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Advancements in Dry Electrode Technologies: Towards Sustainable and Efficient Battery Manufacturing

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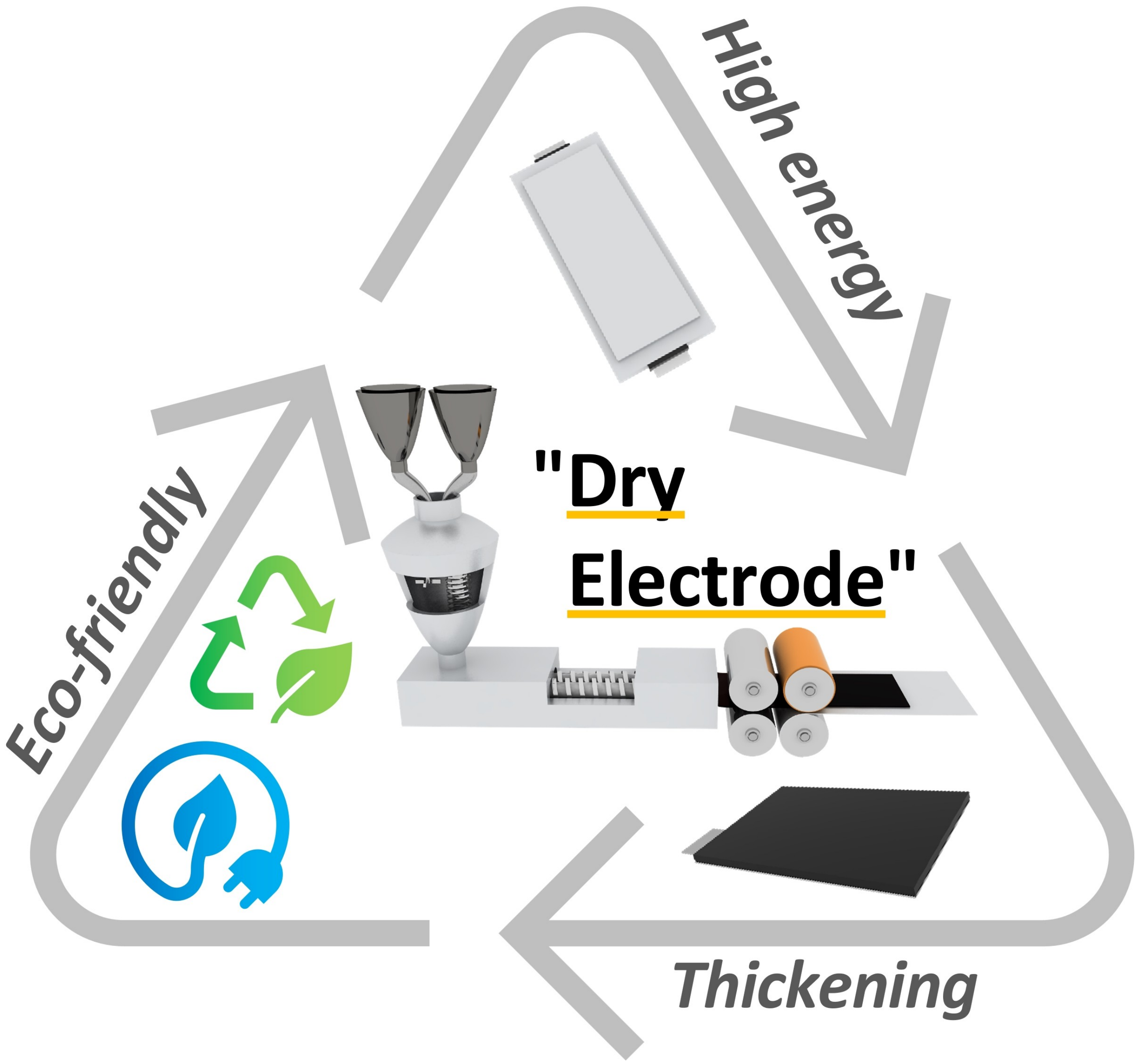

To address the urgent demand for sustainable battery manufacturing, this review contrasts traditional wet process with emerging dry electrode technologies. Dry process stands out because of its reduced energy and environmental footprint, offering considerable economic benefits and facilitating the production of high-energy-density electrodes. We spotlight technological innovations that exemplify the paradigm shift towards eco-friendliness and cost-efficiency. This review synthesizes the latest developments in dry electrode production, comparing the techniques with conventional methods, and outlines future research for further optimization toward a higher technology readiness level. We suggest that the evolution of battery manufacturing hinges on the synergy between process innovation and materials science, which is crucial for meeting the dual goals of environmental sustainability and economic practicality.

1. Introduction

The escalating global energy demands have spurred notable improvements in battery technologies. It is evident from the steady increase in global energy consumption, which has grown at an average annual rate of about 1–2% over the past fifty years.[1] This surge is primarily driven by the growing adoption of electric vehicles (EVs) and the expansion of electricity usage. These findings underscore the critical need for advancements in battery technologies. Since Sony developed the Li-ion battery in 1991,[2] the penetration of energy storage devices with rechargeable batteries, including electric vehicles, has dramatically increased. Simultaneously, the energy density, production rate, and quality of batteries have steadily improved. These advancements are central to the transition towards sustainable, efficient, and cost-effective manufacturing processes. From these perspectives, dry electrodes are vital to developing next-generation batteries that meet increased energy demands and sustainability.

