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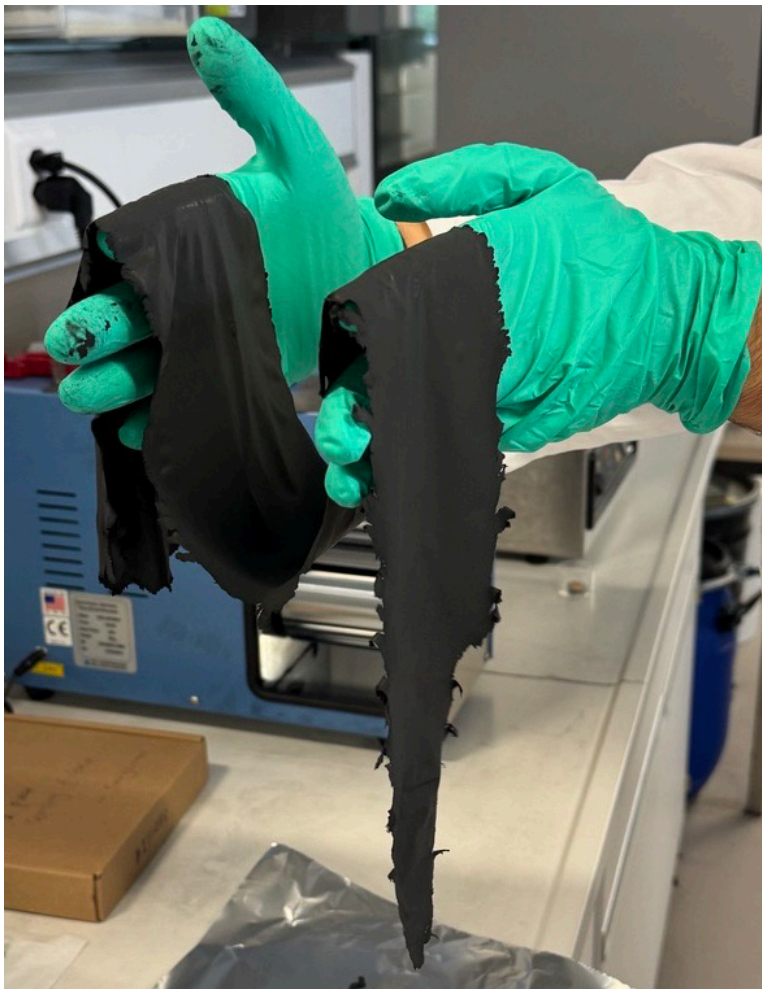
Final report from 05 May 2026

DECIDER

Dry Electrodes for Solid State Batteries



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Summary

To accelerate the market adoption of solid-state batteries, the development of high-capacity and cost-effective electrode materials compatible with solid electrolytes is crucial. The DECIDER project investigated the feasibility of dry battery electrode (DBE) processing as a manufacturing approach for high-loading electrodes (> 4 mAh cm^{-2}) in lithium metal batteries, targeting both liquid and solid-state electrolyte systems. DBE technology was selected for its potential to reduce environmental impact and manufacturing costs while enabling high areal capacities.

Within the project, dry-processed cathodes were successfully fabricated using either a pure PTFE (Polytetrafluorethylen) binder or a hybrid binder system combining PTFE with a biodegradable polymer. In parallel, thin lithium metal anodes (25 µm) were produced in-house using a thermal evaporation technique. These components were subsequently integrated into both coin cells and monolayer pouch cells, which were successfully assembled and electrochemically assessed.

Electrochemical testing revealed limitations in cycling stability. These issues were primarily attributed to the poor compatibility of PTFE-based cathodes with carbonate-based liquid electrolytes, resulting in insufficient electrode wettability, as well as to the high chemical reactivity of lithium metal with this class of electrolytes. The use of an ether-based electrolyte led to improved cathode wettability; however, its limited oxidative stability at high operating voltages restricted long-term cyclability. In addition, dry-processed cathodes were tested against lithium metal anodes using gel polymer electrolytes (GPEs) developed and prepared in-house, for which acceptable and encouraging cycling performance was achieved at slow current rate (0.1C), exceeding 100 cycles.

Overall, the results demonstrated the technical feasibility of DBE processing for high-loading electrodes in lithium metal battery configurations, while highlighting critical interfacial and electrolyte-related challenges. Further optimization of electrolyte formulations and lithium metal coating strategies is required. Post-DECIDER project activities are continuing to address these aspects, with the objective of further improving the cyclability and robustness of high-loading, dry-processed cathodes paired with thin lithium metal anodes using either liquid or gel polymer electrolytes.

Zusammenfassung

Um die Markteinführung von Festkörperbatterien zu beschleunigen, ist die Entwicklung hochkapazitiver und kosteneffizienter Elektrodenmaterialien, die mit Festelektrolyten kompatibel sind, von entscheidender Bedeutung. Das DECIDER-Projekt untersuchte die Machbarkeit der Trockenelektrodenherstellung (Dry Battery Electrode, DBE) als Fertigungsansatz für hochbeladene Elektroden (> 4 mAh cm^{-2}) in Lithium-Metall-Batterien, sowohl für Systeme mit flüssigen Elektrolyten als auch für Festkörperelektrolyte. Die DBE-Technologie wurde aufgrund ihres Potenzials ausgewählt, die Umweltbelastung und die Herstellungskosten zu reduzieren und gleichzeitig hohe flächenbezogene Kapazitäten zu ermöglichen.

Im Rahmen des Projekts wurden trockengefertigte Kathoden erfolgreich hergestellt, entweder unter Verwendung eines reinen PTFE-Binders oder eines hybriden Bindersystems, das PTFE (Polytetrafluorethylen) mit einem biologisch abbaubaren Polymer kombiniert. Parallel dazu wurden dünne Lithium-Metall-Anoden (25 µm) in-house mittels thermischer Verdampfung hergestellt. Diese Komponenten wurden anschließend sowohl in Knopfzellen als auch in einlagige Pouch-Zellen integriert, die erfolgreich assembliert und elektrochemisch charakterisiert wurden.

Die elektrochemischen Tests zeigten Einschränkungen hinsichtlich der Zyklenstabilität. Diese wurden hauptsächlich auf die schlechte Kompatibilität der PTFE-basierten Kathoden mit karbonatbasierten flüssigen Elektrolyten zurückgeführt, was zu einer unzureichenden Benetzung der Elektroden führte, sowie auf die hohe chemische Reaktivität von Lithium-Metall gegenüber dieser Elektrolytklasse. Der Einsatz eines etherbasierten Elektrolyten verbesserte die Benetzbarkeit der Kathoden; jedoch begrenzte dessen eingeschränkte oxidative Stabilität bei hohen Betriebsspannungen die langfristige Zyklenfähigkeit.

Zusätzlich wurden die trockengefertigten Kathoden gegenüber Lithium-Metall-Anoden unter Verwendung von gelpolymeren Elektrolyten (GPEs), die in-house entwickelt und hergestellt wurden, getestet. Dabei wurde bei niedriger Stromdichte (0,1C) eine akzeptable und vielversprechende Zyklenleistung erzielt, mit mehr als 100 Lade-/Entladezyklen.

Insgesamt zeigten die Ergebnisse die technische Machbarkeit des DBE-Verfahrens für hochbeladene Elektroden in Lithium-Metall-Batteriekonfigurationen auf, während gleichzeitig kritische Herausforderungen im Zusammenhang mit Grenzflächen und Elektrolyten identifiziert wurden. Eine weitere Optimierung der Elektrolytformulierungen sowie der Beschichtungsstrategien für Lithium-Metall ist erforderlich. Die Aktivitäten im Anschluss an das DECIDER-Projekt werden fortgesetzt, um diese Herausforderungen gezielt anzugehen, mit dem Ziel, die Zyklenstabilität und Robustheit hochbeladener, trockengefertigter Kathoden in Kombination mit dünnen Lithium-Metall-Anoden unter Verwendung von flüssigen oder gelpolymeren Elektrolyten weiter zu verbessern.

Résumé

Afin d'accélérer l'adoption sur le marché des batteries tout solide, le développement de matériaux d'électrodes à haute capacité, économiquement viables et compatibles avec les électrolytes solides est crucial. Le projet DECIDER a étudié la faisabilité du procédé de fabrication des électrodes de batterie par voie séche (dry battery electrode, DBE) en tant qu'approche de fabrication pour des électrodes à haute capacité (> 4 mAh cm^{-2}) dans des batteries au lithium métal, ciblant à la fois des systèmes à électrolyte liquide et tout solide. La technologie DBE a été retenue pour son potentiel à réduire l'impact environnemental et les coûts de fabrication, tout en permettant d'atteindre des capacités surfaciques élevées.

Dans le cadre du projet, des cathodes produites par voie sèche ont été fabriquées avec succès en utilisant, soit un liant PTFE pur, soit un système de liant hybride combinant le PTFE (Polytetrafluorethylen) à un polymère biodégradable. En parallèle, des anodes fines en lithium métal (25 µm) ont été produites en interne à l'aide d'une technique d'évaporation thermale. Ces composants ont ensuite été intégrés dans des cellules type pile-bouton ainsi que dans des cellules type cellule poche, qui ont été assemblées et évaluées électrochimiquement avec succès.

Les essais électrochimiques ont mis en évidence certaines limitations en termes de stabilité au cours du cyclage. Celles-ci ont été principalement attribuées à la faible compatibilité des cathodes à base de PTFE avec les électrolytes liquides à base de solvants carbonates, entraînant une mouillabilité insuffisante des électrodes, ainsi qu'à la forte réactivité chimique du lithium métal avec cette famille d'électrolytes. L'utilisation d'un électrolyte à base d'éthers a permis d'améliorer leurmouillabilité à la surface des cathodes. Toutefois, sa stabilité oxydative limitée a restreint la cyclabilité à long terme, en particulier à des voltages supérieurs à 4.2V. Par ailleurs, les cathodes fabriquées par voie sèche ont été testées face à des anodes en lithium métal en utilisant des électrolytes polymères gélifiés (GPEs) développés et préparés en interne, pour lesquels des performances de cyclage acceptables et encourageantes ont été obtenues à faible régime de courant (0.1C), dépassant 100 cycles.

Généralement, les résultats ont démontré la faisabilité technique du procédé DBE pour des électrodes à haute capacité dans des configurations de batteries au lithium métal, tout en mettant en évidence des points critiques principalement liés aux interfaces et aux électrolytes. Une optimisation supplémentaire des formulations d'électrolytes et des stratégies de dépôt des revêtements afin d'optimiser l'interface avec le lithium métal est nécessaire. Les activités à la suite du projet DECIDER se poursuivent afin de lever ces verrous, avecl'objectif d'améliorer davantage la cyclabilité, et la stabilité des cathodes à haute capacité préparées par voie sèche, associées à des anodes fines en lithium métal, en utilisant des électrolytes liquides et polymères gélifiés.

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SFOE	Swiss Federal Office of Energy
DBE	Dry Battery Electrode
PTFE	Polytetrafluoroethylene
NMP	N-Methylpyrrolidone
PVDF	Polyvinylidene fluoride
XG	Xanthan Gum
LiM	Lithium metal
a-SEI	artificial solid electrolyte interphase
Al	Aluminium
Cu	Copper
cc	current collector
CNT	Carbon Nanotubes
VGCNF	Vapor Grown Carbon NanoFibers
NMC622	$LiNi_{0,6}Mn_{0,2}Co_{0,2}O_2$
SEM	Scanning Electron Microscopy
PVD	Physical Vapor Deposition
EDX	Energy-Dispersive X-ray spectroscopy
LiFSI	Lithium bis(fluorosulfonyl)imide
GPE	Gel Polymer Electrolyte

1 Introduction

1.1 Context and motivation

Since their commercialization in the 1990s, Lithium-ion batteries have been dominating the energy storage devices market. Simultaneously, high technology applications have been developing and demanding much higher-performance (higher power and energy density) energy storage devices to satisfy their needs (e.g., electric vehicles (EV), drones, etc.).[1] At this stage, Li-ion batteries are reaching their limits, and a need to develop new technologies to satisfy the new energetic demands is developing. Li metal batteries (batteries using metallic-lithium anodes instead of graphite) stand as one of the very promising technologies that can enable higher gravimetric and volumetric energy densities when used with either liquid or solid electrolytes. To achieve this goal huge efforts are made to enhance the cycling stability and safety of the lithium metal anode. However, to attain the full potential of using the lithium metal anode in either liquid or solid-state batteries, high loading cathode materials are required to achieve the highest possible energy density of the battery. Unfortunately, the existing slurry-based electrode preparation processes cannot achieve the required loadings (>4 mAh/cm^2).

The slurry-based processes involve some complex, toxic and high energy consuming procedures. Mostly, the electrode active material, an electronic conductive additive, and a conventional binder such as polyvinylidene fluoride (PVDF) are used with the chemically and thermally stable, but toxic and expensive mixing solvent named N-methyl-2-pyrrolidone (NMP) to enable the slurry preparation.

