REVIEW

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Dry Battery Electrode Technology: From Early Concepts to Industrial Applications

Benjamin Schumm, Arthur Dupuy, Milena Lux, Christian Girsule, Susanne Dörfler, Florian Schmidt, Magdalena Fiedler, Maria Rosner, Felix Hippauf, and Stefan Kaskel**

The increasing demand for clean and efficient energy storage makes the environmentally friendly and cost-effective production of lithium-ion batteries a focal point in current battery research and development. Dry battery electrode (DBE) coatings play a crucial role in future production schemes as this technique does not require the use of toxic solvents and energy-intensive drying steps. This review article focuses on the most advanced DBE method today, based on fibrillated polytetrafluoroethylene (PTFE) binder. PTFE-based DBE coatings are suitable for both laboratory scale and mass production, which places them in a prominent position among DBE methods. The article covers the historical development of the process as well as current research in the field of lithium-ion batteries (LIB) and next-generation batteries such as lithium–sulfur batteries (LSB) and solid-state batteries (SSB). Both the suitability and existing drawbacks of PTFE-based dry coatings for these cell types are discussed. The article also provides insights into production research and describes approaches for scaling the method. Characteristic features and differences of the most important methods, the DRYtraec and Maxwell-process, are outlined. Finally, existing challenges in commercializing the technology are discussed, and an outlook on environmentally friendly PTFE-alternative binders is given.

1. Introduction

The demand for efficient, sustainable, and cost-effective battery technologies is rapidly increasing, driven by the growing need for energy storage solutions in sectors ranging from consumer electronics to electric vehicles (EVs) and grid storage. Traditional wet coating processes, which involve the application of a slurry containing active materials, binders, and solvents onto a metallic foil, are widely used in the manufacturing of battery electrodes.[1,2] However, these processes pose significant environmental and economic challenges, including high energy consumption, extensive use of toxic solvents such as N-Methylpyrrolidone (NMP), and substantial production costs.[3,4] Depending on the cell chemistry, design, and production standards, 57–92 kg CO_2-eq are emitted per kWh of produced battery capacity for today's lithium-ion battery (LIB) cells.[5] In response to these challenges, dry coating technologies have emerged as a promising alternative, offering numerous advantages in the production of battery electrodes.

Dry coating, also known as dry battery electrode (DBE) coating, eliminates the need for solvents by directly applying a dry mixture of active materials and binders onto the electrode substrate. This approach not only reduces the environmental footprint by eliminating solvent emissions but also simplifies the manufacturing process, potentially lowers production costs, and reduces equipment footprint. According to Degen et al., 6–7% of cell production costs can be reduced using dry coating.[6] Moreover, when 10% of battery production is expected to use dry coating in 2030, 480 000 tons of CO_2 emissions could be saved annually.[7]

As highlighted in recent publications, especially for thick electrodes, DBE processing can achieve similar or superior electrochemical performance compared to slurry-coated electrodes (cf. **Table 2**); while, offering substantial reductions in energy consumption and production costs.[6,8–13] The achievements that have been reached with DBE are addressed in several literature review articles. **Table 1** sbriefly illustrates the focus topics and differences of these publications. It becomes obvious that the focus on a clear transfer into industrial applications is missing in the existing literature discussion.

Various methods of dry coating have been explored to optimize the efficiency and performance of battery electrodes. Key techniques include electrostatic dry powder deposition, mechanical

B. Schumm, A. Dupuy, M. Lux, C. Girsule, S. Dörfler, F. Schmidt,
M. Fiedler, M. Rosner, F. Hippauf
Fraunhofer Institute for Material and Beam Technology IWS
Winterbergstraße 28, D-01277 Dresden, Germany
E-mail: benjamin.schumm@iws.fraunhofer.de

S. Kaskel
Chair of Inorganic Chemistry I
TUD Dresden University of Technology
01062 Dresden, Germany
E-mail: stefan.kaskel@tu-dresden.de

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/aenm.202406011

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Table 1. Summary of the published reviews from 2018 to 2024 which are related to dry battery electrode manufacturing.

Review	Content focus topics	Refs.
Dühnen et al., 2020	– Material and technology approaches for greener LIB production – Environmental impacts, including energy consumption and emissions – Critical supply issues of raw materials such as lithium, cobalt, and nickel	[3]
Bryntesen et al., 2021	– Energy consumption in LIB production process steps, particularly drying step – Approaches for optimization of production – Discussion of lab-scale dry coating approaches	[18]
Liu et al., 2021	– LIB manufacturing challenges, focusing on energy consumption – Discussion of potentials for coating, drying, and formation processes – Brief review of lab-scale solvent-free technologies	[1]
Degen et al., 2022	– Comparison of various dry coating methods according to their TRL – Specific analysis of most mature and scalable technologies – Quantification of energy saving potential of dry coating	[7]
Li et al., 2022	– Broad dry coating technology literature review and patent analysis – Comparison of vapor deposition, binder fibrillation, powder spray, and compression – Application fields: LIB and SSB	[8]
Lu et al., 2022	– Advantages of dry coating for SSB – Brief low-TRL technology and binder material review – Effects of dry coating on electrochemical properties and microstructure in SSB	[9]
Wang et al., 2023	– Brief review of dry coating techniques and used binders – Polytetrafluoroethylene (PTFE) as a key binder for dry coating – Discussion of application examples for PTFE-based dry coating on lab-scale	[10]
Bouguern et al., 2024	– General description of dry electrode coating advantages – Comparison of powder spraying, binder fibrillation, and extrusion – Lab-scale results and scale-up concepts discussion	[12]
Zhang et al., 2024	– Review of dry coating techniques: powder deposition, extrusion, 3D Printing, compression, polymer fibrillation; lab-scale examples – PTFE binder application examples and challenges; alternative binders – Material challenges in PTFE-based dry coating	[11]
Liu et al., 2025	– DBE for solid state batteries, advantages and synergies – Evaluation of different manufacturing principles – Approaches for cathode anodes and SEL, binders	[19]
This work	– Fundamentals, historical development, and advancements of PTFE-based DBE – Review of recent lab-scale achievements LIB, Li–S, SSB, and SIB – Review and comparison high-TRL approaches for DBE technology scale-up – Challenges for high-volume and widespread market penetration – Expected future research and development directions	—

dry powder coating, and vacuum deposition methods.[8,14] Among these, the calendar-based dry film coating using fibrillated binders has shown the most promise due to its ability to produce uniform coatings with high material utilization rates.[12,15,16] The technologies summarized under the terms "direct calendaring" and "freestanding film" provide the highest technology readiness level (TRL) at the highest overall performance rating.[7] With the slurry-based wet coating as reference for TRL 9 (system works in an operational environment), the calendar-based methods have been validated or demonstrated in a battery-production relevant environment (TRL 5–6). Continuous double-side coating has been demonstrated.[13] In contrast, most competing dry coating technologies have not yet passed the laboratory environment (single side coating or small samples); and thus, are not considered for commercialization within this decade.

As a key enabler for calendar-based DBE technologies, the used binder material polytetrafluoroethylene (PTFE) plays a major role in electrode properties and process characteristics. This unique polymer serves as an effective binder with characteristic features, such as high chemical and mechanical stability, as well as excellent thermal resistance, which make it an ideal candidate for DBE processes.[10] One particularly noteworthy characteristic of PTFE is its ability to form fibrils under the influence of shear forces during the mixing and coating process.[17] Given these advantages, this review will mainly focus on PTFE-based systems, examining their implementation in dry coating technologies and their impact on the performance of battery electrodes.

In the following, a comprehensive overview of dry coating technologies based on PTFE binders will be provided as this system currently holds the highest TRL; and thus, the greatest chances for imminent market penetration.[7] In contrast to previous reviews in the field, this review will focus on scalable approaches with high relevance for the industrial application. Materials and production technologies which only provide academic value will not be reviewed. Instead, we will point out the high TRL of PTFE-based DBE processing and highlight production research, scaling up processes, and the challenges associated with market introduction. Insights into both the technical and economic aspects that must be addressed for widespread adoption will be provided. This review intends to cover a broad spectrum of topics, from the fundamental principles and historical development of dry coating methods to the latest advancements in electrode development

Table 2. Overview on recent DBE studies on parameters of influence for LIB cathodes.

CAM	Evaluation parameter	Key findings	Ref.
NMC	– Microstructure and electrochemical properties – Qualitative description of fibrillation	– Hierarchical PTFE network – Reduced charge-transfer (−14%) and ionic (−57%) resistance compared to slurry cast electrode	[39]
NMC	– Porosity variation by compression	– Optimal porosity in intermediate range (32% porosity) – NMC fracture at high compression	[40]
NMC	– Fibrillation process of different PTFE by kneading – Microstructure, electrochemical and mechanical properties	– 10 mAh cm^{-2} thick electrodes with 80% capacity retention at 0.5 C – Lowest tortuosity for high extrusion-ratio PTFE	[41]
NMC	– Electrolyte additive influence	– PTFE side reaction observed in thick cathodes (6.6 mAh cm^{-2}) – $LiPF_6$ outperforms $LiClO_4$	[42]
NMC	– CNT versus carbon black (CB) as conductive additive	– ≈30% improved rate capability of CNT-based electrode	[43]
NCA	– CNT coating of NMC by pre-mixing – Electrochemical evaluation	– 99.6% CAM content in electrode – 821 mAh cm^{-3} volumetric capacity – Superior cycling capacity over 300 cycles compared to slurry electrode with 96% CAM	[44]
NCMA	– PTFE/CB composite – Microstructure and electrochemical properties	– 11% increased density and 17% increased volumetric capacity compared to slurry electrode	[45]
LMO	– CNT versus CNF versus CB additive – Microstructure and electrochemical properties	– Electrical conductivity: CNT > CNF > CB – All dry Ni- and Co-free full cell (LTO) – 82.8% capacity retention after 200 cycles	[46]
LCO	– MWCNT versus CB – Microstructure and electrochemical properties	– Co-network from MWCNT and PTFE fibrils improves mechanical stability of electrode during cycling	[47]
LFP	– LFP CNT premix – Electrochemical evaluation	– 90% capacity retention after 90 cycles with 1.5 mA cm^{-2}	[48]
LFP	– 97% LFP dry electrode – Comparison to slurry electrode	– 7.8 mAh cm^{-2} over 300 cycles – 470 Wh L^{-1} (dry) versus 390 Wh L^{-1} (wet)	[49]
LMNO	– Loading and electrolyte optimization	– 4.5 V full cell with 68% capacity retention after 1000 cycles	[50]

for lithium-ion and next-generation batteries, such as lithium-sulfur and all solid-state batteries. By doing so, the authors aim to shed light on the currently most promising avenues for future research and development in the field of dry coating technologies.

2. Historical Background

2.1. PTFE as Outstanding DBE Binder

The ability to form fibrils under the influence of shear forces is a property of PTFE that has been described since the middle of the 20th century.[20]

PTFE is a semi-crystalline polymer and several polymorphs have been identified in characteristic temperature and pressure regions. Above 19 °C, PTFE exhibits a first order phase transition from triclinic to hexagonal crystal structure.[21] The crystal structure of the PTFE phase between 19 °C and 30 °C features helical PTFE chains that are organized in a hexagonal orientation to slide on each other upon deformation and form the polymer fibrils (**Figure** 1a–c).[22] These fibrils create a network-like structure within the electrode, providing exceptional binding properties. This network enables the mechanical integrity and flexibility of the electrode, ensuring long-term stability and performance. The ability to form a fibrillary binder network renders PTFE as an outstanding binder system with superior processability. The effect of fibril formation is promoted not only by temperature or shear but also by the molecular nature of the PTFE itself (Figure 1d–f). High molecular weight PTFE can be achieved using emulsion polymerization.[23] Whereas for molecular weights below 10^5 g mol^{-1}, the polymer chains crystallize in relatively stable rod like structures, molecular weights above 10^6 g mol^{-1} result in the formation of folded ribbons.[24] PTFE fibrils can be easily extracted from a polymer particle with domains organized in a ribbon structure. As a result, emulsion polymerized PTFE with high molecular weight provides a good tendency for fibril formation; and is thus, preferred to be selected as a binder for DBE.

2.2. Historical Application of PTFE in DBE

The exceptional properties of PTFE as a binder have been utilized for decades in the manufacturing of electrodes for electrochemical devices. The evolution of the technology from a method involving PTFE suspensions with a high solvent content, through approaches with reduced solvent content leading to dough-like masses, to completely dry processing, can be traced over the period from around 1960 to around 1980.[25–29] Its ability to form a mechanically robust fibril network under shear forces is a unique feature compared to other polymers.[25,30]

Early reports on the use of PTFE binders for the manufacturing of so-called PTFE-bonded electrodes often mentioned the use of a PTFE dispersion to ensure a homogeneous mixture of the powdered active materials. Niedrach and Alford used PTFE dispersions to deposit platinum black to prepare a catalyst layer for fuel cell applications.[27] They reported the formation of a cream-like slurry which was treated at 350 °C and pressed at 20 MPa after deposition to obtain the final electrode layer. This

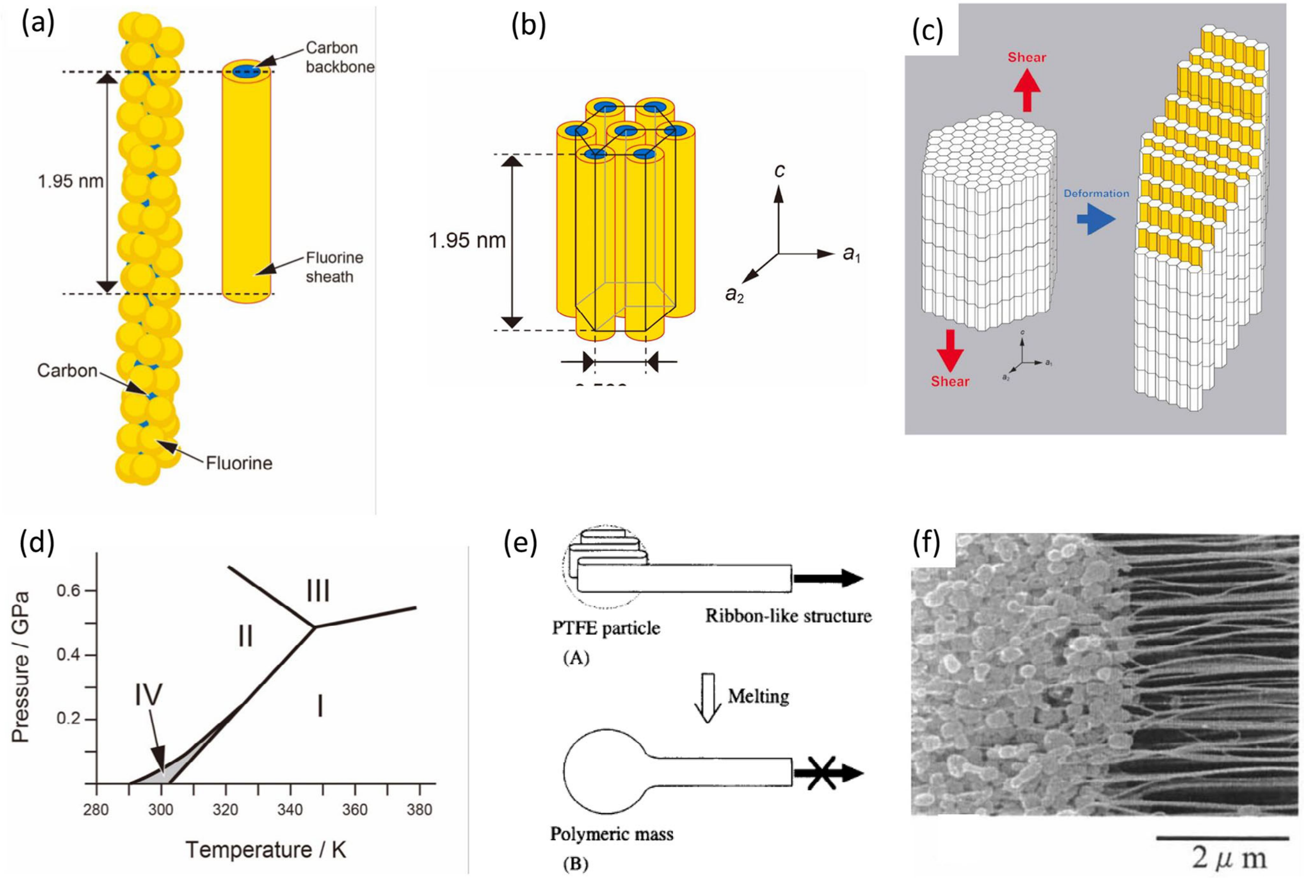

Figure 1. a) The helical structure of an individual PTFE polymer chain in hexagonal phase IV between 19 °C and 30 °C and a cylindrical simplified model of the PTFE chain. Reproduced (Adapted) with permission.[22] Copyright 2000, Wiley; b) hexagonal unit cell structure of the phase IV; [22] c) illustration of the shear-driven deformation of a PTFE crystal with colored slip planes in the PTFE crystal. Reproduced (Adapted) with permission.[22] Copyright 2022, Elsevier; d) phase diagram of PTFE; e) schematic representation of the concept of structure change leading to the development of mechanical strength;[17] and f) SEM image of fibrils formed from PTFE particles in a stretched membrane.[17]

method was further developed by reducing the amount of solvent to produce a dough-like mass which was kneaded until the desired processing consistency was achieved. The mass was then rolled out to the desired thickness and combined with a substrate. A temperature treatment was typically applied to this coating to evaporate the solvent, resulting in the final electrode layer.

