-ray spectroscopy (EDS) further simplifies CAM particle identification. Because of its central functional role, the homogeneity of CAM particle distribution is an important quality criterion.

Solid Electrolyte

During charging and discharging, solid electrolyte particles are constituting the conductive element for ion transport. The interrelation between a composite cathode's microstructural geometry on the ionic conductivity is characterized by the tortuosity factor [57]. This describes the ratio of the length of the effective lithium-ion pathway to the shortest possible pathway. It should be as low as possible for an even flux density and low ionic resistance. To increase ion conduction and therefore keep the tortuosity factor low, the active interface area formed by CAM and SE particles contacts are crucial as they allow lithium ions to enter and leave CAM particles during charging and discharging. The more pronounced the contact,

the lower the ionic interface resistance [45, 53-56]. Process-wise, this parameter is very difficult to adjust. Due to differences in the size of the SE and CAM particles, contacts can be approximated by MIP and identified using SEM. In addition to SE and CAM particle contact, individual SE particles must be in good contact to reduce the ionic resistance of the composite cathode [40]. Due to the easily detectable SE particles, the fulfillment of this criterion can be evaluated using SEM images or detected via MIP spectra. Furthermore, unfavorable SE particle distribution leads to increased tortuosity, and thus lowers the composite cathode ionic conductivity. This results in lower power densities through slower ion transfer and lower energy densities because of inactive CAM particle fractions. It can be qualitatively analyzed rather precisely by humans via particle detection on top-view SEM images as well as cross-sections.

Conductive carbon

To increase the composite cathodes' electrical conductivity resulting from CAM particles, graphite-based CC is added to conduct electrons between the CAM particles and aluminum conductor foil [47]. To achieve the lowest possible electrical resistance, the contact between CAM and CC, as well as between different CC particles should be as pronounced as possible. Similar to the distribution of CAM and SE, the homogenous CC particle arrangement ensures sufficient electrical conduction, lowering electrical resistance [57]. Process-wise, the relatively small carbon particles are easily distributable, but can hardly be identified by SEM images or mercury intrusion porosimetry spectra. Accordingly, EDS analysis is used to identify carbon atoms' locations.

4. Procedure for applying quality criteria to production processes

Since knowledge of the production processes is currently unavailable and expertise from the manufacturing of conventional batteries is hardly transferable, an iterative approach must be taken to design suitable processing routes. Thereby, the knowledge of the quality criteria and their characterization plays a central role. Because only through the systematic comparison of their fulfillment, the influence of routes, processes, and parameters becomes quantitatively comparable.

When applying the quality criteria for production development, a total of three stages must be passed.

1. At first, the object of investigation must be chosen. This may be an entire manufacturing route consisting of multiple processes, an individual step composed of various parameters, or the variation of just a single process parameter.
2. Composite cathodes must be produced, and the quality criteria to be investigated chosen. The focus can be on all criteria at once, but of course also refer to only one single aspect, which is determined by the stage of process development. The former tends to be suited for entire manufacturing routes and initial production trials, the latter for the fine-tuning of individual steps and their parameters. After cathode production, the respective characterization methods are applied.
3. In the last step, the evaluation regarding the fulfillment of quality criteria is conducted. The application of this approach is independent of the production scale under consideration. Thus, it suits both the manual manufacturing of both laboratory-scale components as well as the production of large-format composite cathodes in industrially relevant processes.
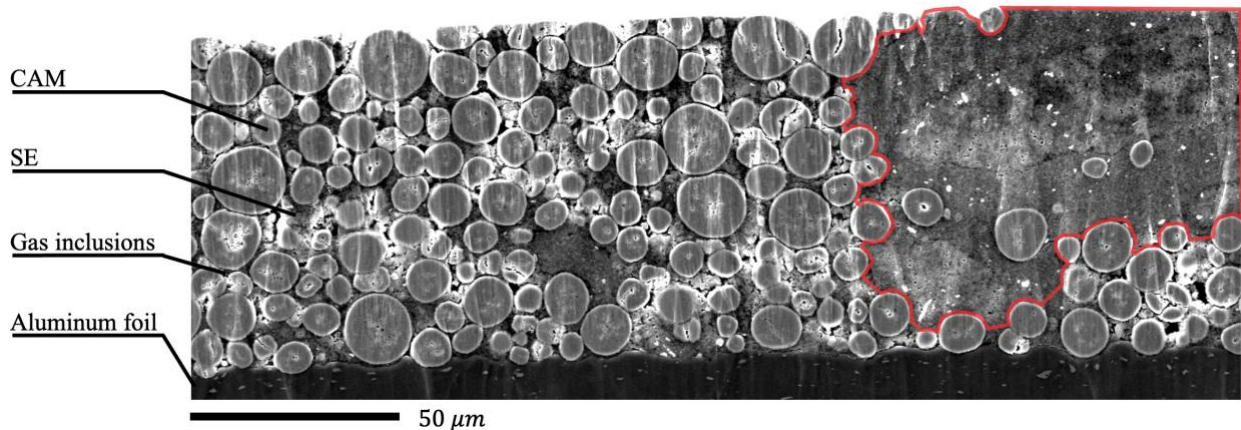
Figure 3: Cross-section of a densified sulfidic composite cathode after processing in a wet chemical route at the Institute of Machine Tools and Industrial Management. On the top right, an SE agglomerate is highlighted in red. The composite cathodes remaining porosity originates from voids between particles.

