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# Conceptual Design of Solid-State Li-Battery for Urban Air Mobility

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The negative impact of internal combustion engines on the environment is a major concern in metropolitan areas due to the continued rapid growth and high overall level in the number of vehicles, population, and traffic congestion. Electric vertical take-off and landing (eVTOL) aircraft promises a new era for urban regional transportation and air mobility to address the challenges mentioned above. Nonetheless, providing electrical energy storage systems, like batteries, is one of the key issues with such aircraft. Here, the non-flammable technology of all-solid-state Li batteries with high theoretical gravimetric energy is an attractive option. Modelling allows for a knowledge-driven assessment of the potential of this technology. We here used a combination of a pseudo-2-dimensional cell model with a microstructure surrogate model approach to acquire a better understanding of the effect of the cathode microstructure on the internal process limitations. This model is incorporated into a global optimisation algorithm to predict optimum battery size with respect to the dynamic load demand of eVTOL. When carbon black and active materials are premixed, the battery performs better than when solid electrolyte and active materials are premixed, particularly for low amounts of carbon black in the cathode combination, i.e., 5%. Further, results indicate that future electrification of transportation powertrains would necessitate optimising the composition and distribution of electrode components to fulfil the high demands for power and energy density. By enhancing transport through the microstructure and improving the material's intrinsic conductivity, it is possible to significantly increase the effective diffusivity and conductivity of ASSB, and hence the mission range.
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The transportation sector in metropolitan areas is confronted with serious challenges in addressing the growing demand for convenient human mobility while decreasing emissions, enhancing safety, and reducing congestion.[1] Also, traffic congestion and land-use restrictions have limited the use of electrification and automated driving to overcome the issues mentioned above.[2] Hence, electric vertical take-off and landing (eVTOL) aircraft have been suggested as an alternative mode of urban transportation and a potential future market to relieve traffic congestion and reduce commuting times in congested areas.[3] eVTOL brings advantages, such as its light-weight, ease of maintenance, low noise, safety, and the lack of a need for additional run-up space.[4] The main types of eVTOL are rotary-wing cruise and fixed-wing cruise; the former is effective in hover and has less complexity, while the latter has superior efficiency at longer mission ranges.[5] Bacchini and Cestino discovered that choosing the ideal configuration of eVTOL among different eVTOL designs is mainly dependent on the mission.[6] Shamiyeh et al.[7] investigated the mission performance of two eVTOL configurations. They indicated that a multicopter design has a higher transport energy efficiency than a fixed-wing cruise configuration at short distances. Finger et al.[8] presented an initial sizing algorithm using a simple model for urban air mobility with a family of transitioning VTOL aircraft. They reported that future mid-range aircraft would need hybrid-electric propulsion systems and fully electric propulsion systems for short-range missions to avoid the complexity of the hybrid design. In another study, Nathen et al.[9] studied the technological feasibility of an eVTOL aircraft in regional and urban air mobility. They demonstrated that there is a need to develop a battery with higher specific energy to increase mission range. Fredericks et al.[10] also analysed the eVTOL performance by determining the mission range at a constant gross take-off mass. They stated that extremely high-power demands are necessary both during take-off and, particularly, during landing; However, because of the lower voltage and lower state-of-charge during the landing phase, it is much harder to meet the high-power demand during landing than take-off. Pradeep et al.[11] reported that the specific energy of commercially available Li-ion polymer batteries is insufficient for long-distance travel with eVTOL aircraft.

As stated in the literature study above, providing batteries with sufficient performance is a critical obstacle to electrifying such aircraft owing to their limited capacity and poor hover performance. The size and design of the battery in the eVTOL aircraft will have a significant impact on the design outcome, such as the number of passengers and mission range.[12] Besides being capable of producing sufficient energy and power to meet mission demands, batteries in such aircraft must be safe, reliable, have broad environmental capability, and reduce weight penalties. High flammability and risk of thermal runaway in current Li-ion batteries motivate to develop next-generation chemistries with non-flammable nature and long life, such as all-solid-state batteries (ASSB) as alternative electrical energy storage systems for eVTOL. ASSB has attracted interest among researchers due to its high theoretical specific energy and high intrinsic stability.[13] However, ASSBs often exhibit low capacities at high (dis)charge rates due to the weak electrode/electrolyte interface. Possible explanations for the weak ionic diffusion over the electrode/electrolyte interface could be small effective contact area for charge transfer, space charge effects, which change the conductivity by redistributing ions near the interface and weak interphases due to electrochemical and chemical and decomposition. However, it is demonstrated in study by Klerk et al.,[14] that the performance of all-solid-state batteries is insignificantly affected by space charge layers. They examined the role of space-charge layers in all-solid-state batteries and evaluated the interface capacitance and resistance due to the space-charge layer. According to their findings, the thickness of the space-charge layer is roughly in the nanometer regime for common electrode-electrolyte combinations, causing negligible resistance for lithium-ion transfer through the space-charge layer. Further, ASSB currently suffers from a significant amount of non-utilised active material (AM), i.e. low capacity. Electrode composition and structure have a crucial impact on three performance-relevant microstructure parameters, i.e.,

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**Table I. Assumption, C-rate demand, and required mass and capacity of the battery for the entire flight time of multicopter.**[7]

| | | |
|---|---|---|
| Assumptions | Number of passengers | 4 |
| | Constant mission range, km | 25.7 |
| | Required battery pack specific energy, Wh $kg^{-1}$ | 250 |
| Required mass and energy | Maximum take-off mass, kg | 1005 |
| | Battery mass, kg | 215 |
| | Battery pack capacity, kWh | 53.9 |
| Required current rate | C-rate required to hover, 1 $h^{-1}$ | 2.71 |
| | C-rate required to cruise, 1 $h^{-1}$ | 1.54 |

effective active area, and effective electronic and ionic conductivities. Improper mixing of the cathode electrode and electrolyte leads to a reduced surface area accessible for the solid electrolyte, which causes high overpotential and low capacity.[15] However, finding solutions to these restrictions experimentally by developing and assessing a diverse set of design parameters to obtain a well-performing ASSB is both time and resource-consuming. Modelling based on physics promises to determine the optimum design of ASSB designs with respect to these restrictions more time and cost-efficient.

