Chemical Science

ROYAL SOCIETY OF CHEMISTRY

REVIEW

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Cite this: *Chem. Sci.*, 2025, **16**, 6598

Sustainable and cost-effective electrode manufacturing for advanced lithium batteries: the roll-to-roll dry coating process

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The transition to electric vehicles motivated by global carbon neutrality targets has intensified the demand for lithium-ion batteries (LIBs) with high energy density. While the innovation of cathode/anode active materials has reached a plateau, development of thick electrodes has emerged as a critical breakthrough to achieving high-energy-density LIBs. However, the conventional wet coating process has intrinsic limitations, such as binder migration during the solvent drying process, which becomes increasingly problematic with thick electrodes. To address these challenges, dry coating processes have been actively explored in three main forms: electrostatic spraying, hot pressing with thermoplastic polymers, and roll-to-roll dry coating utilizing the polytetrafluoroethylene binder. This review highlights the roll-to-roll dry coating process, a scalable and industrially viable approach, by introducing its underlying mechanisms, latest developments, and applications in all-solid-state batteries and lithium–sulfur batteries. By combining technical advancements with manufacturing scalability, the roll-to-roll dry coating process demonstrates significant potential to enable next-generation battery systems.

Received 4th January 2025
Accepted 21st March 2025

DOI: 10.1039/d5sc00059a

rsc.li/chemical-science

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1. Introduction

1.1 Demand for sustainable and cost-effective electrode manufacturing for high-energy-density lithium batteries

As the global drive for carbon neutrality has accelerated the transition from internal combustion engine vehicles to electric vehicles (EVs), the demand for medium- and large-scale high-energy-density lithium-ion-batteries (LIBs), essential for long driving ranges and fast charging, has grown significantly.[1–3] To meet these demands, nations and industries around the world have initiated projects, such as the U.S. Department of Energy's Battery 500, Japan's NEDO RISING II, and China's "Made in China 2025", all targeting energy densities exceeding 500 W h kg^{-1} by 2030 (ref. 4–8) (Fig. 1a). Since Sony's commercialization of lithium–cobalt oxide and graphite-based LIBs in 1991, significant progress has been made in increasing the energy density of LIBs through material innovations.[9,10] Advances such as high-nickel cathode materials, silicon-based anode materials, and high-voltage electrolytes have enabled LIBs to achieve

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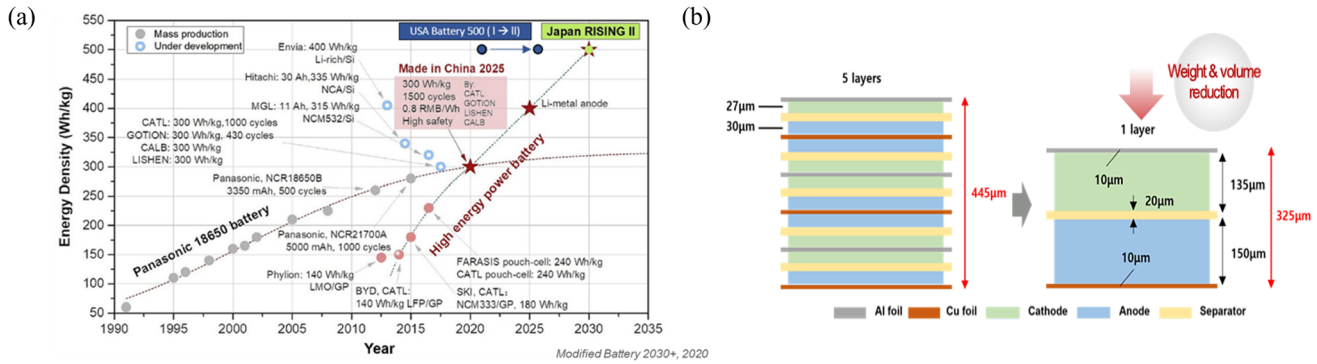

Fig. 1 Demand for sustainable and cost-effective electrode manufacturing for high-energy-density lithium batteries: (a) the history and development of Li-ion batteries according to the worldwide nations and industry. Reproduced with permission from ref. 8. Copyright (2019) Elsevier B.V. (b) Schematic illustration of the advantages of thick electrodes in terms of energy density of lithium rechargeable batteries.

energy densities of approximately 300 W h kg^{-1} for commercial applications and up to 350 W h kg^{-1} in the laboratory scale.[11–15] However, without the introduction of innovative materials that can exhibit a higher capacity than the theoretical capacity of existing cathode and anode materials, further increases in the energy density of LIBs through material innovation alone are limited. The limitation of materials innovation has shifted research attention to electrode design, particularly to thick and dense electrodes with high mass loadings, which can be applied to various battery systems such as lithium metal batteries (LMBs), all-solid-state batteries (ASSBs), and lithium–sulfur (Li–S) batteries.[16,17] Thick and dense electrodes present a promising pathway to achieving higher gravimetric and volumetric energy densities by reducing inactive components such as current collectors and separators.[18–20] For instance, transitioning from conventional electrode thicknesses (~25 μm) to thick electrodes (~200 μm) can increase the fraction of the cell occupied by

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active materials, thereby enhancing energy densities by reducing the stack count required in a battery pack (Fig. 1b).

1.2 Comparison of dry and wet processes

Achieving high-energy-density LIBs with thick and dense electrodes poses significant challenges in the conventional wet coating process. The wet coating process, the industry standard for manufacturing electrodes, is hindered by several limitations that compromise both electrochemical performance and environmental/sustainability/economic.[21,22] The critical issue of the wet coating process is binder migration during the solvent drying process, which induces an inhomogeneous microstructure in the electrode[23–26] (Fig. 2a). During the solvent drying process, the binder migrates toward the top region of the electrode along with the solvent due to capillary forces. Then, the binder blocks the pores in the top region of the electrode while leaving behind large voids within the electrode or causing an uneven distribution of active materials. When the calendering process is applied to such an electrode, localized collisions between active material particles lead to cracks and the difference in composition of electrode components between the top and bottom regions of the electrode further contributes to the formation of an inhomogeneous microstructure. The inhomogeneity of the electrode microstructure becomes more pronounced as the electrode thickness increases because the increase in solvent drying time aggravates the binder migration. Accordingly, wet-processed thick electrodes exhibit significant differences in porosity and density within the electrode after the calendering process[27,28] (Fig. 2b). The inhomogeneity of the electrode microstructure degrades lithium-ion transport kinetics, leading to an increase in ionic resistance and poor electrochemical properties such as cycling stability or rate capability.[29–33] Furthermore, the reliance on organic solvents in the wet coating process, such as *N*-methyl-2-pyrrolidone (NMP), increases energy demand for solvent evaporation/recovery and generates hazardous waste, adding to the environmental and economic costs of the battery manufacturing process.[34,35] In contrast, dry coating processes exclude these drawbacks by removing the solvent drying and recovery processes entirely.[36] The dry coating processes effectively prevent binder migration caused during the solvent drying process, enabling the uniform distribution of active materials, conductive additives and binders within the electrode and resulting in improvements in the homogeneity of the electrode microstructure and electrochemical properties[37,38] (Fig. 2c). Moreover, dry coating processes align with global sustainability goals by removing the use of toxic solvents, reducing energy consumption by approximately 46%, and lowering production costs by up to 19%.[39,40] The advantages of the dry coating process over the wet coating process, as outlined in Table 1, establish the dry coating

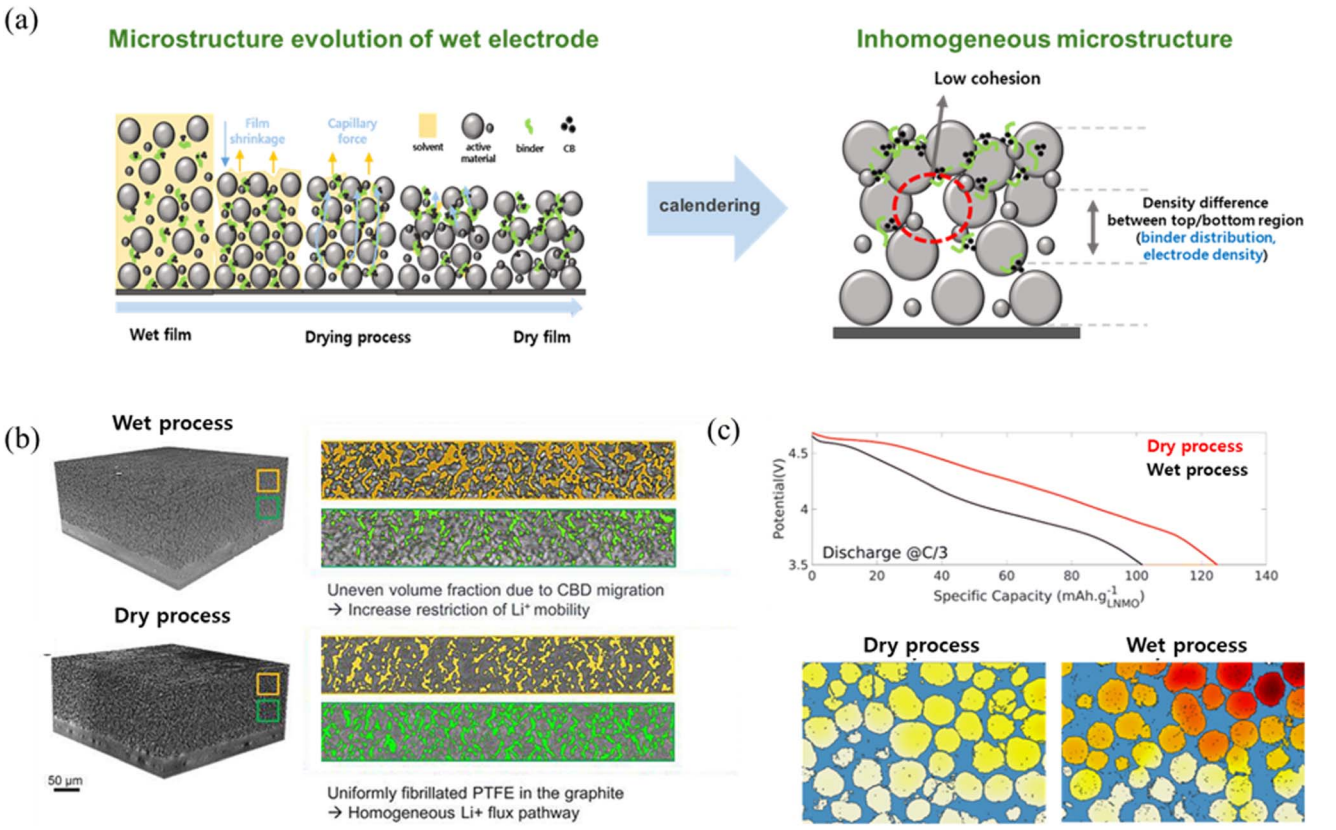

Fig. 2 Comparison of dry and wet processed electrodes: (a) schematic illustration of degradation of wet-processed thick electrodes during the solvent drying and lamination process by binder migration. (b) Comparison of the electrode microstructure and (c) electrochemical performance. Reproduced with permission from ref. 28 and 37. Copyright (2023) Elsevier B.V. and copyright (2023) RSC Publishing.

