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Review article



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Modelling of lithium-ion battery electrode calendering: A critical review

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ABSTRACT

Lithium-ion Batteries (LIBs) are central to modern energy storage, with growing demands for improved performance, safety, and cost efficiency. Electrode calendering, a critical step in LIBs manufacturing, significantly influences the microstructure and electrochemical properties of electrodes. This review explores advances in the modelling of the calendering process over the past few years, focusing on empirical, numerical, and machine learning approaches. Empirical models, though computationally efficient, are limited by oversimplification, while numerical methods, such as Discrete Element Method (DEM) and Finite Element Method (FEM), offer more detailed insights into the structural evolution during calendering but require intensive computational resources. The growing application of machine learning introduces novel data-driven methods for optimising the process by effectively handling multiscale phenomena and high-dimensional data. A comparative analysis of these modelling strategies highlights the need for hybrid approaches that integrate empirical, numerical, and data-driven models to accurately predict electrode behaviour and optimise calendering conditions. Future research should aim to bridge the gap between computational accuracy and practical application to improve the performance and cost-efficiency of LIBs manufacturing.

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List of Abbreviations Artificial Intelligence AI AM Active Material CBD Carbon Binder Domains CNN Convolutional Neural Network DEM Discrete Element Method Deep neural network DNN FDM Finite Difference Method Finite Element Method FEM Lithium Iron Phosphate LFP Lithium-Ion Batteries LIBs LMO Lithium Manganese Oxide MLP Multilayer Perceptron NMC Lithium Nickel Manganese Cobalt Oxide RNN Recurrent Neural Network SEM Scanning Electron Microscopy XCT X-ray Computed Tomography

1. Introduction

In the context of pursuing sustainable development and improving the global climate, energy storage technology has become an essential area of focus for the global automotive industry and economic growth. Over the past decade, Lithium-ion Batteries (LIBs) have become the most widely used product in the energy storage field. However, along with their significant growth in popularity, the requirements for their performance also increase year by year [1]. Consumers and industries are demanding batteries with higher energy densities, faster charging capabilities, longer service life, and improved safety features while seeking to reduce costs [2]. In order to improve cell performance and reduce production expenditure, it is beginning to be recognised as important to investigate the effects of fabrication parameters on electrode structure and electrochemical performance at the microscopic scale [3-7]. Calendering represents a crucial stage in the electrode manufacturing process, where loose electrode particles are compressed to control the porosity of the electrode and enhance adhesion. During this procedure, the connectivity and volumetric energy density of the particles is increased. Furthermore, as the internal structure of the electrode evolves throughout the calendering process, other performance parameters, including mechanical properties and electron and ion transport properties, are also affected [8].

However, quality control and optimisation of the LIBs calendering process still face many challenges. The current limitations of traditional experimental and empirical methods have been identified as a significant obstacle to the development of stable and consistently improved product quality in the context of the highly complex and variable nature of the manufacturing process [9]. Due to their complex electrochemical characteristics, the operational mechanisms of LIBs are difficult to quantify and correlate with specific influencing factors accurately. Experimental-based research methods have drawbacks such as long development cycles, high costs, insufficient depth in microscopic mechanism analysis, and difficulty in evaluating the effects of multivariate synergistic, significantly increasing the challenges in the development of new electrodes [10]. In the past few years, the rapid development of computer simulation and Artificial Intelligence (AI) technologies has provided new opportunities to address these issues. Computational simulations offer an efficient alternative to complex and repetitive experimental optimisation processes, leading to significant savings in time and cost [11]. Therefore, modelling work focused on the calendering process of LIBs has become an essential area of research and practical application.

Currently, simulation studies on the macroscopic scale for LIBs cells, packs, and vehicle systems have become relatively mature, but

modelling research on the microscopic scale remains in its infancy. In this Review, we systematically examine and compare three different modelling approaches, empirical, numerical, and machine learning, for electrode calendering processes. The main contributions include providing a detailed evaluation of the strengths and limitations of each modelling method, highlighting the evolving trend towards hybrid approaches that integrate numerical precision with machine learning efficiency, and identifying critical future research directions focused on developing comprehensive, multiscale, and interpretable models. This comparative analysis uniquely positions our work to guide researchers in selecting and improving appropriate modelling strategies to meet the increasing accuracy and predictive demands of electrode manufacturing. The structure of the paper is as follows. Section 2 introduces the basic principles of electrode calendering and its impact on battery performance, providing a theoretical foundation for subsequent computational simulation studies. Section 3 sequentially presents the current research status of three modelling methods: empirical models, numerical models, and machine learning models. Section 4 offers a detailed comparison and in-depth analysis of these three modelling approaches, summarises the limitations of current research, and discusses future research directions.

This review is of substantial significance for industry, the research community, and policymakers on the regional and global scales. The insights presented here can guide more efficient, cost-effective, and environmentally sustainable production practices, enabling battery manufacturers to optimise processes and improve competitiveness. This review can also serve as a valuable reference for advising strategic investments and formulating targeted regulations aimed at advancing battery technologies. Furthermore, aligning with the United Nations Sustainable Development Goals, this paper underscores the necessity of optimising electrode production processes to reduce environmental footprints, improve resource efficiency, and support a sustainable energy infrastructure.

2. Fundamentals of LIB electrode calendering

2.1. Principles of calendering

Electrode calendering is a process used to customise the microstructure of an electrode to meet specific power density and energy density requirements for a given application [8]. This customisation is achieved by adjusting the thickness and porosity of the electrode. Typically, the conventional calendering method involves a two-roll compaction process, which ensures uniformity in both the surface and volume of the electrodes (Fig. 1). To promote uniform material distribution and minimise residual stresses during processing, this compaction is usually performed at elevated temperatures above the glass transition temperature of the binder [12].

This process, as illustrated in Fig. 2, has been simplified to allow a more detailed analysis of its mechanical behaviour. The electrode is assumed to be a homogeneous material subjected to compression by the action of a uniformly rotating roller. During this process, the electrode coating is subjected to a line load q_L of:

$$q_L = \frac{F_N}{\omega_C} \tag{1}$$

where F_N is the force applied to the coating and ω_C is the width of the coating.

The roll radius r_R , electrode width ω_C and thickness δ_{El,ρ_0} , and gap width δ_{Γ} together determine the effective contact area A_{Γ} of the electrode within the gap:

$$A_{\Gamma} = \omega_C(\sqrt{r_R(\delta_{El,\rho_0} - \delta_{\Gamma})} + \sqrt{r_R(\delta_{El,\rho_i} - \delta_{\Gamma})})$$
(2)

The contact area A_{Γ} and calendering speed v_0 determine the residence time of the coating in the roll gap. Longer residence times mean



Fig. 1. Schematic illustration of the changes in electrode microstructure during the calendering process. The blue spheres represent AM, the black spheres represent carbon, and the yellow stripes represent binders.



Fig. 2. Simplified mechanical diagram of the calendering process, including the roll force F_N , the electrode thickness before (δ_{El,ρ_i}) and after (δ_{El,ρ_i}) the calendering, the in-gap width (δ_{Γ}) , the roll radius r_R , and the inlet and outlet strings S_{I_R} and S_{Out} , Z_C describes the compaction zone and Z_R the relaxation zone of the coating. *Source:* This figure is redrawn with permission from Abdollahifar et al. [14].

more time for particle rearrangement, heat transfer, and a smoother compaction process with less shear stress.

Under the line loading, the particles undergo rearrangement or even fracture, leading to plastic deformation (as shown in Fig. 2 Z_C region) [13], and after leaving the roller, the electrode undergoes elastic recovery as line load decreases (as shown in Fig. 2 Z_R region).

The degree of compaction of the electrode during this process can be expressed as:

$$\Pi = 1 - \frac{\delta_{E,a}}{\delta_{E,initial}} \tag{3}$$

where $\delta_{E,a}$ is the electrode thickness after compaction and $\delta_{E,initial}$ is the initial thickness.

2.2. Impact of calendering on electrode performance

The calendering process affects the electrochemical properties of the electrode in three main ways: thickness, porosity, and tortuosity. Sometimes, particle size is also taken into account. Although calendering does not directly affect the particle size of the electrode composition, these together determine the mesostructure of the electrode.

The thickness of the electrode can affect essential parameters such as the active mass loading, ion transport distance, ohmic resistance, and mechanical integrity. In the absence of other variables, the mass loading of the active materials (AM) is directly proportional to the thickness of the electrode. It can be seen from Fig. 3(a–h) that thick electrodes are relatively more challenging to achieve high critical Crate. Hamed et al. [15] investigated the critical C-rate at which a steep decline in discharge capacity is observed as the current rate increases from 0.2 to 5C for 52 design variants of Lithium Manganese Oxide (LMO) electrodes (Fig. 3(i–k)). The rate-capability coefficient, γ , indicates the multiplier performance of the cell. An increase in active materials results in a reduction in the proportion of non-participating components in the electrode, thereby enhancing the energy density of the cell and reducing production costs (Fig. 4(a–c)) [16,17]. However, as the thickness of the electrodes increases, the ion transport paths and the ohmic resistance also increase. With these two in effect, this leads to an increase in energy density at the cost of losing power density, as shown in Fig. 4(a) [18–20]. The issue is further compounded at elevated C-rates. At the same time, as shown in Fig. 4(b), thin electrodes have better cycling performance because they are less prone to breakage and delamination during fabrication, which gives them better mechanical integrity and lower resistance to ion and electron transport [21].