In 1981, Bernstein and Coffey (MPD Technology) described the production of a "dry film" electrode using a dry process without liquid processing aids in their patent.[26] They highlighted the avoidance of the energy-intensive drying step as a primary advantage of this method. The processes they described bear a strong resemblance to approaches still discussed today. They detailed the use of mills, extruders, and molding machines, where even small amounts of fibrous binder (between 0.5 wt% and 5 wt%) were effective. For processing into flexible films with structural integrity in any desired thickness, roll-type mills were proposed. Moreover, they addressed the combination with current collectors, such as conductive films, wire screens, or expanded metals. Based on this approach, the manufacturing of various types of electrodes for a multitude of electrochemical devices was described in subsequent years.

For application in fuel cells, Schautz and Kordesch developed the rolling method for the manufacturing of PTFE-bonded carbon electrodes.[31] They used a mixture consisting of carbon black, optionally a catalyst, PTFE, and a pore-forming filler (ammonium carbonate or sugar). The paste was processed into a diffusion layer or active layer by rolling and sintering. The sintering step removed the filler and provided the desired porosity.

The importance of introducing sufficient shear forces during the mixing process for achieving well-agglomerated dry mixtures was described by Hayashida et al.[32] Their procedure for the manufacturing of a hydrogen absorption alloy electrode based on $LaNi_5$ family materials explained the formation of 3D entangled PTFE fibers acting as a network for the mechanical fixation of the active material.

Zhong et al. used the dry film approach to manufacture electrodes for nickel-metal hydride (Ni-MH) batteries.[33] They applied a slurry electrode, powder-based electrode without any binder and a dry film electrode onto a copper mesh current collector and compared the electrochemical performance of the three approaches. The lowest resistance and the highest adhesion were observed for the dry film electrode, resulting in the highest cycle life. With the increasing importance of lithium-ion batteries

(LIB) after their commercialization by Sony,[34] the first reports on dry electrode production for this cell system have also been described.

Megahed et al. evaluated various compositions, materials, and electrolytes for optimizing their cell system. They described advantageous behavior of PTFE-based dry calendared electrodes regarding swelling effects during cycling. Further, the authors suggested the processing of the electrodes by continuous extrusion methods.[35]

The electrochemical behavior of PTFE as a binder in dry processed lithium-ion electrodes was described by Li et al. They used carbon fibers with different PTFE amounts to investigate the binder stability at low potentials.[36] Given its significant impact on film formation and electrode properties, we will explore the aspects of PTFE binders on the anode side in greater detail in Section 3.1.

In all these methods, the active material was processed in a dry and solvent-free manner with PTFE, pressed or rolled into a freestanding (= self-supporting) dry film, and then, shaped into electrodes. Sometimes, the dry film was bonded to a current collector or another substrate by pressing or laminating. As it was a powerful tool for efficient electrode manufacturing in research and development, the dry film method with PTFE binder established itself as a standard procedure on a laboratory scale during this period. The main advantages were: minimal material amounts were required and a wide range of materials could be manually processed without complex dispersion and with simple equipment. Today, the use of manually rolled PTFE-based dry films in the research and development of new materials or battery chemistries is more relevant than ever. In Section 3, we will delve into recent reports on this subject.

Approaches to scale the technology for production have also been published in numerous patents and publications. However, large-scale implementation was primarily achieved by Maxwell Technologies, which industrially used the method for mass production of supercapacitor electrodes and was later also demonstrated for LIB.[37] We will discuss the evolution of this process to a scalable coating method avoiding the manufacturing of a freestanding film by the DRYtraec technology,[13,15,16] as well as further recent developments focusing on industrial dry battery electrode manufacturing in Section 4.

3. Recent Developments and Applications

3.1. DBE for Lithium-Ion Batteries

Climate change and the limited availability of oil and gas are the main drivers behind the current change in industry and society toward an electricity dominated economy based on renewable energy. Lithium-ion batteries play a crucial role in this transition by enabling electromobility and stationary storage at affordable costs. On the road to affordable electric cars, economy of scale demands for substantial increase in battery production to reduce cost without sacrificing ecological standards. This growing demand for energy storage systems for electromobility, stationary storage, power tools, mobile devices, and other applications requires an expansion of production capacities for lithium-ion batteries (LIB). At the same time, production costs must be lowered and the carbon footprint in cell production reduced. DBE technology offers an opportunity to address these challenges and revolutionize the production of LIBs.

3.1.1. LIB Cathode

The demand for replacing the slurry process in LIB production is stronger on the cathode side because NMP is crucial in today's cathode production, whereas the anode side already utilizes aqueous processes. This leads to more studies and publications on effects of DBE processing for LIB cathodes. We follow this tendency and will therefore discuss the main principles of structure–property relationships in dry film based electrodes using the example of LIB cathodes. The feasibility of a PTFE based dry coating process has been shown for numerous different cathode active materials (CAM), achieving excellent rate and cycling performances comparable to conventionally fabricated wet electrodes, for example, NMC, NCA, NCMA, LFP, LMO, LMNO, and LCO.[12,38] Lithium nickel manganese cobalt oxide ($LiNi_xMn_yCo_{1-x-y}O_2$, NMC) was the most commonly used material due to its high energy density. Various studies have fabricated PTFE-based NMC cathodes on a lab scale, emphasizing different aspects, which we will address in the following paragraph.

The electrode properties are not only influenced by the chemical composition of the CAM but also by parameters such as particle size distribution, formulation, and additives as the following examples will explain.

Matthews et al. analyzed the microstructure and electrochemical performance of PTFE-based NMC622 electrodes with a focus on a qualitative description of the fibrillation process (**Figure 2a**).[39] They observed the anchoring of PTFE on NMC surface and the following unwinding of crystalline units in SEM images. For 1% PTFE content, a hierarchical structure of primary and secondary fibrils with diameters ranging from a few micrometers down to tens of nanometers forming a web like structure was described. Electrochemical advantages in terms of lower ionic diffusion resistance, slightly better capacity retention in full cells (after 200 cycles at C/3), and rate performance at 0.1–2 C compared to wet electrodes were observed. It could be seen that, adjusting the interaction of CAM with the PTFE binder had a strong influence on the electrode microstructure; and thus, on the electrode performance. The effects of varying the DBE microstructure were also studied by Tao et al.[40] They aimed to provide correlations between electrode structure and electrochemical kinetics for NMC cathodes. The study varied the compression load, resulting in changes of porosity (here: 22%, 32%, and 39%). The compression; however, also led to a fracture of NMC particles. The sheet resistance showed an optimum for medium porosity electrodes and increases for high porosity due to the high pore volume, as well as for low porosity, which was attributed to some of the fractured NMC particle pieces being poorly electronically connected. Charge transfer resistance was also lowest in the medium porosity electrode: It provided sufficient voids to maintain the charge transfer but also had a structure being dense enough to reduce the lithium ion diffusion pathway. The rate capability was highest at 32% porosity, which led the authors to assume an optimum of porosity in that range. However, the particle fracture being induced by this compression study indicates

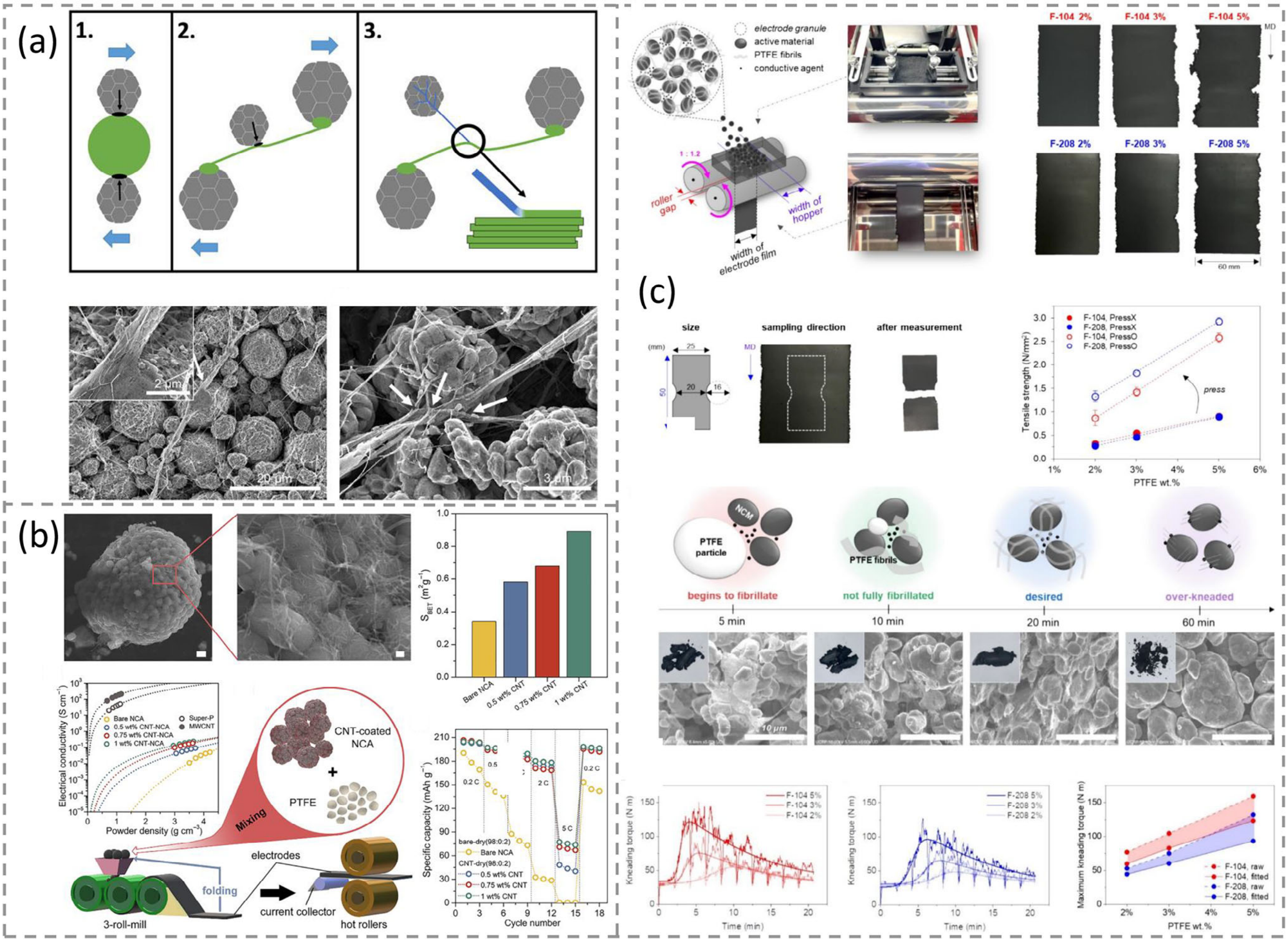

Figure 2. a) Formation of primary and secondary fibrils during compression and shear mixing of NMC and PTFE; Reproduced with permission.[39] Copyright 2024, Frontiers Research Foundation; b) improved rate capability of NCA by CNT coating on the CAM surface; Reproduced with permission.[44] Copyright 2022, Wiley. c) preparation of free-standing NMC films using different PTFE binders with its influence on tensile-strength and evaluation of the impact of fibrillation process of the PTFE binder with NMC, focusing on kneading time, torque, and electrode microstructure. Reproduced with permission.[41] Copyright 2024, Elsevier.

the limits of this approach. Thin DBE coatings with low porosity may not be achievable in good quality according to this finding. Long term cycling will have to be done in order to evaluate potential negative effects of fractured NMC particles at high compression loads. Evaluating the influence of inactive materials on the electrode performance, Oh et al. contributed another important aspect in current literature (Figure 2c).[41] They applied the dry process for two different types of PTFE with similar median particle size and bulk densities in an extrusion-based mixing process. By qualitatively describing the morphological structure of the intermediate products over time as well as electrode properties of the NMC811 cathodes, they demonstrated a slightly easier fibrillation, lower tortuosity and slightly favorable electrochemical performance for the binder with the higher extrusion ratio. For the lowest PTFE content of 2%, a discharge rate capability of 80% was achieved at 0.5 C for electrodes with 10 mAh cm^{-2} loading.

The electrolyte as another crucial inactive component also provides a significant influence on the electrode performance. As PTFE-based electrodes show different interaction with the electrolyte compared to PVDF-based slurry electrodes, a variation of electrolytes was carried out by Tao et al.[51] The study examined the properties of the cathode electrolyte interface (CEI) for $LiPF_6$ and $LiClO_4$ electrolytes, revealing for the first time their effect on PTFE based dry processed electrodes. Despite a thicker CEI for the $LiPF_6$ electrolyte, those cells showcased higher capacities at higher C-rates due to a superior conductivity; while, the performances at lower C-rates were comparable. The study also confirmed that PTFE binder had side reactions in the dry-processed cathode that resulted in LiF side products, requiring further investigations. While this example showed the influence of the electrode's ionic conductivity on the rate capability, the following examples will show the influence of electric conductivity which can be influenced by varying the conductive additive.

