



Mixing duration and performance in NMC electrodes: Insights from 3D imaging and electrochemical simulation

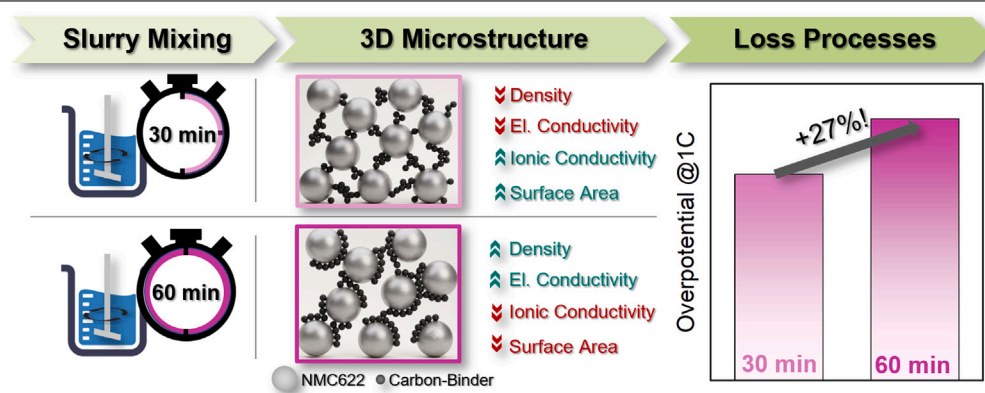
Adrian Lindner^a, Roman Buchheit^b, Maximilian Lechner^c, Fabian Biebl^b,
Wolfgang Menesklou^a, Ulrike Krewer^a,*

^a Karlsruhe Institute of Technology, Institute for Applied Materials - Electrochemical Technologies (IAM-ET), Adenauerring 20b, Karlsruhe, 76131, Germany

^b Math2Market GmbH, Richard-Wagner-Straße 1, Kaiserslautern, 67655, Germany

^c Technical University of Munich (TUM), TUM School of Engineering and Design, Institute for Machine Tools and Industrial Management (iwb), Boltzmannstraße 15, Garching b. München, 85748, Germany

GRAPHICAL ABSTRACT



HIGHLIGHTS

- Long mixing times improve carbon-binder-domain (CBD) distribution, increase electronic conductivity and active material attachment.
- 3D electrode scans are used as input for structurally resolved simulations to determine loss processes resulting from different mixing times.
- Reduced active area and longer diffusion paths resulting from higher CBD attachment reduce performance compared to electrodes with shorter mixing time.

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ABSTRACT

In lithium-ion batteries, energy and power densities depend heavily on the electrode microstructure. Because their structure is determined during the manufacturing process, manufacturing steps must be understood in great detail to tailor the electrodes to the specific needs of the application. The present work investigates the influence of wet mixing time on the conductive networks and consequently on electrode performance. FIB-SEM tomography coupled with 3D-resolved simulations unveils an increased homogeneity of the carbonaceous network inside a NMC622 electrode for longer dispersion times. Quantitative structural analysis is used to correlate microstructure and electrochemical performance using rate capability tests and electrochemical impedance spectroscopy. Longer mixing increases both active material particle coverage in the carbon binder

* Corresponding author.

E-mail address: ulrike.krewer@kit.edu (U. Krewer).

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domain (CBD) and homogeneity of the CBD, leading to higher electronic conductivity and reduced contact resistance. Conversely, the more homogeneous CBD negatively affects ionic transport inside the electrolyte due to pore blockage and lowers available surface area of the active material. This leads to a 21% increase in solid diffusion length and a reduction in surface area of 21% after doubling the wet mixing time from 30 to 60 min. Overall, this contribution untangles the complex interaction between wet mixing duration, microstructural evolution, and electrode performance.

