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Function-Oriented Binder Architectures for Dry-Processed Lithium-Ion Battery Electrodes

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ABSTRACT

The role of binders in lithium-ion battery (LIB) electrodes is undergoing a fundamental reevaluation as dry electrode processing gains momentum. In solvent-free systems, the binder must operate without the aid of liquid-phase dispersion or capillary adhesion, instead enabling structural cohesion, ionic percolation, and mechanical integrity through entirely solid-state mechanisms. These evolving demands render traditional views of binders as passive, inert glues obsolete. While polytetrafluoroethylene (PTFE) remains central to current dry electrode fabrication due to its shear-induced fibrillation, environmental and political regulation and electrochemical and interfacial limitations have underscored the need to move beyond both its chemistry and design logic. Rather than seeking one-to-one material replacements, a growing body of research reframes the binder as a platform of modular functionality, distributing roles such as adhesion, elasticity, and interface stabilization across hybrid, reversible, or sacrificial materials. This function-first approach enables the decoupling of cohesion mechanisms from specific chemistries, opening pathways to dry electrode systems tailored to emerging battery architectures. In this perspective, we explore the structural design principles and interfacial strategies that redefine the binder not as a static material but as an actively engineered system essential to the advancement of sustainable, high-performance dry electrode technologies.

1 | Introduction

Historically, polymer binders in lithium-ion battery (LIB) electrodes have been viewed as passive components that are structurally essential but chemically and electrochemically inert [1]. Within this paradigm, binders have been optimized primarily for processing compatibility and adhesion rather than for functional performance [2]. However, as battery technologies move toward architectures that push the limits of thickness, rate capability, and manufacturability, the limitations of conventional binder roles are becoming increasingly apparent [3, 4]. Emerging demands for high-loading electrodes, faster fabrication routes, and solvent-free processing have exposed the inadequacy of viewing the binder as merely "glue."

One of the clearest examples of this shift is seen in dry electrode manufacturing [5, 6]. In this solvent-free approach, the binder must do more than provide adhesion, as it must sustain mechanical cohesion under shear, enable electrical and ionic percolation in the absence of liquid-phase mixing, and integrate seamlessly into roll-to-roll (R2R) processing. This multifunctional requirement has brought attention to polymers like

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polytetrafluoroethylene (PTFE). While PTFE has historically been a cornerstone of supercapacitor manufacturing due to its unique fibrillation behavior, its adoption in LIB electrodes is a more recent development driven by the push for dry processing [7–11]. Its fibrillation under shear enables a seamless dry "powder-to-film" transition [12–15]. However, therein lies both a technical and regulatory challenge, including that the PTFE is increasingly targeted due to the environmental persistence, bioaccumulation potential, and resistance to degradation [16–18].

As environmental constraints begin to intersect with manufacturing needs, a purely material-centric definition of the binder is proving unsustainable. Instead, a function-oriented approach is needed: one that decouples material identity from performance and reframes binder design around four foundational mandates, mechanical integration, interfacial stability, electrochemical compatibility, and regulatory resilience. In this context, the current research goal of binder materials, especially for dry-processed electrodes, is not simply to replace PTFE but to reconceptualize the binder as a platform of orchestrated functionality.

Recent research illustrates how this reframing opens new directions for binder innovation. For instance, sericin, a silk-derived protein, can be fibrillated under shear to form a fine nanofibrous network capable of binding active material and conductive carbon in a sulfur cathode [19]. In dry-processed lithium–sulfur cells, sericin-based binders have demonstrated comparable capacity and cycling stability to PTFE-based systems, validating the concept of PTFE-free fibrillating binders. However, while these results are promising, they still reflect a material substitution mindset in which the functional paradigm of fibrillation-driven cohesion remains unchanged. To realize more impactful innovation, binder systems must be designed not around material analogs but around performance outcomes: mechanical robustness, particle connectivity, processing compatibility, and interfacial resilience.

A function-first framework has already begun to influence binder design in emerging studies. One notable direction is the use of hybrid or composite binder systems that decouple the mechanical and adhesive roles that are typically handled by a single polymer. For example, in graphite anodes processed via dry methods, a composite binder using PTFE for fibrillation and polyvinylidene fluoride (PVdF) for adhesion has been shown to offer both structural integrity and electrochemical durability [20]. Such task-specific combinations reflect a growing awareness that binder functionality can be distributed across multiple materials, each optimized for a distinct role in the electrode system. Other efforts have taken this idea further by developing transient or sacrificial binders. These materials serve a structural or processing function during fabrication but are removed after electrode formation. Poly(propylene carbonate) and paraffin wax have been used in melt-extrusion and hot-pressing processes to shape thick electrodes and control porosity. They are then thermally or chemically eliminated to yield mostly binder-free architectures [21, 22]. These approaches challenge the long-held assumption that binders must remain in the final device.

Reversible binders, such as low-melting polymers that soften during mixing and reset upon solidification, also offer a flexible and scalable approach to achieve similar outcomes. Phase-change-based systems or viscoelastic pressure-sensitive adhesives (PSAs) are being explored for their ability to form mechanically robust but processable architectures under moderate conditions [23]. These examples illustrate a shift in design logic away from viewing the binder as a permanent polymer to be directly replaced and toward seeing it as a dynamic, multimaterial system whose collective behavior satisfies the required electrochemical and mechanical constraints. Some of these systems still incorporate minor amounts of PTFE or PVdF, but they demonstrate that binder functionality can be deconstructed, redistributed, and ultimately reengineered.

Reconceptualizing binder design in this way is not merely an academic exercise. Scientifically, next-generation battery technologies, including solid-state cells, thick-electrode architectures, and high-rate systems, will impose increasingly specific demands on binder function [1, 24]. A binder designed from first principles rather than inherited from legacy slurry processing can be tailored to accommodate these needs through enhanced mechanical compliance, optimized interfacial contact, and compatibility with novel chemistries such as lithium metal or high-voltage cathodes [25, 26].

At the same time, regulatory alignment is no longer optional. Although some stakeholders have expressed concern about the potential impact of fluoropolymer phase-out on battery production workflows, recent developments suggest that these risks can be mitigated. Several major manufacturers have already commercialized fluoropolymer-free cathodes using water-based binders, removing both fluorinated binder and N-methyl-2-pyrrolidone (NMP) from the production line [27–29]. Proprietary alternatives, such as nanocarbon-reinforced polymers and scalable bio-derived blends, are also under active development and have begun to demonstrate performance metrics comparable to or exceeding fluoropolymer-based controls [30, 31].

Taken together, these advances indicate that function-oriented, PTFE-free dry processing is a possible strategy. In particular, dry electrode technology imposes distinct demands on binder systems: without solvent mediation, binders must ensure sufficient mechanical cohesion, maintain interfacial adhesion across heterogeneous surfaces, and support scalable processing conditions such as pressure-induced densification or shear mixing. Addressing these requirements requires moving beyond conventional material substitutions toward a function-oriented design strategy, in which each property, including cohesion, adhesion, processability, and ionic accessibility, is treated as a core design parameter rather than an incidental outcome. By redefining the binder not as a passive polymer but as a multifunctional, process-adaptive component, innovation in dry processing can simultaneously meet industrial, environmental, and regulatory imperatives, thereby securing the future of sustainable and high-performance battery systems [32].

While recent reviews have recognized the limitations of PTFE and the need for alternative binder systems, binders are often treated as secondary components [33–41]. In particular, there is a lack of systematic discussion on specific binder candidates and the functional requirements they must satisfy under a solvent-free process. To address this gap, this perspective shifts the focus toward the functional redefinition of the binder itself. Rather than evaluating processing routes or system-level strategies, we explore how polymer binders can be reconceptualized as

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modular, multifunctional platforms capable of satisfying the evolving mechanical and interfacial demands of next-generation batteries.

2 | The Role PTFE in Dry Electrode Fabrication and the Limitation

2.1 | Why PTFE Worked: Technical Foundations of Fibrillated Binder Systems

PTFE has played a central role in enabling dry electrode processing, owing to its rare ability to form fibrillar networks under mechanical stress [42]. When subjected to shear stress, PTFE transforms from spherical particles into elongated fibrous strands, which entangle active materials and conductive agents to form a cohesive, nonwoven network even with a small amount of binder (~3 wt%). This structural network not only enhances mechanical integrity but also improves electrode conductivity by minimizing the volume fraction of insulating components.

The widespread adoption of PTFE stems from its unique molecular structure. Its polymer backbone consists of strong carbon–fluorine (C—F) bonds (485 kJ mol^{-1}) [12], imparting exceptional chemical inertness and thermal stability. At the same time, this highly fluorinated architecture induces repulsive van der Waals forces between polymer chains, significantly weakening interchain cohesion. As a result, PTFE chains are prone to slippage under shear stress, enabling deformation through chain alignment and elongation, a phenomenon that drives its nanofibrillation. This chain mobility drives PTFE's nanofibrillation, aided by its intrinsically low shear strength (~0.8 MPa) and hexagonal crystal phase at room temperature (Figure 1a) [42, 43].

PTFE's fibrillation behavior can be optimized by adjusting parameters such as molecular weight, temperature, and mixing conditions. High-molecular-weight PTFE provides long, flexible chains that sustain deformation and form robust fibrous networks, whereas low-molecular-weight PTFE tends to produce brittle fibers with weak cohesion [45–47]. Processing temperature also strongly affects fibrillation through crystalline phase transitions. Between 19 and 30°C, PTFE shifts from triclinic (Phase II) to a hexagonal phase (Phase IV), which has a larger chain repeat distance (1.65 nm → 1.95 nm) and looser packing, facilitating chain sliding and nanofiber formation (Figure 1b).

These molecular and thermal attributes make PTFE particularly suited for industrial-scale roll milling and calendaring, where high shear forces naturally occur. During these processes, PTFE elongates, wrapping around active particles to form binder networks. Its ability to maintain structural integrity without a substrate further enables the fabrication of freestanding electrode sheets, advantageous for lamination- and stacking-based manufacturing.

2.2 | Why PTFE Must be Eliminated From Dry Electrode Processing

PTFE has played a pivotal role in the development of dry electrode technology, owing to its unique ability to form fibrillar networks under mechanical shear stress. This property has made PTFE the standard binder for solvent-free electrode fabrication. However, its advantages are increasingly overshadowed by fundamental limitations. These include electrochemical degradation, poor ionic conductivity, interfacial instability at both electrodes, and process-related limitations. Collectively these issues underscore a growing mismatch between PTFE and the needs of scalable and sustainable battery manufacturing. As dry processing transitions from lab-scale to industrial implementation, continued reliance on PTFE is no longer viable. A material once considered essential must now be critically reassessed and ultimately replaced. The following section outlines four reasons why PTFE should be phased out from dry electrode systems, especially with a focus on function-oriented design and why this transition is not optional but essential for advancing next-generation LIBs.

2.2.1 | PTFE Decomposition Undermines Electrochemical Stability at Both Electrodes

Although PTFE is often described as chemically inert, it undergoes electrochemical degradation under the full range of conditions encountered in LIBs. Its low-lying lowest unoccupied molecular orbital (LUMO) makes it vulnerable to reduction at potentials below 0.5 V vs. Li/Li^+, a regime commonly accessed during the lithiation of graphite anodes (Figure 1c) [48–50]. During initial cycles, PTFE undergoes reductive cleavage, yielding lithium fluoride (LiF) and unsaturated carbonaceous species, as exemplified by the following reaction as reported by Shiraishi et al. [51]:

$$-(CF_2-CF_2)_n + 4n\ Li \rightarrow -(C\equiv C)_n + 4n\ LiF. \quad (1)$$

This irreversible decomposition reduces initial Coulombic efficiency and contributes to long-term capacity fade through binder detachment and microcracking. These issues are further exacerbated in high-capacity anode systems such as silicon oxide (SiO_x), where repeated volume expansion imposes additional stress on the binder matrix.

On the cathode side, PTFE shows signs of oxidative degradation at high voltages above 4.3–4.5 V, even if more subtle than at the anode. Its lack of chemical affinity for the cathode surface, especially in metal–oxide lattices, leads to weak adhesion and increases the risk of cathode electrolyte interphase (CEI) delamination [52, 53]. Additionally, PTFE does not provide protective functional groups against transition metal dissolution (e.g., Ni^{2+}) or HF attack, unlike some functional binders that incorporate chelating groups. This can result in CEI instability and surface reconstruction of cathode particles.