Figure 1 depicts the historical and technological progression of dry electrode technologies.[3] The outline figure presents a chronological sequence of pivotal discoveries, technological advancements, and significant industrial implementations that have shaped the current landscape of dry electrode technology, with a particular focus on calendering methods due to their prominence in current industrial applications and prototype plants. It's important to note, however, that calendering is but one of several dry coating techniques explored in battery manufacturing. Others include electrostatic deposition, spray drying, and roll-to-roll coating, each offering unique advantages and challenges to be reviewed in the following. This innovation, stemming the strategic acquisition of Maxwell Technologies from Tesla in 2019[3f] and subsequent integration of their dry electrode technology, marks a significant shift in battery production techniques. These cells, born out of a technology foundation laid in 2003 by the pioneering patent of Maxwell,[3c] not only enhance energy density but also show potential for reducing both the cost per kilowatt-hour and the environmental impact of battery production. Furthermore, the growing interest in dry electrode technologies is evidenced by recent groundbreaking developments in processing concepts. For instance, the 4680 cylindrical cells of Tesla, manufactured using freestanding dry anodes announced at Tesla Battery Day event in 2020,[3g] illustrate the significant efforts toward efficient and sustainable battery production. This new form factor boosts energy density and potentially reduces the cost per kilowatt-hour and carbon footprint using dry electrode manufacturing.

Our mini-review will delve into the complexities of dry electrode technologies, elucidating the associated economic and environmental innovations, manufacturing advancements, and compatibility with materials. Through a detailed examination of recent literature and a comparative analysis with conventional wet processes, this mini-review aims to provide comprehensive insight into the potential of dry electrode technologies in heralding a new era of sustainable and efficient battery science.

2. Economic and Environmental Comparison for Battery Electrode Manufacturing

Wet processing is a well-established method but poses a host of challenges as depicted in the upper part of Figure 2. Primarily, it requires significant energy consumption due to the extensive drying steps needed to evaporate the solvent used in the slurry-coating process. The drying process in wet electrode fabrication is notably energy-intensive, requiring 30–55 kWh per kWh of cell energy.[4] Additionally, producing a 28 kWh lithium-ion battery can result in CO_2 emissions of 2.7-3.0 tons equivalently, emphasizing the environmental impact of the production process.[5] This high energy demand not only increases the operating expenditure (OPEX) related to production cost with N-methyl-2-pyrrolidone (NMP) solvent in cathode production alone accounting for up to 11.5% of manufacturing costs and over 46% of energy consumption, which is also

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environmentally and biologically hazardous. Meanwhile, despite lower solvent costs,[6] water-based processes face parallel energy challenges for the drying process, which forms a significant part of the OPEX.[7] Furthermore, carbon-binder domain (CBD) migration is a significant issue in wet processes where uneven composition during solvent drying can lead to delamination in thicker electrodes[8] for higher energy density.

Additionally, the inherent inefficiencies in the wet process result in low productivity, worsened by the lengthy drying times. The need for expansive facilities equipped with extensive drying infrastructure further increases the capital expenditure (CAPEX) by approximately 20%, rendering the process less economical, especially in the face of rising demand for battery use in electric vehicles and energy storage applications.[4,12] The equipment footprint for wet processing is significantly larger due to the required solvent recovery systems and multiple drying stages, which are necessary to handle large volumes of hazardous solvents like NMP. These systems increase the physical space required for production for setting up a manufacturing plant.

Unlike wet process, dry electrode manufacturing technologies offer a more sustainable and efficient paradigm for electrode production as illustrated in the lower part of Figure 2.[10b,11b,13] The cornerstone of dry process is its eco-friendliness, eliminating the need for toxic solvents, thereby significantly mitigating the environmental impact by reducing the generation of most hazardous wastes such as NMP. The solvent-free approach curtails energy consumption and makes the production method more efficient. Furthermore, dry process brings economic advantages by considerably lowering the production cost by 10–15%.[13c,14] Eliminating drying steps and solvent recovery systems might significantly reduce the space and energy requirements, translating to a lower CAPEX and OPEX. The reduction in energy consumption, coupled with the


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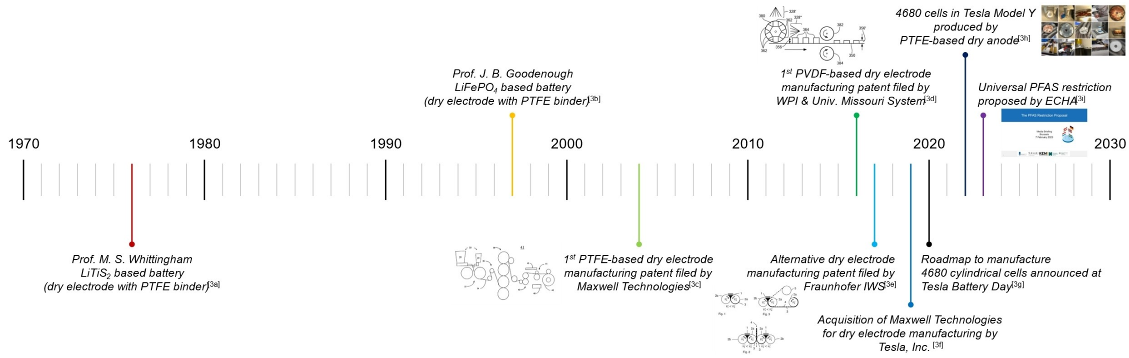

Figure 1. Historical and technological progression of dry electrode technologies for battery manufacturing.[3] Reproduced with permission from.[3h] Copyright (2023) IOP Publishing.