So far, few studies have been conducted on the microstructure modelling of ASSB with physics-based modelling. Cistjakov et al.[16] modelled the influence of cathode particle structures on discharge behaviour in a polymer-based battery with agglomerate and core–shell particles. He demonstrated that cells with homogeneous agglomerate particles have the highest capacity at low C-rates. In contrast, cells with core–shell particles have superior performance at higher C-rates. Laue et al.[17] applied a microstructure model to study the influence of distribution and cathode composition on solid-state electrodes. He showed that cathode distribution and composition of cathode components have a significant effect on the effective ionic and electronic conductivities. He also coupled a microstructure surrogate model for effective microstructure parameters with a pseudo-2 dimensional (P2D) Li-ion cell model with homogenised electrode structures to study the effect of electrode structures more realistically.[18] Extending the P2D model with a microstructure surrogate model allowed for more accuracy in estimating the experimentally observed effect of porosity on performance in comparison with the classical P2D model. In another study, the impact of AM particle size and electrode composition on the ASSB was investigated by Bielefeld et al.[19] using a three-dimensional microstructure model. They expressed that when choosing an AM material particle size, a trade-off between electronic and ionic conductivities needs to be considered. They also applied microstructure modelling to study the ionic and electronic percolation behaviour of cathode composition for ASSB.[15] They indicated that small AM particles are preferable for electronic conduction in the cathode composition with no carbon because they have larger active surface areas and more possibility to create percolating electronic clusters. On the other hand, high surfaces of AM may accelerate degradation processes at the interface of the electrolyte/cathode, lowering cell performance.

Apart from the microstructure modelling of ASSB, there are a few works that have employed P2D models to simulate ASSB. Danilove et al.[20] modelled ASSB with a single-ion conductor electrolyte and provided knowledge on migration processes, concentration profiles, and overpotential contribution within the electrolyte. Their simulations indicated that the solid-state electrolyte has significant transport restrictions, accounting for at least half of the overall overpotential. In another research, Wolff et al.[21] utilised the P2D model to assess the performance of cells with a single-ion conductor and binary liquid electrolytes, finding that both systems are affected by electrolyte conductivity and electrode thickness. Following Wolff's work, our group developed an ASSB model for hybrid electrolytes, including polymer and oxide electrolytes, to estimate the optimal electrode design using a global optimisation algorithm to attain higher specific energy.[22] We found that a hybrid electrolyte based on 12.7 vol% of garnet, i.e. LLZTO, based on the current manufacturing limitations of ASSB, is the best choice. We also provided optimal cell designs without transport limitations for high-energy applications, and high-power and high-energy applications. Our results showed that the optimal identified design for a particular application e.g. for high energy doesn't perform well for higher power applications, and vice versa. We also discussed the main disadvantages associated with oxide-based solid electrolytes including poor contact between the electrode and electrolyte interface, low mechanical flexibility, and poor wettability. These significantly reduce the effective interaction area of the electrolyte/electrode interfaces and sluggish charge transfer between electrolytes and electrodes. To mitigate the above concerns, mixtures of oxide-based electrolytes with polymer electrolytes are used.

Although a few studies have been performed on mechanistic modelling and model-based design of ASSB, and extensive research has been carried out on various designs and battery sizing for eVTOL applications using simple analytical models or equivalent circuit models, no single study exists, that integrates the ASSB with eVTOL based on mechanistic modelling. Therefore, in this study, we use a microstructure-extended P2D-type model to assess possible options and potential limitations when using ASSB for eVTOL. In addition to cathode design, the separator and anode designs can also significantly affect the performance of the battery used in aircraft applications. Our recent study concentrated on the thickness of the separator and various potential materials for it.[22] In the current work, we have applied the separator results from our previous study, and we have given particular attention to the cathode side. Here we examine the impact of different cathode compositions and distributions on the battery performance and propose the best electrode structure. The inclusion of the microstructure surrogate model is used to give a better understanding of the electrode structure and especially the effects of percolation, which cannot be appropriately covered by the classical Bruggeman approach. This model with the identified best cathode structure is then incorporated into a global optimisation approach to determine the optimal design of solid-state electrodes concerning eVTOL C-rate demand. Finally, we give recommendations based on a comparison of the battery performance at the identified optimal and reference electrode design.

## Methodology

In the following, first, the requirements and assumptions for operating the eVTOL with an ASSB are discussed. Subsequently, the microstructure model and the electrochemical model are introduced. Finally, details on implementation, model coupling, and optimisation are given.