5. Exemplary analysis of quality criteria for process development

To demonstrate our approach, it is applied for the analysis of a scalable wet chemical process chain. The chain is built from mixing, coating, drying, and pressing steps which are executed in a similar fashion already described by Sakuda et al. [38]. Also, the composite cathodes material composition is adopted.

The aim of the exemplary analysis is the identification of the influence of the mixing duration on the quality criteria porosity and particle distribution before and after the pressing step. Therefore, the mixing duration is set to 2, 32, and 64 minutes, while all other process parameters as well as the material composition are kept constant. After producing uncompressed uand compressed composite cathodes with varying mixing durations, VMM, SEM, and EDS analyses are carried out to analyze the quality criteria.

For the mixing time of two minutes, the resulting slurry possesses large agglomerates. Therefore, it is not further processed. Measuring the porosity using VMM measurements it is indicated that for mixing times of 64 minutes the resulting cathodes porosity before and after compaction is higher compared to 32 minutes mixed composite cathodes. The SEM images show potential explanation in higher SE agglomeration for the longer mixing times. For all process parameter sets, agglomerates are visible. Through the SE agglomerates the CAM and SE distributions are negatively affected, as visible in figure 4 column A. The local SE exaggeration leads to the formation of SE poorer regions, possessing a high CAM share and mediocre CAM|SE contacts. Tough through compaction CAM|SE and SE|SE contacts are improved by SE deformation; distributions are not. SEM images indicate that for the 2 min mixed processing route much worse SE|SE contacts are achieved compared to the other two parameter sets. The CC distribution and contacts can be investigated by the identification of CC particles on SEM images in combination with EDS analysis. The results show a good distribution for CC particles for all three investigated parameter sets, highlighting the rather easy manageability of its distribution during composite cathode slurry mixing. Conclusively the results indicate poor fulfillment of the derived quality criteria for all three cathodes. This is indicated by high porosities and low distribution and contact qualities for