2.2.2 | PTFE's Poor Wettability Restricts Electrolyte Interaction and Ion Transport

The molecular structure of PTFE consists of a fully fluorinated carbon backbone, which renders its surface highly hydrophobic and chemically nonpolar [54]. This leads to poor wettability with conventional liquid electrolytes, which are typically carbonate-based and polar. As a result, PTFE-coated or PTFE-containing dry electrodes exhibit high contact angles with electrolyte, slowing electrolyte infiltration and wetting, especially in thick or high-areal-loading electrodes. Incomplete wetting introduces ionic transport bottlenecks and uneven formation of the CEI or solid electrolyte interphase (SEI), which in turn exacerbate local

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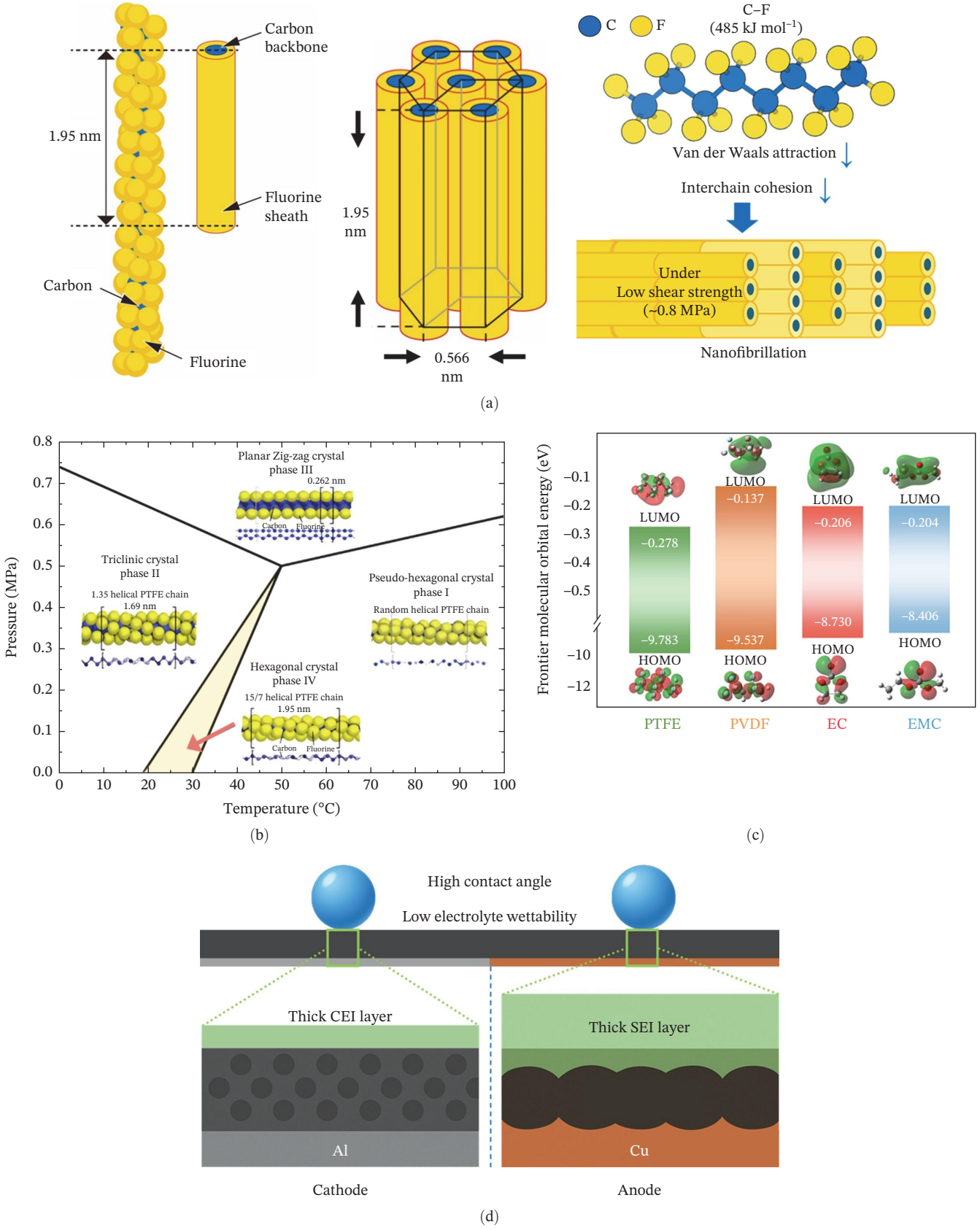
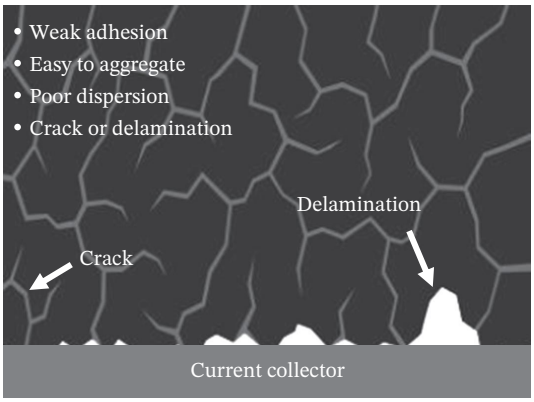
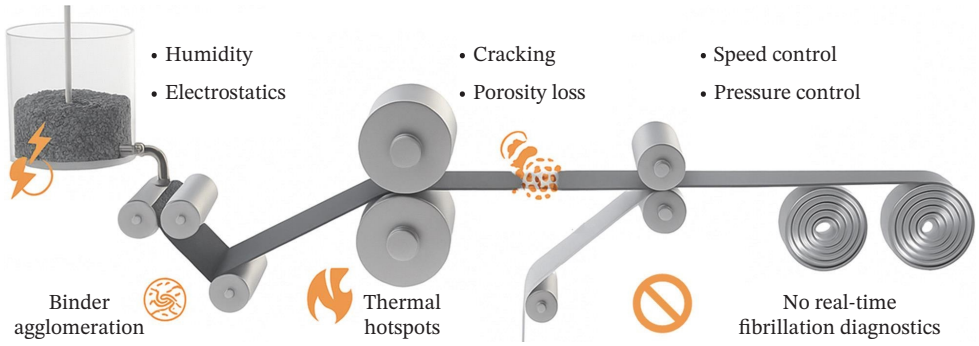


FIGURE 1 | (Continued)

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(e)



(f)

FIGURE 1 | (a) Molecular structure of PTFE and shear-induced nanofibrillation mechanism. Reproduced with permission [42]. Copyright 2022, Elsevier B.V. (b) Phase transition of PTFE. Reproduced with permission [43]. Copyright 2021, Elsevier B.V. (c) Comparison of frontier molecular orbital energies, including the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), for PTFE and representative binders and solvents, highlighting the low reactivity of PTFE. Reproduced with permission [44]. Copyright 2024, Elsevier Inc. (d) Schematic illustration of poor wettability in PTFE-based electrodes, which induces thick and inhomogeneous CEI/SEI layer formation. (e) Crack and delamination caused by poor adhesion and uneven dispersion of PTFE within the electrode composite. (f) Limitations of PTFE scalability in industrial-scale dry electrode manufacturing.

impedance and accelerate degradation (Figure 1d) [55–58]. At the anode, poor wetting delays SEI stabilization and leaves regions of exposed graphite vulnerable to parasitic reactions. On the cathode, particularly those with large secondary particles like $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (NCM811), insufficient electrolyte contact can lead to heterogeneous lithiation, internal cracking, and loss of active surface area. While functional additive binders such as poly(acrylic acid) (PAA), carboxymethyl cellulose (CMC), or polyvinylpyrrolidone (PVP) have been used to compensate, they are often insufficient to overcome the dominant hydrophobicity of PTFE. Additionally, such functional additive binders may only be partially distributed due to PTFE's tendency to aggregate, further compounding the issue of wetting heterogeneity. Ultimately, poor electrolyte–electrode interaction reduces rate capability and increases the risk of lithium plating or surface passivation under fast-charging conditions.

2.2.3 | Poor Adhesion and Dispersion Complicate Electrode Architecture

Another fundamental limitation of PTFE lies in its inherently weak adhesion properties [59–61]. It lacks polar functional groups such as hydroxyl or carboxyl moieties that could form hydrogen bonds or ionic interactions with current collectors (Al and Cu) or active material surfaces [15, 62, 63]. Consequently, electrodes formulated with PTFE exhibit poor mechanical adhesion to both the conductive foil and the bulk composite layer. This issue is particularly acute during calendaring, where delamination may occur under pressure, or during long-term cycling, when interfacial stresses accumulate. To compensate for this, additional adhesive agents are currently employed, but they may increase interfacial resistance and add complexity to the manufacturing process [44, 64].

In addition to poor adhesion, PTFE tends to aggregate during dry mixing due to its low surface energy and nonpolar nature [65, 66]. This results in heterogeneous dispersion within the electrode matrix, with binder-rich and binder-poor regions. Such heterogeneity can lead to localized overinsulation or loss of mechanical integrity. Regions deficient in binder may crack or delaminate, while binder-rich zones may block ion conduction or reduce electronic percolation (Figure 1e). These nonuniformities hinder reproducibility, scale-up, and process consistency in large-format electrodes. Such workarounds undermine the very sustainability and simplicity that dry processing aims to achieve.

2.2.4 | Processing Sensitivities and Scalability Challenges of PTFE in Dry Electrode Fabrication

PTFE readily undergoes fibrillation under mechanical shear, forming an entangled nanofiber network within dry-processed

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electrodes [42]. However, in large-scale manufacturing, controlling the uniformity and extent of fibrillation across wide-area electrodes presents a significant challenge. Small variations in calendaring pressure, temperature, or mixing energy can lead to inconsistent fibril formation, which in turn causes inhomogeneous binder distribution, poor mechanical integrity, or loss of porosity. These effects are particularly problematic for thick or brittle electrodes such as high-Ni $LiNiCoMnO_2$ (NCM) [67, 68]. Moreover, the nonthermoplastic and viscoelastic nature of PTFE limits its adaptability to high-throughput R2R processes, where precise control of pressure and line speed is required to prevent thermal hotspots and ensure consistent microstructure formation. The absence of real-time diagnostics for fibrillation further complicates process stabilization. In addition, PTFE exhibits sensitivity to environmental factors such as humidity, electrostatic charge accumulation, and triboelectric effects during powder handling and mixing, which can undermine process reliability and reproducibility at scale. Consequently, these factors constrain the process window of PTFE and underscore its limited scalability in mass production environments where uniformity, tolerance, and operational robustness are critical (Figure 1f).

3 | Requirements for Function-Oriented Binders for Dry Electrode

Given the critical limitations of PTFE outlined in the previous section, including its electrochemical instability, poor wettability, adhesion issues, and process sensitivity, there is growing consensus that a transition away from PTFE is both necessary and urgent (Figure 2a). This shift, however, is not merely a matter of material substitution. It requires a fundamental redefinition of what a binder is expected to do in dry-processed electrode systems. As the dry electrode community gradually transitions from PTFE-dominant systems to other function-oriented binder configurations, the role of binders is no longer auxiliary. It becomes architecturally central. The binder that replaces PTFE must absorb multiple responsibilities, ensuring cohesion, facilitating ion and electron transport, stabilizing interfaces, and withstanding mechanical and thermal stress during processing and cycling. Importantly, these roles must be fulfilled under solvent-free conditions, where molecular mobility is limited, and material interactions are driven predominantly by mechanical force and interfacial affinity. Furthermore, the binder must also demonstrate compatibility with R2R processes to enable electrode mass production. While it is important to address the limitations and issues of existing binders, their feasibility for actual production is equally critical given the close connection of this field to industrial applications [33, 35]. This section outlines five critical requirements that function-oriented binders for PTFE-free dry electrode systems must meet to enable high-performance and scalable manufacturing.