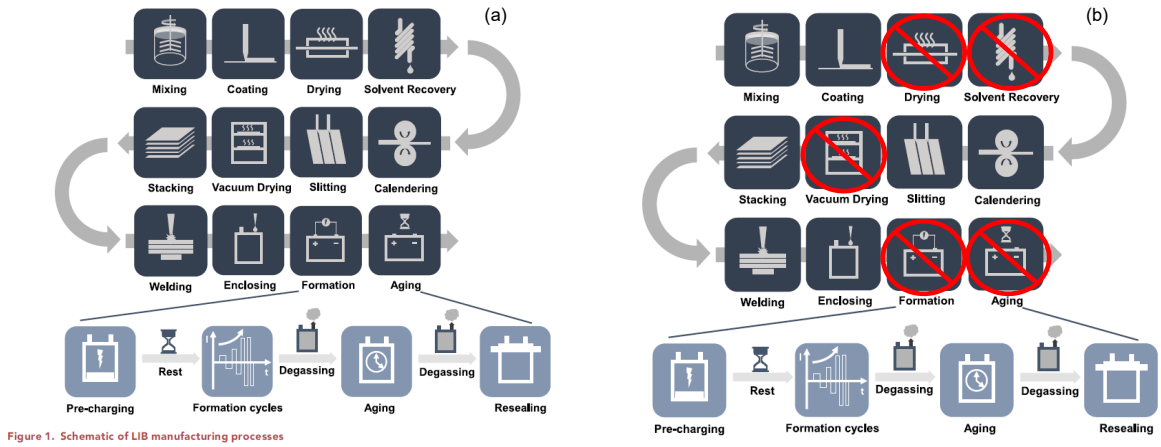

Figure 1. Schematic of LIB manufacturing processes

Figure 1: Fabrication steps for (a) traditional Li-ion batteries and (b) all dry solid-state batteries as targeted in this proposal.

After the mixing and coating steps, a drying process takes place to evaporate the solvent and deliver the dry coated electrodes. This drying step is one of the most limiting factors of the whole process. It adds some extra costs to recover the evaporated solvent, requires a large footprint, and meanwhile leads to some adhesion and cracking problems (during solvent evaporation) when high electrodes loadings are targeted. Moreover, the whole process is not adapted to some new innovative and promising

technologies such as all solid-state batteries. This incompatibility can be resumed in four main points: 1) Solvent sensitivity of the electrolytes, 2) the increase porosity of the electrodes after solvent evaporation, 3) the poor mechanical properties of the required high loading electrodes, and 4) the high production costs of the whole process.[2]

The electrode fabrication process determines the battery performance and is the major cost of the battery, as summarized in Table 1. Today, coating/drying and solvent recovery account for c.a. 20% of the cost per year and > 45% of the energy consumption per cell/kWh.[2]

Table 1: Cost, throughput, and energy consumption of Li ion battery manufacturing processes. [2]

Table 1. Cost, throughput, and energy consumption of LIB manufacturing processes

Manufacturing processes	Cost per year/$* (Nelson et al., 2019)	Percentage %	Throughput (Heimes et al., 2019a)	Manufacturing processes	Energy consumption per cell/kWh	Percentage %
Slurry mixing	7,396,000	7.91%	30 min–5 h	Slurry mixing	0.11	0.83%
Coating/drying	13,984,000	14.96%	35–80 m/min	Coating	0.18	1.36%
Solvent recovery	4,296,000	4.60%	NA	Drying/solvent recovery	6.22	46.84%
Calendering	4,849,000	5.19%	60–100 m/min	Calendering	0.38	2.86%
Slitting	2,891,000	3.09%	80—150 m/min	Slitting	0.71	5.35%
Vacuum drying	2,990,000	3.20%	12–30 h	Stacking	0.77	5.80%
Stacking	8,086,000	8.65%	NA	Welding	0.25	1.88%
Welding	6,864,000	7.34%	NA	Enclosing	0.69	5.20%
Enclosing	11,636,000	12.45%	Depend on the cell design	Formation/aging	0.07	0.53%
Formation/aging	30,482,750	32.61%	Up to 1.5–3 weeks	Dry room	3.9	29.37%

*The manufacturing cost includes equipment depreciation, labor cost, and plant floor space cost.
The labor cost was calculated based on the US average factory worker's salary of $15/h (Economic Research Institute, 2020).
The floor space cost was calculated based on $3,000/$m^2$ per year (includes rent, utility, and management) (Nelson et al., 2019).
The depreciation cost was calculated by 16.7% of capital investment and 5% of floor space cost (Nelson et al., 2019).

To overcome the issues and limitations brought with the wet processing of electrodes, and for more cost effective and compatible procedure enabling high loading electrodes for both liquid and solid-state electrolyte batteries, dry battery electrodes (DBE) processing was presented as an innovative approach. Indeed, DBE can potentially resolve the industrialization concerns of high loading electrodes and solid-state batteries at lower costs. This technology possesses unique advantages compared to the conventional slurry-based methods due to the solvent free process exhibiting environmental friendliness, low cost, enhanced compatibility, high production efficiency, and improved electrode performances. [2,3]

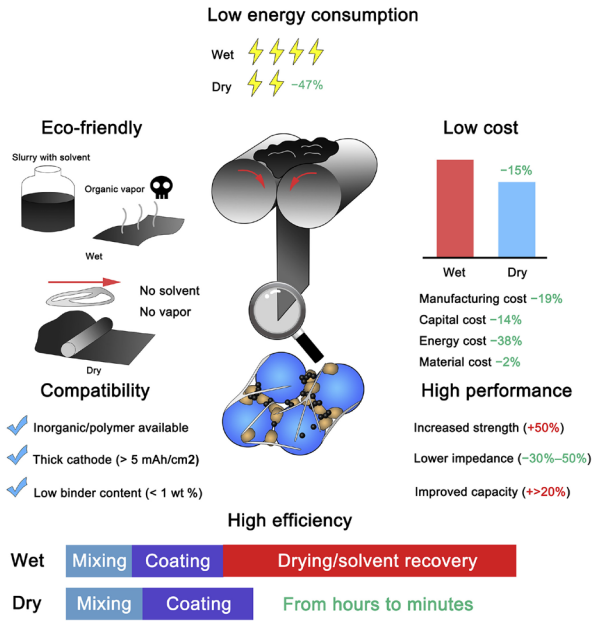

Figure 2: Overview of advantages of applying DBE technology in the battery industry [2]

DBE process consists only of three main steps: 1) Dry mixing of the active material, a conductive additive, and the binder, 2) High temperature pressing of the mixture to form a free-standing film, 3) Lamination of the free-standing film on the current collector to the desired thickness (electrode loading). Here the choice of the binder is very essential to get the required mechanical properties of the electrodes with good adhesion properties without any significant lowering of the active material ratio in the electrode composition. Fibrillation of the binder at high shearing force during the mixing step is a key-step to optimize the adhesion properties of the active material particles, homogeneity of the electrodes and to avoid the agglomeration of the binder which can limit both the ionic and electronic conductivity within the electrode and therefore lower the performance. In this regards, polytetrafluoroethylene (PTFE) is widely used as the binder of choice for cathode materials. [3]

Apart from its high potential to enable high loading electrodes for the next generation liquid based electrolyte batteries, DBE processing can enable the industrialization of the next generation all solid-state batteries without a need to readjust or build new production lines. Current Li-ion batteries have an energy density in the range of 300 Wh/kg (see Figure 3 (a)). Aside the limited energy density, safety, due to flammability of the electrolyte is a major concern for automotive applications. Nowadays, the solid-state battery, as one of the most promising energy storage systems, is developing rapidly, and numerous remarkable achievements have been reported at lab scale level. This technology holds the promise of increasing the energy density up to 500 Wh/kg and improve battery safety by replacing the liquid with a solid electrolyte (see Figure 3 (c)).

However, there are big gaps between lab-level demonstrations and practical applications due to the lack of adapted scale-up lines allowing the transformation to sheet-like forms of both electrodes and solid

electrolyte. To accelerate the market adoption of solid-state batteries, high-capacity materials which are cost effective and compatible with solid electrolytes, together with compatible production lines are essential. Therefore, the interest of developing the DBE processing appropriate to achieve the required transformations.[2,4] The fusion between high-capacity dry electrodes and solid-state polymer electrolytes, will bring new market opportunities where high capacity and safe batteries are needed (EV, aviation, …). As this technology is extremely promising to get high-capacity electrodes, it is really appealing for solid state batteries with Li metal anode.

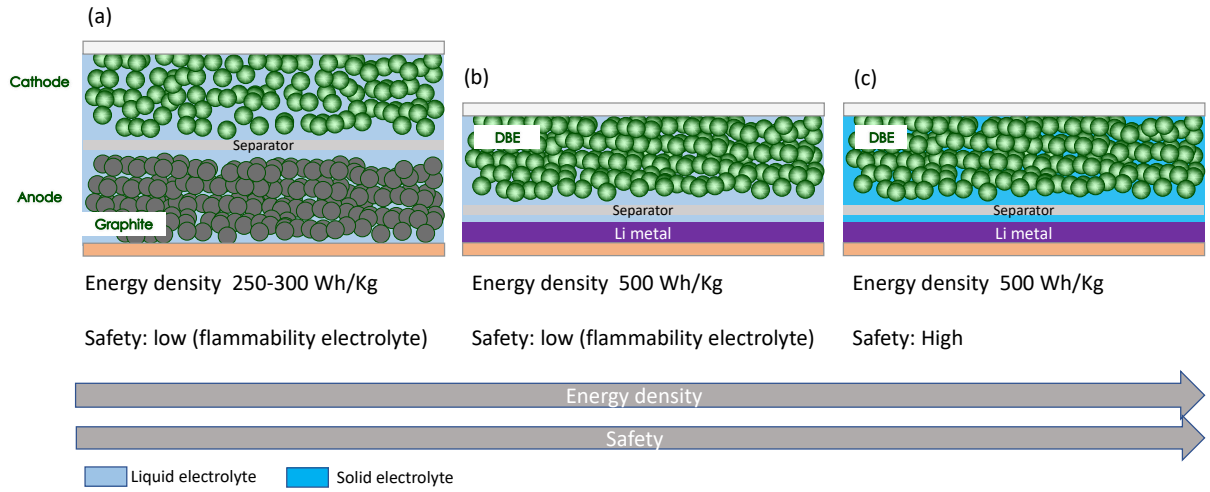

Figure 3: Sketch of traditional (a) Li ion battery, Li metal with DBE and Li metal in liquid (b) and solid (c) electrolytes.

1.2 Project objectives

The objectives of this project can be summarized as following:

- Optimization of the dry processing of electrode using a scalable process (mixing and calendaring conditions).
- Preparation of high loading (>3 mAh cm^{-2}) electrodes via dry process.
- Evaluate the quality of the prepared electrodes (homogeneity, mechanical properties, etc.) and their performance when integrated into battery cells in terms of capacity, cycling stability, safety and environmental profile.
- Investigate the feasibility of using dry electrodes with high loadings against lithium metal anode while using both liquid and polymer (solid) based electrolytes (see Figure 3 (b) and (c)).

2 Approach, method, results and discussion

2.1 Approach and Methods

In this project, our approach relied on the development and optimization of the dry processing of electrodes using an appropriate rotary mixer and a high-temperature calendaring machine. After achieving the targeted properties and performance of the dry-processed electrodes, thin lithium metal anodes were developed using a physical vapor deposition (PVD) method based on thermal evaporation. The developed dry-processed electrodes were then combined with the thin lithium metal anodes to assemble battery cells employing both liquid and solid polymer electrolytes.

Process Optimization

To develop the dry processing method for battery electrode fabrication, a rotary mixer (EL1 mixer from Eirich, an industrial high-intensity mixing machine) was selected, as it enables effective fibrillation of the PTFE binder during the mixing step. At the same time, the mixing conditions were maintained in a manner compatible with scale-up from laboratory to pilot scale. A calendaring machine with controlled temperature and friction was also used to achieve the targeted electrode thickness and optimized mechanical and electrochemical properties.

Screening of Different Binders

Following optimization of the electrode fabrication process, different grades of PTFE binders were screened to evaluate the impact of binder specifications on electrode properties, including mechanical integrity, adhesion, and electrochemical performance. In addition, the potential use of more environmentally friendly binders was assessed. In a first step, a cellulose-based binder was tested for dry electrode processing, then a hybrid binder containing PTFE and a biodegradable second binder was assessed and the properties of the resulting electrodes were analysed and reported.

Screening of Different Current Collectors

After selecting the most suitable binder, a screening of different aluminum (Al) current collectors with various primer coatings was conducted. The objective was to achieve optimal adhesion and electrical contact between the Al current collector and the dry-processed free-standing electrode films.

Choice of the Carbon Additive

To further improve the electrochemical performance of the dry-processed battery electrodes, different carbon additives were screened and analysed. This study aimed to enhance electronic conductivity and overall electrode performance.

Electrode Testing

The prepared electrodes were first tested in battery cells using thick lithium metal counter electrodes (≥ 100 µm) to minimize the influence of anode-side effects, such as lithium side reactions, and to focus specifically on the properties of the dry-processed cathodes.

Thin Lithium Metal Anodes

Thin lithium metal electrodes (25 µm) were prepared using the CSEM thermal evaporation system following a Best-Known Method (BKM) to obtain uniform and dense lithium films. An appropriate thin-film coating was applied to stabilize the interface between the lithium anode and the electrolyte. This type of coatings enables rapid Li^+ transport, mitigated Li^+ concentration gradients at the interface, and promotes a uniform current density distribution during cycling, resulting in more homogeneous lithium plating and stripping behaviour.

Polymer Electrolyte

To enable the use of dry-processed electrodes in all-solid-state battery configurations, a solid polymer electrolyte formulation already available at CSEM was employed (GPE1). A development of a new formulation was carried out to ensure compatibility with the properties of the dry-processed electrodes (GPE2).

Lithium Metal Battery Cell Testing (Liquid and Polymer Electrolytes)

Coin cells were assembled using dry-processed electrodes with the best optimized formulation and thin lithium metal anodes produced by PVD. Cathodes with different areal loadings were evaluated. Subsequently, monolayer pouch cells were assembled to reproduce and validate the results obtained at the coin cell level. Electrodes with dimensions of 4.4 × 5.7 cm^2 were used for pouch cell fabrication. Long-term galvanostatic cycling tests were performed at different current rates (0.1C, 0.2C, 0.5C and 1C).