Porosity is another critical parameter that is influenced by the calendering process and alters the overall performance of LIBs (Fig. 4(d-f)). Higher porosity usually means lower tortuosity, which facilitates the flow of ions through the electrode structure but reduces the electron conduction efficiency [22]. The study found through polarisation analysis that porosity affects not only the overall characteristics of electrical resistance, but also the quality of contact between the active material, the carbon binder, and the electrolyte [15]. Higher porosity is likely to lead to uneven contact within the electrode and a high local current density, which can accelerate local ageing and microstructural degradation, thus affecting long-term cycle stability. Although high porosity facilitates electrolyte wetting and improves active material utilisation, its negative impact on volumetric energy density and power density has led commercial electrodes to typically maintain porosity around 0.3 to balance performance, life expectancy and cost [23]. In addition, low porosity is more favourable for batteries to achieve highrate performance. According to Hamed et al. [15], the increase in electrode porosity significantly increases the critical C-rate of the cell (Fig. 3). From another perspective, the experimental results show that, with the same active material loading, the low porosity (0.3) electrode



Fig. 3. The sensitivity of the critical C-rate of Li-LMO cells to the porosity of the LMO electrodes (0.5 and 0.3) for the four groups of AM loading. (a,e) thin electrodes (AM-loading<18 mgcm⁻²); (b,f) normal electrodes (18<AM-loading<22 mgcm⁻²); (c,g) thick electrodes (22<AM-loading<26 mgcm⁻²); (d,h) ultra-thick electrodes (26<AM-loading<30 mgcm⁻²). The discharge-energy rate-capability coefficients (γ) of the 52 Li-LMO cells as a function of (i) AM loading, (j) thickness, and (k) porosity. *Source:* This figure is quoted with permission from Hamed et al. [15].

can achieve a capacity output that is about twice as high as that of the high porosity (0.4) electrode at a high rate (5C). This indicates that a lower porosity helps to form a denser, continuous electronic conductive network, thereby reducing ohmic polarisation losses and improving charge and discharge efficiency. However, it is important to note that LIBs are not single-purpose devices. Although a lower porosity is beneficial for electronic conduction, if the porosity is too low, it may weaken the ion transport channels in the electrolyte, resulting in local concentration differences and ion transport limitations. Not all applications require batteries to provide high charge/discharge rate capability. In scenarios where volumetric energy density and cell rate are not a priority, the high use of active materials (Fig. 4(e)) and high capacity retention (Fig. 4(f)) brought about by high porosity is an advantage that cannot be ignored. In addition, the content of inactive materials significantly influences the calendering process. Diener et al. [24] demonstrated that an increased binder content in the electrode formulation results in a reduction of the rebound effect due to enhanced plastic deformation, which in turn leads to a marked decrease in porosity. Similarly, Primo et al. [25] investigated the effects of calender pressure and temperature on electrode performance, finding that while porosity and mechanical properties are primarily determined by the applied pressure, the electrode conductivity is predominantly influenced by the calendering temperature. The latter observation is likely attributable to the increased deformability of the binder at higher temperatures. The research results [15] emphasise that the relationship between the macroscopic parameters of the electrode (such as thickness, loading, and porosity) and its performance (not just rate performance) is non-linear. This shows that changes in porosity can affect cycling performance through complex internal polarisation mechanisms

(including solid-state diffusion, electrolyte transport, and contact resistance between particles), requiring that manufacturing processes and subtle changes in microstructure be considered comprehensively in practical electrode design. Therefore, different electrode structures need to be designed for different LIB applications to achieve optimal performance.

The size of the pores and the size of the particles can work together to influence the tortuosity of the electrode. A high degree of tortuosity increases the resistance of the ions to flow through the electrodes in the electrolyte and reduces the wettability of the electrolyte. Fig. 4(g)shows two LiFePO₄ electrodes with the same porosity but different pore diameters; the lower pore diameter results in a lower degree of tortuosity and, therefore, exhibits a higher area capacity [26]. However, the effect of pore size on electrode performance is still poorly understood, and a valid measurement and statistical method is lacking. Similarly, the size of the active material determines the diffusion resistance of lithium ions in the solid state. As the particle size increases, the solidstate diffusion length increases. This is often considered one of the main limiting factors for the high-magnification performance of LIBs [27]. Nanoscale AM particles are suitable for high-rate charging and discharging and improve cycle stability (Fig. 4(h) and (i)). However, too small particles can exacerbate side reactions and consume more lithium ions, resulting in a lower cycle capacity [28,29].

Overall, the electrolyte-filled electrode pores act as a temporary reservoir for lithium ions, ready to be incorporated into the solid active material. Electrodes with smaller pores require faster ion transfer rates to maintain high current densities, as they deplete the ions stored in the electrolyte more quickly, which is extremely challenging. However, it is important to note that the effects of the calendering process do not always affect the anode and cathode in the same way. Compared with



Fig. 4. Effects of architectural parameters (thickness, porosity, pore size) on electrode performance. (a–c) Effect of thickness on (a) energy and power density, (b) cyclability, (c) cost. D–F: Effect of porosity on (d) energy and power density, (e) rate capability, (f) cyclability. (g) Effect of pore size on rate capability. (h–i) Effect of particle size on (h) stability, and (i) cyclability.

Source: This figure is quoted with permission from Zhu et al. [30].

the anode, the Cathode Active Material exhibits a more pronounced electrochemical enhancement effect during the calendering process because of its lower conductivity in the actual performance. In this process, the cathode active material particles are densely packed, which facilitates better integration with conductive additives. Furthermore, the calendering process primarily reduces the plastic deformation energy of the electrode while retaining most of its elastic deformation energy. This effectively minimises irreversible capacity loss during battery cycling, thus improving cycle life and performance [31].

Nevertheless, calendering is not always beneficial; for example, it is essential to maintain an appropriate solid-state diffusion rate during this process. Excess calendering can lead to excessively low porosity, hindering lithium ion diffusion into the electrode matrix and causing polarisation during the initial stabilisation of the solid-electron interaction [8]. This can degrade the high-rate performance of the battery and pose safety risks, particularly for electrodes composed of large particles. Studies [32–35] have shown that higher operating temperatures and smaller AM particles can overcome the reduction in reaction kinetics caused by a decreased porosity. However, this comes at the expense of safety and energy density. Furthermore, a compact pore structure also reduces the surface energy, making the electrode less attractive to the electrolyte. This is not conducive to the infiltration of the electrolyte on the electrode surface and the formation of the SEI layer [36]. The dense electrode particles are also detrimental to the effective wetting of the electrolyte, which is important to ensure that the electrode can participate fully in the electrochemical reaction.

The dramatic changes in the mesoscopic structure of the electrode particles during calendering significantly impact the electrochemical properties compared to other process steps [38]. However, a large number of studies [10,39-43] have demonstrated that the range of densities and porosities that can provide the best mechanical and electrochemical properties is relatively limited. In previous years, balancing the electronic and ionic conductivities and determining the ideal porosity have been the main challenging objectives of the calendering process. Meanwhile, it is important to note that although the application of high pressures during the electrode calendering process can reduce porosity and improve electronic contact and conductivity, it simultaneously introduces significant mechanical stresses within the electrode [44]. The stress concentrations that develop between AM particles and between particles and the binder can promote the initiation and propagation of microcracks. Over repeated cycles, these microcracks may eventually coalesce, compromising the mechanical



Fig. 5. Schematic diagram of the evolution of the microstructure and pore size of the electrode during the calendering process. (a) SEM images showing the evolution of NMC cathode morphology under incremental line load. (b–c) Evolution of the porosity and pore size, (b) porosity and pore size of macropores, (c) pore size of mesopores and average pore diameter of the NMC cathode. (d) The morphology of spherical graphite anode. (e) Evolution of the specific surface area and micropore pore size. (f) Particle rupture due to friction and compression. (g) SEM images of the near-surface layer (the red curve indicates the possible flow paths of the electrolyte). (h) The morphology of the layered graphite anode. (i–j) Electronic conductivity evolution of graphite anode, (i) spherical, (j) layered. (k–l) Relationship between the (k) line load and (l) porosity on the resistance of the NMC cathode. Colour backgrounds highlight the inflexion points of data changes. *Source:* This figure is quoted with permission from Zhang et al. [37].

integrity of the electrode, reducing the effective use of active materials, and accelerating capacity decay [45]. In addition, mechanical stresses can induce fatigue in the binder, weakening the interfacial adhesion between the binder and AM particles. Such interfacial failures not only lead to a loose electrode structure but also disrupt localised electron transport, thereby increasing the internal resistance of the battery and hastening cycle degradation. Moreover, the mechanical stresses induced by the calendering can generate new surface defects within the electrode, which may serve as nucleation sites for the inhomogeneous formation of the solid electrolyte interphase. Although this layer initially acts as a protective barrier, its continuous growth consumes active lithium, resulting in irreversible capacity loss. Furthermore, the nonuniform distribution of the solid electrolyte interphase can exacerbate localised stresses and temperature rises, potentially triggering thermal runaway [44]. Consequently, there is a considerable demand for studies of electrode densification based on theoretical principles rather than experience.

2.3. Experimental studies on electrode calendering

Over the past few years, experimental studies on electrode calendering primarily focused on tracking changes in the internal microstructure of porous electrodes using techniques such as Scanning Electron Microscopy (SEM) and X-ray Computed Tomography (XCT). For instance, Zhang et al. [37] utilised SEM to investigate the microstructural and pore size evolution of NMC622 samples under progressively increasing linear loads (Fig. 5(a)). SEM images clearly reveal that before calendering, the active particles within the cathode are widely spaced, the pore distribution is uniform, and the contact area with the current collector is minimal. As the linear load applied to the electrode surface increases, large AM particles undergo deformation and fracture, forming a more densified structure. When the linear load exceeds 1960 N/mm, significant deformation and fusion occur in the Carbon Binder Domains (CBD), and the pore size decreases markedly. Fig. 5(b–c) details the trends of the changes in pore size and porosity with increasing linear load. With higher calender density, the mode, median, and mean diameters of macropores (>50 nm) decrease exponentially until convergence, indicating a more uniform pore size distribution at lower porosity. In contrast, the diameters of the mesopores (2–50 nm) decrease linearly without a dramatic reduction.

Fig. 5(f) clearly illustrates the fracture of large AM particles due to compression during the calendering process. This fracture increases the specific surface area of the particles (Fig. 5(e)), allowing faster penetration of the electrolyte into the interiors of the large particles, thus enhancing the cycling rate. However, excessively exposed cracks and particle wear can lead to more severe cracking during subsequent charge–discharge cycles, preventing electrolyte transport (Fig. 5(g)). This results in the loss of cyclable AM and accelerates the formation of side reactions, ultimately leading to a decrease in cycling capacity.

For graphite anodes, calendering also affects their interfacial resistance. Spherical graphite, which is difficult to deform plastically (Fig. 5(d)), experiences a significant decrease in resistivity due to particle rearrangement and densification caused by calendering (Fig. 5(i)). However, the situation is markedly different for layered graphite electrodes. Their microstructure changes from a disordered interwoven arrangement before calendering to a distinctly oriented arrangement parallel to the current collector after calendering (Fig. 5(h)). Consequently, their resistance first increases and then decreases to a value comparable to the initial resistance (Fig. 5(j)). This behaviour is attributed to the highly anisotropic electronic conductivity of the graphite layers: the electron conductivity is stronger parallel to the graphite layers and weaker in the perpendicular direction. Therefore, as the linear load increases, the pathways for current to penetrate the graphite in the perpendicular direction become much longer, leading to an increase in resistance. Subsequently, as densification increases, the contact area between the graphite particles enlarges, resulting in a decrease in resistance. Fig. 5(k-l) illustrates the effect of calendering on the resistance of the cathode. Overall, due to conductive additives, even slight calendering can reduce resistance to near-critical levels.