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Table 1 Comparison between wet and dry coating processes

	Wet coating process	Dry coating process	Reference
Environmental effect	Using toxic solvents Excessive CO_2 emissions	Eliminating toxic solvents	41 and 42
Process efficiency	Complex process	Simplified process (no drying/solvent recovery processes)	43 and 44
Cost	High OPEX and CAPEX (using NMP solvent, drying infrastructure)	Low OPEX and CAPEX (estimated reduction of the process and solvent)	22, 45 and 46
Microstructure	Inhomogeneous microstructures attributed to the binder migration during the fabrication of thick electrodes	Homogeneous microstructures associated with uniform binder distribution during the thick electrode fabrication	26 and 47
Areal capacity	Limitations caused by binder migration (<7 mA h cm^{-2})	Increase in thickness and density of electrodes fabricated by the dry process (≥5 mA h cm^{-2})	48 and 49

method as an economically viable and environmentally friendly manufacturing option for medium- and large-scale high-energy-density LIBs.

This review also explores the dry coating processes in next-generation rechargeable LIB systems. In particular, the roll-to-roll dry coating process was highlighted among various dry coating processes with the future research directions to achieving high energy density LIBs that will support the transition to a sustainable energy future. By integrating material innovations with sustainable manufacturing processes, the battery industry can meet the growing demand for high-energy-density and environmentally friendly energy storage systems.

1.3 Introduction of various dry coating processes

Dry coating processes have emerged as transformative technologies in the realm of battery electrode manufacturing, offering a sustainable alternative to the conventional solvent-based wet coating process. These methods eliminate the need for solvents, thereby reducing environmental impacts, improving energy efficiency, and addressing the growing demand for global carbon neutrality. Among the various dry coating processes, electrostatic spray coating, hot pressing, and roll-to-roll dry coating stand out, each offering unique advantages and challenges to meet the specific requirements of electrode manufacturing.

1.3.1 Electrostatic spray coating. Electrostatic spray coating utilizes high-voltage electric fields to precisely deposit a mixture of active materials, binders, and conductive additives onto a substrate[50] (Fig. 3a). The process begins with the preparation of a homogeneous dry mixture, which is introduced through an electrostatic spray nozzle. The high voltage applied to the nozzle generates a fine mist of charged particles, which are directed toward the substrate due to electrostatic attraction. These charged particles adhere uniformly to the substrate's surface, forming a thin and uniform layer. After deposition, a thermal or mechanical stabilization step enhances the mechanical integrity and chemical stability of the deposited layer. The primary advantage of electrostatic spray coating lies in its precision and ability to produce highly uniform layers with precise control over thickness, which is critical for achieving consistent electrochemical performance.[51–53] Additionally, the capability to apply extremely thin coatings makes this technique ideal for advanced battery designs that demand uniformity in layer thickness and material composition. Brandon *et al.* demonstrated the use of this method by preparing a dry mixture of the $LiCoO_2$ active material, carbon black (CB) conductive additive, and PVdF binder using a high-energy mixer, which was deposited onto an aluminum current collector using electrostatic spray coating.[45] The mechanical strength and electrochemical properties of the fabricated electrode were evaluated and compared with a wet-processed electrode of the same composition. The dry electrode exhibited superior performance, which is attributed to the homogeneous distribution of the binder achieved through the electrostatic spray coating process. Similarly, Mohanad *et al.* employed the electrostatic spray coating method to fabricate both positive and negative electrodes and compared their properties to those of wet-processed electrodes with the same composition.[54] They reported that the dry electrodes showed improved properties resulting from the uniform distribution of binders and conductive additives on the surfaces of graphite and NCM811 particles. This uniform distribution also acted as a protective layer, minimizing side reactions with the electrolyte and promoting the formation of stable CEI and SEI layers on the active material surface. Consequently, the dry electrodes achieved high areal capacities (6 mA h cm^{-2}) and demonstrated excellent electrochemical performance. Despite its advantages, the electrostatic spray coating method faces challenges for the commercial production of thick electrodes for medium-to-large secondary batteries, due to the reliance on high-voltage equipment with high complexity and increased operational costs. Additionally, the slow deposition rate limits its suitability for high-throughput manufacturing environments.[55] These limitations currently restrict its application to small-scale production and specialized markets where precise control of the electrode architecture is essential.

1.3.2 Hot pressing. Hot pressing is a dry coating technology that combines high temperatures and pressures to compact

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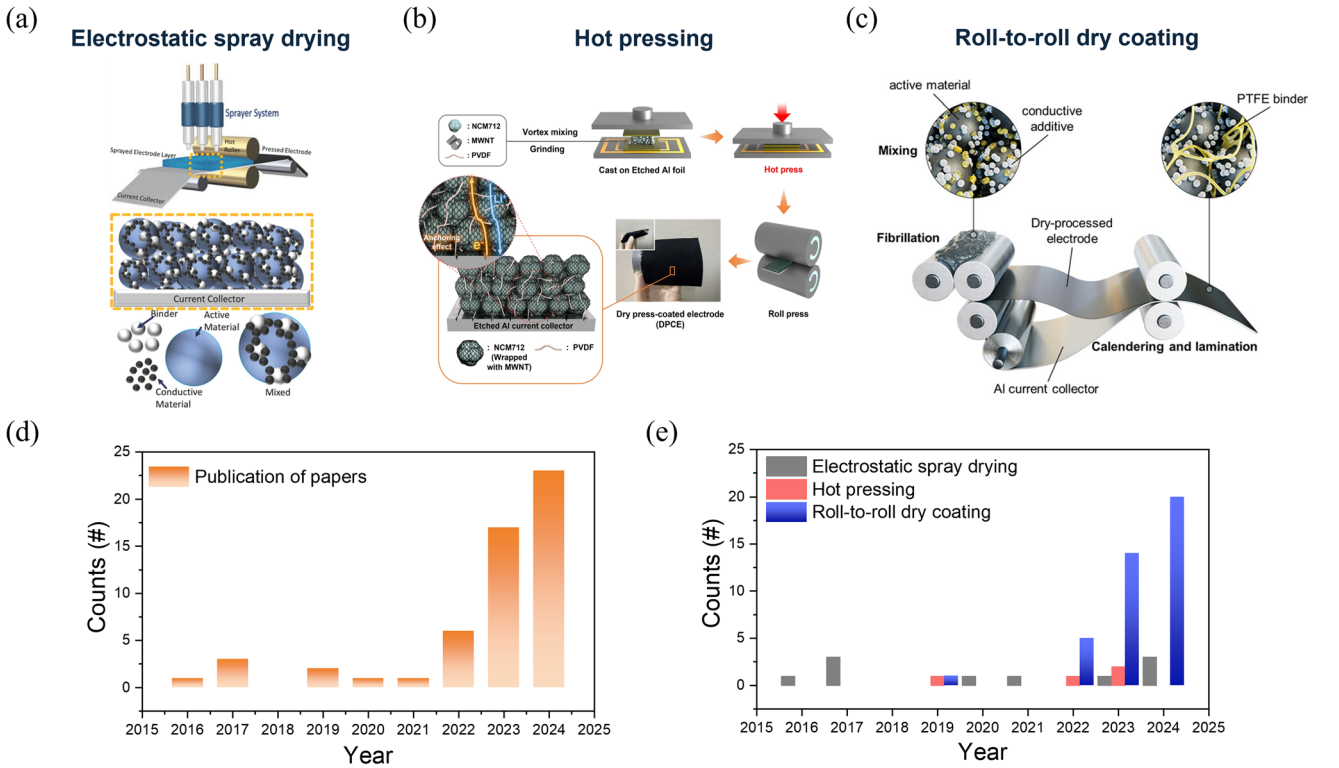

Fig. 3 Various dry coating processes: (a) schematic illustration of electrostatic spray drying, (b) hot pressing and (c) roll-to-roll dry coating processes. Reproduced with permission from ref. 50, 56 and 57. Copyright (2017) WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, copyright (2023) The Author(s) and copyright (2024) The Authors. Small Science published by Wiley-VCH GmbH. (d) The total number of papers on dry coating processes (electrostatic spray drying, hot pressing and roll-to-roll dry coating processes) and (e) papers on each dry coating process (2016–2024).

a dry mixture into dense and mechanically durable electrode films[58] (Fig. 3b). The process begins by preparing a uniform mixture of active materials, conductive additives, and binders, which is then placed into a mold or die. The mixture is exposed to elevated temperatures, typically ranging from 100 °C to 300 °C, and pressures exceeding several MPa. A key advantage of hot pressing is its ability to produce highly compact electrode structures with various thermoplastic binders and retain strong cohesion and mechanical stability with low content of binders.[59,60] Ryu *et al.* utilized hot pressing to fabricate ultra-thick electrodes incorporating multi-walled carbon nanotubes (MWNTs) as conductive additives and polyvinylidene fluoride (PVDF) as a binder.[56] Throughout the hot pressing, electrodes with an areal capacity of 17.6 mA h cm^{-2} and a thickness of 573 μm were developed, demonstrating its suitability for high-energy-density applications. Zhang *et al.* also developed a $LiFePO_4$ cathode using lithium-ion-conductive and biodegradable poly(propylene carbonate) (PPC) as a dry binder. PPC, with its low glass transition temperature (T_g) of 36.5 °C, allowed the fabrication of the dry electrode in a rubbery state, providing strong mechanical properties to the LFP cathode.[61] Additionally, PPC's high ionic conductivity reduced ionic resistance within the electrode compared to conventional dry electrodes, significantly improving its electrochemical performance. Despite its advantages, hot pressing encounters challenges when applied to the commercial-scale production of thick electrodes for medium-to-large secondary batteries. The need for high temperatures and pressures leads to significant energy consumption, increasing manufacturing costs. Additionally, the challenges in the continuous process of hot pressing limit its scalability, making it difficult to be applied in industrial-scale manufacturing.[62]

1.3.3 Roll-to-roll dry coating. The roll-to-roll dry coating method is recognized as one of the most scalable and efficient technologies for large-scale electrode fabrication[57] (Fig. 3c). This method involves preparing a dry mixture of active materials, conductive additives, and the polytetrafluoroethylene (PTFE) binder, which is then continuously fed into a roller or extrusion system, where the PTFE is fibrillized by external shear forces. The resulting electrode film is subsequently conveyed to downstream roller systems, enabling continuous production and making the roll-to-roll dry coating process particularly suitable for mass production, which helps to minimize manufacturing time and costs. The PTFE binder used in the roll-to-roll dry coating process forms a fibrous network within the electrode film, enhancing its mechanical strength and structural integrity.[63–66] Due to these advantages, the roll-to-roll dry coating process has become a critical point of research in both industry and academia. For example, Tao *et al.* developed dry cathodes and anodes with an areal capacity of 6 mA h cm^{-2}

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using the roll-to-roll dry coating process and evaluated their electrochemical properties against wet-processed full cells. The dry electrodes showed superior electrochemical performances.[67] Beyond LIB systems, research on the roll-to-roll dry coating process has expanded to next-generation batteries. Hippauf *et al.* reported NCM cathodes for ASSBs with an areal capacity of 6.5 mA h cm^{-2} using the PTFE binder in concentrations ranging from 0.1 to 1 wt%.[68] Similarly, Hu *et al.* developed dry cathodes for lithium–sulfur batteries with high sulfur loadings (2 mg cm^{-2}) using just 1 wt% PTFE binder.[38] The roll-to-roll dry coating process has also been implemented in industrial-scale production. For instance, Tesla has adopted this method for manufacturing dry-coated anode films, demonstrating its feasibility for large-scale production of advanced battery components.[69] However, challenges remain in further optimizing the roll-to-roll dry coating process. The fibrillization of the PTFE binder, which is crucial for improving mechanical and electrochemical properties, requires precise control. Furthermore, achieving the homogeneous distribution of active materials, conductive additives, and binders within the electrode remains an ongoing area of research.