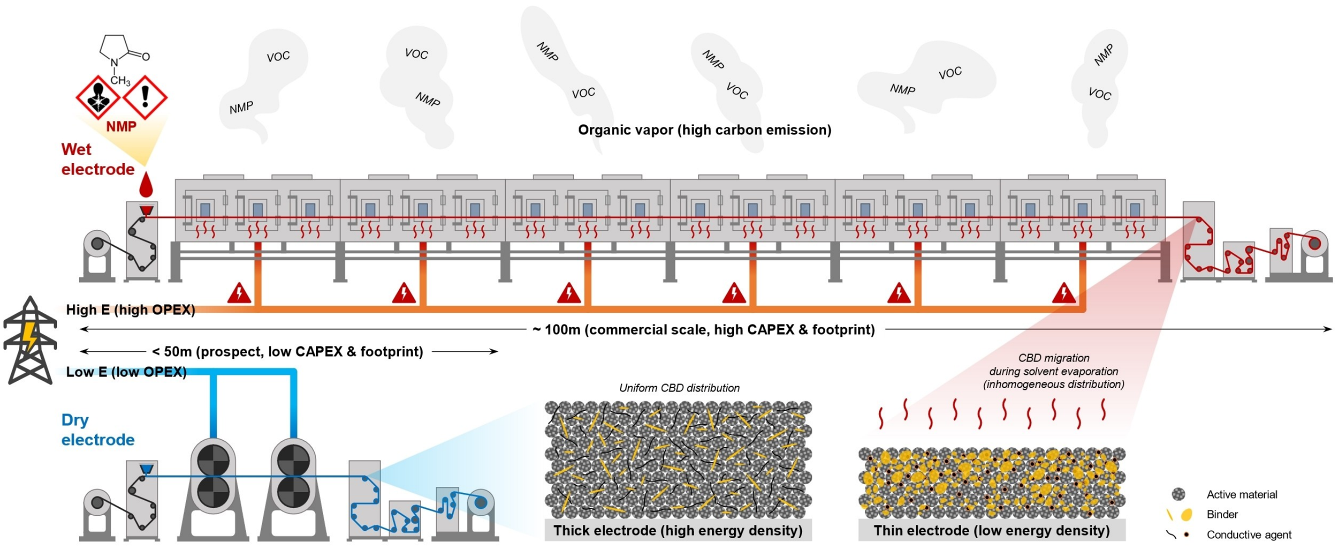

Figure 2. Schematic overview comparing wet and dry electrode manufacturing lines. The top illustrates the traditional wet process with substantial equipment footprint and VOC/NMP emissions, leading to inhomogeneous binder distribution. The bottom shows the streamlined dry process with a smaller footprint, lower CAPEX, and the production of thicker electrodes with uniform composition, enabling higher energy densities.

avoidance of solvent procurement and disposal costs, contributes to the cost-effectiveness of dry electrode manufacturing. Moreover, dry processes facilitate the fabrication of thick electrodes over 5 mAh cm^{-2} due to the absence of CBD migration, crucial for enhancing the energy density of batteries.[10b,11b,15] It also avoids polyvinylidene fluoride (PVDF) gelation problems in wet processing, particularly with Ni-rich NCM cathodes.[10a,11a] In the context of all-solid-state batteries (ASSBs), dry process presents significant benefits, particularly for sulfide-based solid electrolytes. The lack of polar solvents in dry process prevents adverse reactions with these electrolytes, crucial for the structural integrity and performance of ASSBs.[9,10b,16] Several researchers have demonstrated that varying the content of polytetrafluoroethylene (PTFE) in sulfide-based ASSBs influences the quality of the electrodes and electrochemical properties.[17] These aspects make dry process highly suitable for advanced, high-energy-density battery production. The comprehensive comparison of wet and dry electrode manufacturing is represented in Table 1.

The paradigm for constructing electrodes should be innovatively refined to enable carbon neutralization and eco-friendly electrification. As a game changer in the battery field, dry electrode technology has been developed to prevent fast climate change for as long as possible, even in battery manufacturing systems beyond the battery operating environment. In addition, the drying-free process in the dry electrode concept could shorten electrode production time and reduce power consumption for solvent vaporization owing to the unaffected design factor in this system. Therefore, the suggested manufacturing process for dry electrodes is simplified compared with slurry-based electrode fabrication (Figure 3). The typical wet process involves slurry mixing, coating, drying (including solvent recovery), pressing, and slitting. At the end stage, final vacuum drying is optionally performed to clearly remove residual solvent inside the as-prepared electrode. In contrast, the dry electrode fabrication steps can be categorized into dry mixing, electrode film fabrication, pressing, laminating, and slitting; the removal of electrode drying dramatically reduces the time/cost and required plant size, as reported at

Table 1. Comparative analysis of wet and dry electrode manufacturing processes in battery production.

