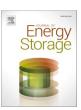
ELSEVIER

Contents lists available at ScienceDirect

#### Journal of Energy Storage

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



#### Research Papers

## Comparative performance assessment of li batteries with solid and highly concentrated electrolytes for aviation applications

Somayeh Toghyani <sup>a</sup>, Florian Baakes <sup>a</sup>, Helmut Kühnelt <sup>b</sup>, Ningxin Zhang <sup>b</sup>, Ulrike Krewer <sup>a,\*</sup>

- <sup>a</sup> Institute for Applied Materials Electrochemical Technologies, Karlsruhe Institute of Technology, 76137 Karlsruhe, Germany
- <sup>b</sup> Center for Low-Emission Transport –Electric Drive Technologies, AIT Austrian Institute of Technology GmbH, 1210 Vienna, Austria

#### ARTICLE INFO

# Keywords: Hybrid solid electrolyte Highly concentrated electrolyte Aeronautic applications P2D model

#### ABSTRACT

Non-flammable next-generation batteries with high power and energy density are essential to meet the high demands of aircraft applications. These cells require electrolytes that not only are stable but that also ensure reliable battery performance under dynamic load conditions. This study employs a physicochemical model to evaluate and compare the electrochemical performance of cells using different cathodic electrolytes: hybrid solid electrolytes, highly concentrated liquid electrolytes, and ionic liquids (ILs). To isolate the intrinsic behavior of each electrolyte under ideal conditions, the evaluation deliberately excludes side reactions, interfacial stability, and contact limitations, focusing solely on gravimetric energy, power output, and mission performance theoretically achievable with such cells. Cells with LiFSI as salt, IL-hydrofluoroether (HFE) as cathode electrolyte or with LiFSA as salt and sulfolane-HFE show superior performance and require less battery mass for aircraft use. To meet the mission requirements of a regional aircraft configuration and when assuming ideal conditions, cathode thicknesses of  $70~\mu m$ ,  $60~\mu m$ , and  $40~\mu m$  for sulfolane-HFE, IL-HFE, and hybrid solid electrolytes, respectively, were determined. This approach helps to identify promising electrolyte candidates, exclude stable candidates that cannot fulfil the dynamic power demand, reduce unnecessary experimental efforts, and support the development of efficient and safe battery systems for aviation.

#### 1. Introduction

Hybrid-electric propulsion systems using Li-based batteries have the potential to reduce CO<sub>2</sub> emissions in airplanes [1]. The main barrier to progress for this kind of aircraft is the low specific energy of batteries. Additionally, to fulfil the requirements of high specific energy and power, as well as reliable operation under extreme environmental conditions, battery chemistry and design need to be optimized [2,3]. This brings additional challenges for the battery community. Liquid electrolytes, which are currently used in many lithium-ion batteries, exhibit high ionic conductivities and enable fast charge transfer. However, because of their high flammability, liquid electrolytes pose a safety issue [4]; liquid electrolyte and their decomposition products decompose at the negative electrode under heat release, causing thermal events [5]. Non-flammable liquid electrolytes allow minimising the risk of battery fire and explosion. Yet they still need to enable high specific energy and performance of the battery [6].

The typically non-flammable solid electrolytes (SE) are therefore a promising alternative for future aircraft. Nevertheless, all-solid-state

batteries (ASSB) currently face high charge transfer overpotentials, low ionic conductivity, and limited capacity, especially at ambient temperatures. They suffer from low specific energy at elevated current rates, as noted in our previous study [7]. Using cells with hybrid electrolytes, which combine liquid electrolytes with inorganic solid electrolyte separators or various SE and polymer electrolytes, is one way to improve ASSB performance. However, there is still a safety concern with hybrid electrolytes, primarily due to the presence of liquid electrolytes.

One approach to potentially reduce flammability and improve the safety of liquid electrolytes is to increase the salt concentration in the electrolyte. Therefore, highly concentrated liquid or ionic liquid (IL) electrolytes have attracted increasing attention for use in lithium batteries [8]. Increasing the salt and thus Li-ion concentration and adding Li-ion containing IL can considerably enhance the electrochemical stability window due to the higher oxidation stability of the IL in the highly concentrated electrolytes (HCE) [9]. However, due to the high viscosity and low ionic conductivity of IL electrolytes in comparison to commonly utilized organic carbonate solvents, most lithium batteries using IL electrolytes have poor rate capability [4]. To address this, diluent

E-mail address: ulrike.krewer@kit.edu (U. Krewer).

<sup>\*</sup> Corresponding author.

solvents like hydrofluoroether (HFE), which are non-flammable and feature low viscosity, can be added to reduce viscosity. HFE could also reduce the cost of IL electrolytes while enhancing the ionic conductivity of IL electrolytes and their wetting capabilities for the separator surface [10]. Therefore, due to these favorable properties, HFE shows strong potential for application in aviation batteries.