Advancements in experimental techniques have contributed significantly to the development of simulation methodologies. Highresolution microscopic imaging techniques, such as SEM and XCT, enable the quantitative characterisation of particle size, shape, distribution, and interparticle contact states. Additionally, these techniques provide critical insight into the distribution of binders and interfacial conditions. The data obtained serve as essential input parameters for physics-based multiscale models, allowing for a more accurate representation of the material heterogeneity and localised damage mechanisms occurring during the calendering process. Furthermore, experimental methods such as microscratch and peel tests facilitate the quantitative assessment of interfacial adhesion strength and damage evolution. These measurements support the development of more precise contact models and fracture criteria, thereby improving the predictive capacity of numerical simulations for crack propagation and failure processes. Moreover, microscale stress and strain distributions, along with interparticle interaction data obtained from experiments, can be used to refine these parameters in models and establish couplings with the mechanical response of continuum media. By systematically comparing experimental results with numerical simulations, model deficiencies can be identified, and parameters can be iteratively refined to enhance the accuracy of the model in representing real-world processes. Experimental data not only serve as validation benchmarks for simulation outputs, such as local stress fields, fracture energy, and deformation behaviour, but also provide a foundation for sensitivity analyses and process parameter optimisation.



Fig. 6. Initial coating porosities and minimally achieved, extrapolated and predicted coating porosities regarding the variation of active material, mass loading, composition and processing.

Source: This figure is quoted with permission from Meyer et al. [46].

3. Modelling approaches for electrode calendering

3.1. Empirical models

The empirical modelling approach focuses on simplified analysis of existing results based on the laws of physics, leading to the derivation of mathematical equations using analytical methods. For example, Meyer et al. [38] constructed an empirical model that can represent the relationship between line load and electrode density ($0 \le \Pi \le 0.25$) based on their study of the electrode calendering process. It can be expressed as:

$$q_L = A(\rho_{C,i} - \rho_{C,0})^B \tag{4}$$

where *A* and *B* are the correction factors, which are related to the electrode components and particle morphology. $\rho_{C,0}$ and $\rho_{C,i}$ are the coating densities before and after calendering, respectively.

For the calendering process of NMC at lower circumferential speeds, the relationship between coating porosity and line load can be fitted by the following formula:

$$\varepsilon_C = \varepsilon_{C,min} + (\varepsilon_{C,0} - \varepsilon_{C,min}) \exp(-\frac{q_L}{\gamma_C})$$
(5)

where $\varepsilon_{C,min}$ is the minimum porosity of the coating, $\varepsilon_{C,0}$ is the initial porosity of the coating and γ_C is the damping correction factor, and the relationship between it and the active mass loading M_C is:

$$\gamma_C = (\mu_0 - \xi T_R) M_C \tag{6}$$

where μ_0 is the mass factor at 0 °C, ξ is the temperature factor, and T_R is the roll temperature.

They found that the electrode calendering process can be reduced to a sphere-filling-based model. Its initial structure can be regarded as a simple cubic pack consisting of rigid spheres of the same size, and its minimum porosity $\epsilon_{C,min}$ is close to the theoretical porosity of a cubic closed pack. Therefore, a compaction factor p can be introduced:

$$\varepsilon_{C,\min} = p\varepsilon_{C,0} \tag{7}$$

The experimental results — Fig. 6 — show that for some common cathode materials, the compaction factor p can be used as a predictor of the minimum coating porosity.

On this basis, Eq. (5) can be simplified using this compaction factor:

$$\varepsilon_C = \varepsilon_{C,0}(p + (1 - p)\exp(-\frac{q_L}{\gamma_C})) \tag{8}$$

However, it should be noted that for NMC, LMO, and graphite anodes, the predicted values deviate significantly from the experimental values. This may be attributed to the fact that the initial structure of NMC and LMO has low sphericity and is not suitable for this spherical simplified model. In contrast, for graphite anodes, the lamellar structure allows for denser compression.

In summary, the results of Meyer et al. have shown the relationship between the calendering load and the porosity of the electrode coating. Their conclusions are expressed in mathematical formulae by constructing a relatively idealised model supplemented by correction factors. However, their limitations are obvious, and the correction factor does not have good applicability and generalisability.

Previous research [21,47–50] has indicated that the lithium ion concentration of the electrolyte within the pores of the electrode is distributed in a gradient during the discharge of the battery. As the current density increases, the decay of the lithium ion concentration gradually increases until the lithium ion concentration around the AM in close proximity to the current collector region is reduced to zero. At this point, the electrodes have reached the maximum current density I^* that allows all AM to participate in the reaction, as there are no lithium ions in the electrolyte available for AM in this region to participate in the electrochemical reaction when it continues to increase.

Based on this principle, Tambio et al. [51] proposed the Penetration Depth Model to analyse the effect of calendering-induced microstructural changes on the electrochemical properties of electrodes. The penetration depth L_d of lithium ions in the electrolyte can be expressed as:

$$L_d = \frac{\varepsilon}{T} \frac{D_0 C_0 F}{(1 - t_{\rm Li})I} \tag{9}$$

where ϵ is the porosity of the electrode, *T* is the tortuosity factor of the matrix, D_0 is the diffusion coefficient of the lithium salt in the electrolyte, $t_{\rm Li}$ is the number of lithium ions transported, *I* is the current density, and *F* is the Faraday constant.

The effect of porosity ε and matrix tortuosity factor *T* on effective ionic conductivity κ_{eff} and diffusivity D_{eff} can be expressed as:

$$\kappa_{eff} = \kappa_0 \frac{\varepsilon}{T} \tag{10}$$

$$D_{eff} = D_0 \frac{\varepsilon}{T} \tag{11}$$

where κ_0 and D_0 is the bulk electrolyte conductivity and diffusivity. If only considering the effect of geometric tortuosity, the matrix tortuosity factor can be expressed as:

$$T = \tau^2 \tag{12}$$

Mathematically, the ratio of the actual capacity Q_I of the cell at a current density above the critical current density to the nominal capacity Q_0 is equal to the ratio of penetration depth L_d to the thickness of the design electrode L:

$$\frac{Q_I}{Q_0} = \frac{L_d}{L} \tag{13}$$

Based on these conclusions, the Penetration Depth Model points out that the capacity of the battery at a high rate can be expressed as:

$$Q_I = Q_0 \frac{\epsilon}{TL} \frac{D_0 C_0 F}{(1 - t_{\rm Li})I} \tag{14}$$

This Penetration Depth Model can quantitatively predict the relationship between the specific discharge capacity and the areal current density based on the measured porosity, tortuosity, and diffusion coefficient (as shown in Fig. 7).

Nevertheless, it is important to note that although the Penetration Depth Model has been shown experimentally to be a practical tool for



Fig. 7. Areal discharge capacities as a function of the areal current density for NA3b, NA7b and NA4b and the corresponding Penetration Depth Model fits (dotted lines) at temperature 22 $^{\circ}$ C.

Source: This figure is quoted with permission from Tambio et al. [51].

evaluating the phenomenon of a sudden drop in the usable capacity of porous electrodes after a critical value of current density is reached, it has some obvious limitations that limit its wider applicability. For example, the model is only well fitted to specific electrode materials (NA3b and NA7b). And, as indicated by Tambio et al. [51], the model can only accurately simulate the electrochemical behaviour of the electrodes near room temperature. At high temperatures, such as 40 °C, the model does not provide reliable predictions. Temperature is a key factor in electrochemical reactions, affecting the kinetics, diffusion rates, and stability of the overall system. The model is unable to account for the effects of temperature variations.

In summary, employing empirical models to study the electrode calendering process is relatively straightforward and efficient. These models require less computational effort and time compared to more complex numerical simulations, making them attractive for preliminary analyses and for gaining fundamental insights into the calendering mechanisms. Empirical models can provide quick evaluations of the effects of various parameters on the overall behaviour of the electrode, which is particularly useful in the early stages of research and development. However, due to the simplifying assumptions inherent in their formulation, they often fail to capture the intricate microstructural changes that occur during the calendering process. In addition, these models typically overlook the non-linear and heterogeneous nature of electrode materials, resulting in very significant limitations.

3.2. DEM-based numerical models

The numerical modelling approach focuses on solving complex differential equations based on numerical models and computer simulation techniques by discretising continuous problems into independent elements or nodes. The commonly used solution methods are Discrete Element Method (DEM), Finite Element Method (FEM) and Finite Difference Method (FDM). Several experimental studies have been conducted to investigate the impact of the calendering process on the electrode structure and cell performance with the assistance of numerical modelling. DEM has been demonstrated to be a valuable approach for investigating the microstructure of electrodes by modelling the motion and forces of particles [53,54]. Ge et al. [52] proposed a DEM numerical model for analysing the structural evolution, mechanical stress, and transport properties of the electrode calendering process (e.g., tortuosity, anisotropic, force network, stress tensor, etc.), based on real cathode microstructure data obtained by using X-ray tomography imaging. The 2D voxelised electrode structure information (as shown in Fig. 8(a)) generated by the simulation results can help predict



Fig. 8. Simulation schematic of electrode structure evolution under different calendering forces. (a) Voxelised 3D electrode structures and 2D cross-sections from DEM simulations under different compression pressures (In the 2D cross-section, the white voxels are AM particles, and black voxels are voids). (b–c) Diagonal components of (b) the deviatoric part of the fabric tensor, and (c) the stress tensor under different compression pressures. (d–e) Tortuosity factor with/without binder phase 2D visualisation of electrode structure parallel to the through-plane direction and normalised potential distribution without/with binder phase (the binder phase is coloured in red). (f) Tortuosity factor plotted as a function of porosity.

Source: This figure is quoted with permission from Ge et al. [52].

changes in key information such as porosity and tortuosity during the calendering process.

The fabric tensor and the stress tensor of the electrode material can be quantitatively analysed by using the simulation data from the DEM model. The fabric tensor Φ_{ij} is used to assess the directionality of the particle contact, which is defined as the average value of the outer product of the normal contact *n*:

$$\Phi_{ij} = \frac{1}{N_c} \sum_{a=1}^{N_c} n_i n_j$$
(15)

where n_i and n_j are the projections of the normal contact n in the Cartesian coordinate system. As shown in Fig. 8(b), as the calender stress increases, Φ'_{xx} and Φ'_{yy} show the same growth trend, while Φ'_{zz} shows a completely opposite trend. Similarly, the stress tensor σ_{ij} , which reflects the interaction force between the particles, shows the same trend (as shown in Fig. 8(c)) and can be described as:

$$\sigma_{ij} = \frac{1}{2V} \sum_{\alpha=1}^{N_c} (r_{1i} F_{1j} + r_{2i} F_{2j}) \tag{16}$$

3.7

where r_{1i} and r_{2i} are the relative positions of two particles in contact with each other and F_{1j} and F_{2j} are the interaction forces. The

difference in the gradual increase of the components along the *Z*-axis reveals the inhomogeneity and anisotropy of the structure during the calendering process. Fig. 8(d) illustrates the effect of the potential distribution in porous electrodes using the level set algorithm with and without considering the bonding phase, respectively. On this basis, the tortuosity-porosity can be fitted according to the generalised effective medium equation $\tau = a\epsilon^b$. The results (Fig. 8(f)) show that the fitted curves are highly close to the Bruggeman equation when the bonded phase is not considered ($\tau = 0.885\epsilon^{-1.068}$), but the experimental results are closer to the fitting results when the bonded phase is considered ($\tau = 1.02\epsilon^{-0.53}$). This work demonstrates for the first time that the tortuosity of calendered electrodes can be predicted by combining DEM simulations and bonded phase generation algorithms.