2.2 Results and discussion

2.2.1. WP1: Dry electrodes preparation and optimization

WP1 covers the preparation and optimization of dry-processed electrodes and is structured into four closely interconnected tasks: T1-1 focuses on formulation development through optimization of the dry-component ratios; T1-2 addresses the optimization of the fabrication process, including mixing and calendaring conditions; T1-3 involves the characterization of electrode morphology, homogeneity, and component distribution; and T1-4 concerns the preliminary testing of the produced electrodes in coin cells using commercial thick lithium anodes to identify the most promising preparation routes. The work conducted across these tasks was carried out simultaneously due to their interdependencies, and the corresponding results are presented in the following sections.

The optimization of the dry-content composition was carried out in parallel with the optimization of the dry electrode processing steps (mixing and calendaring steps). Several binder grades with different fibrillation abilities from multiple suppliers were systematically screened (Table 2). These included PTFE binders (classified as PFAS materials) sourced from Chemours, Daikin and Sigma-Aldrich. In addition, a cellulose-based binder (sericin) was evaluated as an alternative PFAS-free binder to replace PTFE. Finally, hybrid binder systems combining PTFE grade with a biopolymer binder (Xanthan gum) were also investigated. Such hybrid formulations can reduce the overall PFAS content while offering additional advantages, including enlarged processing window and improved electrode wettability.

Table 2: Binders tested during the project (vs suppliers and properties).

Supplier	Binder	Ability to fibrillate?
Chemours	PTFE TE 5447A	Fibrillates easily
	PTFE TE 5448A	Requires high energy to fibrillate
Daikin	PTFE SG	Fibrillates easily
	PTFE 104	Requires high energy to fibrillate
	PTFE 107	Requires intermediate energy to fibrillate
Sigma-Aldrich	PTFE GF53268486	No fibrillation properties demonstrated
Nanografi	Sericin	No fibrillation properties demonstrated
TCI	Xanthan Gum	No fibrillation properties demonstrated but can be used in combination with PTFE grade to build hybrid binders

In this context, various electrode formulations were investigated, starting from compositions with relatively high binder and conductive carbon contents (approx. 5%wt) and progressively reducing these fractions while increasing the cathode active material content throughout the optimization process. Table 3 and Figure 4 summarize the different formulations that were tested during the project. Several types of carbon grades were tested, as well as two types of carbon-coated aluminium current collectors

(Al-CC), as shown in Table 3.

Table 3: Screening parameters for the tested formulations.

	Active material (NMC622)	Binder	Carbon Additive	Carbon type	Al-CC type
Minimal value	90%wt	1%wt	1%wt	C45 C65 CNT VGCNF Mix C45/CNT	Carbon-coated Al CC from 2 suppliers (Xiamen and Armor Battery Films)
Maximal value	98%wt	7%wt	5%wt		
Optimal value	**95.5%wt**	**1.5%wt**	**3%wt**		

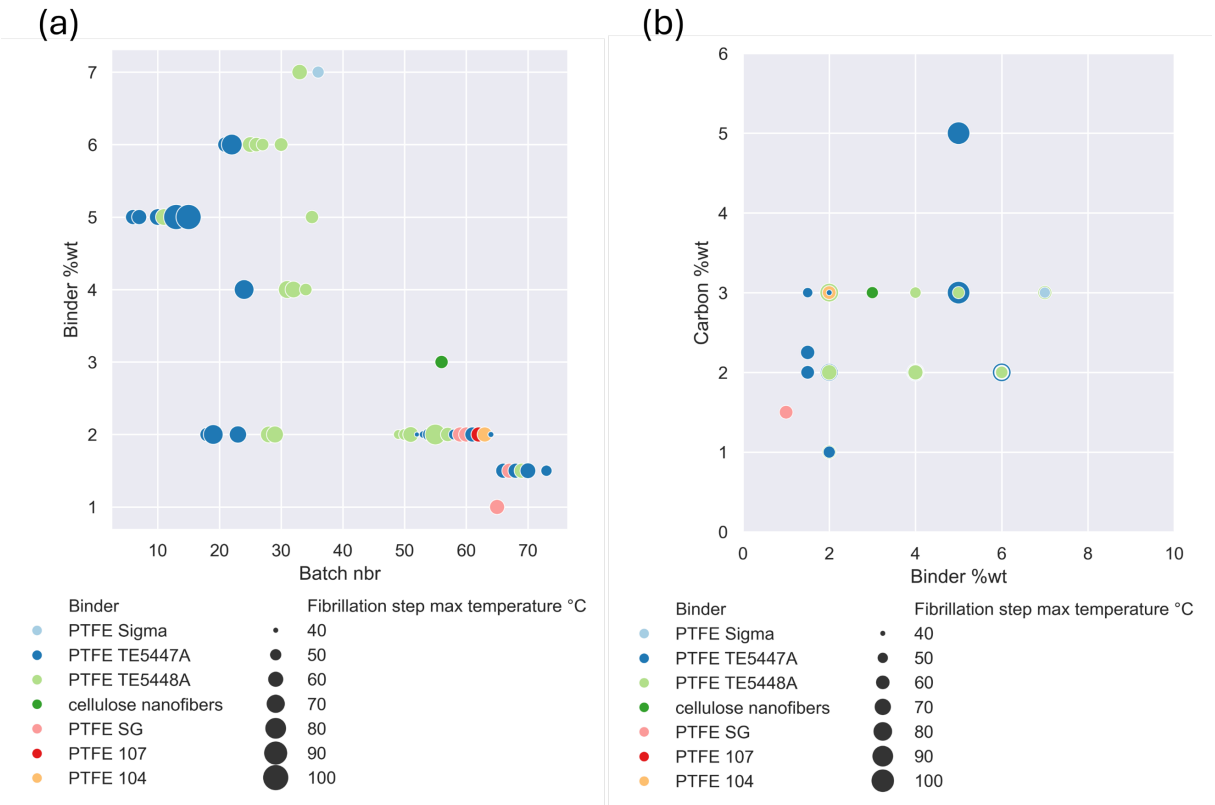

Figure 4: Summary of the different formulations tried vs PTFE grade and mixing conditions (to reach max temperature during the fibrillation step).

This work primarily focused on optimizing mixing parameters across the different formulations investigated. The mixing process was divided into four successive steps: (i) a premixing step to coat the NMC622 particles with the carbon additive, (ii) a low-speed mixing step for homogenous binder addition, (iii) a fibrillation step performed at a fixed high mixing speed and duration, during which different

temperatures could be reached, thereby influencing PTFE fibrillation, and (iv) a grinding step to optimize the size of the resulting aggregates (Figure 5). For each batch, powder samples were collected at different process stages and subsequently analysed by SEM to assess, for example, the efficiency of the carbon coating, and the presence and structure of the PTFE fibres.

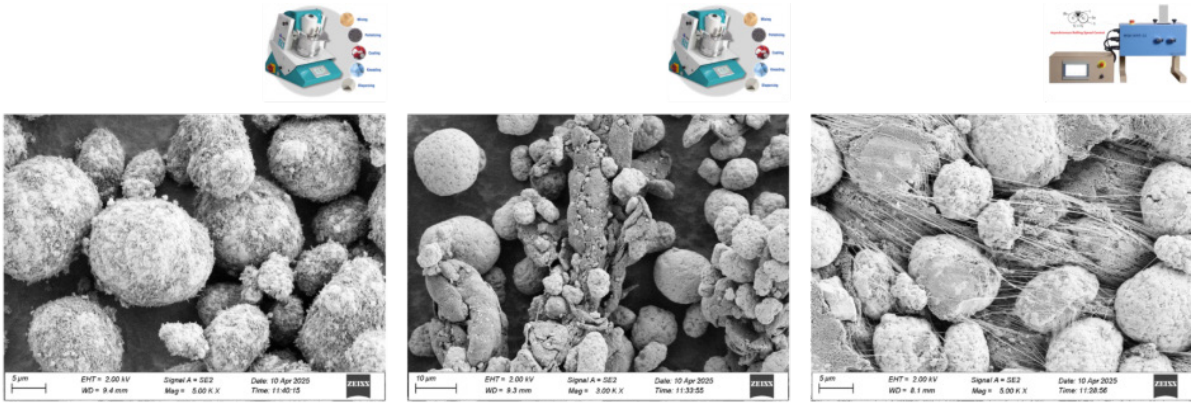

Figure 5 : Illustration of the typical steps in a solvent-free manufacturing process (a) Premixing of active cathode materials and carbon additives; (b) Addition of a polymer binder (PTFE) to the mixture and fibrillation step; (c) Film formation and lamination onto a current collector to obtain a dry-process cathode. The equipment used for each step is shown at the top of the figure.

In addition, the calendaring temperature, rolls gap, and applied friction during the calendaring process were systematically adjusted to obtain a homogeneous film with the targeted thickness and properties. During process development, multi-step calendaring strategies were also investigated while maintaining constant friction to achieve films with adequate areal loading. Tables 4 and 5 summarize the investigated processing conditions, while Figure 6 illustrates an example of calendaring process windows screening for a single batch formulation. For each condition, the ability to form a continuous film was first assessed. When successful, the resulting films were subsequently characterized by SEM (top and cross-section views), peel tests, and the electrochemical cycling performances in coin-cells.

Table 4: Mixing process optimization conditions that have been investigated.

	Fibrillation step / Mixing time (min)	**Fibrillation step / Max temperature reached (°C)**
Minimal value	5	40
Maximal value	120	100
Optimal value	**10**	**45-60**

Table 5: Calendaring process optimization conditions that have been investigated.

	Rolls Temperature (°C)	Applied friction (%)	Rolls gap (um)	Process steps
Minimal value	80	(-) 25	100	1 step
Maximal value	160	(-) 83	300	4 steps
Optimal value	**160**	**(-) 55**	**180**	**3 or 4 steps**

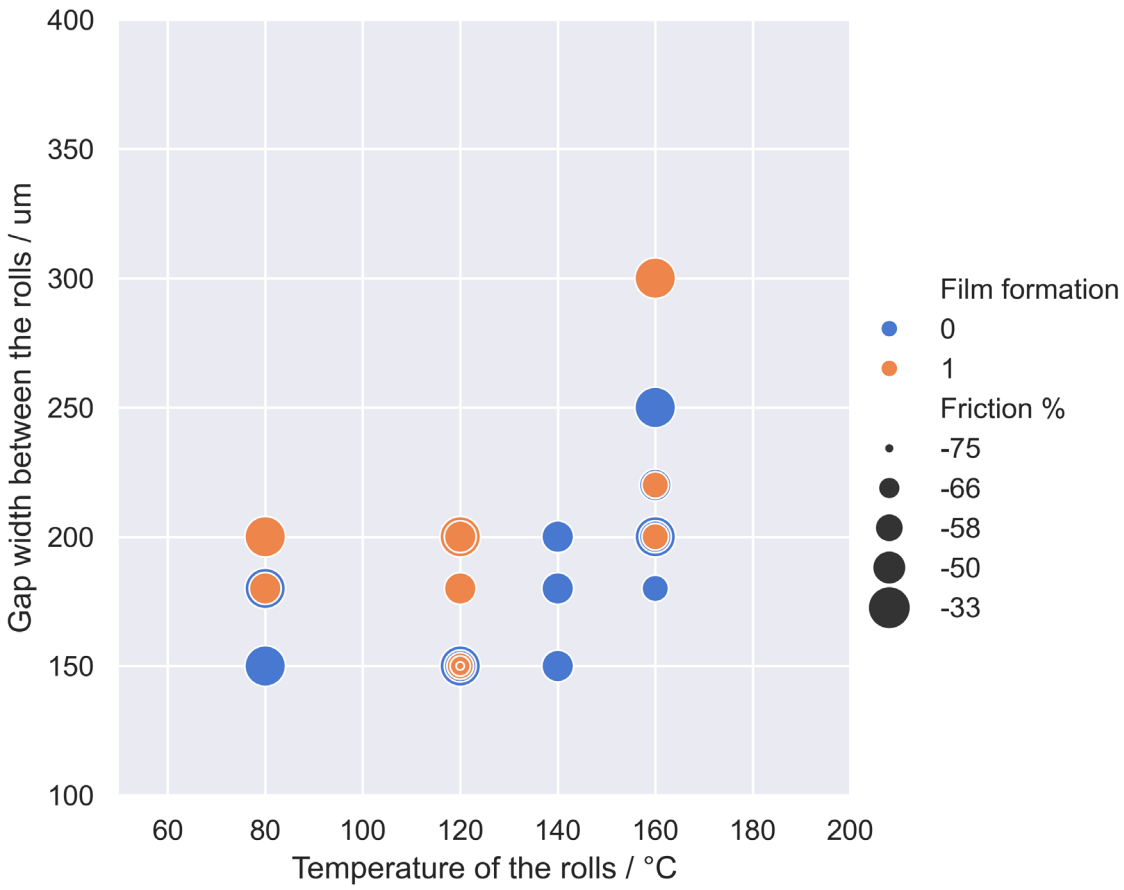

Figure 6 : Example of process windows screening for optimizing the calendaring process conditions for a fixed formulation.

The first part of the work was to test all the binders described in Table 1 and evaluate their fibrillation potential during the mixing and calendaring trials.

The PTFE binders supplied by Chemours and Daikin, which were specifically designed for dry processing applications, exhibited a clear fibrillation behaviour after mixing, evidenced by the formation of micron scale fibre networks (Figure 7a), and this fibrillation was further enhanced during the calendaring

steps. The incorporation of biopolymers such as xanthan gum to form hybrid binder systems did not hinder the fibrillation capability of these PTFE grades, indicating good compatibility and preservation of the properties responsible for the fibres formation.