Park et al. described a mortar-based dry process comparing electrochemical performance of NMC electrodes with different conductive additives, carbon nanotubes (CNT), and carbon black (CB).[43] Showing a much better capacity retention (75% after 100 cycles at 0.1 C) and better conductivity as well as higher mechanical stability, electrodes containing CNTs were described to be beneficial for the process. However, the relatively high carbon additive (10%) and binder (5%) amount reduced the practical relevance of this study.

The beneficial influence of CNT on the DBE performance was also described for other CAM. Shin et al. utilized NCA, investigating the effect of a pre-mixing step of active material with the CNT as conductive additive to achieve a homogenous CNT-coating layer on the CAM particles (Figure 2b).[44] The cathodes achieved an extremely high (CNT-coated) CAM content of 99.6%. Adding also a premixing step for carbon black and PTFE, a study on NCMA by Choi et al. improved the distribution of inactive materials.[45] Both studies achieved slightly better cathode conductivities and cycling performances over comparable wet coated electrodes. Nickel- and cobalt-free CAM lithium manganese oxide (LMO) were used for DBE by Sadan et al.[46] They tested various conductive additives (multi-walled CNT, carbon black, and carbon nanofibers CNF), also emphasizing that CNT electrodes prove best in terms of electric conductivity, followed by CNF and CB. Consequently a better rate performance at higher C-rates resulted for the CNT-based electrodes; while, all three variants maintained a similar behavior at lower C-Rates. An all-dry Ni- and Co-free full cell (with LTO anode) was built and delivered a stable performance (capacity retention of 82.8% after 200 cycles at 0.7C). The impact of CNTs as a conductive additive was further studied by Wang et al.[21] In their study, dry LCO cathodes containing multi-walled CNTs showed a superior mechanical and electrochemical performance with 91% capacity retention after 100 cycles at C/3 and no evident cracks in contrast to dry LCO cathodes containing CB with only 70% capacity retention under the same conditions.

As a CAM of huge interest due to its beneficial cost structure, lithium iron phosphate ($LiFePO_4$, LFP) is of interest for DBE R&D. Zhang et al. fabricated LFP cathodes with areal capacities of 3.1 mAh cm^{-2} utilizing CNT in order to achieve good conductivities.[48] In subsequent full cell cycling tests with a dry coated hard carbon counter electrode, the dry coated cells showed a better rate performance than slurry-coated electrodes. In addition, dry coated electrodes provided a good mechanical stability at higher thicknesses; while, slurry electrodes developed cracks when the material loading was increased.

Even higher loadings were reported by Kwon et al., showing an LFP electrode with 97% active material content and a high areal capacity of 7.8 mAh cm^{-2}.[49] This electrode outperformed a comparable slurry coated cathode in terms of rate performance by providing homogeneous dispersion of conductive additive and binder, excellent cathode-electrolyte interfacial stability, and electronic resistance. In addition, a more efficient lithium ion diffusion was observed in the PTFE-based dry electrodes, which was attributed to a higher specific surface area of the powder due to spot contact and bridge-like connections between LFP and PTFE; while, PVDF covered the active material of the conventional wet cathode hindering ionic diffusion pathways.

With LMNO as another CAM, Yao et al. demonstrated the possibility to fabricate ultra-high loading cathodes (9.5 mAh cm^{-2}).[50] The authors showed long-term stability in full cells with 68% capacity retention after 1000 cycles at C/3 in (at 3 mAh cm^{-2} vs graphite anode); which was ascribed to the combination of robust mechanical properties, a well-connected PTFE network, and stable cathode electrolyte interphase.

The wide variety of CAM that has been successfully used in DBE, as well as the positive results in terms of mechanical and electrochemical stability, present PTFE-based dry cathodes as a promising alternative over slurry cathodes to achieve sustainable battery production without sacrificing performance. The discussed results show that high-quality cathodes can be produced using DBE. However, the direct comparison to slurry-based electrodes is challenging due to a lack of standards regarding electrode properties (composition, loading, porosity, etc) or methods (configuration of test cells, used components, and test conditions). Accordingly, the achieved values must be critically evaluated, and the proper reference electrodes and methods should be taken into account. Nevertheless, the ability to produce highly loaded electrodes with good rate capability using DBE can objectively be considered as a major advantage at the electrode level compared to slurry-based processes.

3.1.2. LIB Anode

Graphite is the dominant material in LIB anode production due to its natural abundance, affordability, long cycling stability and safety, high specific capacity, and high electrical conductivity.[52] In the field of dry battery electrodes, most studies have been conducted with graphite as anode material with a focus on the electrochemical degradation mechanisms in PTFE binder which will be discussed later in this chapter. However, few reports provide insights into the DBE processing of alternative anode materials. Lithium titanate (LTO) is an alternative negative electrode offering minimal volume change, enhanced Li-ion mobility, no lithium plating, and fast charging but a low electrical conductivity and limited energy density.[53] Zhou et al. produced DBE LTO anodes for a fully dry hybrid "supercabattery" (with LFP cathode).[54] They demonstrated excellent long term cycling stabilities (86.2% capacity retention after 5000 cycles and the possibility to perform at temperatures as low as −40 °C). Promising results for full cells with LTO anodes were also shown by Matthews et al.[39]

Suh et al. reported outstanding electrochemical performances of dry coated graphite anodes in full cells outperforming conventional slurry processed anodes in terms of rate capability and capacity retention (88.2% after 300 cycles at 0.5 C).[55] Compared to wet coated anodes, the dry electrodes show a lower cross-sectional porosity gradient and exhibit lower tortuosity and ionic resistance as well as less Li-plating, indicating their suitability for fast charging applications. The PTFE content was varied between 0.5% and 4% and set to 2% as a compromise among electrode resistance, adhesion strength, and decrease of initial coulombic efficiency due to side reactions between PTFE and lithium ions (**Figure 3c**).

As already discussed for CAM, the particle morphology also influences the anode DBE process. A recent study of Hwang et al. evaluated correlations between the particle shape of graphite and the anode fast charging capability.[56] A slightly better fast charging capability at 5 C is attributed to spherical graphite due to its lower ionic resistances and tortuosity compared to flake-like particles. Morphological differences between dry coated and comparable slurry coated anodes are more pronounced for the spherical graphite.

In contrast to the stable cycling performances of dry anodes reported by Suh et al. and Hwang et al., Liu et al. described

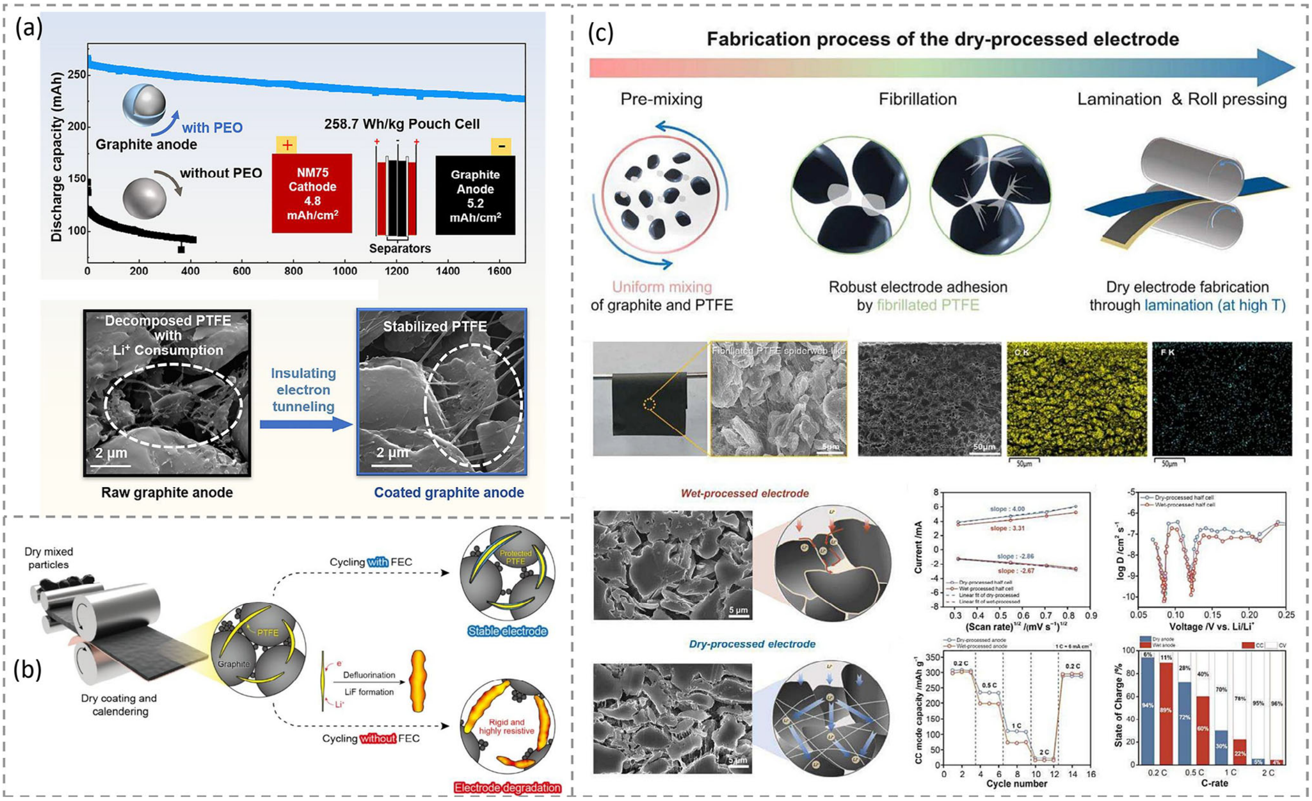

Figure 3. a) Prevention of PTFE degradation in anodes by PEO coating of graphite; Reproduced with permission.[59] Copyright 2024, Elsevier; b) schematic illustration of PTFE degradation in dry-processed anode without and with FEC. Reproduced with permission.[61] Copyright 2024; Elsevier; and c) fabrication process of the dry PTFE-based graphite anode and its electrochemical comparison with wet-processed anode. Reproduced with permission.[55] Copyright 2023, Elsevier.

superior cycling stabilities of slurry coated anodes in half cells especially for thick electrodes.[57] The electrochemical decomposition of PTFE leads to low initial coulombic efficiency and poor electrode integrity but the use of VGCF (vapor grown carbon fibers) as a conductive additive in dry electrodes instead of Super P enhances cycling performance and could be a promising amendment. According to the authors, VGCF with its high aspect ratio and fibrous structure, enhances particle interlocking within the powder composite. Thereby, as evidenced by higher tensile strength measurements, it improves electrode cohesiveness. In addition, the VGCF-based electrode remains crack-free after film formation; and therefore, demonstrates superior cycling stability compared to the Super P-based electrode.

Similar stability issues were shown by Wu et al. comparing the cycling stability of graphite anodes with PTFE binder to carboxylated nitrile (XNBR) binder.[58]

When used in negative electrodes, PTFE reacts with lithium ions, generating lithium fluoride, destroying the PTFE fiber network, and leading to a rapid decline in electrochemical performance of the cell, as well as a degradation of electrode integrity and the formation of cracks due to volume changes of graphite during lithiation and delithiation processes.[10] This phenomenon was first raised in 1996 by Li et al.[36] To overcome this limitation, studies have been assessing the impact of a polymer coating on the graphite particles to mitigate electrochemical degradation of PTFE.[11] Materials that can be used for coating graphite particles should be electrochemically stable against reduction reactions, inhibit electron transfer between graphite and PTFE, and be a conductor for lithium-ions in order to maintain the cell reaction. Wei et al. applied polyethylene oxide (PEO) layers with different thicknesses, finding an optimum at 1.25% PEO content, with improved initial coulombic efficiency and moderate interphase resistance (Figure 3a).[59] The corresponding pouch cells showed a good capacity retention of 87.3% after 1700 cycles, a C/3, and increased initial coulombic efficiency of 84% instead of 47% for uncoated graphite. Lee at al. showed similarly good results for a PVDF coating, achieving an initial coulombic efficiency of 78% and decreased electrode resistance by mitigating the bulk resistance resulting from PTFE degradation.[60]

Another way of improving PTFE stability was demonstrated in 2024 by Han et al.[61] Fluoroethylene carbonate (FEC) was introduced as an electrolyte additive forming a stable electrolyte–binder interface (Figure 3b). The authors assessed the degradation of PTFE via nanoindentation and measured adhesion force: The cells containing FEC exhibited a higher elastic recovery, as well as stronger adhesion, which is ascribed to less PTFE degradation. Capacity retention and initial coulombic efficiency could also be improved significantly.

Zhang et al. evaluated the effect of hard and soft carbon as an active material instead of graphite.[62] According to the

Table 3. Overview on strategies for increasing the anode stability, indicating the increase in initial coulombic efficiency.

Evaluation parameter	CE increase	Refs.
PEO coating on graphite particles	47.1% → 83.7%	[59]
PVDF-based coating on graphite particles	67.2% → 77.8%	[60]
FEC electrolyte additive	72.5% → 83.8%	[61]
Hard/soft carbon instead of graphite	Not exactly mentioned	[62]
PVDF/PTFE co-binder blend	85.6% → 89% (w/prelithiation)	[63]

authors, the PTFE in hard or soft carbon electrodes was less prone to be reduced because the fibrils maintained their position due to low volume changes in the active material, reducing the tendency to contact the active material directly. The hard and soft carbon electrodes thus maintained their mechanical stability and showed a cycling stability comparable to slurry coated electrodes.

In a subsequent study, Zhang et al. produced a graphite anode containing two binders, PTFE and PVDF, to make use of the synergistic interactions between both polymers.[63] PVDF acted as the functional binder; while, PTFE was used as a processing aid to form a self-supporting electrode film. The electrodes showed a 95% capacity retention in half cells after 50 cycles at 0.23 mA cm^{-2}, with a loading of 4.22 mAh cm^{-2}, and kept their integrity despite PTFE reduction. The binding capability of PVDF led to good mechanical stability; while, a comparable electrode without PVDF showed a cracked microstructure due to PTFE fiber decomposition.

Stabilizing the PTFE binder on the anode side has long been the primary research focus for dry coating with graphite. The approaches presented in this work offer complementary solutions to this technological challenge. As summarized in **Table 3**, the incorporation of electrolyte or polymer additives within the electrochemical setup enhances coulombic efficiency and can counteract the effects of PTFE degradation. Therefore, it can be stated that the previously known negative impact of PTFE on battery performance is no longer the main obstacle in using DBE for the production of LIB.

With the development status presented for LIBs, high-quality cathodes and anodes can be produced. A variety of cathode materials can be combined with graphite anodes. In future, reports on how the challenges of the volume change of silicon-based anodes will be countered with DBE can be expected in order to increase energy density.

3.2. DBE for Next-Generation Batteries

3.2.1. Lithium–Sulfur Batteries

Lithium–sulfur battery (LSB) research has gained significant attention due to its potential for cost-efficient energy storage. Emerging in the 1960s, LSB offers a high specific energy, theoretically five times greater than traditional lithium-ion batteries.[64] This makes them an attractive option for applications requiring lightweight and efficient power sources, such as electric vehicles, drones, and renewable energy storage. In addition, sulfur is abundant and cost-effective, enhancing the sustainability and economic viability of these batteries. Current research aims to overcome challenges such as shuttling, metal anode degradation, limited cycle life, and sulfur's insulating nature, as well as cost-efficient production in order to pave the way to widespread adoption in various industries.

