

Performance improvement of hybrid electrolyte technology batteries for electrical propulsion in aerospace industry

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Abstract

In the aerospace industry, electrical propulsion promises to completely revolutionize the industry, with brand new architectures and ecosystems becoming tomorrow's reality. High storage capacity and low weight batteries are clearly crucial to all-electric and hybrid-electric aircrafts, and in order to begin to allow the reaction of products with commercially viable payload-range characteristics, it is generally accepted that electrical storage systems would need an energy density of at least 500 Wh/kg after 2025.⁶ Currently, the battery market is dominated by conventional Li-ion technology (LIB), which is reaching its theoretical limits (<300 Wh/kg).

Hybrid electrolyte with metallic lithium and Ni-rich cathode batteries are among the most promising candidates due to their many advantages, such as high specific energy density, high operating voltage, low self-discharge rate and no memory effect. However improvement of performance must be achieved through the optimal design of electrolyte and cathode materials.

In this regard, optimizing electrode design and hybrid electrolyte transport properties is a straightforward approach to increase the cell energy density. For this purpose, numerical modeling has been previously applied to understand the electrochemical system when defining the best electrode and electrolyte design. The operation of a lithium ion battery (LIB) follows porous electrode theory and electrochemical reaction thermodynamics and Newman et al. developed the governing equations¹³⁸ that later the community used for electrode design improvement purposes. For instance, *LiFePO₄* and nickel rich layered material *LiNi_{1-x-y}Mn_xCo_yO₂* (NMC) thick electrodes were optimized by a combined experimental and simulation approach^{7,5}. *LiNi_{0.8}Co_{0.15}Al_{0.05}O₂* (NCA)/graphite cell stack energy density was improved by a mathematical model.¹ Furthermore, the influence of the negative electrode design (in terms of porosity and electrode thickness) was studied under Newman continuum model¹².

The main aim in this work is to provide in-depth understanding of the cell performance and design improvement of the current status of the hybrid electrolyte technology through a novel approach relying on non-dimensional parameters. This approach arises as an appropriate tool to study the electrode performance since it brings the opportunity to identify the limiting factors in the battery performance by analyzing a somewhat reduced number of dimensionless parameters, which are obtained by condensating the huge number of physical, dimensional parameters involved in the description of the electrochemical phenomena taking place in the cell. Here, a parametric study is developed in order to identify the limiting mechanisms in each region of the parametric space of different Hybrid Electrolyte Batteries. For this purpose, battery consisting of porous cathodes (i.e NMC622), hybrid electrolyte and a lithium metal anode is examined and investigation of a much larger set of operation conditions of hybrid electrolyte based battery performance is also performed. By analyzing the resulting dimensionless parameters, the main physical phenomena that limit battery performance are identified (and rough estimates of voltage drops associated with each transport mechanism are obtained). Our resulting outcomes can be used as quantitative guidelines in the development and design of hybrid electrolyte batteries.

1. Introduction

There have been consistent upward trends in the electrification of aircraft systems and research into Electrical Propulsion. It is well known that electrification not only offers the capability to reduce emissions, but could also unlock the

HYBRID ELECTROLYTE TECHNOLOGY BATTERIES FOR ELECTRICAL PROPULSION IN AEROSPACE INDUSTRY

potential for more energy-efficient aircraft and brand new architectures and use cases. In this scenario, high battery storage capacity and low weight are clearly crucial to all-electric and hybrid-electric architectures, and in order to begin to allow the creation of product with commercially viable payload-range characteristics, it is generally accepted that electrical storage systems need an energy density of at least 500Wh/kg .⁶

Lithium-ion battery (LIB) has been a leading energy storage technology over the past decades because of its high energy density and long cycle life. However, recently LIBs have been carefully examined because of their apparent incompatibility between safety and high energy density.²

Metallic lithium anode in conventional LIBs with liquid electrolytes can present high energy density. However, the probability to suffer for leakage of the electrolyte if any holes are present is high; this is one of the main drawback of the conventional Li-ion battery. Another problem inherent in the liquid electrolyte battery is the formation of dendrites of lithium, which make it prone to explosion. In order to surmount these problems, a solid electrolyte can be positioned between the electrodes, being less hazardous and having a less flammable electrolyte-electrode system and better storage capacity. All solid state batteries are well established because of safety, lifetime, and achievable energy density. Substitution of liquid electrolyte by a solid allows simplification of the cell structure, and many restrictions in terms of architecture and safety are eliminated. Also the solid state electrolytes tend to last longer, as they undergo less wear and tear during operation, are more proof against shocks and vibrations, and can operate within a larger temperature range, up to about $200\text{ }^{\circ}\text{C}$. However, they have several disadvantages as well. Solid state batteries, are not suitable for use in low and ambient temperature conditions, and the power and current output is generally lower. This is because of the large ionic resistance of the electrolyte at room temperature, whereas this does not occur at elevated temperatures. In addition, at room temperature, the stress created at the electrode-electrolyte interface due to continuous contact with the solid electrolyte tends to reduce the longevity of the battery.

