REVIEW ARTICLE

# Revolutionizing Battery Manufacturing: The Role of Dry Electrode Technology in Sustainable Energy Storage Solutions

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## Abstract

**The growing demand for high-performance energy storage, particularly in electric vehicles and grid applications, has highlighted the limitations of conventional lithium-ion battery manufacturing. Traditional wet-processing electrode fabrication, though widely used, faces challenges such as solvent waste, limited scalability, and inconsistent microstructures. Dry electrode technology (DET) offers a promising alternative by eliminating solvents and drying steps, enhancing sustainability, cost-efficiency, and performance. This review provides a comprehensive overview of DET, emphasizing key microstructural advantages including uniform material distribution, low ion-transport tortuosity, and superior mechanical strength. Various dry fabrication methods, such as binder fibrillation and dry powder spraying, are examined for their potential to enable scalable production of high-areal-capacity, high-energy-density batteries. Additionally, we explore DET applications across lithium-ion, all-solid-state, and lithium-sulfur chemistries, highlighting its effectiveness in addressing challenges related to electrode stability and interface optimization. Despite these advantages, widespread adoption faces hurdles in binder selection and process control. Future developments will require novel materials, improved interfacial engineering, and scalable manufacturing approaches to fully realize DET's potential in next-generation batteries.**

**Keywords**: Lithium-ion batteries, dry electrode technology, all-solid-state batteries, dry-processed electrodes, sustainable energy storage.

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## 1 Introduction

The global demand for sustainable energy and electric transportation is driving rapid advancements in electrochemical energy storage technologies, particularly in the electric vehicle (EV) market. Global EV sales are projected to exceed 17 million units in 2024, reflecting a 25% increase from the previous year, and are expected to account for 20% of global car sales. This growth is supported by policies in over a dozen countries that have committed to banning the sale of new gasoline and diesel vehicles by 2035 or earlier. As the electric vehicle fleet expands, the demand for high-performance, cost-effective, and sustainable energy storage solutions, especially lithium-ion batteries (LIBs), is projected to continue to rise. The International Energy Agency (IEA) forecasts that the global stock of EVs will increase from 16.5 million in 2021 to nearly 350 million by 2030, with energy demand from EV batteries projected to surge to 14 TWh by 2050 — a 90-fold increase from 2020. This exponential growth emphasizes the urgent need for innovations in battery technology to meet the evolving energy demands of the transportation sector and support renewable energy integration in stationary grid applications [1].

Despite significant progress in lithium-ion battery technology, several performance limitations persist, particularly in terms of energy density, cycle life, and manufacturing cost. Current commercial LIBs typically achieve energy densities between 150 and 250 Wh/kg, but applications such as electric vehicles and large-scale storage systems require densities exceeding 500 Wh/kg [2]. To meet these targets, new materials and manufacturing methods are needed, as well as significant improvements in the design and architecture of battery electrodes. The most common electrode manufacturing technique—slurry-based wet processing—remains the dominant approach in the industry [3]. However, this process has several inherent challenges that hinder the performance, scalability, and sustainability of the resulting batteries [4].

The conventional wet electrode manufacturing process involves mixing active materials, conductive agents, and binders into a slurry, which is then coated onto the current collector and dried [5]. While effective, this method has notable drawbacks. First, it requires the use of solvents, typically N-methyl-2-pyrrolidone (NMP), which must be recovered and disposed of, adding both cost and environmental concerns [6]. Second, during the drying process, capillary forces cause binders and conductive agents to migrate toward the electrode surface, leading to uneven distribution of these materials [7]. This migration increases internal resistance, reduces the overall conductivity of the electrode, and may result in cracks and defects due to the volume shrinkage that occurs as the solvent evaporates [8]. Additionally, the migration of materials creates microstructural defects that reduce the overall cycling stability and longevity of the battery [9–12].

The challenges of wet electrode processing are compounded as the demand for higher areal capacity and thicker electrodes increases [13–16]. As batteries are designed to store more energy per unit area (increasing areal capacity), ensuring uniform distribution and stable performance across the electrode thickness becomes more difficult [17]. High areal capacity electrodes are more prone to issues such as poor ion transport, uneven electrolyte wetting, and mechanical degradation. These challenges limit the ability to scale up the manufacturing of next-generation batteries that require thick electrodes for high energy density applications, such as electric vehicles with long driving ranges or large-scale grid storage systems [18–20].

Building on its technical advantages, dry electrode technology (DET) is particularly well-suited to enable the development of next-generation batteries, such as solid-state, sodium-ion, and high-nickel cathode-based LIBs [21–23]. DET eliminates the need for solvents and the energy-intensive drying process, using physical processes such as dry powder mixing, binder fibrillation, and electrostatic powder deposition to create electrodes without the use of liquid-based slurries [24, 25]. This process has several advantages, including a reduction in energy consumption, a smaller carbon footprint, and lower production costs due to the absence of solvent recovery and drying stages. Moreover, by avoiding solvent migration during drying, DET can achieve more uniform distribution of active materials and a more stable electrode microstructure [26–28].

One of the key advantages of DET is its ability to directly produce self-supporting electrodes with optimized three-dimensional (3D) electron/ion conduction networks [29, 30]. These electrodes have uniform pore distribution, low tortuosity, and enhanced mechanical toughness, all of which contribute to improved electrochemical performance [31]. Additionally, dry processing

**Table 1.** Comparison of slurry casting technology and dry electrode technology across key performance metrics.

| Metric | Dry Electrode | Wet Electrode (Slurry Casting) |
|---|---|---|
| Production Cost | Low energy consumption, no solvent recovery, reduced material cost | High energy consumption, solvent recovery required, higher material cost |
| Production Efficiency | High (continuous roll-to-roll process) | Low (batch processing, drying time bottleneck) |
| Environmental Impact | Solvent-free, no toxic emissions | Uses toxic solvents (e.g., NMP), requires solvent recovery and disposal |
| Adhesion Strength | High (mechanical interlocking via fiber network) | Moderate (binder migration reduces bottom adhesion) |
| Electrode Thickness | >500 $\mu$m, crack-free achievable | Typically <100 $\mu$m, prone to cracking at higher thickness |
| Areal Capacity | >10 mAh cm$^{-2}$ demonstrated | Limited by thickness constraints |
| Porosity Control | Adjustable via calendering parameters | Limited control, binder migration creates gradient |
| Microstructural Uniformity | High (uniform component distribution) | Low (binder/conductive agent migration during drying) |
| Tortuosity | Low (aligned pore channels) | High (tortuous pathways from binder segregation) |
| Compatibility with Solid Electrolytes | Excellent (solvent-free, no chemical degradation) | Poor (solvents react with sulfide/oxide electrolytes) |
| Energy Consumption | 40-70% lower than wet process | High (drying and solvent recovery) |
| Equipment Footprint | Compact (no drying ovens, solvent recovery systems) | Large (drying ovens, solvent handling equipment) |

methods have been shown to enhance cycling stability, reduce internal resistance, and improve charge/discharge rates, making them particularly well-suited for high-performance applications such as electric vehicles and fast-charging batteries [32].

In addition to its technical advantages, DET also offers significant economic and environmental benefits. The elimination of solvents and the reduction in energy consumption make dry electrode manufacturing a more sustainable and cost-effective alternative to traditional methods [33]. This is particularly important as the demand for batteries continues to rise, placing increasing pressure on manufacturers to reduce production costs while meeting the growing performance requirements. As the automotive and energy storage industries continue to scale up, the adoption of DET could be a key factor in achieving the affordability, scalability, and sustainability required for next-generation battery technologies [34, 35].

In conclusion, DET represents a disruptive innovation that has the potential to transform the battery manufacturing landscape (Table 1). By addressing the limitations of traditional wet electrode processing and enabling the development of high-performance, cost-effective, and environmentally friendly batteries, DET could play a pivotal role in the widespread adoption of electric vehicles and large-scale energy storage systems [36]. This review aims to explore recent advancements in DET, emphasize its potential benefits, and critically assess the challenges that must be addressed for successful industrial adoption [37, 38].

## 2 The Microstructural Superiority of Dry-Processed Electrodes

The revolutionary potential of DET resides in its capacity to form a near-ideal electrode microstructure a feat challenging to replicate via wet-processed routes [39, 40]. The formation of this structure is attributed to its solvent-free manufacturing essence, allowing it to avoid inherent drawbacks of wet processes—such as solvent evaporation, component migration, and interfacial residues—from the very start, thus yielding unique structural advantages [41, 42].

