



Exploring the role of carbon binder domain morphology in enhancing the electrochemical performance of Li-ion battery

Wei Sun^a , Chun Huang^{a,b,c,*}

^a Department of Materials, Imperial College London, London, SW7 2AZ, UK

^b The Faraday Institution, Quad One, Becquerel Ave, Harwell Campus, Didcot, OX11 0RA, UK

^c Research Complex at Harwell, Appleton Laboratory, Didcot, OX11 0FA, UK

HIGHLIGHTS

- Couple XCT imaging and numerically generated carbon binder domain (CBD) morphology.
- Explicit and implicit CBD electrochemical simulations.
- Analysing impact of CBD morphology on interface area and discharge performance.
- Identifying ionic transport limit of CBD at high-rate conditions.
- Providing guidance on CBD morphology optimisation for electrode design.

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ABSTRACT

The electrochemical performance of Li-ion batteries (LIBs) is significantly influenced by electrode microstructure. The carbon and binder domain (CBD) plays an important role as it supports electronic conduction and maintains mechanical integrity. Despite its importance, the impact of CBD morphology on electrochemical performance remains poorly understood. One of the reasons is that CBD requires higher resolution to resolve the nanostructures, and techniques such as most X-ray computed tomography (XCT) machine cannot resolve CBD. This study establishes a workflow that incorporates active material (AM) particles obtained from XCT with numerically generated CBD morphology to build a 3D image-based and physics-resolved model to predict its electrochemical performance. Our model considers both explicit and implicit configurations of dense and nanoporous CBD during the electrochemical modelling process. Results indicate that the interface between AM and electrolyte significantly limits discharge capacity under explicit dense CBD configurations. In contrast, film-coating-like nanoporous CBD could enhance LIB longevity due to locally homogeneous lithiation in AM particles. Our findings also suggest that Li-ion transport is a major limitation in achieving higher (dis)charge rate capacities (5C) under implicit CBD considerations. This study highlights the crucial role of CBD morphology in optimising electrode design for improved battery performance.

1. Introduction

Li-ion batteries (LIBs) have been widely adopted in electronic devices, electrical vehicles (EVs) and grid-scale power stations due to their high energy and power densities [1–3]. However, challenges remain in achieving high-rate capability during (dis)charging for cost-effective electrodes [4,5]. Among the various approaches to resolving the above challenges, tailoring electrode microstructure has become promising in achieving fast (dis)charging without significantly inducing degradation

[6,7].

Typically, LIB electrodes are porous microstructures consisting of electrochemically active material (AM) particles, carbon black additives, polymeric binder (e.g. polyvinylidene fluoride, PVDF), and pores deposited on the electronic conductive current collector [8]. The combination of electronic conductive carbon lack and polymeric binder is known as the carbon binder domain (CBD) [9]. The energy and power densities of LIBs are notably influenced by the microstructural configuration of the electrode and mass loading of its components [10–13].

* Corresponding author. Department of Materials, Imperial College London, London, SW7 2AZ, UK.

E-mail address: a.huang@imperial.ac.uk (C. Huang).

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Although experimental results have shown that reducing the proportion of CBD while increasing the proportion of AM particles can enhance energy density [14,15], the specific role of CBD in modelling remains underexplored in existing literature. This gap highlights the need for detailed investigations into the influence of CBD on the electrochemical performance of LIBs, which is crucial for advancing electrode design and overall battery efficiency.

The morphology observation and microstructure characterisation of CBD have been primarily conducted using scanning electron microscopy (SEM) from a 2D imaging and focused ion beam-SEM (FIB-SEM) from a 3D perspective due to their superior resolution capabilities down to nanometers [8,16–18]. Notably, a 2D SEM image led to a reported nanoporosity of 0.27 for CBD [16], whereas a 3D microstructure of CBD reconstructed using seven images obtained from FIB-SEM suggested a higher nanoporosity of 0.47 [17]. Recently, a novel approach involving stand-alone scanning of CBD and electrode films was introduced, using nanoscale X-ray computed tomography (XCT) at a resolution of 63–126 nm [11,19]. This method integrated the CBD phase within the AM particles phase based on greyscale histogram analysis. Nevertheless, this technique has raised concerns regarding the accuracy of resulting porosity measurement and subsequent electrochemical simulations. For example, porosity measurements for the electrode with large AM particles, calendered by 22 %, was determined to be 0.188, and for the electrode with small AM particles, calendered by 24 %, the porosity was measured to be 0.195. Those porosity values significantly diverge from the analytical predictions of 0.34 and 0.36, respectively, as estimated using the equation in Refs. [10,12]. This discrepancy may be attributed to the measurements reflecting the macropore volumes, while the impact of nanoporosity within the CBD phase remains unclear. Recent advancements have seen the adoption of micro-XCT, which offers resolutions of 370 nm–700 nm [10,20–23], facilitating an improved characterisation of electrodes over a larger field of view characterising electrodes [24,25], such as in the study of electrodes measuring 700 μm in thickness [21,22]. Subsequently, the CBD phase within the scanned electrode can be extracted using the advanced machine learning (ML) segmentation method [12,23,26–28]. Interestingly, such a method has identified a macroporosity of 0.11 in the uncalendered electrode, although the nanoporosity of the CBD phase remains to be unclarified [27]. Therefore, it is imperative to investigate further the effect of the CBD phase on the electrochemical performance during the 3D microstructure-resolved modelling of the electrodes.

Besides the experimental method to characterise the CBD phase, CBD can be artificially generated within the complementary volume of the AM particles imaged by XCT [9] or created by the discrete element method (DEM) [27]. For example, an interfacial energy-based approach was developed to control the effect of the long-range and short-range transport properties within the battery electrode [29]. A level-set approach was proposed to generate a bridge-like CBD between the AM particles [9,30,31]. Subsequently, the transport properties of those generated electrodes were further investigated under different calendaring degrees [31]. A recent work adopted a random field method to generate the CBD with tunable pore size [27]. They presented that the pore size distribution of CBD has substantial effects on the simulated battery performance. Furthermore, the CBD was generated during the modelling of the electrode manufacturing process, such as slurry, drying and calendaring, using coarse-grained molecular dynamics (CGMD), DEM and multi-phase smoothed particles (MSMP) [32,33]. However, uncertainties persist in the actual distribution of CBD due to incomplete knowledge of the binder migration mechanisms during the drying process. For example, the generated CBD network is non-connective in the electrode for the 5 % mass loading of CBD within the electrode, resulting in a fragmented electronic transport path within the electrode considering the AM particles [33].

The advancement of computational capabilities has significantly enhanced the popularity of 3D microstructure-resolved modelling of electrodes, providing researchers with critical insights to optimise the

microstructure of LIBs [8,11,12,19,26,34–36]. In these models, the CBD is typically represented either as an explicit phase [19,34,35], where it is distinctly resolved within the microstructure with its specific properties, or as an implicit phase [8,12,26,27,36], where it is integrated with the electrolyte. Despite the advantages of explicit modelling, significant variations have been observed in the predicted specific capacities, leading to potential uncertainties in simulation outcomes. For instance, specific capacities in such models have ranged from 40 to 88 mAh g^{-1} for electrodes with stochastically generated CBD, particularly when discharged at 3C [35]. This variability underscores the need for a refined model that better explains the role of CBD in explicit configurations and enhances the consistency and reliability of the simulation results.

Here, a 3D image-based and physics-resolved model is developed to explore the role of CBD in the electrochemical performance of LIBs, in which the CBD morphology within the electrode is generated using advanced ML segmentation technique and numerical methods. The resulting microstructures form the basis for further explicit and implicit CBD consideration during the modelling of electrochemical performance. The model allows to understand the electrochemical process of electrodes and provides new insights into the underlying microstructural behaviour when changing the CBD morphology. We present a tool that enables the design of electrode microstructures with higher capacity and improved transport properties.

2. Material and methodology

2.1. Material and data source

An uncalendered polycrystal $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (NMC532) cathode with a mass loading of 22.6 mg cm^{-2} and a thickness of 104.8 μm was used in this study. The mass ratio of the constituents was 90:5:5 for AM particles (NMC532), C45 carbon black and PVDF binder, respectively. The electrode microstructure was characterised by XCT using Zeise-radia 520 with an isotropic voxel of 397 nm and referred to as “3-UNCAL” within the open source library provided by the National Renewable Energy Laboratory (NREL) [20]. An uncalendered electrode was selected to avoid structural modifications induced due to the calendaring process. Among the available uncalendered samples, we chose 3-UNCAL for consistency with our previously published work [12]. The advancement here is that we further investigate the effects of the CBD phase which is challenging to be distinguished by using traditional imaging techniques.

2.2. Image processing and segmentation

The raw XCT image dataset was reoriented, transformed and filtered with Avizo software. Subsequently, this pre-processed image data with the three-phase electrode (AM particles, CBD and pore) was segmented using the ML-based, open-source software Ilastik [37]. The detailed image processing and segmentation procedure and the corresponding results can be referred to our previous publication [12,38]. The segmentation process yields a ternary electrode image with the following volume fractions: AM particles at 43.8 %, CBD at 13.0 %, and macropores at 43.2 %.