In contrast, the PTFE powders sourced from Sigma Aldrich, for which no specific suitability for dry electrode processing was indicated, did not show any signs of fibrillation, either after mixing or following calendaring experiments (Figure 7b). A similar absence of fibrillation was observed for cellulose-based binders, where no fibrillation was observed neither after mixing, nor after the calendaring process.

These differences can be attributed to the intrinsic properties of the PTFE material, such as primary particle size, particle morphology, molecular weight, crystallinity, and the degree of resin agglomeration, which strongly influence the ability of PTFE particles to undergo deformation and fibrillate under specific applied shear force. PTFE grades optimized for dry processing typically consist of fine, loosely agglomerated particles with high molecular weight, enabling particle elongation and interconnection during shear, whereas conventional PTFE powders tend to fracture or remain particulate under similar processing conditions.

Overall, these results underline the critical importance of binder grade selection for achieving effective fibrillation and, consequently, mechanically robust and homogeneous dry processed electrodes.

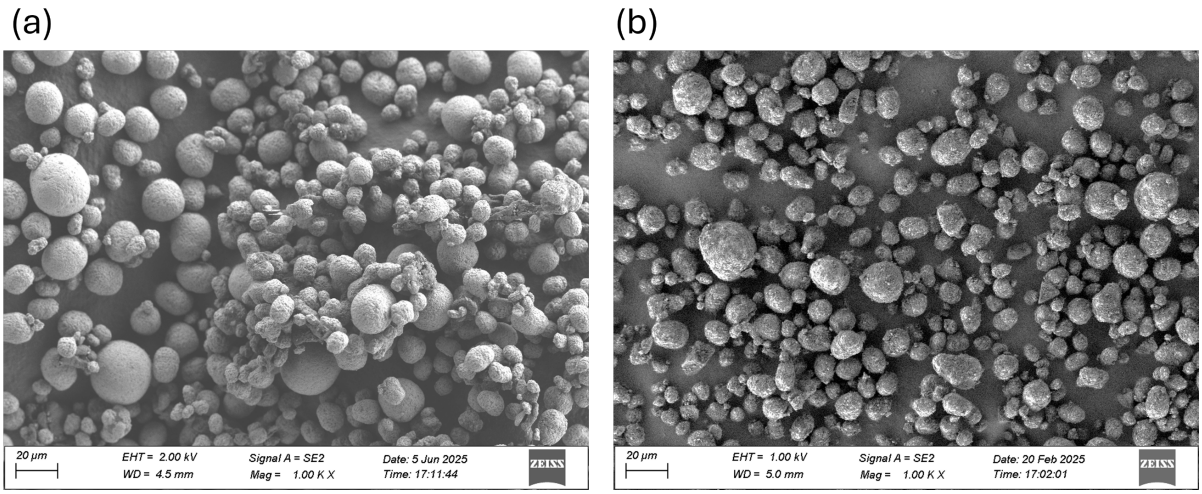

Figure 7: Examples of SEM images of dry-p powders after mixing step (a) using a binder with fibrillation abilities (b) using a binder with no fibrillation potential.

It should also be noted that both the carbon type and its content in the dry formulation strongly influenced the powder properties during and after the mixing step, particularly the extent of PTFE fibrillation. Consequently, the calendaring conditions had to be adjusted accordingly to obtain homogeneous films with the desired quality for each newly investigated formulation.

Several carbon additives were evaluated, including C45, C65, carbon nanotubes (CNTs), and vapor-grown carbon nanofibers (VGCNFs), either individually or in combination, using different ratios relative to the active material and binder. Although multiple carbon types were screened, the C45 grade

was retained for most of the experimental work, based on material availability and the electrochemical cycling performance obtained. In addition, the carbon content was found to directly impact the rheological properties of the mixed powder, thereby influencing the subsequent calendaring behaviour and processing window. Beyond its primary role of enhancing the electronic conductivity of the electrode and thus its electrochemical performance, the carbon content also played a critical role in determining processability.

Taking all these factors into account, and with the objective of maximizing the active material fraction in the electrode, several formulations were tested, as summarized in Table 3. Based on this systematic screening, a carbon content of 3 wt% was identified as the optimal compromise (using our processing tools) and was therefore selected for the final optimized formulation.

The mixing process parameters were also optimized, with particular emphasis on the fibrillation stage, which was identified as the most critical step. Key parameters investigated included mixing speed, mixing duration, the relative rotation direction of the mixing head versus the pan (powder container), and the type of mixing head employed. Figure 8 presents an example of mixing optimization for a fixed formulation, in which the mixing time, and therefore the maximum temperature reached by the powder mixture, was progressively increased while all other process parameters were kept constant. SEM analysis of the corresponding electrodes revealed the onset of well-developed fibrillar networks after 20 min of mixing (Figure 9, left), whereas excessive mixing times (40 min) led to over-fibrillation, characterized by the formation of large PTFE/carbon agglomerates (Figure 9, right).

For batches prepared using PTFE binders supplied by Chemours, an optimal maximum temperature of approximately 45 °C reached during the fibrillation step was identified. This temperature provided sufficient fibrillation while avoiding binder agglomeration and was achieved after 10 min of mixing for grade TE5447A and after 20 min for grade TE5448A.

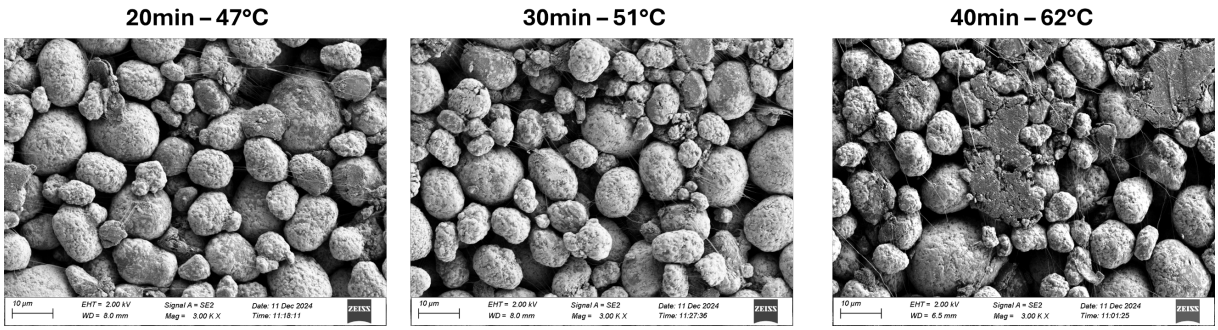

Figure 8: SEM images (top-view) of dry-p NMC622 cathodes vs mixing time and max reached temperature during the fibrillation step (97 wt% NMC622, 2 wt% PTFE TE5448A, 1 wt% carbon (C45)).

Meanwhile, different aluminium (Al) current collectors were evaluated, including a bare Al foil and two grades of carbon-coated Al current collectors (one supplied by Xiamen and one by Armor). The bare Al current collector exhibited very poor adhesion when the free-standing electrode film was laminated onto its surface. As a result, it was not possible to punch electrodes for electrochemical testing, as the active

material readily peeled off during handling. In contrast, both carbon-coated Al current collectors demonstrated significantly improved adhesion properties. Peel tests indicated comparable adhesion performance for the two carbon-coated grades (Figure 9a). To further identify the most suitable current collector for the developed dry-processed electrodes, electrochemical testing was conducted on electrodes fabricated using the different carbon-coated current collectors under identical cell assembly and testing conditions. The obtained results (Figure 9b) showed that the carbon-coated Al current collector from Xiamen provided the most favourable electrochemical performance and was therefore selected as the preferred current collector for preparing the dry-processed electrodes of this project.

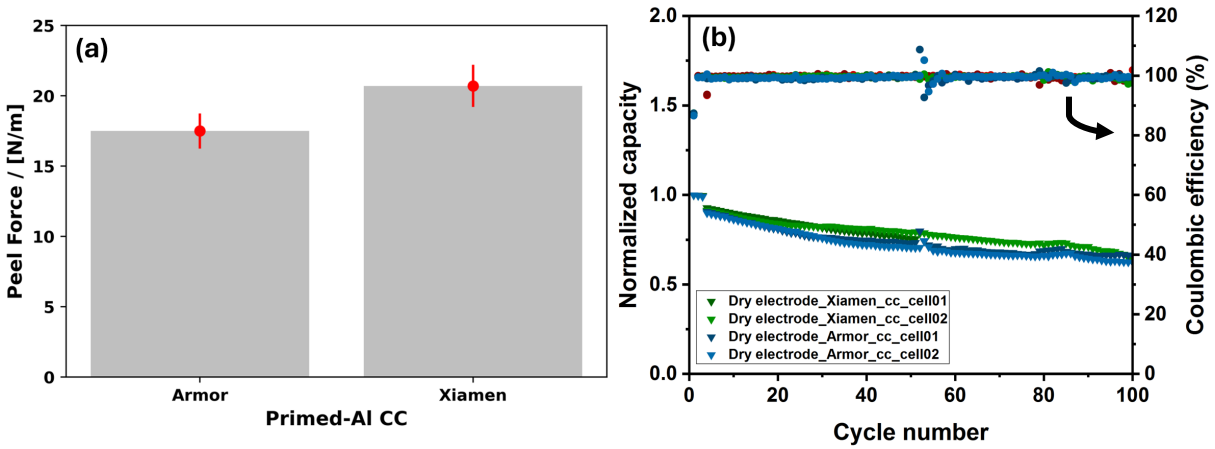

Figure 9: Peel test (a) and electrochemical cycling performance (b) of dry processed electrodes using Xiamen and Armor carbon coated Al current collectors.

Dry processed electrodes versus 200 µm Li

In parallel with the optimization of processing parameters and materials composition, preliminary electrochemical testing was carried out on the prepared dry-processed electrodes using coin cells assembled versus thick lithium metal anodes (200 µm). These electrochemical evaluations constituted the final screening step to identify the most suitable formulation and processing conditions, ultimately enabling the selection of optimized dry-processed electrodes.

Figure 10 illustrates the cycling performance of NMC622 dry-processed electrodes fabricated using the optimized formulation and processing conditions defined in WP1. The electrodes were evaluated using both carbonate-based and ether-based electrolytes. Owing to an improved wettability and better interfacial compatibility with the lithium metal anode, the ether-based electrolyte (Figure 10b) resulted in superior electrochemical performance, particularly in terms of capacity retention, despite the limited oxidative stability of this electrolyte system at high voltages (≥ 4V). The observed performance trends were reproducible across three independent cells for each electrolyte.

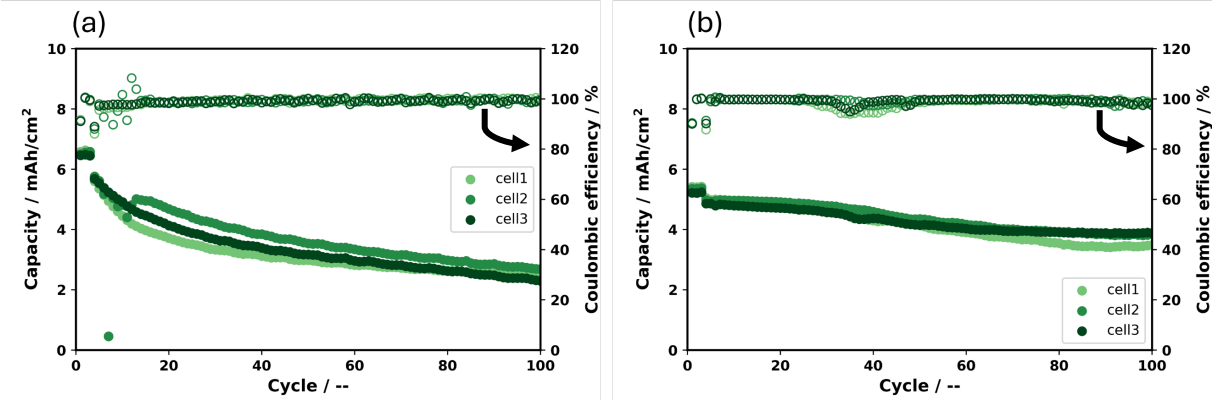

Figure 10: Galvanostatic cycling performance of dry-processed NMC622 coin cells (47A formulation, 1.5 wt.% C45, 3 wt.% binder) versus Li metal (200 µm) in (a) carbonate-based and (b) ether-based liquid electrolytes. Cells were cycled using 3 formation cycles at C/10, followed by 100 charge–discharge cycles at C/2 within a voltage window of 3-4.3 V.

Benchmark versus wet-based

Figure 11 summarizes the galvanostatic cycling performance of dry-processed electrodes manufactured using the optimized formulation reported in Table 3. Their performance was benchmarked against reference slurry-based electrodes produced in-house using the same active material and conductive additive, i.e., 95 wt.% NMC622 and 3 wt.% C45, respectively. For the slurry-based electrodes, the binder was adapted to a solvent-based one (2 wt.% PVDF) and N-methyl-2-pyrrolidone (NMP) was used as the processing solvent.

It should be noted that the two electrode sets were evaluated using different liquid electrolyte systems: the dry-processed electrodes were tested in an ether-based electrolyte, whereas the slurry-based ones were tested in a carbonate-based electrolyte. This difference in electrolyte chemistry accounts for the shorter cycling lifetime observed for the cells with slurry-based electrodes under the same testing conditions. Despite this limitation, the results indicate that, when combined with a compatible electrolyte, the electrochemical performance of the dry-processed electrodes can be comparable to that of slurry-based electrodes, including at high areal loadings. In this work, the loading of the electrodes that were tested was in the range of 6 mAh cm^{-2}.