3.1 | Mechanical Cohesion and Interfacial Adhesion

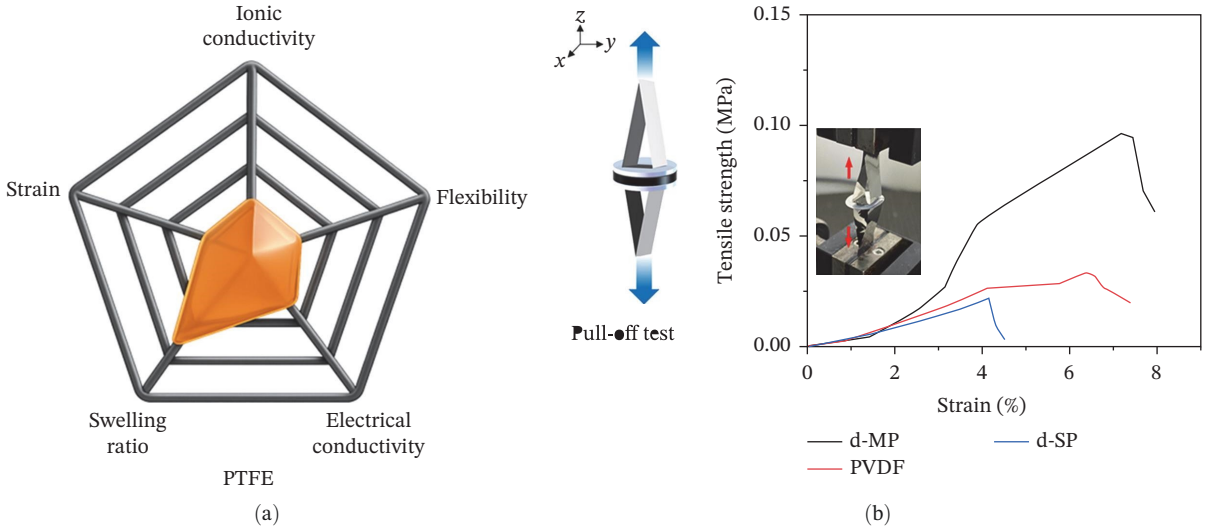
A primary function of any binder system is to preserve the mechanical integrity of the electrode, both within the network of active and conductive particles and at the interface between the electrode and current collector (Figure 2b), because mechanical failure ultimately leads to severe capacity degradation. The enhancement of mechanical properties is generally attributed to the improved uniformity and the elastic behavior of the binder within the electrode structure [74]. In slurry-based electrodes, such mechanical robustness is well established due to capillary forces generated during solvent evaporation, which aid in the initial cohesion of particles [4]. However, dry-processed electrodes lack such assistance and must instead rely exclusively on binder-mediated mechanical entanglement and interparticle adhesion. PTFE, in particular, imparts this functionality through in situ fibrillation under shear, creating an interwoven fibrous mesh that physically locks particles into a cohesive network [75–77].

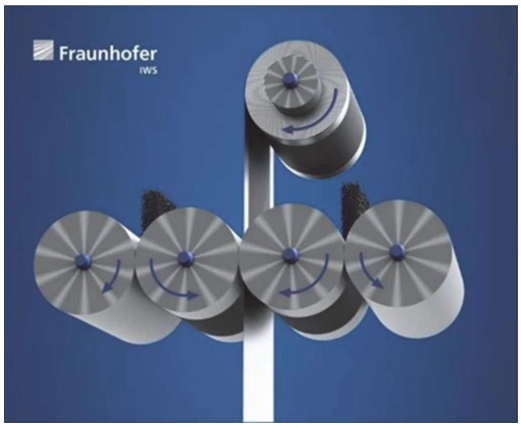
Upon substitution of PTFE, binders must compensate for the loss of fibrillar cohesion through alternative interaction mechanisms. One effective strategy involves the use of polymeric additive binders bearing multiple functional groups, such as hydroxyl, carboxyl, or amine moieties, which can engage in hydrogen bonding, coordinate with surface oxides on cathode particles, or interact electrostatically with native SEI components on anode [1, 15, 61]. Recent studies have shown that hydrogen bonding interactions can remain effective even in solvent-free dry-processed electrodes. For example, Sung et al. [59] introduced PAA as a hydrogen-bonding co-binder in PTFE-based dry cathodes, reporting significantly enhanced interfacial adhesion and reduced contact resistance, despite the absence of solvent. This behavior was attributed to the formation of hydrogen bonds between PAA and the surfaces of cathode particles and current collectors, as confirmed through interfacial adhesion and mechanical testing under dry conditions. Such hydrogen bonding strategies have also been successfully applied in graft copolymers combining PAA and CMC, which demonstrate improved mechanical robustness, attributed to their hybrid backbone rigidity and multivalent interfacial adhesion [53]. Further enhancement can be achieved by incorporating branched or hyperbranched polymer architectures, which increase the effective contact area per macromolecule and facilitate improved particle wetting and network uniformity [78]. These architectures also promote better packing and stress dissipation during calendaring, thus maintaining electrode integrity over prolonged cycling [79]. In parallel, the adhesion between the electrode layer and current collector remains a critical consideration. PTFE's inherent chemical inertness results in limited interfacial affinity toward common metallic substrates such as copper and aluminum, often leading to delamination under mechanical or electrochemical stress. Function-oriented binder possessing polar functional groups, such as imide, sulfonate, or carbonyl moieties, can engage in dipole–dipole interactions or hydrogen bonding with metal surfaces, thereby reducing interfacial resistance and enhancing mechanical durability [1].

3.2 | Elasticity and Structural Flexibility

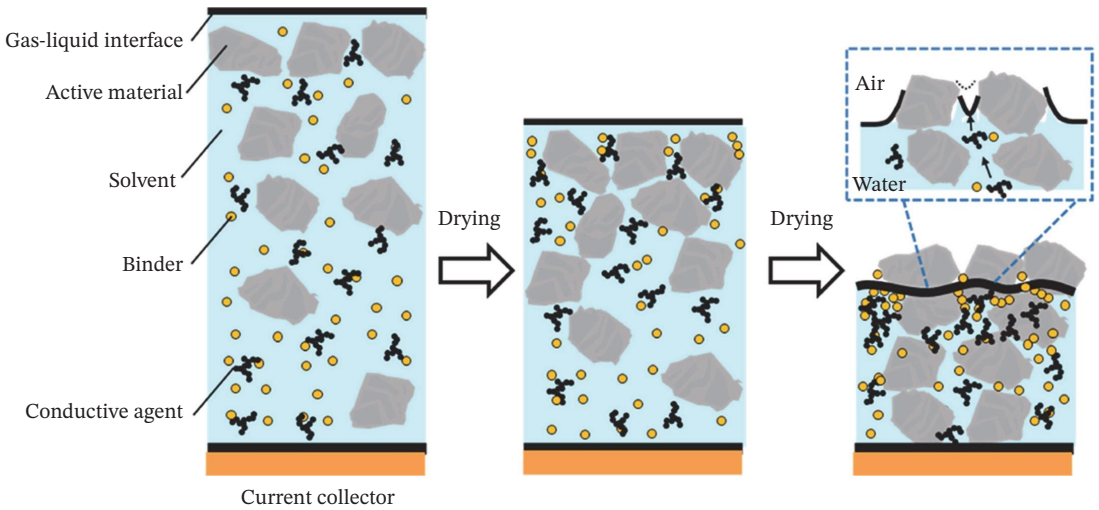
Electrode integrity is continually challenged by mechanical stress during calendering, cycling-induced volume changes, and thermal fluctuations. In dry electrode processing, particularly within high-throughput R2R fabrication lines, electrodes must withstand severe mechanical loading such as continuous bending and rolling stresses encountered as they pass through multiple roller assemblies (Figure 2c). Binders must therefore exhibit a critical balance of stiffness to maintain the structural architecture and high elasticity to accommodate these dynamic processing strains without mechanical failure, such as delamination or cracking. To meet these demands, hybrid binders can be used

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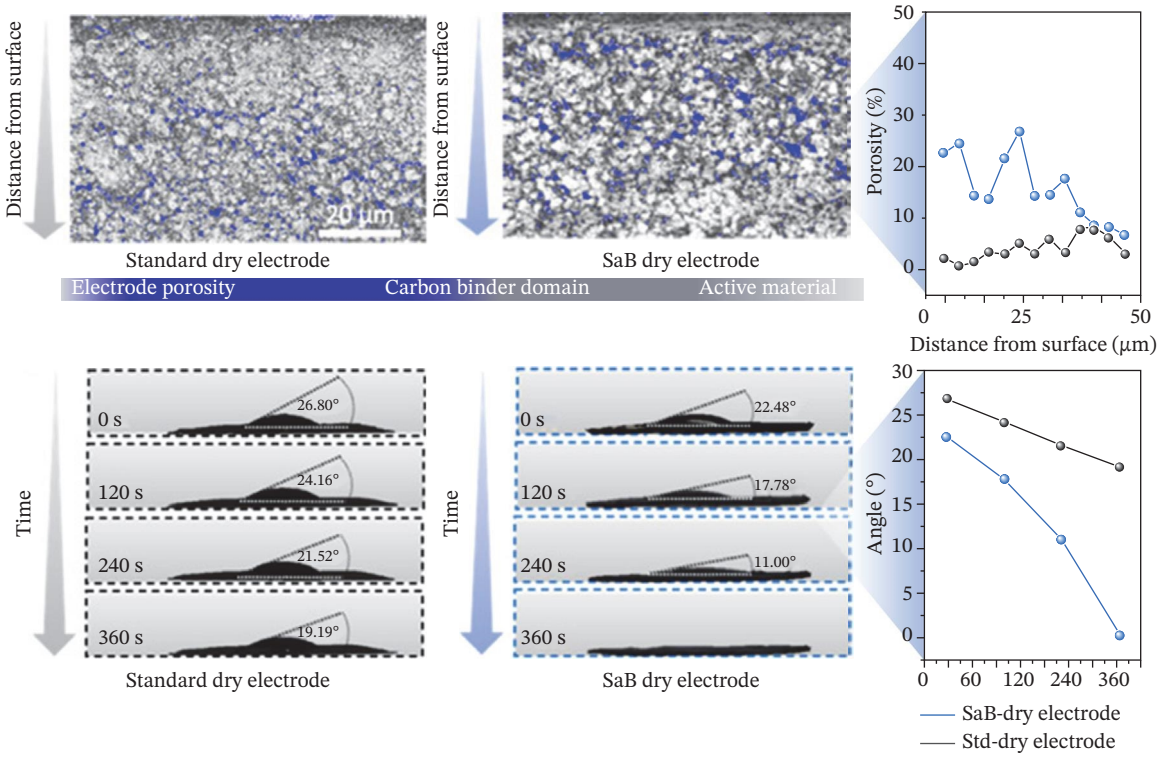
(c)



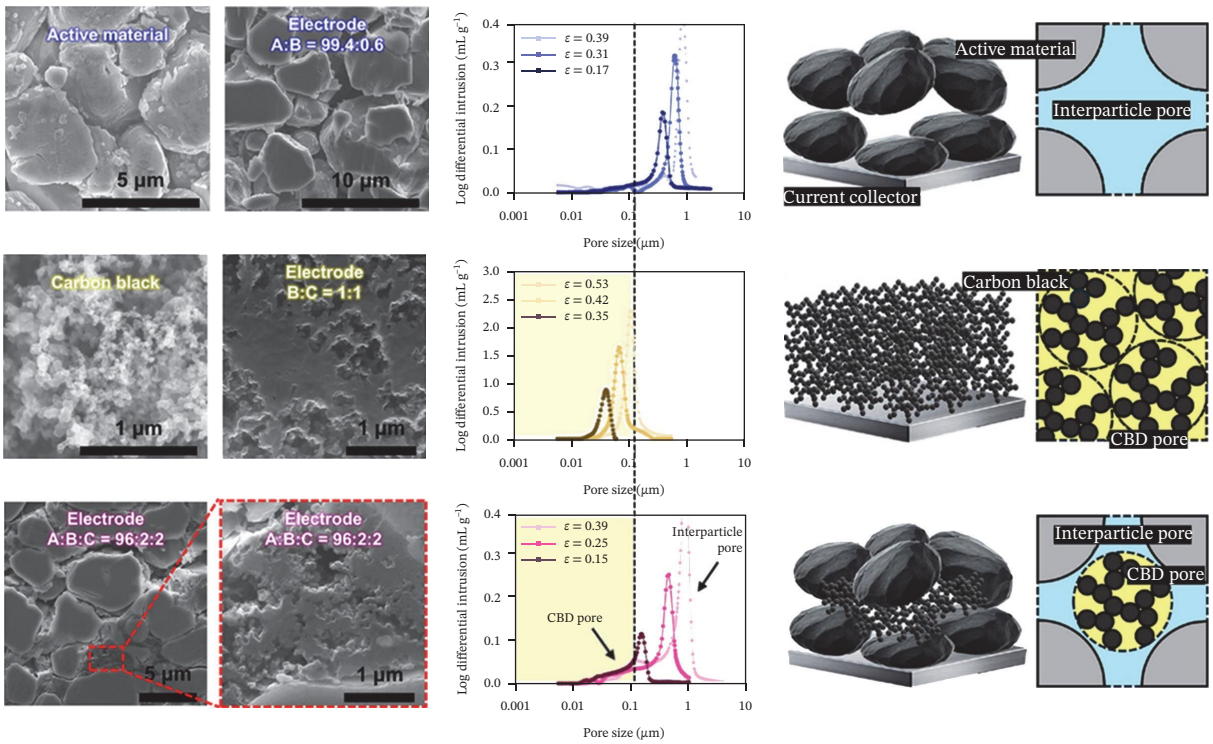
(d)