2.3. Volume fraction of CBD in the electrode

To validate the segmented results, the solid volume fraction of each electrode phase was calculated using the known mass fractions of the constituents from the electrode manufacturing details along with their material mass densities. The solid volume fraction of the CBD phase ($V_{\text{CBD-solid}}$) was determined using the following formula

$$V_{\text{CBD-solid}} = V_{\text{AM}} \left(\left(\frac{w_C}{1 - w_C - w_B} \right) \left(\frac{\rho_{\text{AM}}}{\rho_C} \right) + \left(\frac{w_B}{1 - w_C - w_B} \right) \left(\frac{\rho_{\text{AM}}}{\rho_B} \right) \right) \quad (1)$$

where w_C and w_B represent the mass fractions of carbon black and binder, respectively, and ρ_{AM} , ρ_C and ρ_B are the material mass densities of AM particles, carbon black and binder, respectively. The volume fraction of AM particles (V_{AM}) was extracted from ML segmentation results, calculated as 0.438. Upon inputting the mass density of each phase ($\rho_{AM} = 4.74 \text{ g cm}^{-3}$ [39], $\rho_C = 1.78 \text{ g cm}^{-3}$ and $\rho_B = 2 \text{ g cm}^{-3}$ [10]), the resulting V_{CBD_solid} was 0.122. This value is consistent with the result obtained from ML segmentation.

Considering the above-calculated solid volume fraction of CBD and reports of its intrinsic nanopores ranging from 5 to 150 nm [17], it is crucial to consider the potential nanoporous nature of CBD, especially in light of the current XCT scan resolution of 397 nm. This suggests that CBD in the electrode may exhibit in either dense or nanoporous configurations. Accordingly, the volume fraction of explicitly nanoporous CBD is determined to be 0.23, reflecting a nanoporosity of 0.47 as characterised by the FIB-SEM technique [17].

2.4. Numerical generation of CBD

This section describes the numerical methodologies used to generate CBD configurations within the electrode matrix, utilising the MATBOX toolbox in MATLAB [40]. Two distinct approaches were employed to model the spatial distribution and interaction of CBD within the electrode, which are briefly introduced in the following subsections: the deterministic bridge approach and the interface energy-based approach. Both methods ensure that the generated microstructure aligns with experimental observations and theoretical requirements for explicit dense and nanoporous CBD configurations.

2.4.1. Deterministic bridge approach

The deterministic bridge approach was employed using MATBOX to generate the “bridge-like” CBD configurations, assuming CBD preferentially locates between particles that are not far from each other [13, 40]. Using the ML segmented AM particles mentioned in section 2.2, the remaining phases of the electrode were classified into either CBD or void/electrolyte. Meanwhile, it is necessary to ensure that the generated CBD phase matches the prescribed solid volume for either the explicit dense or the nanoporous CBD.

2.4.2. Interface energy-based approach

The formation of electrodes is typically governed by an evaporation-driven process in which a colloidal slurry loses solvent and forms a porous structure. This structure is significantly influenced by the interfacial and cohesive energies of the AM particles and CBD, which dictate the final electrode microstructure. The interface energy-based approach, also incorporated in MATBOX, was adopted to generate the CBD additive based on energy toward deposition on the pre-existing network of ML segmented AM particles or the pre-deposited new additive CBD phase [29,40].

A user-defined morphology parameter, denoted as ω (dimensionless, ranging from 0 to 1), directs the energy deposition for the candidate CBD voxel (e_k), determining its preferential deposition onto either an existing AM particle ($\omega = 0$) or the pre-deposited CBD phase ($\omega = 1$):

$$e_k = \frac{\omega}{6} N_{CBD} + \frac{(1-\omega)}{6} N_{AM} \quad (2)$$

where N_{CBD} and N_{AM} denote the number of voxels adjacent to the candidate voxel that belong to either CBD or AM phases, respectively. The local deposition energy is normalised by dividing by six due to the cuboid voxel representation of the microstructure. The deposition energy is maximised when ω takes values of 0 or 1, leading to either a thin layer of CBD coating on the AM particles ($\omega = 0$) or agglomerated CBD phase ($\omega = 1$). Intermediate values of ω enable a controlled transition between these two deposition behaviours, allowing for microstructural tuning.

This parameter plays a crucial role in optimising the electrode microstructure by directly influencing the porosity and connectivity of the deposited CBD phase which in turn will influence electrochemical performance and transport properties. Once the cumulative energy deposition of candidate CBD voxels is determined, a finite subset of these voxels is selected for deposition based on their energy values. This iterative process continues until the desired volume fraction of dense or nanoporous CBD is achieved.

2.5. Electrochemical simulation setup in COMSOL multiphysics

As illustrated in Fig. 1(a–c), Simpleware, a commercial software specialising in model meshing, was used to mesh those numerically generated electrode microstructures as described in section 2.4. This resulted in approximately 0.9–4.8 million linear tetrahedral elements. The meshed volumes were then imported into the software COMSOL Multiphysics for electrochemical simulation, utilising the embedded battery package. The governing equations, boundary conditions and input parameters used in the present study are listed in Tables S1–S3 in Supplementary Information (SI). Briefly, the generalised Nernst-Plant equation and Fick’s law were employed to describe the mass conservation for Li-ion transport in the electrolyte and Li transport in AM particles, respectively. Ohm’s law and the electroneutrality equation were adopted to describe the charge conservation in the AM particles and electrolyte, respectively. The Butler-Volmer equation was used for interfacial charge transfer reactions occurring at the interface of Li metal, as well as the interface between AM particles and nanoporous CBD and electrolyte. The local exchange current density was linked to the concentration of reactants in the electrolyte and AM particles.

CBD is considered in explicit or implicit scenarios to analyse the role of CBD in the electrochemical performance comprehensively. In the explicit scenario, the conducting network of CBD is explicitly distributed. In the implicit scenario, electrolyte-CBD integration is captured without explicitly resolving CBD morphology (see Fig. 1(a–c)). It is important to note that the CBD could be either dense or nanoporous. Therefore, the electrochemical reactions occur solely at the interface between AM particle and electrolyte where the part of the AM particle surface is not covered by CBD in the dense CBD scenario. In contrast, in the nanoporous CBD scenario, the electrochemical reactions occur at the interface between AM and electrolyte, and some Li-ions diffuse through the nanoporous CBD phase and react at the interface between the nanoporous CBD and electrolyte, albeit the latter with a restricted electrochemical reaction rate. In the implicit CBD scenario, the electrochemical reaction takes place at the interface between the AM particle and lumped CBD and electrolyte. The modelling definitions for the lithiation process are illustrated in Fig. 1(e–f).

The above-built models were discharged at 1 and 5 C rates from the fully charged state, where all AM particles had an initial SOL state of 0.38. The parallel direct sparse solver (PARDISO) was employed to solve the discretised transport and electrode kinetics equations. A segregated approach was taken involving solving the coupled field in a sequential, staggered way. Time stepping was managed using 2nd order backward Euler differentiation. A stop condition was set for the cell to reach 3 V, marking the end of the cell discharge.

3. Results and discussion

3.1. Numerically generated CBD morphology comparison

Fig. 2 showcases the two approaches for generating CBD phase microstructures: the deterministic bridge approach and the interface energy-based approach. In Fig. 2(a), the dense CBD phase is illustrated. The first image demonstrates CBD morphology utilising the bridge approach, exhibiting high connectivity among neighbouring AM particles. The subsequent five images showcase the interface energy-based approach at different morphology factors ($\omega = 0.999, 0.9, 0.5, 0.1$,