Among the various dry coating techniques (electrostatic spray coating, hot pressing, and roll-to-roll dry coating), each offers distinct advantages and limitations, making them suitable for different applications. Electrostatic spray coating is suitable for precision and uniformity, making it well-suited for small-scale or research-focused applications. Hot pressing excels in producing dense and mechanically robust films, making it valuable for high-performance electrodes on a smaller scale. Roll-to-roll dry coating, in contrast, is the most promising technique for industrial-scale production due to its scalability, efficiency, and compatibility with high-throughput manufacturing. The roll-to-roll dry coating process has gained significant attention in recent years driven by the increasing demand for high-performance and cost-effective LIBs, demonstrated by the number of publications related to dry coating processes in lithium rechargeable batteries since the early 2010s (Fig. 3d and e). The combination of environmental sustainability and industrial scalability of the roll-to-roll dry coating process would position it as a cornerstone technology for next-generation battery production.

2. Roll-to-roll dry coating process

2.1 PTFE binder for the roll-to-roll dry coating process

The roll-to-roll dry coating process represents a novel technology in electrode manufacturing, particularly for producing thick electrodes essential for high-energy-density lithium rechargeable batteries. However, its successful application requires an understanding of the materials and processes involved, particularly the uniform dispersion of electrode components and the behavior of PTFE fibrillization. PTFE fibrillization plays a crucial role in determining the mechanical and electrochemical properties of dry electrodes, making it vital to explore how this material transforms into a fibrous structure under the shear forces applied during the process. PTFE has a unique molecular structure characterized by a linear carbon–carbon backbone with symmetrically arranged fluorine atoms that form a helical conformation around the backbone[70,71] (Fig. 4a). Unlike many other polymers, PTFE can deform under shear forces even at room temperature due to the change in the crystalline structure of PTFE, which undergoes phase transitions at relatively low temperatures.[72] At 19 °C, PTFE transitions from a low-temperature triclinic phase to an intermediate hexagonal phase, and above 30 °C, it further transforms into a pseudo-hexagonal phase[73] (Fig. 4c). The phase transition significantly affects the material's viscoelastic properties. As the temperature increases, the storage modulus, which represents the material's ability to store elastic energy under deformation, decreases. This change is attributed to the structural transformation of PTFE's crystalline regions. In the hexagonal phase, fluorine atoms are symmetrically aligned around the carbon backbone, and the polymer chains are oriented along the *c*-axis. This alignment reduces structural density and weakens intermolecular interactions, making PTFE more susceptible to shear deformation along the *c*-axis[71] (Fig. 4b). Accordingly, as the temperature increases, the elastic modulus of PTFE decreases, indicating a reduced ability to recover from deformation caused by shear forces[74] (Fig. 4d). The temperature- and pressure-induced transformation of PTFE's crystal structure, coupled with its fibrillization under shear forces at elevated temperatures, is directly applicable to the roll-to-roll dry coating process[75,76] (Fig. 4e). During the roll-to-roll dry coating process, PTFE undergoes deformation due to the shear forces exerted during the fibrillization process, which occurs above room temperature. A crucial point to note is that PTFE, a polymer with a low coefficient of friction, cannot easily undergo deformation independently. Therefore, an intermediary capable of transmitting external shear forces to the PTFE binder is required. In the electrode system, active materials or conductive additives, components of the electrode, can act as intermediaries to transfer external shear forces to the PTFE binder. These shear forces, transmitted through the interaction with active materials or conductive additives, cause the PTFE binder to deform and develop a fibrous network[77] (Fig. 4f and g). The PTFE fibrous network is critical for enhancing mechanical strength and ensuring the homogeneity of the electrode, contributing to its overall electrochemical properties. Accordingly, the control of the fibrillization behavior of the PTFE binder is a key factor in the PTFE binder-based roll-to-roll dry coating process. While equipment parameters influence fibrillization behavior, as mentioned earlier, the interaction between the PTFE binder and other electrode components, such as conductive additives or active materials, is considered particularly crucial. Stronger interactions between the PTFE binder and these intermediaries are expected to facilitate the effective transmission of external shear forces to the PTFE binder, thereby enhancing the fibrillization of the PTFE binder. Despite its importance, fundamental research on the particle interaction-driven PTFE fibrillization process remains limited. Therefore, future studies are needed to explore this fundamental aspect and deepen the understanding of PTFE fibrillization behavior in the roll-to-roll dry coating process.

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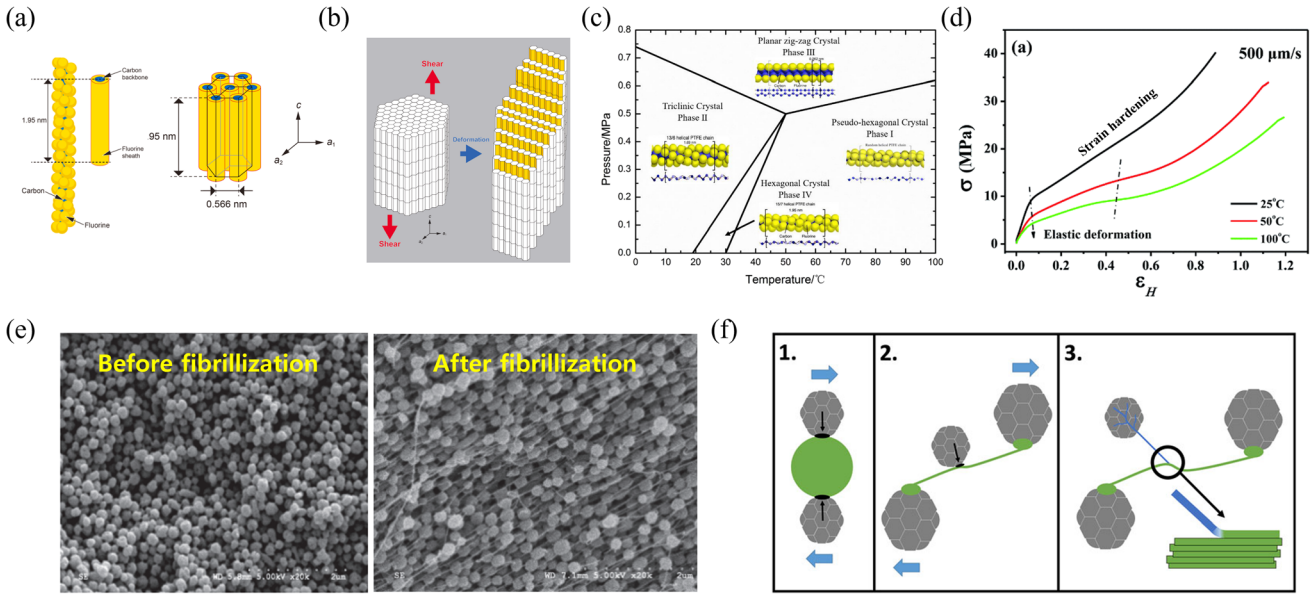

Fig. 4 PTFE binder for the roll-to-roll dry coating process: (a) The helical structure of an individual PTFE polymer chain in phase IV and (b) the hexagonal unit cell structure of phase IV PTFE. Reproduced with permission from ref. 71, copyright (2022) Elsevier Ltd. (c) Phase diagram of PTFE. Reproduced with permission from ref. 73, copyright (2020) Elsevier Ltd. (d) The true stress–strain curves of PTFE according to the temperature. Reproduced with permission from ref. 74, copyright (2021) RSC Publishing (e) SEM images of PTFE before and after the fibrillization process. Reproduced with permission from ref. 75, copyright (2017) Elsevier Ltd. (f) Schematic illustration and (g) SEM images of PTFE fibrillization in the roll-to-roll dry coating process. Reproduced with permission from ref. 77, copyright (2024) Matthews, Wheeler, Ramírez-González and Grant.

2.2 Main processes of the roll-to-roll dry coating process

The roll-to-roll dry coating process typically comprises three major processes: dry mixing, fibrillization, and free-standing film formation (Fig. 5). Each process plays a pivotal role in improving the mechanical and electrochemical properties of the electrode. The dry mixing process induces the uniform distribution of active materials, conductive additives, and binders. This uniformity is essential for superior electrochemical performance, as it is directly related to the ionic and electrical resistances of dry electrodes.[78–80] The fibrillization process aims to promote PTFE fibrillization by applying shear forces, transforming the PTFE particle into fibrous structures. The enhancement of PTFE fibrillization improves the mechanical and binding properties of dry electrodes, promotes pore formation, and enables uniform dispersion of conductive additives, collectively reducing ionic and electrical resistances.[81] The free-standing film formation process produces a mechanically robust free-standing electrode film, which is subsequently adhered to the current collector.

To achieve high energy density lithium rechargeable batteries, the development of thick dry electrodes is required. However, as the electrode thickness increases, ionic and electronic resistances (R_{ion} and R_{ct}) within the electrode also increase.[82] R_{ion} and R_{ct} represent the resistances of lithium-ion transport through the pore channel within the electrode and lithium-ion reactions at the interface of active materials[33] (Fig. 6a). As the electrode becomes thicker, the transport distance of lithium-ions from top to bottom regions increases, leading to an increase in R_{ion}. This, in turn, reduces the lithium-ion concentration in the bottom region of the electrode, thereby increasing R_{ct} (Fig. 6b and d). As a result, the overall increase of ionic resistance within the dry thick electrode led to degradation of rate capability (Fig. 6c). To alleviate these issues, it is necessary to minimize the lithium-ion transport distance within the dry electrode by improving the homogeneity of the electrode microstructure. Through the engineering of the three processes (dry mixing, fibrillization, and free-standing film formation), the homogeneity of the electrode microstructure could be achieved with proper pore size and distribution, improved PTFE fibrillization and crack-free cathode active materials, thereby improving the overall electrochemical properties of the dry electrode (Fig. 6e).