The application of dry battery electrodes in lithium–sulfur batteries was inspired by supercapacitors in which the carbon porosity plays a key role for the resulting performance, and wet-film processing might alter or even block carbon pores that are crucial for the supercapacitor storage mechanism. In related development, PTFE/ethanol/water-based kneading and rolling of small clay-like electrode sheets was common at that time, as described by Korenblit et al.[65]

Thieme et al. further developed this process to a complete dry process without the use of ethanol or water, and the PTFE content was drastically lowered.[66,67] They reported for the first time, the usage of dry film sulfur cathodes based on PTFE and micro-mesoporous carbons as host with sulfur as an active material (**Figure 4a**). For this process, the carbon/sulfur material is blended in mortar with PTFE spheres until agglomerated flakes form. The flakes are then further rolled out by a stainless-steel roll until the desired film thickness is reached. Within this procedure, the film is folded several times. The resulting freestanding film (the film is mechanically self-sustaining due to its high mechanical stability) is typically laminated onto primered current collector foil for further electrode preparation. The primer layer is an ≈1 µm thin conductive and adhesive layer enabling adhesion and improving interface resistance. This manual dry film method has been widely employed for material development in LSB R&D because it is well suited especially for small carbon material amounts with precisely adapted porosity.[68–71] As a key advantage over slurry processing, dry formulation avoids sulfur sublimation as no drying step is required. The risk of sulfur evaporation after slurry coating and subsequent drying arises as most solvents have high boiling points, temperatures at which sulfur has significant vapour pressure.[72,73] For cathode active material development using manual DBE processed electrodes, ZnO-templated micro-mesoporous carbons without[70] and with[74] nitrogen doping have been reported. Especially for air- or moisture-sensitive active materials such as Li_2S[75] or Na_2S,[76] the manual dry film process is particularly suitable as it can be easily carried out in a glove box and the binder chemistry or pH value does not need to be adapted for the respective active material. Over the last 10 years, it has become standard practice to preferably carry out LSB material and component development in pouch cells, as recommended in a perspective paper by Oxis Energy, for example.[77] In consequence, the need to carry out dry film manufacturing at elevated scale was emphasized, enabling high component

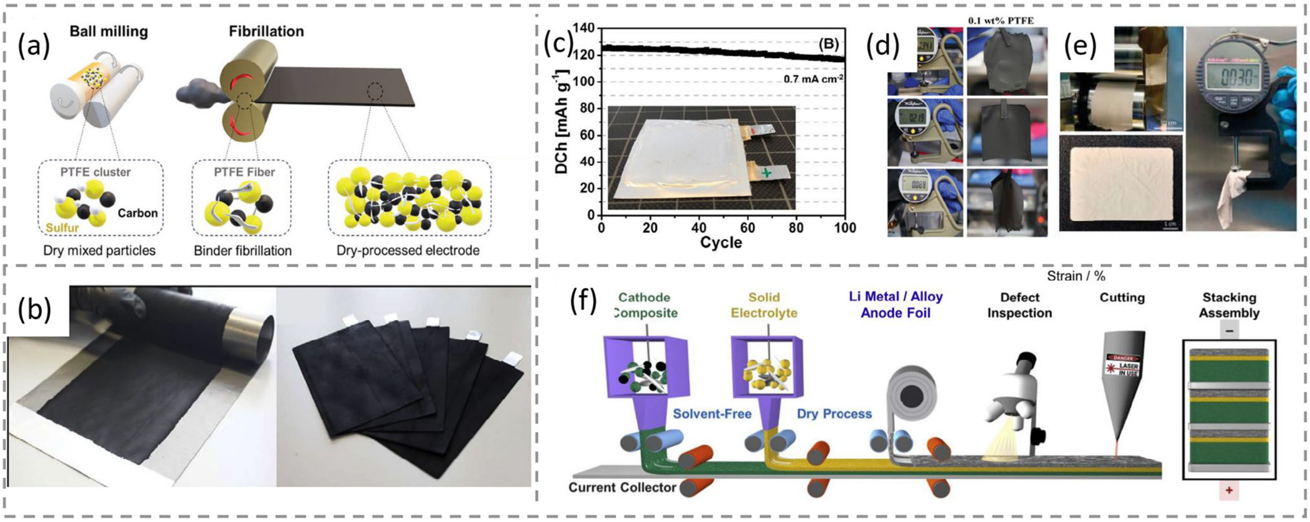

Figure 4. a) Schematic procedure for manufacturing dry-processed carbon/sulfur electrodes by PTFE fibrillation; Reproduced with permission.[68] Copyright 2024, Wiley; b) roll-to-roll processed carbon sulfur electrodes for pouch cell evaluation; Reproduced with permission.[78] Copyright 2020, Elsevier. c) all-dry processed sulfide-SE based solid-state battery pouch cell; Reproduced with permission.[83] Copyright 2019, Elsevier; d) carbon/sulfur/solid-electrolyte composite electrode sheets prepared with 0.1 wt% binder at different thicknesses; Reproduced with permission.[84] Copyright 2023, Wiley. e) 30 µm thin sulfidic solid electrolyte layer. Reproduced (adapted) with permission.[85] Copyright 2021, American Chemical Society. f) Schematic dry SSB manufacturing. Reproduced with permission.[86] Copyright 2022, Elsevier.

homogeneity; and thus, evaluation in multi-layered pouch cells is recommended. Using nitrogen-doped carbon blacks, Kensy et al. were the first to report a comparable study on sulfur cathodes produced by both a manual dry film and electrodes from a continuous DBE process (Figure 4b).[78] A drastic swelling of the dry processed films with expansion of up to 200% was noticed, and consequently, further investigated by Schmidt et al. using confocal microscopy.[79] Accordingly, the cell stack pressure is a key performance indicator for sulfur cathodes produced by dry film coating as analyzed in a follow-up study.[80] In addition, by adjusting the cathode composition and using different porous carbon black to carbon nanotube ratios, the swelling effect can be controlled leading to decent specific energies over 400 Wh kg^{-1} in pouch cells.[81]

As a sustainable binder alternative to PTFE, sericin was reported to create fibrillar structures under certain processing conditions. Decent performance of sericin-based carbon/sulfur dry film electrodes was demonstrated in pouch cells.[81]

With the increasing interest in DBE over the past 3 years, dry coating for sulfur cathodes has attracted further researchers in the battery community. Horst et al. reported on the influence of the mixing procedure for a binder-free dry cathode manufacturing on the resulting electrode performance.[82] Manthiram et al. recently published a comparative study on a manual method for sulfur and Li_2S DBE cathodes.[68] With 1 wt% PTFE binder content, extremely high sulfur loadings of up to 12 mg cm^{-2} at a relatively low E/S ratio of 6 with remarkable cycle and rate performance could be demonstrated.

Future developments for lithium–sulfur-batteries may benefit from the studies mentioned above. One example is the all-solid-state concept of the lithium–sulfur battery chemistry with sulfidic solid electrolyte, which will be discussed in the following section.

3.2.2. Sodium-Ion Batteries

Sodium-ion batteries (SIB), the main advantage of which is the low cost of raw materials, are likely to represent an alternative to LFP-based batteries.[87–89] Surprisingly, at the time of writing this review, only few reports exist on dry-coated electrodes for sodium-ion batteries.[62,76,90,91] The cathode active materials $Na_3V_2(PO_4)_3$ (NVP) and $Na_{0.75}Ni_{0.25}Fe_{0.25}Mn_{0.50}O_2$ (NFM) were processed into dry films with PTFE, as well as hard carbon for anodes. Kühn et al. compared the electrochemical performance of dry-coated versus wet-coated NMF cathodes in multi-layer pouch cells with slurry-based hard carbon anodes.[91] They observed comparable cycling stability over 400 cycles. The rate-capabilities of both electrodes provided advantages for the wet-processed cathode, which was related to a different electrode density (dry: 1.8 g cm^{-3}; wet: 1.3 g cm^{-3}).

With increasing pressure on the cost structure of rechargeable batteries, strongly increasing numbers of publications in the field of SIB can be expected over the coming years.

3.2.3. Solid-State Batteries With Sulfidic Solid Electrolyte

State of the art lithium-ion batteries opened a wide field of applications ranging from consumer electronic to electric vehicles and energy storage. Their combination of high energy density, power density, and long term stability comes with a steady decrease in production costs. However, further improvements are limited to incremental steps, whereas solid-state batteries (SSB) promise to enable great opportunities due to new electrode chemistries such as zero-excess or silicon anodes. This can lead to a huge increase in energy density by up to 70%.[92,93] Solid state technology is an enabler to rethink electrode interfaces and cell chemistries fundamentally, leading to a playground of new possibilities for scientists.

3.2.4. SSB Cathodes

One crucial difference in the preparation of solid-state batteries is that the solid electrolyte (SE) must be considered in the electrode structure from the beginning, and it does not simply fill every void during infiltration. The electrode morphology plays an even more important role on the battery's performance. Many research studies report data without any binder by just pressing pellets, which are several hundreds of microns thick, having a drastic influence on energy density.[94] However, for a transition from the lab scale to the production of larger electrodes, a binder has to be used to assure processability.[9] The binder adds another component changing the morphology of the electrode. The contact area of the active material with the solid electrolyte is already lower than for liquid cells and the classic wet coating approach reduces it even more. Nam et al. demonstrated in GITT measurements that the surface coverage of a dry processed NMC cathode with solid electrolyte reaches values of only 25% at moderate electrolyte contents. A similar wet processed cathode exhibits a reduced coverage of ≈18%.[95] This directly influences partial ionic and electric conductivities of cathode and separator composites.[96,97] In a wet coating, the binder polymer dissolves and obstructs the surface of the active material particles. Unlike swelling with liquid electrolyte to function as a gel electrolyte, it merely acts as an insulator, impeding electron and ion flux.

Dry processing is a perfect solution for solid state batteries, especially for systems using sulfidic solid electrolytes.[19] These materials are very sensitive to moisture and their ionic conductivity decreases when used as a slurry. In dry coating, PTFE can be used in very low amounts of ≈0.1 wt%.[98] The PTFE fibrils spanning like a spider web across the electrode particles basically do not interfere with the electrode morphology and compaction.[99] Hence, electrode performance can be preserved perfectly even for high areal loading of ≈6 mAh cm^{-2}, as demonstrated by Hippauf et al. in 2019 (see Figure 4c).[83] They realized the lowest binder content at that time for an SSB cathode and the successful fabrication of an all-dry pouch cell consisting of a dry-processed cathode, separator, and anode. For the first time, PTFE was shown as a suitable binder for all components of a solid-state battery despite its reactivity at low potentials. Since then, this technology has found widespread application by other research groups. The method was adapted by Lee et al. in their publication about the zero-excess anode system resulting in a high-energy long-cycling solid-state lithium metal battery, where a demonstrator cell delivered a record-breaking energy density of >920 Wh L^{-1}.[100,101] After Cangaz et al. combined dry-processed cathodes with 100% columnar silicon anodes in 2020, Tan et al. combined dry cathodes with μm-silicon electrodes to prepare high energy full cells.[86,102] Some studies even suggest that the shearing induced during dry processing can increase surface coverage of the CAM and allow increasing rate capability because the porosity after compaction is drastically reduced.[103] This demonstrates that the electrode morphology vastly influences the battery performance. As the PTFE fibrils do not interfere with electrode morphology, a well-directed design of high areal loading electrodes with carefully adjusted ionic conduction pathways becomes possible.[104] Several research groups demonstrated that areal loading above 8 mAh cm^{-2} can be realized using dry electrode processing, which is crucial for achieving the highest energy densities.[105,106] This is one major advantage compared to slurry based coatings. The latter have their limitations when it comes to high loading due to higher impedance in SSB cells and the formation of cracks that occur during drying.

3.2.5. Lithium–Sulfur-Solid State Batteries (LSSB)

Beyond solid-state lithium-ion batteries, solid-state cell concepts such as lithium–sulfur-batteries based on conversion chemistry gained interest in recent years. Lithium–sulfur cathodes typically consist of three components, solid electrolyte, sulfur, and a carbon matrix, which are intensively mixed, for example, by ball-milling to ensure triple-phase-boundaries. Consequently, the particles are present in nm-scale, leading to a cathode morphology which strongly differs from particular lithium-ion-cathode morphology, as discussed previously. Nonetheless, dry electrode processing of solid-state lithium-sulfur-cathodes was achieved in literature. Hu et al. demonstrated dry film cathodes consisting of sulfur/carbon/argyrodite with PTFE contents down to 0.5 wt%. The dry film cathodes are characterized by lowered electronic and ionic resistances compared to wet-processed electrodes and uncompromised electrochemical performance compared to powder cathodes.[107] Fiedler et al. demonstrated that free-standing cathodes could be realized, even with low binder contents down to 0.1 wt% PTFE (Figure 4d).[84] Further, the long-term stability was shown, realizing 400 cycles by usage of a dry film cathode. Additional research from Fiedler et al. indicated that the dry film process was feasible for all-solid-state lithium–sulfur cathodes with various carbon materials, resulting in different morphologies with particles in μm-scale.[108] After the feasibility of dry processing of lithium–sulfur-cathodes was demonstrated, Yang et al. focused on high-areal-capacity cathodes, aiming for 5.0 mg(S) cm^{-2}.[109] They prepared sulfur cathode sheets with a high sulfur loading of 4.5 mg cm^{-2}, demonstrating outstanding electrochemical performance with high reversible capacity, good cycling stability, and rate performance. Further, they demonstrated an all-solid-state sheet-type $Li_{3.75}Si$/LiSiPSCl/S cell with sulfur cathode sheet, LiSiPSCl electrolyte membrane, and $Li_{3.75}Si$ anode sheet and exhibited remarkable electrochemical performance.

Besides LSSB, a wide range of cell chemistries has been addressed by the scientific community. Shen et al. demonstrated dry film processing of conversion cathodes containing different CAMs. Using FeS_2 as active material, they achieved sheet-type cathodes with low PTFE contents down to 0.5 wt%, which were cycled for more than 200 cycles in pouch cells.[110] A promising alternative was also suggested by Kirchhoff et al., who introduced a novel semiliquid–solid Li–S-battery cell concept, using an argyrodite-based LSSB cathode, a lithium anode, and liquid electrolyte, enabling specific energy above 600 Wh kg^{-1}. [111]

The preparation of a dry film-based solid electrolyte layer (SEL) as a separator in SSB has been addressed in the above-mentioned studies. As this topic is of specific interest for solid state battery manufacturing, we will discuss it in more detail in the following section.