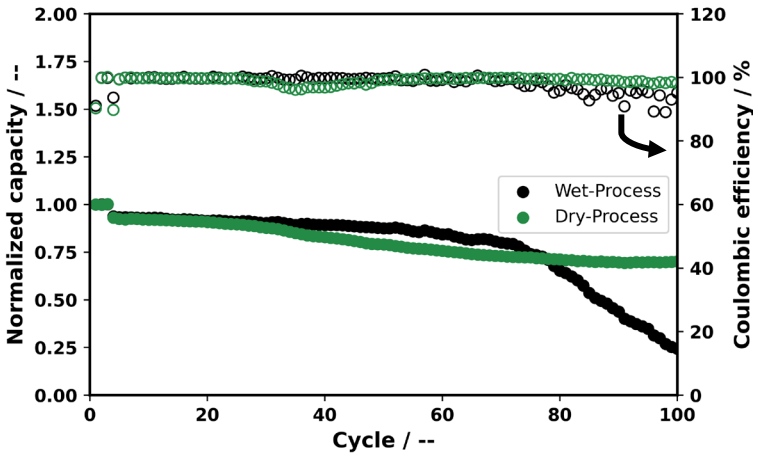

Figure 11: Galvanostatic cycling performance of coin cells using dry-processed NMC622 electrodes (**Green curves**; 47A formulation with 1.5 wt.% C45 and 3 wt.% binder) tested with an ether-based liquid electrolyte, compared with slurry-processed NMC622 electrodes (**Black curves**; 2 wt.% PVDF and 3 wt.% C45) evaluated in a carbonate-based liquid electrolyte, both versus 200 µm Li metal. Cells were cycled using 3 formation cycles at C/10, followed by 100 charge–discharge cycles at C/2 within a voltage window of 3-4.3 V.

2.2.2. MS1: Identification of a DBE process enabling cathodes with areal loading > 3 mAhcm^{-2}

MS1 aimed to identify a dry battery electrode (DBE) processing route enabling the fabrication of cathodes with an areal capacity exceeding 3 mAhcm^{-2}. This milestone was fully achieved, as a robust DBE process was successfully established, consistently delivering cathodes with areal loadings above the targeted threshold.

2.2.3. WP2: Electrodes testing with liquid electrolyte

WP2 covers the experimental activities related to the preparation and electrochemical evaluation of lithium-metal anodes and is organized into three main tasks: T2-1 focuses on the preparation of lithium-metal anodes with protective coatings; T2-2 addresses the assembly and testing of coin cells using electrodes with different areal loadings cycled against PVD deposited lithium metal; and T2-3 involves the post-mortem analysis of cycled electrodes and the correlation of these results with the electrochemical performance, although this task was shifted to WP4 to allow the analysis of more representative data from larger cells (pouch-cells). The work conducted across these tasks (T2-1 and T2-2) is described in the following sections.

T2-1: Preparation of Li metal anode with protective coatings

Within this task, we have used our expertise at CSEM to fabricate and optimize thin lithium metal electrodes with a targeted thickness of approximately 25 µm. The lithium electrodes were produced via a thermal evaporation process, in which metallic lithium granules were placed in a crucible and heated to

approximately 480 °C under high vacuum (10^{-6} mbar). Under these conditions, lithium evaporates and is deposited onto a rotating copper foil substrate until the targeted thickness is achieved. Scanning electron microscopy (SEM) images presented in Figure 12a reveal a dense and homogeneous morphology of the deposited lithium layer. In addition, energy-dispersive X-ray spectroscopy (EDX) analysis (Figure 12c) confirms the high purity of the evaporated lithium, with no detectable contamination traces.

To further validate the deposited lithium thickness, electrochemical lithium plating was performed on copper electrodes using a liquid electrolyte at a current density of 1 mA cm^{-2} (Figure 12b). The full plating of the lithium reservoir delivered a capacity of approximately 5.33 mAhcm^{-2}, which corresponds to an equivalent lithium thickness of about 25.8 µm. This result confirms that the targeted lithium thickness (~25 µm) was achieved, with only minor variation.

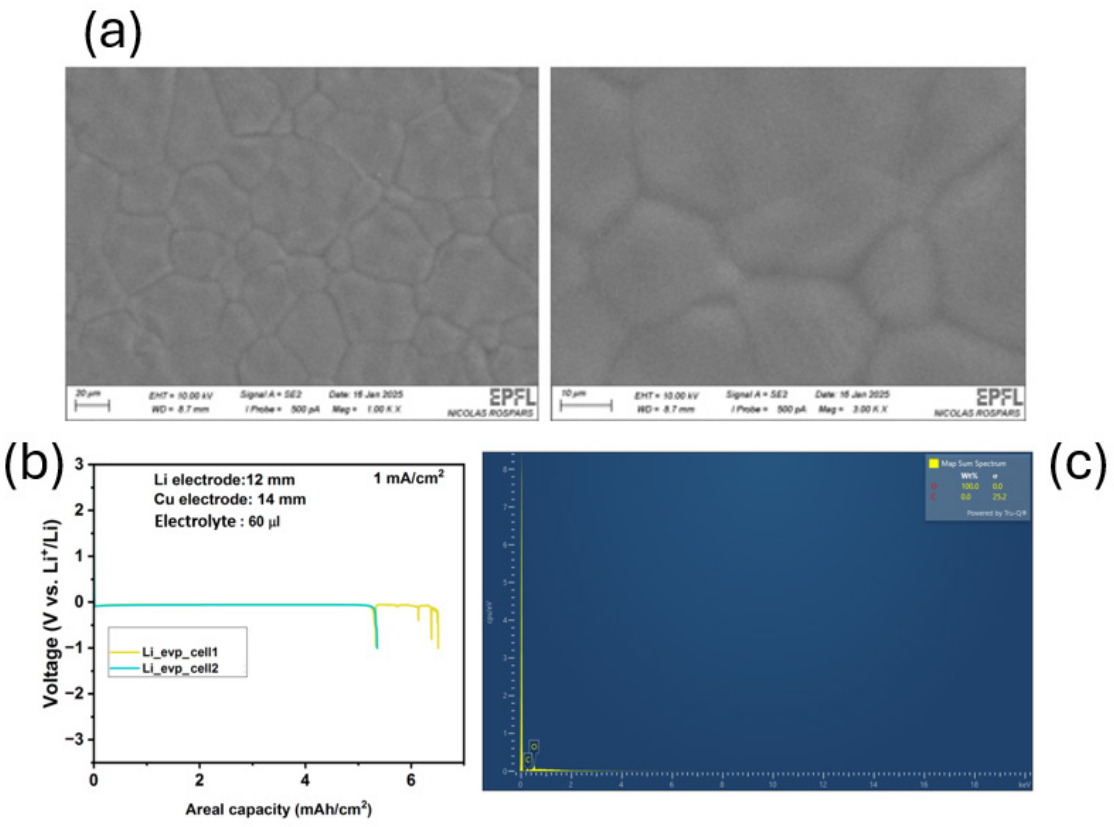

Figure 12: (a) SEM images of the fabricated thin Li metal anode; (b) electrochemical plating of the full lithium reservoir of the fabricated Li metal anode on Cu electrode; (c) EDX results highlighting the chemical composition of the evaporated Lithium metal confirming the absence of any contamination elements.

In addition, different coatings have been investigated to improve the Li metal stability and prolonged its cycle life. Among several coatings, composite coatings containing a lithiophilic metal and a Li-halide showed promising results when investigated using a low loading and low voltage cathode (LFP with a loading of 2 mAhcm^{-2}). Figure 13c shows an example of coated Li metal anodes cycled in full cells versus LFP cathode, confirming the improved cycling performance which can be enabled by this type

of coatings. Moreover, SEM images (Figure 13a) illustrate the morphology of the coated anode, for which the bare lithium morphology was preserved, but the chemical composition on the surface was modified. EDX results (Figure 13b) confirm the chemical modification of the lithium interface while the homogenous distribution of both elements (element 1, and element 2) constituting the composite coating is ensured.

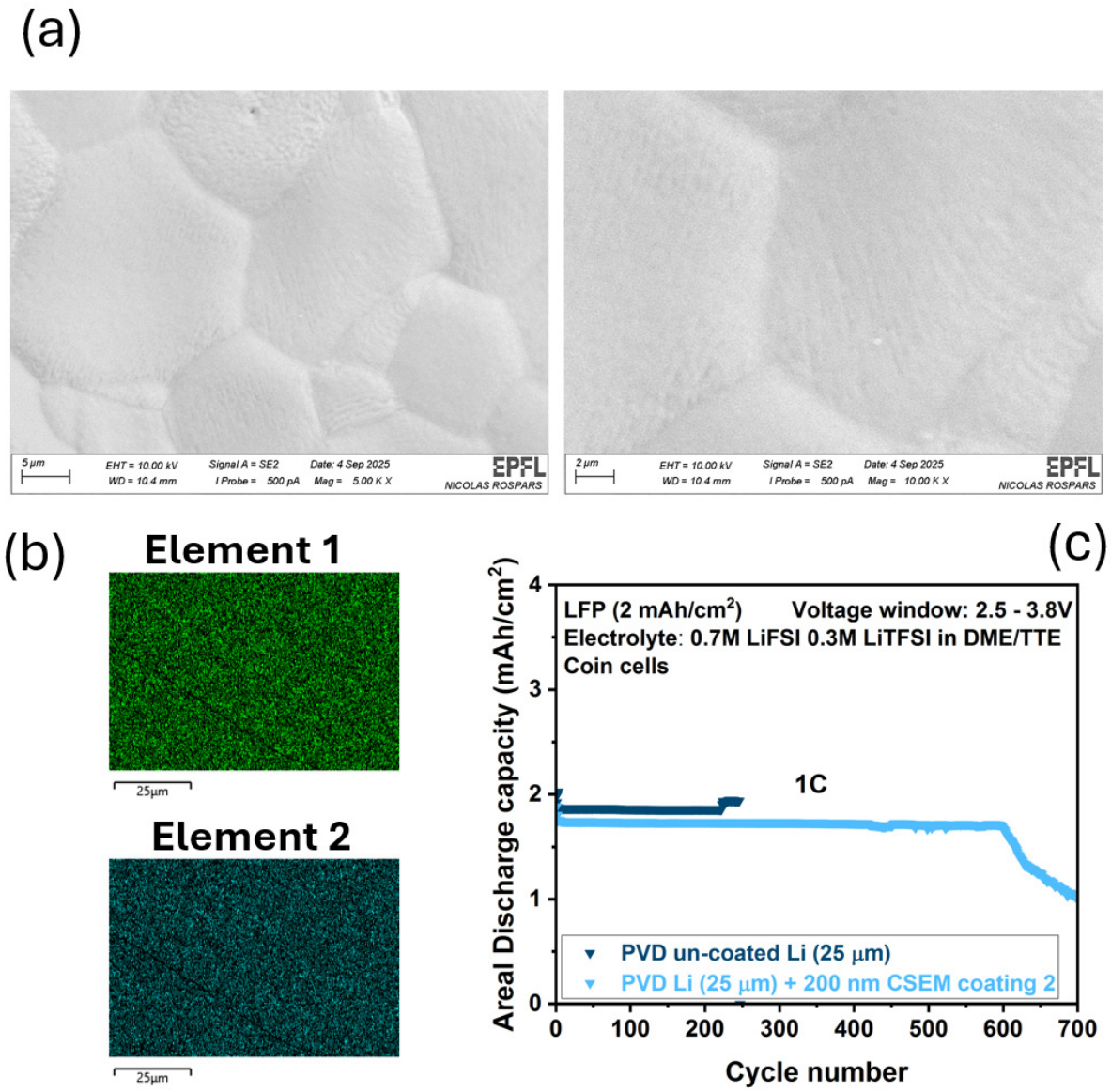

Figure 13: (a) SEM images of coated Li; (b) EDX mapping of the coated Li showing a homogenous distribution of the coating elements (element1 and element 2) of the composite coating; (c) cycling performance of the coated Li compared to reference uncoated one using liquid ether-based electrolyte.

T2-2: coin cells assembly and testing: electrode materials with different loading versus PVD Li

Once the parameters controlling the preparation of the dry-processed electrodes had been fully optimized, the best-performing electrodes were evaluated in full cells (coin-cell format) using thin lithium metal anodes (25 µm) fabricated within Task T2.1.

Figures 14 and 15 present the galvanostatic cycling performance of NMC622 cathodes prepared using the optimized formulations and processing conditions described in WP1 in combination with both carbonate-based and ether-based electrolytes. The reported results were achieved using dry-processed NMC622 cathodes, employing either a PTFE binder or a hybrid binder system combining PTFE with a biopolymer (Xanthan gum). The use of the hybrid binder offers two main advantages: it enabled the fabrication of slightly thinner cathodes (with areal capacities of approximately 5 mAh cm^{-2} compared to around 6 mAh cm^{-2} when only PTFE is used as binder) and promotes improved electrolyte wettability at the cathode surface. It should be noted that the limited loading (cathode thickness) that was achieved in this project, is partially related to limitations of the calendaring tool used in this project.

Moreover, an electrolyte additive (Additive D2) supplied by Daikin was incorporated into the carbonate-based liquid electrolyte with the aim of improving the wettability of the dry-processed electrodes and, consequently, enhancing their electrochemical performance. However, galvanostatic cycling results obtained from coin cells employing PVD lithium-metal anodes (Figure 14b) revealed inferior cycling performance, characterized by a pronounced capacity drop when transitioning from the C/10 formation cycles to long-term cycling at faster C-rate (C/2). This significant capacity loss was primarily attributed to insufficient electrolyte wettability of the dry-processed electrodes, which was inferior to that achieved with the initial electrolyte formulation. The latter had enabled improved cycling stability with a markedly lower capacity drop upon increasing the C-rate (Figure 14a). It should be noted that Daikin's recommendation for Additive D2 was based on improved cyclability observed in a different carbonate-based electrolyte formulation, highlighting the strong formulation-dependent effect of this additive.