2.2.1 Dry mixing process. Dry thick electrodes inherently have long pathways for lithium-ion and electron transport, which increases ionic and electrical resistance. To overcome these challenges, the dry mixing process should be designed to ensure a dry mixture in which the active materials, conductive additive and PTFE binder are uniformly dispersed. Accordingly, researchers have conducted extensive studies to optimize the dry mixing process. Tao *et al.* investigated the effects of blade speed and mixing time on the distribution of $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (NMC), carbon black, and PTFE[84] (Fig. 7a). Similarly, Shin *et al.* addressed the aggregation problem of carbon nanotubes (CNTs) during the dry mixing process, inducing the degradation of electrochemical properties of CNT applied dry electrodes[85] (Fig. 7b). CNTs tend to clump together due to their high surface energy, leading to poor distribution within the dry electrode. To overcome this issue, CNT-coated $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA) was introduced in the roll-to-roll dry coating process, where the wet coating method was applied to coat CNTs onto the surface of NCA particles, improving their

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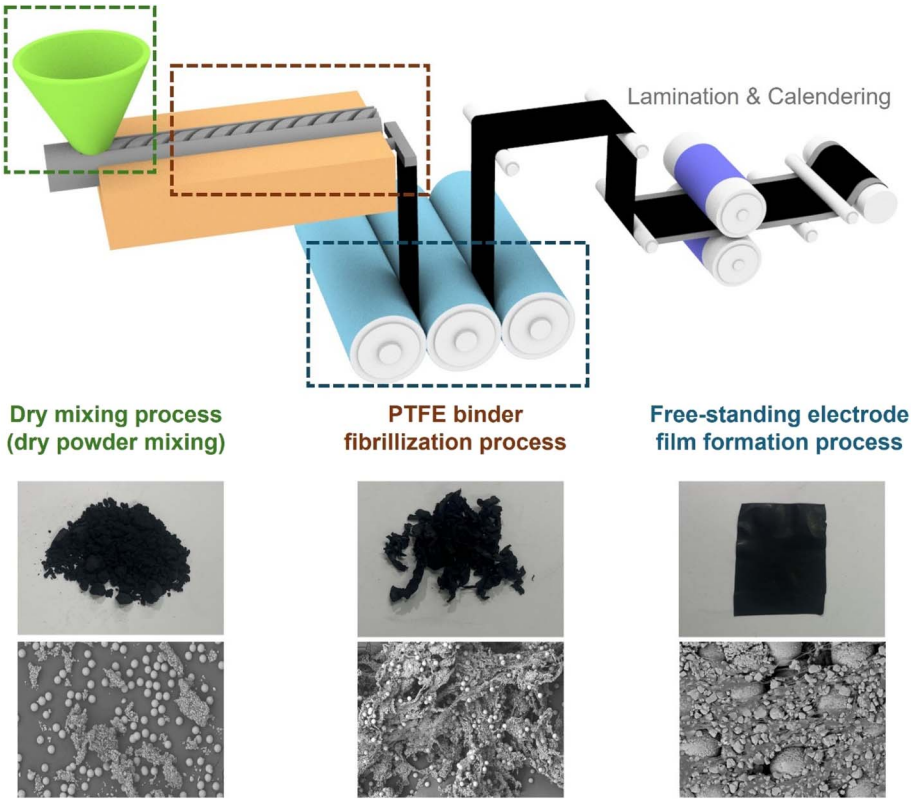

Fig. 5 Main processes of the roll-to-roll dry coating process.

dispersion and reducing the formation of aggregates. Throughout the modification of the surface of NCA, the electrical conductivity and mechanical properties of dry electrodes were improved, associated with uniform distribution of CNTs within the dry electrode. Controlling the PTFE distribution within the dry electrode has also been explored. Choi *et al.* took a different approach by modifying the surfaces of CB and PTFE with ionic surfactants[86] (Fig. 7c). This modification increased electrostatic interactions between the CB and PTFE, resulting in a more uniform distribution of CB and PTFE. Lee *et al.* used cryogenic freezer milling to reduce the PTFE particle size, which allowed for uniform dispersion of PTFE within the electrode[87] (Fig. 7d). This approach not only improved the uniformity of the binder but also reduced ionic resistance within the electrode. These studies above highlight the importance of addressing dispersion issues of active materials, conductive additives and the PTFE binder in the dry mixing process to provide the reliability of thick dry electrodes with superior electrochemical properties.

2.2.2 PTFE fibrillization process. The fibrous network formed by PTFE fibrillization enhances the mechanical strength of the electrode film and uniformity of pore distribution within the dry electrode, closely related to the tortuosity, resulting in robust durability of the electrode film during cell manufacturing processes such as winding and lamination, as well as a homogeneous microstructure of the dry electrode with improved ionic resistance. Various approaches have been explored to enhance PTFE fibrillization. Oh *et al.* investigated the behavior of PTFE fibrillization as a function of the kneading time by measuring the applied torque during the process[88] (Fig. 8a). They found that increases in the kneading time induced excessive fibrillization of PTFE, leading to the thin PTFE fiber and resulting in brittle characteristics of the dry electrode film. Similarly, Kim *et al.* observed the temperature dependence for the PTFE fibrillization behavior and suggested the temperature region that can retain the morphology of the PTFE fibrous network within the dry electrode[83] (Fig. 8b). They reported that while higher temperatures facilitated fibrillization, excessive heating could cause the fibrous structures to aggregate, affecting the morphology of the PTFE film and reducing the tensile strength of the dry electrode film. Research on engineering the PTFE fibrillization process is not limited to cathode materials such as high nickel cathode materials. Studies on lithium–iron phosphate (LFP), a cathode material with low ionic conductivity, have shown that its small particle size increases the surface area and shear forces exerted on PTFE, leading to excessive fibrillization. This excessive fibrillization makes the electrode film brittle, lowering its mechanical properties. Wiegmann *et al.* addressed this issue by adjusting the screw configuration in extrusion equipment to control the number of fibrillization zones, thereby suppressing the excessive fibrillization and improving the mechanical properties of the electrode film[89] (Fig. 8c). Beyond LIBs, fibrillization of PTFE has been investigated for the fabrication of solid-state electrolyte membranes. Lee *et al.* studied the effects of PTFE binder content, temperature and direction of the fibrillization process

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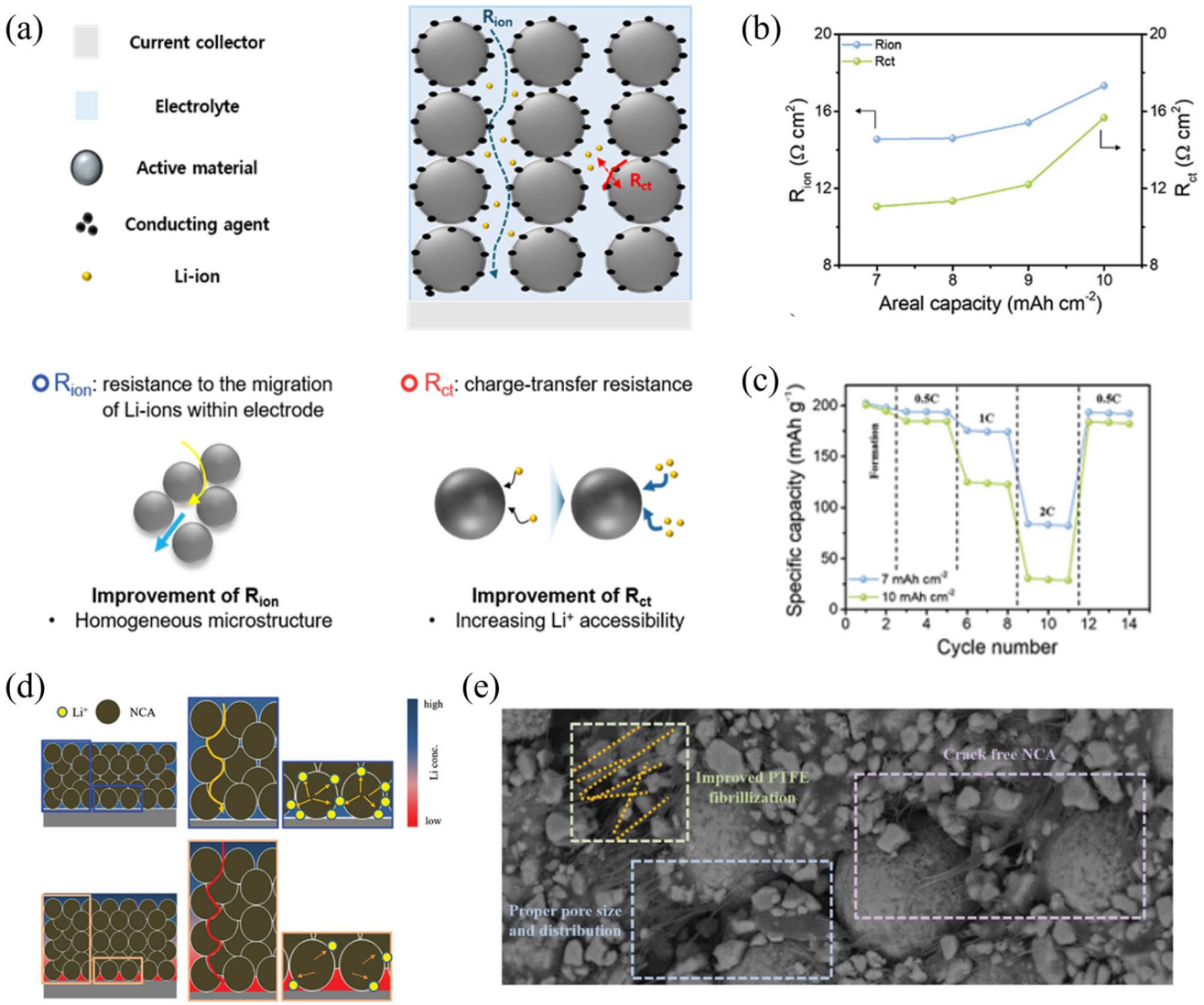

Fig. 6 Relationship between ionic resistance and electrochemical performance of dry thick electrodes: (a) schematic illustration of ionic resistance (R_{ion} and R_{ct}). (b) Ionic resistance and (c) C-rate capability of dry electrodes depending on the areal capacity. (d) The schematic illustration of the issue of ultra-high thick electrodes and (e) requirement of main characteristics for dry electrodes with homogeneous microstructures. Reproduced with permission from ref. 83, copyright (2024) Wiley-VCH GmbH.

on the fibrous network morphology of PTFE in solid-state electrolyte membranes[90] (Fig. 9a). Based on how the morphology of PTFE within solid-state electrolyte membranes influenced the electrochemical and mechanical stability of the ASSB, a durable and electrochemically stable bi-layer electrolyte membrane was developed. Furthermore, Yoon *et al.* conducted a study to examine the fibrillization behavior of PTFE as a function of molecular weight[91] (Fig. 9b). They utilized XRD analysis to quantify fibrillization under varying shear forces and temperatures for PTFE with different molecular weights. Despite the significant advancements in understanding the PTFE fibrillization process and its influence on the electrochemical performance of dry electrodes, further fundamental research is necessary to fully understand the behavior of PTFE fibrillization during the roll-to-roll dry coating process. Such studies are essential to address remaining challenges and enable the successful commercialization of the roll-to-roll dry coating process, particularly in ensuring consistent fibrillization behavior and its correlation with the mechanical and electrochemical properties of dry electrodes.

2.2.3 Free-standing electrode film formation process. The free-standing electrode film was formed and laminated onto the current collector through the roll pressure to achieve the desired electrode thickness and density during the rolling and lamination process. During the rolling and lamination process, proper control of the rolling parameters is essential to prevent crack of active materials, ensure strong cohesion between electrode components, and improve adhesion between the electrode film and the current collector.[83]

The roll-to-roll dry coating process shows great potential for producing high-energy-density and cost-effective electrodes. However, its success depends on optimizing each process.