Numerous studies have been conducted on HCE. Wang et al. [10] demonstrated the high safety of IL electrolytes composed of lithium bis (fluorosulfonyl) imide (LiFSI) salt in N-methyl-N-propyl-piperidinium bis(fluorosulfonyl)imide ([PP13][FSI]): they feature low volatility and nonflammability. Using this type of electrolyte in batteries demonstrated cycling stability exceeding 800 cycles [10]. They also examined a highly concentrated TFSI-FSI dual-anion electrolyte based on LiTFSI, Nethyl-*N*-methyl-pyrrolidinium bis(fluorosulfonyl)imide ([C2mpyr] [FSI]), and dimethyl carbonate (DMC) [11]. Their results showed long cycle life for LiFePO<sub>4</sub>/Li cells with 80 % capacity retention after 500 cycles, while Li/Ni<sub>5</sub>Co<sub>2</sub>Mn<sub>3</sub>O<sub>2</sub> cells with TFSI-FSI dual-anion electrolyte retained 95 % capacity after 100 cycles. Ren et al. [12] indicated that highly concentrated LiFSI-sulfolane (SL) electrolytes could considerably reduce electrolyte decomposition when in contact with the Li metal anode, enabling stable cycling of Li-ion batteries at a high cathode voltage of 4.9 V. In another study, Nakanishi et al. [13] showed that adding HFE to SL-based HCE significantly improved diffusion coefficients and reduced viscosity by nearly two orders of magnitude. Additionally, HFE decreased viscosity by nearly two orders of magnitude. Further, the transport characteristics of HCE consisting of Li salt and LiBF<sub>4</sub>-SL electrolytes were studied by Dokko et al. [9]. They attributed the enhanced rate capability of LiBF<sub>4</sub>/SL (1:2 volume ratio) to reduced concentration polarization and higher limiting current. Matsumoto et al. [14] exhibited that a battery with a high concentration of LiTFSI could operate without degradation. Tong et al. [15] computationally studied the physicochemical characteristics of HCE using two organic solvents, DMC and DEC, and four IL solvents ([Cnmim][BF4] and [Cnmim][TFSI] (n = 2, 4)). Their simulations revealed that 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide [BF4]) had the highest conductivity tested IL-based electrolytes. A nonflammable electrolyte based on IL (0.8Pyr14FSI-0.2LiTFSI) was designed by Wu et al. [16]. A full cell with this electrolyte and a Li metal anode and NMC cathode could provide a specific energy of more than 560 Wh/kg at 0.1C.

Although extensive experimental research has been carried out on HCE, there is no single experimental or modelling study that demonstrates the capability of such electrolytes in high-power, high-energy applications like aeronautics. Battery performance for aviation strongly depends on the design of the anode, separator, and cathode; in this work, we focus specifically on cathode design.

In our previous work, we conducted a comprehensive analysis of various material combinations and separator thicknesses for Li-ASSB using NMC811 as the cathode material. The model developed in that study was validated against experimental data across different C-rates, confirming its reliability for assessing cell-level performance under diverse operating conditions [7]. Among the solid electrolytes examined, PEO-LiTFSI/LLZTO (12.7 vol%) delivered the best performance and compatibility with Li-metal, and is therefore adopted in this study as the solid separator. However, ASSB alone remains insufficient to meet the high specific energy demands of aviation, particularly under high current densities and thick electrode configurations [7]. Their major limitation lies in the low ionic conductivity within thick cathodes, especially at elevated C-rates. Therefore, employing two distinct electrolytes within the battery is a promising strategy to enhance ASSB performance using a stable solid electrolyte at the anode and a highly conductive one at the cathode.

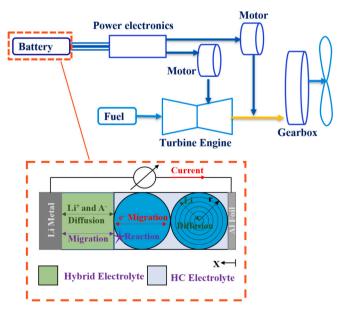
In this study, we implement HCE as the cathodic electrolyte to enhance energy and power performance. Developing such dualelectrolyte cells experimentally would require significant time and resources, and testing all HCE-SE combinations would be impractical. To overcome this, we use physics-based modelling to quickly assess the theoretical potential of various highly concentrated liquid or ionic cathodic electrolytes. We purposely omit any potential interface issues due to electrolyte reactivity, as these could be mitigated in the future through additives or interface engineering. Therefore, this study focuses on the electrochemical behavior of candidate cathodic electrolytes under idealised conditions. The goal is to identify non-flammable electrolytes with good compatibility with high-voltage cathodes like NMC811 and sufficient performance for aviation applications. Based on the best-identified cathodic electrolyte, we also examine the limiting processes in the cell and the battery performance under aircraft power demands to give an overview of the feasibility of utilising these electrolyte alternatives for future aircraft technology. Finally, some recommendations for the battery manufacturing sector are provided.

Although several of the selected electrolytes have been investigated previously, this study contributes a unique performance comparison under aviation-specific mission profiles, using a physically consistent model framework. The focus is on combining non-flammable cathodic electrolytes with a hybrid solid electrolyte separator to reduce pack-level battery mass while maintaining high power and energy output. We restrict our scope to Li-metal/NMC chemistry, as it currently offers higher maturity in terms of stability and manufacturability compared to alternatives like lithium-sulfur.

#### 2. Methodology

To assess battery performance in aviation, a conservative concept for a hybrid-electric regional aircraft is evaluated [1]. A parallel-hybrid architecture is employed, which is described in detail in [17]. For propulsion, each turbo engine is assisted by two electrical motors, as illustrated in Fig. 1. The electric motors powered by batteries, assist the turbo engine during take-off, climb, and cruise phases of the mission. Here, we applied the mission profile of this regional aircraft configuration, with battery power requirements at various flight stages shown in Fig. S1 [17]. In this mission profile, a fully electric operation is used during the taxi-out and taxi-in phases, while the battery load decreases throughout the descent to provide power for taxi-in. Additionally, batteries provide electric power for the aircraft's non-propulsive subsystems in most flight phases.