Characteristic	Wet Electrode	Dry Electrode	References
CO_2 Emission	2.7-3.0 tons CO_2 per 28 kWh battery	Substantial reduction (exact figures vary based on process specifics)	[4–6]
CAPEX	Higher, up to 20% more due to solvent recovery and drying infrastructure	Reduced by approximately 10–15% due to streamlined processes	[7–8]
OPEX	NMP solvent accounts for up to 11.5% of manufacturing costs and over 46% of energy consumption	Estimated reduction of 10–20% in energy and solvent costs	[4,9]
Equipment Footprint	Larger, requiring significant space for solvent recovery systems and drying stages	Smaller footprint, potentially reducing factory space needs by 20–30%	[7,10]
Electrode Thickness	Limited due to CBD migration, typically under 4 mAh cm^{-2}	Facilitates production of electrodes over 5 mAh cm^{-2}	[10b,11]

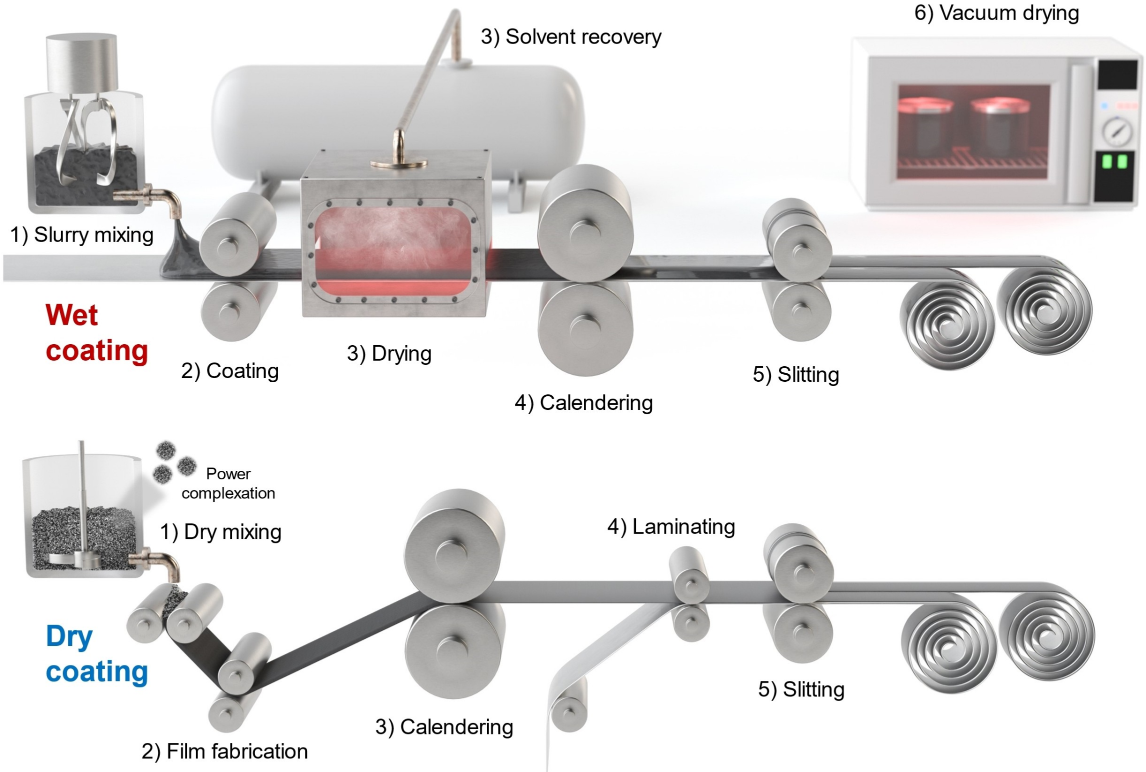

Figure 3. The comparison of typical slurry-based (upper) and dry-based (lower) electrode manufacturing process. Slurry-based wet process: 1) Slurry mixing, 2) Coating, 3) Drying/Solvent recovery, 4) Calendering, 5) Slitting, and 6) Vacuum drying. Dry process: 1) Dry mixing, 2) Electrode film fabrication, 3) Calendering, 4) Laminating, and 5) Slitting.

Battery Day by Tesla held in 2020.[3g] Similarly, the emergence of DRYtraec® technology by Fraunhofer IWS also represents a promising dry electrode manufacturing alternative.[18] With its advanced processing concept promise of productivity and lower cost, DRYtraec® is being developed toward a higher technology readiness level (TRL) for the pilot scale. In addition, other dry electrode manufacturing techniques are being developed, such as AM Batteries (a spin-off from Worcester Polytechnic Institute), which uses the electrostatic spraying concept, further diversifies the dry electrode process techniques by referencing mature methods of different industries.[13b,c]

3. Process Factors for Designing Dry Electrodes

3.1. Manufacturing Technologies of Dry Electrode Film

Various dry electrode fabrication methods have been designed and maturated as proof of concepts at both the academic and industrial levels. As an example, a simple and batch-scale procedure was designed using a hot-press machine (Figure 4a).[11b] The as-mixed cathode materials under dry conditions displayed electrode integrity achieved through hot pressing to realize efficient thermal activation of the internetworked binder. Then, conventional roll pressing was used to complete the dry electrode fabrication process. Because hot pressing induces the phase transition of the binder such as forming a molten state, it provides numerous anchoring sites across the entire electrode to construct a robust electrode framework. However, this technical concept has yet to be introduced into the continuous electrode manufacturing system as it diminishes the advantage of the dry electrode process in terms of enabling a high production rate. In addition, it is difficult to expand the polymer-dependent design factor to the commercialization stage because the electrode steadily requires an active material-enriched system to effectively emphasize the advantage of the dry electrode in terms of uniform thick electrode fabrication.