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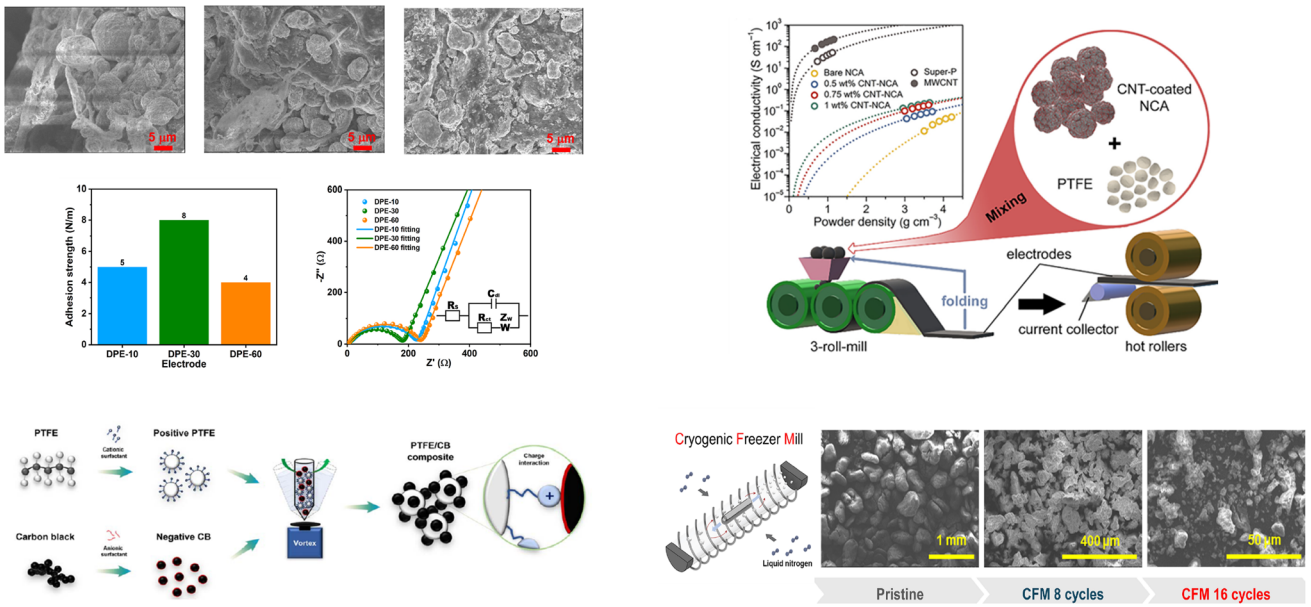

Fig. 7 Engineering of the dry mixing process: (a) control of shear force by the dry mixing time. Reproduced with permission from ref. 84. Copyright (2023) Springer. (b) The schematic illustration of improvement of CNT dispersion in the dry electrode by wet coating of CNTs on NCA. Reproduced with permission from ref. 85. Copyright (2022) John Wiley & Sons Ltd. (c) The schematic illustration of surface modification of PTFE and carbon black for improvement of PTFE and CB dispersion. Reproduced with permission from ref. 86. Copyright (2023) Springer. (d) Controlling the size of PTFE to improve the dispersion of the PTFE binder. Reproduced with permission from ref. 87. Copyright (2023) Springer.

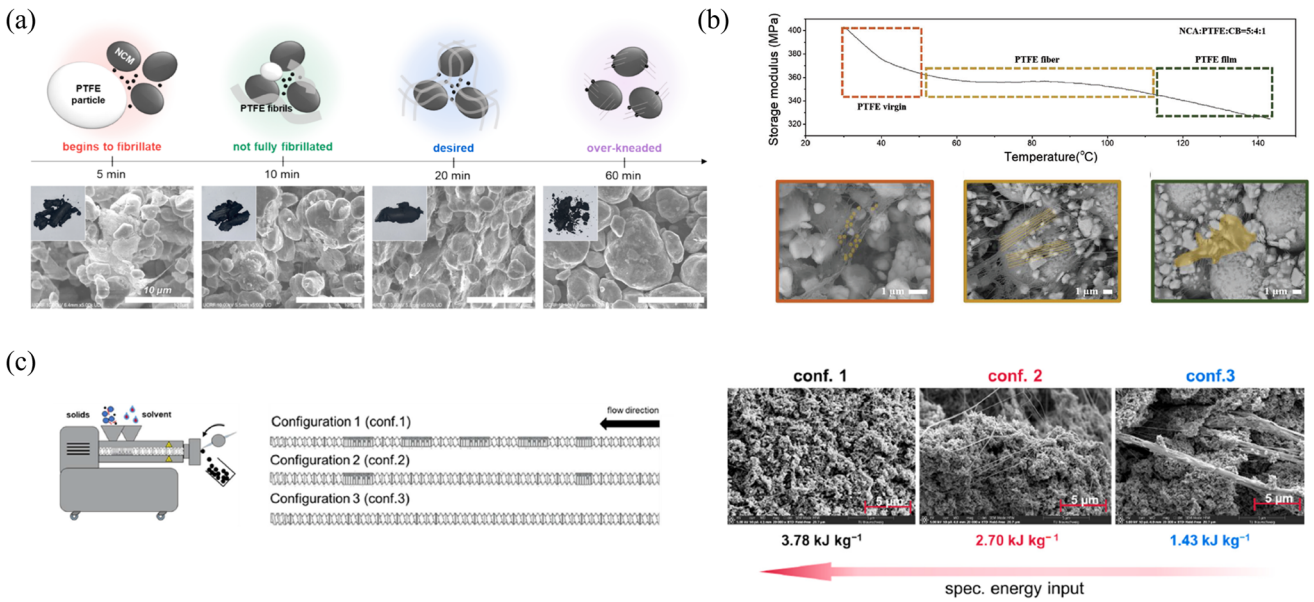

Fig. 8 Engineering of the PTFE fibrillization process (LIBs). Investigation of PTFE fibrillization behavior according to the (a) fibrillization process time, (b) fibrillization process temperature and (c) the number of kneading configurations. Reproduced with permission from ref. 83, 88 and 89. Copyright (2024) The Authors. Published by Elsevier B.V., copyright (2024) Wiley-VCH GmbH and copyright (2023) The Authors. Licensee MDPI, Basel, Switzerland.

While significant progress has been made in understanding and improving these processes, challenges remain. For example, PTFE fibrillization behavior, influenced by particle properties such as size, shape and surface characteristics, as well as processing conditions, requires further research for a comprehensive understanding. Accordingly, establishing a database of PTFE fibrillization behavior under various conditions could support the improvement of the roll-to-roll dry coating process, accelerating the commercialization of thick dry electrodes and their integration into next-generation battery technologies.

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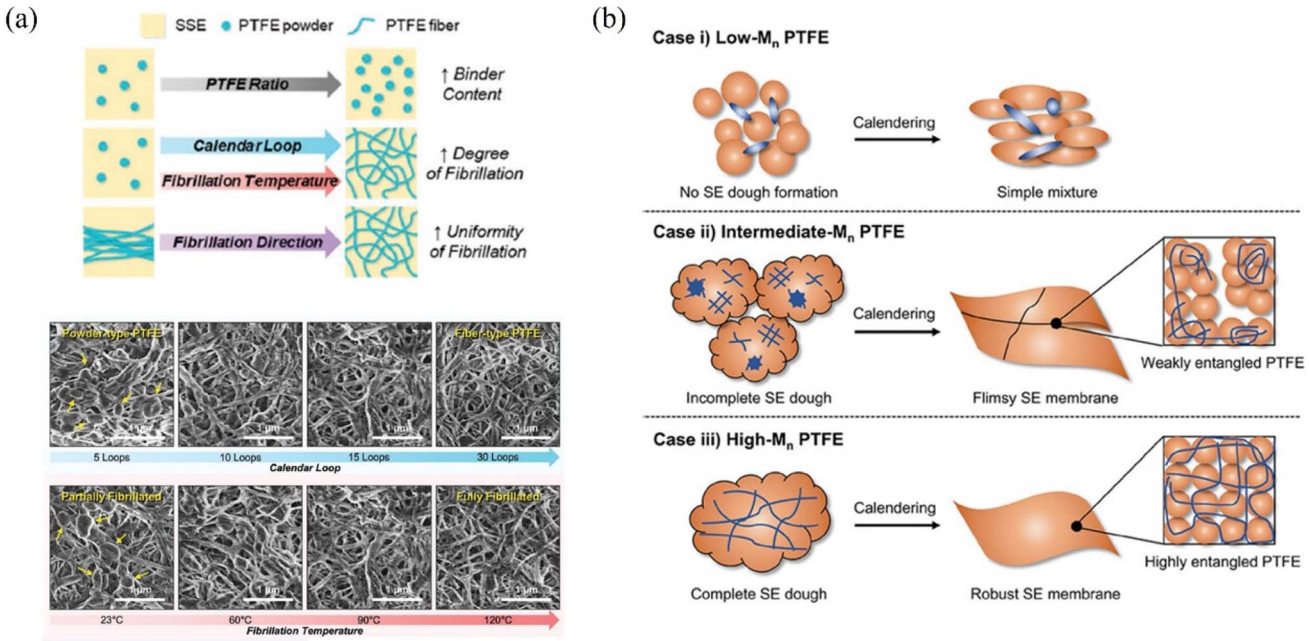

Fig. 9 Engineering of the PTFE fibrillization process (ASSBs). Investigation of PTFE fibrillization behavior in the solid electrolyte film according to (a) various process parameters and (b) the molecular weight of PTFE. Reproduced with permission from ref. 90 and 91. Copyright (2023) Wiley-VCH GmbH and copyright (2024) Wiley-VCH GmbH.

3. Applications for the roll-to-roll dry coating process

3.1 Lithium-ion batteries

The roll-to-roll dry coating process has been introduced to improve the performance of LIBs, yet it still faces several challenges, including issues related to ionic and electrical resistance, chemical stability, and mechanical stability. This section discusses the major issues and the solutions to address them.

3.1.1 Improvement of lithium-ion & electron resistance and chemical stability of dry electrodes. Dry electrodes face significant technical challenges in terms of ionic and electrical resistance and chemical stability. In particular, the roll-to-roll dry coating process faces significant dispersion issues due to the absence of solvents, leading to non-uniform dispersion of conductive additives.[84] This issue is particularly pronounced with conventional conductive additives such as CB and CNTs.[92–95] When CB or CNTs are introduced into a cathode composite through physical mixing without solvents, the aggregation of conductive additives occurs, lowering electrical conductivity within the dry electrode and hindering the reaction of lithium ions with active materials. To address the issue of aggregation of conductive additives, studies have focused on the modification of their surface characteristics. For example, Kim *et al.* proposed CNTs treated with ozone (O_3) surface treatment as a conductive additive for the roll-to-roll dry coating process to enhance the dispersion of CNTs within the dry electrode[96] (Fig. 10a). The ozone treated CNTs have high affinity with NCM811, inducing the uniform dispersion of CNTs on the surface of NCM 811 and resulting in superior electrochemical properties. The morphology of active materials can also affect the ionic resistance of dry electrodes, associated with the different microstructures according to the morphology of active materials. Hwang *et al.* reported that the arrangement of graphite within the dry electrode, whether in-plane (parallel to the *yz*-plane) or through-plane (perpendicular to the *yz*-plane), depends largely on morphology of graphite, resulting in variations in porosity and tortuosity[97] (Fig. 10b). Dry electrodes with fine spherical graphite (FSG) exhibited a higher degree of alignment along their longest principal axes parallel to the current collector compared to dry electrodes with small natural graphite (SNG). Moreover, dry electrodes with SNG exhibited a more pronounced pore gradient, resulting in lower tortuosity compared to dry electrodes with FSG and improved fast-charging capability. Furthermore, the different behaviors of electrical resistance within the dry electrode were reported depending on whether the active materials are single- or poly-crystalline. Tao *et al.* showed that dry electrodes with single-crystal active materials (SC DPE) exhibited more uniform distribution of CB compared to dry electrodes with poly-crystalline active materials (PC DPE), resulting in improved electrical conductivity[98] (Fig. 10c). It is attributed to the different conductive pathways where the conductive pathway in PC DPE was anisotropic along the electrode thickness direction, whereas the conductive pathways are more isotropic in SC DPE. In addition to studies on the ionic and electrical resistance of dry electrodes, studies have also been reported on the electrochemical stability of the PTFE binder. Tao *et al.* reported the side reaction of PTFE within the dry cathode during cycling by conducting cycle tests of dry electrodes with electrolytes containing either $LiPF_6$ or $LiClO_4$ (ref. 99) (Fig. 10d). By eliminating the fluorine source ($LiPF_6$) in electrolyte and applying $LiClO_4$ as a lithium salt, the decomposition behavior of PTFE was