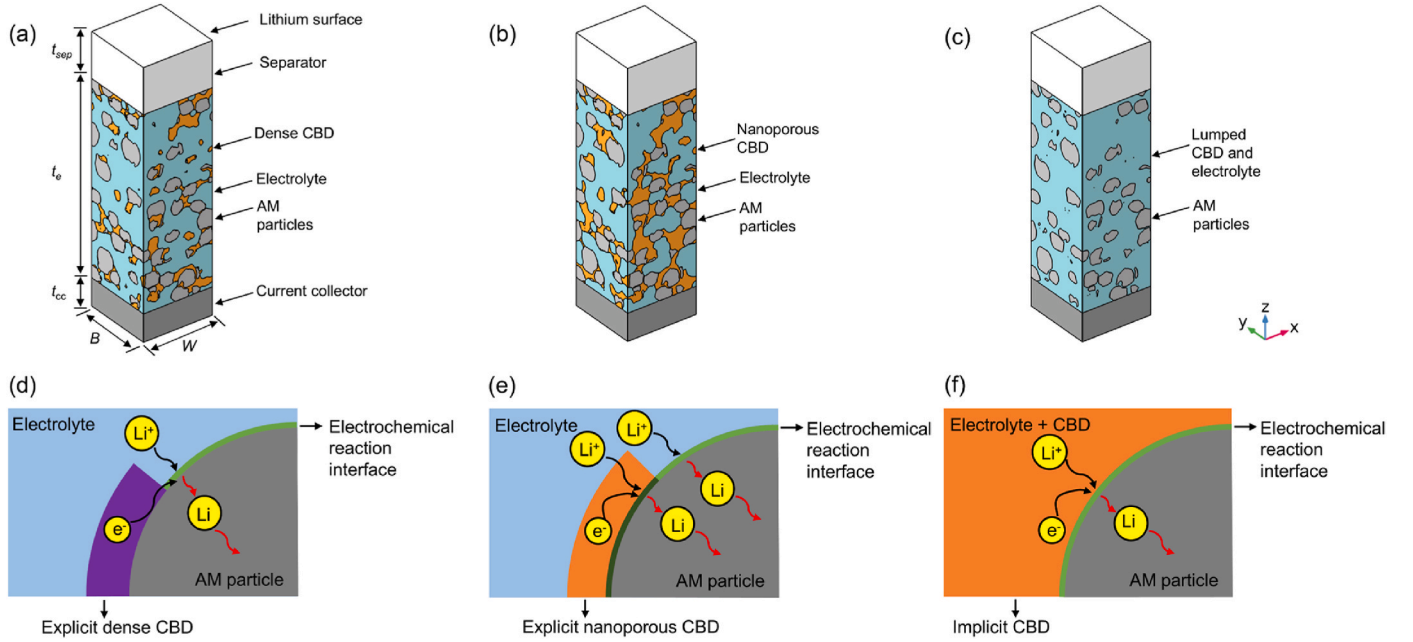


Fig. 1. Illustrations of electrochemical models and lithiation processes used in COMSOL Multiphysics: electrode configured with an explicit representation of CBD featuring numerically generated (a) dense CBD morphology; (b) nanoporous CBD morphology, both employing a deterministic bridge approach. (c) Electrode configured with an implicit representation of CBD with lumped CBD and electrolyte. (d) Explicit dense CBD scenario showing lithiation only at the interface between AM particles and the electrolyte. (e) Explicit nanoporous CBD scenario illustrating lithiation occurring at the interfaces between AM particles and electrolyte, and some Li-ions diffuse through the nanoporous CBD phase and react at the interphase between nanoporous CBD and electrolyte. (f) Implicit CBD scenario showing lithiation across the interface of lumped electrolyte and CBD with AM particles.

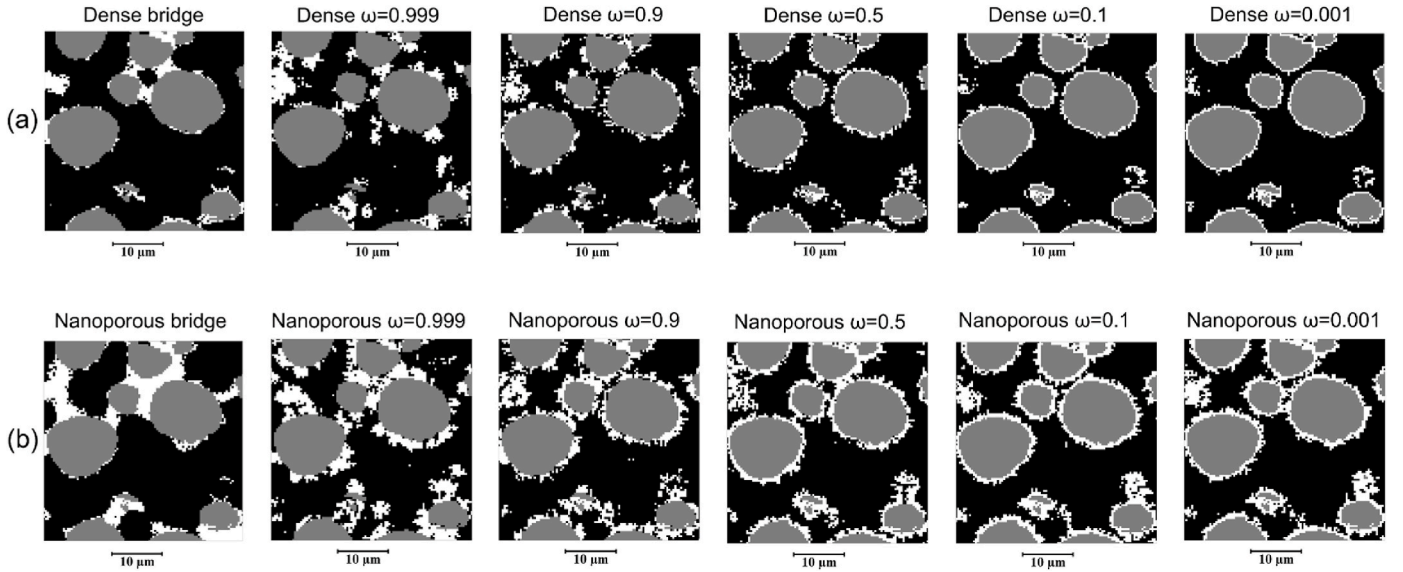


Fig. 2. CBD morphology comparison on a 2D slice through the 3D domain. AM particles are shown in grey, CBD in white and void/electrolyte in black across two configurations: (a) dense CBD scenarios and (b) nanoporous CBD scenarios. Each configuration is analysed using a deterministic bridge approach and an interfacial energy-based approach with ω ranging from 0.999 to 0.001.

0.001). As ω decreases, the CBD structure transitions from agglomerate-like to more film-like configurations.

Similarly, Fig. 2 (b) features the nanoporous CBD phase. The first image again utilises the bridge approach to depict the CBD morphology, followed by five images using the interface energy-based approach with varying ω values. These illustrations also depict a morphological transition, emphasising the consideration of nanoporosity, resulting in an almost doubled volume fraction in the corresponding morphologies.

In practice, the explicit dense and nanoporous CBD phases could

correspond to different electrode calendaring conditions, e.g. an uncalendered electrode (0 MPa) exhibits a nanoporous CBD microstructure, whereas a highly calendered electrode (1000 MPa) shows a densified CBD structure [41]. The dense CBD phase represents a highly percolated and agglomerated carbon-binder network, facilitating electronic conduction, while the porous phase signifies a more open and dispersed structure, allowing better ionic accessibility.

3.2. Geometric and physics comparison of the electrode with numerically generated CBD

The morphological changes in dense and nanoporous CBD configurations, influenced by the different generation approaches and varying ω values, inevitably affect structural properties while the morphology of AM particles remains unchanged. To quantitatively analyse those effects, the discussion includes the interfacial area percentage for the interfaces between AM particles and the electrolyte and between AM and either dense or nanoporous CBD. Additionally, the analysis covers both ionic and electronic tortuosity within the electrode.

Fig. 3 compares the percentage distribution of the AM/CBD interface and the AM/electrolyte interface for both dense and nanoporous CBD phases using two different approaches: the deterministic bridge approach and the interface energy-based approach. In Fig. 3 (a), for the dense CBD configurations, the distribution comparisons include the ML-segmented CBD (first column) and the deterministic bridge method (second column) versus the interface energy-based approach across various morphology factors ($\omega = 0.999, 0.9, 0.5, 0.1$). The ML CBD configuration exhibits a 39 % AM/CBD interface and 61 % AM/electrolyte interface. In contrast, the bridge approach reveals a 54 % AM/CBD interface and 46 % AM/electrolyte interface. Notably, as ω decreases from 0.999 to 0.1, a significant shift in the distribution occurs, demonstrating a 35 % AM/CBD and 65 % AM/electrolyte interface at $\omega = 0.999$, shifting to 53 % AM/CBD and 47 % AM/electrolyte at $\omega = 0.9$, with further increases to 78 % at $\omega = 0.5$ and 98 % at $\omega = 0.1$ for the AM/CBD interface, while the AM/electrolyte interface correspondingly decreases to 22 % and 2 %, respectively.

For the nanoporous CBD configuration illustrated in Fig. 3 (b), the bridge approach indicates a 71 % AM/CBD interface and 29 % AM/electrolyte interface. The energy-based approach at $\omega = 0.999$ shows the distribution of 64 % AM/CBD interface and 36 % AM/electrolyte interface. As ω decreases, the trend continues with an increase in the AM/CBD interface percentage: 85 % at $\omega = 0.9$, 98 % at $\omega = 0.5$, and reaching 100 % at ω values of 0.1 and 0.001. Those results highlight that the interface distribution within the electrode varies significantly with different morphology factors and approaches, clearly demonstrating the profound influence of CBD morphology on the structural properties and potentially on the performance of the electrode, which will be discussed in section 3.3.

Fig. 4(a–b) evaluates the ionic and electronic tortuosities in electrodes composed of both dense and nanoporous CBD, assessed along the

Z-direction (through the electrode thickness) using the Taufactor in MATLAB [42]. Ionic tortuosity is determined using the pore network, and electronic tortuosity is assessed through the CBD network within the electrode. This study differentiates between explicitly dense and nanoporous scenarios, where the CBD phase serves exclusively as the pathway for electronic transport. As shown in Fig. 4 (a) for dense CBD configurations, ionic tortuosity values range from 1.4 to 1.2, showing a declining trend as the parameter ω decreases. Similarly, in nanoporous CBD, a decrease in ω correlates with a decrease in ionic tortuosity, attributed to the presence of more macropores. Fig. 4 (b) reveals that electronic tortuosity in dense CBD is significantly higher than in its nanoporous counterpart due to a less interconnected CBD network structure.

The ionic and electronic tortuosities were evaluated along the Z-direction because this is the primary direction of the Li-ion transport within the electrode during charging and discharging, between the separator and the current collector. In practical battery operation, transport is dominated along this axis due to the applied potential gradient [43]. While the in-plane (X- and Y-directions) transport could be evaluated, it is less relevant to the overall cell performance. However, we acknowledge the importance of assessing anisotropic effects and will consider this in future studies.