3.2.6. Solid Electrolyte Layers (SEL)

Aside from electrodes, there is an increasing interest in dry processing of solid electrolyte layers (SEL). These layers act as ion-conducting separator between anode and cathode and consist almost entirely of SE. As solvent-processed SEL (e.g., by slurry casting) show substantially reduced ionic conductivities (loss of ≈50% compared with the pristine solid electrolyte material), a strong motivation is given for developing manufacturing processes sustaining the SE conductivity.[86,112]

In recent years, dry processed highly conductive and thin SE layers have been reported, such as a 30 µm thick sulfide SEL with high room temperature conductivity of 8.4 mS cm^{-1}.[85] This is of particular interest because thin SEL are crucial for manufacturing high-energy density SSB (Figure 4e). Recent studies have also used a variety of scaffold materials to increase the mechanical stability and structural integrity of thin SE films, such as cellulose or nylon membranes, non-woven, or electrospun fabrics.[101,109,113–118] Using PTFE as a binder and a cellulose mesh to reinforce the mechanical properties, flexible SE membranes of 30 µm thickness with high mechanical strength (8.1 MPa) and superior ionic conductivity (6.5 mS cm^{-1}) were achieved.[119]

As discussed above, while PTFE is currently the dominant binder of choice, it is electrochemically unstable at the low potentials on the anode side.[11,120] When combined with Li metal, PTFE is readily defluorinated and reduced to carbyne-type carbon.[121,122]

Nevertheless, there are several reports on successful cycling of solid-state pouch cells employing a dry processed SEL containing PTFE binder. Combining such a SE dry film with a 100% columnar Si anode resulted in stable cycling and an excellent CE.[102] Even with Li anodes, PTFE-based SE dry films have been successfully implemented by introducing a protective lithium iodide (LiI) layer between the SE and Li anode.[121]

However, the dry film processing of SEL is typically performed by small-scale calendaring. For upscaling, the major challenge of continuous and scalable processing of thin SEL has to be solved.[86]

With next-generation battery technologies promising great chances for improved battery performance, they also provide the opportunity for introducing new manufacturing technologies, such as dry coating. The advantages of combining dry coating and next-generation cell chemistries on the battery properties will catalyze this development in the near future. Significant improvements are still required in solid electrolyte production and scalable processing technologies to enhance the development and adoption of solid-state batteries. While extensive research has already been conducted on both cathode and anode dry electrodes, further advancements are expected in the coming years for solid-state systems. In addition, a key question remains regarding the ideal anode material for SSBs as neither silicon-based nor lithium-based electrodes have yet emerged as the definitive solution for the future.

4. Technological Approaches Toward A Scalable Production

Section 3 showed the multitude of cell chemistries and the versatility that DBE allows. Advantages regarding achievable thicknesses, rate capabilities, and material compatibilities were pointed out. However, a successful implementation of DBE in battery production requires scalable processes. In the field of battery electrode manufacturing, it is crucial to differentiate between dry coating and dry mixing processes. A common misconception in many studies and reviews is the conflation of these two distinct aspects. For instance, in the perspective of dry battery electrode, extrusion is often incorrectly identified as a dry coating method; while, calendaring is frequently relegated to a post-compaction step, primarily associated with slurry-based electrodes. In this Section, we will review the literature focusing on scalable mixture preparation and coating for the mechanical dry powder coating.

4.1. Dry Mixture Preparation

DBE manufacturing is strongly dependent on the powder preconditioning process or mixing step. This step is crucial to ensure not only the quality of the intermediate product before coating but also the structure of the electrode and the resulting electrochemical performance. Most academic publications are conducting mixing process at a lab scale in a mortar mill.[39,43–46]. First and foremost, it is important to recall that mixing powders involves setting the individual particles in motion within a mixer. The external energy required for this process can be supplied through various means: the movement of a vessel, the action of a blade passing through the bulk, gravity, or a combination of these three mechanisms. Depending on their chemical and mechanical properties, powders can exhibit highly variable responses to these forces. Ideally, they mix uniformly, but in some cases, they may instead tend to aggregate.

It is generally accepted that three primary mixing mechanisms exist, as illustrated in **Figure 5a**:[123,124]

i. Diffusion mixing: It induces the rearrangement of individual or small groups of particles. It is related to blade-to-bulk movement, to particle-to-particle, or particle-to-wall interactions. This is a slow process that promotes mixing at the particle scale.
ii. Convection mixing: It activates the movement of particle clusters, typically driven by a blade. This is a fast process that enhances spatial homogeneity and increases the contact surface between components.
iii. Shear mixing: It arises when a high velocity gradient forms between clusters of particles. This leads to the creation of shear surfaces, resulting in particle reorganization. Shear mixing is often difficult to separate from convection in practical mixing processes.[125]

The choice of mixing mechanism significantly influences powder behavior. In some cases, instead of achieving uniform mixing, granular materials may undergo a phenomenon named "segregation." This aspect, particularly relevant to DBE applications, can negatively impact the overall composite properties. To date, more than 19 different segregation mechanisms have been identified,[126] largely attributed to differences in particle size, density, vessel shapes, and energy applied during mixing.[124]

While such studies have been extensively conducted in industries such as coal processing, mining, and metallurgy, there is

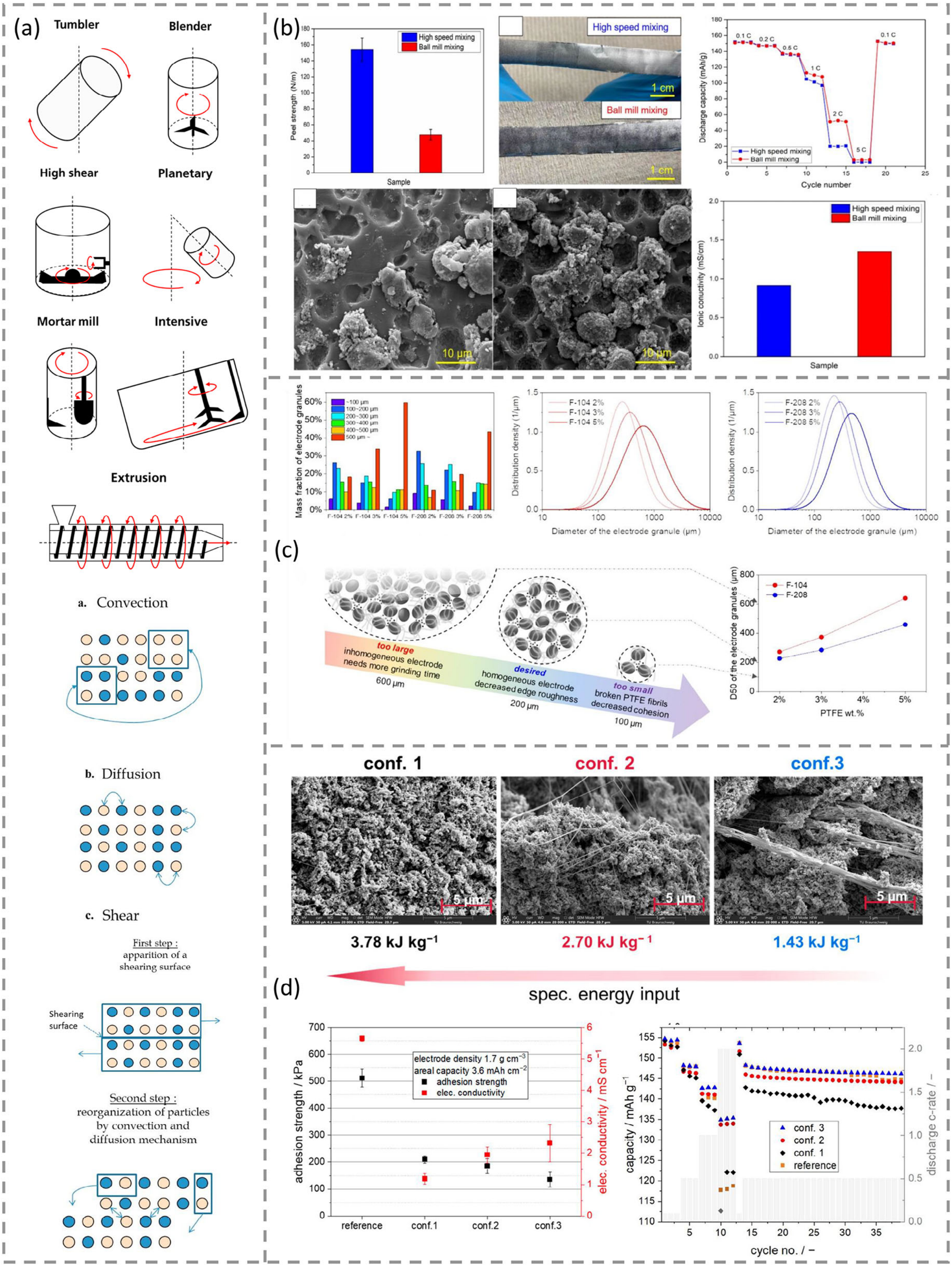

Figure 5. a) Schematic illustration of the different mixing setup and the three primary mixing mechanisms. Reproduced with permission.[124] Copyright 2015, Elsevier; b) Influence of different mixing protocols on peel strength, cycling, and ionic conductivity measurements of NMC-based calendared electrodes. Reproduced with permission.[129] Copyright 2023, Electrochemical Society; c) Influence of grinding and sieving step of PTFE-based powder to electrode granules properties. Reproduced with permission.[41] Copyright 2024, Elsevier; d) Impact of extrusion screw configurations on LFP electrode structure and electrochemical properties. Reproduced with permission.[137] Copyright 2023, MDPI.

a notable lack of research specific to DBEs, despite the comparable impact of mixing on final product performance. Therefore, the mechanisms outlined above have been integrated into the present section, which aims to explore mixing processes suitable for scale-up to pilot and industrial levels. Moreover, to clarify the differences between the mixing setups used in the literature and industry, schematic illustrations are provided in Figure 5a.

4.2. Batch Mixing

Various types of discontinuous mixers have been employed for the homogenization of electrode materials and PTFE fibrillation. In addition, mixing has been a key focus in numerous investigations involving conventionally slurry-processed electrodes. In the same way, the influence of dry mixing active materials and conductive additives on powder and electrode properties has been particularly well-researched. For instance, Bockholt et al. examined the efficiency of dry mixing NMC and carbon black using different mixer types, including a rotary drum, a high-shear, an intensive, and a planetary mixer.[127] It has been shown that the interaction between carbon black and NMC varies significantly depending on the mixing energy, which is influenced by the type of mixer used. For example, high-shear intensive mixing resulted in carbon black forming a coating layer on the active material; while, smaller agglomerates remained when using intensive and planetary mixers. Further, both intensive and planetary mixers achieved the highest electrical conductivity, whereas high-shear mixtures exhibited the highest powder packing density. The decrease in electrical conductivity for the well-coated NMC in the high-shear mixture was attributed to a strongly increased number of particle contacts, and simultaneously, a reduced contact area. This result indicates the existence of an optimum in terms of mixing degree, where dispersion of the material as well as its specific contact with the other material within the composite matters. Similar investigations have been made by Wenzel et al. in 2024.[128] In parallel, Wang et al. have differentiated the impact of high speed and ball mill mixing, equivalent to intensive and tumbler mixing setup, respectively, to the electrode properties. They demonstrated that ball milling can provide a more conductive film by homogeneously distributing the carbon additive into a porous layer (Figure 5b).[129] Chauhan et al. developed a numerical DEM model on the impact of intensive dry mixing on cathode microstructure and electrochemical performance establishing a link between bulk density and performance in half cells.[130] Changes in bulk density and structural characteristics due to reduced agglomeration of carbon black were identified as key performance determinants.

The dry mixing step of only active material and carbon black was also studied by Lischka et al.[131] They investigated the possibility to characterize the quality of dry mixing via the parameter bulk density. Therefore, NMC622 and carbon black were mixed in an intensive mixer, resulting in the powder bulk density increasing with increasing mixing time and speed. The study revealed that prolonged mixing does not enhance electrical conductivity due to the destruction of long-range carbon structures, which hinders the formation of continuous electrical pathways within the samples. The authors introduced a dimensionless number to predict the bulk density of the mixture based on mixing parameters. They concluded that higher powder conductivity is achieved at lower bulk densities.

In 2024, Oh et al. reported a scalable mixing process for PTFE-based NMC811 cathodes.[41] They evaluated intermediate products throughout the manufacturing process, including during mixing and before/after the lamination of the freestanding electrode films. The process involved pre-mixing, fibrillation in a kneader to create an electrode dough (monitored through torque measurements), followed by grinding in a powder mill to enhance processability. The torque required for mixing increased as the PTFE content rose. In addition, characterization was performed on the granule size of the ground electrode mass and the tensile strength of the freestanding films, with variations analyzed for different PTFE types and contents (Figure 5c). As expected, higher PTFE content increased the tensile strength of the films as well as granule size, but also ionic resistance and tortuosity. Edge roughness of the freestanding films was shown to be lower for decreased PTFE content. The ideal kneading duration was defined by SEM analysis at different mixing times to determine the limit between under- and over-fibrillated dough. A similar kneading technique was used by Hsieh et al.[132] The authors tested PTFE grades with varying fibrillation tendencies at temperatures ranging from 40 °C to 100 °C to compare the maximum kneading torque and torque rise time during the mixing process. Kneading torque increased as PTFE was sheared, driven by the formation of fibrils. Once fibrillation reached completion at maximum torque, the fibrils stretched and oriented within the electrode dough, reducing internal friction, and subsequently, lowering the kneading torque.

Depending on the PTFE grade, different kneading torques were required, and the time to reach maximum torque decreased significantly with increasing temperature—from 30 min at 40 °C to less than 5 min at 100 °C. While this study demonstrates the dependence of the mixing process and mixing parameters on material quality, the effects of these parameters on the direct properties of the fibrillated powder are still unknown and represent the main challenge currently facing the scientific community.

Discontinuous fibrillation of PTFE was also possible in a jet mill, as shown by Zhang et al. and Zhou et al.[48,54] After premixing the powders, they were subjected to a high speed air stream, resulting in a cotton-candy like dry mixture, as described by the authors. Lee et al. conducted the mixing process for anodes with polymer coated graphite in a planetary centrifugal mixer.[60] Tao et al. investigated PTFE fibrillation in a ball mill with a variation of mixing time between 10 and 60 min.[133] Insufficient mixing for shorter durations resulted in low PTFE fibrillation; while, excessive mixing led to the breakage of fibrils, forming a PTFE-carbon black film. The tensile strength of the free-standing films was highest after prolonged mixing, whereas adhesion peaked after 30 min of mixing. Although the ohmic resistance of the three electrodes remained similarly low, the charge transfer impedance was significantly reduced for the electrode mixed for 30 min. Both under- and over-mixing degraded the mechanical properties of the electrodes and compromised their rate performance. In contrast, an optimal mixing time enhanced PTFE fibrillation, mechanical properties, and electrochemical performance. The study concluded that a moderate degree of mixing is required for good processability and performance of dry electrodes. While some studies focus on the binder impact on dry mixing process, other publications address the influence of active material particle morphology. Horst et al. compared graphite, LFP, and NCM regarding the fibrillation behavior of PTFE depending on the active materials used during the dry mixing process.[134] Under identical mixing conditions, PTFE fibrillated more slowly with graphite due to its platelet-like structure, which favored sliding over fibrillation. LFP, with its high fine particle content, caused prolonged PTFE fibrillation as the small particles were intermittently absorbed by the PTFE. In contrast, NCM, containing fewer fine particles, exhibited faster PTFE fibrillation.

Another notable study on mixing for dry electrode fabrication was conducted by Gyulai et al.[135] The experiments were conducted with PVDF as a binder but general findings might be transferable to PTFE electrodes. The authors processed dry powders in a planetary centrifugal mixer, showing the importance of mixing parameters for the subsequent calendaring steps. They investigated the processability window of electrode mixtures for cathodes with 3% PVDF by varying the mixing intensity among low, medium, and high intensity mixing with different rotational speeds. SEM images and powder conductivity measurements showed that for low intensity mixing, most carbon black agglomerates were still intact. Due to insufficient binder distribution, the process window in a roll-to-roll calendar, in terms of roll speed and shearing ratio, was smallest for the low-intensity mixed powder. Sufficient cohesion within the electrode film was achieved over a larger process window for medium- or high-intensity mixed samples, which also resulted in better capacity retention and thinner electrodes. Yonaga et al. also examined the effect of mixing intensity in a high-shear mixer on electrode structure and electrochemical performance.[136] While capacity retention was similar at low C-rates, the medium-intensity mixed electrodes exhibited the best performance at 20 C. Both studies highlight the importance of considering the entire process chain in dry coating as the various process steps are highly interdependent.