***Approach and assumptions.***—We aim to implement a dynamic load demand of eVTOL into ASSB modelling to investigate whether ASSB can deliver appropriate electrical power and energy to meet mission requirements, identify the most promising electrode design, and provide recommendations to increase performance. To this end, we first use the microstructure modelling approach of Laue[17] to determine effective diffusivities, active areas, and conductivities for

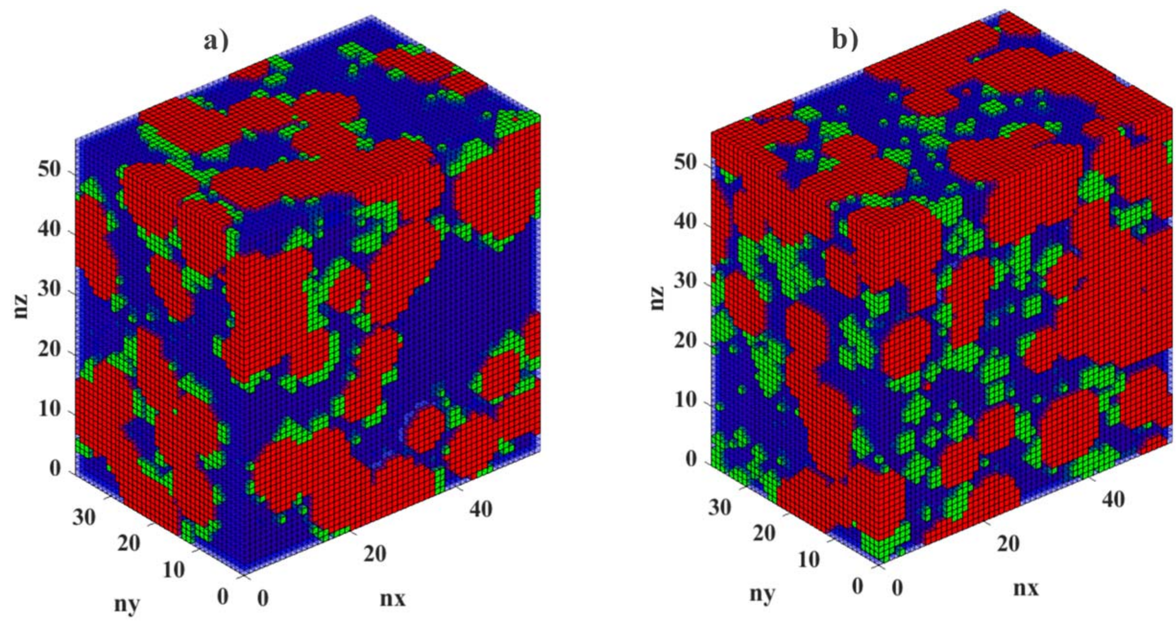


**Figure 1.** Microstructure containing active material (red), electrolyte (blue), and carbon black particles (green) with (a) attached distribution, (b) even distribution.

different cathode structures with the here proposed electrolyte and the AM of the cathode electrode. The dependency of the parameters on electrode composition, which is the output of the microstructure modeling, is cast into polynomial equations of the three performance-relevant microstructure parameters. These are then fed into the cell model for electrode structure optimisation. For identifying the best battery electrode design for eVTOL, we require the dynamic load demand of such aircraft. Shamiyeh et al.[7] calculated the C-rate demand of a multicopter configuration, as shown in Table I, for a constant mission range of 25.7 km and four passengers. We use the same current profile for our investigation. In their research, a specific energy of Li-ion batteries at the pack level of 250 Wh $kg^{-1}$ was assumed, and the battery capacity and mass were determined based on this specific energy. In this study, we apply the same dynamic load demand to ASSB modelling to see whether conventional Li-ion batteries can be substituted with ASSB to complete the mission range while maintaining a specific energy density of 250 Wh $kg^{-1}$ at the pack level. Table I summarises the assumption and required mass and battery capacity, and the current demands for the entire flight time of the multicopter.

For integrating the microstructure parameters into the P2D model for ASSB with respect to the dynamic load demand of eVTOL, the following assumptions for the battery are considered.

- A steady temperature of 60 °C is considered for the ASSB.
- The particle size of the cathode's AM is considered to be uniform.
- The Li metal electrode is modelled as a boundary condition and delivers the quantity of Li-ion that corresponds to the applied current density.
- Degradation effects for calculating the pack level specific energy from the cell level is considered equal to 0.742.[23]
- Microstructure effects and distribution of carbon black or electrolyte are considered via the microstructure surrogate model.
- The properties of all phases are isotropic.
- No voids in the electrode, no space-charge layers at the electrode-electrolyte interface.

Laue et al.[17] employed Li-$FePO_4$ as AM and a solid polymer as the electrolyte in his microstructure model. In the present study, we use NMC811 as AM to achieve higher specific energy and a hybrid oxide-polymer electrolyte. We use a hybrid electrolyte with 12.7 vol% LLZTO, as this configuration yielded the best performance among hybrid electrolytes at a relatively high C-rate, i.e., 1C.[22] The electrolyte properties of this hybrid electrolyte are based on reported experimental data in Ref. 24, which accounts for any impurity or degradation effects present in real material. We employed an experimentally feasible separator layer thickness of 40 $\mu$m for the hybrid electrolyte containing 12.7 vol% LLZTO, which allowed to suppress lithium dendrite growth and protect the cell from internal short circuits according to Ref. 22. It is possible to achieve higher specific energy from hybrid electrolytes by reducing separator thickness, but there is a risk of reduced solid electrolyte mechanical strength, Li dendrite growth and membrane separator fracture. Therefore, we did not investigate the effect of separator thickness on battery performance in this study. A brief description of the microstructure surrogate model and electrochemical modelling of ASSB is given below.