To adapt explicit CBD scenarios for modelling 3D microstructure-resolved electrodes with implicit CBD, the effective transport coefficient ($\delta = \epsilon/\tau^2$) was utilised [44], where ϵ represents the volume fraction of CBD or the pore phase within the combined CBD and electrolyte phase. Fig. 4(c–d) depict the effective ionic and electronic transport coefficients. As illustrated in Fig. 4 (c). The effective ionic conductivity in the dense CBD configuration generally exceeds that in the nanoporous setup due to lower ionic tortuosity and a higher volume fraction of the pore phase. The highest effective ionic transport coefficient was observed in the ML-segmented CBD configuration.

In the nanoporous CBD scenario, the bridge CBD configuration exhibited the lowest effective ionic transport coefficient, correlating with the highest tortuosity values noted in Fig. 4 (a). As ω decreased from 0.999 to 0.001, there was an increase in the effective ionic transport coefficient, attributed to the film-like nature of CBD at $\omega = 0.001$, which enhances the pore phase availability for ionic transport. Fig. 4 (d) provides a comparative analysis of effective electronic transport coefficients, demonstrating that nanoporous CBD exhibits a significantly higher transport efficiency than dense CBD due to the reduced CBD volume fraction and increased electronic tortuosity within the

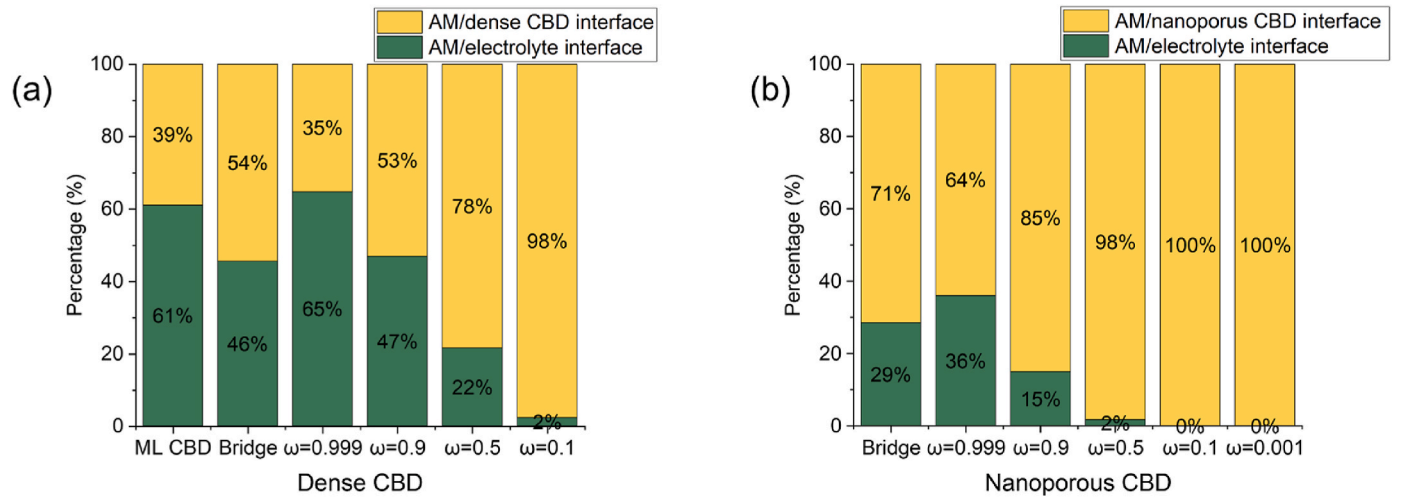


Fig. 3. Percentage distribution of AM/CBD and AM/electrolyte interfaces for dense and nanoporous CBD configurations: (a) dense CBD configurations using the bridge approach (second column) and the energy-based approach at various morphology factors ($\omega = 0.999, 0.9, 0.5, 0.1$), the ML segmented CBD is included for comparison (first column); (b) nanoporous CBD configurations using the bridge approach and the interface energy-based approach at various morphology factors ($\omega = 0.999, 0.9, 0.5, 0.1, 0.001$).

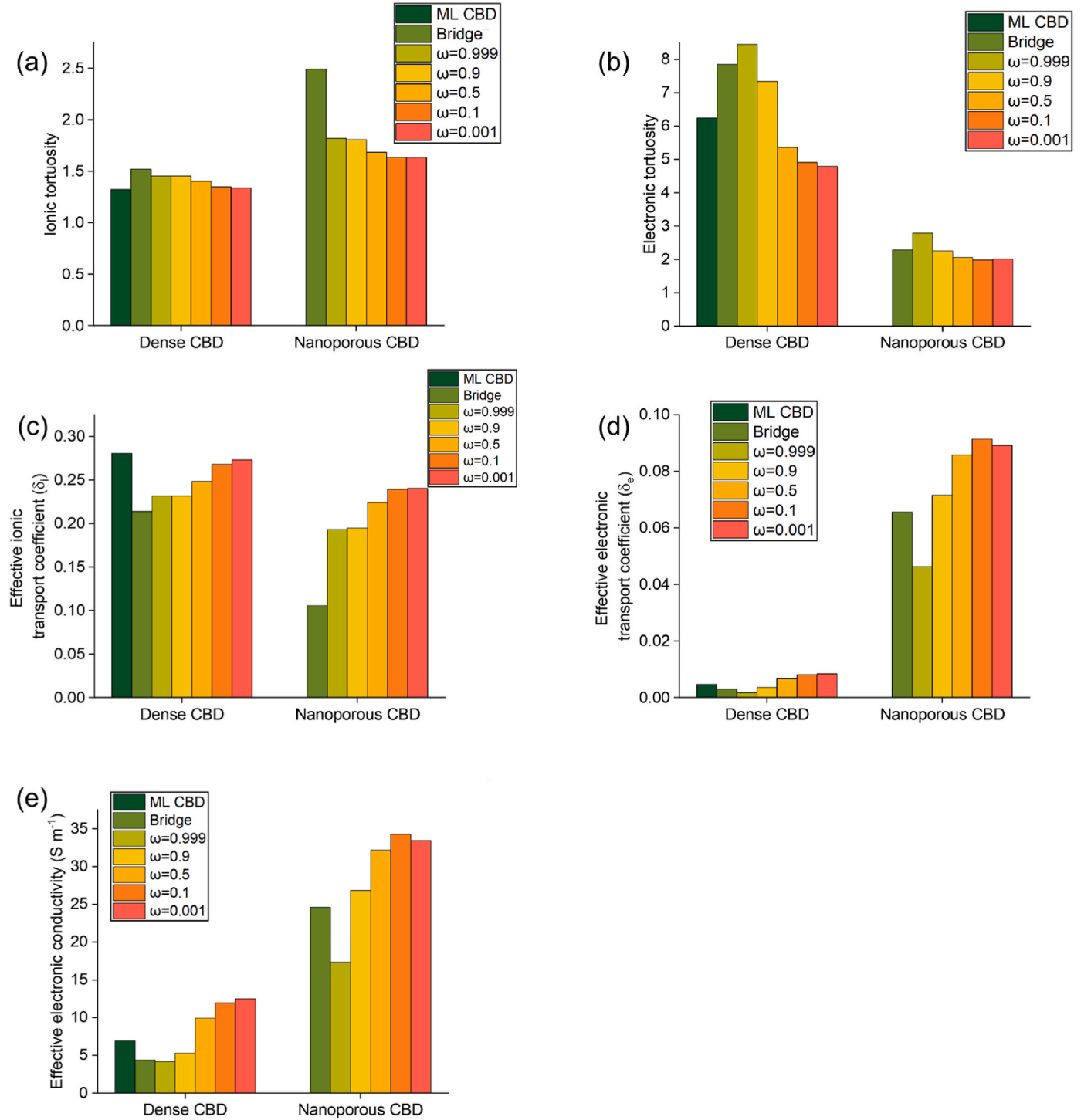


Fig. 4. Analysis of ionic and electronic transport in dense and nanoporous CBD configurations along Z-direction: (a) ionic tortuosity, determined using the pore network within the electrode; (b) electronic tortuosity determined using the generated CBD network within the electrode; (c), (d), and (e) display the effective ionic and electronic transport coefficients, as well as the effective electronic conductivity, respectively, in the electrodes featuring dense and nanoporous CBD configurations using ML-segmented, deterministic bridge, and interface energy-based approaches.

nanoporous configuration.

Fig. 4 (e) presents the effective electronic conductivity of the electrode for different CBD morphologies. These values were obtained by multiplying the effective electronic conductivity coefficient with the bulk electronic conductivity of dense ($1490\ S\ m^{-1}$) and nanoporous ($375\ S\ m^{-1}$) CBD, as reported in Ref. [8]. As shown in Fig. 4 (e), the transition from explicit dense to explicit nanoporous CBD morphologies leads to a noticeable increase in effective electronic conductivity. For

example, in the bridge CBD morphology, the effective electronic conductivity increases from $4.4\ S\ m^{-1}$ to $24.6\ S\ m^{-1}$. This is significantly higher than the bulk electrolyte ionic conductivity ($0.9\ S\ m^{-1}$ at $1200\ mol\ m^{-3}$ [45]), suggesting that electronic conduction is well-facilitated regardless of CBD morphology.