CAM and SE, which are expected to lead to reduced ionic conductivities. Though the cause of agglomerate occurrence cannot be deduced, quality criteria can be used for further hypothesis-driven experimentation and evaluation. These could potentially investigate if the agglomerates are induced by the supplied SE materials and give further insights into whether they grow through the mixing process.
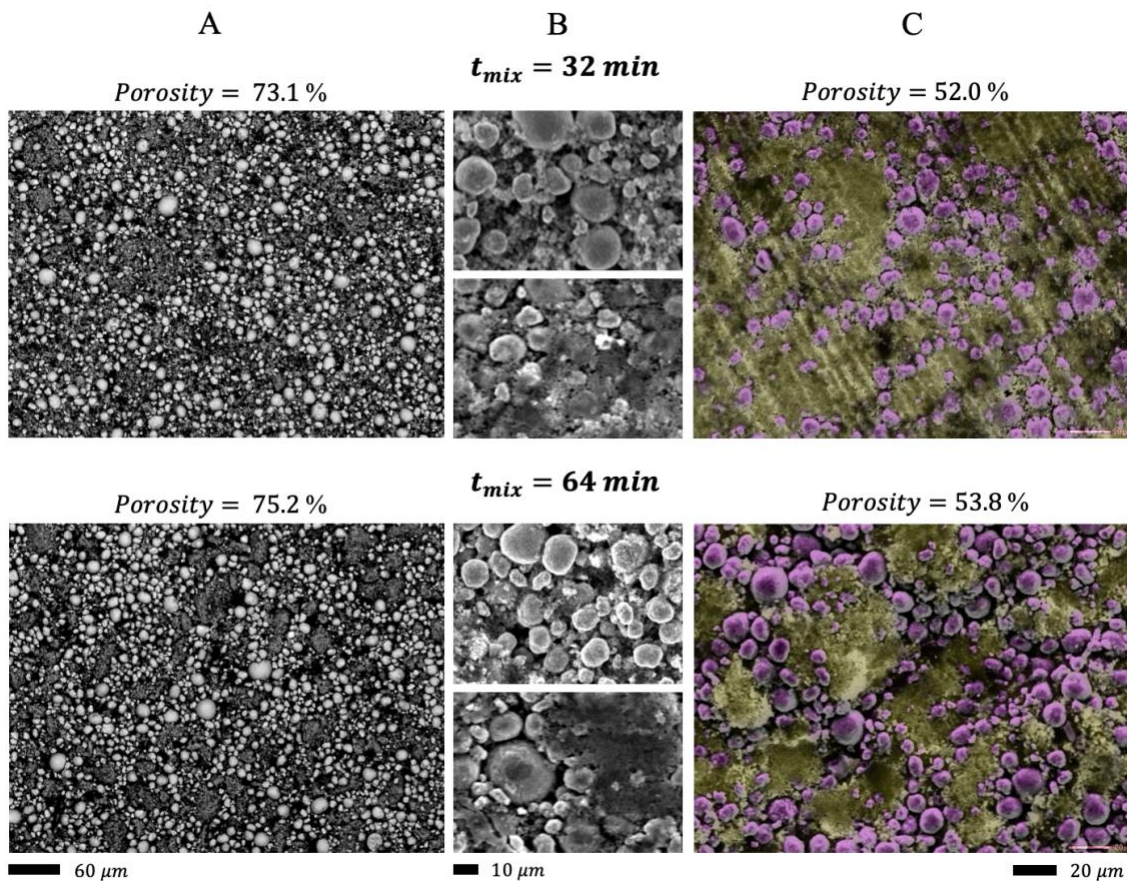
Figure 4: In column A backscatter SEM images of the uncompacted composite cathodes are shown, especially highlighting CAM distribution. In column B, secondary electron detector images indicate the particle contacts, associated with the non-compressed and compressed cathodes. The third column shows EDS analysis indicating the CAM and SE distribution after the compaction step, the active material is marked purple, while SE olive. Porosities are calculated using averaged VMM measurements for 8 samples, scales are introduced per column.

6. Conclusion and outlook

Sulfidic ASSBs represent a promising candidate for the next generation of electrochemical energy storage systems. However, industrial-scale production technology necessary for their application is currently unavailable. To support the identification of suitable manufacturing routes, processes, and their parameters, microstructural quality criteria are presented focusing on composite ASSB cathodes. For each criterion, possible measurement techniques are listed for their quantification. An exemplary use case focusing on the evaluation of the mixing durations influence on selected criteria is described to show the approach's applicability.

Further investigations may focus on improving and automating quantification methods for the proposed quality criteria. In the long term, this could be used as feedback in an automated process parameter optimization loop. In addition, aspects such as the sensitivity of the quality criteria regarding process steps and their parameters may be analyzed.

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Biography

Thorben Prein is a graduate student in mechanical engineering at the Technical University of Darmstadt and is currently working as student research assistant at TUMint. Energy Research GmbH and Institute for Machine Tools and Industrial Management (*iwb*) at the Technical University of Munich.

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