***Microstructure surrogate model.***—Our previous work[17] describes the algorithm for generating electrode structures and subsequently evaluating various distributions and compositions of electrodes for a polymer-based battery. This method is adopted and adjusted here to allow for the modelling of the new chemistry of ASSB, as stated above.

As in our prior work, voxel-based particle architectures are used as an efficient technique for generating shapes with different electrode compositions and distribution of particles and particle shapes. To predict the effective electronic and ionic conductivities of the resulting microstructure, the voxel-based structure is converted into a node-based resistor network. Every node is placed in the middle of each voxel and each connection to neighbor nodes transforms into a resistor. The connector conductivity between nodes is influenced by the present material and particle/particle interfacial resistance.[17,18] Further, the effective conductivity of a full structure can be calculated using the macroscopic voltage drop between the first and last nodes. By counting the area at the interface between the electrolyte and the AM, the volume-specific surface area is determined.

Figure 1 depicts two scenarios for the distribution of the cathode components in this study, similar to Laue's work. In the first scenario, the premixing of carbon black (CB) and AM are assumed,

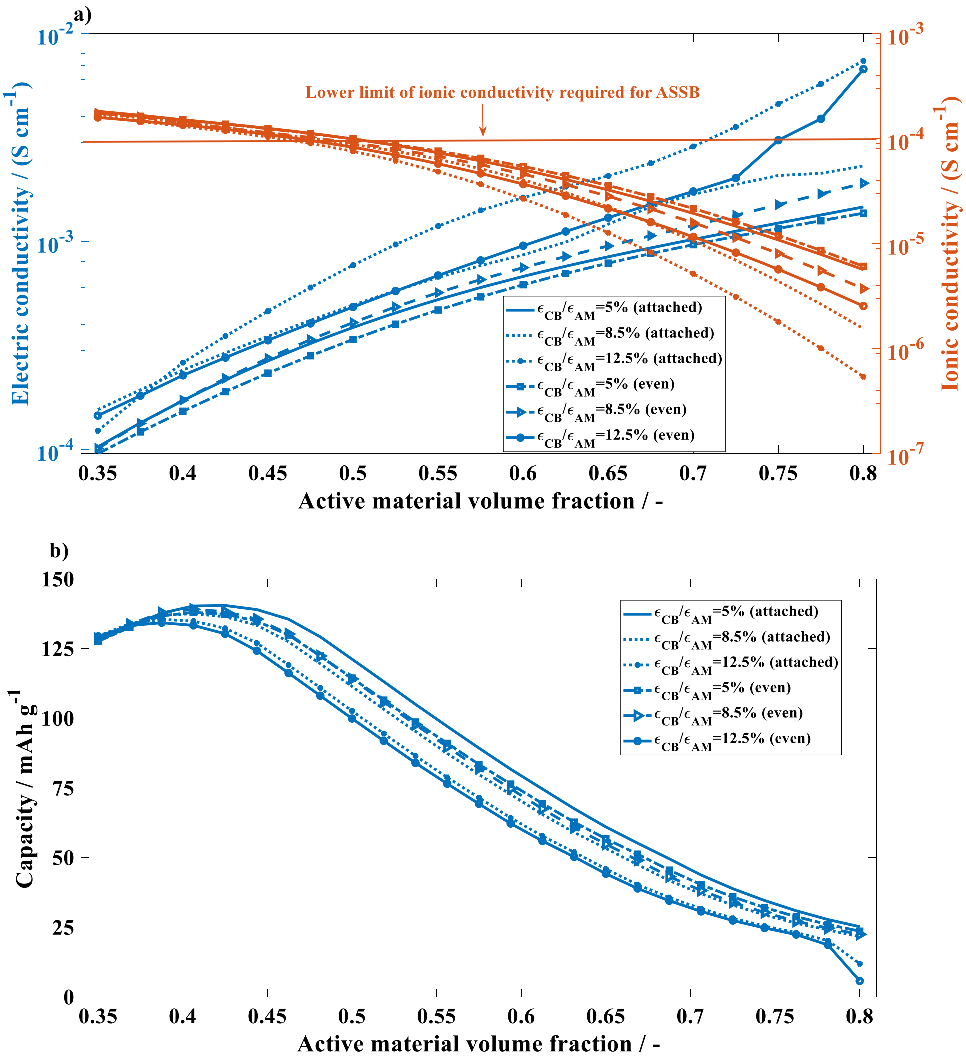


**Figure 2.** Impact of active material volume fraction and cathode structure on (a) effective ionic and electronic conductivity, and (b) the resulting capacity of the battery at 1 C for Li-ASSB with oxide-based hybrid electrolyte, and NMC811 as the cathode. Tables S1 and S2 display the relevant parameters.

thus CB particles are entirely attached to AM. In the second scenario, CB is uniformly distributed within the electrolyte, assuming the premixing of AM and solid electrolyte.