Extrusion-based electrode processes have been adopted as they have been widely used in industrial fields such as injection molding (Figure 4b).[10b] The flowability of polymeric binder in the dry-mixed powder provides is induced by the mechanical pressure during extruding. The semi-solid rheological property allows three different electrode materials to blend with each other, similar to a slurry-based system despite the milder conditions. Once the electrode film is prepared using the extrusion process, it undergoes calendering and laminating processes with the current collector. The manufacturing process using extrusion is greatly affected by the physical properties and component ratio of the polymeric binder,[21]; thus, similar to hot pressing, this suggested approach is limited by the restricted polymer portion in the electrode. Even though the polymer-poor system can be applied by reforming or developing polymer properties, a high solid (active and conductive materials) ratio results in an overload pressure in the extrusion machine, implying explosive issues; in addition, the low operating rate presents an obstacle for mass production.

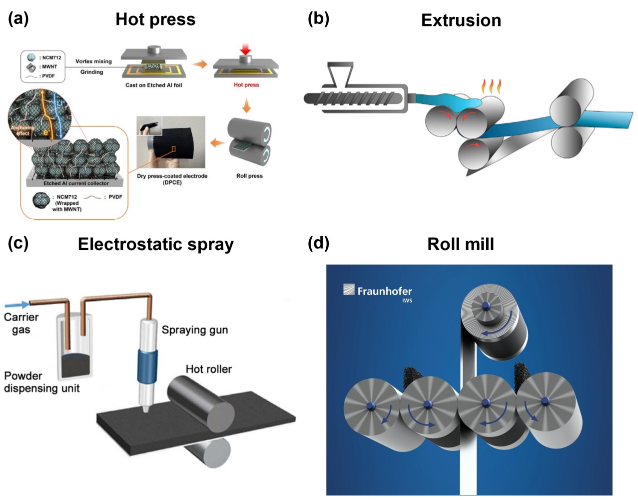

Figure 4. Conceptual methods to fabricate dry battery electrodes. Suggested concepts of electrode film fabrication for dry electrode production; (a) Hot press. Reproduced with permission from.[11b] Copyright (2023) Springer (b) Extrusion. Reproduced with permission from.[10b] Copyright (2023) Elsevier (c) Electrostatic spray. Reproduced with permission from.[19] Copyright (2021) Elsevier (d) Roll mill. Reproduced with permission. Copyright (2023) Fraunhofer IWS.[20]

Electrostatic spraying is also often applied for thick film and uniform coating on substrates, facilitating fast production and large-scale coverage. Dry electrode fabrication using electrostatic spray coating has been widely used in the battery field. The so-called "powder-to-electrode" technology has been representatively suggested by AM Batteries for direct coating on a current collector to eliminate the possible risk of free-standing film formation, followed by hot rolling to reinforce the adhesion with the current collector via hot melting and thermal-crosslinking of polymers (Figure 4c).[22] However, a critical challenge remains in this manufacturing process that the uniform powder coating is hardly guaranteed when fine powder or a complicated powder compositions were used.

Finally, the roll-milling-included procedure has been adopted as one of the most remarkable concepts for designing dry battery electrodes. The shear force created by the calender gap, pressing, and rotation ratio between rolls causes the dry-mixed power to experience additional mixing and dispersion, resulting in a smooth electrode film. The powder can be consecutively attached to the fast-rotated roll; thus, the roll-milling concept facilitates continuous battery electrode production. In industry, Maxwell–Tesla selected a roll-milling-based dry coating process to fabricate freestanding electrode films by using two roll mills, and Fraunhofer IWS recently patented DRYtraec® technology consisting of multiple roll mills toward cost-saving electrode production (Figure 4d). The advance of their technologies additionally stems from the equipment configuration, where the deployed rolls with different roles enable the unification of the process along with direct electrode pressing and lamination. The roll-mill-based method is likely to be used in the mainstream development of dry battery electrode procedures. However, the shear force depends on the particle or granular size, requiring sensitive control to minimize film rupture, swelling, and edge deformation during the entire process and finally produce fine dry battery electrodes. Additionally, although the roll-mill process is relatively unaffected by the binder ratio, the binder material is currently severely restricted to PTFE, enabling polymer fibrilization, as elucidated in section 4.