However, it should be noted that the results of the DEM simulations still differ considerably from the experimental values, which Ge et al. [52] attribute to the fact that the actual AM particles do not stay ideally spherical after compression and that the model does not take into account the effects of particle fragmentation. In a subsequent report [55], this team attempted to use ellipsoids instead of spheres in order to simulate the deformation and directionality of the particles.

The DEM simulation results are compared with the XCT results. The two methods of calculating the pore size distribution, i.e., Continuous



Fig. 9. Electrode microstructures and pore size distributions before and after calendering based on tomography reconstructions and analytical approximations. (a) Electrode structures from tomography scans, spherical approximations and ellipsoidal approximations. Porosity distribution and 50% pore radius for both (b–c) uncalendered and (d–e) calendered structures based on Continuous Pore Size Distribution and Mercury Intrusion Porosimetry Pore Size Distribution. *Source:* This figure is quoted with permission from Ge et al. [55].

Pore Size Distribution and Mercury Intrusion Porosimetry Pore Size Distribution, are also considered. The comparison results are shown in Fig. 9. The Continuous Pore Size Distribution method calculates the cumulative pore size distribution by gradually adding spheres of increasing radius to the pores, while the Mercury Intrusion Porosimetry Pore Size Distribution method simulates the intrusion of mercury into the porous electrode. Both show the same trend in the results, but Mercury Intrusion Porosimetry Pore Size Distribution decreases slightly more in the middle phase. From the final statistics, the structure of the perfect spherical approximation is closer to the real performance than the ellipsoidal approximation. Therefore, this problem has not yet been effectively solved.

In order to further explore the effect of the shape of the AM particles on calendering simulations, Nikpour et al. [56] attempted to create particles with different morphologies to explore their potential effects. Fig. 10 illustrates the mechanical properties, including coating thickness, porosity, particle surface area, and Young's modulus, of AM particles with various shapes following drying and calendering processes. In addition, it displays their electrochemical properties, such as electronic conductivity and the MacMullin number N_M , which is the ratio of the ionic conductivity of the pure electrolyte to the ionic conductivity of the electrolyte with separators. The simulation results show that the disk particles have the highest thickness variation and porosity variation compared to the spherical and rod particles, which indicates a higher degree of stacking. It reflects the same trend in the electrochemical performance-related parameters. However, irregular AM particles induce the formation of a looser and softer porous structure, leading to a reduction in stiffness. In conclusion, calendering has different effects

depending on the shape of the active material. However, the increase in electronic conductivity and MacMullin number after calendering is mainly due to the compression of the micropores inside the CBD phase rather than the AM particle shape difference itself, which is more sensitive to differences in particle surface area.

Similarly, Xu et al. [57] carried out a more detailed study of this problem to resolve the situation where particle deformation could not be observed in the DEM simulation. They used the XCT results of the NMC111 sample as the basis for the shape data and converted the 2D scanning phase diagram into a 3D matrix. Subsequently, small spherical particles were used to populate this matrix in the DEM model to construct AM particles that matched the true morphology. The initial structure of the electrode coating model is generated by the random rotation and displacement of a large number of AM particles with different shapes (as shown in Fig. 11(a)).

The comparison of the simulation results with the experimental data is shown in Fig. 11(b) and (c). It can be seen that the model can better predict the changes in porosity and tortuosity of the electrode structure during the calendering process. Fig. 11(h–i) visualises the deformation of AM particles constructed from small spherical particles during calendering. However, it is also important to note that the accuracy of the model decreases as the AM content decreases.

In addition, Xu et al. [58] attempted to use a bonding model on this basis to simulate the effect of particle fracture. The maximum normal and shear stresses of the bond can be calculated from the following equations:

$$\sigma_B^n = -\frac{|F_B^n|}{A_b} + \frac{|M_B^r|R_{min}}{I}$$
(17)



Fig. 10. Effect of AM morphology on electrode performance. (a–c) Simulated cross sections for (a) disk, (b) sphere, and (c) rod AM shapes at three stages of coated, dried, and calendered. LD and HD denote high and low-density CBDs. (d–i) The simulated dried and calendered (d) film thickness, (e) porosity, (f) AM surface area, (g) electronic conductivity, (h) MacMullin number, and (i) Young's modulus.

Source: This figure is quoted with permission from Nikpour et al. [56].

$$\sigma_B^t = \frac{|F_B^t|}{A_b} + \frac{|M_B^n|R_{min}}{J}$$
(18)

where *A* is the contact area, *I* and *J* are the moment of inertia and polar moment of inertia of the section, respectively. M_B^n and M_B^t represent the axial and shear incremental moments of the bond, respectively. *F* is the Hertzian elastic force between two pairs of particles in contact:

$$\Delta F_B^n = \frac{E_b A_b}{l_b} v_r^n \Delta t \tag{19}$$

$$\Delta F_B^t = \frac{(E_b/2(1+P_{Ob}))A_b}{l_b} v_r^t \Delta t$$
(20)

where E_b and P_{Ob} are Young's modulus and Poisson's ratio of the bond, respectively. v is the relative velocity, and l_b is the length of the bond.

Fig. 12 shows the evolution of the shape of a randomly selected AM particle (Fig. 12(c)) during the calendering process, as well as the bonds to be broken (blue region in Fig. 12(d)). The fracture of the

AM particles was found to not depend on the linear variation of the applied force. When the pressure exceeded 120 MPa, the bond breakage rate within the particles reached more than 10% (as shown in Fig. 12(b)), and there was an increased probability of particle fracture. The distribution of cracks within the electrode is not uniform and is related to the distribution or orientation of the AM particles in the coating and the applied force.

In conclusion, this study presents a novel electrode microstructure model that offers a more precise representation of the actual particle shape and is more closely aligned with the actual situation than previous models based on spherical particles. This model is capable of tracking the deformation and orientation changes of the AM particles throughout the calendering process, and its predictions are in good agreement with experimental data. In addition, this model has the potential to predict and visualise the pressure-induced fragmentation of AM particles during the calendering process. However, this modelling



Fig. 11. The process of DEM numerical model construction and simulation results considering the real shape of the active material in the calendering process. (a) Schematic representation of the NMC111 AM structure generation procedure. (b) Comparison between experimental and simulated porosity for both electrode compositions as a function of compression degree. (c) Tortuosity factor as a function of the porosity during the simulated calendering and comparison with μ -XCT data. (d–e) The 3D structure resulting from the calendering model. (f–g) Pore, CBD and AM volume distribution along the thickness direction, represented by black, red and blue curves respectively. (h–i) Cross-sectional view for the electrodes. The blue represents the CBD, and the red, from light to dark, represents different sizes of AM.

Source: This figure is quoted with permission from Xu et al. [57].

approach requires the generation of a large number of primary spherical particles and is therefore computationally inefficient (requiring up to 216 h for a single run on a 2 of Intel Xeon E5-2680 v4, 2.4 GHz 14 cores platform) and does not allow for more complex simulations due to computational limitations.

It is worth noting that most of the literature reviewed so far is based on the study of parameters such as electrode porosity, tortuosity, and conductivity. Compared to these fundamental parameters, the force and elastic recovery behaviour of the electrodes during calendering is more difficult to simulate accurately. In order to study in detail the evolution of the mechanical behaviour of electrodes during calendering, Schreiner et al. [59] proposed a method for parameterising the calendering model and discussed in detail the effects of various parameters of the model on the load–displacement curves.

Schreiner et al. [59] point out that the mechanical parameters of the model can be calibrated by nanoindentation experiments, including compressive strength, elastic rebound, and initial particle cohesion. Nanoindentation experiments are a method for determining the mechanical properties of a material. This is achieved by applying a tiny indenter to the electrode material, which allows measurement of the displacements and corresponding forces generated during compression. The study of particle contact parameters between different materials is particularly important because the introduction of contact models and corrective parameters, such as virtual bonds, makes it impossible to accurately calculate specific values for each parameter directly from mechanical tests. The study indicated that for three common types of offset in simulation results, the loading curve of the model could be adjusted by correcting the parameters of critical shear or normal stress and surface energy (Fig. 13(a)), the maximum compression pressure by correcting Young's modulus parameter (Fig. 13(b)), and the elastic recovery curve by correcting the contact plasticity ratio parameter (Fig. 13(c)).

In conclusion, the model parameterisation concept and calibration methodology proposed in this study are of considerable significance and a far-reaching influence, extending the research scope for DEM numerical models. Nevertheless, it is important to note that there is still a notable discrepancy between the simulation results and the experimental values obtained in this study. This highlights the need for further research to investigate the evolution of the mechanical behaviour of the electrodes.

Furthermore, Lundkvist et al. [40] indicated that electrode coatings have a pressure-sensitive behaviour, i.e. they have different mechanical properties under tension and compression. Existing models have treated CBD as extremely small spherical particles that adhere around AM particles. However, this makes it difficult to relate the mechanical behaviour of the active layer to the contact properties of the CBD and AM, and thus, to explain the differences in tensile and compressive processes. To solve this problem, they innovatively considered the CBD as flat cylinders and controlled the stiffness of the CBD by adjusting the radius ratio of the cylinders to characterise the mechanical properties of the electrode coatings in tension and compression (shown in Fig. 14(a)). Compared to spherical particles, cylindrical particles have a larger contact area with neighbouring particles, and their mechanical behaviour is directional and anisotropic.

Four types of CBD particles with different stiffnesses were selected to investigate the effect of CBD elastoplastic behaviour on the active layer load-displacement curve during calendering. For cylindrical CBD with different radius ratios, the number of interparticle contacts is similar and increases with higher strain levels. The simulation results (Fig. 14(b)) show that the calender pressure is linearly related to the square of the binder radius ratio, while the elastic rebound remains relatively constant. Studies of the plastic behaviour of adhesives have similarly demonstrated that plastic deformation of binders, whether in tension or compression, does not significantly affect the stiffness of the unloading (Fig. 14(c) and (e)). This indicates that plastic deformation occurs only at the elastic-binder-particle contact. In addition, the model successfully reproduced the experimentally observed range of different variations in unloaded stiffness under tensile and compressive conditions (Fig. 14(d)). However, an accurate numerical reduction was not achieved. This modelling framework provides a good basis for further analyses of the mechanics of the electrode layer, such as the expansion of the AM particles due to electrochemical loading.