The power profile of the hybrid-electric regional aircraft is imple-



**Fig. 1.** Hybrid-electric propulsion with Li metal anode and hybrid electrolyte separator, highly concentrated electrolyte as the cathodic electrolyte, and NMC811 cathode materials.

mented in the Pseudo-Two-Dimensional (P2D) model to evaluate the performance of a single battery cell. In reality, due to battery management systems, contacting technology, sensors, and other factors, it is impossible to maintain the same specific energy at the cell and system levels. As a result, specific energy decreases from the cell level to the system level. This reduction occurs because the battery system includes additional components such as housing, thermal management, wiring, and battery management systems, which add mass without contributing directly to energy storage. For example, while a single cell might deliver approximately 250–270 Wh/kg, the system-level specific energy typically drops to 150–180 Wh/kg due to these overheads [18,19]. To quantify this, the efficiency rate can be defined as follows:

Efficiency rate = 
$$\frac{Specific\ energy\ at\ pack}{Specific\ energy\ at\ cell}$$
 (1)

The efficiency rate of the battery from cell level to pack level is considered equal to 0.74 [18]. Using this value and knowing the total charge delivered by the battery from the mission profile, we can estimate the total battery mass required for the mission. As mentioned earlier, due to the main barriers of ASSB, i.e., the limited ionic conductivity of the electrolyte on the cathode side, they cannot operate efficiently at high C-rates. One approach to improve cell performance in hybrid-electric propulsion systems is the use of composite cathodic electrolytes combining inorganic solid electrolytes with highly concentrated liquid or ionic electrolytes. We here use a hybrid solid electrolyte (PEO-LiTFSI/LLZTO (12.7 vol%)) as the reference cathodic electrolyte and then compare its performance with HCE containing either highly concentrated Li-ion containing liquid or ionic electrolytes in the cathode. All these electrolytes contain Li-ions of high mobility. In our model, the electrolyte is treated as a single, lumped phase with effective ionic transport properties that represent the combined behavior of all electrolyte components. This simplification is appropriate within the P2D modelling framework, which focuses on cell-scale performance and typically assumes homogeneous electrolyte properties to reduce computational complexity.

The aim of this work is to identify promising cathodic electrolytes that could improve battery performance in regional hybrid-electric aircraft. We use the same separator layer (PEO-LiTFSI/LLZTO), Li metal anode, and NMC811 cathode active material for all simulations. The reason for the choice of an oxide-based solid electrolyte in the separator is that such electrolytes effectively inhibit lithium dendrite growth and protect the cell from internal short circuits when in contact with the Li metal anode [20]. Furthermore, in Ref [7], we showed that this electrolyte has the highest specific energy in the medium power application, given the current manufacturing limitations of ASSB compared to other hybrid electrolytes including polymer and oxide-based solid electrolytes.

## 2.1. Physicochemical model of hybrid electrolytes and non-flammable highly concentrated electrolytes

The P2D model is a common approach in battery modelling, allowing for a more detailed representation of the electrochemical processes within the battery. This approach, described in detail in our previous work, was used to model Li-ASSB with hybrid electrolytes [7]. The electrode microstructure is crucial for mass transport through the electrolyte phase and the solid-phase electrical conductivity [21,22]. Therefore, the classical P2D model, which assumes ideal particles and a homogeneous electrode, is extended by microstructure surrogate models, which incorporate the influence of microstructure on effective surface area and transport parameters into the homogeneous P2D framework. We concluded from our previous research that premixing conductive additives and active materials (AM) into the cathode structure results in a higher capacity than when carbon black is evenly distributed throughout the electrolyte [23,24]. The electrodes produced by premixing additives with AM and the corresponding surrogate model

[24], as detailed in Table S1, are used in this study. Furthermore, our prior work identified that a cathode composition with a low CB/AM volume ratio of 5 % performs best by balancing ionic and electronic conductivities for ASSB. Hence, we adopt the same cathode composition in the present study.

The governing equations of the P2D model are provided in Table S1. The concentrated solution theory is employed to describe Li-ion transport in the electrolyte phase [25]. To predict the electrolyte diffusion coefficient of the hybrid electrolyte, we used the Einstein relation; however, this relation is only valid for dilute solutions. Therefore, to estimate the diffusion coefficient of the highly concentrated electrolyte, we utilized experimentally derived diffusion coefficients from the literature, as listed in Table S2. The initial lithium concentration is a critical parameter in battery modelling as it influences the electrochemical behavior and overall performance. For the hybrid electrolyte, the initial lithium concentration is set to 1200 Mol/m<sup>3</sup>, while the initial concentrations for highly concentrated electrolytes are given in Table S2, e.g., 2100 Mol/m<sup>3</sup> for LiFSI in IL-HFE and 2000 Mol/m<sup>3</sup> for LiFSA in sulfolane-HFE. These values are based on reported experimental formulations. Additionally, concentrated electrolytes can affect the electrode-electrolyte interface properties, influencing parameters such as double-layer capacitance and charge transfer kinetics. It is important to note that these effects are complex and depend on multiple factors, including electrolyte composition, electrode materials, and operating conditions. Experimental studies and detailed modelling are typically required to fully understand battery behavior with such electrolytes. For simplicity and to elucidate intrinsic limitations of catholytes due to electrochemical performance during a mission, this study omits material-specific interfacial effects.

To estimate the specific energy,  $E_G$ , and energy density,  $E_V$  of a single battery cell, Eqs. (2) and (4) are used [7].  $V_{\rm cell}$  and  $M_{\rm cell}$  denote the cell volume and mass of the cell, respectively, accounting for the mass and volume of components including cathode and anode electrodes (c, a), solid separator (s), solid electrolyte (SE), and current collectors (cc: cathode, ac: anode).