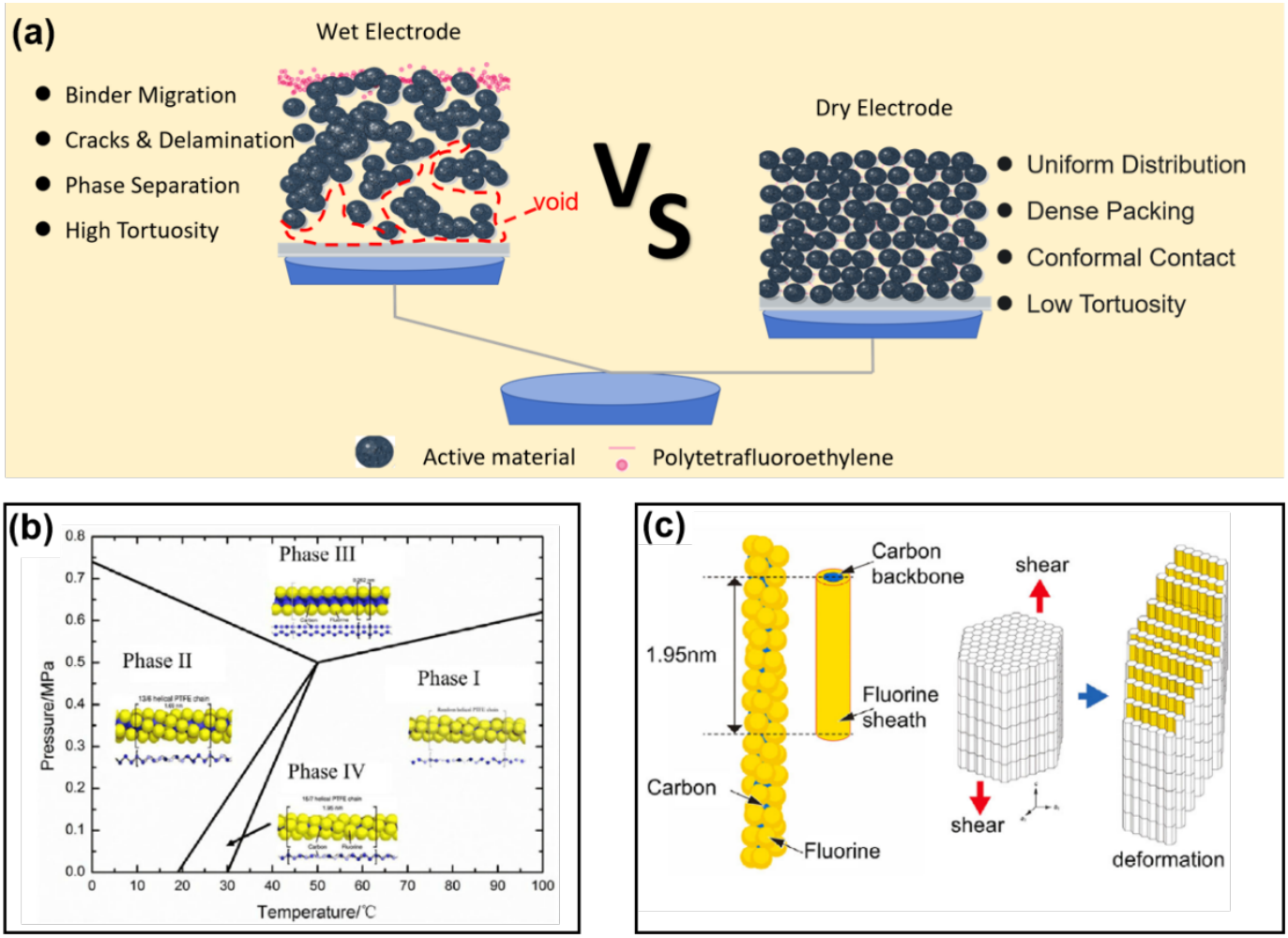


**Figure 1.** a) Comparison of the dry (left) and wet (right) coating electrode processes. b) Phase diagram of PTFE and c) Structural transformation during pre-fibrillation [43, 44].

### 2.1 The Genesis of a Solvent-Free Microstructure

Whether based on binder fibrillation or powder spraying, the core of DET resides in the direct integration of all electrode components into an integrated structure in a dry state via physical forces [45, 46].

In the binder fibrillation process, binders such as Polytetrafluoroethylene (PTFE) undergo a distinctive physical transformation under shear stress [47]. When PTFE particles exceed their phase transition temperature of 19°C, their crystal structure transitions from Phase II to Phase IV(Figure 1(b)) [44], accompanied by molecular chain slippage [43]. This process stretches the spherical particles into microfibers with diameters ranging from 20 to 100 nm. These fibers then entangle and branch extensively, forming a three-dimensional interconnected network that permeates the entire electrode (Figure 1(c)). Through physical entanglement and mechanical interlocking, this network firmly immobilizes active material and conductive agent particles, acting as a porous "polymer scaffold" for the electrode [48, 49].

In the powder spraying process, polyvinylidene difluoride (PVDF) serves as the primary binder [50, 51]. Charged dry powder particles are uniformly deposited onto the current collector under the influence of an electrostatic field, forming a loosely packed powder layer. Subsequent hot pressing induces melting of the thermoplastic binder particles, which forms robust "weld joints" at particle contact points, thereby consolidating the entire structure. This "outside-in" assembly strategy enables particle-level precision stacking, resulting in a pore structure with highly homogeneous distribution.

In contrast, the wet-process route exhibits inherent limitations. During slurry drying, capillary action and diffusion mechanisms associated with solvent evaporation drive the directional migration of binders and low-density conductive agents dispersed in the solvent, following the liquid-phase flow [52]. These

components ultimately accumulate on the electrode surface and within pore channels, while active materials tend to precipitate—resulting in significant concentration gradients along the electrode thickness direction [53]. This phenomenon is particularly prominent in thick electrodes. Consequently, insufficient binder content at the electrode bottom leads to facile peeling of active materials, whereas excessive coverage of binders and conductive agents at the electrode top forms an ion transport barrier. Simultaneously, volume shrinkage during drying introduces internal stresses, which are highly susceptible to inducing electrode cracking and current collector warpage [54].

### 2.2 Key Microstructural Characteristics and Their Impacts

The unique formation mechanism of DET endows them with several key structural features that determine electrochemical performance:

#### *2.2.1 Low-tortuosity ion transport pathways*

By avoiding migration and local enrichment of binders and conductive agents, dry-processed electrodes exhibit more uniform and continuous pore distribution, forming ion diffusion channels with better straightness [55]. This advantage is particularly prominent in PTFE-fibrillated electrodes. Studies have shown that electrodes fabricated via dry printing technology, due to their distinctive microstructure, exhibit significantly lower tortuosity, achieving 69% capacity retention at 4C high rate, far exceeding the 52% of conventional wet-processed electrodes [56]. This is directly attributed to the ability of $Li^+$ ions to transport rapidly through more direct, lower-resistance pathways. In high-areal-capacity $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) dry-processed electrodes, even with an areal capacity as high as 9.5 mAh $cm^{-2}$, their optimized pore structure and conductive network still support stable full-cell cycling, retaining 68% capacity after 1000 cycles—demonstrating exceptional ion migration capability under extreme loading conditions [57].

#### *2.2.2 Uniform interfacial contact and stress distribution*

Dry-processed electrodes achieve uniform distribution of active materials, conductive agents, and binders in three-dimensional space—this characteristic brings revolutionary advantages to both LIBs and ASSBs [58]. The high-intensity shear force in dry processes serves dual functions: it not only mixes components but also induces plastic deformation of ductile sulfide solid electrolytes (e.g., $Li_6PS_5Cl$), enabling them to tightly wrap active material particles and form superior "surface contact." Quantitative studies highlight this advantage: dry-processed electrodes exhibit interfacial coverage as high as 67.2%, whereas wet processes only form isolated "point contacts" with a coverage of merely 33.3% [8]. Such intimate contact drastically reduces solid-solid interfacial impedance, a key factor in achieving high-performance solid-state batteries [59]. For $LiFePO_4$ systems, dry-processed electrodes not only improve Li+ transport but also, through their uniform microstructure, suppress electrolyte side reactions, forming a more stable cathode-electrolyte interface (CEI)—allowing the electrode to maintain excellent structural integrity after 300 cycles [60, 61].

#### *2.2.3 Superior mechanical properties*

The 3D continuous fiber network constructed via PTFE fibrillation endows dry-processed electrodes with unparalleled mechanical properties, laying the foundation for their applications in thick electrodes, flexible devices, and solid-state batteries [32, 62, 63]. Direct mechanical test comparisons show that $LiCoO_2$ electrodes prepared by dry powder spraying (with 5% PVDF) exhibit a mechanical strength of up to 148.8 kPa, significantly higher than the 84.3 kPa of wet-processed controls [20]. More notably, even when the binder content is reduced to 1 wt%, the strength of dry-processed electrodes still surpasses that of traditional wet-processed counterparts. This intrinsic mechanical robustness enables the fabrication of ultra-thick electrodes. For example, $LiFePO_4$ dry-processed electrodes prepared via melt extrusion reach a thickness of 500 $\mu$m, with areal capacity increased to 13.7 mAh $cm^{-2}$ without cracking—breaking the limitations of wet processes [61]. Ultra-low binder content in solid-state battery applications maximizes the advantages of PTFE fibrillation technology. Studies have successfully fabricated NCM self-supporting electrodes with binder content as low as 0.1 wt%, achieving an areal capacity of 6.5 mAh $cm^{-2}$ and exhibiting rate performance comparable to binder-free electrodes—validating the ultra-high bonding efficiency of this network [64].