Microstructure modelling is carried out with a mean particle size of 5.85 μm. The edge length of the cubic voxels is 0.531 $\mu$m, and the electrode fragment is composed of 125 substructures, which contain $42 \times 60 \times 60$ voxels, yielding a total of 18.9 million voxels. The electronic conductivity of CB is assumed to be $5\ \mathrm{S\ cm^{-1}}$ [17] and the ionic conductivity of hybrid electrolytes with 12.7 vol% LLZTO $5.63 \times 10^{-4}\ \mathrm{S\ cm^{-1}}$,[22] respectively. The volume fraction of AM changes between 0.35 and 0.8. To examine the impact of the distribution of cathode components on the battery capacity, and thus on the eVTOL performance, we run the microstructure model with three different CB/AM volume ratios, with low, medium, and high amounts of CB in the ASSB cathode electrode. Table S1 in the supplementary material contains the surrogate model, i.e. the empirical polynomials for effective ionic and electronic conductivities and effective surface area, which are derived from the microstructure model with different electrode compositions and distribution. The estimated parameters that correspond to the empirical polynomials for effective transport parameters and effective surface area are also listed in Table S2. Then, for the best-identified electrode structure, battery performance considering the C-rate demand of eVTOL is evaluated. The simulation of the microstructure model is conducted in MATLAB 2020a on the workstation at 2.30 GHz with 384 GB of RAM.

***Electrochemical model of ASSB.***—The microstructure surrogate model is integrated into the P2D model for a realistic evaluation of performance and transport characteristics as a function of the electrode structure. This coupling between the P2D model and microstructure surrogate model will help us to comprehend the electrode structure and, particularly the impacts of percolation, which the Bruggeman relation cannot adequately describe.

The P2D model for modelling Li-ASSB with hybrid electrolytes is described in detail in our prior work.[22] Table S1 displays the governing equations of the P2D model including solid and electrolyte diffusion, solid and electrolyte potential, electrolyte migration, and Butler-Volmer kinetics. Table S2 lists the geometrical, physical, and electrochemical parameters. The finite volume method is used in the numerical calculation of the P2D model to discretise partial differential equations, and the ODE15s solver is used to solve time derivatives. The electrode and separator are divided into 10 computational domains along the length scale. To solve the solid phase diffusion numerically, each particle is divided into 10 computational domains along the r-direction from the particle center to the particle surface. The P2D model for Li-ASSB is implemented in MATLAB 2021b and executed on a Laptop at 2.60 GHz with 16 GB of RAM.

***Optimisation.***—We conducted global optimisation using the optimisation function as follows:

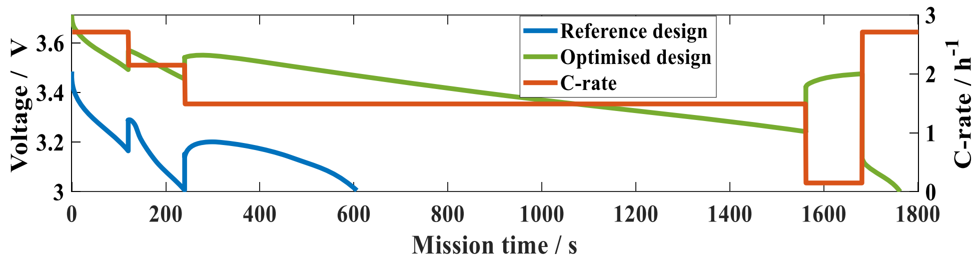


**Figure 3.** Comparison of the cell voltage evolution during the 25.7 km eVTOL mission for the reference (blue) and optimised (green) electrode design for attached CB distribution with a CB/AM ratio of 5% for Li-ASSB cell. Tables I, S1, and S2 display further relevant parameters.

$$\text{maximise specific energy } (x)$$
$$\text{subject to } x_{\text{lower bound}} \leqslant x = \{\delta_{cat},\ \varepsilon_{AM}\} \leqslant x_{\text{upper bound}} \qquad [26]$$

The objective function is to maximise specific energy to enhance the mission range of the multicopter. Since we aimed to optimise the battery without changing the material properties, we considered the cathode parameters, i.e., the AM volume fraction, $\varepsilon_{AM}$, and cathode thickness, $\delta_{cat}$, as decision parameters for the optimisation. These two factors have significant influences on ASSB performance, according to the sensitivity analysis of our recent study.[22]

## Results

In this section, we first examine the performance of batteries with different cathode structures: two cathode distributions (attached and even) with three distinct cathode compositions—low, medium, and high levels of CB inside the cathode electrode. The best structure is then chosen, and it is used as the employed structure in the reference cell. Next, we assess if the reference battery can meet the mission requirement by incorporating the multicopter's dynamic load requirement into the battery model. The optimal design of solid-state electrodes with respect to eVTOL C-rate demand is then identified using a global optimisation technique to improve battery performance. Finally, comparisons between reference and optimal electrode designs are made, and recommendations are given.

***Effect of microstructure on constant current discharge performance.***—Ionic conductivities $>10^{-4}\,\mathrm{S\,cm^{-1}}$ are considered adequate for the development of solid-state batteries with high energy densities, thick electrode designs, and rapid charging/discharging.[25] In the following, we evaluate the hybrid electrolyte of 12.7 vol% of garnet, i.e. LLZTO, which was found best as separator material in our prior study.[22] We examine the effective transport capabilities of various cathode structures using the microstructure surrogate model. To achieve this, we first take into account three possible cathode compositions with different contents of CB in the cathode electrode: low ($\varepsilon_{\mathrm{CB}}/\varepsilon_{\mathrm{AM}} = 5\%$), medium ($\varepsilon_{\mathrm{CB}}/\varepsilon_{\mathrm{AM}} = 8.5\%$), and high ($\varepsilon_{\mathrm{CB}}/\varepsilon_{\mathrm{AM}} = 12.5\%$). We also consider two distinct cathode distributions, CB attached to active material, and CB evenly distributed in the electrolyte for each cathode composition.