$$E_G = \frac{\int_0^{t_{cutoff}} U_{cell} I dt}{M_{cell}}$$
 (2)

$$\mathbf{\textit{M}}_{cell} = (\delta_{ac}\rho_{ac}\mathbf{\textit{A}}_{ac} + \delta_{SE}\rho_{SE}\mathbf{\textit{A}}_{SE} + \delta_{c}\rho_{c}\varepsilon_{s}\mathbf{\textit{A}}_{c} + \delta_{c}\rho_{e}\varepsilon_{e}\mathbf{\textit{A}}_{c} + \delta_{a}\rho_{a}\mathbf{\textit{A}}_{a} + \delta_{cc}\rho_{cc}\mathbf{\textit{A}}_{cc})$$
(3

$$E_V = \frac{\int_0^{t_{cutoff}} U_{cell} I \, dt}{V_{cell}} \tag{4}$$

$$V_{cell} = (\delta_{ac} A_{ac} + \delta_{SE} A_{SE} + \delta_c \varepsilon_s A_c + \delta_c \varepsilon_e A_c + \delta_a A_a + \delta_{cc} A_{cc})$$
(5)

where  $\rho$  is the density,  $\delta$  the thickness,  $\epsilon$  is the volume fraction and A is the geometric area of the various battery components and can be computed using the parameters listed in Table S2. The electrode is discretised into 10 equidistant parts along the length scale; further, each particle is also discretised in 10 layers along the radial-direction from the particle surface to the particle center. The P2D modelling of the Libattery with various cathodic electrolytes is executed in MATLAB 2022b [26]. All physical, electrochemical, and geometrical parameters used in the P2D model, as well as parameters linked to the empirical polynomial equations for effective surface area and transport properties, are provided in Table S2.

#### 3. Results and discussion

This section first evaluates the performance of various highly concentrated electrolytes and hybrid solid electrolytes to identify those with promising gravimetric energy and power density suitable for aviation applications. Next, we use the aircraft power demand during a mission for a regional hybrid aircraft and couple it with the P2D model

to evaluate how these electrolyte types influence battery performance in realistic aviation scenarios.

#### 3.1. Comparative performance study of electrolytes

We here present a comprehensive study on the impact of different highly concentrated electrolytes to identify the most suitable ones for aircraft application. This comparison uses the same materials for the anode, i.e., lithium metal, NMC811 as cathode active materials with PEO- LiTFSI/LLZTO (12.7 vol%) electrolyte used in the separator layer. The only difference between cells is the cathodic electrolyte composition. It should be noted that the mass fraction of active material is kept at 0.42, as our earlier work demonstrated that this ratio provides optimal ionic conduction networks for medium power applications [7]. We applied electrolyte parameters for the modelling, such as the lithium transference number, ionic conductivity, electrolyte diffusion coefficient, and initial electrolyte concentration based on experimental values reported in the literature, as listed in Table S2. Since hybrid SE performs well at a temperature of 60 °C [7], the Arrhenius relation is used to account for the temperature dependence of electrolyte properties when those properties are not available at the target temperature [27]. The highly concentrated electrolytes listed in Table S2 are selected based on existing experimental data from the literature. The goal of this comparison is to evaluate their performance under ideal conditions and identify on this basis electrolytes are promising options for aviation applications. By examining these electrolytes, we aim to provide valuable insights for future experimental investigations, which will be necessary to evaluate their practical applicability under real-world operating conditions. This information is crucial for guiding future research efforts, preventing unnecessary investment of time and resources into electrolytes that are unlikely to perform well under the demanding conditions of aviation applications.

Fig. 2 illustrates the performance of the various cathodic electrolytes at different current rates. As discussed in the introduction, 2 M LiFSA in sulfolane/HFE and 2.1 M LiFSI in IL/HFE ((IL = [PP13][FSI])) are the most promising cathodic electrolytes on their electrochemical characteristics. These electrolytes exhibit high ionic conductivity and notably high electrolyte diffusivity, which together facilitate efficient lithiumion transport within the cathode. The addition of HFE increases both ionic conductivity and electrolyte diffusivity, leading to more efficient lithium-ion transport and improved electrochemical performance of the cathodic electrolyte [9,10,13]. Moreover, LiFSI-based ionic liquids like [PP13][FSI] demonstrate excellent safety characteristics, including low volatility and nonflammability, along with cycling stability exceeding

800 cycles. In addition, their superior electrochemical and thermal stability compared to conventional carbonate-based electrolytes ensures reliable performance under the demanding conditions of high-power aviation systems [10]. In the simulated Ragone plot, both electrolytes outperform the others, delivering relatively high specific energy even at elevated C-rates. This makes them strong candidates for high-power aviation applications. Their superior performance is attributed to the presence of HFE, which enhances ionic conductivity and diffusion [10].

Among these, the highly concentrated LiFSA-sulfolane/HFE electrolyte shows the best performance. However, its lower experimentally reported cycling stability compared to LiFSI-IL-HFE is an open challenge [9]. This is due to the presence of LiFSI salt in the electrolyte solution, which shows higher cycling performance compared to LiFSA [28].

The second best-performing electrolyte, the highly concentrated LiFSI-IL-HFE electrolyte has an experimentally reported high electrochemical stability in contact with high cathode voltage materials like NMC811: it currently features a relatively high cycling number of 800 [10]. Therefore, due to its outstanding properties, we selected this electrolyte as a promising candidate for detailed evaluation and comparison against the baseline electrolyte.

The areal capacity of a single cell with the three selected cathodic electrolytes at different C-rates is depicted in Fig. 3. As shown in this figure, at low discharge rates, the differences in cell performance between the various cathodic electrolytes are insignificant. This is because kinetic and mass transport limitations are less pronounced under low current conditions. At high discharge rates, PEO-LiTFSI/LLZTO performs significantly worse than the other electrolytes. The high ionic transport overpotential of the hybrid electrolyte increases the slope of the discharge curve, resulting in a low and fast-degrading voltage, and consequently, a reduced discharge capacity. Replacing the hybrid solid electrolyte with a highly concentrated electrolyte improves ionic conductivity and the Li-ion diffusion coefficient, thereby mitigating diffusion limitations at elevated discharge rates. To gain deeper insight into these differences, we also examine the Li-ion and solid concentration profiles at the end of discharge for the best- and worst-performing cathodic electrolytes.