## 3 Dry Electrode Technology Portfolio: A Unified Manufacturing Language

Among the various technical routes for dry electrode manufacturing, binder fibrillation and powder spraying technology stand out due to their excellent capability in achieving a uniform

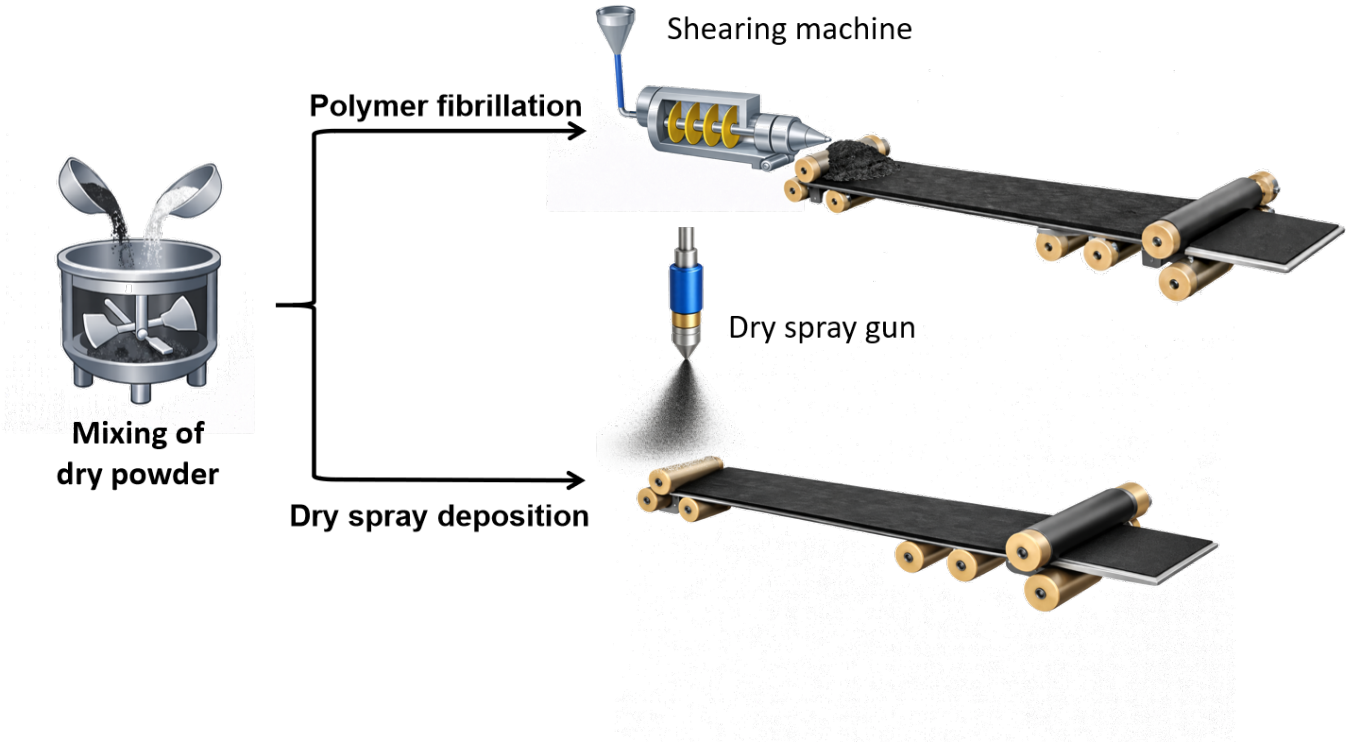


**Figure 2.** Schematic of representative dry electrode fabrication methods.

electrode microstructure, compatibility with large-scale roll-to-roll production, and significant commercialization potential [65, 66]. Although their process principles are distinct, both approaches converge toward a common goal: under solvent-free conditions, achieving a uniform and strong composite of active materials, conductive agents, and binders (for all-solid-state batteries (ASSBs), this also includes solid-state electrolytes (SSEs)), ultimately constructing an ideal electrode structure with low tortuosity and high mechanical integrity, as discussed earlier (Figure 2) [67, 68].

### 3.1 Binder Fibrillation

Binder fibrillation technology, particularly the PTFE-based roll milling process, is currently the most mature dry electrode route with the highest potential for scale-up [31]. The fibrillation of PTFE is driven by both thermodynamics and mechanics. When shear stress is applied to PTFE particles, their crystal structure undergoes a phase transition from Phase II to Phase IV at temperatures above 19°C. The molecular chain slippage stretches the spherical particles ( 500 $\mu$m) into nanoscale fibers (ranging from 20 to 100 nm in diameter) [43, 69]. Dynamic mechanical analysis (DMA) reveals that within the temperature range of 60-110°C, the storage modulus of PTFE exhibits a plateau, which is ideal for fibrillation (Figure 3(a)). At this stage, a fiber network with optimal length and interconnectivity can form, providing the electrode with excellent mechanical strength and electrical conductivity [70].

Two mainstream modes have been developed for this process: one is represented by Maxwell/Tesla [71], which involves first preparing self-supporting films followed by lamination; the other is the Drytrace technology developed by Fraunhofer IWS, which directly forms and transfers electrodes via differential rolls. This latter technology avoids the issue of brittle dry films and is more suitable for continuous production [32].

The microstructure formed by this process is dominated by physical entanglement. Acting as "scaffolds," PTFE fibers mechanically lock active material particles into a highly porous and continuously interconnected network. This structure directly contributes to the electrode's outstanding performance: the fiber network provides exceptional mechanical toughness, enabling the fabrication of crack-free thick electrodes with a thickness of up to 500 $\mu$m and an areal capacity exceeding 13.7 mAh cm$^{-2}$ [61]. Moreover, its "point-contact" bonding mode preserves open and interconnected ion transport channels, which is critical for achieving low tortuosity and excellent rate performance [72].

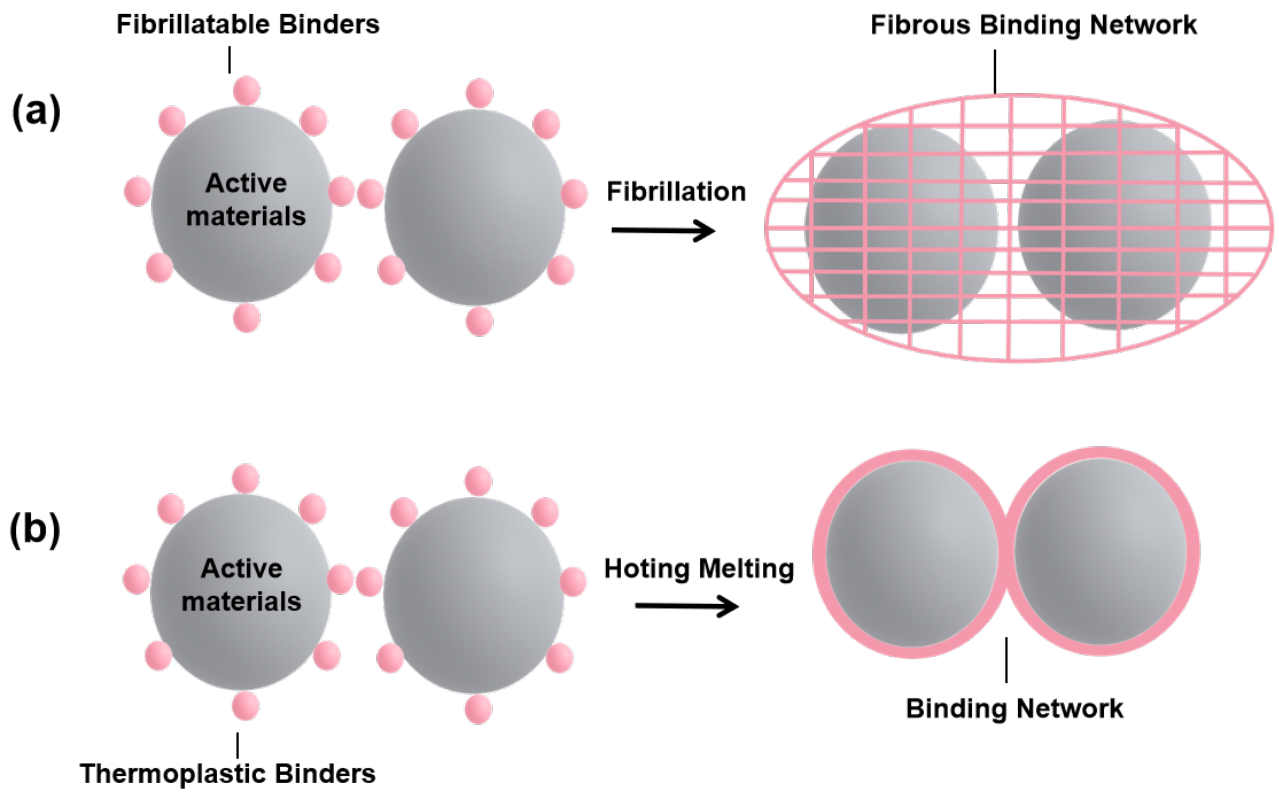


**Figure 3.** Schematic illustration of solvent-free bonding mechanisms in dry electrode technology. a) The bonding via polymer fibrillation relies on shear force to induce PTFE to form a three-dimensional fibrous network, thereby achieving physical entanglement and tightly binding the active particles through mechanical action. b)Spray deposition utilizes thermal effect to promote the melting of the binder and the wetting of particles, followed by curing to form a continuous polymer network, which further realizes the mechanical interlocking effect.