This detailed examination underscores the pivotal role of CBD structure in determining ionic and electronic tortuosities, and consequently, the ionic and electronic transport properties of electrode

materials, which are crucial for optimising battery performance.

3.3. Electrochemical performance of explicit CBD consideration

Fig. 5(a and b) depict the discharge curves at 1C and 5C for the half-cell featuring explicit dense and nanoporous CBD electrodes, respectively. These discharge curves were generated using COMSOL Multiphysics. Initially, the model computed the voltage as a function of discharge time (h). To express the x-axis in terms of specific capacity, the discharge time was multiplied by the applied current density ($A\ m^{-2}$) at the current collector surface, determined based on the specified C-rate (see Eq. (18) in SI). The resulting charge per unit area ($Ah\ m^{-2}$) was then multiplied by the inlet surface area of the electrode to obtain the total charge (Ah), which was subsequently divided by the mass of AM particles within the electrode to obtain the specific capacity ($mAh\ g^{-1}$).

In Fig. 5(a), the discharge performance varies significantly across different dense CBD morphologies, i.e. ML segmented CBD, bridge and varying ω at 0.999, 0.9, 0.5 and 0.1. The electrode with ML segmented CBD demonstrates specific capacities of $114\ mAh\ g^{-1}$ at 1C and $50\ mAh\ g^{-1}$ at 5C. Comparatively, the dense bridge CBD electrode shows lower specific capacities of $104\ mAh\ g^{-1}$ at 1C and $38\ mAh\ g^{-1}$ at 5C. Notably, as the value of ω decreases, a marked decline in performance is evident, with the $\omega = 0.1$ scenario delivering the lowest capacities of $45\ mAh\ g^{-1}$ at 1C and a mere $2\ mAh\ g^{-1}$ at 5C. These trends are linked to variations in the AM/electrolyte interface area, emphasising its critical role in electrochemical reactions during Li insertion.

Fig. 5(b) illustrates consistent discharge results across the nanoporous CBD configurations. The simulation results show that the discharge capacities for the various nanoporous CBD configurations are $125\text{--}135\ mAh\ g^{-1}$ at 1C and $75\text{--}80\ mAh\ g^{-1}$ at 5C, consistent with the experimental measured capacities of $\sim 130\ mAh\ g^{-1}$ at 1C [46] and $\sim 75\ mAh\ g^{-1}$ at 5C [47] for the same NMC532 cathode materials using standard testing protocols. It is extremely challenging to experimentally test all the nanoporous CBD configuration, whereas our simulation results show that the morphology with $\omega = 0.001$ exhibits the smallest capacity and hysteretic discharge curve, suggesting impediments to ionic transport in the nanoporous CBD surrounding the AM particles. This indicates that the structural configuration of the CBD significantly impacts the efficiency of ionic transport and the utilisation of the AM particles, which is crucial for optimising battery performance, especially at high discharge rates.

The transition from a dense (nanoporosity = 0) to a nanoporous (nanoporosity = 0.47) CBD configuration can be interpreted as an increase in nanopore diameter. The discharge capacity of the electrode with nanoporous CBD increases compared with the electrode with dense CBD because the additional reaction area between the AM and nanoporous CBD phase enhances electrochemical reactions, whereas reactions are limited to the electrolyte-AM interface in the dense CBD phase. In our current study, we did not explicitly investigate the effect of varying nanopore size within the CBD on electrochemical performance. However, identifying the optimal nanopore size will be an interesting direction for our future research.

In the study of electrodes with explicit dense CBD configurations, Fig. 5(c–d) depict the spatial distribution of SOL in AM particles and Li-ions in the electrolyte at the end of 5C discharge, respectively. Fig. 5(c) reveals that in dense CBD configurations, the utilisation of AM particles is primarily limited to their surface areas, suggesting the limited Li diffusion into the AM particles. This surface-focused lithiation evidences the inherent limitations of dense CBD morphologies, which could reduce the electrochemical accessibility of AM particles. Consequently, this restricted Li-ion access restrains the effective utilisation of the entire AM particle during rapid discharge conditions, leading to diminished capacity contributions from the internal regions of the AM particles. For instance, these electrodes with $\omega = 0.999$ and $\omega = 0.9$ CBD configurations show higher specific capacities at 1C and 5C than other electrodes with $\omega = 0.5$ and $\omega = 0.1$ CBD configurations (see Fig. 5(a)), offering a

favourable Li-ion transport in the electrolyte and better utilisation of AM particles. Notably, the dense film-like CBD morphology demonstrates significant underutilisation, correlating with inferior specific capacity performance. As illustrated in Fig. 5(d), the Li-ion distribution in the electrolyte exhibits minimal polarisation, further suggesting that the effective active area for electrochemical reactions or Li insertion is crucial in 3D microstructure-resolved modelling.

Concerning electrodes with explicit nanoporous CBD configurations, Fig. 5(e–f) show the spatial distribution of SOL in AM particles and Li-ion concentrations in the electrolyte at 5C discharge for $42.5\ mAh\ g^{-1}$, respectively. Fig. 5(e) indicates that AM particles in all configurations were initially utilised near the separator side, displaying electrode polarisation. Li-ions preferentially insert at the interface between the electrolyte and AM particles, indicated by the red colour areas, and exhibit severely heterogeneous utilisation across individual particles. This localised heterogeneity is mitigated in configurations with film-like coating CBD, particularly at $\omega = 0.1$ and $\omega = 0.001$. The Li-ion concentration in the electrolyte, as depicted in Fig. 5(f), mirrors this polarisation, with higher concentrations near the separator.

As the discharge progresses, arriving at the end of 5C discharge, the utilisation patterns of AM particles and Li-ion concentrations in the electrolyte are presented in Fig. 5(g–h). Fig. 5(g) shows that the utilisation of AM particles across all electrodes with nanoporous CBD remains relatively consistent. This behaviour is attributed to the ionic transport constraints imposed by the nanoporous CBD phases, where ionic transport exhibits hysteresis at the AM/nanoporous CBD interface. The pronounced polarisation of Li-ion concentrations in the electrolyte, shown in Fig. 5(h), underscores the significant role of the explicit nanoporous CBD configuration in influencing ionic transport and AM particle utilisation, which may impact long-term electrode degradation.

It is observed that while the Li-ion concentration in the electrolyte exhibits a gradient along the current collector–separator direction (Fig. 5(d) and (h)), the SOL in the AM particles does not follow the same trend (Fig. 5(c) and (g)). This discrepancy arises due to the distinct timescales and governing mechanisms of Li-ion transport in the electrolyte and lithium interaction into the AM particles. The Li-ion concentration gradient is primarily influenced by ionic transport limitations in the electrolyte, which depends on its intrinsic transport properties, i.e. ionic conductivity and diffusivity, and structural properties of the porous electrode, including tortuosity, porosity and thickness. In contrast, the SOL of the AM is governed by interfacial reaction kinetics and solid-state diffusion coefficient, which do not necessarily mirror the electrolyte concentration profile.

Similar behaviour can occur in practical electrodes, depending on their microstructure and ionic transport properties, local porosity variations and the interplay between ionic transport and intercalation kinetics. However, inhomogeneities in practical electrodes may differ from the idealised cases considered in our model. Future experimental validation, such as operando X-ray tomography [48], could provide further insights into the spatial lithiation behaviour under realistic operating conditions.

Fig. 5(i–j) quantifies the distributions of state of lithiation (SOL) in AM particles and Li-ion concentration in the electrolyte at the end of 5C discharge, analysing those metrics as a function of normalised distance from the current collector across the electrodes with different dense CBD morphologies. As illustrated in Fig. 5(i), most AM particles within the dense bridge and $\omega = 0.9$ configurations exhibit full lithiation, while only a subset of particles display a SOL range from 0.38 to 1. Notably, AM particles in the dense $\omega = 0.1$ CBD configuration largely remain unlithiated, highlighting significant underutilisation. Fig. 5(j) contrasts these observations, presenting a modest gradient of Li-ion concentration in the electrolyte for dense $\omega = 0.1$ CBD, as opposed to the steeper gradient seen in the dense bridge and $\omega = 0.9$ CBD configurations. This quantitative analysis underscores the fundamental role of the interface area between AM and electrolyte in influencing Li-ion dynamics within these configurations.