In summary, the development of DEM numerical models for the electrode calendering process remains in its infancy. While numerous studies have employed DEM to simulate the evolution of electrode microstructures during calendering, significant challenges persist. The inherent complexity of DEM modelling has hindered the accurate and efficient simulation of the deformation, fracture, and fusion processes of AM. There is also a lack of attention to the stresses and strains experienced by the electrodes and the movement of the particles during



Fig. 12. Schematic diagram of the calendering evolution for the fracturable DEM model and its simulation results. (a) Cross-sectional diagram of the volume. The position of the studied particles within the electrode is indicated. (b) The pressure applied on the simulated electrode and the number of bonds break of this individual particle. (c) Particle shape evolution during the calendering. (d) The change of the bonds within the selected particle during the calendering. The blue indicates the bonds that will break during this compression, while purple indicates bonds that remain intact throughout. *Source:* This figure is quoted with permission from Xu et al. [58].



Fig. 13. Experimental values and simulation results for a typical NMC622 load-displacement curve. (a) Typical load-displacement curve for an NMC622 electrode before, during and after compaction. (b) Comparison of the load-displacement curves for the simulation and experiment and the number of broken bonds. *Source:* This figure is quoted with permission from Schreiner et al. [59].

calendering. Moreover, the CBD, being a nanoscale microporous material with particle sizes enormously different from those of AM particles, presents substantial difficulties in accurately representing its physical properties within DEM simulations. Consequently, there is still a lack of convincing DEM numerical models capable of precisely replicating the various mechanical behaviours exhibited by electrodes during calendering. Addressing these challenges is urgent and represents a critical focus for future research endeavours.

3.3. FEM-based numerical models

In the simulation of the electrochemical performance of electrodes, FEM holds a crucial position due to its effectiveness in modelling complex electrochemical phenomena. FEM is particularly suitable for simulating electrochemical behaviour because it allows for detailed analysis of spatially varying fields such as potential, concentration, and current density within complex electrode geometries. It facilitates the coupling of multiple physical processes, including ion transport, electron conduction, and electrochemical reactions, providing a comprehensive understanding of the behaviour of the electrode under various operating conditions. However, since each phase within the porous electrode microstructure requires individual modelling and meshing, the computational cost of this method becomes extremely high when full-sized batteries are modelled. Consequently, FEM was initially predominantly used to study various behaviours of electrodes at macroscopic scales.

For example, He et al. [60] employed COMSOL Multiphysics to develop a 3D model of a soft-pack Lithium Iron Phosphate battery



Fig. 14. The electrode modelling process using columnar particles to represent CBD and its mechanical performance compared to the experiment. (a) The RVE of the positive electrode for the different steps in the simulation process: random packing, uncalendered electrode layer, calendering process, and in-plane mechanical loading. (b) Calendering surface pressure to surface height for four binder radius ratios. (c) Calendering pressure for the elastic and elastic–plastic binder. (d) Unloading stiffness for four binder radius ratios compared with the experimental results. (e) Unloading stiffness for the elastic and elastic–plastic binder compared with the experimental results. *Source:* This figure is quoted with permission from Lundkvist et al. [40].

(LiFePO₄). In their model, they neglected the volume changes of electrode particles during charging and discharging, as well as the formation of the SEI and lithium dendrites. They assumed that the interior of the battery could be represented as a continuum comprising overlapping solid and electrolyte phases. Using this model, they investigated the electrolyte diffusion behaviour and lithium ion transport within porous electrode materials under high current rates. Fig. 15(a–b), respectively, present comparisons between simulated and experimental results for voltage and temperature variations of the battery under different C-rates. The simulation results closely match the experimental data, demonstrating the validity of the model. It is evident that distinct voltage plateau are observed at various discharge rates, and the voltage plateau decreases as the discharge rate increases. This indicates that the model effectively captures the electrochemical dynamics of the battery under different operating conditions.

Fig. 15(c-e) (anode) and (f-h) (cathode) illustrate the distribution of the overpotential within the electrodes at a discharge rate of 5C. The overpotential is a critical parameter in electrochemical systems, defined as the difference between the actual electrode potential under load and its equilibrium (open-circuit) potential. The energy levity arises because of kinetic barriers such as charge-transfer resistance, mass-transport limitations, and ohmic losses within the electrode and electrolyte. The overpotential directly influences the cell voltage, efficiency, and overall battery performance. As the depth of discharge increases, the anode and cathode electrodes exhibit entirely opposite trends in overpotential distribution. This behaviour reflects the differing electrochemical reactions and kinetics of the two electrodes during discharge. However, regardless of the electrode type, at the initial stage of discharge, the regions near the current collectors (tabs) are highly utilised first due to shorter electronic conduction pathways and lower resistance. The regions farther from the tabs begin electrochemical reactions more slowly, likely due to increased internal resistance and longer pathways for ion and electron transport. This non-uniform utilisation can lead to uneven degradation and reduced efficiency. Furthermore, the current density distributions at the electrode-separator interfaces (Fig. 15(ik) for the anode electrode and Fig. 15(l-n) for the cathode electrode) corroborate this observation, showing higher current densities near the tabs and lower densities farther away.

Over the past few years, most FEM simulations of LIBs are concentrated on the macroscopic scale, with relatively few studies focusing on the mesoscopic or even microscopic scales. However, macroscopic continuum models face significant challenges in capturing the microstructural changes induced by calendering. Specifically, these models often struggle to accurately represent the intricate details of particle rearrangement, deformation, fracture, and contact mechanics that occur at the micro level during the calendering process. The homogenisation inherent in macroscopic models overlooks the heterogeneities and anisotropies present in the electrode materials, such as variations in particle size distribution, binder distribution, and porosity. This simplification can lead to less accurate predictions of mechanical stresses, electrochemical performance, and overall battery behaviour under operational conditions. Moreover, macroscopic models typically assume uniform material properties and neglect the local interactions between particles and phases, thus ignoring phenomena such as crack initiation, propagation, and the formation of conductive pathways. As a result, these models may fail to predict localised failures or performance degradation mechanisms that originate from microstructural features. The inability to capture these detailed microstructural effects limits the effectiveness of macroscopic models in optimising electrode design and processing conditions. In this context, some of the outstanding advances in FEM simulations over the past few years at smaller scales will be reviewed to highlight new insights and identify future research directions.

Boyce et al. [61] employed XCT images alongside the standard filtered back-projection algorithm [62] to reconstruct the threedimensional morphology of electrodes with high precision. Subsequently, the discrete transport and electrode dynamics equations were solved in FEM software to investigate the influence of electrode thickness and porosity on its discharge performance. The XCT sample and the model construction process are shown in Fig. 16(a–c).

The simulation results show that the discharge performance of the electrode is strongly affected by the electrode thickness and porosity. When other factors are kept constant, the discharge capacity at 1C increases significantly when porosity increases (Fig. 16(e)). When the electrode thickness increases, the discharge capacity decreases dramatically (Fig. 16(f)). In addition, this phenomenon is more pronounced



Fig. 15. The continuous medium electrochemical numerical model and the overpotential and current density distribution map. (a–b) Comparison between simulation results and experimental data, (a) variation of voltage and (b) variation of temperature with time. (c–h) Overpotential distribution within the electrodes at different times at the discharge rate of 5C, (c–e) positive electrode, and (d–h) negative electrode at t = 60, 360, and 660 s, respectively. (i–n) Current density within the electrodes at different times at the discharge rate of 5C, (i–k) positive electrode, and (l–n) negative electrode at t = 60, 360, and 660 s, respectively. (i–n) Current density within the electrodes at different times at the discharge rate of 5C, (i–k) positive electrode, and (l–n) negative electrode at t = 60, 360, and 660 s, respectively. *Source:* This figure is quoted with permission from He et al. [60].

under high-rate discharge conditions. Fig. 16(g) shows that when the electrode thickness exceeds 160 μ m or the porosity is less than 0.25, the electrode can hardly meet the 5C discharge rate.

This can be attributed to the fact that as the length of the diffusion path increases, the resistance of lithium ions to traversing the tortuous path through the macrostructure during discharge also increases, resulting in a decrease in discharge capacity. In contrast, as the macroporosity increases, the tortuosity decreases, resulting in more uniform diffusion and electrolyte within the particles. The increased porosity also provides a larger surface area for exposure to the active particles, which promotes a higher utilisation rate of the active material. Fig. **16**(d) shows the lithium-ion concentration distribution and the lithiation state of electrodes with different porosities at the same discharge rate.

Similarly, Sun et al. [63] developed a FEM numerical model based on XCT images to study the evolution of the electrode structure and electrochemical performance during calendering. Fig. 17(a) illustrates the pre-processing of the XCT scans to extract the necessary data suitable for the construction of the FEM model, which includes: image filtering, normalisation, phase segmentation, particle labelling, structural reconstruction, etc. Fig. 17(b) illustrates the simulation results of the electrode microstructure cross-section compared to the experimental results.

The experimental results reveal that when the degree of calendering is below 10%, compression of the electrode occurs predominantly in the upper surface region. As the degree of calendering reaches 20%, this compression trend extends to the lower layers, indicating the initiation of self-arrangement phenomena among AM particles. Beyond a calender degree of 30%, particles become densely compacted, leading to plastic deformation. This effect is especially pronounced in small and medium-sized particles, which experience stress concentration because of their proximity to larger particles. The emergence of these processes significantly impairs the electrochemical performance of electrodes with high calendering degrees, particularly under high-rate discharge conditions. For example, as illustrated in Fig. 17(g), at a low discharge rate of 0.5C, electrodes with a degree of calendering 40% still exhibit a specific capacity of approximately 146 mAh/g. This suggests that at low rates, lithium-ion diffusion is primarily limited by diffusion within the solid AM particles themselves. However, as the current rate increases, the performance deteriorates markedly. At a high rate of 5C,



Fig. 16. Electrode discharge model on the basis of XCT image data and the results of its simulation. (a) The sample slice from an X-ray nano-CT scan with particles (white), CBD (grey) and pores (black). (b) Segmented and model geometry of the base sample where $H = 40 \mu m$. (c) An example of a computationally mirrored sample where the electrode thickness is 160 μm . The macro-porosity is 20% in B and C, and $B = W = 30 \mu m$, and $t_{cc} = t_{sep} = 15 \mu m$ in all cases. (d) State of lithiation and Li⁺ concentration profiles at 100% degree of discharge for 160 μm electrodes of varying macro-porosity. All electrodes were discharged at 1C. (e) Discharge response of an electrode with varying macro-porosity, a fixed thickness of 160 μm and a discharge rate of 1C. (f) Discharge response of an electrode with varying thickness, fixed macro-porosity of 25% and discharge rate of 1C. (g) Specific capacity as a function of macro-porosity for a variety of electrode thicknesses and discharge rates.

uncalendered and lightly calendered electrodes retain about 58% of their maximum capacity, while electrodes with a degree of calendering 40% retain only about 44 mAh/g of specific capacity. This indicates that a high degree of calendering leads to significant hindrance of ion transport, likely due to reduced porosity and increased tortuosity within the electrode microstructure.