### 3.2 Powder Spray & Electrostatic Deposition

Dry powder spraying technology draws on the principles of industrial powder coating, and achieves the precise arrangement and solidification of dry powders through the synergy between electrostatic action and hot-melt bonding [73–75]. The technology starts with the preparation of uniform dry-mixed composite powders. Subsequently, in an electrostatic field (via corona charging or triboelectric charging), the charged powder particles are accurately and uniformly adsorbed onto the grounded current collector [76, 77]. The final hot calendering step is the key to activating bonding: during this process, thermoplastic binders (e.g., PVDF) melt, flow, wet the particle surfaces, and form strong "bonding bridges" at the contact points. Process parameters such as electrostatic voltage, powder flow rate, hot pressing temperature, and pressure collectively determine the thickness, density, and porosity of the electrode (Figure 3(b)). Studies have shown that through the precise control of these parameters, even when the binder content is as low as 1 wt%, mechanical strength (e.g., up to 148.8 kPa for $LiCoO_2$ electrodes) and electrochemical performance that far exceed those of wet-processed electrodes can be achieved [20].

The microstructure formed by this process is dominated by hot-melt welding. Binders exist in the form of discrete, localized "welding points"—a configuration that minimizes the blockage of ion transport channels by inactive materials while ensuring efficient electronic conduction between particles [78]. This structural feature enables the electrode to maintain high electronic conductivity while also achieving excellent ion accessibility. For instance, NCM811/graphite full cells fabricated via this method exhibit a cycle life twice that of wet-processed cells, benefiting from an optimized microstructure that enables the formation of a more uniform and stable CEI/SEI.

In summary, binder fibrillation technology, particularly the roll milling process, has emerged as the dominant direction and focus of current scaled development of dry electrodes. It offers inherent advantages in achieving an ideal electrode microstructure, excellent mechanical properties, and the potential for seamless integration with the existing battery manufacturing

industry chain. Meanwhile, the search for new environmentally friendly fibrillable binders capable of replacing PTFE remains crucial for advancing the sustainable development of this technology [79].

## 4 Application Across Battery Chemistries

DET is not limited to a specific type of battery chemistry. Its core value lies in its ability to serve as a universal platform for manufacturing electrode microstructures, offering unique solutions to address the common challenges faced by various next-generation battery systems [80]. To date, dry electrodes have been successfully applied in a range of systems, including LIBs [81], ASSBs [82, 83], and aqueous batteries.

### 4.1 Lithium-Ion Batteries

One of the most direct approaches to achieving high energy density in LIBs is to enhance areal capacity. However, when fabricating thick electrodes using traditional wet-process technology, issues such as binder migration and electrode cracking caused by solvent evaporation severely degrade charge transport kinetics [84].

DET provides a fundamental solution to this challenge through its solvent-free nature and uniform microstructural characteristics, as demonstrated in a study by Tao et al. [85] (Figure 4(a)). They successfully fabricated thick electrodes with areal capacities of 6.6 mAh $cm^{-2}$ (graphite anode) and 6.0 mAh $cm^{-2}$ (NMC622 cathode) using dry powder mixing and roll pressing. Structural analysis revealed that dry electrodes exhibited significantly lower tortuosity (e.g., 2.05 for DP-Gr, compared to 4.59 for traditional wet-processed C-Gr), which provides a more direct pathway for ion transport. Although the PTFE binder leads to low initial Coulombic efficiency in the graphite anode, the assembled dry-process full cell still demonstrated excellent rate performance and long cycle life, with a capacity retention rate of 63.6% after 800 cycles (Figure 4(b)). This result underscores the mechanical robustness of the dry electrode structure during long-term cycling.

Moreover, the work by Kim et al. [86] has pushed the performance of dry-process thick electrodes to new heights (Figure 4(c)). By precisely regulating the fibrillation temperature of PTFE (80°C), introducing graphene-based multidimensional conductive agents to optimize pore distribution, and employing a two-step calendering process to prevent active particle cracking, they successfully fabricated a high-performance dry-process cathode with a homogeneous microstructure, achieving an areal capacity of 10 mAh $cm^{-2}$. This electrode delivered a high volumetric energy density of approximately 710 Wh $L^{-1}$ and exhibited excellent rate performance (capacity retention rate of 59.5% at 2C/0.2C) (Figure 4(d)) and cycling stability (capacity retention rate of 89.5% after 100 cycles). These results strongly confirm that dry electrode technology, through proactive microstructure engineering, can fundamentally address the charge transport challenges in thick electrodes and serves as a key enabler for the next generation of high-energy-density lithium-ion batteries.

Beyond the PTFE fibrillation approach, the dry spray deposition method has also emerged as an efficient technique for manufacturing high-loading and fast-charging batteries, primarily due to the optimized distribution of electrode components (Figure 4(e)). Wang et al. [94] developed a scalable dry spray deposition technology compatible with roll-to-roll manufacturing systems to produce electrodes specifically designed for fast-charging applications. The dry-processed electrodes exhibited superior rate performance (69% capacity retention at 4C) compared to commercial standard electrodes. Analysis confirmed that the specific microstructure of dry-processed electrodes results in significantly lower tortuosity and faster ion diffusion paths. Additionally, the introduction of a surface coating composed of PVDF and conductive carbon effectively protects the cathode surface from harmful electrolyte side reactions [56, 87].

Thus, dry spray deposition technology can be applied to nearly all particle-based manufacturing processes, including the fabrication of both cathodes and anodes. Fu et al. [3] further explored the application of this technology in manufacturing ultra-high-loading NCM811 and graphite electrodes (Figure 4(f)). The optimized microstructure of the dry-processed electrodes enhances electrolyte penetration and minimizes $Li^+$ diffusion tortuosity, improving rate performance at high current densities. Moreover, this innovative electrode manufacturing method enables more uniform CEI and SEI formation and growth. Compared to conventionally SCT-processed batteries, this effectively doubles the cycle life of single-layer pouch cells equipped with dry-processed electrodes [3].