As shown in Fig. 4-a, there is a lithium deficiency at the end of discharge for the PEO-LiTFSI/LLZTO electrolyte, even at medium current ranges of 1C. In contrast, cells with 2 M LiFSA in sulfolane/HFE and 2.1 M LiFSI in IL/HFE do not exhibit signs of Li-ion depletion on the cathode side, even at a high discharge rate. A low Li concentration in the active material is observed for the hybrid electrolyte, Fig. 4-b, which is attributed to the limited lithium content in the electrolyte and insufficient Li-ion transport toward the end of the electrode. This results in low

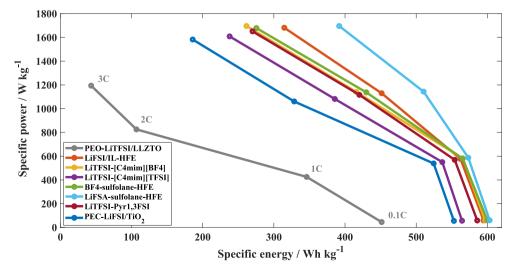


Fig. 2. Specific power vs. specific energy for various cathodic electrolytes at different C-rates. Operation at a temperature of 60  $^{\circ}$ C with Li and NMC811 electrodes. All cathodes feature a thickness of 75  $\mu$ m and an active materials volume fraction of 42  $^{\circ}$ C.

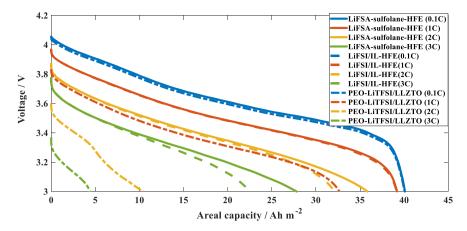


Fig. 3. Voltage vs. areal capacity for cells with 1.2 M LiTFSI in PEO/LLZTO, 2 M LiFSA in sulfolane/HFE, and 2.1 M LiFSI in IL/HFE as cathodic electrolyte at various C-rates. The cell contains Li metal as the anode, PEO-LiTFSI/LLZTO as the solid separator, and NMC811 as the cathode with a thickness of 75 μm.

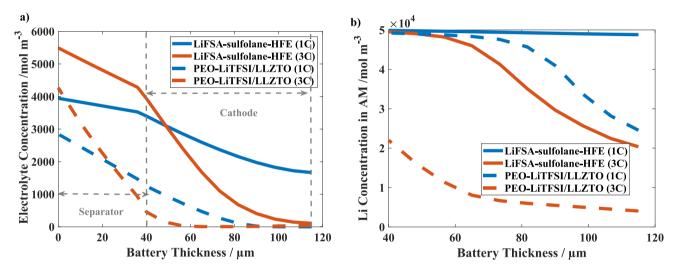


Fig. 4. a) Li-ion concentration profile within the electrolyte, b) Li concentration in the active materials for cells with 1.2 M LiTFSI in PEO/LLZTO or 2 M LiFSA in sulfolane/HFE as cathodic electrolyte at the end of discharge at various C-rates for a cell with Li metal as the anode, hybrid SE based on LLZTO as the solid separator, and NMC811 as the cathode with a thickness of 75  $\mu$ m.

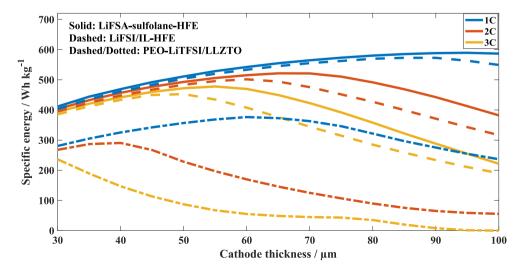


Fig. 5. Specific energy vs. cathode thicknesses for cells with 1.2 M LiTFSI in PEO/LLZTO, 2 M LiFSA in sulfolane/HFE, and 2.1 M LiFSI in IL/HFE as cathodic electrolyte at constant current operation with Li metal as the anode, PEO-LiTFSA/LLZTO as the solid separator, and NMC811 as the cathode.

active material utilisation. In addition, we examine the Li-ion and solid concentration profiles at the end of discharge for IL-based HCE, i.e. 2.1 M LiFSI in IL/HFE; unlike the hybrid electrolyte, no Li shortage is observed at 1C, as seen in Fig. S2. However, at higher C-rates, a minor lithium depletion appears near the current collector, whereas for the hybrid electrolyte, such depletion is more severe and occurs at the beginning of the cathode.

Increasing electrode thickness can enhance the proportion of active material and, consequently, the specific energy. However, as discussed, the excessive thickness may result in underutilisation of active material, especially at high C-rates. Thus, we analyse how cathode thickness affects performance across various current rates and cathodic electrolytes.

As illustrated in Fig. 5, the specific energy of the cell with LiFSA-sulfolane-HFE and LiFSI/IL-HFE increases with increasing electrode thickness, reaching 70  $\mu m$  and 60  $\mu m$  for 2C, and 55  $\mu m$  and 50  $\mu m$  for 3C operation. Beyond this critical thickness, the specific energy decreases significantly. A similar trend is observed for the hybrid solid electrolyte: the optimum cathode thickness for 2C is around 40  $\mu m$ . For 3C operation, however, there is a continuous decline in specific energy with increasing thickness. This decline is attributed to mass transfer limitations caused by longer diffusion paths in thicker electrodes (see Fig. 4). Moreover, it is evident that cells employing highly concentrated electrolytes achieve significantly higher specific energy compared to those using hybrid solid electrolytes.