In contrast, studies on volumetric capacity exhibit an opposite trend. Due to the higher volumetric usage in highly calendered electrodes, the volumetric capacity increases with increasing calendering degrees, especially at low discharge rates below 2C. As the porosity decreases to 25%, the volumetric capacity increases almost linearly, reflecting the denser packing of active materials, which enhances energy storage per unit volume. However, at high rates such as 5C, further increases in the calender exacerbate overpotential phenomena, leading to a decline in volumetric capacity. Specifically, at 5C and a porosity of 25%, a peak volumetric capacity of approximately 209 mAh/cm³ is achieved, beyond which the capacity decreases due to kinetic limitations. In general, this study provides a systematic analysis of how calendering degree influences battery performance metrics such as specific capacity, volumetric capacity, energy density, and power density. It elucidates the intricate interplay between electrode microstructure and electrochemical performance across different C-rates. The findings highlight that, while increased calendering can enhance the volumetric energy

density at low discharge rates by reducing porosity and increasing the density of active materials, it can be detrimental at high rates due to impeded ion transport and increased polarisation. These insights are crucial for optimising electrode design to balance energy and power requirements in practical battery applications.

In conclusion, FEM is a powerful tool for analysing the impact of the electrode calendering process on electrochemical performance. By coupling multiple physical fields, such as mechanical deformation, thermal effects, and electrochemical reactions, FEM enables an in-depth investigation of the intrinsic changes and operational principles occurring during battery charging and discharging processes. The meshing process inherent in FEM allows for detailed spatial discretisation of the electrode material, facilitating simulations that account for the complex, irregular geometries typical of electrode structures. This detailed modelling bridges the gap between theoretical predictions and practical observations, making FEM particularly well-suited for studying electrodes with non-uniform morphologies.

However, current FEM studies focusing on electrode microstructures are relatively limited and largely depend on XCT imaging results. This reliance constrains the development of data-driven, selfiterative optimisation models that can adapt and improve based on simulation outcomes. Therefore, integrating DEM models with FEM presents a highly promising research direction. Using DEM to generate



Fig. 17. The electrochemical numerical model reconstructed by XCT to analyse the relationship between its electrochemistry and structural evolution. (a) Flow chart of image processing, ML segmentation and 3D reconstruction for electrode XCT image dataset: raw image dataset (the bright regions are AM particles or CBD, the dark region is pore); image after using non-local means filter; image after using the operation of normalise grayscale; image after ML segmentation; labelled AM particles; 3D reconstruction of the electrode (only AM particles) and selected cubic ROI for further meshing and FE modelling (highlighted by orange box). (b) Comparison between simulation and experimental examination for microstructure evolution of the electrodes: the surface and cross-section views, respectively, from the mechanical simulation at different calendering degrees, and the surface and cross-section views, respectively, from the mechanical simulation at different C-rates. Comparison of specific discharge performance of electrode at different calendering degrees: (c) uncalendered, (d) 10% calendered, (e) 20% calendered, (f) 30% calendered, (g) 40% calendered, (h) comparison of the simulated rated specific capacity of the electrode against the C-rates, (i) comparison of the simulated rated volumetric capacity is calculated by considering the mass of AM particles in the electrode, while the volumetric capacity is determined by including the volume of the electrode (AM particles, macropore and CBD phases).

Source: This figure is quoted with permission from Sun et al. [63].

electrode-calendered structures and analyse their mechanical evolution and subsequently employing FEM for electrochemical analysis, a more comprehensive and predictive modelling framework can be established. This coupling leverages the strengths of both methods, the capability of DEM to simulate particle-level interactions and mechanical behaviour, and the proficiency of FEM in handling complex multiphysics problems, which is expected to deepen the understanding of how mechanical processes affect electrochemical performance. Such an integrated approach will hold significant potential for the advancement of battery technology through the optimised design of electrode materials and structures.

3.4. Machine learning and data-driven models

Currently, the rapid advancement of AI has introduced new horizons for the optimisation of computational models. Traditional modelling approaches are often impeded by complex modelling procedures, lengthy simulation times, and limitations in handling the multiscale and multiphysics nature of electrode materials. These challenges make it difficult to optimise electrode performance effectively. Additionally, traditional models may struggle with scalability and adaptability when dealing with the irregular and heterogeneous structures of electrodes, further complicating the optimisation process. Moreover, model validation is heavily dependent on a large amount of experimental data, which is both labour-intensive and time-consuming. This dependency not only slows down the development cycle, but also limits the ability to perform extensive parameter studies and iterative improvements.

The emergence of data-driven models based on machine learning offers a promising solution to these challenges. Machine learning algorithms can efficiently process large datasets to identify complex patterns and relationships that are difficult to capture with traditional modelling techniques. By incorporating techniques such as deep learning and neural networks, these models can handle high-dimensional data and non-linear interactions inherent in electrochemical systems. This approach can significantly reduce simulation times and simplify the modelling workflow by automating feature extraction and model training processes. On this basis, machine learning models can enhance predictive accuracy and generalisation by learning from both simulation and experimental data. They enable self-iterative optimisation by continuously updating the model as new data become available, thereby improving reliability and performance over time. This reduces the need for extensive experimental validation and accelerates the development of optimised electrode materials.

Deep neural networks (DNNs) represent one of the most prominent machine learning architectures today and are widely applied across diverse fields. A DNN is a type of artificial neural network characterised by the presence of multiple hidden layers between the input and output layers. In essence, an artificial neural network is composed of interconnected units that aggregate several inputs to produce a single output. The term 'deep' in DNN signifies both the increased number of layers and the increased complexity within each layer, enabling these networks to model intricate non-linear relationships and hierarchical features in the data. This depth facilitates the extraction of highlevel abstractions from raw input, which is particularly beneficial for complex pattern recognition tasks.

Among the various types of DNN, multilayer perceptron (MLP), convolutional neural networks (CNN), and recurrent neural networks (RNN) have emerged as the most popular in recent years. The MLP, which is the most fundamental form of deep neural networks, is composed of a sequence of fully connected layers. In an MLP, each layer transforms the weighted sum of outputs from the previous layer using a set of non-linear activation functions, thereby allowing the network to capture complex patterns within structured data. This architecture makes MLPs especially suitable for tasks that involve inputs with fixed dimensions and independent features. Over the past few years, MLPs have been employed by researchers to predict the microstructural evolution of slurries during drying and calendering processes. For example, Tan et al. [64] pointed out that the electrode manufacturing process involves multiple subprocesses, each characterised by highly nonlinear behaviours and interwoven with high-dimensional data. To address this limitation, they proposed a data-driven machine learning model that bypasses the need for an in-depth understanding of complex physical phenomena (as shown in Fig. 18(a)).

In their study, Tan et al. employed an experimental design method that combines both experimental and computational approaches. They generated a dataset based on experimental data to train a general machine learning model to study electrode manufacturing. To enhance the density and diversity of the dataset, they expanded it through computational means. This approach ensured that the model had sufficient data to learn from. Recognising the significant correlations between different Intermediate Product Properties, the model first used a graph clustering algorithm to group these properties. This method involved analysing the correlation coefficients between the properties and grouping highly correlated attributes together (as shown in Fig. 18(b) and (c)). This reduced redundancy and multicollinearity in the dataset, improving the predictive accuracy of the model, and streamlining the learning process. Subsequently, on the basis of the clustered data, appropriate machine learning algorithms which are well-suited for modelling complex nonlinear relationships and can handle high-dimensional data effectively were employed to establish the electrode production process model. Specifically, algorithms such as Multilayer Perceptron, Support Vector Regression, and Gaussian Process Regression were used to predict the relationships between process parameters and Intermediate Product Properties. The structure and training process of the model was optimised through hyperparameter tuning, which involved adjusting parameters like learning rates, regularisation coefficients, and network architectures to achieve the best performance. This comprehensive approach was able to reveal the impact of variations in process parameters on electrode characteristics and identify the Key Process Parameters (as shown in Fig. 18(d-i)). This study demonstrates the potential of machine learning models to effectively navigate the complexities of electrode manufacturing processes, providing high-dimensional data correlations that traditional physical models cannot capture.

Convolutional neural networks (CNNs) constitute another prominent category within DNNs. Unlike fully connected layers found in MLP, CNN models incorporate one or more convolutional layers that extract features from input data through convolution operations. In these layers, a set of nonlinear functions is applied to a weighted sum calculated over spatially adjacent subsets of the outputs of the preceding layer. This approach facilitates the repeated use of weights across different spatial locations, thereby reducing the number of trainable parameters and enhancing computational efficiency. The hierarchical application of convolution and pooling operations enables CNN to learn high-level abstract representations of the input, making them particularly well suited for image processing tasks. Recent studies have used models based on convolutional neural networks to predict characteristic changes in electrode microstructures, including variations in contact surface area, porosity, diffusivity, and bending coefficients. For example, Galvez-Aranda et al. [65] present a novel deep learning framework to bridge the computational gap between detailed simulations based on physics and the need for rapid process optimisation in lithium-ion battery manufacturing. In doing so, the study seeks to facilitate real-time adjustments in manufacturing parameters to achieve the desired electrode properties. They integrate a physics-informed deep learning approach with a time-series analysis framework. Specifically, a one-dimensional convolutional neural network is established, which is trained on a dataset generated through a sequential computational workflow that couples Coarse-Grained Particle Dynamics for simulating the slurry preparation and drying stages with DEM simulations for the calendering process. The microstructures, initially represented as threedimensional voxelised data, are reconstructed into a time-series dataset via a sliding window method. This transformation enables CNN to capture sequential dependencies and local spatial correlations inherent to the dynamic calendering process (as shown in Fig. 19(a)).