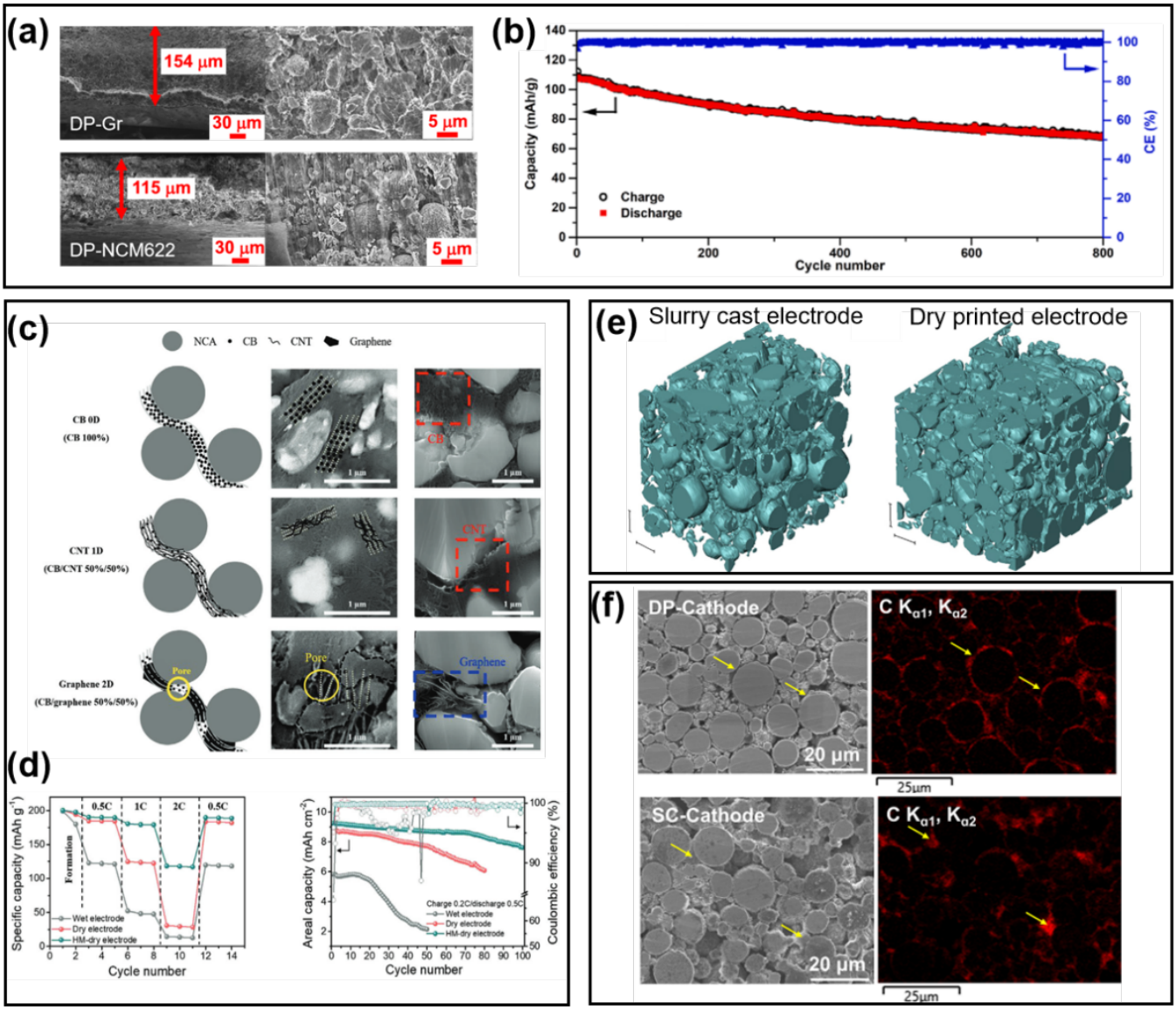


**Figure 4.** a) SEM images of DP- Gr and NMC622 electrodes. b) cyclic performance study of the full-cells after aging at a current density of 0.1C for three cycles [85]. Copyright 2023, Elsevier Ltd. c) SEM images of the electrodes introducing multi-dimensional conducting agent [86]. Copyright 2024, American Chemical Society. d)Discharge rate capabilities of wet, dry, and HM-dry electrodes (charging 0.2 C). Cycle stability ofwet, dry, and HM-dry electrodes (charging 0.2 C/discharging 0.5 C). e) 3D NMC electrode structures reconstructed from X-ray nanotomography.Carbon signal distribution on the cross-section of the [56]. Copyright 2023, Elsevier Ltd. f) SCT-based and DET-based electrodes [3]. Copyright 2024, Elsevier Ltd..

### 4.2 All-Solid-State Batteries

The commercialization of ASSBs is severely hindered by instability and contact loss at solid-solid interfaces. DET, particularly its solvent-free nature and ability to apply high shear force, provides a unique platform for constructing ideal electrode-solid electrolyte interfaces [88–90].

The co-roll pressing dry process developed by Lee et al. [91] serves as a paradigm in this field (Figure 5(a)). This technology abandons the challenging approach of independently fabricating ultra-thin SE films; instead, it directly co-roll presses thick SE feedstock with cathode feedstock. This not only forms a 50 $\mu$m ultra-thin SE layer and a high areal capacity cathode (5 mAh cm$^{-2}$) but also in-situ constructs a robust and intimate interface. Studies have shown that the shear force during the roll pressing process induces fibrillation of the PTFE binder and conductive agent at the interface, forming a 3D interconnected network. This achieves far superior interfacial mechanical strength and integrity compared to traditional lamination methods. Benefiting from this optimized interfacial microstructure, the ASSB

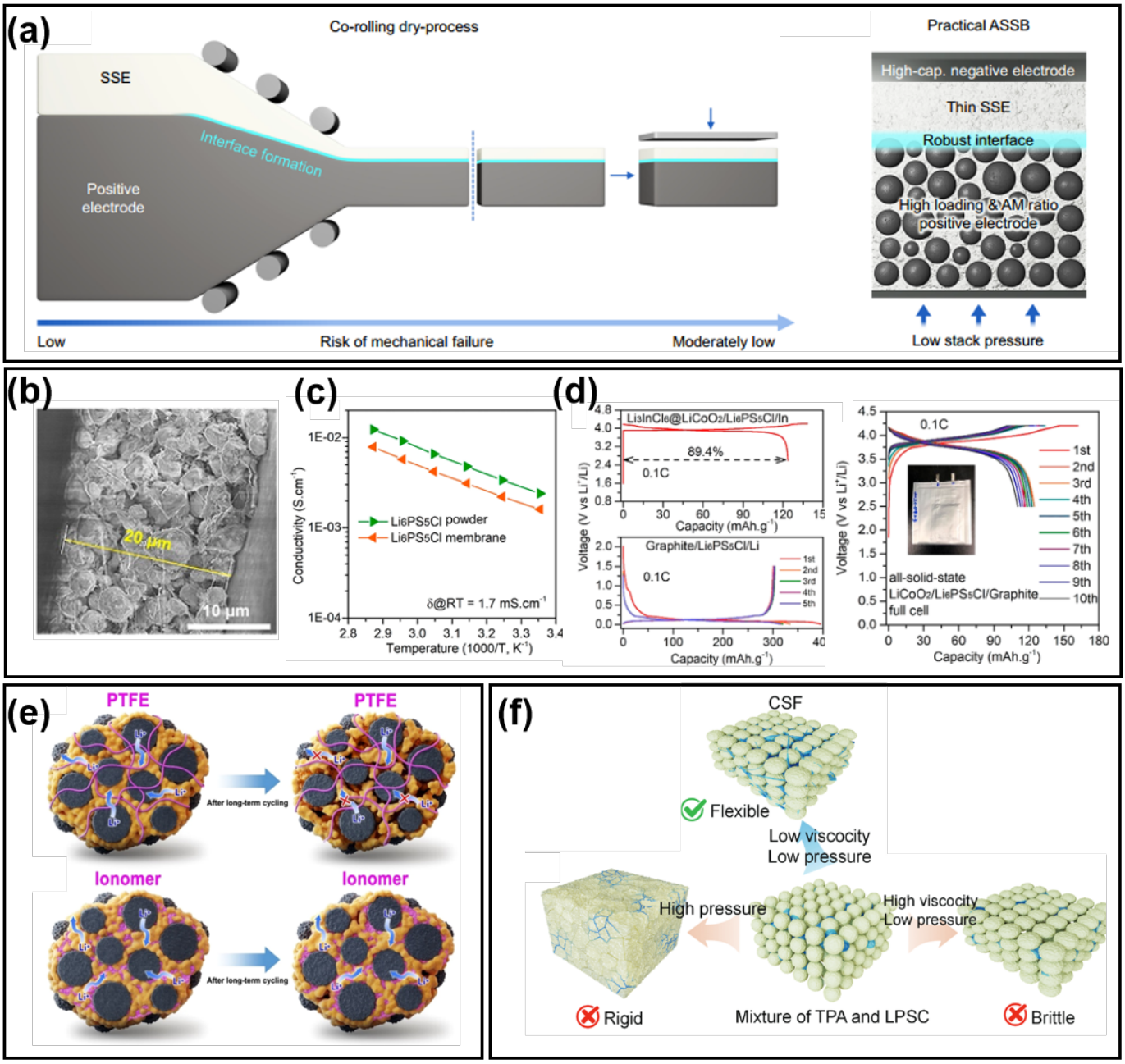


**Figure 5.** a) Fabrication design of co-rolling dry-process, Cell design and consideration for practical ASSB [91]. b) Cross-sectional SEM image of a 20 $\mu$m LLZTO membrane. c)Arrhenius plot of $Li_6PS_5Cl$ powder and its membrane. d) Initial charge-discharge curves of the freestanding electrode $Li_3InCl_6$@$LiCoO_2$ (top) and initial 5 charge-discharge curves of Graphite@$Li_6PS_5Cl$ electrodes (bottom), Initial 10 charge-discharge curves of ASSBs with a configuration of $Li_3InCl_6$@$LiCoO_2$/$Li_3InCl_6$+$Li_6PS_5Cl$/Graphite@$Li_6PS_5Cl$ [94]. Copyright 2021, American Chemical Society. e) Schematic of PTFE and ionomer distribution in the composite cathodes [95]. Copyright 2022, American Chemical Society. f) Effect of viscosity of fused binder and preparation pressure on the flexibility [96]. Copyright 2024, Wiley-VCH.

based on this co-rolled film achieves stable cycling over 500 cycles (capacity retention > 80%) under extremely low stacking pressure (2 MPa) and has successfully been fabricated into pouch cells with high energy density (310 Wh $kg^{-1}$, 805 Wh $L^{-1}$). This work compellingly demonstrates that dry electrode technology can proactively design and precisely control the most critical electrode-electrolyte interface in ASSBs, providing a key manufacturing and design paradigm for realizing high-performance ASSBs suitable for low-pressure operation [91].