When the same electrolyte containing Additive D2 was tested with dry-processed electrodes incorporating a hybrid binder system (PTFE + Xanthan gum) (Figure 14c and d), a clear improvement in cycling performance was observed, confirming the positive impact of the biopolymer on the electrochemical performance. Among the two hybrid-binder formulations, the electrode with a lower PTFE content (Figure 14d) exhibited a higher reversible capacity when cycled at a C/2 rate. This enhanced performance can be attributed to the reduced PTFE fraction in the dry formulation, while maintaining a constant total binder content of 1.5 wt.%, combined with the presence of hydrogen-bonding interactions introduced by the xanthan gum. The functional groups (such as –COOH and –OH) associated with the biopolymer are expected to promote stronger particle–particle and particle–current collector adhesion, reduce interfacial resistance, and improve electrolyte wettability. Together, these effects effectively mitigate the intrinsic limitations of conventional (only) PTFE binder systems in dry-processed electrodes.

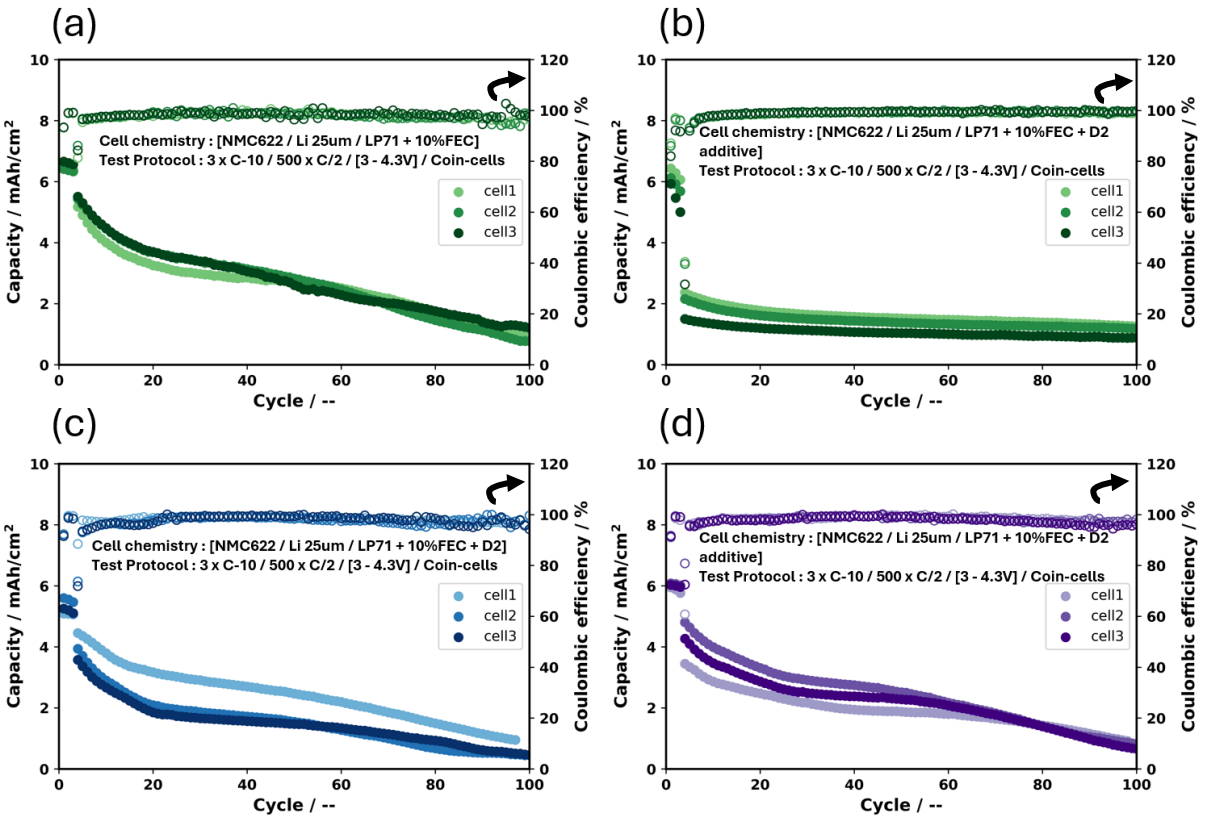

Figure 14: Galvanostatic cycling performance of coin cells assembled with dry-processed NMC622 cathodes paired with 25 µm lithium-metal anodes, using a carbonate-based liquid electrolyte. The investigated cathode formulations include: (a, b) dry-processed NMC622 with 1.5 wt.% PTFE (47A grade) and 3 wt.% C45; (c) dry-processed NMC622 with 1.5 wt.% PTFE (47A grade), 0.5 wt.% Xanthan gum, and 3 wt.% C45; and (d) dry-processed NMC622 with 1.0 wt.% PTFE (47A grade), 0.5 wt.% xanthan gum, and 3 wt.% C45. Cells were cycled using 3 formation cycles at C/10, followed by 100 charge–discharge cycles at C/2 within a voltage window of 3-4.3 V.

In addition to the electrochemical testing performed using carbonate-based electrolytes (Figure 14), further electrochemical evaluations were conducted using ether-based electrolytes (Figure 15). Owing to their superior wettability and improved interfacial compatibility with thin lithium-metal anodes, the ether-based electrolyte, particularly when combined with dry-processed cathodes employing the hybrid binder system (Figure 15b), delivered the most promising electrochemical performance when considering the initial 40 cycles. However, beyond the 40th cycle, a progressive degradation in performance was observed. This degradation was attributed to the well-known corrosion phenomena of the stainless-steel coin-cell casing induced by the ether-based electrolyte formulation (oxidation at high voltage), specifically related to the presence of the LiFSI salt.

In contrast, cells assembled using formulations containing PTFE as a single binder exhibited inferior electrochemical performance, primarily characterized by a pronounced areal capacity loss of approximately 3-4 mAh cm^{-2} relative to the discharge capacity achieved during the formation cycles, which was around 5 mAh cm^{-2}. Upon the introduction of the Xanthan gum as a secondary binder additive, the capacity loss was significantly reduced to approximately 1 mAh cm^{-2}. These results clearly confirm the

beneficial effect of incorporating a biopolymer binder into the PTFE-based system, leading to enhanced capacity retention and overall cycling stability.

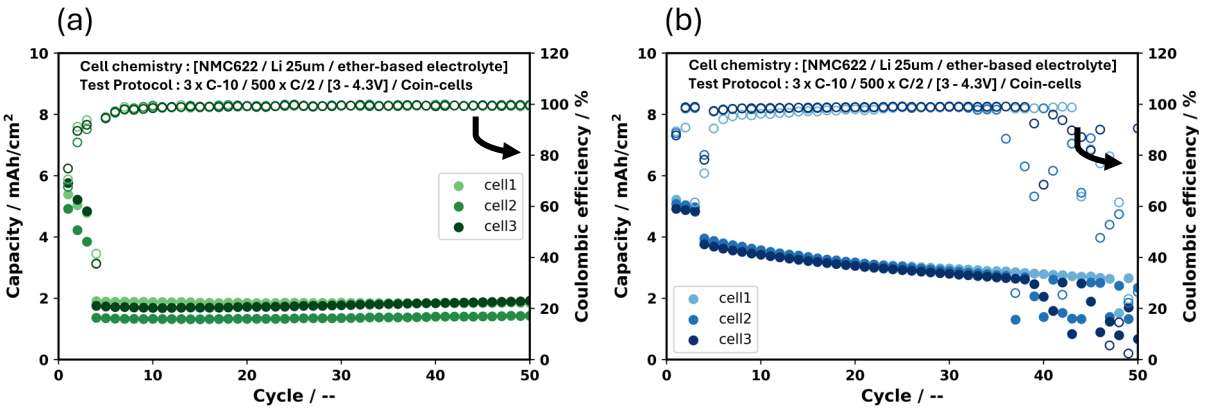

Figure 15: Galvanostatic cycling performance of coin cells assembled with dry-processed NMC622 cathodes paired with 25 µm lithium-metal anodes, using an ether-based liquid electrolyte. The investigated cathode formulations include: (a) dry-processed NMC622 with 1.5 wt.% PTFE (47A grade) and 3 wt.% C45; (b) dry-processed NMC622 with 1.5 wt.% PTFE (47A grade), 0.5 wt.% xanthan gum, and 3 wt.% C45. Cells were cycled using 3 formation cycles at C/10, followed by 100 charge–discharge cycles at C/2 within a voltage window of 3-4.3 V.

When coated thin lithium metal anodes were tested against dry-processed cathodes, unexpected cyclability issues were encountered. Specifically, the assembled cells exhibited an uncontrolled charge behaviour during the initial formation cycle, resulting in an indefinite charging profile that reaches the predefined internal safety limits established for this type of cell (coin cells). Consequently, all tested cells were automatically terminated during the first charge, preventing any further cycling. At this stage, the root causes of this behaviour have not yet been fully identified, and a detailed investigation is still required. This analysis will be carried out as post-DECIDER project work, with the objective of enabling the successful integration of dry-processed, high-areal-loading cathodes with the coated thin lithium-metal anodes developed at CSEM.

2.2.4. MS2: Coin cell demonstration with Li metal anode and DBE cathodes

MS2 aimed to demonstrate coin cells based on a lithium metal anode and dry battery electrode (DBE) cathodes achieving more than 300 cycles with 80% capacity retention at a 0.5C charge and discharge rate. This milestone was partially achieved, as stable cycling was demonstrated for up to 100 cycles instead of the targeted 300 cycles.

The limited cycle life observed was attributed to the following technical factors:

- Electrode balancing constraints: The cathodes were prepared with a high areal loading (>5 $mAh \cdot cm^{-2}$), while the lithium metal anode was limited to a thin foil thickness of 25 µm, corresponding to approximately 5 $mAh \cdot cm^{-2}$. As a result, the cells operated with an N/P ratio below 1, which imposed a strong limitation on cycling stability and lithium inventory retention.
- Electrolyte compatibility issues: The liquid electrolyte used showed limited compatibility with the PTFE binder employed in the dry-processed cathodes, as well as with the lithium metal anode, contributing to interfacial degradation during cycling.

- Lithium metal coating-electrolyte interactions: The compatibility of the prepared lithium metal anode coatings with the selected liquid electrolyte system was not optimal, further impacting long-term electrochemical stability. In addition, the high cathode loading limited the impact of the used thin (200 nm) coatings.

Despite these limitations, the results demonstrated the technical feasibility of combining lithium metal anodes with high-loading DBE cathodes and liquid electrolyte under realistic current densities, while clearly identifying the key bottlenecks to be addressed to reach the targeted cycling performance.

2.2.5. WP3: Electrodes testing with CSEM gel polymer electrolyte

WP3 includes the experimental activities dedicated to evaluating electrode–electrolyte compatibility using CSEM's polymer electrolyte and is structured into three main tasks: T3-1 focuses on the preparation and optimization of the polymer electrolyte formulation for the targeted electrodes; T3-2 involves the preparation of lithium-metal anodes equipped with protective coatings (developed in WP2); and T3-3 covers the assembly and testing of coin cells using electrodes with different areal loadings cycled against PVD lithium. The work conducted across these tasks, mainly within T3-1 and T3-3, is reported in the following sections.

The high areal loading of dry-processed cathodes presents specific challenge for electrolyte integration, particularly in terms of wettability, interfacial contact, and ionic transport. To address these limitations, our solid electrolyte development strategy focused on gel-polymer electrolytes (GPEs), and more specifically on systems formed through in-situ polymerization. This approach has gained increasing attention in recent years for next-generation lithium-metal batteries due to its ability to combine the advantages of liquid electrolytes, such as high ionic conductivity and facile processing, with the mechanical stability and improved safety profile of solid-like polymer networks.

in-situ-polymerized GPEs offer several advantages over traditional solid polymer electrolytes. They are introduced into the cell in a liquid precursor form, which ensures effective wetting and deep infiltration into the porous structure of the dry-processed electrode. This liquid-phase processing promotes intimate interfacial contact both at the cathode-electrolyte interface and within the porous network of the active material, thereby facilitating efficient lithium-ion transport. Following cell assembly, a thermal-curing step initiates polymerization, converting the precursor into a quasi-solid electrolyte with enhanced mechanical integrity and stability. This hybrid liquid-to-solid transition makes GPEs highly compatible with dry-processed electrodes while maintaining scalability and industrial relevance.

In the other hand, GPEs can significantly improve cycling stability of lithium metal batteries, suppress lithium dendrite growth, and reduce interfacial resistance, making them strong candidates for integration with high-loading cathodes. Considering all these attractive properties, two different GPE formulations were selected and evaluated within the DECIDER project to assess their interfacial behaviour, ionic accessibility, and overall electrochemical performance when coupled with dry-processed high-loading NMC622 cathodes.

The first GPE candidate (GPE1) consisted of a vinylene carbonate (VC) monomer mixed with lithium

bis(trifluoromethanesulfonyl)imide (LiTFSI) salt and 2,2′-azobis(2-methylpropionitrile) (AIBN), which is used as a thermal polymerization initiator. To assess the cycling performance of this in-situ-polymerized electrolyte, reference coin cells were assembled using commercial slurry-based NMC622 cathodes with areal capacities of 2 mAh.cm^{-2} and 3.5 mAh cm^{-2}. In addition, the compatibility of the same GPE with our high-loading dry-processed cathodes (≈ 6 mAh cm^{-2}) was evaluated. Each cell was filled with 60 µL of the precursor solution. Following cell assembly and sealing, polymerization was initiated by thermal curing at 60 °C for 18 hours, effectively solidifying the VC-based matrix and ensuring intimate interfacial contact across all cathode loadings. The resulting electrochemical performance is shown in Figure 16a and Figure 17a. The reference cells with lower-loading cathodes demonstrated good cycling behaviour, indicating acceptable compatibility between the developed GPE, the thin lithium-metal anode, and the slurry-based cathodes. In contrast, when the high-loading dry-processed cathodes were tested, the cells exhibited significantly reduced cycling performance, particularly after the formation cycles during higher C-rate cycling. This limited cyclability can be attributed to the combination of high electrode loading and the inherently poor wettability of PTFE-rich dry-processed electrodes toward carbonate-based solvents such as VC.