FIGURE 2 | (Continued)

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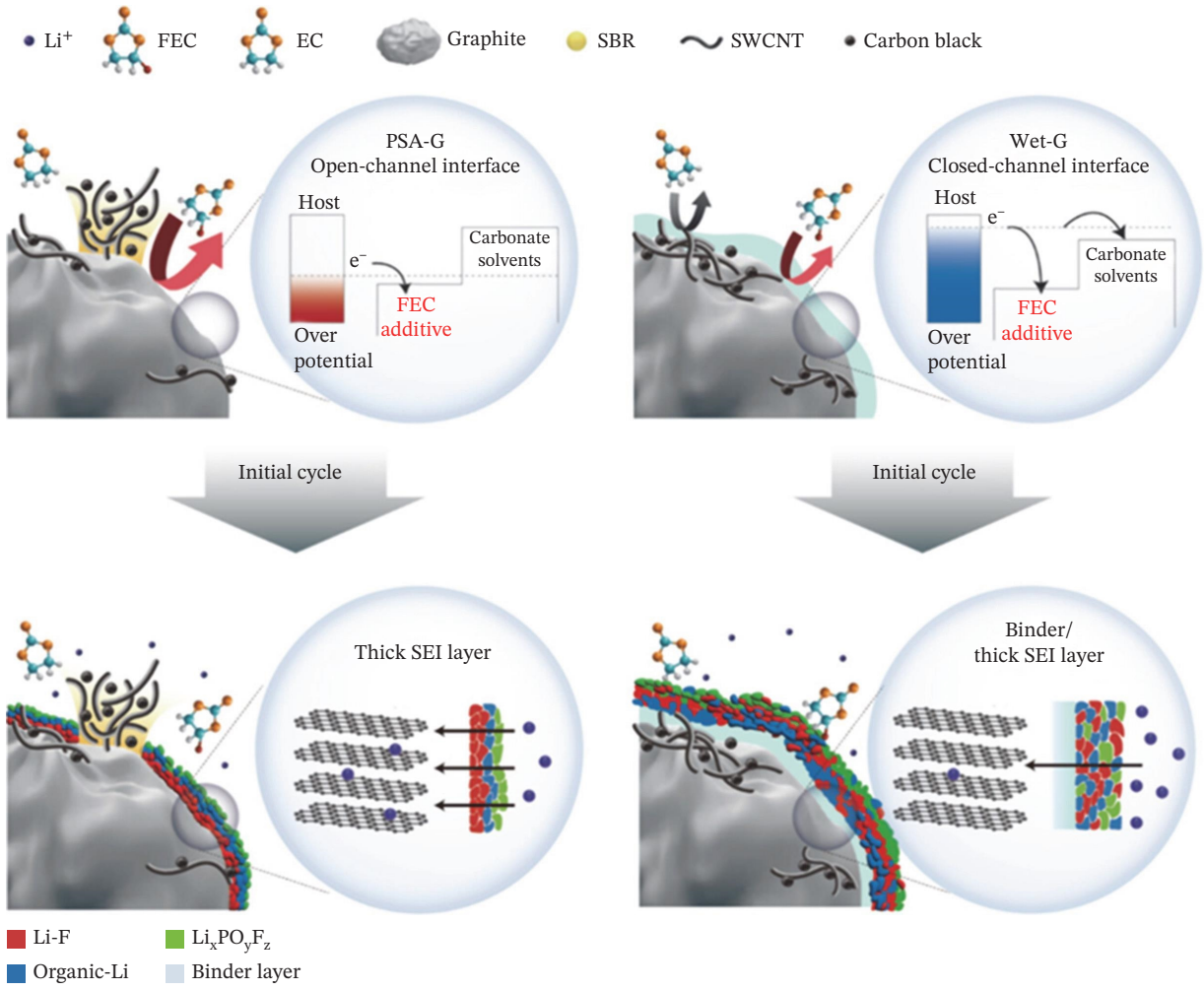
(e)



(f)

FIGURE 2 | (Continued)

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(g)

FIGURE 2 | (a) Radar chart summarizing the overall performance of PTFE binders across key parameters. Reproduced with permission [53]. Copyright 2025, Wiley-VCH GmbH. (b) Mechanical cohesion and interfacial adhesion for maintaining structural integrity under solvent-free processing and ensuring strong contact with current collectors. Reproduced under the terms of the CC-BY 4.0 license. Copyright 2023, Ryu et al. [69], published by Springer Nature Ltd. (c) Elasticity and structural flexibility to endure continuous bending and rolling stresses inherent in high-throughput roll-to-roll (R2R) processing and to accommodate mechanical strain during electrochemical cycling. Reproduced with permission from [70]. Copyright 2023, Elsevier. (d) Structural instabilities and nonuniform component distribution caused by capillary-driven binder migration during slurry drying. Reproduced with permission from [71]. Copyright 2023, Elsevier B.V. (e) Ionic conductivity and electrolyte affinity to facilitate Li^+ transport in electrodes and enhance wettability. Reproduced with permission from [72]. Copyright 2024, Elsevier. (f) Hierarchical pore distribution and dual-scale architecture characterizing the coexistence of large interparticle and fine carbon-binder domain (CBD) pores to optimize ion transport pathways. Reproduced with permission [73]. Copyright 2025, Wiley-VCH GmbH. (g) Electrochemical and interfacial stability to suppress parasitic reactions and stabilize CEI/SEI structures under cycling. Reproduced with permission from [23]. Copyright 2025, American Chemical Society.

by combining rigid domains (e.g., cellulose nanofibers or aramids) with soft segments (e.g., PEG or rubbery chains), creating strain-dissipating networks. These properties are especially vital for anodes composed of alloying materials like silicon, where volumetric expansion can exceed 300%. In such systems, binders with sacrificial hydrogen bonds, reversible ionic crosslinks, or dynamically reconfigurable chain segments can prevent cracking and delamination [15]. For example, PAA chains that undergo physical entanglement can unwind and re-entangle under cyclic strain, providing recoverable elasticity [80]. The introduction of zwitterionic moieties is also being explored to create "slideable" networks that can dissipate stress without permanent damage [81]. Rigid binders with high T_g are prone to brittle fracture or particle detachment during these steps, whereas binders with a lower T_g provide the ductility and elasticity necessary to accommodate compressive and shear stresses without compromising electrode integrity [82, 83]. Since dry-processed electrodes can also experience local heat buildup during calendaring or fast charging, binders must maintain their mechanical resilience across a broad temperature range. Therefore, binders with a soft glass transition (T_g) within 0–40°C are preferred, as they remain sufficiently elastic to absorb and disperse stress during both fabrication and battery cycling, while simultaneously ensuring adequate adhesion and structural cohesion for reliable dry electrode performance.

3.3 | Ionic Conductivity and Electrolyte Affinity

In thick electrodes, through-plane lithium-ion transport across the porous electrode is the rate-limiting step [84, 85]. To fully

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leverage the advantages of high-loading dry electrodes, effective through-plane wetting is essential to activate the entire depth of the electrode [86]. As shown in Figure 2d, conventional slurry-cast electrodes are inherently prone to capillary-driven binder migration during solvent evaporation, which leads to vertical nonuniformity and structural instabilities that block efficient ion transport [71]. While dry processing is fundamentally free from such solvent-driven migration issues, function-oriented binders must still address other critical hurdles to effectively enable high-performance thick electrodes. Specifically, PTFE binder systems suffer from hydrophobic barriers that restrict electrolyte uptake and ion diffusion [54]. The electrically insulating properties of PTFE provide no support for ionic or electronic conductivity, further slowing charge transport during operation. To overcome PTFE's limitation, function-oriented binder must actively support electrolyte wetting and facilitate through-plane transport (Figure 2e). This is often achieved through the inclusion of polar or ionic functionalities, such as ether, carboxylate, sulfonate, or carbonate groups, which can interact favorably with liquid electrolytes [15, 87]. Polymers incorporating ethylene oxide units, which are representative of ether functional groups, can form transient coordination complexes with lithium ions, supporting Li^+ transport via segmental motion [88, 89]. These functionalities also lower the interfacial contact angle, improving electrolyte wetting kinetics and minimizing the soaking time required during cell manufacturing.

Beyond these chemical modifications, researchers are actively investigating ways to optimize transport pathways by precisely managing the interactions between electrode components. Recent research has demonstrated that regulating the interfacial interactions and ratios between the binder and conductive agents can optimize the carbon-binder domain (CBD) to enhance electrolyte affinity in dry-processed thick electrodes (Figure 2f) [73]. Transport pathways can be further optimized by utilizing conductive agents with high aspect ratios to prevent structural collapse during dry consolidation while simultaneously controlling calendaring parameters, such as roll temperature and line pressure, to regulate pore volume and avoid excessive densification that would otherwise impede electrolyte penetration in high-loading films [90, 91]. In addition to enhancing ionic transport, some functional additive binder systems have explored the integration of conductive additives or intrinsically conductive polymers to support electronic percolation within the electrode film [92, 93]. While the binder's primary function remains structural, its capacity to support charge transport, either by minimizing impedance or facilitating uniform electrolyte distribution, has become increasingly important for high-performance designs.

3.4 | Electrochemical and Interfacial Stability

As the binder's role in dry electrode systems expands beyond mechanical cohesion, its ability to influence electrode–electrolyte interfacial behavior is gaining increasing importance. In both cathodes and anodes, interfacial degradation, driven by parasitic reactions, gas evolution, or metal dissolution, can compromise long-term performance and safety. A growing body of research suggests that functional binders can serve as active stabilizers at these interfaces, shaping the composition, structure, and transport properties of the CEI and SEI (Figure 2g).

In high-voltage cathodes, oxidative decomposition of electrolyte components and transition metal dissolution are key degradation pathways that accelerate capacity fading and promote cathode–anode crosstalk. For example, functional groups in the binder could scavenge HF generated from electrolyte degradation or trap transition metal ions (like Ni^{2+}) leaching from the cathode, thereby protecting the cathode surface and current collector and mitigating crosstalk from the dissolved metal ions. For example, CMC has demonstrated HF-neutralizing capabilities by coordinating with fluorine-containing species, thereby alleviating acid-driven degradation pathways in phosphate-based cathodes [94]. Similarly, Ma et al. [95] reported a biomass-derived binder containing electron-rich moieties capable of scavenging free radicals and capturing dissolved transition metal ions, significantly enhancing the interfacial stability of 5 V-class $LiNi_{0.5}Mn_{1.5}O_4$ cathodes under oxidative conditions.

On anodes, excessive SEI growth and unstable interfacial reactions remain key contributors to capacity fade and low coulombic efficiency. To mitigate these issues, certain binders are molecularly engineered to undergo controlled electrochemical decomposition at predefined potentials, thereby forming artificial interphases with optimized ionic conductivity and mechanical resilience. For instance, the use of PVP as a binder in dry-processed anodes has been shown to promote the formation of a stable, Li_3N-rich SEI layer, resulting in enhanced interfacial stability, accelerated lithium-ion transport, and improved long-term cycling performance [96].

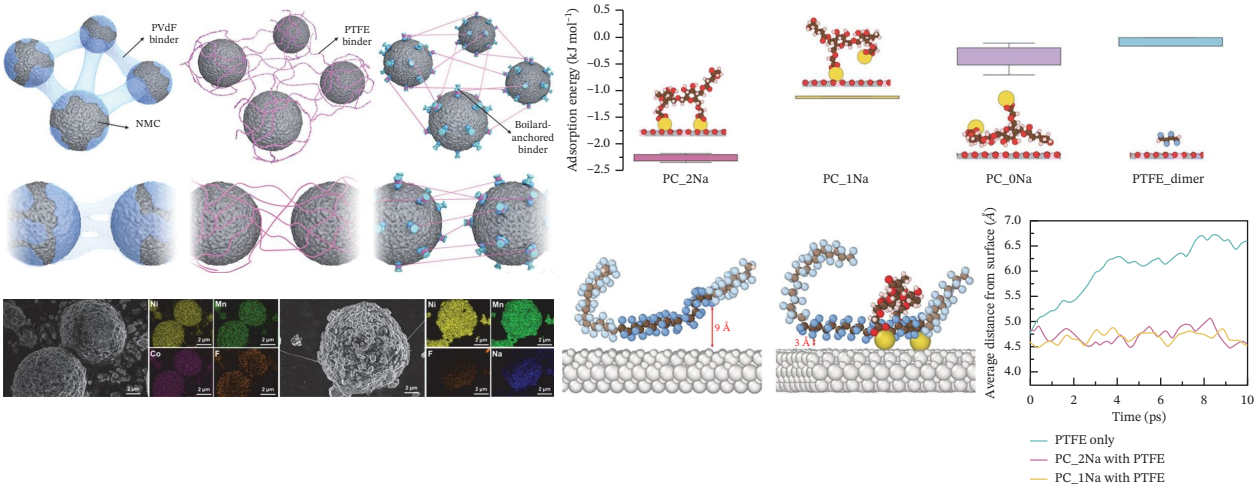
3.5 | Dispersion Control and Morphological Uniformity

Binders must assume a proactive role in promoting morphological uniformity across the electrode film. Compared to conventional slurry-based methods, solvent-free dry processing presents intrinsic challenges in achieving uniform dispersion, especially for nanoscale conductive agents which exhibit a strong tendency to aggregate. Since PTFE can undergo excessive fibrillation during prolonged mixing, potentially compromising its binding functionality, precise control over the mixing environment is essential [97].