DET is a core solution to overcome the manufacturing bottlenecks of ASSBs. Its solvent-free nature fundamentally avoids the chemical reactions between polar solvents (in traditional slurry casting) and sulfide/oxide SEs (e.g., the complexation

degradation of sulfide SEs with oxygen/nitrogen elements in solvents) [92]. Moreover, through precise microstructural regulation, it constructs low-impedance ion transport channels and electrode/electrolyte systems with high mechanical stability, addressing three core challenges of ASSBs—"SE-solvent incompatibility", "poor solid-solid contact", and "easy breakage of thin SE films"—while providing a universal route for the large-scale fabrication of high-energy-density ASSBs [93].

Wang et al. [94] demonstrated a solvent-free approach to fabricate freestanding and ultrathin inorganic solid electrolyte membranes by utilizing PTFE shear fibrillation. They dry-mixed and sheared different types of SE particles (including $Li_6PS_5Cl$ (sulfide), $Li_3InCl_6$ (halide), and $Li_{6.5}La_3Zr_{1.5}Ta_{0.5}O_{12}$ (LLZTO, oxide)) with PTFE powder. This process caused PTFE to form a three-dimensional fibrous network that "weaves" the SE particles, resulting in large-area ($8 \times 6\,cm^2$), ultra-thin (15–20 $\mu$m) self-supporting SE films (Figure 5(b)). Due to the absence of solvent residues and the uniform fiber-SE composite structure, these films exhibit room-temperature ionic conductivities exceeding 1 mS $cm^{-1}$ (e.g., 1.7 mS $cm^{-1}$ for the $Li_6PS_5Cl$ film (Figure 5(c))). The assembled ASSB pouch cells (e.g., double-layer $Li_6PS_5Cl$ / $Li_6PS_5Cl$ film matched with NCM cathode) deliver a discharge capacity of 124.3 mAh $g^{-1}$ at 0.1C with an initial Coulombic efficiency of 89.4% (Figure 5(d)). Additionally, the LLZTO-based film shows excellent flame-retardant safety. This work demonstrates that DET is compatible with multiple inorganic SEs and enables the low-cost fabrication of high-performance SE films [94].

To address the issue that traditional PTFE binders have low ionic conductivity and easily increase the interface impedance of ASSB cathodes, Hong et al. [95] designed a $Li^+$-conducting PTFE-based ionomer (polytetrafluoroethylene-co-perfluoro (3-oxa-4-pentenesulfonic acid) lithium salt) and used it as the dry binder for sulfide-based ASSB composite cathodes (Figure 5(e)). This ionomer retains the shear fibrillation capability of PTFE, enabling the formation of a uniform composite structure with active materials, conductive carbon, and $Li_6PS_5Cl$ SE. Meanwhile, its lithium sulfonate groups provide additional $Li^+$ transport channels, significantly improving solid-solid interface contact. The ASSB based on this cathode delivers a discharge capacity of 180.7 mAh $g^{-1}$ at 0.1C and maintains 90% capacity retention after 300 cycles at 0.5C. Compared to cathodes with traditional PTFE binders, the ion transport impedance is reduced by 40%, solving the problem of "difficulty in balancing mechanical integrity and ionic conduction" for dry-process cathodes in ASSBs [95, 106].

To further meet the demand for "thin SE films + thick electrodes" in high-energy-density ASSBs, Hu et al. [96] proposed a thermoplastic polyamide (TPA)-assisted dry fusion bonding technology (Figure 5(f)). After dry-mixing low-viscosity TPA binder with $Li_6PS_5Cl$ SE, low-temperature hot pressing ($\leq$5 MPa) was applied to make TPA form a permeable network between SE particles, fabricating flexible composite SE films with a thickness of $\leq$25 $\mu$m. The thermoplastic property of TPA not only endows the SE film with excellent stress dissipation capability (avoiding interfacial delamination during cycling) but also enables intimate interfacial bonding with thick NCM811 cathodes (areal capacity > 4 mAh $cm^{-2}$). The assembled ASSBs exhibit a stable areal capacity of 2.5 mAh $cm^{-2}$ at 0.2C, with no significant capacity fading after 1400 cycles (over 9200 h), and achieve energy densities of 390 Wh $kg^{-1}$ and 1020 Wh $L^{-1}$. This advances the application of DET from "basic SE fabrication" to "full-system integration of high-energy-density ASSBs" [96].

### 4.3 Lithium-Sulfur

Thieme et al. [105] were the first to apply binder fibrillation to the fabrication of sulfur cathodes (Figure 6(a)). They used sulfur-impregnated carbon materials, multi-walled carbon nanotubes (MWCNTs), and PTFE binder, and induced PTFE fibrillation via hot rolling, successfully preparing self-supporting dry-process sulfur cathodes. At a loading of approximately 3 mg $cm^{-2}$ this electrode achieved a reversible capacity of around 1000 mAh $g^{-1}$ outperforming electrodes fabricated via the conventional slurry method [97].

Sul et al. [98] further optimized this process and developed a dry-process sulfur cathode containing only 1 wt% PTFE (Figure 6(b)). This low binder content significantly enhanced the battery's energy density. Studies showed that the dry-process electrode avoided the binder migration issue common in slurry processes and formed a denser, more uniform structure. As a result, it achieved an areal capacity of 10 mAh $cm^{-2}$ at a high loading of 12 mg $cm^{-2}$, exhibited excellent cycling stability, and successfully demonstrated its applicability in large-scale pouch cells [98].