Solid polymer electrolytes based on poly(ethylene oxide) (PEO) have been intensively studied so far, because PEO can solvate various lithium salts, through interaction of its ether oxygens with cations. Nevertheless, their low ionic conductivities at ambient temperature preclude their practical applications for use in lithium batteries that operate at room temperature. In this paper, a high performance cell is studied based on a flexible hybrid solid electrolytes proposed by Park and col.¹⁰ and composed of poly(1,4-butylene adipate) (PBA)-based solid polymer electrolyte and Li^+ conductive LAGP, which exhibited superior electrochemical properties and good interfacial contact towards electrodes in the cell.

Among the different cathode materials available, NMC-type layered oxides are the most established positive electrode material due to their high properties, in terms of voltage and capacity. Research towards the improvement of NMC cathode materials have focused on the optimization of its specific energy by increasing the amount of nickel within the cathode slurry, such as in the case of the $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ material.

In the present work, we propose to perform the analysis on this high energy type cell based on the novel metallic lithium anode combined with a nickel rich cathode (NMC622) and PBA-based hybrid solid electrolyte. The present document is structured in the following way: in section 2, the expected energy densities of the selected cells are presented, in the section 3 the p2D model simulations estimating cell real performance under certain conditions are introduced, in section 4 cell design improvement based on the dimensionless work is presented and lastly in the section 5, main conclusions of the proposed work are enumerated.

2. Design of a high energy cell

Gravimetric energy density requirements are the huge challenges among many of the requirements of batteries for electric propulsion of the aerospace sector. In this regards, several approaches have been used to increase the energy density of Li-ion batteries in the past, such as developing new active materials and optimizing cell engineering, materials processing, and quality control.

For a given active material, energy density of the electrode could be improved by engineering approaches, including increasing electrode thickness, reducing electrode porosity and decreasing the content of inactive materials (polymeric binder and conductive carbon) in the electrode components. Thus, thickening electrodes in batteries while making current collectors and separators thinner is one effective approach to continuously increasing the active material content for higher energy density batteries. In this line several notable works³⁴¹¹⁴ have demonstrated that modifications on electrode thickness, porosity and inactive material content can considerably improve battery performance.

HYBRID ELECTROLYTE TECHNOLOGY BATTERIES FOR ELECTRICAL PROPULSION IN AEROSPACE INDUSTRY

Today's best available Li-ion battery cells have an energy density of around 250 Wh/kg^{11,9}. Current traction batteries are to a large extent based on Li-ion chemistry which is expected to remain the technology of choice for still many years but their maximum energy density will be limited to approximately 300-350 Wh/kg. As mentioned previously, solid-state batteries incorporating a lithium metal anode and a nickel rich cathode have the potential to address the energy density issues of conventional lithium-ion batteries. Here, we are considering lithium metal anode combined with a NMC622 cathode and PBA-based hybrid solid electrolyte cell (Li-PBA-NMC622) as a baseline to demonstrate that the novel approach we are proposing (the study based on the non-dimensional model parameters) provides in-depth understanding on limiting factors, and guides on cell design optimization reducing the number of required experiments for the developing procedure.

Firstly proposed cell's theoretical energy density is calculated as a function of the cathode thickness. For this purpose the equation 1 is used and the theoretical capacity of $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ is assumed to be 175 mAh/g.

$$E = \frac{Q * V_{nom}}{A \sum_{i=1}^3 (L_i \rho_i)} \quad (1)$$

being Q , the assumed 175 mAh/g multiplied by the cathode material mass, V_{nom} nominal voltage of the cell, A the area of the cell, L_i thickness of each of the components, such as anode, cathode and separator and lastly ρ_i refers to the density of the components.