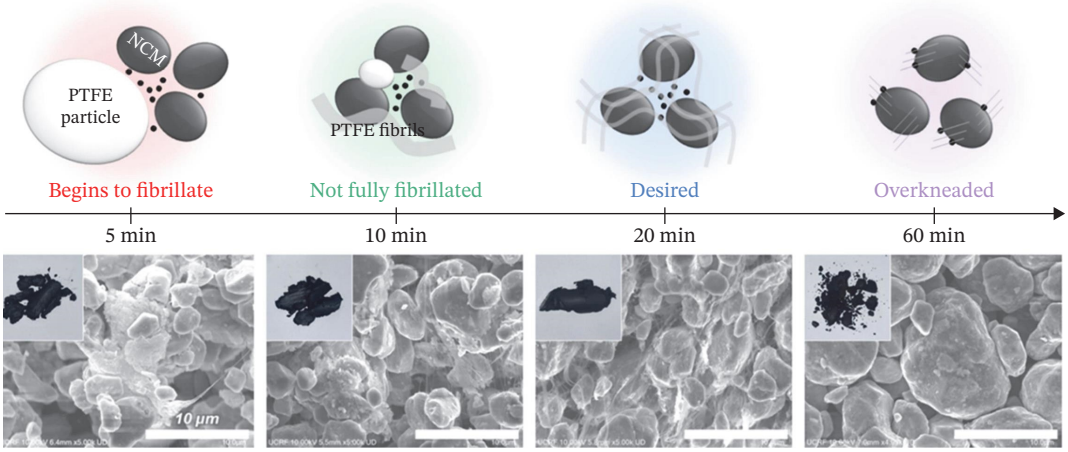
To address these challenges, various strategic approaches have been developed to balance dispersion quality with binder integrity. From a material properties perspective, managing the binder's physical state, such as mixing above its glass transition temperature (T_g), can enhance spreading and contact before the onset of intense fibrillation [98]. Furthermore, tailoring the interface between the binder and active materials provides another innovative avenue; for instance, anchoring bollard-type functional binders onto the surface of active materials can significantly increase the density of available anchoring sites, thereby promoting robust interactions and facilitating more uniform component distribution during solvent-free processing (Figure 3a) [53].

In addition to these material-level considerations, recent research has emphasized the necessity of a systematic sequence involving individual unit processes. This includes granule formation (encompassing mixing, kneading, and grinding), film formation, calendaring, and lamination, each designed with specific functions to maintain a homogeneous particle distribution and ensure structural integrity (Figure 3b) [97]. Building upon these foundational steps, the timing and sequence of component

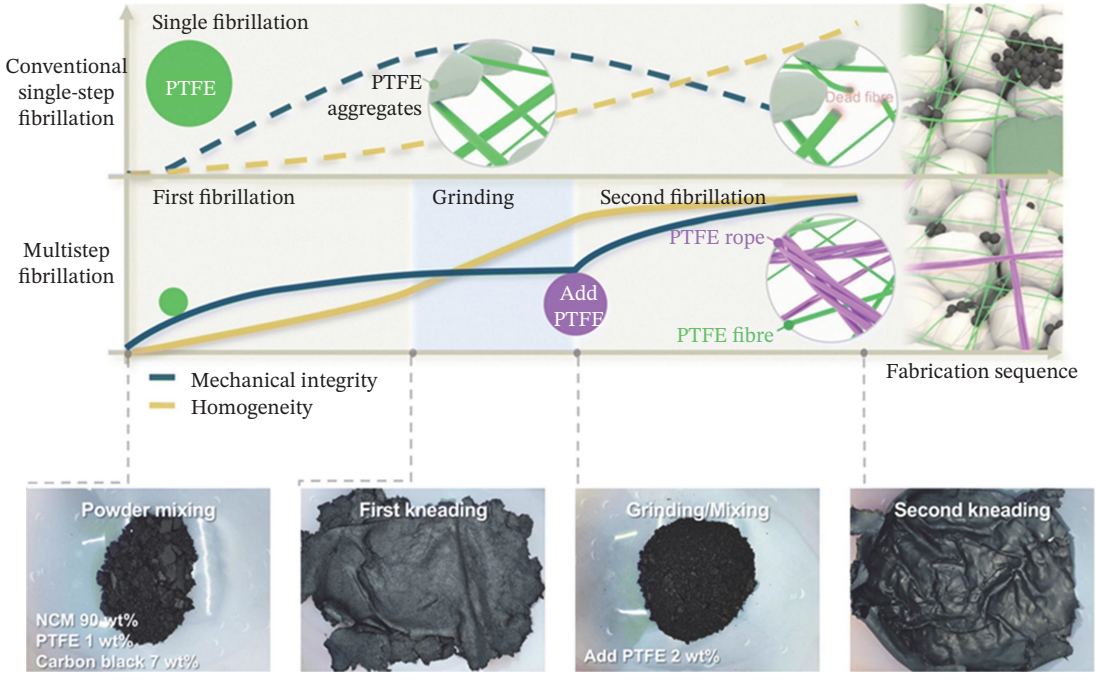
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(a)



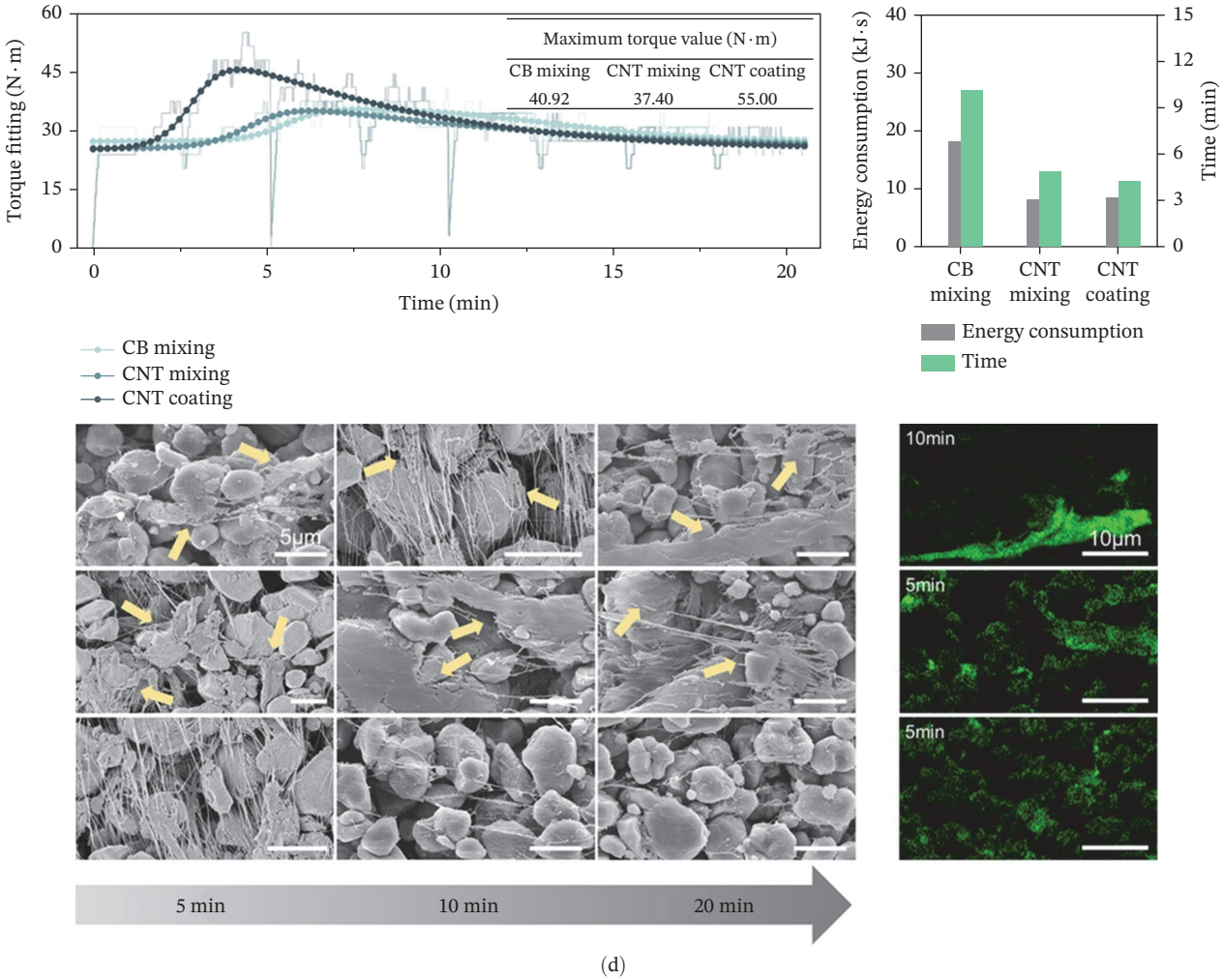
(b)



(c)

FIGURE 3 | (Continued)

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(d)

FIGURE 3 | (Continued)

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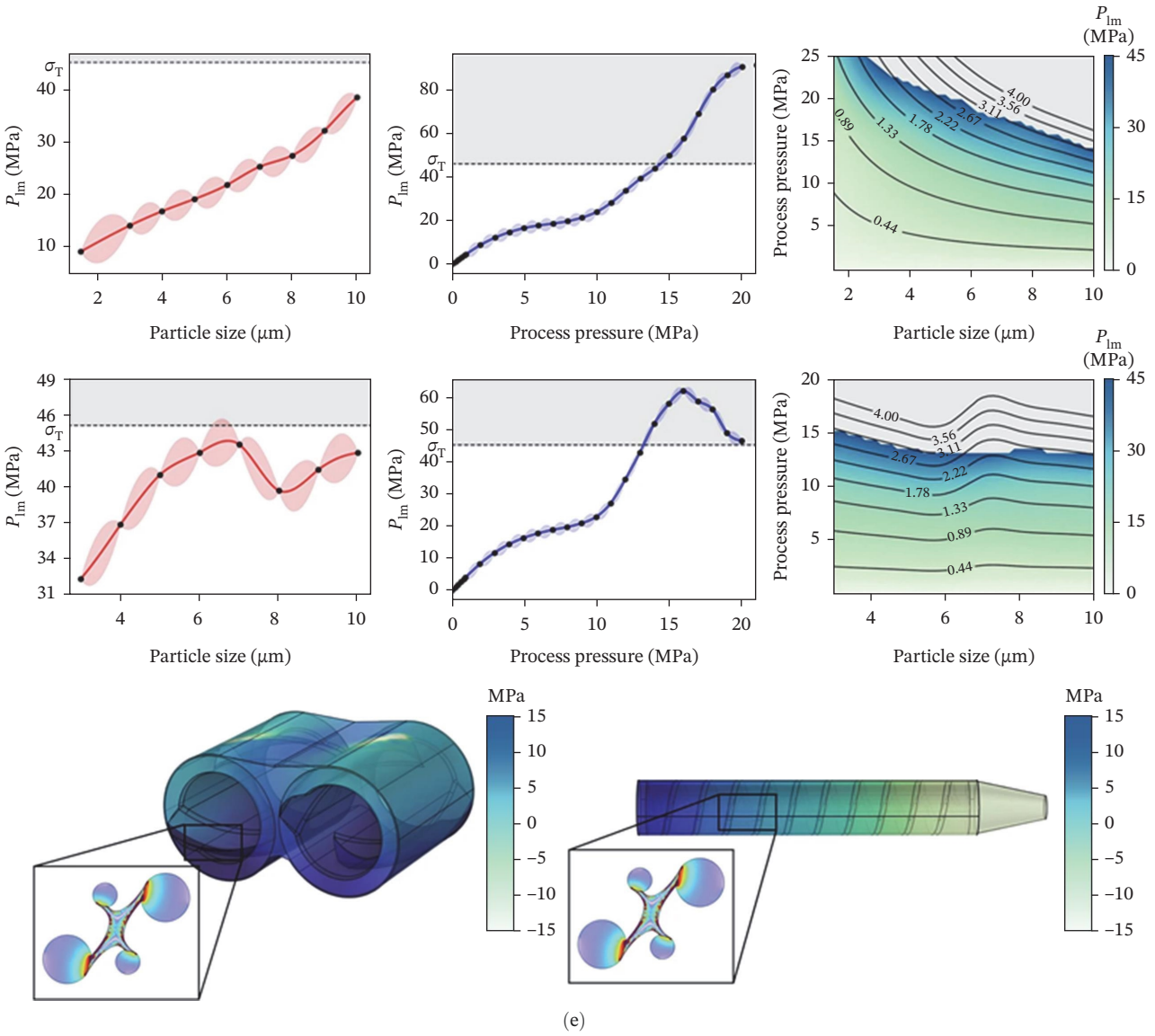