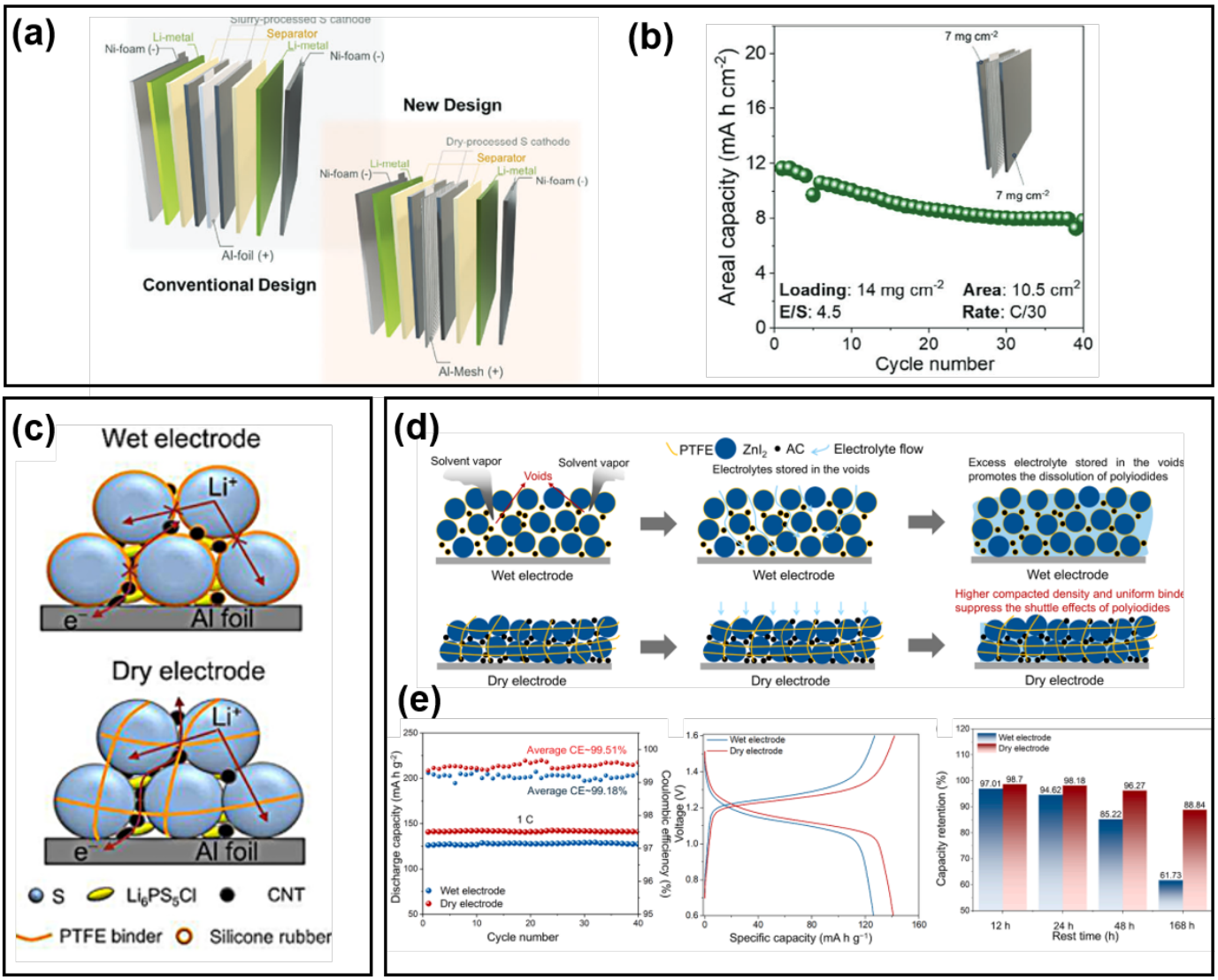


**Figure 6.** a) Schematic of the conventional multi-layer pouch cell and novel pouch cell design with the dry-processed electrodes. b) Cycling performance of dry-processed multi-layer pouch cell [98]. Copyright 2024, Wiley-VCH. c)Schematic diagrams of electrode microstructures by conventional wet-slurry casting and dry electrodet echnology [99]. Copyright 2022, Elsevier Ltd. d)Schematic illustration of structural differences in wet and dry electrodes and impacts on the shuttle effects of polyiodides. e) Cycling performance of wet and dry electrodes in 2 M $ZnSO_4$ electrolyte (Zn metal as anode). The Galvanostatic charge-discharge (GCD) test of wet and dry electrodes in 2 M $ZnSO_4$ electrolyte. Self-discharge performance of batteries using wet or dry electrodes after 12, 24, 48, and 168 h [100]. Copyright 2025, Elsevier Ltd..

Hu et al. [99] successfully integrated dry-processed sulfur cathodes with dry-processed $Li_6PS_5Cl$ solid electrolyte films, constructing high-performance pouch-type all-solid-state lithium-sulfur batteries (Figure 6(c)). This study highlights the unique advantage of DET in constructing robust solid-solid interfaces: the 3D network formed by fibrillation not only provides continuous electron conduction pathways but also establishes tight mechanical interlocking and ionic contact with solid electrolyte particles, effectively reducing interfacial impedance. Operating at 60°C and 0.05C, the battery achieved a total capacity of 10 mAh, fully demonstrating the practical potential of the dry-process electrode structure in ASSBs.

### 4.4 Batteries with Aqueous and Special Electrolytes (Na, Zn)

As a promising alternative to LIBs, sodium-ion batteries (SIBs) have been extensively investigated owing to their low raw material costs and high resource availability. However, the primary challenges faced by SIBs, including relatively low energy density and power density, have to some extent limited their widespread practical application. To enhance the performance of SIBs, improving the quality

and loading level of electrodes has emerged as one of the key strategies. In this regard, DET has demonstrated tremendous potential. Beyond cost reduction, optimizing the compatibility between active materials and dry fibrillation processes is of critical importance. Studies on dry-processed $NaCrO_2$ cathodes indicate that micron-sized particles (>4.4 $\mu$m) exhibit significantly superior electrochemical performance compared to submicron particles. The high specific surface area of small particles leads to excessive consumption of PTFE binder, resulting in incomplete network formation and poor mechanical strength. In contrast, large particles enable more effective coating and conductive network penetration even at an extremely low binder content (0.5%), forming dense electrode sheets with a low porosity of 27.9%. Based on this strategy, they fabricated self-supporting electrodes with an active material content as high as 96.5% and a mass loading of 45 mg $cm^{-2}$. This work highlights that the design of "dry-process compatible materials", especially particle size optimization, is a critical pathway for advancing dry electrode technology in SIBs.

In addition, DET has shown great potential in addressing the inherent challenges of aqueous halogen batteries, such as active material sublimation and dissolution. Wu et al. [100] developed a solvent-free preparation strategy for zinc-iodine ($Zn$-$I_2$) batteries (Figure 6(d)), achieving an unprecedentedly high areal capacity of 15.8 mAh $cm^{-2}$ (with a mass loading >100 mg $cm^{-2}$). Unlike wet-processed electrodes, which generate pores during solvent evaporation and thus exacerbate polysulfide (polyiodide) shuttling, dry-processed electrodes possess a dense, void-free microstructure constructed by a fibrillated PTFE network. This unique architecture effectively reduces excessive electrolyte wettability and physically restricts the diffusion of polyiodide species, thereby significantly lowering the self-discharge rate. Combined with the in-situ formed polymer SEI film on the anode side, the assembled Ah-scale pouch cells achieved 750 stable cycles (Figure 6(e)), highlighting the advantages of DET as a scalable solution for high-energy-density aqueous batteries [100].

## 5 Universal Challenges and Microstructure-Based Solutions

While DET demonstrates great potential in avoiding solvent-related defects, its industrialization process still faces several core challenges. Fundamentally, these challenges revolve around how to precisely construct and control the electrode microstructure under solvent-free conditions. From the perspective of microstructure design, this chapter will reexamine these universal challenges and discuss the corresponding solutions [101].

### 5.1 The Binder Dilemma: Beyond PTFE

As an indispensable binder in current dry electrodes, PTFE's unique fibrillation capability is key to constructing 3D microstructure scaffolds. However, from the perspective of microstructure integrity, PTFE has inherent drawbacks [102, 103]. First, its hydrophobicity and chemical inertness result in weak interfacial adhesion with active materials, conductive agents, and current collectors. Relying solely on van der Waals forces makes it difficult to maintain stable interfacial contact during cycling—especially in electrodes with high volume variation (e.g., silicon-based anodes)—leading to particle detachment and electrode failure [104]. Second, achieving uniform dispersion of PTFE is a significant challenge. Its low surface energy causes it to easily agglomerate during dry mixing, forming insulating binder clumps that disrupt the continuity of the conductive network and induce inhomogeneity in the electrode microstructure. More critically, PTFE undergoes electrochemical reduction at the low potential of the anode (~1.2 V vs. Li/Li+), generating LiF and consuming active lithium. This not only causes irreversible capacity loss but also destroys the integrity of the fibrillated network, triggering electrode cracking and rapid capacity fading [27].

To address this dilemma, strategies are shifting from simple "replacement" to "functional synergy." PFAS-free approaches aim to minimize PTFE content while introducing auxiliary functional binders [105]. For instance, the PAA-g-CMC polymer developed by Kang et al. [31] acted as a "binder anchor". Through abundant carboxyl and hydroxyl groups, it formed strong bonds with active particles and anchored PTFE fibers. By reducing PTFE content by 70%, this strategy further enhanced the electrode's mechanical integrity and electrochemical performance. Font et al. [72] utilized hydrogen bonding interactions between PAA and both current collectors and particles to improve the electrode's adhesion and cohesion. For anodes, interfacial protection strategies are particularly crucial. Studies by Lu et al. [29] and Doerrer et al. [107] have shown that coating the surface of graphite particles with a layer of polyethylene oxide (PEO) or

fluorinated copolymer can form an ion-conducting yet electron-insulating barrier. This isolates PTFE from direct contact with the low potential, significantly suppressing its decomposition and improving initial Coulombic efficiency and cycling stability.

### 5.2 The Conductive Network Reformation

In dry processes, constructing efficient and stable mixed conductive networks (electronic and ionic) presents unique challenges. The difficulty with electronic conductive networks arises because conductive agents—especially carbon nanotubes (CNTs) or graphene, which have high aspect ratios—are highly prone to agglomeration in the dry state [108]. This makes uniform dispersion difficult, unlike in slurries, where solvation and shear forces facilitate dispersion [96]. Uneven distribution of conductive agents leads to localized high current density and increased impedance. Current solutions include:1) Pre-composite strategy: Nanoscale conductive agents are pre-coated on the surface of active material particles via methods such as spray drying to form a core-shell structure, ensuring intimate contact at the source [109]. 2) Optimized mixing processes: Adopting high-intensity dry mixing or sequential feeding methods can effectively break up agglomerates, form finer conductive pathways, and significantly reduce electrode resistance.

Challenges for ionic conductive networks stem from the complexity of the pore structure in dry electrodes [110]. Although dry electrodes typically exhibit lower tortuosity, the hydrophobicity of PTFE severely hinders the wetting of liquid electrolytes—especially in thick electrodes. Therefore, hydrophilic components can be introduced. For example, incorporating polymers with polar functional groups (such as polyacrylic acid (PAA) or PAA-grafted CMC (PAA-g-CMC)) into the binder system can effectively improve the electrode's electrolyte wettability. Alternatively, pore structure design can be optimized: by regulating calendering process parameters (pressure, number of passes), sufficient inter-particle contact is ensured while retaining an appropriate amount of interconnected pores, which serve as channels for ionic transport.