3.2. Importance of Powder Mixing in the Dry Electrode

For developing high-energy-density batteries, it is essential to fabricate thicker electrodes with better electrochemical performance. As shown in Figure 5, during the production of high-loaded electrodes using wet processes, the rheological properties of the slurry during solvent evaporation in the coated electrode film can cause conductive agents and binders to unevenly accumulate on the surface due to their low-density characteristics.[23] This non-uniform distribution compromises both the mechanical and electrochemical properties of the electrode, thereby limiting their density and loading level for

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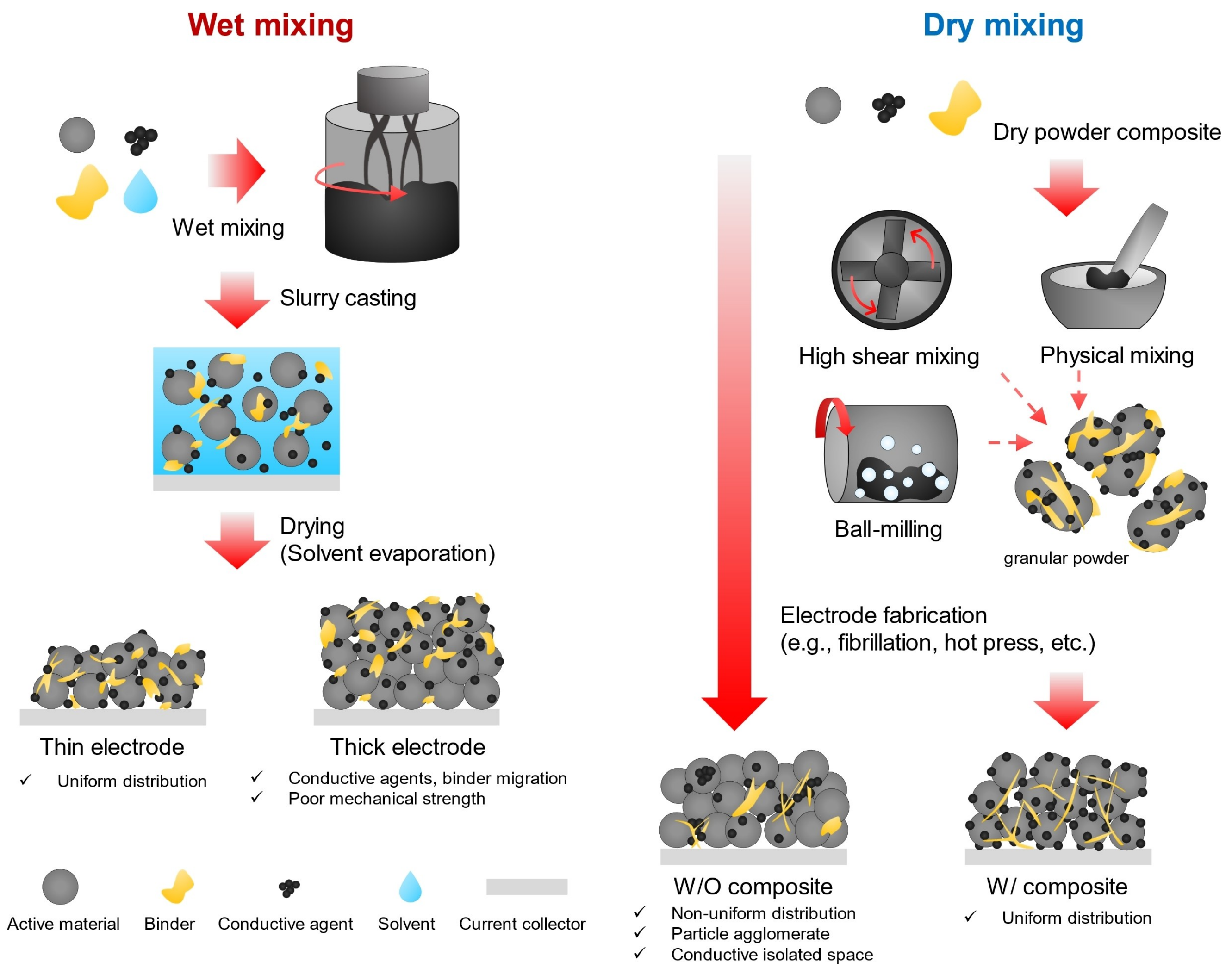

Figure 5. Comparison schematic for mixing process between wet and dry electrodes. Conventional wet processing incorporates solvent, which disperses the active materials and conductive agents while dissolving the binder. Dry electrode processing utilizes high energy physical mixing for uniform distribution of materials without the aid of solvents.

high-energy-density batteries. On the other hand, in the fabrication of dry electrodes, the aim is to reduce or eliminate solvent use, preventing the same phenomenon as slurry-based system at high-loaded electrodes. The dry process requires consideration not only of chemical stability against the adopted electrolyte in cell evaluation but also of distinct physical properties, such as high dispersion, fibrillization, low melting point, and powder flowability.