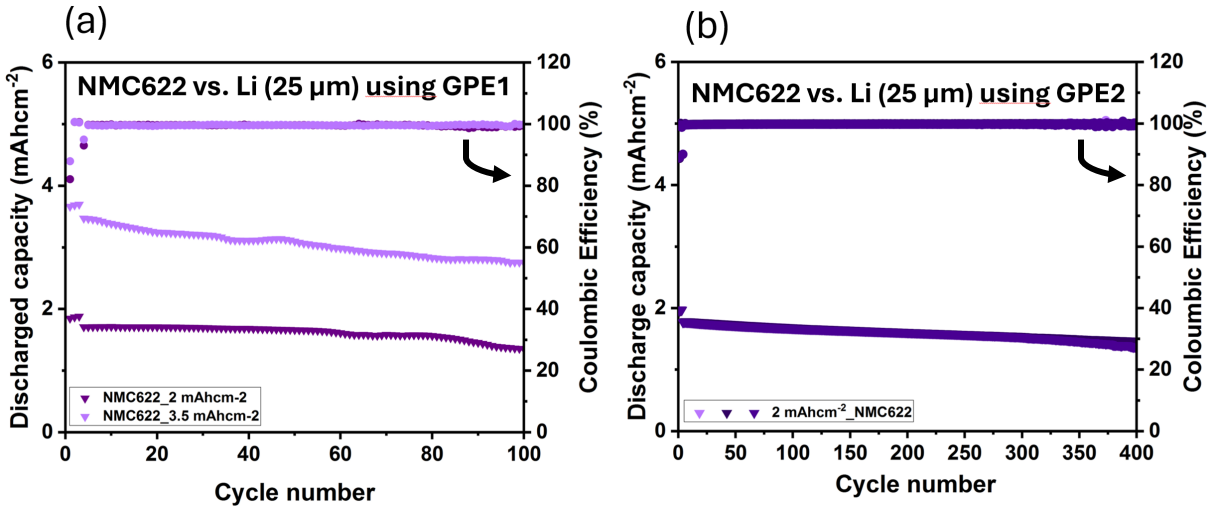

Figure 16: Reference cycling performance of the developed GPE versus slurry-based low loading cathodes. a) results obtained using GPE1; b) cycling performance obtained using GPE2. The cycling protocol includes formation (3 cycles of (C/20)) and long-term cycling (C/4) to 4.3V for charging, C/2 to 3.0 V for discharging).

Moreover, a second GPE candidate (GPE2) was developed. The formulation of this electrolyte is not disclosed, as it is being considered for future patent protection..

The cycling performance of GPE2 was first evaluated using an NMC622 slurry-based cathode with an areal capacity of 2 mAh cm^{-2}. Under relatively moderate cycling conditions (C/4 charge and C/2 discharge), the cell demonstrated stable long-term performance with a capacity retention of approximately

80% after 400 cycles (Figure 16b). These conditions were chosen to be less demanding than those used for GPE1 (Figure 16a), enabling a clearer assessment of the stability of the newly developed polymer system. When tested with high-loading dry-processed electrodes (Figures 17b and 17c), GPE2 exhibited significantly improved performance compared to GPE1. Under the same cycling conditions applied for GPE1, the GPE2 containing cells delivered higher reversible capacities, reaching approximately 1.8 mAh cm^{-2} for the first 40 cycles before capacity decay occurred. At very slow cycling rates (C/10), the reversible capacity closely matched the electrode areal loading (around 6 mAh cm^{-2}), indicating that the polymer electrolyte provides sufficient ionic transport when rate limitations are minimized.

The progressive capacity decay observed during continued cycling can be attributed to the continuous consumption of lithium, a process that becomes increasingly significant at higher electrode loadings (high amount of Li being plated/dissolved). Moreover, the limited cyclability at faster C-rates (Figure 17b) is likely linked to the restricted wettability of the thick, PTFE-rich dry-processed cathodes, which hinders uniform electrolyte infiltration and ion transport within the cathode microstructure.

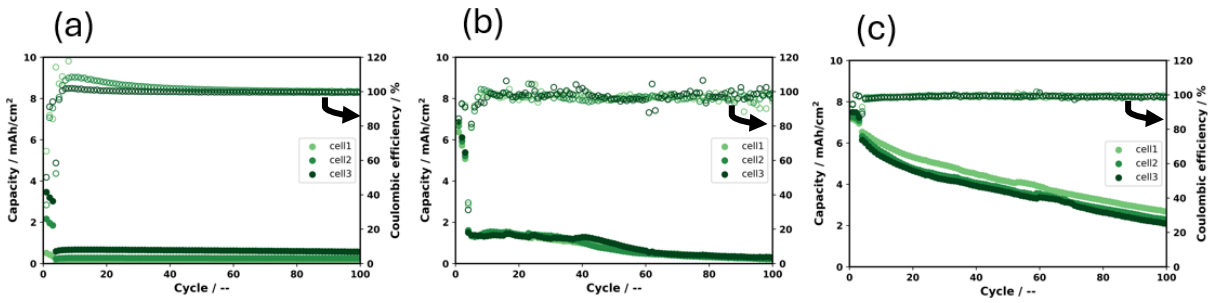

Figure 17: Galvanostatic cycling performance of coin-cells using dry-processed cathodes (NMC622 / 47A 1.5% C45 3%wt). a) using GPE1; b) using GPE2; c) using GPE2 and very low C-rate. (Cell chemistry: [NMC622 / Li 25um / Gel Polymer Electrolyte]) Test Protocol (a, b): 3 x C-10 / 100 x C/2-D/2/ [3 - 4.3V]). Test Protocol (c): 3 x C-50 / 100 x C/15-D/10/ [3 - 4.3V]).

Based on the obtained results, and before implementing these tests in pouch cells format, further optimization of the GPE formulation is still required to ensure full compatibility with high-loading dry-processed electrodes and lithium metal anode, particularly to enhance cycling performance at higher C-rates and at room temperature. In parallel, the use of a hybrid binder system represents a promising strategy to improve electrode-electrolyte interactions. Reducing the PTFE content, while maintaining or even improving adhesion between the dry-processed cathode and the current collector, could significantly increase electrode wettability and mitigate the rate-limiting ionic transport effects. This approach will therefore be considered in future work to further improve the overall cyclability of dry-processed high-loading cathodes in combination with advanced GPE formulations.

2.2.6. MS3: Coin cell demonstration with Li metal anode and DBE cathodes using a polymer electrolyte

MS3 aimed to demonstrate coin cells based on a lithium metal anode and dry processed cathodes achieving more than 100 cycles with 80% capacity retention at a 0.5C charge and discharge rate while employing a polymer electrolyte system. This milestone was partially achieved.

Cycling up to 100 cycles was demonstrated; however, the performance was only obtained either with limited discharge capacity or with insufficient capacity retention when lower current rates were applied (0.066C during charge and 0.1C). Operation at the targeted 0.5C rate did not meet the targeted performance when both GPE1 and GPE2 are used.

The limited electrochemical performance can be primarily attributed to the low compatibility between the prepared polymer electrolyte and the dry-processed cathodes. In particular, poor electrolyte wettability is expected due to the high cathode areal loading and the use of PTFE as the binder, which hindered effective interfacial contact and ionic transport within the electrode. These compatibility issues resulted in increased polarization and accelerated performance fade during cycling.

2.2.7. WP4: Scale-up to pouch-cells

WP4 addresses the scale-up of dry-processed electrode concepts to pouch-cell level and is structured into several tasks: T4-1 focuses on upscaling the production of cathode sheets; T4-2 covers the assembly and testing of pouch cells using liquid electrolytes; and the originally planned T4-3 activity, which was intended to evaluate pouch cells with polymer electrolytes, was not conducted due to the limited cycling performance observed in coin-cell testing (WP3) and the need for further electrolyte development. Instead, a new task was introduced within WP4, shifted from WP2 to benefit from the more representative pouch-cell format, corresponding to T4-3: post-mortem analysis of cycled electrodes and correlation of the results with cell performance. The work performed across these tasks is presented in the following sections.

T4-1: Upscaling the cathode sheets production

Following the optimization work and the extensive testing performed in coin-cell format, a scale-up phase was carried out focusing on the optimized formulations using either PTFE-only or the hybrid PTFE–Xanthan gum binder system. Due to the requirements of the calendaring process, specifically, passing the free-standing film three times under identical friction conditions while progressively reducing the roll gap, the maximum achievable electrode sheet length was approximately 20 cm (Figure 18). Each sheet enabled the cutting of around four cathodes suitable for pouch-cell assembly. It is worth noting that a few defective spots could have been identified on the surface of the prepared sheets, which can

be attributed to localized carbon-PTFE agglomerates.

These prepared electrode-sheets enabled the preparation of a sufficient number of cathodes for the pouch-cell assembly, which allowed the evaluation of the electrochemical performance of the optimized dry-processed electrodes under scaled-up and more practical, cell-level cycling conditions.

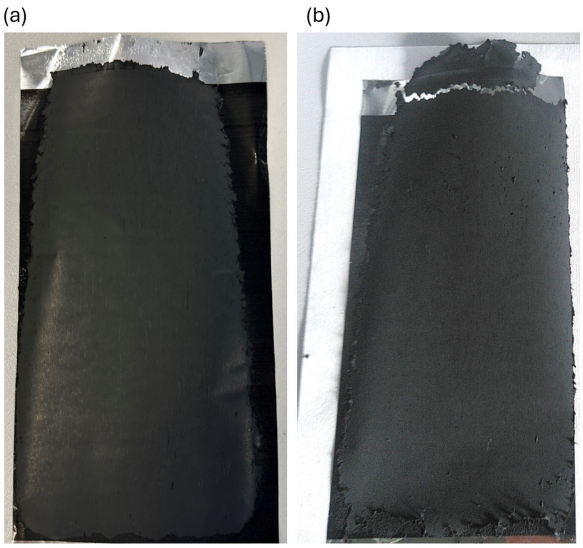

Figure 18: a) Picture of a dry-processed NMC622 cathode sheet prepared with 1.5 wt% PTFE (47A grade) and 3 wt% C45. The areal loading of this cathode sheet was around 6.2-7 mAhcm^{-2}. b) Picture of a dry-processed NMC622 cathode sheet prepared with 1.5 wt% PTFE (47A grade), 0.5 wt% Xanthan gum, and 3 wt% C45. The areal loading of this cathode sheet was around 5.2 mAhcm^{-2}

T4-2: Pouch cells assembly and testing: Liquid electrolyte

The dry-processed cathodes were then evaluated in a ≈ 25 cm^2 pouch-cell configuration using a 25 µm evaporated lithium-metal anode (5 mAh cm^{-2}) and a liquid electrolyte (Figure 19). First, we have investigated the influence of applying external pressure on the cycling behaviour and overall cell cycling stability. Figure 19a shows that both cells, with and without applied external pressure, exhibited stable cycling performance over more than 100 cycles. However, applying external pressure led to a noticeable improvement in reversible capacity, with an increase of approximately 1 mAh cm^{-2} compared to the pressure-free configuration. In both cases, the dry-processed electrodes successfully endured cycling at high current densities (7 mA cm^{-2} discharge), demonstrating their robustness under harsh

operating conditions.

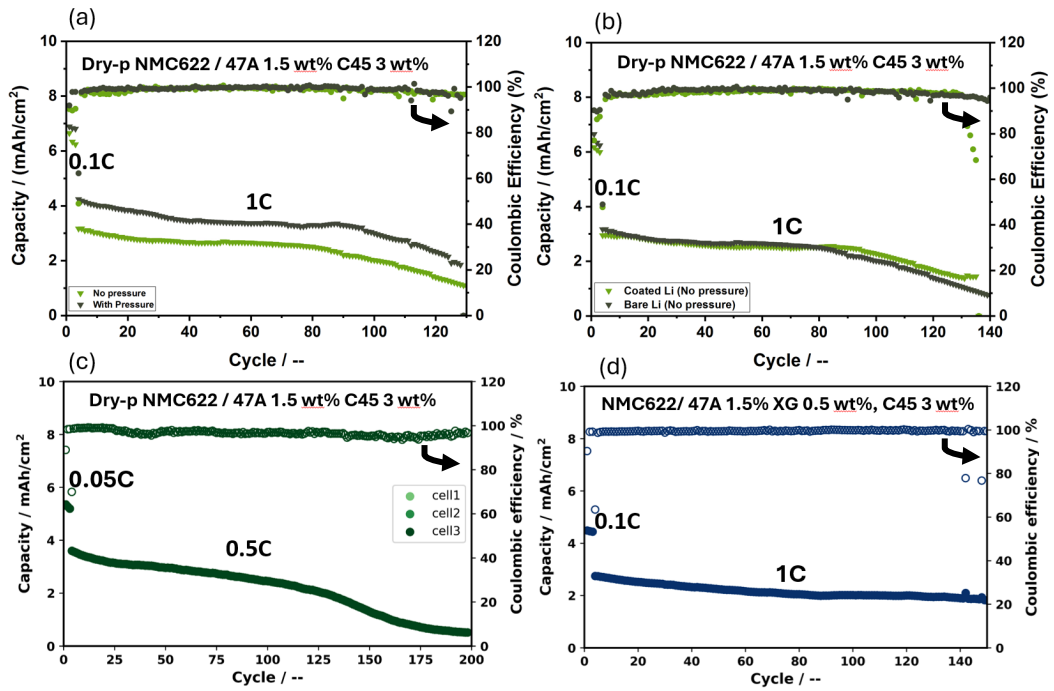

Figure 19: Cycling performance of dry-processed NMC622 cathodes paired with thin lithium-metal anodes in pouch-cell format (25.08 cm²). a) Comparison of the cycling performance of the same dry-processed electrode in pouch cells operated with and without externally applied pressure (0.5 C charge and 1 C discharge). b) Cycling behaviour of dry-processed electrodes paired with coated and uncoated thin lithium-metal anodes under zero applied pressure (0.5 C charge and 1 C discharge). c) Cycling performance under slower cycling conditions (0.2 C charge, 0.5 C discharge) following three formation cycles at 0.05 C. d) Cycling performance of dry-processed cathodes prepared with the hybrid binder system (0.5 C charge and 1 C discharge).