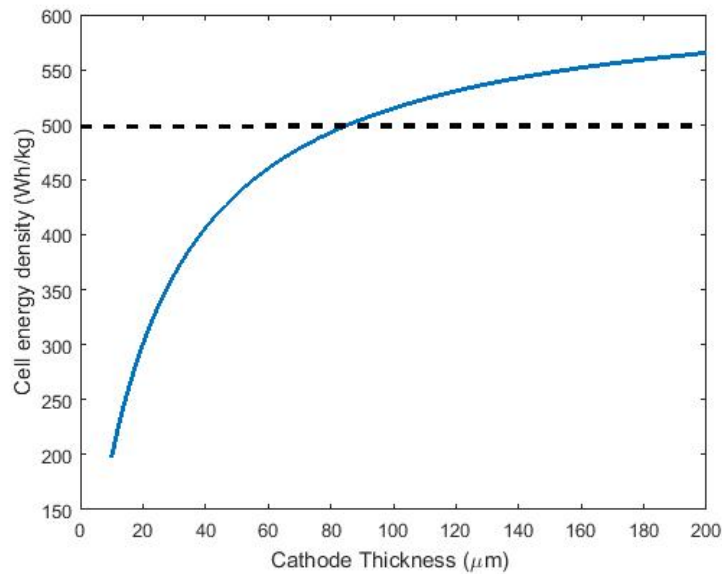


Figure 1: Theoretical Energy density prospect with respect to cathode thickness

According to the Figure 1, a direct relationship between the theoretical energy density and the cathode thickness is revealed. Looking at low values, corresponding to thin cathodes, the energy density is facing significant increase while in the case of thick cathodes, the growth of the energy density is reduced. In the same Figure (in dash line) the previously mentioned energy density target in aerospace industry is also included. The intersection of these two lines (theoretical energy density with the target value), could provide the estimation of the minimum required value of electrode thickness for meeting the target. Thus, in this case considering the selected cathode, we could anticipate that with a design of $87\mu m$, one could presumably reach the target of having a 500 Wh/Kg cell.

3. Cell performance under different load conditions

In the present work, a continuum pseudo-two-dimensional (p2D) model (hereafter full p2D) is used to quantify the effect of cathode thickness on the expected cell energy densities. This model was firstly developed by Newman's group and is relying on a physics based model using porous electrode and concentrated solution theories.⁸ The full p2D model is composed by two charge transport equations (one for the electronic current in the active material and another for the ionic current in the electrolyte phase) and two lithium transport equations (one for the transport in the electrolyte and

another one for the lithium diffusion in the active material particles). In addition, lithium intercalation kinetics (both at the surface of the active material particles and at the surface of the lithium foil) are included in order to close the model.

Firstly, the full p2D model is established for a Li-PBA-NMC622 battery. Table 1 shows the physical parameters used for the simulations, the ones related to the solid phase of the cathode material were measured experimentally in the lab, while the parameters related to the ionic phase such as the ionic conductivity were taken from Park and col.¹⁰ and electrolyte diffusion coefficient and transference number were set fixed as $2.4 \times 10^{-12} \text{ m}^2/\text{s}$ and 0.2, respectively.

Table 1: Physical parameters considered for p2D simulations

Parameter	Separator	Positive electrode	Source
Thickness (μm)	25	10 – 80	Assumed
Particle radius (m)	–	4.5×10^{-6}	Estimated
Solid phase diffusion coefficient (m^2/s)	–	1×10^{-13}	Estimated
Kinetic constant ($\text{mol}^{2.5}m^{-0.5}$)	–	5.4×10^{-11}	Estimated
Active material volume fraction	–	0.379	Estimated
Electronic conductivity of solid active material (S/m)	–	40	Estimated
Maximum solid phase concentration (mol/m^3)	–	48450	Calculated
Stoichiometric window	–	0.01 – 0.99	Literature
Charge transfers coefficients	–	0.5	Literature
Electrolyte phase volume fraction	1	0.399	Assumed
Initial salt concentration (mol/m^3)	1000	100	Assumed
Transference number (m)	0.2	0.2	Assumed
Electrolyte phase diffusion coefficient (m^2/s)	2.4×10^{-12}	2.4×10^{-12}	Assumed
Ionic Conductivity (S/m)	0.0038	0.0038	Literature ¹⁰

Similar to the work proposed by Mei and col.¹⁵ one can use the full p2D model simulations for understanding the cell limitations. Running our full p2D model implemented in MATLAB environment one can predict cell performance in voltage profile like shown in Figure 2, and later calculate the energy density as a result of this performance. Additionally, full p2D model simulations results provide better understanding on cell behaviour due to the fact that one can analyze the evolution of the internal variables such as ionic potential, electronic potential as well as the concentration along the electrolyte and solid phase, and therefore gain understanding on limiting factors.