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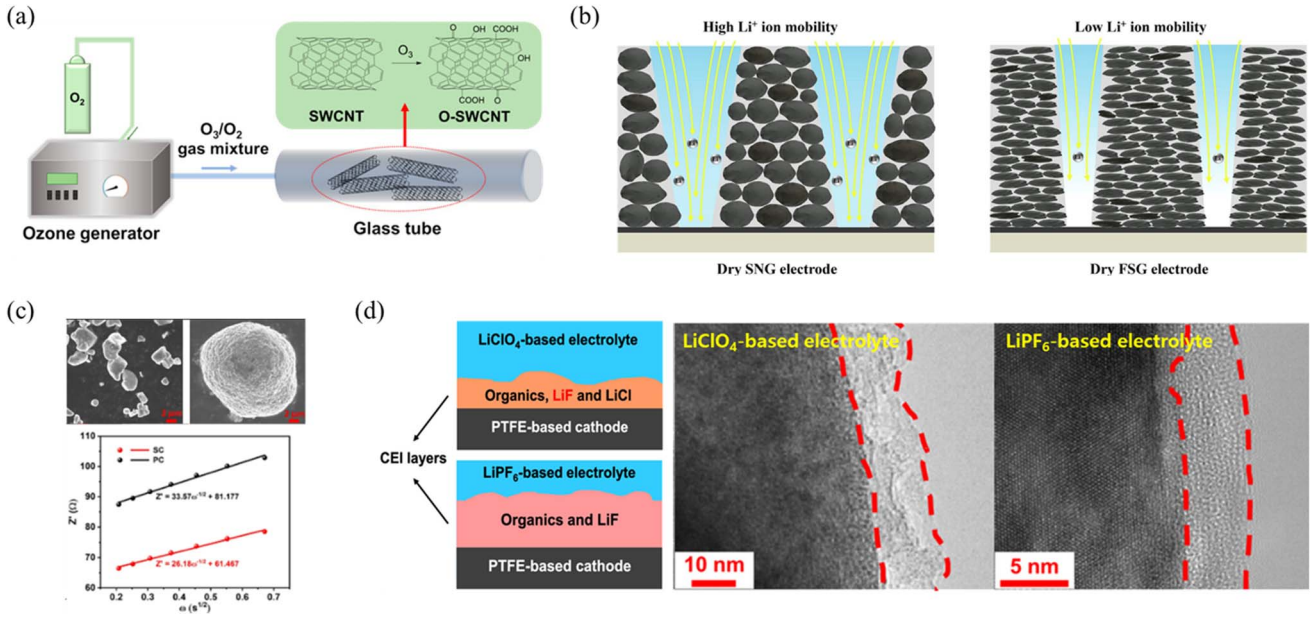

Fig. 10 Improvement of lithium ion/electron resistance and chemical stability of dry electrodes (LIBs): (a) ozonation of the SWCNT and its application to the dry cathode process. Reproduced with permission from ref. 96. Copyright (2023) American Chemical Society. (b) Pore structure of dry-processed SNG and FSG graphite electrodes. Reproduced with permission from ref. 97. Copyright (2024) The Author(s). Published by Elsevier B.V. (c) SEM images and linear curve fittings of Z' *versus* $\omega^{-1/2}$ in the low-frequency region of SC and PC NMC. Reproduced with permission from ref. 98. Copyright (2024) Elsevier B.V. (d) Schematic illustration of CEI layers and TEM images of the $LiPF_6$-cycled and $LiClO_4$-cycled DPEs. Reproduced with permission from ref. 99. Copyright (2023) American Chemical Society.

investigated, where the detection of LiF in cells with $LiClO_4$ confirmed that PTFE undergoes side reactions during cycling.

3.1.2 Improvement of mechanical properties of dry electrodes. Dry electrodes with insufficient fibrillization of PTFE exhibit poor mechanical properties and structural stability. To address this issue, extensive research has been conducted from both material and process perspectives to enhance the fibrillization of PTFE. However, significant limitations remain and accordingly, efforts have been made to improve the mechanical properties of dry electrodes by introducing additives beyond conventional materials. For example, He *et al.* developed a high-performance dry electrode (DP-F electrode) incorporating flour as a partial binder[100] (Fig. 11a). The addition of 1 wt% flour (DP-1% F electrode) results in superior mechanical properties, excellent rate performance and high cycling stability. The DP-1% F electrode demonstrated a tensile strength of 1.12 MPa (high mechanical strength) and a minimum elastic modulus of 8.04 MPa (high flexibility), attributed to the cross-linking between proteins and starches in the flour. In the context of structural stability for dry electrodes, the calendering process poses challenges due to the high pressure required to densify thick, free-standing electrode films and increase electrode density. This high pressure often causes cracks in polycrystalline active materials, compromising the electrode's mechanical properties. To address this issue, Embleton *et al.* introduced a solvent-assisted binder dry coating process, which mitigates microstructural problems arising from such cracks[101] (Fig. 11b). Using a low-boiling-point solvent such as ethanol (<3 wt%), this approach enhanced electrode processability and PTFE fibrillization, with the solvent removed through elevated roll press temperatures. In this way, small amounts of solvents (additives) have been utilized to enhance the mechanical properties of dry electrodes. However, to fully achieve the advantages of the dry coating process, such as low manufacturing cost and scalability, it is ultimately necessary to improve the mechanical properties and structural stability without relying on additives.

3.2 Next-generation batteries

Next-generation batteries, such as ASSBs and Li–S batteries, have benefited significantly from the roll-to-roll dry coating process utilizing the PTFE binder, which has brought transformative advancements by addressing challenges in stability, energy density, and scalability. In particular, traditional ASSBs with wet coating processes suffer from undesirable side reactions between solvents used in wet coating processes and solid electrolytes. The roll-to-roll dry coating process eliminates the need for solvents, thereby preventing side reactions, while forming uniform and robust electrode and electrolyte films that improve structural and electrochemical stability.[102–104] Additionally, binder migration induced by solvent evaporation during the wet coating process often results in electrode cracking for thick electrodes designed to achieve high energy density. The roll-to-roll dry coating process effectively addresses this issue, establishing itself as a key technology for the development of high-capacity next-generation batteries.

3.2.1 All-solid-state batteries (ASSBs). All-solid-state batteries (ASSBs) have been extensively studied as next-

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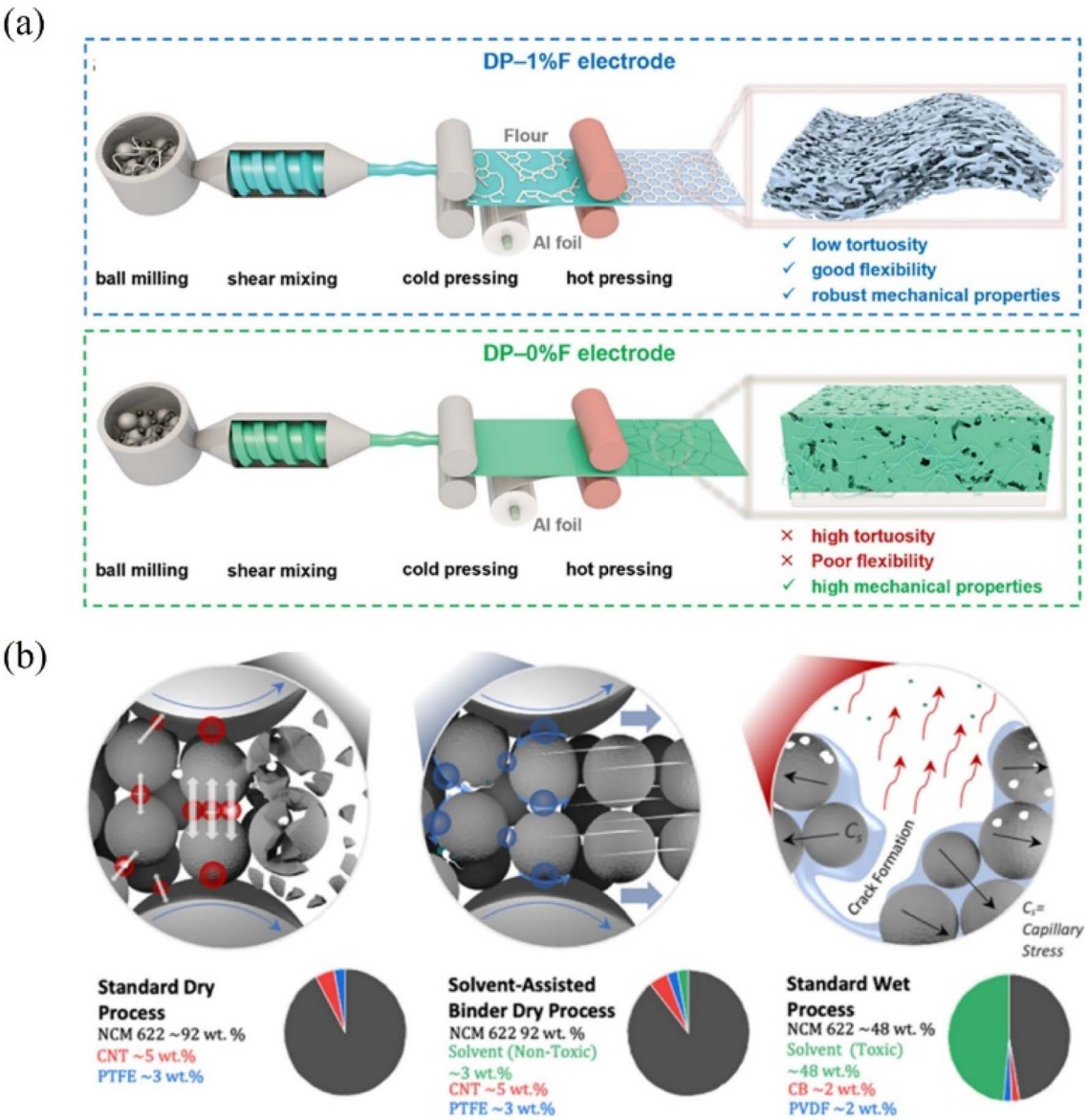

Fig. 11 Improvement of mechanical properties of dry electrodes (LIBs): (a) fabrication procedures of DP-1% F and DP-0% F electrodes. Reproduced with permission from ref. 100. Copyright (2024) Wiley-VCH GmbH. (b) Fracturing of active materials, effects of the solvent-assisted-binder (SaB) dry process, and capillary stresses in relation to mixture composition during dry processing. Reproduced with permission from ref. 101. Copyright (2023) The Authors. Published by Elsevier B.V.

generation battery technology due to their high energy density and exceptional safety. Sulfide-based solid electrolytes, which offer good malleability and high ionic conductivity at room temperature, are commonly employed.[105] However, sulfide-based solid electrolytes present challenges in electrode fabrication due to their narrow electrochemical window and poor chemical compatibility with polar solvents.[106] In particular, their poor chemical compatibility with polar solvents induces side reactions between the solid electrolyte and the solvent during electrode fabrication, limiting the choice of solvents in wet processing. To address these issues, a roll-to-roll dry coating process, which eliminates the use of solvents, was adopted. Nevertheless, the dry coating process also faces challenges, such as contact issues between the solid electrolyte and active materials, inhomogeneous electrode microstructures, and low ionic conductivity of the PTFE binder. Lee *et al.* systematically investigated the effect of shear force on coverage, the percentage of electrolyte-covered surface area of the active materials, by comparing wet and dry electrodes for ASSBs[107] (Fig. 12a). Since the slowest lithium-ion movement in ASSBs is solid-state diffusion between active materials, coverage plays an important role in electrochemical performance such as rate performance. Dry electrodes exhibited significantly higher coverage compared to wet-processed electrodes, attributed to the deformation of the ductile sulfide solid electrolytes by shear forces, resulting in high ionic conductivity. However, the PTFE binder faces another challenge related to its intrinsic low ionic conductivity. To overcome the limitation of the PTFE binder