When slower cycling conditions were applied, using formation cycles at 0.05 C followed by long-term cycling at 0.2 C during charge and 0.5 C during discharge (3.5 mA cm^{-2}), and without applying any external pressure (Figure 19c), an improvement in cycling performance was observed. Under these milder conditions, the reversible discharge capacity reached approximately 3.8 mAh cm^{-2}, compared to about 3 mAh cm^{-2} obtained at 1C discharge rate (7 mA cm^{-2}) (Figure 19a). This enhancement of the performance at reduced C-rates indicates that the performance of the high-loading dry-processed cathodes is strongly limited by a rate-dependent ionic transport constraints (can be a result of the limited electrode wettability). These limitations become particularly pronounced at higher current densities, where insufficient electrolyte infiltration and restricted ion mobility across the electrode thickness hinder efficient capacity utilization.

Furthermore, the impact of our optimized coating on the cycling stability of the lithium-metal anode was evaluated. As shown in Figure 19b, the cycling performance obtained with the coated lithium anode was

comparable to that of the uncoated (bare) one when tested against high-loading (6.2 - 7 mAh cm^{-2}) dry-processed cathodes. These results indicate that the current coating formulation still requires optimization in terms of both coating thickness and the composition ratio of the composite layer to ensure effective compatibility with high-loading cathodes delivering large reversible capacities. Moreover, the impact of applying external pressure to evaluate the impact of the coating under the coin-cell like conditions is still required. In parallel, the electrolyte composition must also be refined, as one of the coating components is soluble in the liquid electrolyte. Its dissolution and resulting concentration changes can strongly influence the cycling behaviour and performance of the lithium metal anode and may partially diminish the intended benefits of the coating.

Finally, dry-processed electrodes formulated with the hybrid binder system containing PTFE and xanthan gum were also evaluated in pouch-cell configuration. The cells assembled with this cathode and a thin 25 µm lithium-metal anode exhibited very good cycling stability, sustaining up to 150 cycles at a fast 1C discharge rate. However, the reversible capacity, consistent with previous observations, remained limited to less than 3 mAh cm^{-2} when no external pressure was applied. In addition to the rate-dependent ionic-transport constraints discussed earlier, the reduced adhesion strength of the dry-processed cathode to the current collector, compared with that of slurry-based electrodes, may further contribute to the observed limitation in reversible capacity.

To gain deeper insight into the origins of these performance losses and to identify the mechanisms constraining the integration of high-loading dry-processed cathodes with lithium-metal anodes in pouch cells, a post-mortem analysis of the cycled cells was performed.

T4-3: Post-mortem analysis of cycled electrodes and correlation of the results with performance of the cells

After long-term cycling of the pouch cells assembled using dry-processed cathodes prepared with the hybrid binder formulation, one of the cells was transferred into an argon-filled glovebox (O_2 < 0.1 ppm, H_2O < 0.1 ppm) for post-mortem analysis. The pouch was opened, and the Li/separator/cathode stack was recovered, as shown in Figure 20a. Examination of the anode revealed that most of the 25 µm lithium reservoir had been consumed, leaving behind brittle black residues. These residues correspond to passivated, electrochemically inactive “dead lithium,” which is a key factor limiting the achievable cycle life when high-capacity cathodes are paired with thin lithium-metal anodes.

On the cathode side (right-hand photograph in Figure 20a), the active material layer detached easily from the carbon-coated aluminium current collector and adhered strongly to the separator. This observation highlights the poor adhesion of the dry-processed free-standing film to the current collector compared to the slurry-based cathodes, which likely contributes significantly to the limited reversible capacity and reduced cyclability observed in the pouch-cell tests. Further structural insights were obtained from SEM imaging of the recovered cathodes (Figure 20b), which revealed well-developed PTFE fibrillation

within the electrode matrix. However, cracked NMC622 particles were also identified. Such particle cracking may result from the harsh calendaring conditions required to achieve the targeted electrode thickness, as well as from mechanical and chemical degradation of the NMC cathode materials during prolonged cycling under demanding operating conditions.

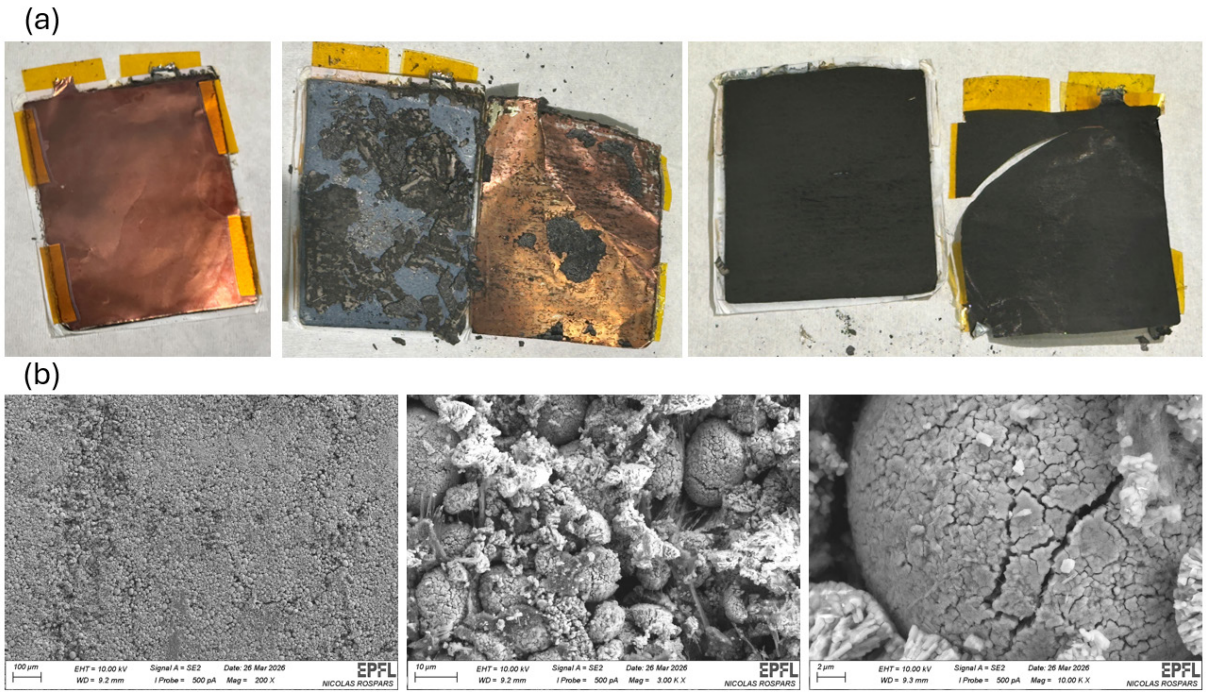

Figure 20: a) Photos of pouch cells electrodes (cathode and anode) and separator after pouch cell cycling; b) SEM images of cycled dry processed electrode at different magnifications.

3 Conclusions and outlook

The DECIDER project successfully demonstrated the feasibility and strong potential of manufacturing high energy density lithium metal batteries using dry electrode processing, without the use of solvents. A complete dry manufacturing route for cathodes was developed from scratch as an alternative to conventional slurry-based coating methods.

Through extensive optimization of the full process (formulation and processing), reproducible dry-processed cathodes with areal capacities exceeding 5 mAh cm^{-2} were achieved. These cathodes were fabricated using a PTFE-based binder system, as well as innovative hybrid formulations combining PTFE with biodegradable biopolymers, significantly reducing PFAS content while delivering improved electrochemical performance and enhanced sustainability.

In parallel, thin lithium metal anodes were reliably produced in-house via thermal evaporation, allowing precise control of the lithium metal anode thickness and surface purity. The compatibility of dry-processed electrodes with lithium metal anodes was validated using both liquid (carbonate and ether based) and gel polymer electrolytes at both coin-cell and monolayer pouch-cell levels, demonstrating the scalability of the proposed approach. The electrochemical testing confirmed a promising cycling behaviour, including the successful operation of solid-state coin cells using our in-house developed gel polymer electrolyte for more than 100 cycles. These cells can achieve a specific energy density of up to 490 $Wh.kg^{-1}$, nearly doubling that of conventional lithium-ion technologies. Furthermore, pouch-cell prototypes were successfully developed using liquid electrolytes, establishing a critical intermediate step toward fully dry processed solid state pouch cells.

Table 6: the best cells chemistries prepared within the project and the corresponding projected energy density.

Cell chemistry	Calculated energy density (Wh/kg)	Number of cycles
DBE NMC622/Li/GPE	485	100 (50% capacity retention)
DBE NMC622/Li/LE	400	100 (80% capacity retention)

The table above (Table 6) summarizes the two best cell chemistries developed within the DECIDER project, together with their projected energy density and the maximum number of cycles achieved so far. The combination of thin Li metal anodes with high-loading, dry-processed NMC622 cathodes enabled particularly high potential energy densities, corresponding to an increase of approximately +30% when using a liquid electrolyte and up to +60% when using a gel polymer electrolyte, compared to state-of-the-art Li-ion cell chemistries such as NMC622/Si–graphite. Despite the relatively low cycle life demonstrated at this stage (around 100 cycles), these results clearly highlight the strong potential of the developed cell concepts, which is suitable for applications where weight and compactness are critical,

such as drones, robotics, and medical devices. Further optimization of interfacial stability, electrolyte formulation, and electrode processing is expected to significantly improve cycling performance in the next stages of development.

Table 7 resumes the achievement of the targeted milestones of DECIDER.

Table 7: list of the project milestones and their actual state at the end of the project

Milestone	Milestone description	Success criteria	Status	Comments
MS1	Identification of a DBE process leading to cathodes with areal loading > 3 mAh·cm-2	Demonstrate a dry processed cathode with areal loading above 3 mAh·cm-2	**Achieved**	A DBE process was successfully developed, enabling cathode areal loadings exceeding 3 mAh·cm^{-2} and satisfying the milestone objectives.
MS2	Demonstration of coin cells with Li metal anode and DBE delivering >300 cycles at 80% capacity retention and 0.5C	≥ 300 cycles, ≥80% capacity retention, 0.5C charge/discharge	**Partially achieved**	Coin cells based on Li metal anodes and dry processed cathodes were demonstrated. While stable cycling was obtained, the full target of ≥300 cycles at 80% capacity retention was not reached due to some compatibility issues with the electrolyte and thin lithium metal anode. Only 100 cycles have been demonstrated.
MS3	Demonstration of coin cells with Li metal anode and DBE delivering >100 cycles at 80% capacity retention and 0.5C	≥ 100 cycles, ≥80% capacity retention, 0.5C charge/discharge	**Partially achieved**	Coin cells up to 100 cycles have been demonstrated, but the reversible capacity and capacity retention was limited when cycling at 0.5C current rate. When the current rate was decreased to 0.1C for discharge, the reversible capacity has improved, and a capacity retention of about 45-50% has been achieved

DECIDER laid the groundwork for future optimization steps to improve the cycling performance of dry processed cathode using either liquid or solid electrolyte, then scale up the resulting cell components to build high-capacity pouch cells, targeting capacities of up to 1 Ah while maintaining the high cycling performance demonstrated at smaller scales.

Overall, the outcomes of the DECIDER project provided experimental evidence that dry battery electrode technology represents a viable, scalable, and environmentally attractive pathway for next-generation lithium metal and solid-state batteries. By combining solvent-free processing, reduced environmental footprint, high energy density, and manufacturing cost advantages also the project established

a solid technological foundation for future scale-up and optimization, and encourages the initiation of new collaborations with industrial companies in the field including: tool manufacturers such as BUSS or Coperion K-Tron, and active material and binder providers such as Vianode, Chemours, and Daikin. An Innosuisse between Medtronic, BUSS and CSEM targeting the use of dry processed electrodes is in preparation. The advances in dry processing cathodes and all solid-state lithium metal batteries directly support the acceleration of sustainable, high-performance battery solutions and open the door to new applications such as drones, electric aircraft, and other high energy demand mobility systems.

4 National and international cooperation

This project supported CSEM to initiate the activity on dry processing of electrode a develop from scratch a procedure to prepare dry battery electrodes. Meanwhile, new collaboration with binder suppliers such as **Chemours and Daikin** and current collector supplier such as **Armor Battery Films** was established**.** Furthermore, discussions for future collaborations with other partners such as: anode material supplier such **Vianode** (graphite supplier) or mixing equipment suppliers such as **Buss** and **Coperion K-Tron** or final users such as **Medtronic** were initiated.

5 Publications and other communications

- A poster contribution is planned for the Dry coating Forum (Dresden, German), September 2026.
- A scientific paper resuming all the development occurred within this project is also planned to be submitted by the end of 2026.

6 References

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