Thus, dry mixing, which combines the active materials, conductive agents, and binders in a solid state, presents challenges in terms of realizing a uniform distribution in the entire electrode. Achieving homogeneity demands substantial energy input due to the difficulty in thoroughly mixing these components. Consequently, relatively simple processes result in uneven material distribution, similar to wet electrode fabrication, thereby adversely affecting cell performance. Uneven distribution causes several issues, including 1) disruption of electron- and ion-transport pathways, increasing internal resistance; 2) localized high stress or insufficient adhesion, leading to poor electrode performance and potential mechanical failure; 3) fluctuations in the charge-storage capacity and cycling stability due to uneven active material loading; and 4) over- or under-utilization of specific electrode areas. Therefore, meticulous attention is necessary in material mixing for dry electrodes, especially when compared to wet processes and strategic approaches are suggested such as hot-melt mixing, secondary mixing to realize well-mixed composites and uniform distribution in electrode. Design factors such as the mixing equipment, mixing strength, mixing protocols, and material properties significantly impact the electrode characteristics. Noh et al.[24] compared five different mixing protocols for an LCO/96(78Li_2S–22P_2S_5)–4Li_2SO_4/Super P composite cathode. Their study concluded that protocols that aid in concentrating carbon at the active particle interface can lower the interfacial resistance and increase the discharge capacity.

The choice of binder size and type affect the characteristics of the dry mixing, uniformity of the mixture, and physicochemical stability of the dry electrode.[25] Moreover, the particle size

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differences in the dry electrode process not only influence the electrochemical properties but also result in variations in the manufactured electrode quality.[21] To ensure interfacial contact and uniformity among electrode materials, researchers have explored the pre-mixing of active materials, conductive agents, and binders before electrode coating using various methods. For example, Zhen and Ludwig et al.[19,26] firstly mixed the active and conductive agents and then conducted secondary mixing with PVDF binder, subsequently coating the electrodes using electrostatic spraying. This method achieved a solvent-free uniform distribution of the binder and conductive agents. Additionally, some researchers have compounded active materials, conductive agents, and thermoplastic binders prior to dry coating using hot pressing.[11b,23c,27] Understanding the significance of binders and their effect on electrode fabrication in dry processes is essential, as the procedures for dry mixing and electrode fabrication can differ based on the specific binder employed.

4. Rational Binders for Designing Dry Electrodes

4.1. Polytetrafluoroethylene

Polymer fibrillation of binders, especially in large-scale production, stands as a promising technique for solvent-free electrode manufacturing.[29] Among various fluorocarbon polymer binders such as PTFE, fluorinated ethylene propylene (FEP), perfluoroalkoxy (PFA), and ethylene tetrafluoroethylene (ETFE), only PTFE can undergo fibrillation with the application of an external mechanical force.[30] The polymer structure of PTFE consists of a carbon backbone with four fluorine atoms in each monomer, forming robust carbon–fluorine bonds to enable an extended and fibrous framework (Figure 6a). This unique structure allows PTFE to fibrillate under minimal shear stress owing to its low van der Waals forces and loose molecular stacking structure.[31]

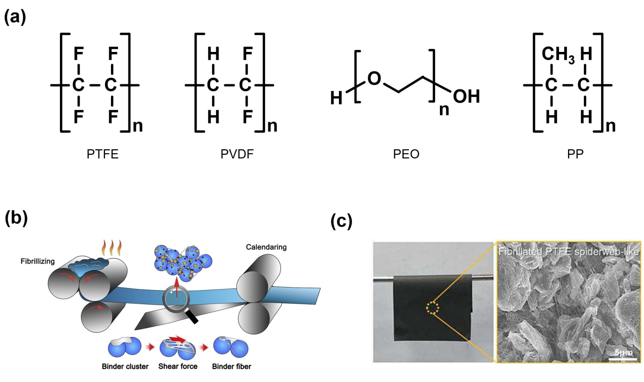

Figure 6. Rational binder properties for dry electrodes. (a) Representative polymers utilized as binders for dry electrodes. (b) Typical procedure for fabricating dry electrode through binder fibrillation. Reproduced with permission from.[10b] Copyright (2023) Springer (c) Free-standing electrode produced through the PTFE fibrillation and its SEM image. Reproduced with permission from [28]. Copyright (2023) Elsevier.

Significantly, the technologies of Maxwell–Tesla[32] and Fraunhofer[3e,33] have received authorized patents unveiling the dry binder electrode paradigm as mentioned above, reinforcing its advanced energy-storage credentials. Their manufacturing traits necessitate using PTFE as an irreplaceable binder to effectively apply shear force for dry mixing. Subjecting PTFE to blending and shear forces transforms its beads into fibrils, establishing a matrix that effectively blends and supports the dry-based powder (Figure 6b,c). The binder fibrillation process is affected by the physicochemical properties of the binder, the particle size of the active material, the equipment type, and the force-transmission parameters.[21,28–29] The temperature also affects PTFE fibrillation, transitioning through distinct phases beyond 19 °C, rendering the molecular chains pliable and easily extractable as fibrils with minimal shear force.[34]

Even though PTFE binder has the potential to realize a solvent-free system for the design of dry electrodes, poor adhesion between the current collector and dry electrode film reinforces the need for a primer coating to be introduced on the current collector in advance. This coating requires a wet process, thereby generating volatile organic compounds (VOCs), making it currently impossible to achieve a completely solvent-free process using PTFE.[35] Furthermore, dry electrodes employing PTFE binders are trending toward thick and large-area electrode design, raising concerns about poor electrolyte wettability. This is attributed to the strong electronegativity of fluorine, low surface energy, and hydrophobic properties in PTFE binders, which can potentially reduce the affinity between electrolytes and electrodes.[36] The low value of the lowest unoccupied molecular orbital (LUMO) for PTFE makes it thermodynamically susceptible to electrochemical side reactions at the potential of the anode, leading to structural degradation during battery charging.[37] Thus, the irreversible reduction at the anode presents challenges for simultaneously employing PTFE as a versatile binder in both the anode and cathode. Moreover, environmental regulation for PTFE use was proposed in 2023 following a report by the European Chemicals Agency (ECHA), aiming to restrict over 10,000 types of per- and polyfluoroalkyl substances (PFASs).[38] PFAS regulation extends beyond the EU, with the United States Environmental Protection Agency (EPA) leading legislative efforts and states undergoing processes to regulate PFAS, leading to ongoing disputes concerning these compounds.