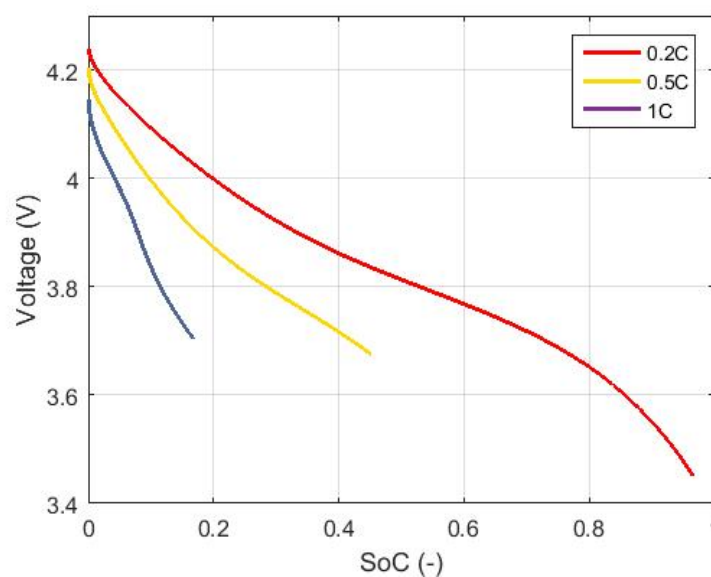


Figure 2: Cell performance for $30 \mu\text{m}$ cathode under different operating conditions

Here, we propose to use our p2D model simulations for calculation of energy density as a function of different parameters and operating conditions. Thus, the effects of the electrode design parameters (electrode thickness) and

HYBRID ELECTROLYTE TECHNOLOGY BATTERIES FOR ELECTRICAL PROPULSION IN AEROSPACE INDUSTRY

C-rate conditions on the battery performance (energy density) are initially investigated. Figure 3 shows the resulting values together with the theoretical energy density coming from Figure 1.

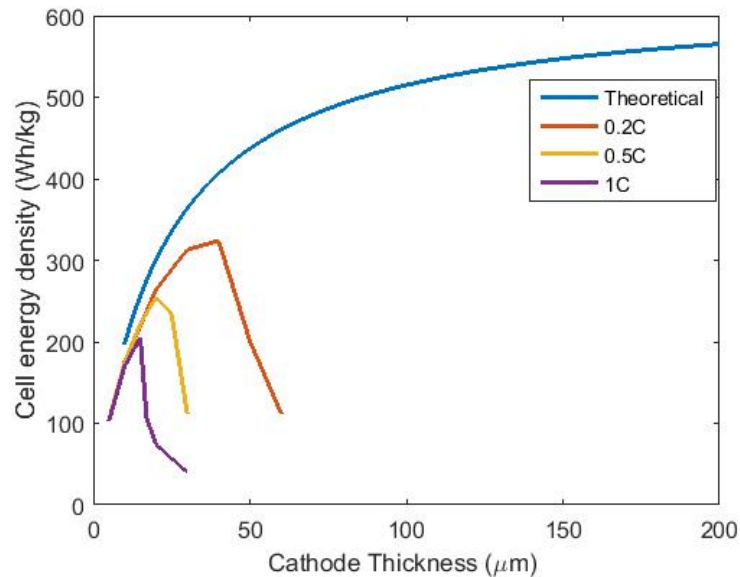


Figure 3: Energy Density

Figure 3 shows the energy density versus electrode thickness under three different C-rates, providing a good indication of the selected cell expected performance. Looking at the above Figure it can be predicted that thicker electrodes lead to higher energy density until a critical electrode thickness for each of the C-rate discharge profiles is reached, leading to maximum points in energy density versus thickness curves.

At low C-rate discharge conditions (0.2C), the energy density maximum point is achieved at $30\mu\text{m}$; while, with an increase in the C-rate, the limiting factors were more profound in thick electrodes, moving to a maximum point at $15\mu\text{m}$.

Energy density improvement is based on the increased utilization of active materials and decreased cell polarization, and here is another goal of this work, propose new cell designs minimizing cell polarizations by addressing the understanding of individual cell polarization once the limiting factors are identified.