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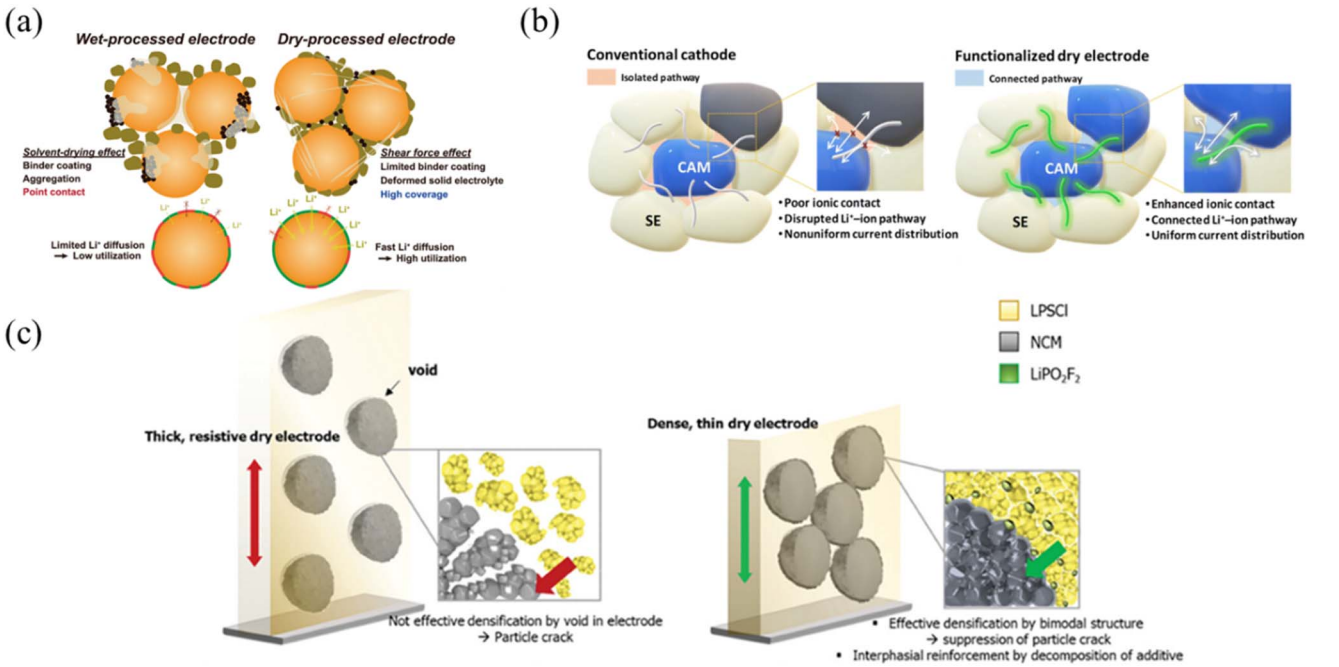

Fig. 12 Next-generation batteries (ASSBs): (a) disparity in electrode coverage and Li^+ diffusion between the wet process and dry process. Reproduced with permission from ref. 107. Copyright (2024) The Author(s). (b) The conventional cathode with an ionically insulating PTFE binder and SIL-infiltrated EC. Reproduced with permission from ref. 108. Copyright (2024) RSC Publishing. (c) The role of $LiPO_2F_2$ as an additive in dry electrodes for ASSBs. Reproduced with permission from ref. 109. Copyright (2023) Wiley-VCH GmbH.

with low ionic conductivity, Lee *et al.* developed a functional dry binder system by introducing the solvate ionic liquid (SIL)-infiltrated ethyl cellulose (EC) into the roll-to-roll dry coating system, which enabled the fabrication of functional dry electrodes (FDEs)[108] (Fig. 12b). By incorporating the SIL-infiltrated EC into the dry electrode, enhanced charge-transfer kinetics and effective contact area among particles were achieved in ASSBs. Despite these efforts to enhance the ionic conductivity of ASSBs, residual voids within the dry electrode remain a challenge, preventing uniform pressure distribution during the calendering process. To suppress the residual voids within the dry electrode, a practically applicable dual-functional electrode additive was introduced. Kim *et al.* introduced dry electrodes with dense bimodal structures by using argyrodite solid electrolytes and lithium difluorophosphate ($LiPO_2F_2$) additives with different particle sizes[109] (Fig. 12c). $LiPO_2F_2$ fills the interparticle voids within the dry electrode, ensuring uniform pressure distribution during the calendering process and reducing residual voids within the dry electrode. Accordingly, the addition of $LiPO_2F_2$ enables the fabrication of dry electrodes with high ionic and electronic conductivity. Despite these advantages of the roll-to-roll dry coating process in ASSBs, further research on functional binders, electrode additives, and manufacturing techniques will be essential for optimizing dry electrode fabrication and accelerating the commercialization of high-performance ASSBs.

3.2.2 Lithium–sulfur (Li–S) batteries. Lithium–sulfur (Li–S) batteries have garnered significant attention as promising next-generation batteries due to their high theoretical energy density (2600 W h kg^{-1}) and high specific capacity (1672 mA h g^{-1}). Additionally, sulfur (S) is earth-abundant, nontoxic, and low-cost, further enhancing its potential as a next-generation battery.[110] To achieve high energy density in Li–S batteries, several factors must be considered, such as the electrolyte-to-sulfur ratio (E/S), negative-to-positive capacity ratio (N/P), and sulfur loading.[111] Among these factors, high sulfur loading has recently attracted attention, as seen in LIBs and ASSBs. However, achieving high energy density through high sulfur loading has critical challenges such as cracks and delamination of thick electrodes, associated with solvent evaporation during the wet coating process, resulting in sluggish reaction kinetics.[112] To address these issues, Sul *et al.* developed high-sulfur-loading electrodes (>7 mg cm^{-2}) through the roll-to-roll dry coating process[113] (Fig. 13a). Moreover, the Li–S batteries were integrated with ASSBs for high energy density lithium rechargeable batteries. Shen *et al.* reported that PTFE fibrillization of the PTFE binder facilitates the interweaving of Li_6PS_5Cl particles and FeS_2 cathode active materials, resulting in the formation of flexible cathodes for all-solid-state-lithium–sulfur-batteries[114] (Fig. 13b). Hu *et al.* also reported dry electrode technology for scalable and flexible high-energy sulfur cathodes in all-solid-state lithium–sulfur batteries[115] (Fig. 13c). The fibrous network of the PTFE binder effectively enables the formation of lithium-ion and electron conductive networks within the dry sulfur cathode, leading to the improved electrochemical performance. The direct current (DC) and alternating current (AC) tests of electrodes demonstrated the improved ionic and electrical conductivity of dry sulfur cathodes in ASSBs

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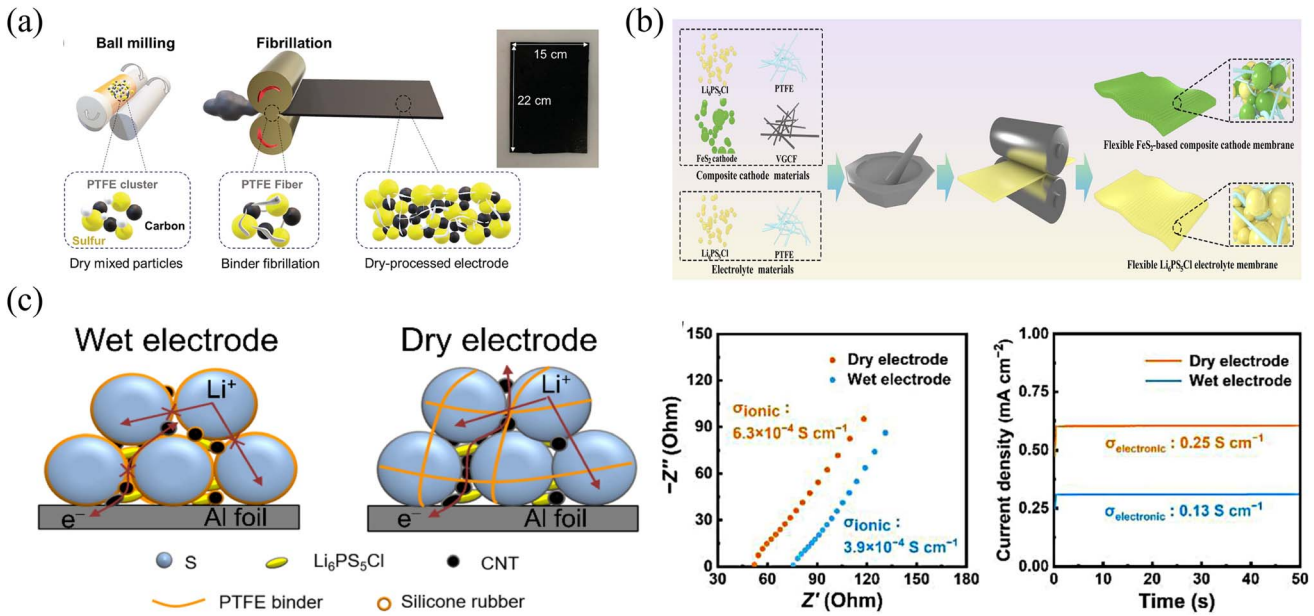

Fig. 13 Next-generation batteries (Li–S batteries): (a) dry electrode fabrication procedure for a sulfur cathode with high mass loading. Reproduced with permission from ref. 113. Copyright (2024) Wiley-VCH GmbH. (b) Schematic illustration of the preparation of the LPSCl electrolyte membrane and FeS_2 cathode by the roll-to-roll dry coating process. Reproduced with permission from ref. 114. Copyright (2024) Elsevier Inc. (c) Schematic illustration and measurement of ionic/electrical conductivity of wet- and dry-processed sulfur cathodes. Reproduced with permission from ref. 115. Copyright (2022) Science Press. Published by Elsevier B.V.

due to the fibrous network of the PTFE binder. These studies demonstrate the potential of the roll-to-roll dry coating process in advancing the scalability and performance of Li–S batteries, paving the way for their practical implementation in next-generation energy storage systems.

4. Summary and outlook

4.1 Summary

Dry coating processes represent a paradigm shift for high energy density lithium rechargeable batteries with thick electrodes, offering a sustainable alternative to conventional wet coating methods. Among these, the roll-to-roll dry coating process emerges as the most promising technique for industrial-scale production, thanks to its scalability and production efficiency, which aligns with the growing demand for high-energy-density and cost-effective batteries. Despite its potential, significant challenges remain on the path to commercialization. Addressing these challenges necessitates advancements in the main processes of the roll-to-roll dry coating process, including the dry mixing process, PTFE binder fibrillization process, and free-standing electrode film formation process, and materials such as the PTFE binder, conductive additives, active materials, and additives to achieve the low resistance dry electrode with a homogeneous microstructure. The process and material advancements not only promise to overcome current barriers but also extend the applicability of the roll-to-roll dry coating process to next-generation batteries such as all-solid-state batteries (ASSBs) and lithium–sulfur (Li–S) batteries. This provides a versatile foundation for further research and development, paving the way for the commercialization of advanced battery systems. Ultimately, the roll-to-roll dry coating process is poised to become a cornerstone technology for fabricating high energy density lithium rechargeable batteries with cost efficiency and environmental sustainability.