Furthermore, another key methodological innovation in this work is the rigorous and efficient hyperparameter tuning using the Optuna approach. This optimisation process not only ensured rapid convergence of the model, but also mitigated overfitting, as evidenced by the low mean squared error and high R² scores observed during validation (as shown in Fig. 19(b-g)). In particular, the model successfully captures intricate phenomena such as elastic recovery (spring-back effect) after compression, thereby reproducing both the transient and steady-state microstructural characteristics of the electrode. This study introduces the first known application of a time-dependent deep learning model to predict the three-dimensional evolution of battery electrode microstructures during calendering. By combining physics-based simulation data with an efficient CNN architecture, they achieve a dramatic reduction in computational cost, transitioning from simulation times on the order of tens of minutes per time step to approximately 15 s per step with the deep learning model. The model not only replicates key electrode



Fig. 18. Flow diagram of the process for modelling the electrode manufacturing process based on machine learning and the results for the calendering process. (a) Methodology of electrode production process modelling, including data generation, data clustering, machine learning modelling, and post-modelling analyses. (b–c) Correlation analysis of calendered electrode IPAs. (d–i) Responses of calendered electrode IPAs to over temperature and residence time. *Source:* This figure is quoted with permission from Tan et al. [64].

functional metrics (e.g. porosity, diffusivity, and tortuosity) with high fidelity but also provides a robust tool for potential integration into high-throughput screening and optimisation loops in battery manufacturing. Among the three primary types of deep neural networks, RNN has not yet been reported in the literature for modelling the electrode calendering process. Nevertheless, both RNNs and state-ofthe-art architectures such as the Transformer, which excels at capturing long-range dependencies and sequential patterns, may offer valuable avenues for future research, particularly in scenarios where temporal dynamics play a critical role.

In addition, compared to complex deep learning algorithms that learn intricate non-linear mappings through numerous neurones and multilayered structures, some researchers have explored alternative methodologies. For example, methods based on compressed sensing, such as Sure Independence Screening and Sparsifying Operator, have been employed to perform feature selection and sparse regression. SISSO is capable of extracting the most informative low-dimensional



Fig. 19. Workflow and simulation results of a CNN-based machine learning model for the evaluation of the calendering process and its structural evolution. (a) Schematic representation of the framework, starting from the 3D microstructure data reconstruction from DEM data, to train a CNN over a time series. After training, the model performance is assessed through evaluation and electrode functional metrics. (b–g) Evolution of the volume of the four different phases (AM, CBD, Pores and Void) in the number of voxel units versus time of the testing data, microstructure II at (b) 35% calendering degree and microstructure III at (d) 25% and (f) 45% calendering degree. R2 score comparing DEM target versus deep learning predicted over time of the testing data, microstructure II at (c) 35% calendering degree and microstructure III at (e) 25% and (g) 45% calendering degree. *Source:* This figure is quoted with permission from Galvez-Aranda et al. [65].

descriptors from a large pool of candidate descriptors, and it yields explicit mathematical formulations. These approaches not only simplify the model, but also enhance interpretability by providing clear, analytical expressions that can be directly related to the underlying physical processes. Duquesnoy et al. [66] proposed an empirical model based on experimental results to fit the relationship between the initial electrode parameters and the porosity after calendering. With the help of this curve, a large number of electrode structures were randomly generated using an electrode generation programme to form a training dataset. Based on this dataset, a machine learning algorithm was trained to identify and map the dependence between the manufacturing process and the electrode properties (as shown in Fig. 20(a)).

This machine learning model is mainly used to analyse the effect of the calendering pressure on the structure of the electrode. Fig. 20(b–e)

shows the evolution of the porosity after calendering electrodes with different initial porosities and AM contents. The simulation results show that electrodes with higher initial porosities and lower AM content can exhibit lower final porosities after calendering. This indicates that as the AM content increases, the degree of freedom in the electrode structure increases, and can more easily rearrange to form a denser solid-phase network when pressure is applied. Fig. 20(f–i) shows the variation in the tortuosity of the solid network predicted by the machine learning model. The tortuosity of the solid network increases with increasing AM mass fraction, indicating that a higher AM content leads to larger pore sizes, increasing the tortuosity of the solid phase paths. When pressure was applied, the volume fraction of the solid phase increased, as did the contact between particles, resulting in a larger



Fig. 20. Workflow and simulation results of a data-driven machine learning model for the evaluation of the calendering process and its structural evolution. (a) The hybrid methodology workflow. Experimental data is used to develop a mathematical equation, which is then embedded into a data-driven stochastic electrode generator that calculates the electrode mesostructure properties associated with different process conditions. The developed dataset is used to train and validate the ML algorithm, which allows to map and identify the interdependencies between electrode properties prior to the manufacturing, process parameters (reported in the Figure as electrode and process variables) and electrode properties after the manufacturing process is investigated. (b–e) ϵ_{cal} Obtained by polynomial fitting of the experimental dataset, in terms of the applied calender pressure and the AM mass fraction for different initial electrode porosities (ϵ_{inil}). As a reference, the electrode porosity after the calendering (ϵ_{cal}) is reported next to the cross symbols for AM mass fractions equal to 93% and 96% at pressures equal to 40, 80, 120 and 160 MPa. (1–m) ML-predicted active surface in terms of the pressure applied during the calendering and the AM mass fraction for different initial electrode porosities (ϵ_{inil}). As a reference, the electrode porosity after the calendering (ϵ_{cal}) is reported next to the cross symbols for AM mass fraction for different initial electrode porosities (ϵ_{inil}). As a reference, the electrode active surface in terms of the pressure applied during the calendering and the AM mass fraction for different initial electrode porosities (ϵ_{inil}). As a reference, the electrode porosity after the calendering (ϵ_{cal}) is reported next to the cross symbols for AM mass fraction for different initial electrode porosities (ϵ_{inil}). As a reference, the electrode porosity after the calendering (ϵ_{cal}) is reported next to the cross symbols for AM mass fractions equal to 93

Source: This figure is quoted with permission from Duquesnoy et al. [66].

and more connected solid network, which reduced the tortuosity. Fig. 20(j-m) shows the percentage of the current collector area covered by CBD as predicted by the machine learning model. The results show that at lower calender pressures, it increases with pressure up to a specific limit, after which the value almost stabilises. In addition, the effect of initial porosity on the percentage of current collector covered by CBD is small, indicating that, as the electrode structure becomes denser, an increase in initial porosity has no significant effect. Fig. 20(n-q) shows the evolution of the area of the AM phase in contact with the electrolyte. This is an essential mesoscale characteristic of the electrode, as it determines the effective rate of the lithium-ion reaction under kinetically limited conditions (such as high current density operation). Simulation results show that a higher initial porosity leads to a decrease in contact area, especially under high calendering conditions. Although it helps to reduce the final porosity after calendering and is beneficial for improving the volumetric energy density and power density of the battery, it is detrimental to the contact area between the AM phase and the electrolyte, which means limiting the lithium-ion reaction rate.

In a subsequent study by the same research group, Duquesnoy et al. [67] extended the multi-objective optimisation machine learning model to identify optimal parameters for electrode manufacturing. They expanded the scope of their empirical models by incorporating additional computational methods, such as Coarse-Grained Molecular Dynamics and the Lattice Boltzmann Method. The inclusion of Coarse-Grained Molecular Dynamics allowed them to simulate larger systems over longer timescales by simplifying the representation of molecular interactions. The Lattice Boltzmann Method, on the other hand, provided a powerful tool for simulating fluid dynamics and mass transport within the porous electrode structures, enabling a more comprehensive understanding of ionic diffusion and electrolyte flow. By integrating these methods, they generated a much larger and more diverse dataset. They then used Sobol sequence sampling to efficiently explore the highdimensional parameter space associated with the extended model (as shown in Fig. 21(b)).

Sobol sequences are a type of quasi-random, low-discrepancy sequence that ensures more uniform and thorough coverage of the parameter space compared to pure random sampling methods. This approach



Fig. 21. Model based on SISSO machine learning algorithms for evaluating the optimisation parameters of the electrode calendering process and its output results. (a) Schematic representation of the link between the different software for the handling of the 3D electrode mesostructures for the data acquisition generated by the physics-based modelling workflow. (b) Design of experiments for the synthetic dataset through the Sobol sequences after the sampling modifications within the input space. A 2D representation of the generated sequences based on the CD% values as a function of SC% values displays the quasi-randomness of the input space. Filling. (c–f) Radar chart plots comparing the optimised electrode properties with extreme cases from the synthetic dataset. Each plot is interested in obtaining the optimal case (red) instead of having a high-performance electrode roport. Replace electronic conductivity by conductivity, tortuosity factor by tortuosity, and active surface area by active surface in this Figure. (g–i) Partial dependence plots allowing to interpret the GP model's predictions. A 2D representation was straightforward to better visualise how manufacturing parameter values influence values for the search for the optimal manufacturing condition. The results are colour-coded where warmed values suggest less variability in the predictions when changing the hidden input parameter values, contrary to cooler values which suggest higher variability. The yellow stars point out the pairwise optimal manufacturing conditions predicted by the BO framework. The black dots represent each new possible candidate of manufacturing parameters to be explored as the solution in the optimisation loop, with most concentrated around the global minimum.

Source: This figure is quoted with permission from Duquesnoy et al. [67].

improves the convergence and robustness of the sampling process, providing a more reliable training set for machine learning algorithms. Based on the synthesised dataset, they conducted deterministic training of the machine learning algorithm, specifically focusing on supervised learning techniques that map the relationships between manufacturing parameters and electrode performance metrics. Deterministic training ensures that the learning process is reproducible and that the model converges to a consistent solution, which is essential for predictive accuracy in optimisation tasks. Finally, they used Bayesian optimisation methods to perform multi-objective optimisation of the electrode properties. Bayesian optimisation is particularly well-suited for optimising complex functions that are expensive to evaluate, as it builds a probabilistic model of the objective function and selects the most promising candidates based on expected improvement. This method allows for efficient balancing of multiple competing objectives. By iteratively updating the probabilistic model with new data, Bayesian optimisation converges towards the global optimum in the parameter space. Fig. 21(g–i) shows the optimal manufacturing conditions calculated by the optimisation algorithm. The result aims to maximise the effective electronic conductivity, density, and active surface area between the AM and the pores of the electrode, and to minimise the tortuosity factor. Fig. 21(c–f) compares the parameters of the optimised electrode with the edge of the dataset. However, it is important to note that, in contrast to the aforementioned deep neural network algorithms, the Sure Independence Screening and Sparsifying Operator method is primarily suited for small datasets. Additionally, SISSO faces limitations such as a reduced capacity for capturing highly non-linear or complex interactions in large-scale data, sensitivity to noise, and potential challenges in generalising beyond the specific conditions of the training set. Consequently, this method is currently considered an alternative

to deep learning models, particularly in scenarios where interpretability and the derivation of explicit mathematical relationships are prioritised over predictive performance in large datasets.