In the present work, we have built a non-dimensional p2D model based on the full p2D model and we are proposing to analyze the generated non-dimensional parameters defined in Table 2 for the understanding of the existing limitations in the cell, which is the scope of this work.

Table 2: Obtained dimensionless parameters

$\frac{1}{\delta_\sigma}$	$\frac{\sigma^{eff} RT}{L_0 \alpha F I_0}$	~ electronic conduction / intercalation kinetics
$\frac{1}{\delta_K}$	$\frac{K^{eff} RT}{L_0 \alpha F I_0}$	~ ionic migration / intercalation kinetics
$\frac{1}{\delta_{k_D}}$	$\frac{K_D^{eff} C_e^{ref}}{L_0 I_0 C_e^0}$	~ ionic diffusion/ intercalation kinetics
$\frac{\Delta C_e^{ref}}{C_e^0}$	$\frac{I_0 L_0}{D_e^{eff} F C_e^0}$	~ Current induced concentration difference / initial salt concentration
τ_e^{eff}	$\frac{t_e D_e^{eff}}{\epsilon_e L_0^2}$	~ discharge characteristic time / electrolyte diffusion characteristic time
τ_s	$\frac{t_e D_s}{r_p^2}$	~ discharge characteristic time / solid diffusion characteristic time
S_s	$\frac{I_0 r_p^2}{3 \epsilon_e L_0 F C_s^{max} D_s}$	~ intercalation kinetics / solid diffusion flux
\hat{k}	$\frac{\alpha_s F k_0 L_0}{C_e^0} (C_e^0)^\alpha (C_s^{max})^{2\alpha}$	~ dimensionless preexponential factor / intercalation kinetics
\hat{k}_{Li}	$\frac{F k_{Li}}{I_0} (C_e^0)^\alpha$	~ preexponential factor / Li stripping–plating kinetics

According to the Figure 1, around $87\mu\text{m}$ thickness cathode would be enough to conduct a 500Wh/kg cell performance when no limiting factors are presented in the cell. However, the electrochemical cells are a multi-scale, multi-phase systems that involve multiple processes occurring simultaneously, and from the p2D model simulation results it can be anticipated that the performance of the selected cell would be very far from the theoretical values. Hereafter, non-dimensional p2D parameters are calculated to a $87\mu\text{m}$ thickness cathode $\text{Li} - \text{PBA} - \text{NMC622}$ cells with exactly the same parameters shown in Table 1 in order to get a more detailed understanding of the electrochemical system constrains when thick electrodes are used.

Table 3: Obtained dimensionless parameters

Condition	$\frac{1}{\delta_\sigma}$	$\frac{1}{\delta_K}$	$\frac{1}{\delta_{k_D}}$	$\frac{\Delta C_e^{ref}}{C_e^0}$	τ_e^{eff}	τ_s	S_s	\hat{k}	Φ	\hat{k}_{Li}
0.2C - $87\mu\text{m}$	1469,008	0,160	0,496	3,86	4,322	8,888	0,028	5148176,074	0,094	0,366
0.5C - $87\mu\text{m}$	587,603	0,064	0,496	9,661	1,729	3,555	0,071	2059270,429	0,094	0,146
1C - $87\mu\text{m}$	293,801	0,032	0,496	19,323	0,864	1,777	0,142	1029635,215	0,094	0,073

Let us consider first the **0.2C - $87\mu\text{m}$ case** in Table 3. From the obtained non-dimensional numbers, it is readily anticipate that polarization from electronic transport can be assumed negligible. Since $\frac{1}{\delta_\sigma}$ is lower than 1 and $\frac{\Delta C_e^{ref}}{C_e^0}$ higher than 2, polarization from ionic transport will show up as well as Li^+ depletion in the electrolyte phase phenomena might appear. From the large value of τ_e^{eff} (i.e. a small ratio of electrolyte diffusion time to discharge time) results a quasi-steady lithium concentration profile in the electrolyte. Non-dimensional number τ_s (ratio of discharge time to particles diffusion time) is large and S_s number quite small predicts that polarization from solid-phase diffusion will be small. Concerning kinetics, value of \hat{k} anticipates a rather low polarization associated to surface reaction on the particles (until saturation of these particles begins to happen). \hat{k}_{Li} instead is not large, and a noticeable polarization due to lithium stripping/plating at the lithium foil surface will be observed.