4.2 Future work

As discussed, the roll-to-roll dry coating process utilizing the PTFE binder is advantageous for the commercialization of thick electrodes. While significant research has been conducted to achieve the commercialization of the roll-to-roll dry coating process, there remain areas requiring further study.[116–118] First, the methodology for the analysis of the degree of PTFE fibrillization is required. Until now, several studies have analyzed the degree of PTFE fibrillization in dry electrodes by measuring the storage and elastic modulus of electrode films through DMA (Dynamic Mechanical Analysis) and crystallinity of PTFE within dry electrode films through DSC (Differential Scanning Calorimetry) or evaluating the mechanical properties using a UTM (Universal Testing Machine)[71,83,119–123] (Fig. 14a and b). XRD (X-ray diffraction) measurement was also employed to analyze the degree of deformation of PTFE according to the intensity of specific peaks affected by deformation of PTFE[71] (Fig. 14c). These methods allow for indirect comparison and evaluation of PTFE fibrillation within electrodes. However, for underlying understanding, more precise measurements for the analysis of the degree of PTFE fibrillization are necessary. Therefore, developing a methodology to quantitatively assess the degree of PTFE fibrillation in electrode films is an important area for

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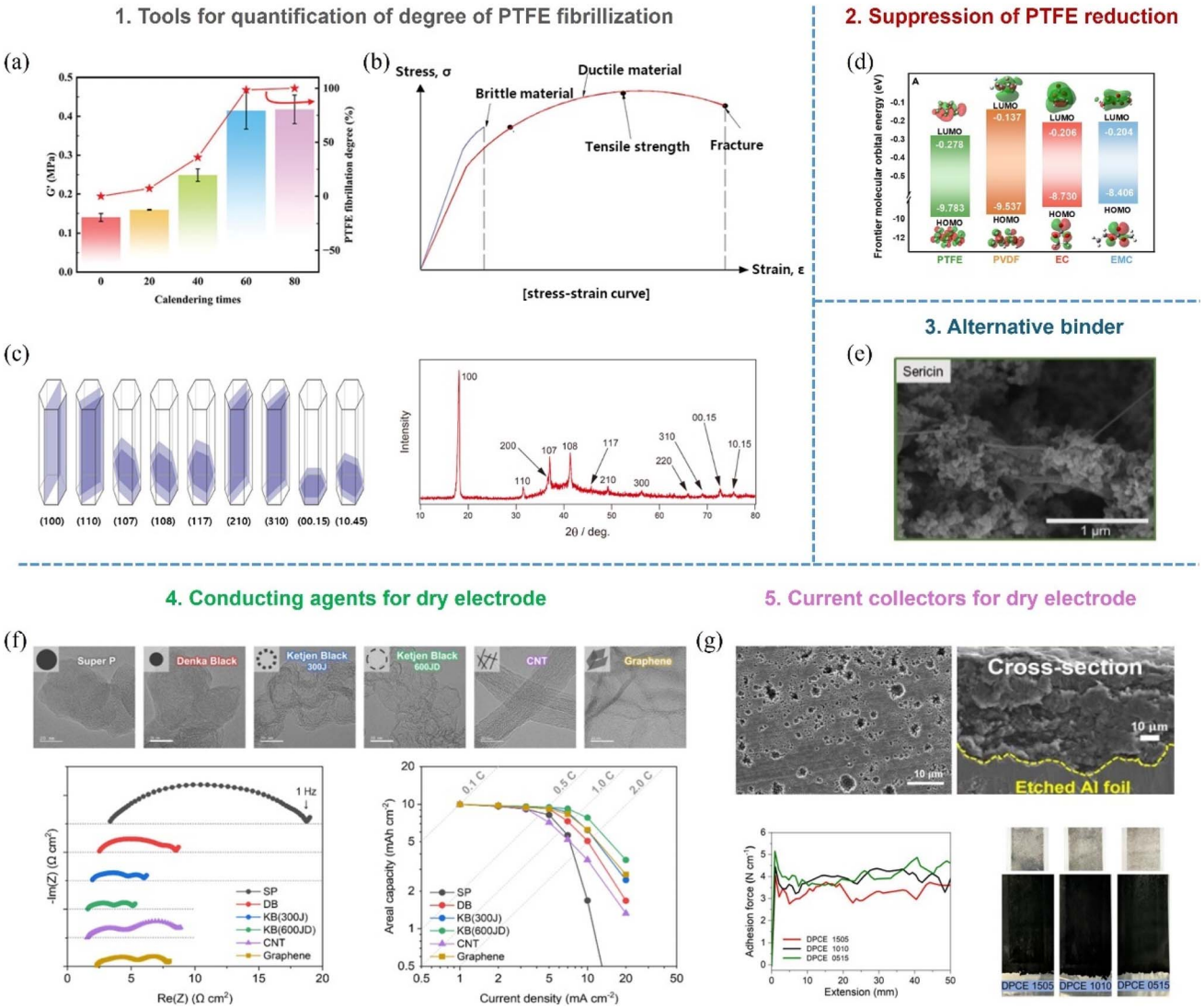

Fig. 14 Future work on the roll-to-roll dry coating process: (a) storage modulus from DMA measurement. Reproduced with permission from ref. 121. Copyright (2024) Wiley-VCH GmbH, (b) stress–strain curve from UTM measurement and (c) (107) and (108) planes from XRD measurement of the electrode film. Reproduced with permission from ref. 71. Copyright (2022) Elsevier Ltd. (d) The LUMO and HOMO energies of polymers and electrolyte solvent. Reproduced with permission from ref. 124. Copyright (2024) Elsevier Inc. (e) Development of sustainable binder (sericin) for replacing the PTFE binder as a dry coating process binder. Reproduced with permission from ref. 129. Copyright (2022) The Authors. ChemSusChem published by Wiley-VCH GmbH. (f) Application of conductive additives with various dimensions and specific surface areas for the roll-to-roll dry coating process. Reproduced with permission from ref. 130. Copyright (2025) RSC Publishing. (g) Application of etched Al current collectors to improve the adhesion between the dry-processed cathode and current collectors. Reproduced with permission from ref. 131. Copyright (2023) The Author(s).

future research. Second, the PTFE binder exhibits a lower Lowest Unoccupied Molecular Orbital (LUMO) level compared to polyvinylidene fluoride (PVDF), commonly used in wet coating processes, and solvents such as ethylene carbonate and ethyl methyl carbonate[124] (Fig. 14d). This property allows PTFE to attract electrons more readily. As a result, when the PTFE binder is applied to anodes, it can react with lithium and electrons during lithiation (charging), leading to the reduction and side reaction of PTFE. The reduction of PTFE increases the irreversible capacity within the electrode, thereby resulting in degradation of reversible capacity.[64,125] While several studies have proposed strategies to mitigate the reduction of the PTFE binder, such as coating of the polymer with high ionic conductivity on the surface of graphite active materials to block the electron path to the PTFE binder or modification of electrolyte additives to form the stable solid electrolyte interface (SEI), a complete solution has yet to be achieved and further development of strategies to address this challenge is needed.[126,127] Third, the PTFE binder belongs to the category of per- and polyfluoroalkyl substances (PFAS), which may be subject to regulatory restrictions.[128] PFAS regulations currently focus on limiting the use of materials where one or more carbon atoms are fully or partially fluorinated. Today, while the primary targets are low-molecular-weight PFAS compounds, such as

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PFOA and PFOS, which have direct environmental and health issues, high-molecular-weight substances such as PVDF and PTFE are not currently subject to direct regulation. However, given the uncertainty regarding how future PFAS regulations will evolve, it is essential to develop alternative binders capable of fibrillization to replace the PTFE binder. The alternatives could ensure compliance with future regulatory changes while retaining the advantageous properties of PTFE[129] (Fig. 14e). While the PTFE binder holds significant potential for thick electrode manufacturing in the roll-to-roll dry coating process, overcoming challenges related to quantification of fibrillization, reduction issues, and regulatory compliance will be addressed carefully for its successful commercialization. Additionally, as mentioned earlier, due to the side reaction issues of the PTFE binder on the anode side, developing alternative binders suitable for dry anode applications is essential. Fourth, conductive additives are crucial for the enhancement of the electrical conductivity of electrodes. In conventional wet coating processes, a key challenge is the dispersion of conductive additives in the solvent. Poor dispersion can degrade the electrical conductivity of the electrode, necessitating the use of conductive additives with low specific surface area or a high concentration of dispersants to improve dispersion of conductive additives.[130] However, the suppression of agglomeration of conductive additives in the solvent remains difficult, significantly limiting the selection of suitable conductive additives for the conventional wet coating process. In contrast, dry processes eliminate solvents, reducing dispersion-related constraints and broadening the range of applicable conductive additives (Fig. 14f). However, 1D and 2D conductive additives, such as CNTs and graphene, are still prone to agglomeration in the dry coating process. Fortunately, strong shear forces applied during the dry coating process can mitigate this issue, making agglomeration less problematic than in conventional wet coating processes. Consequently, exploring and developing conductive additives optimized for dry processes is essential. As various conductive additives are applicable in dry processes, it is crucial to investigate their impact on the fibrillization behavior of the PTFE binder, which significantly influences the mechanical properties and electrode microstructure. Moreover, understanding how different characteristics of conductive additives affect ionic and electrical resistances in dry electrodes is vital for developing optimized conductive additives for dry processing. Fifth, adhesion between the free-standing electrode film and the current collector in dry processes is primarily based on mechanical interlocking rather than chemical bonding, which often results in poor adhesion.[131] To address this, advanced current collectors, such as etched or carbon-coated copper or aluminum foil, are commonly used to improve the adhesion[56] (Fig. 14g). However, these current collectors significantly increase production costs. Therefore, developing cost-effective current collectors that retain strong adhesion with the free-standing electrode film is crucial, and investigating new surface treatment techniques or alternative coating materials that enhance mechanical interlocking while reducing costs will be essential for the advancement of the dry coating process. Focusing on achievements in conductive additives and current collectors for the dry coating process will drive innovation, improve electrochemical performances, and reduce the costs of dry electrode manufacturing, facilitating their broader industrial adoption. Addressing these issues through continued research and innovation will pave the way for advancing the roll-to-roll dry coating process and next-generation battery applications.

Data availability

This review does not include any primary research results, software, or code, and no new data were created.

Author contributions

Joonhyeok Park and Jiwoon Kim: conceptualization, draft preparation, reviewing & editing, visualization. Jaeik Kim: visualization. Minsung Kim: reviewing. Taeseup Song and Ungyu Paik: funding, supervision, reviewing.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This work was supported by the Human Resources Program in Energy Technology of the Korea Institute of Energy Technology Evaluation and Planning (KETEP), which was granted financial resources from the Ministry of Trade, Industry & Energy, Republic of Korea (No. 20214000000520). This work was also supported by the Technology Innovation Program (or the Industrial Strategic Technology Development Program-Materials & Components Technology Development Program) (20024261, Development of thick film electrode and cell manufacturing technology for high-performance lithium iron phosphate battery with energy density of over 200 W h kg^{-1}) funded by the Ministry of Trade, Industry & Energy (MOTIE, Korea).

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