The impact of the cathode structure on the effective transport parameters is shown in Fig. 2a, the lower limit of ionic conductivity required for ASSB—which we described above—is shown as a solid red line. Ionic conductivity decreases steadily with increasing active material fraction, similarly as observed in our prior work.[17] At low AM loading and a low amount of CB, the effect of the distribution of components (even vs attached) on the effective ionic conductivity is insignificant. However, for high AM volume fractions, when conducting additives are attached to AM, the effective ionic conductivity drops more than for the even distribution of CB in the electrolyte. The effect becomes even starker for a high amount of CB loading. The stronger loss in ionic conductivity for the AM-surface-bound CB might be caused by the blockage of active material's surface by CB, leading to higher tortuosities as for the CB being homogeneously distributed in the electrolyte. The surface-bound CB also leads to a significant reduction in effective area, as shown in Fig. S1, high amounts of CB block the surface, and thus, reduce the active area, i.e. the contact area of electrolyte and AM, and the electrochemical reaction. The dependence of active area on active material fraction shows a clear maximum at medium active materials fractions. Active area decreases with carbon black content. The decrease at high fractions is attributable to overlapping active material particles, see also Laue et al.[17]

Furthermore, the effective electronic conductivity rises with a higher CB content in the electrode, as seen in Fig. 2a. It is shifted to lower values at lower carbon black content and for even distribution. However, the effective ionic conductivity drops for higher AM fraction. Because the volume fraction of electrolyte declines as AM loadings grow, thus, the electrolyte is insufficient for forming well-connected ionic conduction networks. From the studies, it is clear that AM fraction and distribution and amount of CB heavily impact effective conductivities and active area. For the first time, we show in the following for ASSB the predicted effect of these parameters on battery performance.

Figure 2b shows the battery capacity for the various simulated cathode structures at a discharge rate of 1 C. The capacity of the battery first increases proportional to AM volume fraction, until at around 43% AM fraction, a maximum is reached and capacity strongly drops. The maximum and subsequent decrease is caused by insufficient effective ionic conductivity of PEO-LLZTO 12 vol% electrolyte: it falls below the above-given threshold of $10^{-4}\,\mathrm{S\,cm^{-1}}$ for AM volume fractions higher than 0.46 and 0.5 for high and low CB content, respectively, as shown in Fig. 1a. The loss due to ionic conductivity cannot be compensated by the still increasing effective surface area (maximum at ca. 0.55 AM fraction), and the increasing electric conductivity. Analysing the structure dependence deeper, we see that the cell with CB attached to AM and low CB content performs best. It shows better electric conductivity than the even distributed one, whereas the low CB content allows for high ionic conductivity and active area. For the best cell, all three parameters, i.e., ionic and electric conductivity, and the surface area thus impact performance and need to be adjusted. Interestingly, at high CB loading, the distribution of CB does not impact performance anymore: there is almost the same capacity for uniform and AM-attached CB distribution. At a high amount of CB in the electrode, there is less effective surface area, and worse electrolyte conductivity than low CB content, leading Li-ion transport to be severely hampered and a high gradient Li-ion concentration within the battery. Further, the blockage of active material's surface by a high content of CB within the electrode results in losing effective surface area and decreasing exchange current density of the electrochemical. This results in a high solid concentration gradient at high CB loading, as shown in Fig. S2.

Summarising, ASSB with low CB content of 5% which is attached to AM performs better than one with a uniform distribution. According to Giménez's work,[26] this amount of CB is sufficient for

**Table II. Design parameters and resulting ASSB performance during the eVTOL mission for the reference and optimised design, assuming AM- attached CB distribution with a CB/AM ratio of 5% for Li-ASSB, and results of utilising Bruggeman relation to predict transport properties for ideal spherical morphology. Tables I, S1, and S2 display relevant parameters.**

| | $\delta_{\mathrm{cat,}}$ $\mu$m | $\varepsilon_{\mathrm{s}}$, − | $_{\sigma \mathrm{e,eff}}$, S cm$^{-1}$ | $_{\sigma \mathrm{s,eff}}$, S m$^{-1}$ | as | $\tau$, − | Predicted pack specific energy, Wh kg$^{-1}$ | Mission time |
|---|---|---|---|---|---|---|---|---|
| **Results referring to electrode design with MS** | | | | | | | | |
| Reference design | 55 | 0.46 | 0.018 | 0.0289 | $7.66 \times 10^5$ | 2.42 | 54.6 | 606 |
| Optimised design | 40.1 | 0.42 | 0.022 | 0.0207 | $7.41 \times 10^5$ | 2.26 | 236.84 | 1760 |
| **Results referring to electrode design with Bruggeman relation** | | | | | | | | |
| Electrode design | 40.1 | 0.42 | 0.037 | 0.0473 | $2.29 \times 10^5$ | 1.33 | 264.3 | 1909 |