Now, looking at the table for non-dimensional numbers corresponding to the **0.5C - $87\mu\text{m}$ case** and comparing these numbers with those for the **0.2C - $87\mu\text{m}$ case** some predictions can be made: (a) polarization contribution from electronic transport is still negligible, (b) contributions from lithium foil surface reaction will be essentially the same, (c) contribution from surface reaction on particles surface is smaller (since \hat{k} is larger) and slightly higher polarization contribution from liquid-phase transport as well as higher electrolyte depletion phenomena are expected in this case (remark that $1/\delta_K$ is lower and $\frac{\Delta C_e^{ref}}{C_e^0}$ higher than in the case of **0.2C - $87\mu\text{m}$**). Similar quasi-steady lithium concentration profile in the electrolyte is expected and the values of τ_s and S_s indicates that the ratio of discharge time to particles diffusion time has started to decrease and still the polarization from solid-phase diffusion will be small.

Non-dimensional numbers for the **1C - $87\mu\text{m}$ case** anticipate relatively large polarization contributions from several mechanisms. Namely, (a) large polarization contribution from lithium foil surface reaction is expected (since \hat{k}_{Li} number is quite small), (b) contribution from particle surface reaction is negligible, (c) solid phase polarization

contributions are started to be noticeable for wider ranges of cell utilization (since neither $1/\tau_s$ is getting close to 1 and S_s is increasing), and (d) liquid-phase transport polarization contribution is expected (since $1/\delta_K$ is very low). The latter being the most relevant one. Concerning the values of $\frac{\Delta C_e^{eff}}{C_e^0}$ ($\gg 2$) it should be emphasized that an important lithium depletion will show up in the electrode. This reduction of lithium in the electrolyte will, in turn, affect to the surface reaction giving rise to even larger polarization contribution since overpotential must compensate the reduction of the pre-exponential factor (proportional to a power of C_e).

All in all, the main limiting factors for achieving high performance in thick electrodes solid electrolyte cells can be summarized to be the high ionic polarization and the depletion of Li^+ concentration along the electrolyte phase.

4. Cell design improvement

In our work, all the factors that minimize the above limiting factors could be considered. However, with the aim of improving cell performance we have concentrate our effort on cathode design improvement through cell engineering.

Focusing in the identified two main limiting phenomena, i.e. ionic charge transport polarization and electrolyte depletion, possible approaches include but are not limited to increasing Li^+ ionic conductivity (K), to increasing porosity (ϵ_e), to decreasing tortuosity (τ_e), to increasing Li^+ diffusion coefficient (D_e) and/or to increasing salt concentration (C_e^0). In the present work, we have consider three electrode design parameters, namely, electrolyte salt concentration, porosity and tortuosity. With a simple script built in MATLAB for calculating the non-dimensional parameters as a function of these three design parameters, one can gain in-depth understanding on how to minimize the aforementioned limiting factors. In particular, starting from a set of parameters created as linearly spaced vectors going from 1000 to $2500\text{mol}/\text{m}^3$ as salt concentration, from 0.3 to 0.4 as porosity in the cathode electrode, and from 2 to 0.5 as tortuosity, all the possible combination solutions are calculated. The effective parameters are calculated following the Equation 2.

$$X^{eff} = X \frac{\epsilon_e}{\tau} \quad (2)$$

being X parameters such as ionic conductivity (K) and electrolyte diffusion coefficient (D_e). From all possible combinations solutions, especially considering the tortuosity and porosity parameters, the effective ionic conductivity values are calculated. Figure 4 shows the influence of the effective ionic conductivity on the dimensionless parameter $1/\delta_K$ at different C-rate conditions. In the same Figure, a threshold value of the non-dimensional value is also added, meaning that above this value (~ 1), the limitation of the ionic charge transport could be minimized.