In conclusion, the integration of machine learning models has elevated the optimisation research of electrode parameters to a higher dimension. Using their exceptional data analysis and processing capabilities, machine learning algorithms can simultaneously balance the relationships among multiple parameters and performance outcomes. This provides significant assistance and support in optimising electrode performance. However, data-driven machine learning models require large datasets for effective training and calibration, which presents a significant challenge in the context of electrode calendering. Acquiring such extensive datasets through experiments alone is often impractical due to the time-consuming and costly nature of experimental procedures. Moreover, the complexity of electrode calendering processes, with their high dimensionality and nonlinear interdependencies among Key Process Parameters, further complicates data collection efforts. Consequently, existing data-driven machine learning models for electrode calendering predominantly rely on generating training datasets through a combination of experimental data and empirical models. This hybrid approach involves supplementing limited experimental observations with synthetic data derived from empirical or semi-empirical models that approximate the behaviour of the system. While this method helps in amassing sufficient data for model training, as previously discussed, empirical models have inherent limitations, often involve oversimplifications, and may not accurately capture the complex microstructural phenomena occurring during electrode calendering, which means it may introduce inherent biases and inaccuracies associated with the simplifications and assumptions of empirical models. Therefore, while machine learning offers powerful tools for optimisation, utilising models trained on such datasets may not fully realise their potential. A substantial journey remains ahead in the effective application of machine learning models to optimise the electrode calendering process. Future research should focus on incorporating more accurate and representative data sources, such as those obtained from DEM or FEM simulations, to enhance the reliability and applicability of machine learning in this domain.

4. Comparative analysis of modelling approaches for calendering

In the preceding sections, we have thoroughly reviewed the application examples of three modelling methods in electrode calendering processes: empirical, numerical, and machine learning models. This section will provide a detailed comparative analysis of the advantages and disadvantages of these three modelling approaches.

Empirical models were the first to be developed and have been relatively thoroughly studied. They use simple mathematical functions to describe the physical phenomena observed in experiments. By continuously introducing new correction factors, these models adjust their function curves to fit the experimental results as closely as possible. This modelling approach is straightforward and efficient, offering good interpretability because the relationships between variables are explicitly defined. However, the inherent philosophy of empirical modelling limits its broad applicability and accuracy. Since this approach requires simplifying and abstracting complex, tangible phenomena to extract critical information, it tends to overlook the intricate structural changes and various nonlinear behaviours occurring during the electrode calendering process. This method is more suitable for quickly assessing the overall behaviour of the system, and has extremely low computational resources and time costs. Therefore, empirical models are typically suitable for the initial stages of research, serving as tools for gaining a preliminary understanding of electrode calendering theory or for applications where high model precision is not critical.

Numerical models have been the focus of research interest in the past few years. They employ computer simulations and numerical solution methods to discretise continuous problems into independent elements or nodes, solving complex differential equations that govern the behaviour of the system. This modelling approach can effectively track complex structural changes, offering excellent applicability and interpretability. However, numerical models face significant challenges. High-precision models consume enormous computational resources, leading to very slow solution speeds, which can be prohibitive for long-term or large-scale simulations. Furthermore, substantial differences exist between various numerical modelling techniques. For example, mechanical studies of the calendering process often rely on the DEM, which models individual particles and their interactions, while electrochemical studies typically depend on the FEM, which solves continuum equations for fields like electric potential and ion concentration. Coupling these different models is relatively difficult due to their fundamentally different formulations and scales, and this remains a key focus of current research. Developing efficient and accurate multiscale and multiphysics coupling strategies is essential to advance the predictive capabilities of numerical models in this area. For example, Sandooghdar et al. [68] are attempting to combine the DEM with computational fluid dynamics to simultaneously capture the mechanical deformation of electrode structures and the fluid flow through the resulting porous networks. In their study, DEM was employed to simulate the calendering process, thereby generating detailed microstructural configurations at varying degrees of compression. Subsequently, computational fluid dynamics was applied at the pore scale to address electrolyte transport phenomena, enabling the computation of key parameters such as porosity, tortuosity, and permeability. This integrated approach established a direct connection between structural evolution during calendering and the subsequent transport properties for the first time. Moreover, the predicted pore-scale characteristics can be incorporated into three-dimensional electrochemical-thermal models, facilitating an in-depth exploration of the relationship between microstructural changes induced by calendering and overall battery performance metrics, such as discharge behaviour and thermal management. Finally, it is essential to note that the accuracy of numerical models is highly dependent on thorough calibration and validation using experimental data or in situ measurements. However, obtaining high-quality, comprehensive experimental data remains a significant challenge in many practical engineering applications. This limitation represents one of the primary obstacles in the transition of numerical models from laboratory research to industrial implementation.

Data-driven machine learning models have emerged in recent years, along with the rapid advancement of AI. Through continuous selfiterative optimisation, they can effectively handle potential correlations among multi-dimensional parameters, uncovering complex relationships that may be difficult to model explicitly. This modelling approach does not aim to reproduce the structural changes during the calendering process but focuses solely on learning the mapping logic between inputs and outputs from data, making it highly efficient and widely applicable. However, the black-box nature of machine learning models results in a significant loss of interpretability. Since the internal workings of these models are often opaque, it is challenging to extract meaningful insights into the underlying physical processes or to explain why certain predictions are made. This lack of transparency can hinder the acceptance of machine learning models in scientific research and engineering applications where understanding causality is important. It should be noted that machine learning models require large and high-quality datasets for training, which can be difficult to obtain in experimental contexts. Moreover, pure data-driven methods often ignore physical laws. In the future, it is urgent to incorporate physical information into machine learning (e.g., physical information neural networks) to improve the physical consistency of models and prediction accuracy.

The progression from empirical models to numerical models and eventually to machine learning models reflects the growing demand for increased accuracy and predictive capabilities in electrode calendering modelling. In light of this, the integration of numerical models with machine learning models holds promise as a new direction for future

Comparison of different modelling approaches based on various evaluation dimensi	ons
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Evaluation dimension	Empirical model	DEM model	FEM model	ML model
Model accuracy	Low	High	High	Moderate-High
Computational efficiency	High	Low	Moderate	High
Interpretability	High	High	High	Low
Data requirement	Low	Moderate	Moderate	High
Complex-System applicability	Low	Moderate	High	High
Microstructure sensitivity	Low	High	High	Moderate-High
Multiscale capability	Low	Low	Moderate	High
Scalability and Flexibility	Low	Moderate	Moderate	High
Ease of Use	High	Low	Low	Moderate
Method maturity	High	Moderate	Low	Low

research. This combined approach leverages the strengths of both methods: the detailed physical insights and interpretability of numerical models, as well as the efficiency and predictive power of machine learning algorithms. By using data generated from numerical simulations to train machine learning models, researchers can develop surrogate models that approximate complex system behaviours at a fraction of the computational cost. These surrogate models can then be used for rapid optimisation, sensitivity analysis, and real-time predictions. On this basis, developing novel architectures that couple multiple algorithms to offset the inherent limitations of individual machine learning methods can offer improved solutions to the challenges mentioned above. For example, Vijay et al. [69] are developing an ensemble deep learning model, Slurry-NN, which employs a data-driven approach using simulation data from coarse-grained molecular dynamics and DEM to predict the final microstructure of electrodes. The model incorporates two CNNs and two MLPs, which are tailored to process spatio-temporal features (such as particle coordinates and image descriptors) and static categorical features (such as particle radius and type), respectively. This modular architecture allows each branch to specialise in capturing distinct aspects of the simulation data, which is expected to improve both the interpretability and predictive accuracy of the overall model. In addition, machine learning techniques can help overcome the coupling difficulties between different numerical models. For instance, machine learning algorithms can learn the relationships between parameters in DEM and FEM simulations, facilitating a more seamless integration of mechanical and electrochemical analyses. This hybrid modelling strategy will not only improve computational efficiency but also enhance model accuracy by capturing the multifaceted interactions within the electrode during calendering (see Table 1).

5. Conclusion

In conclusion, this review has highlighted various modelling approaches, including empirical, numerical, and machine learning-based models, each of which has a unique role to play in the calendering process.

- 1. Empirical models provide simplified yet practical relationships between calendering parameters and electrode properties, but face limitations in addressing the complex nature of real materials.
- 2. Numerical approaches, such as DEM and FEM, provide deeper insight into microstructural evolution and mechanicalelectrochemical interactions, but at the cost of high computational demands.
- 3. The integration of DEM and FEM emerges as a promising research direction, capable of linking microstructural mechanics with macroscopic electrochemical performance. This synergistic approach could provide a more comprehensive and predictive modelling framework.
- 4. Machine learning models promise rapid, data-driven optimisation by handling complex, non-linear relationships and highdimensional data. The application of machine learning represents an exciting frontier in this field, promising increased efficiency and adaptability in the face of growing manufacturing challenges.

However, it should be noted that although this review provides a systematic and comprehensive analysis of the prevailing modelling methodologies used for electrode calendering, several limitations remain. Firstly, the methodological approach relies heavily on existing published literature, potentially overlooking the complexity or unsolved coupling issues inherent to each modelling strategy. For instance, the unresolved multiscale coupling challenges between DEM and FEM may not be thoroughly captured. Secondly, the comparative analysis presented in this review involves a degree of subjectivity. The classification of empirical models solely as preliminary tools, for example, may underestimate their potential value in rapid, industryorientated decision-making contexts. Thirdly, this review primarily concentrates on traditional modelling paradigms (empirical, numerical, machine learning), thereby neglecting emerging technologies with significant potential. Quantum computing, digital twin technologies for real-time process optimisation, and generative artificial intelligence for microstructure generation exemplify promising alternatives that were not discussed herein. Lastly, there exists a notable gap between theoretical academic research emphasised in this review and practical industrial applications. Addressing industrial practicality, feasibility, and direct application scenarios, along with clearly demonstrating how theoretical insights translate into tangible improvements in manufacturing processes, would greatly enhance the practical relevance of this work. Future reviews could further bridge this divide by incorporating industry-driven case studies and investigating real-world applicability alongside theoretical advancements.

Looking ahead, future research should focus on the continued integration of these modelling strategies, especially in bridging the gap between computational accuracy and practical applicability. The development of hybrid models that draw on the strengths of empirical, numerical, and data-driven approaches to provide a reliable predictive model for LIBs electrode calendering will be the focus of future research. In summary, the comparative analysis of the modelling methods outlined here highlights the pathways to achieve significant improvements in battery performance while reducing environmental impacts, aligning closely with global priorities such as climate change mitigation and carbon neutrality. Ultimately, by facilitating more intelligent and data-driven optimisation in electrode calendering processes, this review aims to promote more sustainable environmental, social, economic, and governance practices within the energy storage sector.

CRediT authorship contribution statement

Jiashen Chen: Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. Maryam Asachi: Writing – review & editing, Supervision. Ali Hassanpour: Writing – review & editing, Supervision. Meisam Babaie: Writing – review & editing, Supervision, Project administration, Conceptualization. Masoud Jabbari: Writing – review & editing, Supervision, Project administration, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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