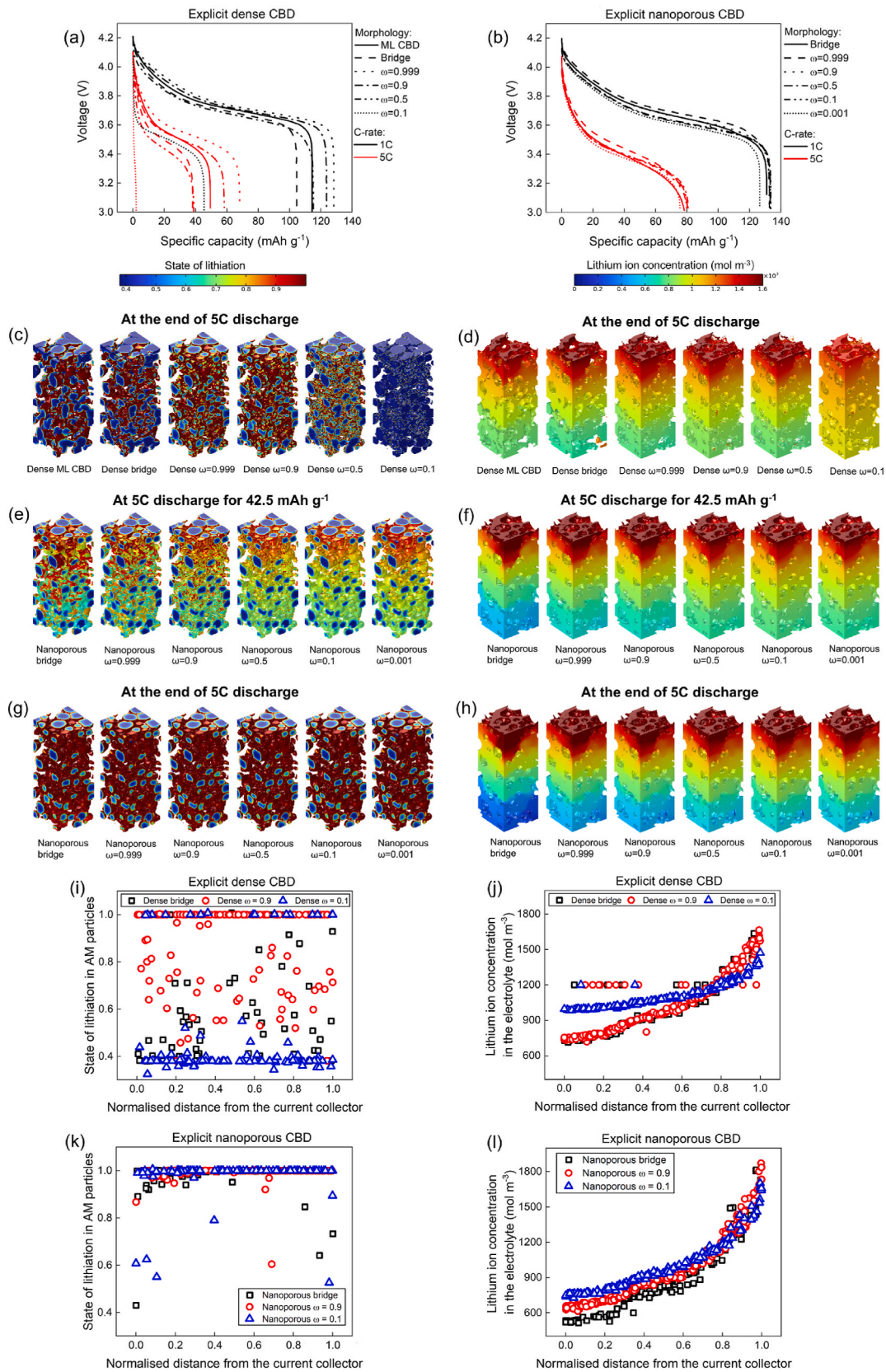


Fig. 5. Electrochemical performance of electrodes with explicit CBD configurations. Specific capacity at 1C and 5C discharge rates for: (a) explicit dense and (b) explicit nanoporous CBD; for explicit dense CBD configurations: (c) spatial distribution of SOL in AM particles and (d) Li-ion concentration in the electrolyte, both at the end of a 5C discharge; for explicit nanoporous CBD configurations: (e) spatial distribution of SOL in AM particles and (f) Li-ion concentration in the electrolyte at a 5C discharge for 42.5 mAh g^{-1} , with (g) and (h) displaying these metrics at the end of 5C discharge; (i) SOL of all AM particles and (j) Li-ion concentration in the electrolyte, as a function of normalised distance from the current collector for dense bridge, dense $\omega = 0.9$, and dense $\omega = 0.1$ CBD morphologies at the end of 5C discharge; (k) and (l) providing same measures for nanoporous bridge, nanoporous $\omega = 0.9$, and nanoporous $\omega = 0.1$ morphologies at the end of 5C discharge.

Fig. 5 (k-l) similarly quantifies the SOL distribution in the AM particles and Li-ion concentration in the electrolyte for electrodes configured with nanoporous CBD. Fig. 5 (k) shows uniform lithiation levels across AM particles at the end of 5C discharge, reflecting consistent utilisation and discharge capacity. Correspondingly, Fig. 5 (l) depicts a consistent range of Li-ion concentration in the electrolyte,

approximately from 500 to 1900 mol m⁻³, with minor discrepancies appearing only near the current collector among different nanoporous morphologies.

Overall, the results from Fig. 5(a-l) indicate that the microstructural configuration of CBD within the electrode, whether dense or nanoporous, plays a significant role in defining the ionic transport pathways

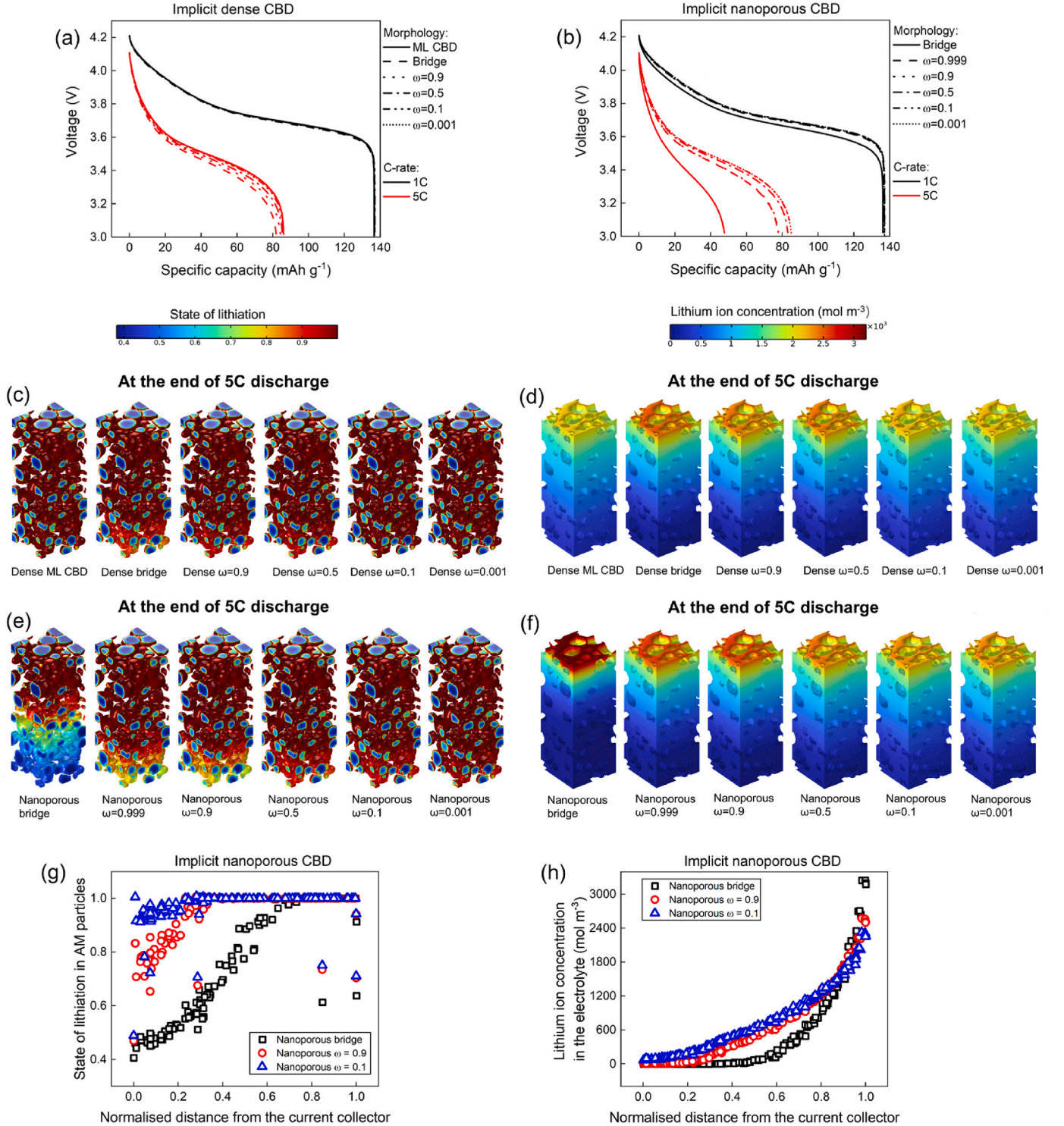


Fig. 6. Electrochemical performance of electrodes with implicit CBD configurations. Specific capacity at 1C and 5C discharge rates for: (a) implicit dense and (b) implicit nanoporous CBD; for implicit dense CBD configurations: (c) spatial distribution of SOL in AM particles and (d) Li-ion concentration in the electrolyte, both at the end of a 5C discharge; for implicit nanoporous CBD configurations: (e) spatial distribution of SOL in AM particles and (f) Li-ion concentration in the electrolyte, both at the end of 5C discharge; (g) SOL of all AM particles and (h) Li-ion concentration in the electrolyte, as a function of normalised distance from the current collector for nanoporous bridge, nanoporous $\omega = 0.9$, and nanoporous $\omega = 0.1$ CBD morphologies at the end of 5C discharge.

and the utilisation efficiency of AM particles. These structural effects are crucial for optimising battery performance under high discharge rates, emphasising the need for careful design and optimisation of the electrode architecture to enhance capacity retention and operational efficiency across varying conditions. This comprehensive analysis underscores the profound impact of electrode microstructure on the performance and longevity of LIBs.