Thus, mixing processes significantly impact the final electrode properties, but they must also be considered alongside the downstream processes needed to shape the powder into an electrode. Therefore, more research should be dedicated to scalable technologies for continuous mixing, as will be discussed in the following section.

4.3. Continuous Mixing

A continuous process for dry electrode mixing and PTFE fibrillation by shear forces is extrusion. Continuous mixing in an extruder provides the advantage of theoretically being able to feed the mixed product continuously into the calendar gap or coating feedstock. In the past, extrusion has been used for slurry mixing, studying important parameters such as temperature and screw configuration (Figure 5d).[137] Rotational speed,[138] as well as the resulting degree of filling and residence time within the extruder, have also been investigated.[139] Many studies were made for a slurry-based process, but effects of certain parameters can be expected to be similar in dry processing. The feasibility of extrusion mixing with PTFE binder in a completely solvent-free process was reported by Huber et al.[140] for graphite, and by Quérel et al.[141] and Tao et al.[40,51,142] for NMC. The latter applied extrusion mixing in a 20 mm twin screw extruder at a feed rate of 1.5 kg h^{-1} and a temperature of 45 °C, producing a flaky powder mixture.

Wiegmann et al. investigated a semi-dry extrusion process for PTFE-based LFP cathodes processing with 1% PTFE and an increased solids content compared to conventional methods.[137] Screws with different kneading intensity were utilized, and the resulting electrodes were tested for adhesion strength, electrical conductivity, and elastic deformation behavior via nanoindentation. Semi dry granules extruded with a strong screw showed a large number of finer fibrils, leading to higher adhesion strength, lower electrical conductivity, and a greater proportion of elastic deformation, but lower discharge capacities.

Detailed studies on dry PTFE fibrillation in an extruder and the impact of the mixing process on intermediate products along the process chain as well as on cell performance are a subject of recent research and can be expected from future publications in the field.

Based on the state-of-the-art research in this field, a list of criteria can be established to ensure maximum efficiency in scaling up and achieving a successful mixing process:

1) Homogeneity of the mixture: to distribute uniformly the different materials within the powder and ensure specific particle size distribution
2) Particle integrity: to avoid breaking or altering the shape of active material particles and to prevent thermal degradation or changes in surface chemistry
3) Binder activation: to ensure the binder adheres adequately to the active material and conductive additives, as well as to coat homogeneously the particle to improve adhesion
4) Material flowability: to maintain free-flowing powder properties to facilitate calendaring/pressing downstream processes and minimize the formation of aggregates that can impact uniform coating or electrode properties
5) Powder density: to achieve densities suitable for high-performance battery electrodes without causing over-compaction and to ensure reproducibility through uniform packing density
6) Electrochemical properties: To maintain electrochemical electrode integrity (e.g., specific capacity, stability) and to ensure the absence of contamination during the mixing process (metallic impurities)
7) Process scalability: to maintain quality; while, scaling up the mixing process from lab-scale to industrial-scale production and to establish controllable parameters (torque, speed, time...).
8) Compatibility with downstream processes: To exhibit properties that facilitate uniform deposition on current collectors during dry coating process and to ensure the effective compression and shearing during calendaring/pressing
9) Environmental and safety considerations: to minimize dust generation to ensure a safe working environment
10) Monitoring and quality assurance: to implement techniques such as spectroscopy or imaging for real-time quality control

Table 4. Summary of the different mixing technologies with their pros, cons, and specific examples.

Mixing	Mechanisms	Pros	Cons
Tumbler	– Diffusion	– Simple and cost-effective setup – Minimal energy consumption – Gentle on sensitive materials	– Poor dispersion for small particles – Limited control over distribution – High risk of agglomeration
Blender	– Diffusion, convection	– Versatile and widely available – Small-scale powder blending – Low cost and easy operation	– Poor dispersion for small particles – Limited scalability for industry
High-shear	– Convection, shear	– Excellent dispersion of powders – Effective for breaking up agglomerates – Faster mixing time	– Potential material degradation – Higher energy consumption – Rapid equipment wear
Planetary	– Diffusion, convection	– Uniform and consistent mixing – Small-batch materials	– Limited scalability for industry – High maintenance requirements
Mortar mill	– Diffusion, shear	– Effective for breaking up agglomerates – Gentle on sensitive materials – Fine control over particle size	– Poor dispersion for small particles – Labor-intensive – Limited scalability for industry
Intensive	– Convection, shear	– Superior mixing efficiency – Precise control over material dispersion – Scalable for industrial production	– High CapEx – Batch process – Complex maintenance and operation
Extrusion/kneading	– Diffusion, convection, and shear	– Continuous operation for high production rates, scalable – Good material dispersion and powder compacting	– High CapEx – Complex process control – Wear issues and limited flexibility

and to maintain tight tolerance for material specifications and mixing parameters

It is also possible to provide a comparison between the different mixing setup, which is summarized in the following **Table 4** as a function of their mixing mechanisms[123–126,143,144] and pros/cons regarding dry battery electrode.

Based on the previous discussion, it is clear that measuring and controlling the mixing and fibrillation processes of PTFE-based powder for DBE requires careful consideration of several factors. For mixing, efficient dispersion of materials to ensure homogeneous electronic contact between particles is crucial, without damaging the carbon additives' structures or the already-formed PTFE fibers. Under-mixed powder would result in poor coating abilities and electrochemical performance; while, over-mixed powder could lead to the mechanical degradation of its components, negatively impacting the final electrochemical properties. An optimal mixing condition exists for each composition, but identifying and measuring this optimum remains a challenge due to the lack of available technology.

Regarding fibrillation, the main focus is currently to determine the degree of fibrillation; while, applying a recipe. Promising results have been achieved by measuring the torque or mechanical resistance of the powder using a kneader. An under-fibrillated powder will not provide sufficient mechanical stability or particle bonding for effective coating on the electrode, whereas an over-fibrillated powder may increase the composite hardness too much and cause plasticization, reducing its coating capability.

Advancements should still be made in the mixing and fibrillation processes to enhance our understanding and control of the mechanisms involved. However, these steps should always be considered in relation to the dry coating technologies applied afterward. This aspect will be further discussed in the following section.

4.4. Dry Coating

Degen et al. have reviewed various dry coating methods in terms of technology readiness level and performance potential.[7] They concluded that mechanical dry powder coating using calendar rollers is the most advanced technology today (**Figure 6**). Great progress toward industrial-scale commercialization of calendar-based DBE coating has been reported over the last several years.[13,15,16] Now, a distinction should be made between conventional calendaring, which is used solely to compact an already formed electrode, and direct calendaring, where an electrode is created directly from a dry mixture. Given this clarification, it becomes apparent that mainly two techniques are currently being investigated as true dry coating processes: Maxwell-type and IWS DRYtraec.

Both techniques make use of fibrillated PTFE as binder and process the dry film in a calendar device. While with the Maxwell-type process, freestanding films are prepared,[37] the DRYtraec avoids the formation of a freestanding film and provides the film formation on the calendar roller, followed by a direct transfer to the current collector foil.[13] Both techniques will be explained in more detail later in this chapter. Other methods, such as extrusion, hot pressing, powder spraying, or gas-phase processes, are at a much lower TRL today and far away from an industrial-scale of commercialization for battery production.

The influence of the calendaring process on electrode performance has been a significant area of investigation, particularly as a post-fabrication step for both slurry- and dry-coated electrodes. Calendaring, originally a compaction process, plays a crucial role regarding the mechanical and electrochemical properties of electrodes.

In the realm of dry electrode manufacturing, Gyulai et al. have notably explored direct calendaring process.[135] The authors identified two distinct zones within the calendaring setup, between the rolls: the "feed zone," where powder transport and

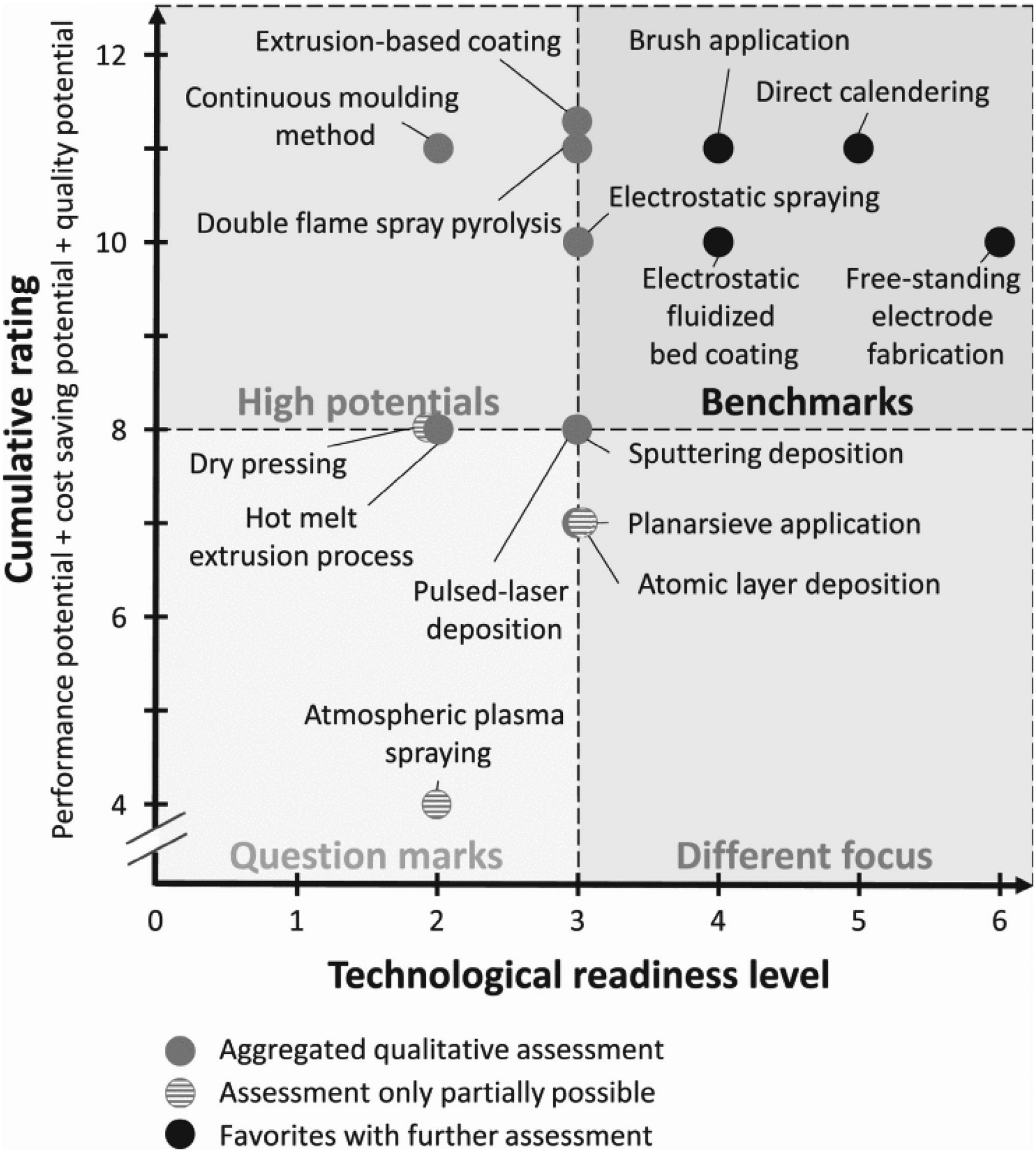

Figure 6. Attractiveness of dry coating technology in terms of TRL and overall cumulative rating. Reproduced with permission.[7] Copyright 2022, IEEE.

internal friction occur, and the "nip zone," where the powder experiences high normal and shearing forces, leading to compaction (**Figure** 7a). Interestingly, their study draws more from the field of metal powder compaction and granular solids theory rather than from traditional slurry-based science. This highlights a gap in the scientific understanding of calendaring for dry powder systems as most references are older than a decade.

Regarding calendaring as a post-fabrication step for electrode densification, several key aspects were highlighted in recent years, which are still equally relevant for DBE dry direct calendaring science, such as:

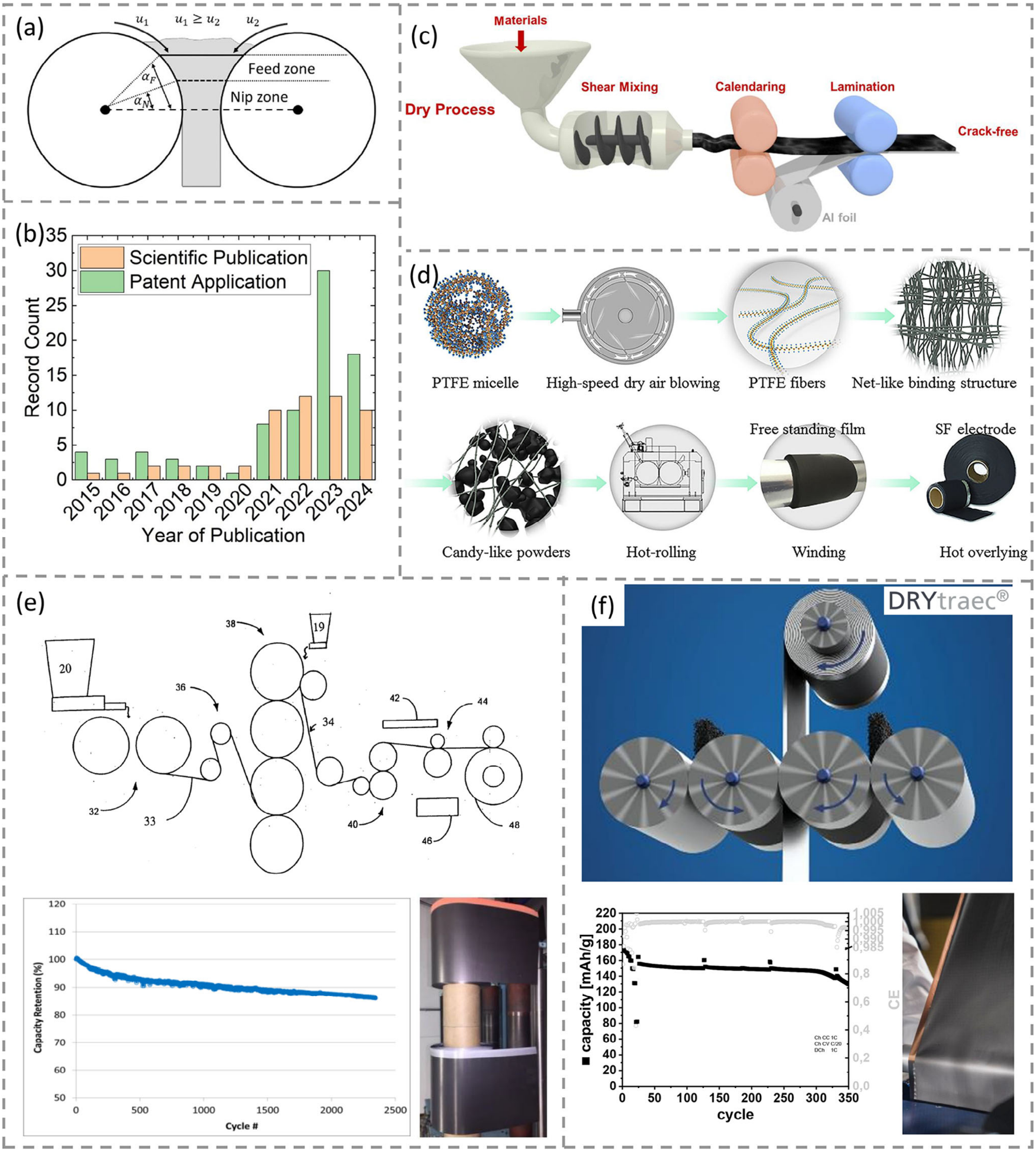

Figure 7. a) Scheme of a vertical powder calendaring process with the distinct gap zones and characteristic parameters. Reproduced with permission.[135] Copyright 2023, American Chemical Society; b) Comparison of the annual publication figures for scientific publications and patent applications using the key word over time "Dry Battery Electrode AND Calendaring" (Sources are from https://patentscope.wipo.int and https://www.webofscience.com). c) Scheme of dry electrode cathode fabrication procedure using extrusion-like mixing. Reproduced with permission.[50] Copyright 2023, Royal Society of Chemistry; d) Schematic representation of the fabrication route for the DBE process using a high-speed dry air mixing step.[54] e) Maxwell-type dry film process and cycle performance NMC/graphite electrode produced by this method. Reproduced with permission.[38,151] Copyright 2018, Power Sources Conferences. f) Shear-based coating process DRYtraec and cycle performance of NMC/graphite full cell produced by this method. Reproduced with permission. [155] Copyright 2024, Electrochemical Society; Fraunhofer IWS.[15,16,155,165]