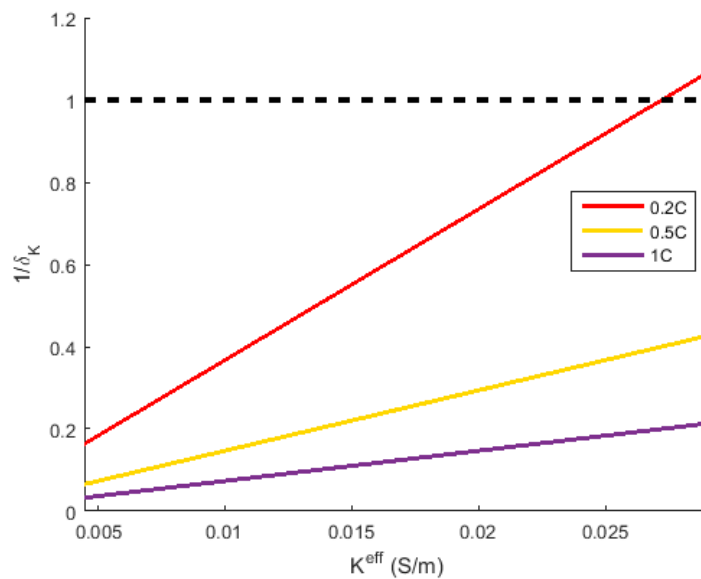


Figure 4: Non dimensional $1/\delta_K$ vs Cathode electrode effective ionic conductivity (S/m)

Taking into account the values in Figure 4, it can be predicted that at high C-rate conditions (i.e. 0.5C and 1C), there is no opportunity to avoid low values of $1/\delta_K$, meaning that there is no way to skip the ionic charge transport

limitation. Focusing on low C-rate conditions, it is estimated that we would need to manufacture a cathode with an effective ionic conductivity of 0.028 in order to minimize the ionic transport limitation. In other words, and looking at the Figure 5, where the relationship between cathode's effective ionic conductivity and design parameters such as porosity and tortuosity is shown, the values in these two parameters should be around 0.4 and 0.5, respectively for ensuring the minimization of the limiting factors.

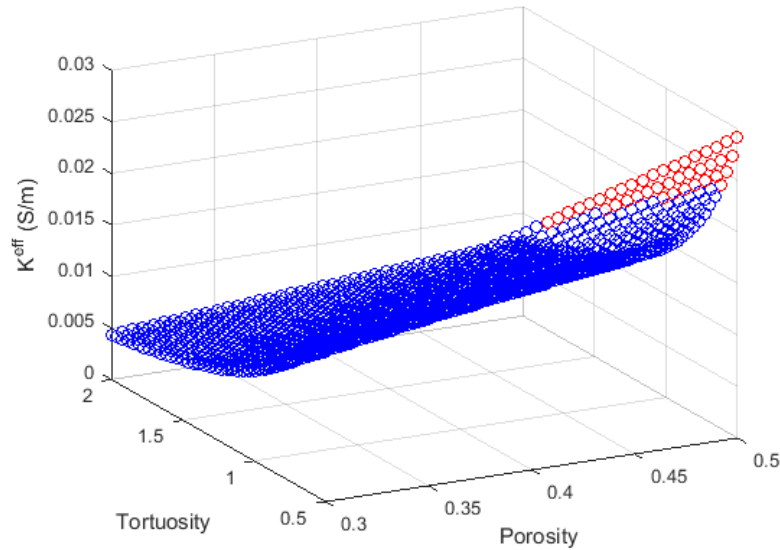


Figure 5: Calculated cathode electrode effective ionic conductivity as a result of the set tortuosity and porosity values

Additionally, to minimize the limiting factor related to the Li^+ concentration depletion, the influence of the new initial salt concentrations was studied. In Figure 6 we depict the correlation between the initial salt concentration and the non-dimensional parameter $\Delta C_e^{ref}/C_e^0$ for each of the applied currents.

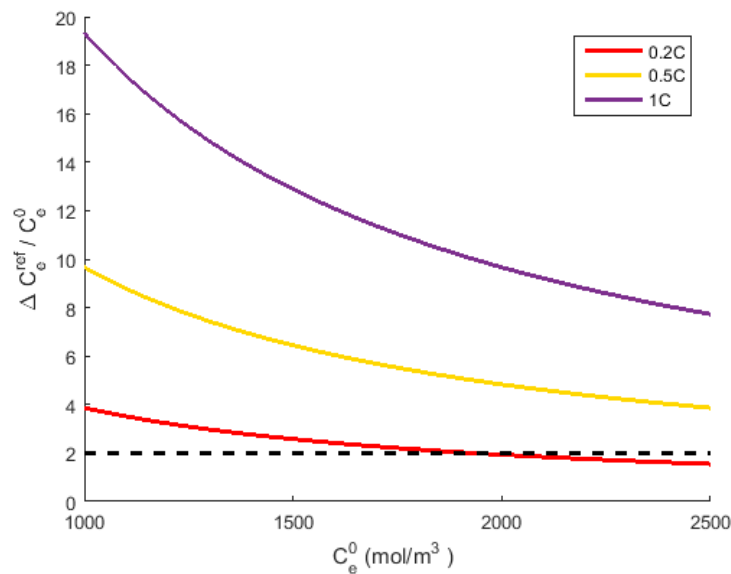


Figure 6: Calculation of the $\frac{\Delta C_e^{ref}}{C_e^0}$ as a function of the initial salt concentration and different C-rate conditions