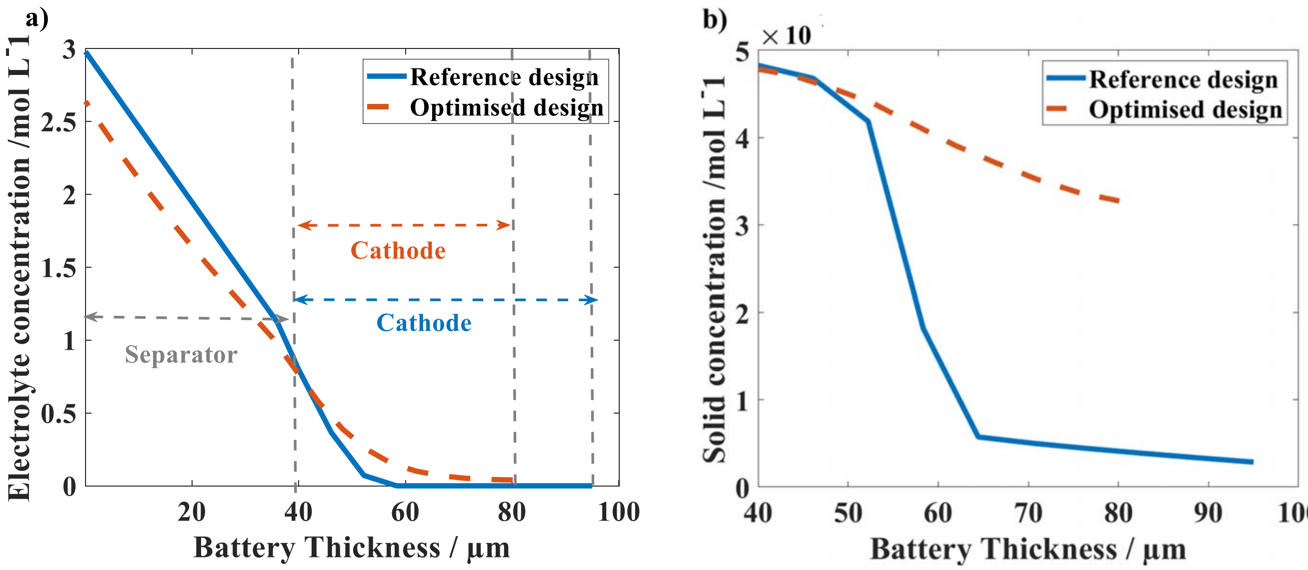


**Figure 4.** State of the ASSB at the end of the eVTOL mission profile of: (a) Li-ion concentration profile in the electrolyte, and (b) solid-phase surface concentration at the particle for the reference and optimised design for AM-attached CB distribution with a CB/AM ratio of 5%.

providing good ionic and electronic conductivities for ASSB. Therefore, in the following section, cells with a 5% CB attached to AM are evaluated for application in the multicopter the cathode.

***ASSB performance during EVTOL mission.***—Here, we first evaluate the ASSB performance with respect to the current demand of the multicopter. Then, to predict the best battery size with consideration of the C-rate requirement of the multicopter, we optimise the battery structure. The cathode design parameters, volume fraction of AM and thickness are set to obtain a cell with high performance, i.e. high specific energy, to fulfill the mission.

Figure 3 illustrates the battery performance for the reference cell during a flight mission. As can be observed, the reference cell breaks down early in the mission, and meets only 33% of the operating time under the operation. Since the battery with reference electrode design would not be able to satisfy mission requirements of eVTOL, global optimisation is used to identify the optimal ASSB electrode design to achieve mission criteria. With an AM volume fraction of 42% and a cathode thickness of 40.1 $\mu$m, the optimum electrode design could significantly increase flight time from 600 to 1760 s at the optimum design point and meets about 98% of mission requirements. It fails in the final landing phase, where high C-rates meet a low SOC. As shown in Table II, the effective ionic conductivity of the optimal electrode design is better than that of the reference cell. This is due to a ca. 20% thinner electrode and 8.7% smaller AM volume fraction, i.e. a lower AM loading in the optimum electrode structure in comparison to the reference design, which leads to better-connected ionic conduction networks. Smaller cathode thickness and a lower AM loading in the optimised design result in shorter effective ion transport pathways, which improves battery performance significantly. Although the reference design with high AM loading results in a higher electronic and effective surface area, the loss in effective ionic conductivity cannot be compensated.

Figures 4a and 4b show the concentration profile of Li-ions in the electrolyte and of Li in the active material at the particle surface for the reference and optimised electrode design at the end of eVTOL mission. As illustrated in Fig. 4a, there is Li-ion starvation at the end of eVTOL mission for the reference design. This can be attributed to the thicker cathode and lower effective ionic conductivity. Higher effective ionic conductivity arises from the optimal electrode design with low AM loading than the reference electrode, resulting in no Li-ion depletion within the electrode. Furthermore, the local exchange current density depends on the local electrolyte concentration. As a result of the difference in electrolyte concentration, local changes in current density and solid concentration will be observed over the electrodes, as exhibited in Fig. 4b. The reference electrode demonstrates a higher solid concentration gradient within the electrodes than the optimum electrode due to a higher Li-ion concentration gradient in the electrolyte and a lack of enough Li-ion at the end of electrode length at the end of eVTOL mission.

In summary, neither the ASSB battery pack with the reference nor with the optimised electrode design is able to fulfil the demand of the mission profile. One reason may be the low ionic conductivity of ASSB, which is still not high enough for high-power and high-energy applications. In this regard, in the following section, we investigate the possible ways to improve the ionic conductivities of the ASSB to meet flight mission requirements.

***ASSB properties needed for fulfilling the eVTOL mission.***—In this section, we aim to assess two possible approaches to improve the effective ionic conductivity of ASSB design with hybrid electrolyte for better performance for urban air mobility applications. One of the possible solutions to reach sufficient specific energy at elevated discharge rates is improving the intrinsic conductivity of the material. Therefore, we examine the impact of increasing the ionic conductivity of hybrid electrolytes on the mission performance, with a focus on the ionic conductivity needed to fulfil the mission time of 30 min, and the specific energy of ASSB, as illustrated in Fig. 5.