FIGURE 3 | (a) Bollard-anchored functional binders increasing anchoring sites on active material surfaces to promote robust component interactions and suppress particle agglomeration. Reproduced with permission from [53]. Copyright 2025, Wiley-VCH GmbH. (b) Dispersion control is essential to prevent the agglomeration of conductive agents, ensure morphological uniformity, and maintain homogeneous particle distribution throughout the electrode film. Reproduced with permission from [97]. Copyright 2024, Elsevier. (c) Dispersion control achieved through multistep mixing, where controlled grinding and kneading regulate PTFE fibrillation to improve morphological uniformity under solvent-free processing. Reproduced with permission from [99]. Copyright 2025, Royal Society of Chemistry. (d) Dispersion control optimized by regulating kneading duration, with torque evolution acting as a key indicator of the fibrillation state and resultant component uniformity. Reproduced with permission from [100]. Copyright 2025, Wiley-VCH GmbH. (e) Dispersion control refined by optimizing particle size and processing pressure through Bayesian optimization and FEM modeling. Reproduced under the terms of the CC-BY 4.0 license Copyright 2025, Kang et al. [101], published by Springer Nature.

integration have proven equally critical. For instance, dividing the binder addition into multiple stages during multistep mixing has been shown to significantly enhance the mechanical and electrochemical properties of dry electrodes by ensuring more uniform component integration and preventing premature binder degradation (Figure 3c) [99]. Furthermore, the kinetics of this process can be monitored in real-time; for instance, torque evolution during kneading serves as a key physical indicator of the binder's fibrillation state, allowing researchers to optimize the kneading duration to reach the desired level of component uniformity (Figure 3d) [100].

Complementing these iterative experimental strategies, data-driven and computational frameworks have been proposed to further refine these processes. Recent advancements have integrated Bayesian optimization and finite element method (FEM) modeling to precisely optimize particle size and processing pressure (Figure 3e) [101]. These tools allow for a predictive understanding of how localized stress distribution affects dispersion, moving the field toward a more scientific basis for process control. From this perspective, dispersion of uniformity in dry electrodes should not be treated solely as a consequence of increased mixing intensity or prolonged kneading. Instead, effective dispersion

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requires binder-mediated control over particle separation and redistribution under shear, while avoiding excessive structural degradation. Binder systems that can support uniform dispersion during solvent-free mixing, without relying on aggressive fibrillation, are therefore essential to suppress phase segregation and maintain morphological homogeneity throughout electrode fabrication.

4 | Future Directions for PTFE-Free Binder Architecture

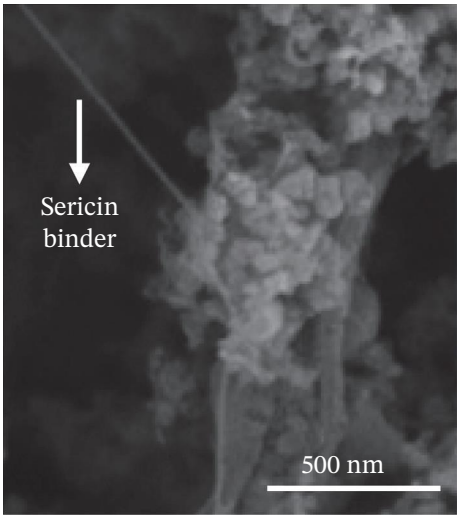
The development of PTFE-free binder systems remains a central challenge for the advancement of dry electrode processing. The complete removal of fluorinated components, particularly PTFE, requires a fundamentally new approach to binder design. Currently investigated fluorine-free binder candidates include high-molecular-weight rubber-based polymers [23], thermoplastic elastomers [22, 98], and aliphatic polycarbonates [21]. These materials exhibit favorable mechanical properties such as toughness and elasticity, and early studies suggest that they possess the potential to support dry electrode fabrication under solvent-free conditions. In addition, natural polymers capable of forming fibrous or mesh-like structures in situ have attracted growing attention, although they may require additional processing steps such as surface modification or moisture mediation. Notably, recent studies have demonstrated that PTFE-free binders can offer not only sufficient mechanical integrity but also structural and electrochemical performance advantages when designed from a function-oriented perspective. Beyond these systems, other nonfluorinated binder concepts, including self-healing polymer networks [102], electronically conductive binders [103], and high-temperature engineering polymers such as polyimides, have also been proposed [104, 105]. While these fluorine-free binder categories are conceptually attractive for dry electrode processing, systematic experimental validation under solvent-free, R2R-compatible conditions remains limited. Accordingly, this section focuses on PTFE-free binder systems for which experimental viability under scalable dry processing conditions, ranging from high-pressure calendaring to R2R assembly, has been demonstrated.

Schmidt et al. [19] developed a fully fluorine-free, dry-processed cathode using sericin, a biodegradable silk byproduct, as the binder (3 wt%) Under mechanical shear, sericin undergoes fibrillation similar to PTFE, forming a nanofibrous network that maintains structural cohesion within the electrode (Figure 4a). It makes a spiderweb-like structure connecting individual carbon aggregates. Mechanistically, the polypeptide backbone of sericin, rich in —OH and —COOH groups, forms hydrogen bonds with both the native Al_2O_3 layer on the current collector and the surfaces of sulfur and carbon particles. These interactions enable a PTFE-free binder system based solely on sericin to achieve an average peel strength of ~4.9 N cm^{-2}, sufficient for R2R processing and mechanical handling (Figure 4b). Although this is lower than the 7.6 N cm^{-2} achieved by PTFE-based electrodes, the performance of sericin is notable considering its current level of molecular weight, fibril morphology, and lack of formulation optimization. The polar functional groups of sericin also enhance interfacial interactions with the electrolyte, improving wettability and supporting more uniform ion transport. Therefore, sericin exhibits mechanical resilience under swelling-induced stress, suggesting that its polar surface chemistry modulates interactions with the electrolyte to mitigate performance loss associated with volume change. This facilitates even distribution of Li_2S discharge products and contributes to a stable Coulombic efficiency of 97.9% over 50 cycles (Figure 4c). The uniform deposition of Li_2S supports consistent electrochemical reactions and improved utilization of active materials. Under various electrolyte conditions, both sericin- and PTFE-based electrodes maintained discharge capacities above 1050 mAh g^{-1} after 50 cycles. Taken together, these results suggest that sericin, a fully biodegradable and fibrillating polymer, holds meaningful potential as a viable alternative to PTFE in dry-processed cathodes. Instead of merely replicating the function of PTFE, sericin-based systems introduce distinct interfacial and mechanical characteristics that may support future development of fluoropolymer-free electrode designs.

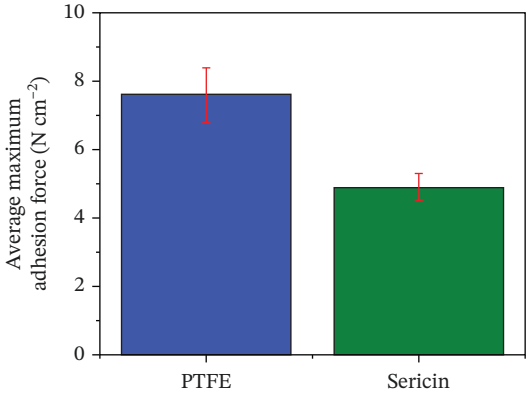
In another approach, Kim et al. [106] utilized parafilm, a commercially available polyolefin-paraffin wax mixture, to propose a universal PTFE-free dry processing strategy. Leveraging the thermoplastic nature of its polyolefin components, parafilm undergoes flexible deformation during dry mixing and pressing, effectively creating robust mechanical interlocking between active material and conductive agent particles. Mechanical cohesion profiling across the electrode depth revealed that parafilm-based electrodes maintain significantly higher and more uniform cohesion throughout the film compared to PTFE or PVDF benchmarks (Figure 4d). Furthermore, the interfacial adhesion strength reached ~2.4 N cm^{-1}, representing a more than two-fold improvement over PTFE-based electrodes (Figure 4e). This structural integrity supports efficient ion transport by maintaining interconnected pathways without excessive particle coverage, resulting in lower ionic tortuosity (Figure 4f) and reduced internal resistance across various current densities (Figure 4g). As a result, parafilm-based full cells delivered a high areal capacity exceeding 5.6 mAh cm^{-2}, highlighting the practical potential of using commodity polymers for high-performance PTFE-free dry electrodes (Figure 4h).

On the anode side, similar efforts have been made to develop PTFE-free binders that ensure both mechanical stability and electrochemical performance under dry processing conditions. Kim et al. [23] introduced styrene–butadiene rubber (SBR) as a PSA binder to enable structural and electrochemical stability in PTFE-free, high-loading dry electrodes (Figure 4i) Under ambient conditions, SBR behaves as an elastomer, but when subjected to mechanical pressure above ~ 1.0 MPa, it transitions into a viscoelastic and highly tacky state, even at room temperature, and reverts to its original form upon pressure release. This reversible tackiness facilitates interparticle adhesion without solvents, forming a point-contact-based binder network between graphite and conductive particles (Figure 4j). The resulting microstructure enhances mechanical cohesion while preserving interparticle porosity through minimal binder coverage, thereby generating an open-channel interfacial architecture. This structural configuration contributed to a notable reduction in charge transfer resistance (R_{ct}), with the PSA electrode exhibiting a low R_{ct} of 19.8 Ω cm^2 compared to 51.7 Ω cm^2 for its slurry-based counterpart. Depth profiling via ToF-SIMS and XPS revealed that the PSA electrode formed a compact, LiF-rich, and thin SEI