In a similar way, as occurred for the ionic charge transport limitation understanding, in the case of the high C-rate conditions there is no option to minimize the limiting factor due to the electrolyte depletion phenomena, in the whole range of the initial concentration values, $\frac{\Delta C_e^{ref}}{C_e^0}$ is always superior to 2. However, for the case of low C-rate conditions,

HYBRID ELECTROLYTE TECHNOLOGY BATTERIES FOR ELECTRICAL PROPULSION IN AEROSPACE INDUSTRY

we would need to build a cathode with solid polymer with $1800\text{mol}/\text{m}^3$ salt concentration to avoid the aforementioned limitations.

All in all, the estimated optimized electrode design parameters are around $2000\text{mol}/\text{m}^3$ of salt concentration, 0.5 of tortuosity and 0.4 of porosity. Once the new design parameters are identified, a full p2D model simulation is run incorporating the new parameters as well as the new cathode thickness, which was identified as $87\mu\text{m}$. By doing this new simulation, we aim to confirm the minimization of the main predicted limiting factors such as the ionic charge transport limitation and electrolyte depletion. Thus, full p2D model was launched under 0.2C condition and compared to the original cell performance in Figure 7. According to the results, it is confirmed that the cell performance was improved significantly, by minimizing the main limiting factors.

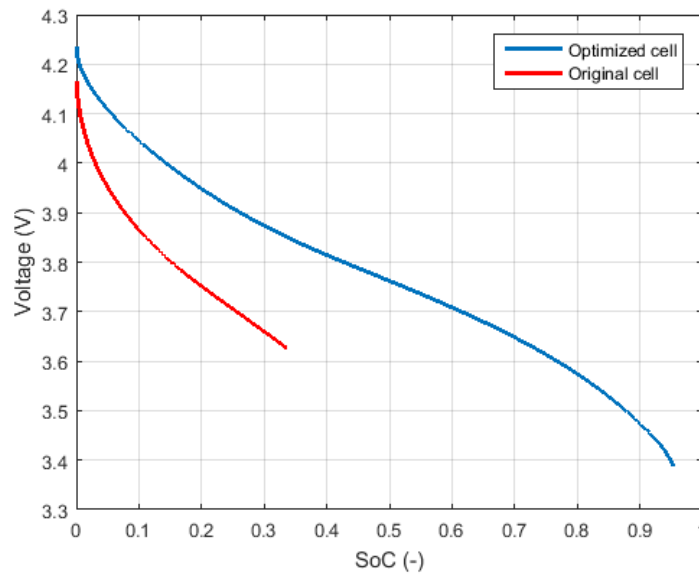


Figure 7: Voltage profile of an optimized cell design compared to the original cell

According to the simulation shown in Figure 7 the cell energy density reached above the $430\text{ Wh}/\text{kg}$, being significantly higher the one predicted by the initial p2D model with the original cell design. In this way, we are demonstrating that establishing synergies between laboratory testing experiments and this novel method based on the non-dimensional analysis will provide a more guided experimental work based on predictions and setting out an efficient experimental matrixes, all this thanks to a greater understanding of material properties and cell behaviour.

5. Conclusion

With the aim of designing a high energy cell for aerospace industry and to avoid today's ad-hoc rules in the development procedure, a novel method for better understanding of material properties and cell behavior, and therefore reducing the number of trial and error experiments, is presented in this work. Thus, a non-dimensional p2D model was conducted to investigate the limiting factors of the energy density of hybrid electrolyte technology batteries with thick cathodes.

With an increase in C-rate, the limiting factors were more profound in thick electrodes, leading to maximum points in energy density versus thickness curves. According to the study main cell polarization, such as ionic charge transport polarization, was found to offset the advantage of a high volume ratio of active materials in thick electrodes. Additionally Li-ion depletion in the electrolyte phase was predicted by the dimensionless analysis leading to a possible underutilization of active material.

Moreover, possible engineering approaches for cathode design improvement, such as increasing lithium salt concentration in the electrolyte, increasing porosity and reducing tortuosity parameter were demonstrated to overcome the limiting factors and extract the maximum energy density predicted by the full p2D model simulations.

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