4.2. Polyvinylidene Fluoride

PVDF is a well-known binder in wet processes, exhibiting superior electrochemical stability compared to PTFE binders in both the anode and cathode.[35c,37,39] However, PVDF encounters limitations, not effectively constructing a dry electrode because of its solidified structure with no fibrillation during the mixing of dry powders compared with PTFE when the same binder ratio is adopted.[40] Therefore, ensuring the even distribution of PVDF between the active and conductive materials is essential, maintaining an appropriate ratio to strike a balance between binding and resistive forces to achieve acceptable dry electrode and battery performance. In this regard, PVDF, one of the thermoplastic polymers that is readily melted by external heat, could undergo a hot melt process using methods such as hot pressing or spraying before and after the dry mixing process to achieve uniform complexation of the electrode materials.[23c]

4.3. Other Binders

A variety of binders have been used in dry electrodes, including poly(ethylene oxide) (PEO),[41] polypropylene,[42] polylactic acid,[43] acrylonitrile–butadiene–styrene (ABS),[43] and paraffin wax,[42] all falling under the category of thermoplastic binders.[10b] The type of polymer can be selectively adopted as the binder depending on the electrode manufacturing processes (e.g., hot pressing, electrostatic spraying, extrusion), as elucidated in the previous section. Despite their potential, to date, these alternative materials have been less recognized than PTFE and PVDF. Therefore, we suggest that researchers focus on the following directions in binder development: 1) studying binder modifications that can ameliorate the weaknesses of PTFE and 2) exploring methods to enhance and apply thermoplastic binders for mass production in electrode manufacturing.

5. Summary and Outlook

As the global thrust towards more sustainable and efficient battery manufacturing intensifies, dry electrode technologies have emerged as pivotal drivers in this transformation. This review has underscored the significant strides made in this domain, particularly in the realms of dry coating methods and innovative process developments for the transition from proof-of-concept to scale-up. These advancements signify a shift towards more eco-friendly manufacturing practices and promise to reshape the economic landscape of battery production.

However, the journey toward optimizing dry electrode technologies still needs to be completed. Key areas requiring further exploration and development are the dry mixing technique and the powder feeding concept.[44] Both of these areas, while promising, are still in the nascent stages of TRL. The dry mixing technique presents a critical avenue for research, given its potential to further streamline manufacturing processes and reduce costs. The inherent challenge lies in refining this technique to ensure consistent and high-quality electrode material production. Similarly, the powder feeding concept, crucial for enhancing manufacturing speed and efficiency, demands concerted efforts to elevate its TRL. The quest to achieve a faster, more efficient manufacturing process is inseparably linked to the evolution of these technologies.

Moreover, exploring alternative new dry binders that are stable for both cathodes and anodes is paramount. The conventional reliance on binders that contribute to the risk of PFAS pollution needs to be addressed.[38,45] Developing binders that reduce the fluorine content without compromising the structural and electrochemical integrity of the electrodes is a crucial step to aligning dry electrode technologies with environmental safety standards. This endeavor supports the sustainability agenda and aligns with the increasing regulatory and public awareness regarding PFAS risks.

It is imperative to acknowledge the critical yet often underrated role of process development in advancing battery technologies. Manufacturing innovation and materials engineering must progress in balance to meet energy storage demands. As the industry progresses, equal emphasis on refining manufacturing techniques will be essential to fully harness the evolution of next-generation batteries. This balanced focus on process and materials will be the cornerstone of future breakthroughs, ensuring that battery technology continues evolving in an environmentally responsible and economically feasible direction.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

Keywords: Thick electrode fabrication · Dry electrode · Solvent-free process · Lithium-ion batteries

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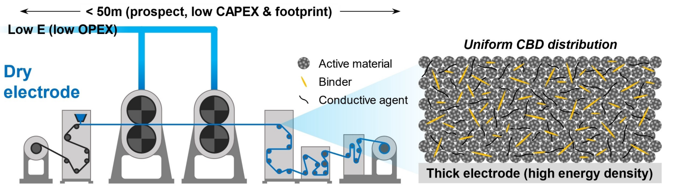
REVIEW



Developing a process for dry electrode fabrication is required to achieve high-energy-density batteries and carbon neutralization through thick electrode construction and organic solvent removal, respectively.

This review highlights promising concepts focused on manufacturing processes and binder materials of dry electrode to substitute slurry-based electrode.

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Advancements in Dry Electrode Technologies: Towards Sustainable and Efficient Battery Manufacturing