### 5.3 Process Control and Scalability: The Pursuit of Microstructural Uniformity

The scale-up challenges of dry electrodes primarily involve maintaining the uniformity and consistency of the electrode microstructure at both macroscopic and microscopic scales during high-speed continuous manufacturing processes [108, 111].

Dry powder mixing is highly sensitive to the size, shape, density, and surface properties of particles, making segregation prone to occur [28]. As discussed in Section 2.1, the morphology of active materials (e.g., flaky graphite hinders PTFE fibrillation) significantly affects the final structure. Therefore, developing intelligent multi-stage mixing processes and technologies for online monitoring of powder mixing quality is crucial [112]. Meanwhile, regulating the degree of fibrillation is critical, as it directly determines the mechanical strength of the electrode film. Inadequate shearing results in an incomplete network and brittle electrodes, while excessive shearing may cause fiber breakage or active particle damage. This requires precise and stable control of shear force, speed, and temperature during the roll milling/calendering processes [65].

Additionally, the weak adhesion between PTFE and metal current collectors presents another bottleneck in large-scale production [113]. To achieve reliable lamination, relatively high pressure or temperature is often required, which may damage the electrode structure. The development of dry-process-compatible interfacial adhesive layers or surface modification of current collectors (e.g., plasma treatment) represents potential directions for future exploration [114].

## 6 Forward-Looking Perspectives

To push DET beyond its current bottlenecks and unlock its full potential, the future development requires several key paradigm shifts, as summarized in Figure 7 [57].

### 6.1 Material Design: From "Slurry-Compatible" to "Dry-Process-Optimized"

Future material design must break free from the mindset of traditional wet-process technology and be tailored specifically for DET. This includes: Active materials: Designing particles with specific morphologies (e.g., spherical, porous) and surface chemical properties to enhance uniformity during dry mixing and improve interfacial interactions with binders. Specialized binders: Developing novel non-PTFE-based polymers with fibrillation capability (such as the recently reported sericin protein [88]), or designing multifunctional binders with intrinsic ionic/electronic conductivity. Functional conductive agents: Researching conductive agents that are more easily dispersible in the dry state, or materials with

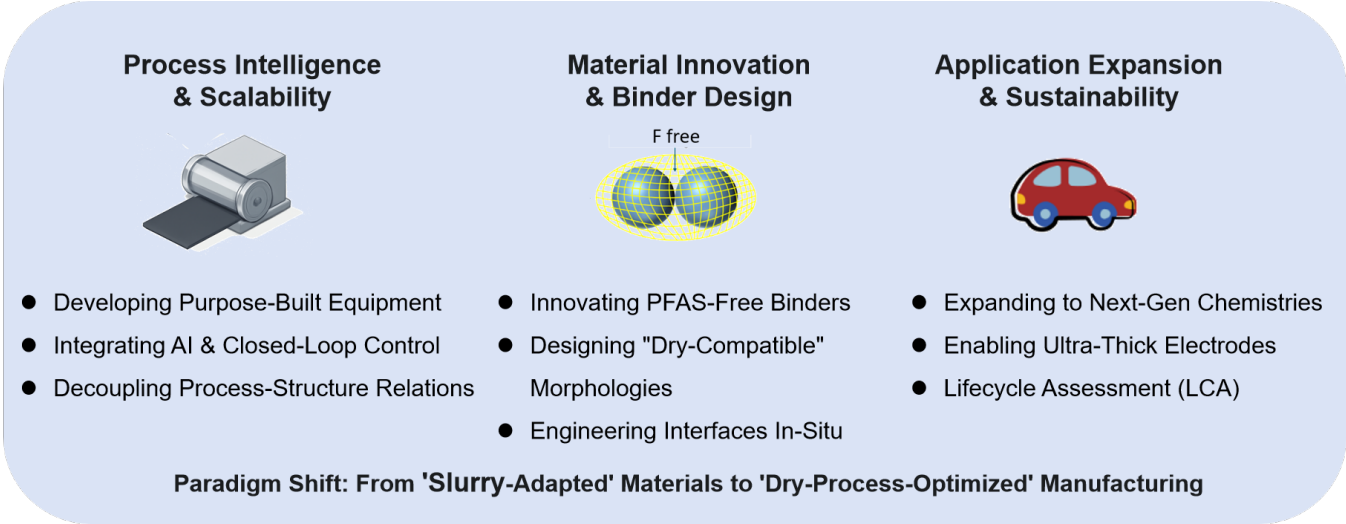


**Figure 7.** Challenges and outlook of dry electrode technology.

specific electrical properties to assist the electrostatic deposition process.

### 6.2 Interfacial Engineering in the Dry State

With the maturation of bulk microstructure control technology, the research focus will shift toward more refined dry-state interfacial engineering. The goal is to directly construct stable interfaces during electrode fabrication under solvent-free conditions. For instance, exploration will be conducted to form artificial CEI /SEI layers or ion-conductive coating layers on the surface of active particles via in-situ reactions or physical vapor deposition during dry mixing or calendering processes. This pre-stabilizes the interfaces prior to battery assembly, thereby improving initial Coulombic efficiency and cycle life.

### 6.3 The Role of Artificial intelligence (AI) and Advanced Characterization

Understanding and optimizing the complex relationship between "process parameters-microstructure-performance" is central to advancing DET [115]. AI and machine learning will play a pivotal role here: by analyzing massive volumes of process data and performance results, they can reverse-recommend optimal combinations of process parameters and design schemes for new materials. Meanwhile, advanced in-situ/quasi-in-situ characterization techniques—such as synchrotron X-ray tomography and in-situ electrochemical testing—can real-time reveal the dynamic evolution of electrode microstructures during fabrication and cycling, providing crucial validation data for theoretical models and AI.

### 6.4 Sustainability and Lifecycle Thinking

The value of DET should be evaluated within a broader framework of sustainability and full life cycle. Future research should not only focus on achieving fluorine-free designs to thoroughly address environmental concerns but also conduct comprehensive environmental footprint analysis of the dry process—spanning raw material acquisition, manufacturing, and battery recycling. By quantifying its advantages in energy consumption, carbon emissions, and hazardous substance emissions, DET can truly establish its position as a core technology for next-generation green battery manufacturing. The roadmap presented in Figure 7 encapsulates the transformative potential of DET across materials, interfaces, characterization, and sustainability dimensions.

## 7 Conclusion

Dry electrode technology is not merely a solvent-free manufacturing upgrade but a fundamental paradigm shift in electrode architecture. By eliminating solvent evaporation-induced issues such as binder migration and capillary damage, DET enables the construction of microstructures with an ideal 3D conductive network, low ionic tortuosity, and robust mechanical integrity. This structural advantage positions it as a key enabling technology for high-areal-capacity electrodes and challenging chemistries (e.g., silicon and lithium metal), aligning it with the new energy industry's demand for high-energy-density and sustainable energy storage solutions.

DET has now advanced from laboratory scale to pilot production, becoming a strategic focus

of the global battery industry. While dedicated manufacturing equipment has been localized and validated, the technology remains in a phase of mature pilot development with pending large-scale commercialization. Wet processes still dominate mainstream production, as low-cost mass production of DET across the entire value chain has not yet been realized. Despite its potential, DET faces significant material and engineering hurdles. Materially, the heavy reliance on PTFE binders creates bottlenecks in interfacial adhesion, electrochemical stability, and environmental sustainability (PFAS concerns). From an engineering perspective, equipment incompatibility with existing wet production lines, escalated capital expenditure, and the lack of standardized in-line quality control for microstructural parameters (e.g., fibrillation integrity) remain critical barriers to scalable production.

In the short term, industrialization will depend on PFAS-reduced formulations and retrofitted production lines. Long-term success, however, hinges on three pillars: the development of high-performance PFAS-free binders, the standardization of dedicated equipment and processes, and the co-design of materials tailored for dry processing. The ultimate maturation of DET will require interdisciplinary integration across materials science, mechanical engineering, and automation, coupled with a life-cycle approach to manufacturing.

In summary, DET offers a universal electrode solution spanning liquid Li-ion to all-solid-state batteries. Despite current challenges, its inherent advantages in energy efficiency, environmental impact, and electrochemical performance solidify its role as a core candidate for next-generation battery manufacturing. As material and process innovations continue, DET is poised to transition from lab-scale innovation to industrial reality, driving the sustainable evolution of global energy storage systems.

## Data Availability Statement

Data will be made available on request.

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## Conflicts of Interest

The authors declare no conflicts of interest.

## AI Use Statement

The authors declare that no generative AI was used in the preparation of this manuscript.

## Ethical Approval and Consent to Participate

Not applicable.

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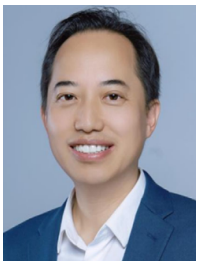

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