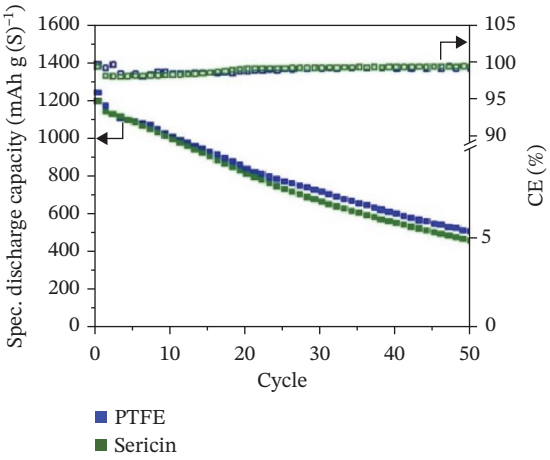
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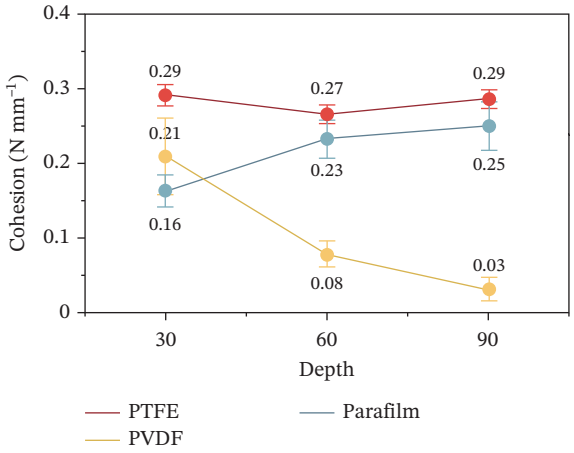
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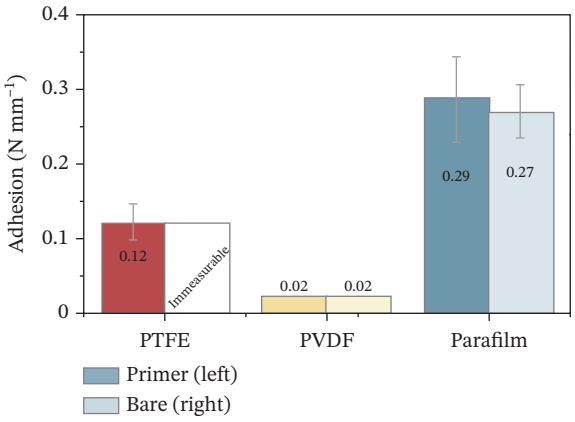
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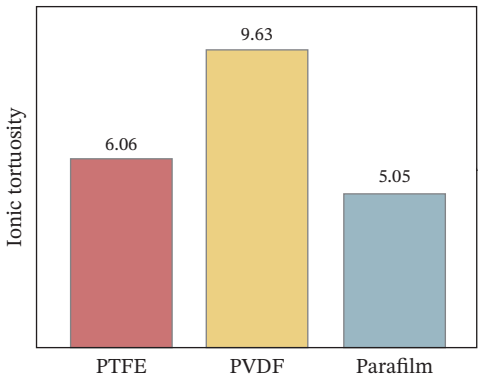
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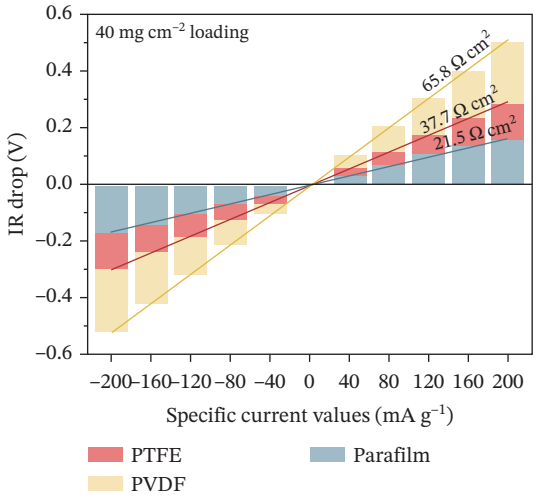
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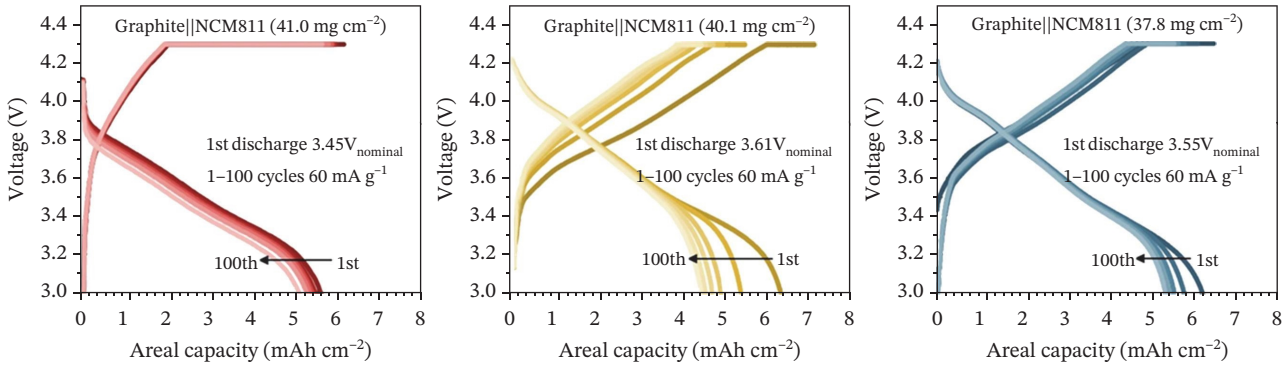
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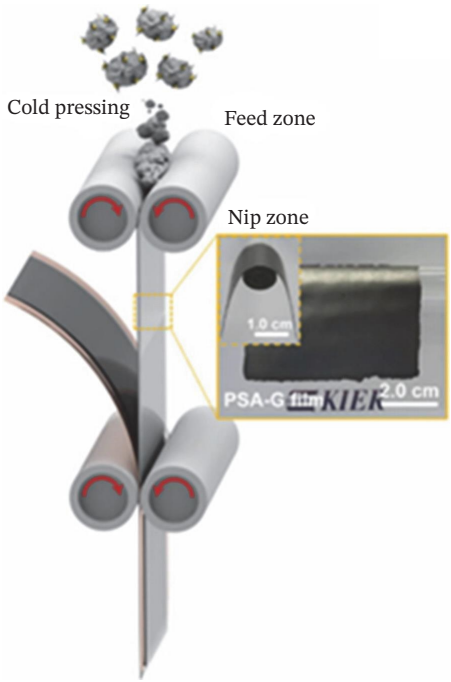
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(g)



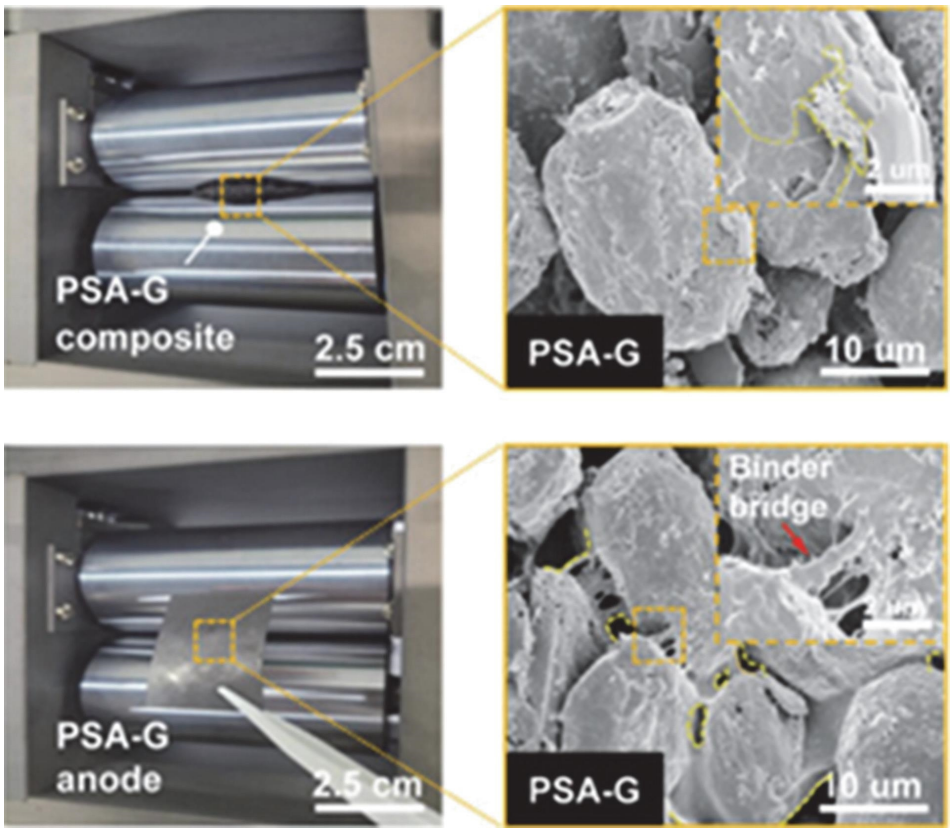
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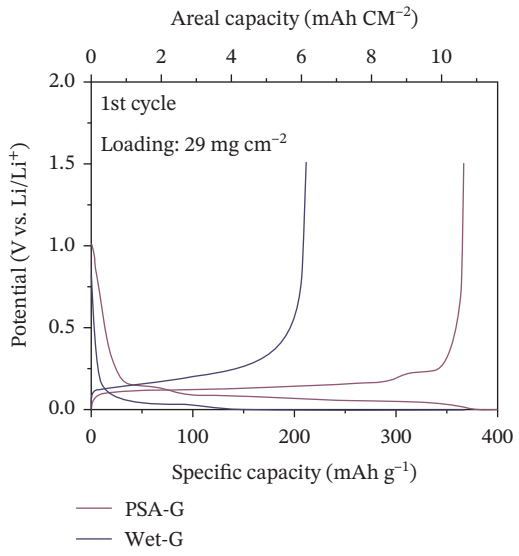
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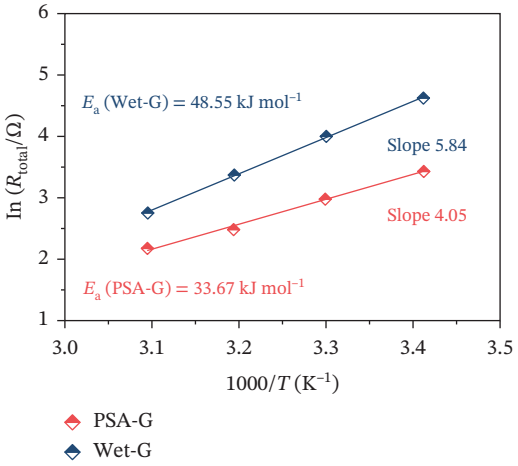
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(j)



(k)



(l)

FIGURE 4 | (Continued)

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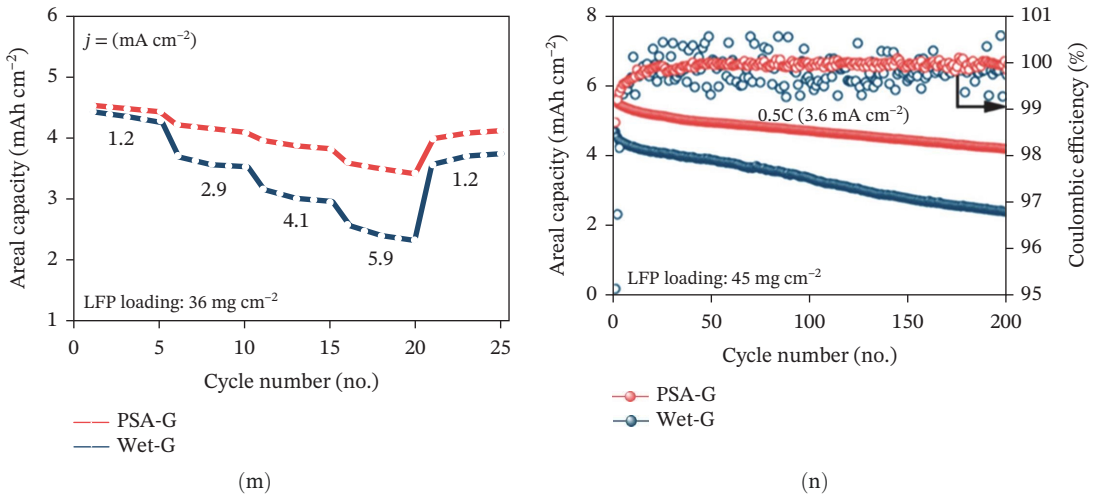


FIGURE 4 | (a) SEM images of a dry-processed electrode containing a sericin binder, with a white arrow indicating the location of fibrillated sericin. (b) Adhesion strength of PTFE- and sericin-based cathode films on substrates. (c) Long-term cycling stability profiles of both binder systems. Reproduced under the terms of the CC-BY license, ref.. Copyright 2022, Schmidt et al. [19], published by Wiley-VCH GmbH. (d) Mechanical cohesion profiling across electrode depth comparing parafilm, PTFE, and PVDF binders. (e) Interfacial adhesion strength of dry-processed electrodes. (f) Comparative ionic tortuosity values demonstrating enhanced ion transport pathways in parafilm-based electrodes. (g) Analysis of IR drop and internal resistance across different binder systems under various specific currents. (h) Electrochemical voltage profiles and areal capacity delivery of high-loading full cells. Reproduced under the terms of the CC-BY 4.0 license ref.. Copyright 2025, Kim et al. [106], published by Springer Nature. (i) Schematic illustration of the dry processing with SBR binder (PSA). (j) FE-SEM images of the SBR-graphite (PSA-G) composite powders and anode. (k) Initial formation behavior of PSA-G and conventional Wet-G anodes at 29 mg cm^{-2} (l) Arrhenius plots derived from impedance data after the formation step. (m) Rate performance comparison between PSA-G- and Wet-G-based full cells. (n) Cycling stability of full cells comprising PSA-G anodes and PSA-LFP cathodes with high loading (45 mg cm^{-2}). Reproduced with permission [23]. Copyright 2025, American Chemical Society.

originating from the internal interface, effectively suppressing excessive electrolyte decomposition.