1) Material-dependent structural response: Understanding the link between calendaring and the structural response of materials is essential to optimize processes for various active materials and applications. For dry coatings, the size, morphology, and amount of binder (typically PTFE) are critical in controlling the porosity and density of the electrode. No optimal condition can be universally applied to a single active material; rather, it must be tailored to specific formulations with secondary products such as binders and additives.[145]
2) Compaction resistance: This parameter is crucial for understanding the electrode coating's resistance to compaction. It has been found to be highly dependent on materials, formulations, and pore structures.[146]
3) Porosity and mass loading: Studies have focused on the compaction of electrode coatings, analyzing factors such as porosity, compaction resistance, and the dependency on mass loading. Equations and models based on Heck's work[147] have been used to characterize the compaction process, noting a linear relationship between compaction resistance and mass loading. This relationship is influenced by factors such as temperature and additive content, with compaction resistance decreasing at higher temperatures due to the thermoplasticity of the binder material.[148]
4) Effects on separator microstructure: In slurry-based systems, the effects of calendaring on layers such as LPS and LPSCl have been studied concerning line load, roll temperature, and roller speed. The "spring back" effect after densification can negatively impact the separator microstructure, affecting ionic conductivity. Pre-densification effects on separators can lead to decreased specific ionic conductivity with increased densification, impacting lithium-ion transport.[147]

Despite the advancements in understanding calendaring processes, there remains a significant gap in the scientific literature regarding the direct calendaring of dry electrodes. Only a few papers mention its use, and these often lack detailed descriptions of the manufacturing process.[49,50] This gap underscores the need for further research to bridge the divide between current calendaring technologies and their scientific understanding, particularly for dry powder systems.

An important difference in terms of publications has to be considered between the key words "Dry Electrode Battery" and "Dry Battery Electrode AND Calendaring." The numbers of publications for the latter indicate the lack of studies and innovation about calendaring science and technology in these last years. However, the amount of patent applications is still higher than scientific publications regarding these domains (Figure 7b). This comparison reflects the industry's major interest in the technology, and more specifically, the race for intellectual property rights to manifest a position in the dry battery electrode market.

4.4.1. Maxwell-Type Process

The classical process for calendar-based DBE coating has been intensively utilized by the company Maxwell Technologies. They have built up a broad patent portfolio originating from supercapacitor electrode production, but also including DBE for LIB, which has later been acquired by Tesla.[149–154] Even though it followed a long list of historical approaches (see Section 2), this technology was the first to manufacture free-standing PTFE-based dry film supercapacitor electrodes on a large industrial scale. The published manufacturing stages included the raw material mixing and PTFE fibrillation using a jet mill.[151] Subsequently, the powder was fed into a calendar-type roll-mill, resulting in the formation of a thick free-standing film (Figure 7e). Using a compaction step in a calendar cascade, the film was rolled down to the desired thickness. A final lamination step was used to combine the dry film with the current collector and form the electrode.

As mentioned in Section 3, many research groups have been adopting Maxwell-type technology for lab-scale developments. However, few publications used the process to fabricate large-sheet electrodes for both lithium-ion and solid-state batteries. For instance, LNMO electrodes were produced by Maxwell-type process showing excellent cycling stability for 3.0 mA h cm^{-2} areal capacity with 68% capacity retention after 1000 cycles in full cell setup (Figure 7c).[50] Zhou et al. used the Maxwell-type process for the fabrication of LFP electrode for a hybrid supercapacitor, showing a capacity retention of 92% over 5000 cycles, and a fibrous PTFE structure was maintained after 1000 cycles.[54] The fabrication route followed by the authors to obtain the free-standing electrode is shown in Figure 7d. Regarding NMC-based electrodes, Duong et al. have demonstrated a 4 mAh cm^{-2} dry coated NMC/graphite pouch cell that delivered 90% capacity retention after 2000 cycles at 0.5 C charge / 1 C discharge (Figure 7f).[38]

The impact of PTFE fibrillation on the Maxwell-type dry process was also briefly examined. Recently, Horst et al. demonstrated how the degree of powder fibrillation and electrode calendaring vary as a function of particle morphology and physical properties.[134] They also showed that while materials with rapid fibrillation, such as NMC—due to its spherical and fine particle shape—may exhibit fast fibrillation, this does not necessarily simplify the calendaring process. Instead, calendaring is more influenced by the material's compressive strength and compaction speed, which are notably high in the case of NMC. Consequently, each composite material and powder must be studied and optimized independently.

Many challenges remain regarding the application of Maxwell-type process for battery production, whether in terms of the minimum thickness that the method can achieve, film quality (surface degradation due to excessive mechanical stress), adhesion to the current collector, or even the mechanical strength of the film, essential for handling the film without breaking it. Especially this last point represents the most important bottleneck for a wider use of the Maxwell-type process in the industry for both LIBs and SSBs applications.

4.4.2. DRYtraec Technology

With the major drawback of handling a fragile freestanding film with thicknesses < 100 µm and at high process speeds in a scaled process, a need for alternative dry coating approaches was desired by the industry. In the labs of Fraunhofer IWS, Althues et al. developed the so called DRYtraec process which can avoid the formation of a freestanding film (Figure 7f).[13,15,16] Different rotational speeds of the used calendar rollers induced

Table 5. Comparison of the most advanced DBE technologies: Maxwell-type, DRYtraec, and electrostatic powder spray.

Dry coating	Pros	Cons
Maxwell-Type (freestanding, self-supporting film)	– Already commercialized, for example, for supercapacitors – Equipment available from several companies – Well suited for soft carbonaceous materials and thick films (>200 µm) – Substrate-independent – Simplicity allows application at lab-scale – Tandem coatings by multi-webs possible – Compaction decoupled from film formation	– Thin films poorly achievable – Roller cascade needed for thin films – Multiple process steps – Risk of material over-densification – Risk of production downtime due to cracks – Mechanically self-sustaining films required, leading to narrow binder classes – Patent protected
DRYtraec (roller-supported)	– Scalable and continuous process – Single-step process, reduced footprint – Large process window (40,… 500 µm), even for ceramic materials – Versatility for broad active material and binder classes reduces electrode compaction – Tandem coatings possible – Combination with roller cascade possible, but not necessarily required – Films processed even with low mechanical stability or binder content – Technology accessible by licensing	– Emerging technology not yet in production – High-precision calendar equipment required – Production equipment to be established – Complex parameter optimization – Primed or perforated substrate required – Film adhesion on roller surface required – Risk of roller wear due to friction
Electrostatic powder spray	– Established in various industries – Simple equipment setup – Various thermoplastic binders can be applied – Simple pre-mixing can be applied – Throughput can be increased with multiple coating guns	– Material distribution strongly depends on size and conductivity of individual particles – Overspray leads to poor edge quality and homogeneity – Occupational safety and health issues related to toxic dust and ATEX – Subsequent thermal compaction needed – Not yet demonstrated at > TRL5 for batteries

additional shear forces in the calendar nip. Further fibril formation along the direction of rotation was the consequence, enabling the adhesion of the resulting dry film on the roller surface. The film remained on the roller before being laminated to a current collector foil. Thus, it was always mechanically supported, either by the roller or by the substrate, and had not necessarily to be self-sustaining. DRYtraec allowed the formation of films in one process step down to 50 µm without the need of a calendar cascade. The risk of production downtime due to film tear could be reduced by this approach, providing a real scalable dry coating technology alternative to the Maxwell-approach. The technology had been demonstrated for roll-to-roll coating of LIB-electrodes, as well as for cathodes for SSB, SIB, and LSB.[155] In this work by Kaskel et al., full-cell testing of NMC and graphite electrodes produced using the DRYtraec process demonstrated a cycle life of 300 cycles at 1 C with a capacity of 3.2 mAh cm^{-2} (Figure 7f). However, the DRYtraec process is still an emerging technology that has yet to be extensively studied. For instance, the impact of PTFE anisotropically oriented fibrillation in the DRYtraec process has not yet been studied. As a result, a detailed comparison between the DRYtraec and Maxwell-type processes, in terms of the mechanical and electrochemical properties of the electrodes, requires deeper analysis and a comprehensive data basis. Further studies on composition and process optimizations are expected to be reported in the coming years using this technology. With their advanced TRL, calendaring methods will be the first methods to find their way into DBE mass production. However, process alternatives with lower TRL such as spray coating or hot pressing should be mentioned as well.[156–164] They are considered especially for non-PTFE binder systems and may gain higher relevance in future once a basic proof of electrode quality and process scalability is reached.

In recent years, dry coating technologies have gained significant attention, paralleling the growing interest in dry battery electrode (DBE) techniques. The scalability and improved coating capabilities achieved through roll-to-roll systems, whether using Maxwell-type or DRYtraec processes, have demonstrated their potential for widespread use in the industry. As summarized in **Table 5**, each of these processes has its own strengths and weaknesses, signaling their future role in battery manufacturing. For example, Maxwell-type technology is already commercialized for supercapacitors and is well-suited for thick films, making it a straightforward option for both lab-scale and industrial applications. Many public communications were already made regarding its application to DBE. However, it faces challenges with thin film production and is prone to issues such as material over-densification and production downtime caused by cracking. In contrast, the DRYtraec process is, as most roll-to-roll methods, scalable with a broad process window. Its key advantage lies in its streamlined, one-step process, which significantly reduces the time required for dry electrode production. In addition, the use of the roll as a mechanical support during electrode lamination helps mitigate the risk of low mechanical stability associated with low binder content. However, as an emerging technology, DRYtraec requires high-precision equipment and complex parameter optimization; and is therefore, not yet fully mature for industrial integration.

From a technical perspective, the limitations specific to each process are well-understood, and it is only a matter of time before these issues are addressed. Nevertheless, there are still unresolved obstacles that directly affect the overall DBE concept. These challenges, along with potential future solutions, will be discussed in the following section.

5. Challenges and Future Directions

The advantages of dry coating are obvious: lower production costs, reduced space requirements, compatibility with next-generation battery technologies, and so on. This has been substantiated in the preceding Sections. Although the first pilot plants for dry coating have been put into operation,[166–169] the implementation in production plants on a GWh a^{-1} scale seems to bring some challenges.[170,171] In this Section, we will address possible causes, obstacles, and challenges that currently hamper mass production of DBE.

5.1. Alternative Binder Systems

The use of PTFE as a binder in dry battery electrodes offers several advantages. PTFE provides excellent mechanical properties to electrodes, making it a reliable choice for ensuring high-performance and long-lasting battery performances. By maintaining the overall electrodes structures and ensuring a strong adhesion to primered current collectors, PTFE binders enable efficient ionic and electric transports within electrode, mandatory for high energy and high powder density battery applications. However, despite these advantages, the use of PTFE also presents several challenges.

First, as a fluorinated polymer, PTFE raises environmental concerns due to the potential release of harmful fluorinated compounds such as PFOA during its production and disposal. These compounds are persistent in the environment and can contribute to pollution. Under the REACH regulation, certain harmful fluorinated substances, including perfluorooctanoic acid (PFOA), face increasing restrictions.[172] Similarly, the EU's F-gas regulations aim to phase out fluorine-based chemicals to mitigate environmental impacts.[173] These regulations will lead to a need for improved PTFE production processes and certification for using PTFE in industrial applications such as battery production. Considering the battery as closed system, the release of F-polymers into the environment could be neglected. However, the same has to be reached during the production of the polymers where the main risk of environmental impact is assumed nowadays. With recent discussions on the carbon footprint of PTFE versus PVDF, it is expected that the polymer manufacturers will publish their data with a life cycle analysis of their materials. At the moment, only outdated data is available from public sources or databases. Initiatives and regulations such as the battery passport will catalyze the publication of certified and recent data.

Second, it has been shown that PTFE reduces during lithiation on anode side, participating to capacity lost over cycling[36] (as discussed in Section 3 of this review). The demand for modified PTFE or binder alternatives is rising. However, for the moment, there is no ideal solution to replace PTFE. The aim of this section is to briefly summarize the latest advances in alternative binders for dry battery electrode manufacturing.

Several studies address the search for alternative binders by hot pressing of mixtures containing thermoplastic binders, temperature-curable resins, or waxes.[174–177] However, these materials may not be applicable as sole binder in DBE production using direct calendaring methods, but may be considered as co-binders or when using alternative DBE methods. Regarding the preparation of solid state batteries using polymer-based solid electrolytes, PEO (polyethylene oxide) may be used as both, binder and solid electrolyte. This was reported, for example, by Helmers et al. who demonstrated the scalable LFP/PEO electrode production by extrusion and calendaring with manufactured pouch cells achieving 140 mAh g^{-1} at 0.1 C and 75/50 mAh g^{-1} at 1 C discharge rate for extruded-calendared and directly calendared electrodes, respectively.[178]

Following the prominent calendar-based DBE technology, the research from Schmidt et al. introduced a biopolymer as alternative to PTFE.[179] Using the polypeptide sericin, the authors demonstrated comparable binding performance in lithium–sulfur batteries with a discharge capacity of 1136 mAh g^{-1}(S) after 35 cycles. They showed that the sericin binder provides fibril formation under the influence of shear forces; and thus, offers a similar binding mechanism when compared to PTFE.