So far, we have examined the cell-level performance of HCE and hybrid solid electrolyte cells. To analyse battery performance at the pack level under the mission requirements, it is necessary incorporate the efficiency factor, as discussed in the methodology section. To do this, we first estimate the specific energy at the cell level and then compute the specific energy at the pack level using an efficiency factor of 0.742. Next, the total battery capacity required to meet the mission energy demand is calculated based on the power profile of the REG-CON aircraft. Finally, the battery mass corresponding to different cathodic electrolytes at their optimal cathode thicknesses is determined, as summarized in Table 1 for 2C and 3C operation.

For PEO-LiTFSI/LLZTO, the theoretically optimized cathode thicknesses under ideal conditions are 40  $\mu m$  at 2C and 30  $\mu m$  at 3C, which require a larger number of cells to meet the power profile. Similarly, to ensure sufficient energy for the flight mission, the ideal cathode thicknesses for IL-based and sulfolane-based cells are 60  $\mu m$  and 50  $\mu m$ , and 70  $\mu m$  and 55  $\mu m$  at 2C and 3C, respectively. Compared to a battery pack with the hybrid solid electrolyte in the cathode, a 2C-optimized battery pack employing LiFSA-Sulfolane/HFE and LiTFSI in IL/HFE electrolytes results in a 44 % and 41 % lower battery mass, respectively. At 3C, the weight savings are even more pronounced—only half the mass is required for battery packs using sulfolane- and IL-based electrolytes. These findings clearly demonstrate the substantial mass advantage

**Table 1**Specific energy and total battery mass with respect to aircraft mission profile for PEO-LiTFSA/LLZTO, 1.2 M LiTFSI in IL/HFE, and 2 M LiFSA in sulfolane/HFE as cathodic electrolyte at 2C and 3C operation with Li metal as the anode, PEO-LiTFSA/LLZTO as the solid separator, and NMC811 as the cathode.

Cathodic Electrolyte	Cathode thickness, µm	C- rate	Specific energy, Wh kg <sup>-1</sup>	Battery mass, kg
2 M LiFSA in sulfolane/HFE	70	2	386.87	2222.91
1.2 M LiTFSI in IL/ HFE	60	2	370.74	2319.92
PEO-LiTFSI/ LLZTO	40	2	215.64	3987.99
2 M LiFSA in sulfolane/HFE	55	3	354.46	2426.22
1.2 M LiTFSI in IL/ HFE	50	3	334.76	2569.26
PEO-LiTFSI/ LLZTO	30	3	175.11	4911.14

offered by HCE electrolytes.

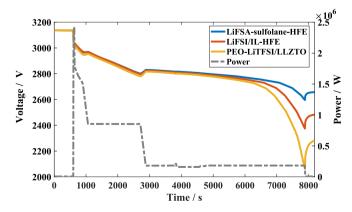
### 3.2. Battery performance under mission-specific demand in a regional hybrid aircraft

Until now, we have assessed the battery performance of various cathodic electrolytes under constant current conditions. To examine how these electrolytes behave under dynamic and especially application-specific load demand, we apply the power demand profile of a regional hybrid aircraft. This mission profile includes a high power load during take-off, a cruise time, and a long landing phase including taxi operations (see Fig. S1).

We use the optimum cathode thickness for each determined under 2C operation: 70 µm for LiFSA-Sulfolane/HFE, 60 µm for LiFSI-IL/HFE, and 40 µm for PEO-LiTFSI/LLZTO. Fig. 6 shows the performance of all three battery types under dynamic power demand in REG-CON hybrid aircraft. While a thinner cathode layer in the hybrid solid-state electrolyte battery allows it to meet mission energy demands, it requires substantial oversizing of the battery pack. This results in increased system mass due to the low specific energy of such batteries, making them unsuitable for aviation applications. In addition to the larger battery mass (see Table 1), the hybrid solid-state battery exhibits significant voltage drops during the mission. This is because of the high concentration gradient and poor ionic conductivity, which slow down ion transport by creating more barriers to ion movement and ultimately leading to an increasing ohmic drop. In contrast, HCE-based cathodic electrolyte batteries—with their higher specific energy—impose significantly less mass on the aircraft system while maintaining stable voltage output, making them more viable for aviation use.

#### 4. Conclusion

To reduce CO<sub>2</sub> emissions and fuel consumption in aviation, hybridelectric propulsion systems represent a promising solution. However, the feasibility of such systems critically depends on battery performance, where electrical energy storage remains a limiting factor. As a precondition, the electrolytes used in batteries for such applications should be non-flammable and have a suitable electrochemical stability window when in contact with high cathode voltage materials; this excludes typical electrolytes used in Li-ion batteries. Next-generation batteries like ASSB technologies have emerged as attractive candidates for future aviation applications due to their superior safety and energy density characteristics. However, ASSB with hybrid electrolytes that allow dual-ion conduction often suffer from Li depletion at the cathode side during cycling. To overcome these limitations, this study explores a



**Fig. 6.** Battery performance under dynamic power demand for REG-CON aircraft with various cathodic electrolytes with Li metal as the anode, hybrid SE based on LLZTO as the solid separator, and NMC811 as the cathode with a thickness of 70  $\mu$ m for LiFSA-Sulfolane/HFE, 60  $\mu$ m for LiFSI-IL/HFE and a thickness of 40  $\mu$ m for hybrid solid electrolyte.

dual-electrolyte concept, using a stable solid-state separator in contact with Li-metal and a high-conductivity cathodic electrolyte to enhance performance.

We conducted a comparative, model-assisted evaluation of various highly concentrated liquid and ionic electrolytes, focusing on mission-relevant conditions. By neglecting interfacial and aging issues, the study isolates the ideal electrochemical behavior of candidate electrolytes. Cells incorporating LiFSA–sulfolane–HFE and LiFSI–IL–HFE outperform hybrid solid-state cells, showing improved energy and power output with reduced diffusion limitations. Under idealised conditions, these cells achieved battery mass reductions of 44 % and 41 %, respectively, compared to hybrid solid electrolyte systems. These gains highlight the potential of highly concentrated cathodic electrolytes for aviation applications.