Furthermore, the ω parameter, which determines the preferential deposition behaviour of CBD onto AM or pre-deposited CBD, plays a crucial role in optimising electrode microstructure for enhanced electrochemical performance. Controlling ω through fabrication parameters, including slurry formulation, drying conditions, and processing sequence, allows fine-tuning of interface area, transport properties, and CBD connectivity. For example, faster solvent evaporation promotes CBD agglomeration and increases ω , whereas slower drying results in a more uniform CBD distribution on AM, leading to lower ω [49]. Additionally, modifying surface interactions through binder chemistry and dispersant concentration can improve CBD adhesion to AM, further tuning ω [50]. Other processing factors, such as mixing strategy, solid content, and shear rate, also influence CBD dispersion and deposition behaviour, affecting ω [51]. While this study primarily focuses on computational optimisation, our future work will incorporate experimental validation to refine processing conditions further and determine the optimal ω that maximises electrode performance.

3.4. Electrochemical performance of implicit CBD consideration

Using the effective ionic and electronic transport coefficients obtained from Fig. 4(c–d), the ionic and electronic transport properties in the lumped CBD and electrolyte (cf. Fig. 1(c)) phase were corrected and adopted as the varying input parameters in the modelling. This correction facilitated the conversion from explicit CBD configurations to implicit ones. The discharge curves of half-cells with implicit dense and nanoporous CBD configurations were illustrated in Fig. 6(a–b) at 1C and 5C discharge rates. As depicted in Fig. 6(a), the half-cell with an implicit dense CBD configuration demonstrates a discharge curve with significantly higher specific capacity performance than its explicit counterpart shown in Fig. 5(a). This enhancement is attributed to the assumption of homogeneous distribution of CBD around the AM particles in the implicit modelling setting, leading to nearly identical utilisation of AM particles and distributions of Li-ion in the electrolyte, as observed in Fig. 6(c–d) at the end of a 5C discharge.

Fig. 6(b) reveals that the discharge curves for the implicit nanoporous CBD configurations are consistent at 1C across different morphologies (ω), from agglomerate-like ($\omega = 0.999$) to film-like structures ($\omega = 0.001$), while the specific capacities increase as ω decreases at 5C discharge. The electrode with nanoporous CBD bridge shows a notable reduction in specific capacity at a 5C rate, nearly halving compared to other CBD configurations. This decrease correlates to the effective ionic transport coefficients shown in Fig. 2(c) and is mirrored by severe polarisation in the utilisation of AM particles and significant electrolyte polarisation, as depicted in Fig. 6(e–f). As reported in our previous studies [12], such electrolyte polarisation severely impedes Li-ion transport, leading to underutilisation of AM particles. Fig. 4 shows the simultaneous decline in ionic transport coefficient underscores that ionic transport governs electrochemical performance and plays a dominant role in optimising electrode microstructure, which is markedly influenced by the morphology of CBD within the electrode. Additionally, as shown in Fig. 6(b) and 6(e) for the electrode with film-like nanoporous CBD morphology ($\omega = 0.1$ and $\omega = 0.001$), it indicates that surface coating could enhance the utilisation efficiency of AM particles by facilitating Li-ion transport. This improvement can be achieved through various methods such as the sol-gel process [52], chemical vapour deposition [53], physical vapour deposition [54], and atomic layer deposition [55].

Fig. 6(g–h) quantifies the distributions of SOL in all AM particles and

Li-ion concentrations in the electrolyte for electrodes with implicit nanoporous CBD, measured at the end of a 5C discharge. As observed in Fig. 6(g–h), AM particles in electrodes with implicit nanoporous $\omega = 0.9$ and $\omega = 0.1$ CBD configurations show high SOL and modest polarisation of electrolyte concentration, indicating effective and uniform utilisation of both AM particles and electrolyte, which is essential for achieving high capacity and efficiency. In contrast, the electrode featuring the implicit nanoporous bridge CBD morphology displays significant polarisation in AM particle utilisation and Li-ion concentration in the electrolyte, evidenced by nearly zero Li concentrations from 0 to 0.4 normalised distance from the current collector.

In the implicit CBD scenario, Li-ion transport is governed by the effective ionic transport coefficient for the pore phase, which influences overall transport resistance. At higher discharge rates, a lower effective ionic transport coefficient increases resistance, causing significant concentration polarisation in the electrolyte and a non-uniform Li-ion distribution. Consequently, this leads to a gradient in the SOL across AM particles, particularly near the separator where Li-ion availability is limited. This effect is further pronounced in cases with lower transport coefficients, such as the nanoporous bridge CBD case ($\delta_i = 0.11$), where severe transport restrictions result in the lowest discharge capacity. These observations underscore the critical impact of CBD morphology on electrode performance, highlighting how structural variations can profoundly affect the efficiency and uniformity of Li-ion dynamics within the battery.

In practical electrode designs, explicit and implicit CBD configurations can coexist within the same electrode. The spatial distribution of these configurations will influence performance by affecting electronic and ionic transport pathways, and will in turn affect the electrochemical characteristics. For example, if the top half of the electrode (near the separator) features an explicit dense CBD configuration while the bottom half (near the current collector) adopts an implicit CBD representation, the utilisation of AM particles during discharge would become non-homogenous in the top half. Although this study does not explicitly model the coexistence of these configurations, future studies will further explore the optimised hybrid structures to enhance electrochemical efficiency.

4. Conclusions

This study thoroughly examined the impact of carbon binder domain (CBD) morphology on the electrochemical performance of LIB electrodes, employing both explicit and implicit representations. The findings from these representations have exposed critical aspects of electrode microstructure design.

Analysis of explicit CBD morphology revealed that the interface active area between the AM particles and the electrolyte is crucial, particularly under scenarios of dense CBD. This configuration was found to significantly influence the specific discharge capacity, which varied markedly with changes in the interface area. Moreover, when CBD was modelled as a nanoporous structure with limited ionic transport capabilities, a stabilised discharge capacity emerged, suggesting that a film-like CBD facilitates locally homogeneous lithiation of AM particles.

Conversely, when transitioning from explicit to implicit CBD morphology, the investigations underscored the substantial role of the ionic transport coefficient in determining discharge capacity. A reduction ionic transport coefficient dropped from 0.21 to 0.11 resulted in a 40 % decrease in specific capacity at a 5C rate. The results highlighted CBD morphology influenced effective electronic and ionic transport, and their combined influence on battery performance.

The methodology presented in this study provides essential guidance for 3D microstructure-resolved modelling of LIBs. It demonstrates how incorporating various CBD morphologies into the existing AM particles obtained from XCT scanning, manufacturing modelling, or similar methods can enhance modelling accuracy and efficiency. This integration facilitates a deeper understanding of electrode behaviour and

optimisation, thus aiding in developing batteries with improved performance and longevity.

CRedit authorship contribution statement

Wei Sun: Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Chun Huang:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization, Investigation, Software.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jpowsour.2025.236904>.

Data availability

Data will be made available on request.