The PSA electrode delivered a high initial capacity of 366.8 mAh g^{-1} while the wet electrode exhibited 211.4 mAh g^{-1} (Figure 4k). The activation energy for Li^+ desolvation was measured at 33.67 kJ mol^{-1}, substantially lower than the 48.55 kJ mol^{-1} observed for the slurry-processed system, indicating more favorable ion transport kinetics (Figure 4l). Moreover, the Li^+ diffusion coefficient derived from Randles–Sevcik analysis reached 1.52×10^{-9} cm^2 s^{-1}, more than double that of the slurry-based electrode, highlighting the role of the open-channel SEI structure in facilitating ionic mobility. The benefits of the PSA binder were further validated in full-cell configurations. When paired with an $LiFePO_4$ (LFP) cathode, PSA maintains higher specific capacities even in high C-rates (Figure 4m). For long cycle analysis, the PSA-based cell delivered a discharge capacity of 5.6 mAh cm^{-2} and maintained 74.6% capacity retention over 200 cycles (Figure 4n). Compared to the slurry-based counterpart under identical conditions, the PSA system demonstrated more stable capacity retention. Notably, the performance gap widened at higher C-rates, underscoring the robustness of the open-channel interface and cohesive binder network in enabling fast charge–discharge operation. This research shows that the dry anode binders can deliver competitive performance while retaining compatibility with existing dry processing infrastructure. By eliminating PTFE yet leveraging pressure-activated adhesion, the approach enables structural cohesion and interfacial optimization without requiring fundamental changes to manufacturing. It marks a practical advancement toward scalable, PTFE-free electrode technologies.

PTFE-free binder systems remain an aspirational yet increasingly tangible goal. Although their performance still requires improvement compared to PTFE-based fibrillation, the development of alternative binding mechanisms through function-oriented binders, particularly those exploiting low glass transition behavior or in situ structuring, offers a promising direction for the future progress. To provide a structured overview of these advancements, the various classes of emerging PTFE-free binder systems and their characteristic binding mechanisms are systematically categorized and summarized in Table 1. Continued innovation in polymer chemistry, interfacial science, and dry process engineering will be essential. Each successful step, whether in minimizing fluorine content or demonstrating dry-process-compatible alternatives at the lab scale, brings the field closer to large-format, sustainable, PTFE-free LIBs.

5 | PTFE-Less as a Transitional Strategy

As PTFE remains deeply embedded in the current architecture of dry electrode processing, complete and immediate elimination is not yet practical. Among these, the concept of a PTFE-less strategy was proposed by Kang et al. [53]. These approaches aim to lower the overall fluorine load within the electrode by combining PTFE with functional additive binders. To this end, they introduced hybrid systems incorporating PAA-grafted CMC, which have successfully reduced PTFE content to as low as 0.6 wt%, while maintaining sufficient fibrillar structure and mechanical integrity. These systems retain the critical functionality of PTFE but shift complementary roles, such as adhesion and ionic conduction, to nonfluorinated components. Altogether, PTFE-less systems serve as a pragmatic bridge between the current reliance

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TABLE 1 | Mechanistic comparison of PTFE, PTFE-less, and PTFE-free binder systems for solvent-free dry electrode processing.

Category	PTFE (conventional benchmark)	PTFE-less (hybrid systems)	PTFE, free (alternative systems)
Core design aim	Maximize mechanical cohesion via fibrillation	Reduce PTFE content while retaining fibrillation-assisted cohesion	Eliminate PTFE while enabling nonfibrillating film formation
Representative binders	PTFE [52, 107–110]	PTFE + PAA-grafted CMC; [53] PTFE + functional additive binders [59, 96, 111]	Sericin; [19] PSA-SBR; [23] thermoplastic elastomers;[22, 98] aliphatic polycarbonates; [21] poly(propylene carbonate); [112] parafilm [106]
Dominant cohesion mechanism	Shear-induced fibrillation and fiber entanglement	Partial fibrillation combined with auxiliary adhesive interactions	Pressure-sensitive adhesion, viscoelastic deformation, or chemical bonding
Film formation trigger	High shear during dry mixing	Moderate shear combined with compressive stress	External compressive stress (calendaring/roll-to-roll/lamination)
Role during powder mixing	Generation of fibrous network for cohesion	Controlled fibrillation to assist dispersion and cohesion	Particle decoration and predispersion without fibrillation
Role during calendaring	Fiber consolidation and densification	Fiber-assisted densification with improved process tolerance	Pressure-activated binder bridging and localized adhesion
Sensitivity to processing conditions	High (overfibrillation, shear-history dependent)	Moderate (balance between fibrillation and auxiliary adhesion)	Pressure-, formulation-, and viscoelastic-property dependent
Electrochemical stability	Limited (PTFE decomposition, fluorine-related issues)	Improved relative to PTFE (reduced fluorine content)	High (fluorine-free systems)
Manufacturability (R2R)	Established but narrow process window	Improved robustness compared to PTFE-only systems	Promising, but still under active validation
Key strengths	Proven scalability and mechanical robustness	Transitional pathway for PTFE reduction	PFAS-free, alternative film-formation mechanisms
Key limitations	PFAS concerns and electrochemical instability	Residual PTFE reliance and increased system complexity	Limited long-term industrial standardization
Typical application focus	Conventional dry electrodes	Transitional dry electrodes with minimized PTFE	Next-generation sustainable dry electrodes

on PTFE and a longer-term shift toward PTFE-free binder platforms. They offer a compromise, enabling regulatory risk mitigation and sustainability gains without abandoning the performance backbone of dry processing. Understanding how to optimize these intermediate systems will be key to maintaining production continuity while building toward a fluorine-free future.

6 | Outlook

Dry electrode processing represents a transformative advancement in LIB manufacturing, enabling the elimination of toxic solvents and the creation of high-loading electrodes with reduced environmental and energy costs. At the center of this technological leap has been PTFE, whose fibrillation under shear allows the formation of cohesive, mechanically stable electrode films without the need for solvents or drying. Its unique function has made it a cornerstone of scalable dry processing strategies adopted across both academic and industrial settings.

However, this long-standing reliance on PTFE now encounters an inflection point. As regulatory frameworks gain traction globally, the continued use of fluorinated binders, even those that are chemically stable and nonleaching, faces increased scrutiny. What once empowered dry processing to move beyond conventional slurry methods now threatens to undermine its long-term sustainability and compliance. The challenge is not the failure of dry technology itself, but the evolving definition of acceptable materials within it. Binder systems must now meet not only performance and manufacturability criteria but also align with emerging standards of environmental responsibility and regulatory transparency.

While they do not represent a final solution, PTFE-less systems offer valuable flexibility in the near term by reducing total fluorine content. These strategies continue to rely on PTFE's

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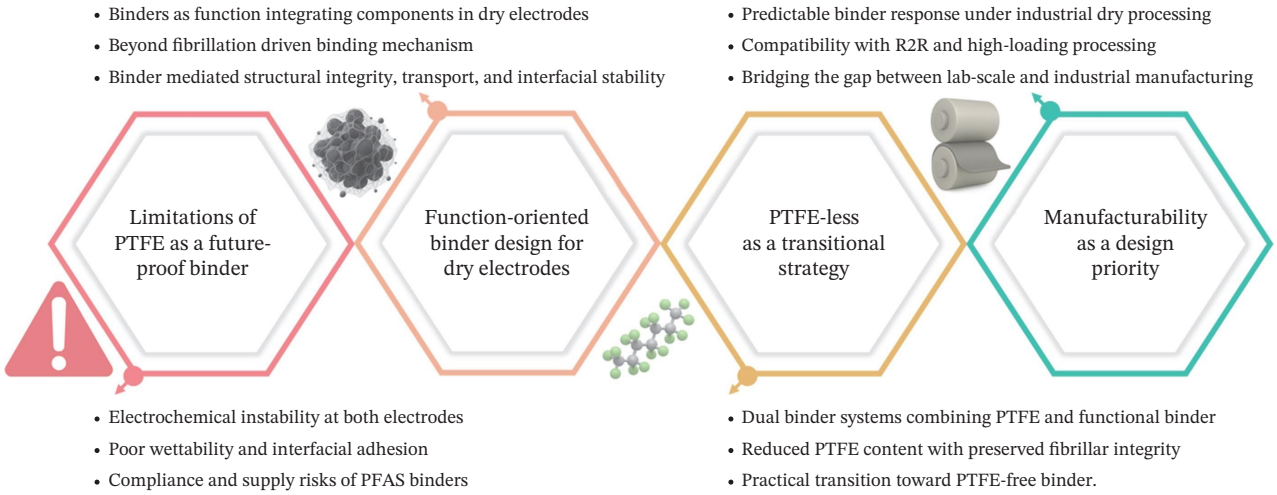


FIGURE 5 | Key considerations for function-oriented binder design in dry electrode processing addressing regulatory risks of PTFE, developing binders based on new mechanisms beyond fibrillation, recognizing that PTFE-less strategies serve only as a transitional solution, and ensuring compatibility with existing dry manufacturing platforms, emphasizing manufacturability as a core design parameter to ensure scalability under industrial dry processing conditions. This framework outlines a comprehensive direction for next-generation PTFE-free binder development.

irreplaceable fibrillation behavior to maintain film integrity, and as such, cannot fully escape the structural and policy risks associated with PTFE. A decisive step forward requires the development of binders that can function independently of fluorinated polymers and operate effectively under the mechanical, thermal, and processing constraints of solvent-free electrode fabrication (Figure 5).

PTFE enabled solvent-free manufacturing, but its role is no longer future-proof; PTFE has served as a foundational enabler for dry electrode technology due to its distinctive ability to fibrillate under shear, forming robust, self-supporting films without solvents. Its chemical inertness, thermal stability, and mechanical performance made it uniquely suited for R2R calendaring and solventless mixing. However, this advantage now conflicts with the accelerating global momentum toward eliminating fluorinated binder materials. Although PTFE does not leach or react under typical battery conditions, its environmental persistence and resistance to degradation have drawn increasing regulatory attention. As restrictions tighten, the continued dependence on PTFE, even in trace amounts, poses substantial risk to long-term process legitimacy and material supply continuity. The very feature that once enabled dry processing may now constrain its future deployment.

PTFE-less binders are a useful bridge, but not the destination: Functional additive binder systems and hybrid architectures in dry electrode systems that minimize PTFE content represent an important intermediate step. These approaches have successfully reduced fluorine concentration by combining PTFE with functional additives binders while maintaining structural integrity and electrochemical performance. However, PTFE-less systems still retain PTFE as the primary fibrillating agent, meaning the regulatory, environmental, and sourcing risks. Moreover, the chemical ambiguity of "PTFE-less" leaves room for interpretation and potential future bans. From a compliance and sustainability perspective, these systems delay rather than resolve the fundamental challenge. Their value lies in buying time for the development of truly fluorine-free alternatives that are both performance-ready and policy-aligned.

Processing and manufacturability constraints must guide material design: One of the most overlooked challenges in binder innovation is compatibility with industrial dry electrode processing conditions. Laboratory-scale demonstrations often use small, manually prepared pellets or discs, where electrode integrity can be maintained under idealized conditions. However, industrial R2R systems impose far greater mechanical and thermal stress. Calendaring pressures, shear-induced heating, and continuous throughput demand materials that respond predictably during high-speed mixing and film formation. Importantly, the development of binders that can form uniform films without relying on shear-induced fibrillation is essential for expanding the dry processing landscape. These emerging alternatives leverage mechanisms such as pressure-activated adhesion (e.g., PSA binders) or thermoplastic and viscoelastic deformation (as demonstrated by materials like parafilm), showing significant potential for compatibility with existing R2R infrastructure. Unlike conventional PTFE, which is highly sensitive to nonuniform shear gradients, these binders respond primarily to the uniform compressive loads of the rollers. This shift from shear-dependent to pressure-dependent film formation ensures structural cohesion and homogeneous component distribution, potentially offering a more stable and scalable process window than conventional fibrillating systems. The focus of future research must therefore move beyond laboratory proof-of-concepts toward the rigorous validation of these new binder architectures within actual industrial-scale manufacturing environments. Therefore, the design of future binders must begin with an understanding of core process parameters the synergy between mechanical and thermal energy activation, dry flow compatibility, and rheological behavior under high compression must be considered as fundamental material properties, not afterthoughts. Without manufacturability, even the most promising material will not scale.

7 | Conclusion

The dry electrode community now faces a pivotal opportunity and an equally urgent responsibility. The goal is no longer simply to maintain performance while minimizing fluorine content but

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to build a new foundation for a cohesion and processability function-oriented binder that is independent of PTFE entirely. This requires elevating binder development to the same level of strategic importance as active material selection or cell design. PTFE-free binders must become a central pillar of solvent-free manufacturing innovation. By moving decisively away from PTFE and toward scalable, regulation-resilient alternatives, the battery field can align itself with the broader transition toward environmentally responsible, high-performance energy storage. The dry electrode process is already a blueprint for clean manufacturing; now, it must complete its evolution with truly clean materials at its core. This transformation is not a limitation but a chance to lead. Binder systems that uphold structural integrity, process compatibility, and environmental responsibility will not only secure the future of dry electrodes but also redefine what sustainable battery technology can be.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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