This model-based screening approach helps to efficiently filter out less viable materials early in the development process, avoiding unnecessary experimental efforts on systems unlikely to meet aviation demands. While the present work focuses on ideal electrochemical performance, real-world considerations—such as interfacial stability, thermal resilience, long-term degradation, and failure risks—need to be addressed in future studies, especially under aviation-relevant conditions such as broad temperature ranges and mechanical stress (e.g., vibration). In addition, since solid electrolytes often require pre-heating to operate efficiently at low temperatures, integrated thermal management strategies must be explored. Further steps include testing single cells under aviation-specific load profiles, and optimising cathode microstructure to improve ion transport and mitigate performance limitations.

This work contributes to a better understanding of the performance limits and trade-offs in dual-electrolyte ASSBs and offers a pathway for designing safer, lighter, and more energy-dense batteries tailored to the needs of next-generation aircraft.

#### CRediT authorship contribution statement

Somayeh Toghyani: Writing – original draft, Software, Methodology, Conceptualization. Florian Baakes: Writing – review & editing, Supervision, Methodology, Conceptualization. Helmut Kühnelt: Writing – review & editing, Methodology, Conceptualization. Ulrike Krewer: Writing – review & editing, Supervision.

## Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this manuscript, the authors used ChatGPT (OpenAI) to assist with grammar correction and sentence clarity. All scientific content, data analysis, and original writing were entirely generated by the authors. The authors reviewed and edited the final version and take full responsibility for the content of the published article.

#### Declaration of competing interest

The authors declare no conflict of interest.

#### Acknowledgments

We would like to acknowledge the funding of the European Union's Horizon 2020 research and innovation program under grant agreement No 875006 IMOTHEP. The authors are extremely thankful to Anais Luisa Habermann from Bauhaus Luftfahrt for providing the necessary information and guidance to pursue this research.

#### Appendix A. Supplementary data

The supporting information contains the following information:

Fig. S1: Power demand of battery at different mission stages for REG-CON aircraft; Table S1: Governing model equations of P2D model for Li-metal battery with hybrid solid electrolyte and highly concentrated electrolyte; Table S2: Geometry, physical, and electrochemical parameters Li-battery with hybrid solid electrolyte and highly concentrated electrolytes; Fig. S2: Li-ion concentration profile within the electrolyte, b) Li concentration in the active materials for cells with 1.2M LiTFSI in PEO/LLZTO or 2.1M LiFSI in IL/HFE as cathodic electrolyte. Supplementary data to this article can be found online at <a href="https://doi.org/10.1016/j.est.2025.118213">https://doi.org/10.1016/j.est.2025.118213</a>.

#### Data availability

The data presented in the manuscript are openly available in the KITopen repository at DOI 10.35097/jvp85q7csvs1dzcp.