References

- [1] J.-M. Tarascon, M. Armand, *Nat* 414 (2011) 359–367.
- [2] T. Kim, W. Song, D.-Y. Son, L.K. Ono, Y. Qi, *J. Mater. Chem. A* 7 (2019) 2942–2964.
- [3] A.M. Boyce, D.J. Cumming, C. Huang, S.P. Zankowski, P.S. Grant, D.J.L. Brett, P. R. Shearing, *ACS Nano* 15 (2021) 18624–18632.
- [4] R. Schmich, R. Wagner, G. Hörpel, T. Placke, M. Winter, *Nat. Energy* 3 (2018) 267–278.
- [5] C. Huang, N.P. Young, J. Zhang, H.J. Snaith, P.S. Grant, *Nano Energy* 31 (2017) 377–385.
- [6] A.M. Colclasure, A.R. Dunlop, S.E. Trask, B.J. Polzin, A.N. Jansen, K. Smith, *J. Electrochem. Soc.* 166 (2019) A1412–A1424.
- [7] C. Huang, M.D. Wilson, K. Suzuki, E. Liotti, T. Connelly, O.V. Magdysyuk, S. Collins, F. Van Assche, M.N. Boone, M.C. Veale, A. Lui, R.-M. Wheeler, C.L. Leung, *Adv. Sci.* 9 (2022) 1–12.
- [8] R. Ge, A.M. Boyce, Y. Sun, P.R. Shearing, P.S. Grant, D.J. Cumming, R.M. Smith, *ACS Appl. Mater. Interfaces* 15 (2023) 27809–27820.
- [9] B.L. Trembacki, A.N. Mistry, D.R. Noble, M.E. Ferraro, P.P. Mukherjee, S. A. Roberts, *J. Electrochem. Soc.* 165 (2018) E725–E736.
- [10] M. Ebner, F. Feldmayer, F. Marone, M. Stampanoni, V. Wood, *Adv. Energy Mater.* 3 (2013) 845–850.
- [11] X. Lu, S.R. Daemi, A. Bertei, M.D.R. Kok, K.B. O'Regan, L. Rasha, J. Park, G. Hinds, E. Kendrick, D.J.L. Brett, P.R. Shearing, *Joule* 4 (2020) 2746–2768.
- [12] W. Sun, C. Huang, *J. Power Sources* 611 (2024) 234774.
- [13] F.L.E. Usseglio-Viretta, A.M. Colclasure, A.R. Dunlop, S.E. Trask, A.N. Jansen, D. P. Abraham, M.-T.F. Rodrigues, E.J. Dufek, T.R. Tanim, P.R. Chinnam, Y. Ha, K. Smith, *J. Electrochem. Soc.* 169 (2022) 070519.
- [14] X. Lu, G.J. Lian, J. Parker, R. Ge, M.K. Sadan, R.M. Smith, D. Cumming, *J. Power Sources* 592 (2024) 233916.
- [15] C.L.A. Leung, M.D. Wilson, T. Connelly, S.P. Collins, O.V. Magdysyuk, M. N. Boone, K. Suzuki, M.C. Veale, E. Liotti, F. Van Assche, A. Lui, C. Huang, *Mater. Today Energy* 31 (2023) 101224.
- [16] S.R. Daemi, C. Tan, T. Volkenandt, S.J. Cooper, A. Palacios-Padros, J. Cookson, D.J. L. Brett, P.R. Shearing, *ACS Appl. Energy Mater.* 1 (2018) 3702–3710.
- [17] L. Zielke, T. Hutzenlaub, D.R. Wheeler, C.-W. Chao, I. Manke, A. Hilger, N. Paust, R. Zengerle, S. Thiele, *Adv. Energy Mater.* 5 (2015) 1401612.
- [18] M. Ghadban, M. Sabharwal, C. Rea, X. Li, A.E. Goode, M. Smith, C. Murphy, M. Secanell, *J. Power Sources* 562 (2023) 232745.
- [19] X. Lu, A. Bertei, D.P. Finegan, C. Tan, S.R. Daemi, J.S. Weaving, K.B. O'Regan, T.M. M. Heenan, G. Hinds, E. Kendrick, D.J.L. Brett, P.R. Shearing, *Nat. Commun.* 11 (2020) 2079.
- [20] F.L.E. Usseglio-Viretta, A. Colclasure, A.N. Mistry, K.P.Y. Claver, F. Pouraghajan, D.P. Finegan, T.M.M. Heenan, D. Abraham, P.P. Mukherjee, D. Wheeler, P. Shearing, S.J. Cooper, K. Smith, *J. Electrochem. Soc.* 165 (2018) A3403–A3426.
- [21] C. Huang, P.S. Grant, *J. Mater. Chem. A* 6 (2018) 14689–14699.
- [22] C. Huang, M. Dontigny, K. Zaghbi, P.S. Grant, *J. Mater. Chem. A* 7 (2019) 21421–21431.
- [23] A.M. Boyce, E. Martínez-Pañeda, A. Wade, Y.S. Zhang, J.J. Bailey, T.M.M. Heenan, D.J.L. Brett, P.R. Shearing, *J. Power Sources* 526 (2022) 231119.
- [24] C. Huang, M.D. Wilson, B. Cline, A. Sivarajah, W. Stolp, M.N. Boone, T. Connelly, C.L.A. Leung, *Cell Rep. Phys. Sci.* 5 (2024) 101995.
- [25] C.L.A. Leung, M.D. Wilson, T. Connelly, C. Huang, *J. Synchrotron Radiat.* 31 (2024) 888–895.
- [26] A.M. Boyce, X. Lu, D.J. Brett, P.R. Shearing, *J. Power Sources* 542 (2022) 231779.
- [27] R. Ge, A.M. Boyce, Y. Shui Zhang, P.R. Shearing, D.J. Cumming, R.M. Smith, *Chem. Eng. J.* 465 (2023) 142749.
- [28] M.J. Shojai, A. Sivarajah, T. Safdar, O.V. Magdysyuk, C.L.A. Leung, C. Huang, *Solid State Ionics* 422 (2025) 1–10.
- [29] A.N. Mistry, K. Smith, P.P. Mukherjee, *ACS Appl. Mater. Interfaces* 10 (2018) 6317–6326.
- [30] B.L. Trembacki, D.R. Noble, V.E. Brunini, M.E. Ferraro, S.A. Roberts, *J. Electrochem. Soc.* 164 (2017) E3613–E3626.
- [31] B.L. Trembacki, D.R. Noble, M.E. Ferraro, S.A. Roberts, *J. Electrochem. Energy Convers. Storage* 17 (2020) 041001.
- [32] A.C. Ngandjong, T. Lombardo, E.N. Primo, M. Chouchane, A. Shodiev, O. Arcelus, A.A. Franco, *J. Power Sources* 485 (2021) 229320.
- [33] C. Liu, T. Lombardo, J. Xu, A.C. Ngandjong, A.A. Franco, *Energy Storage Mater.* 54 (2023) 156–163.
- [34] M. Chouchane, A. Rucci, T. Lombardo, A.C. Ngandjong, A.A. Franco, *J. Power Sources* 444 (2019) 227285.
- [35] M. Chouchane, A.A. Franco, *Energy Storage Mater.* 47 (2022) 649–655.
- [36] X. Lu, X. Zhang, C. Tan, T.M.M. Heenan, M. Lagnoni, K. O'Regan, S. Daemi, A. Bertei, H.G. Jones, G. Hinds, J. Park, E. Kendrick, D.J.L. Brett, P.R. Shearing, *Energy Environ. Sci.* 14 (2021) 5929–5946.
- [37] S. Berg, D. Kutra, T. Kroeger, C.N. Straehle, B.X. Kausler, C. Haubold, M. Schiegg, J. Ales, T. Beier, M. Rudy, K. Eren, J.I. Cervantes, B. Xu, F. Beuttenmueller, A. Wolny, C. Zhang, U. Koethe, F.A. Hamprecht, A. Kreshuk, *Nat. Methods* 16 (2019) 1226–1232.
- [38] W. Sun, Q.M. Li, *Int. J. Mech. Sci.* 216 (2022) 106976.
- [39] T.-T. Nguyen, B. Delobel, M. Berthe, B. Fleutot, A. Demortière, C. Delacourt, *J. Electrochem. Soc.* 169 (2022) 040546.
- [40] F.L.E. Usseglio-Viretta, P. Patel, E. Bernhardt, A. Mistry, P.P. Mukherjee, J. Allen, S.J. Cooper, J. Laurencin, K. Smith, *SoftwareX* 17 (2022) 100915.
- [41] D. Schmidt, M. Kamlah, V. Knoblauch, *J. Energy Storage* 17 (2018) 213–223.
- [42] C. Huang, C.L.A. Leung, P. Leung, P.S. Grant, *Adv. Energy Mater.* 11 (2021) 1–14.
- [43] C. Huang, M.D. Wilson, B. Cline, A. Sivarajah, W. Stolp, M.N. Boone, T. Connelly, C.L.A. Leung, *Cell Reports Physical Science* (2024) 101995.
- [44] W. Sun, Q.M. Li, P. Xiao, P. Carbone, *J. Energy Storage* 97 (2024) 112940.
- [45] A. Nyman, M. Behm, G. Lindbergh, *Electrochim. Acta* 53 (2008) 6356–6365.
- [46] X. Zhu, L. Cheng, H. Yu, F. Xu, W. Wei, L.-Z. Fan, *Journal of Materiomics* 8 (2022) 649–655.
- [47] N. Anansuksawat, T. Sangsanit, S. Prempluem, K. Homlamai, W. Tejangkura, M. Sawangphruk, *Chem. Sci.* 15 (2024) 2026–2036.
- [48] C. Huang, M.D. Wilson, B. Cline, A. Sivarajah, W. Stolp, M. Boone, T. Connelly, C. L.A. Leung, *J. Phys.: Energy* 7 (2025) 025009.
- [49] F. Font, B. Protas, G. Richardson, J.M. Foster, *J. Power Sources* 393 (2018) 177–185.
- [50] J. Entwistle, R. Ge, K. Pardikar, R. Smith, D. Cumming, *Renew. Sustain. Energy Rev.* 166 (2022) 112624.
- [51] L. Ouyang, Z. Wu, J. Wang, X. Qi, Q. Li, J. Wang, S. Lu, *RSC Adv.* 10 (2020) 19360–19370.
- [52] S. Kalluri, M. Yoon, M. Jo, S. Park, S. Myeong, J. Kim, S.X. Dou, Z. Guo, J. Cho, *Adv. Energy Mater.* 7 (2016) 1601507.
- [53] R. Tian, H. Liu, Y. Jiang, J. Chen, X. Tan, G. Liu, L. Zhang, X. Gu, Y. Guo, H. Wang, L. Sun, W. Chu, *ACS Appl. Mater. Interfaces* 7 (2015) 11377–11386.
- [54] M. Salah, T. Pathirana, E. Alvarez de Eulate, C. Hall, B. Stoehr, S. Rudd, R. Kerr, M. Fabretto, *ACS Appl. Energy Mater.* 4 (2021) 12243–12256.
- [55] X. Wang, G. Yushin, *Energy Environ. Sci.* 8 (2015) 1889–1904.