Besides these new binder material approaches, it can be expected that blends of PTFE and PVDF will be of industrial relevance in the near future. A partial substitution of PTFE will allow to sustain the film formation properties; while, the PVDF content will ensure long-term electrochemical stability. It is important to note that there is currently a lack of academic studies specifically focused on these blending concepts for enhanced anode stability. Among a few others, the study by Zhang et al. can be mentioned, where the authors have investigated the impact of PTFE/PVDF mixture into graphite electrode manufacturing, demonstrating 95% capacity retention after 50 cycles with 15 mg cm^{-2} loading.[48,63] Mixing two market-available binders is promising, straight-forward, and should be further exploited. The mentioned gap in academic research presents an opportunity for further investigation and validation of the approach using co-binders, cross-linkers, and binding additives. Despite the current absence of extensive academic evaluation, the strategy of using blended binders holds significant promise for the future of dry battery electrode manufacturing. Consequently, this approach requires continued attention and consideration from both industry professionals and academic researchers.

5.2. Scale-Up

As described in Section 4, methods for scaled dry mixing and dry coating for DBE have been published. However, for scaled production, additional factors need to be considered.

5.2.1. Economic Consideration

Disregarding all other aspects and advantages of dry coating and focusing solely on the pure energy cost advantage, a cost threshold for electricity procurement per kWh can be derived, at which point dry coating becomes superior due to the cost advantage. Below this threshold, the energy cost advantage plays a subordinate role. Consequently, arguments of sustainability through solvent avoidance or advantages in achievable product properties (e.g., high electrode loading) come to the forefront. These considerations influence the location choice for dry coating gigafactories. Conversely, there is higher pressure to introduce an energy-saving technology in countries with high electricity prices. As the leading cell manufacturers produce mainly in countries with comparatively low electricity prices (China, Korea, Japan, USA),[180] there has been relatively low pressure to introduce dry coating in these companies. However, it is foreseeable that with the global ramp-up of battery production capacities, production will also take place in countries with high electricity costs. At the same time, there is significant economic pressure on the battery product from the consumer side, indicating an increased necessity for DBE-based battery production in the coming years. This is reflected in the announcements of major industry players.[168,169,181]

5.2.2. Availability of Turnkey Plants With Successful Validation at TRL-9

Companies along the value chain for battery electrode manufacturing have adapted their products for DBE in recent years and built a corresponding product portfolio. This applies to material manufacturers, as well as equipment or component manufacturers.[182–187] In the equipment engineering sector, the number of companies on the market offering pilot plants for dry coating or mixture production has significantly increased in recent years.[166,185,186,188–190] This trend follows the need of companies in the cell manufacturing sector that want to evaluate dry coating in pilot plants.[168–170,181] Following this development logic, no equipment manufacturer is currently known whose DBE plants at the pilot scale have successfully been evaluated at TRL-9 for the production of LIB cathodes and anodes and who freely offers turnkey solutions for equipping gigafactories. For this reason, significant financial resources are still required both by cell manufacturers and equipment engineers to take the next development step toward production-ready plants and processes. Current obstacles are related to the design and manufacturing of calendar rollers, which provide long-term stability with abrasive materials at high pressures on widths of > 1 meter. Once this gap is bridged and providers have placed corresponding turnkey solutions on the market, the risk and development effort for cell manufacturers will decrease. The entry barrier to DBE production will be significantly reduced, leading to an expansion of DBE production capacities.

5.2.3. Integration and Linking of Sub-Processes

Storing, cooling, dosing, mixing, conveying, coating, or post-treating the electrodes—these are all sub-processes in battery electrode manufacturing. In modern battery production with a high degree of vertical integration, all process steps are coordinated and linked.[1,6] Material flows and plant utilization are optimized. For dry coating, the respective process steps have so far only been described separately. Aspects of the interdependence of the sub-processes and their linkage have not yet been reported. As explained in Section 4, it is being investigated how an optimal dry mixture can be produced. However, how it is then stored, conditioned (cooled, loosened), and fed into the coating process has hardly been discussed in the technical literature. Scientific data is required on the behavior of the used powders, for example, feeding and dosing a dry mixture which has the tendency to be highly cohesive, especially without cooling. To address this need, it is expected that production researchers and factory planners will increasingly begin to focus on the specific challenges of linking sub-processes for DBE.

5.2.4. Characterization and Methodology

In today's battery R&D and production, several standard tools have been established for product development and quality control. Slurry rheology, materials distribution, conductivity, adhesion, or electrochemical stability can be named as representatives for product and intermediate properties which are evaluated with similar setups in battery labs around the world.[129,134,145,191–193] Some of these methods can also be applied for DBE.[134] However, there are some new requirements for dry coating R&D analytics as well as for process and quality control, which cannot be addressed with the conventional tools. No at-line or in-line techniques are available which are dedicated to the requirements of DBE powder preparation. Few studies started to evaluate the behavior of the powder before or during mixing, with shear cell test[130,131,135] and kneading torque measurement.[41] Although further studies and expertise are needed on these aspects, such approaches are promising if the relationship between the state of the powder before fibrillation and the electrode characteristics after formation are demonstrated. As an example of a recently published study, Horst et al. explored an intriguing approach by examining how particle morphology influences PTFE fibrillation during mixing and calendaring.[134] To achieve this, they analyzed various structural, compaction, flowability, and physical properties. Notably, shear cell and uniaxial compression tests proved particularly useful in linking PTFE fibrillation effects to the intrinsic characteristics of the active material, whether NMC, LFP, or graphite.

Despite the increasingly systematic link between the properties of materials and their shaping, no studies have yet provided predictive measures for key aspects such as the degree of fibrillation or the ability of a dry mixture to form a homogeneous film. In consequence, dedicated methodology and tools are required for in-depth characterization of the mechanical and rheological properties of dry mixtures for DBE applications. Moreover, a demand from the industry for dedicated in-line process control during powder handling, mixing, coating, and so on can be expected. Today, efficient electrode formation is mostly governed by empirical optimization. A predictive design making use of rationalized structure–property relationships, atomistic simulations, or advanced AI based tools is in its infancy.

5.3. Authors' Perspectives

The presented publications show that there has been immense progress in the field of DBE in recent years. Several research institutions and companies are working on innovative solutions along the value chain. It is to be expected that fibrillated-binder-based technology will increasingly enter mass production in the coming years. Accordingly, some innovations that have recently been realized on a laboratory scale are on the verge to be industrially implemented.

From the outstanding results described in the area of DBE cathode development, a clear trend toward highly loaded electrodes can be observed. As the production of thick layers represents a significant advantage of DBE over slurry, positively affecting energy density, it can be assumed that this innovation will find rapid industrial application. For the associated challenges regarding ionic and electrical conductivity, the described approaches for incorporating CNTs with a focus on an accessible microstructure will be applied. Although the approaches described on a laboratory scale will be associated with process-engineering compromises, such as enabling a homogeneous distribution of CNT, they still offer outstanding potential, which, in addition to the advantages of DBE regarding energy savings, will bring clear product advantages that will catalyze disruption in electrode manufacturing. More broadly, 3D structured additives appear to be a promising solution for enhancing the mechanical stability of electrodes; while, ensuring adequate electrical conductivity.

The processing of graphite electrodes as DBE is mechanically–rheologically simpler than the cathode coating process; however, the issue of PTFE decomposition on the anode poses a technical challenge. It is expected that this challenge will be addressed in the medium term with at least one of the described PTFE-based approaches (or a combination of several approaches). To suppress PTFE decomposition, the use of binder blends, modified PTFE (with co-polymers), or electrolyte additives are considered the most promising options. Moreover, while reducing the amount of PTFE appears to be the most suitable short-term strategy, no scalable process has yet demonstrated the ability to achieve a binder content below 0.5 wt% without compromising electrode mechanical stability. Consequently, it is becoming increasingly clear that alternative approaches should be further explored. The use of fluorine-free binder alternatives will be intensively researched in the coming years; however, it can be assumed that a substitute binder with nearly similar properties to PTFE will be challenging to find, and industrial use is therefore not foreseeable.

The enormous advancement of next-generation batteries is another motor for DBE industrialization: For SSB, LSB, SIB, and others, only few more years may be required before mass production at gigafactory scale can begin. Thus, it is increasingly likely that by the time of their market launch, DBE will also have already matured toward mass production. Consequently, innovative start-ups in the next-gen battery sector will increasingly be loath to use outdated slurry technology and instead invest directly in emerging DBE technologies. This trend is particularly identifiable in SSB with sulfide solid electrolytes: The slurry-based processing of solid electrolytes leads to reduced ionic conductivity. At the same time, the discussed works show that highly loaded SSB cathodes can be achieved with DBE; while, maintaining high ionic conductivity of the electrode. Therefore, the authors assess the use of DBE for the commercialization of SSB with sulfide solid electrolytes as very likely. However, whether DBE will be the method of choice for producing the SEL will largely depend on what minimum layer thickness can be achieved with the method. In this discipline, the procedure has significant disadvantages compared to the slurry process, calling for further scientific investments to address this challenge in the coming years.

The discussed scalable production techniques on the way to mass production still leave much room for innovation. We expect parallel developments to differentiate competitors. As discussed, for example, both batch or semi-batch high-intensity mixers and continuous extrusion or kneading processes can lead to the desired result in the production of the dry mixture. However, this must always be seen in combination with the coating method used. To produce freestanding electrodes using the Maxwell-type process, a different preconditioning of the mixture is needed compared to processing in the DRYtraec process with roller-supported film formation due to different rotational speeds. Therefore, adapting specific powder preparation methods and processes to the different components of the final battery setup appears necessary. For instance, the extrusion process shows promise for both cathode and anode electrodes with minimal PTFE when integrated into the DRYtraec process, particularly suitable for low-binder amount mixtures. However, for freestanding solid electrolyte layers in solid-state batteries, which require mechanical stability, low porosity, and compatibility with anode lamination, a high-intensity mixing combined with the Maxwell-type process could be a beneficial combination. While the Maxwell-type process has been used in mass production of supercaps and is currently ramped-up for battery application, the DRYtraec approach has gained significant interest recently but needs to be validated in pilot plants being currently established.

To summarize this author's perspective view, DBE technology is rapidly advancing toward mass production, driven by its ability to enable thick, highly loaded cathodes; while, improving energy efficiency. Its adoption in next-generation batteries, particularly solid-state designs, is increasingly likely. However, challenges remain in optimizing conductivity, binders, and solid electrolyte processing. The integration of DRYtraec and Maxwell-type processes could further enhance scalability, but mass production feasibility still needs demonstration. Continued innovation and collaboration will be key to establishing DBE as a standard in future energy storage solutions.

6. Summary and Conclusions

Dry battery electrode (DBE) technology is on the verge to industrialization and an implementation in future gigafactories is highly relevant. Starting from early development applications using PTFE dry coating in the 1960s to the 1990s in various electrochemical components, the technology was further advanced and further adopted in lithium-based battery electrode research. By establishing fundamental formulations and processing methods today, the relatively simple processing is applied for a wide variety of battery active materials and additives. The effective demonstration of high-quality electrodes lays the foundation for the widespread adoption of the DBE method in numerous research laboratories. Several cathode active materials have been processed using DBE techniques at the lab scale. For lithium-ion batteries, most studies examine the interplay between cathode active material and its particle type and form with other electrode components such as binders and conductive additives. Their focus is primarily on the impact on rate capability, cycle stability, and mechanical stability. As a conclusion of these studies, PTFE-based dry cathodes for lithium-ion batteries can be produced as high-quality alternatives to slurry cathodes without performance loss. Anode system studies are less diverse in terms of active materials used, with graphite dominating except for a few studies with LTO. Binder stability is the most critical factor here. However, DBE processed graphite anodes are feasible for production.

Further, dry coating is most promising for next-generation battery technologies. Especially, lithium–sulfur batteries profit as heat treatment after slurry coating becomes obsolete, avoiding sulfur sublimation. DRYtraec is the method of choice for all solid-state batteries. In particular, electrodes containing sulfide solid electrolytes and solid electrolyte layers (SELs) benefit from dry coating approaches as degradation of the solid electrolyte caused by solvent contact can be avoided. However, producing thin SELs (<30 µm) with high mechanical stability is still challenging.

The most widely used DBE-technologies for dry coating battery electrodes promise a significant reduction in footprint and manufacturing costs by eliminating the energy-intensive drying step. Moreover, the avoidance of toxic solvents is a major driver for sustainable production.

Considering advances in industrial implementation, dry mixing and electrode production through dry coating with the dry mixture are distinct processing schemes. In comparison to the number of publications on lab-scale electrode development, production research and technology scaling are in its infancy. For dry mixing, achieving homogeneous distributions of binder and carbon black is a key target. The order of component addition, setting the desired fibrillation degree, optimizing mixing tools, and the impact on bulk properties and electrical conductivity are crucial investigation components. Unfortunately, an accepted reference mixing method has not yet emerged. Continuous kneading and extrusion processes are highly relevant, as well as high-intensity batch mixers. Storage conditions for the dry mixture and dosing capability remain to be targeted scientifically for advancing production capabilities.

For dry coating, two distinct methods are on the verge to industrial large scale implementation. Both processes rely on layer formation of the dry mixture in the calendar gap. The conventional, industrially implemented method (Maxwell process) involves producing free-standing dry films as intermediates, which are laminated onto the target substrate after an additional rolling step. The second method (DRYtraec process), not yet fully implemented in industry, uses different rotational speeds in the calendar gap. Film formation is supported through additional shear forces, avoiding fragile free-standing films that are usually obtained in Maxwell process at low binder content

In summary, dry coating has a high potential for technological disruption. However, challenges remain on the path to mass production. Equipment availability and binder chemistry are important areas for advancing the field and commercialization. Characterization standards and process quality measures are important targets, as well as predictive and rational approaches toward structure–property relationships.

As dry processing is currently among the most dynamic battery topics in academia and industry, this review can only provide a snapshot of the most relevant features. Interdisciplinary research, encompassing materials and binder development, including functional rheological additives, as well as cell implementation, mass production, metrology, and machine engineering, poses a wide open arena in a field with significant ecologic and economic relevance.

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Conflict of Interest

The authors declare no conflict of interest.

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battery production, calendar, dry battery electrode, dry coating, lithium-ion battery, next-generation battery, PTFE

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Benjamin Schumm is Division Manager for Particle Technology at Fraunhofer IWS. He studied Chemistry at Dresden University of Technology, received his Diploma in 2009, and his Ph.D. in 2012, focussing on thin film technologies. In 2012, he joined the Fraunhofer Institute for Material and Beam Technology (IWS) as research associate in the field of chemical surface technologies. B.S. became group leader for Chemical Coating Technologies at IWS in 2015 and focused on coating developments for battery applications from lab scale to prototype scale. He co-invented the DRYtraec technology and was the main driver behind the progress of this technology.

Arthur Dupuy obtained his Materials Science Engineering degree at Polytech Marseille in 2016. He then joined the Université de Sherbrooke in Canada to study the electrochemical etching of semiconductors and their application in lithium-ion batteries. He received his Ph.D in 2021. In the same year, he joined Fraunhofer IWS as research associate, with the purpose to link materials science to process development for dry electrode manufacturing, especially for solid-state batteries.

Stefan Kaskel studied chemistry and received his Ph.D. in Tübingen (1997). After a postdoc in the group of J. D. Corbett, he was a group leader at the Max Planck Institute for Coal Research. Since June 2004, he is Full Professor of Inorganic Chemistry at TU Dresden, and since 2008, also the head of the technology field Battery Technology at the Fraunhofer IWS. His research interests are focused on porous and nanostructured materials for applications in energy storage, catalysis, batteries, and separation technologies.