#### References

- Project of the European Union, H2020 IMOTHEP. https://www.imothep-project. eu/.
- [2] V. Viswanathan, A.H. Epstein, Y.M. Chiang, E. Takeuchi, M. Bradley, J. Langford, M. Winter, The challenges and opportunities of battery-powered flight, Nature 601 (2022) 519–525, https://doi.org/10.1038/s41586-021-04139-1.
- [3] J. Hoelzen, Y. Liu, B. Bensmann, C. Winnefeld, A. Elham, J. Friedrichs, R. Hanke-Rauschenbach, Conceptual design of operation strategies for hybrid electric aircraft, Energies 11 (2018), https://doi.org/10.3390/en11010217.
- [4] M. Weiss, F.J. Simon, M.R. Busche, T. Nakamura, D. Schröder, F.H. Richter, J. Janek, From liquid- to solid-state batteries: ion transfer kinetics of heteroionic interfaces, Electrochem. Energy Rev. 3 (2020) 221–238, https://doi.org/10.1007/ s41918-020-00062-7.
- [5] F. Baakes, M. Lüthe, M. Gerasimov, V. Laue, F. Röder, P.B. Balbuena, U. Krewer, Unveiling the interaction of reactions and phase transition during thermal abuse of Li-ion batteries, J. Power Sources 522 (2022) 230881, https://doi.org/10.1016/j. ipowsour.2021.230881.
- [6] H.Q. Pham, H.Y. Lee, E.H. Hwang, Y.G. Kwon, S.W. Song, Non-flammable organic liquid electrolyte for high-safety and high-energy density Li-ion batteries, J. Power Sources 404 (2018) 13–19, https://doi.org/10.1016/j.jpowsour.2018.09.075.
- [7] S. Toghyani, F. Baakes, N. Zhang, H. Kuehnelt, W. Cistjakov, U. Krewer, Model-based design of high energy all-solid-state Li batteries with hybrid electrolytes, J. Electrochem. Soc. 169 (4) (2022) 040550, https://doi.org/10.1149/1945-7111/20553h
- [8] H. Niu, L. Wang, P. Guan, N. Zhang, C. Yan, M. Ding, X. Guo, T. Huang, X. Hu, Recent advances in application of ionic liquids in electrolyte of lithium ion batteries, J Energy Storage 40 (2021) 102659.
- [9] K. Dokko, D. Watanabe, Y. Ugata, M.L. Thomas, S. Tsuzuki, W. Shinoda, K. Hashimoto, K. Ueno, Y. Umebayashi, M. Watanabe, Direct evidence for Li ion hopping conduction in highly concentrated sulfolane-based liquid electrolytes, J. Phys. Chem. B 122 (2018) 10736–10745, https://doi.org/10.1021/acs. jpcb.8b09439.
- [10] Z. Wang, F. Zhang, Y. Sun, L. Zheng, Y. Shen, D. Fu, W. Li, A. Pan, L. Wang, J. Xu, J. Hu, X. Wu, Intrinsically nonflammable ionic liquid-based localized highly concentrated electrolytes enable high-performance Li-metal batteries, Adv. Energy Mater. 11 (2021) 1-8, https://doi.org/10.1002/aenm.202003752.
- [11] Z. Wang, Y. Sun, Y. Mao, F. Zhang, L. Zheng, D. Fu, Y. Shen, J. Hu, H. Dong, J. Xu, X. Wu, Highly concentrated dual-anion electrolyte for non-flammable high-voltage Li-metal batteries, Energy Storage Mater 30 (2020) 228–237, https://doi.org/10.1016/j.ensm.2020.05.020.
- [12] X. Ren, S. Chen, H. Lee, D. Mei, M.H. Engelhard, S.D. Burton, W. Zhao, J. Zheng, Q. Li, M.S. Ding, M. Schroeder, J. Alvarado, K. Xu, Y.S. Meng, J. Liu, J.G. Zhang, W. Xu, Localized high-concentration sulfone electrolytes for high-efficiency lithium-metal batteries, Chem 4 (2018) 1877–1892, https://doi.org/10.1016/j.chempr.2018.05.002.
- [13] A. Nakanishi, K. Ueno, D. Watanabe, Y. Ugata, Y. Matsumae, J. Liu, M.L. Thomas, K. Dokko, M. Watanabe, Sulfolane-based highly concentrated electrolytes of lithium Bis(trifluoromethanesulfonyl)amide: ionic transport, Li-ion coordination, and Li-S battery performance, J. Phys. Chem. C 123 (2019) 14229–14238, https://doi.org/10.1021/acs.jpcc.9b02625.
- [14] K. Matsumoto, K. Inoue, K. Nakahara, R. Yuge, T. Noguchi, K. Utsugi, Suppression of aluminum corrosion by using high concentration LiTFSI electrolyte, J. Power Sources 231 (2013) 234–238, https://doi.org/10.1016/j.jpowsour.2012.12.028.
- [15] J. Tong, X. Xiao, X. Liang, N. Von Solms, F. Huo, H. He, S. Zhang, Insights into the solvation and dynamic behaviors of a lithium salt in organic- and ionic liquid-based electrolytes, Phys. Chem. Chem. Phys. 21 (2019) 19216–19225, https://doi.org/ 10.1039/c9cn01848d
- [16] F. Wu, S. Fang, M. Kuenzel, G. Kim, F. Wu, S. Fang, M. Kuenzel, A. Mullaliu, J. Kim, X. Gao, Article Dual-anion ionic liquid electrolyte enables stable Ni-rich cathodes in lithium-metal batteries Dual-anion ionic liquid electrolyte enables stable Ni-rich cathodes in lithium-metal batteries, Joule 5 (2021) 2177–2194, https://doi.org/10.1016/j.joule.2021.06.014.

- [17] A.L. A. Habermann, M.G. Kolb, P. Maas, H. Kellermann, C. Rischmüller, F. Peter, Seitz, Study of a regional turboprop aircraft with hybrid-electric turboshaft assistance, Aerospace 10 (6) (2023) 529.
- [18] H. Löbberding, S. Wessel, C. Offermanns, M. Kehrer, J. Rother, H. Heimes, A. Kampker, From cell to battery system in BEVs: analysis of system packing efficiency and cell types, World Electr. Veh. J. 11 (2020) 1–15, https://doi.org/ 10.3390/wevj11040077.
- [19] S. Xu, M. Raghavan, The Influence of Vehicle Mass and Cell Type on eVTOL Battery Pack Designs, in: Eur. Conf. Mech. Sci, Springer, 2024, pp. 383–394.
- [20] R. Wei, S. Chen, T. Gao, W. Liu, Challenges, fabrications and horizons of oxide solid electrolytes for solid-state lithium batteries, Nano Select 2 (2021) 2256–2274.
- [21] D. Witt, D. Wilde, F. Baakes, F. Belkhir, F. Röder, U. Krewer, Myth and reality of a universal lithium-ion battery electrode design optimum: a perspective and case study, Energ. Technol. (2021), https://doi.org/10.1002/ente.202000989.
- [22] D. Witt, F. Röder, U. Krewer, Analysis of lithium-ion battery state and degradation via physicochemical cell and SEI modeling, Batter. Supercaps 5 (2022) e202200067.

- [23] V. Laue, N. Wolff, F. Röder, U. Krewer, Modeling the in fl uence of mixing strategies on microstructural properties of all-solid-state electrodes 1801049 (2020) 1–12, https://doi.org/10.1002/ente.201801049.
- [24] S. Toghyani, W. Cistjakov, F. Baakes, U. Krewer, Conceptual design of solid-state Li-battery for urban air mobility, J. Electrochem. Soc. 170 (10) (2023) 103510.
- [25] J. Chiew, C.S. Chin, W.D. Toh, Z. Gao, J. Jia, C.Z. Zhang, A Pseudo Three-dimensional Electrochemical-thermal Model of a Cylindrical LiFePO 4 / Graphite Battery, 2019, pp. 1–36.
- [26] MATLAB, 9.13.0.2126072 (R2022b). MA MathWorks Inc. https://www.math works.com, 2022.
- [27] J. Mao, W. Tiedemann, J. Newman, Simulation of temperature rise in Li-ion cells at very high currents, J. Power Sources 271 (2014) 444–454.
- [28] J.S. Ko, T. Hamann, J. Fortunato, V. Augustyn, Recent advances in electrolytes for enabling lithium-ion batteries across a wide temperature range, J. Phys. Chem. C 128 (8) (2024) 3113–3126.