As depicted in Fig. 5, an ionic conductivity of $7.5 \times 10^{-4}$ S cm$^{-1}$, which is an increase of 54%, is needed to increase the practical specific energy during the mission from 236 Wh kg$^{-1}$ to a minimum of 251 Wh kg$^{-1}$ at the pack level, which allows to fully complete the mission. This clearly motivates future research in the solid electrolyte to enable ASSBs usable profile of eVTOL applications.

Thus, to obtain sufficient ASSB performance for eVTOL application, highly conductive SE materials with a conductivity of at least $7.5 \times 10^{-4}$ S cm$^{-1}$ should be addressed in material design. As stated in Ref. 22 such a high conductivity might be achieved by employing a hybrid elecrolyte of oxide and polymer electrolytes or by using a combination of polymerised ionic liquid (PIL) or poly (propylene carbonate) (PPC) with oxide-based electrolytes instead of poly (ethylene oxide) (PEO). It should be noted, that more insight into and development of hybrid electrolytes are needed. Conductivity seems frequently not to be a linear combination of

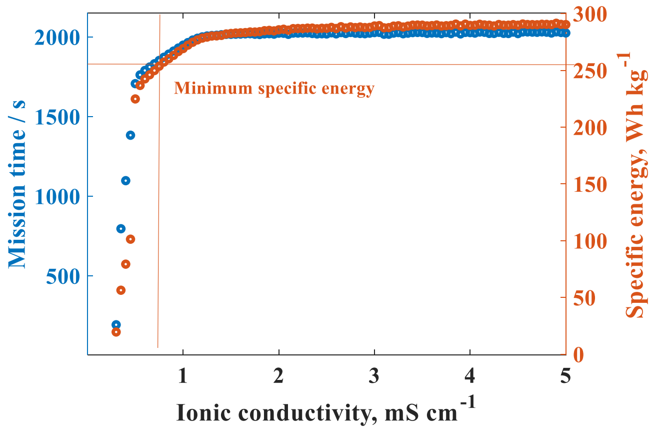


**Figure 5.** Impact of ionic conductivity of PEO-LLZTO 12 vol% electrolyte on the mission time (blue colour) and the predicted practical specific energy during the mission (red colour) for an ASSB with optimised cell design (see Table II) with AM-attached CB distribution with a CB/AM ratio of 5%.

that of the single electrolytes, but rather microstructure, the chemical nature of both electrolytes, and space-charge effects may be highly influential. Significantly higher conductivities of hybrid electrolytes than single-solid electrolytes were reported for some electrolytes.[27–29]

A second way to improve the effective ionic conductivity of ASSB is increasing the transport through the microstructure, e.g. via more spherical particles. The development of high-energy and power-density ASSB necessitates the ability to design electrode microstructures. Because a high tortuosity factor in real porous electrodes is associated with lower delivered energy and power densities; hence, highly spherical particles shape may be a potential way of lowering tortuosity.[30] We seek to apply spherical particle morphology instead of the real structure used in the microstructure surrogate model. The Bruggeman relation with a coefficient of 1.5 corresponds to effective transport parameters of electrode structures with perfect spherical particles and no overlap.[31] We compare the battery performance using this Bruggeman approach with that of the more complex microstructure to see the sensitivity and need for further modifying the particle and electrode structure. As shown in Table II, using an electrode structure with highly spherical particle morphology and a resulting 68% higher effective conductivity would result in an increase in specific energy, and subsequently mission range.

## Conclusions

eVTOL aircraft have the potential to reduce environmental issues caused by internal combustion engines while also reducing commute times. Safety and low specific energy of batteries are major challenges, which may be addressed using solid-state batteries. In this work, ASSB design using hybrid electrolyte (polymer + oxides) are investigated and optimised with electrochemical modelling to identify promising structures that allow eVTOL operation. The cathode microstructure has a considerable impact on performance and specific energy via three performance-relevant microstructure parameters, namely effective electronic and ionic conductivities, effective surface area. Thus, we here extended the P2D model with a microstructure surrogate model, to gain a deeper understanding of the electrode structure. Then we assessed different cathode structures to identify the most promising design for medium-power applications, i.e., 1 C. The optimal design of structures with respect to eVTOL C-rate requirement is finally predicted by linking this model into a global optimisation approach. According to our findings, when CB and AM are premixed, the battery performs better than when SE and AM are premixed, especially for low amounts of CB in the cathode mixture, i.e., 5%. Further, current solid-state battery technology makes it difficult to fully match the mission profile even at an optimal design point. Here, similar to other studies,[19,32] we figured out that enhancing the ionic conductivity of ASSB is required for the battery to successfully improve specific energy at elevated C-rates. Therefore, solid electrolytes with higher ionic conductivity are a precondition for enabling ASSB-powered medium-power applications such as small aircraft. Furthermore, there are two ways to improve effective conductivity. First is to improve the intrinsic conductivity of the material, second is to enhance the transport through the microstructure, e.g. via more spherical particle. Using highly spherical particles instead of uneven ones may increase further effective diffusivity and conductivity, and thus mission range. Further, thinner separators may contribute to the mission goal. Therefore, studying the effect of separation thickness on multicopter performance is valuable for future work. All-in-all, modelling will remain an essential tool to assess in an early stage what are the material and design requirements. Future studies may especially focus on the complexity inside the electrolyte and effects like space-charge layers in hybrid electrolyte, and the contact area between electrolyte and electrode. A better understanding and improved electrolyte models will allow to further improve predictability of the models.

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