1. Introduction

Establishing a successful battery value chain in Europe will require a deep and quantitative understanding of the relevant processing steps and their interaction with cell quality and performance. This includes ways to ensure suitable conductive networks inside the porous electrodes, enabling fast electronic conduction without limiting ion transport or active surface area. This becomes especially critical for positive electrode active materials with low intrinsic electronic conductivity like lithium-nickel-manganese-cobalt-oxide (NMC) [1,2]. Multiple studies have reported the importance of the carbon-binder-domain (CBD) in the positive electrode [3–5]. While studies exist that correlate the three-dimensional CBD structure to performance [6,7], the question remains whether the CBD structure can actually be tailored directly during the electrode manufacturing process. Since electrode manufacturing is a complex and multi-step process with significant interactions and uncertainties [8], stages like mixing and dispersion need to be studied in isolation to quantify their impact.

Firstly, the electrode formulation defines the types and amounts of conductive additives and binders used. Studies in this field often compare the influence of different types and amounts of additives on electrode conductivity and performance without addressing how processing steps like mixing, coating, drying, and calendaring affect CBD distribution and morphology [9,10]. In addition to an improvement in electronic electrode properties, Tran et al. showed how graphite acts as a lubrication additive, improving calendaring and reducing damage to electrode sheets during densification [11]. Studies in recent years identified the mixing processes, both dry and wet, as critical factors in defining CBD properties during electrode manufacturing. Dry mixing refers to the powder homogenization process containing active material and conductive additives like graphite, carbon black, graphene, or carbon nanotubes and was previously shown to influence particle size distribution and morphology during positive [12] and negative electrode [13] manufacturing. To support findings from experimental studies on the dry mixing process, Asylbekov et al. also employed discrete element method modeling to understand changes in carbon black morphology during high-intensity dry mixing [14]. Weber et al. recently investigated the structuring of carbon black (CB) during dry and wet mixing in a twin-extruder process and were able to increase specific discharge capacities by fine-tuning CB dispersion [15]. While this and other previous studies often contain information on particle sizes and electronic and electrochemical performance, the real three-dimensional electrode structure was not investigated. Including three-dimensional information about the CBD structure would allow a beneficial correlation between performance and the production process. However, due to the challenging nature and poor accessibility without specialized imaging techniques, the 3D CBD structure is often neglected.

Following the dry mixing process, wet mixing, sometimes referred to as dispersion or slurry mixing, is the process responsible for homogenization of the binder solution with the dry-mixed components. Rheological properties, responsible for how the slurry behaves during the coating process as a function of slurry preparation, have already been investigated extensively [16–20]. As conductive additives and binder solution come in contact for the first time during this step, the wet mixing process is often discussed when CBD networks are investigated. Many attempts were made to deepen the understanding

of the processability of the wet mixing process and to find optimal process parameters. Hoffmann et al. used different mixing and dispersion strategies to successfully tune the manufacturing of ultrathick positive electrode sheets and provide experimental data on electrode performance [21]. However, they only include cross-sectional 2D imaging, making a correlation to the actual 3D microstructure difficult. And while they show how mixing conditions can influence processability of the slurry and the discharge performance of electrodes, they do not establish a link between the three-dimensional CBD network and electrode behavior, leaving the question of how the changed CBD network would affect specific loss processes like surface kinetics and solid diffusion open, which we answer in our study. In contrast, Kremer et al. combined experimental results with virtually generated 3D electrode structures based on synchrotron imaging to elucidate the manufacturing process of thick electrode structures and its influence on electrode performance. While the study uses 3D data, the CBD distribution had to be virtually added into the structure based on 2D energy-dispersive X-ray spectroscopy data. They were able to show that mixing with lower shear rates leads to higher specific discharge capacities across a high range of current densities, stating that mixing at high shear rates promotes binder migration, negatively affecting electrode performance [22]. In a more rheology-focused study, Wang et al. could demonstrate how altering the mixing sequence can positively influence rheological properties of the slurry, ultimately having an effect on NMC microstructure after drying. They found better rate capability of the electrodes when adding CBD and NMC in two separate mixing steps, which leads to a more porous CBD after drying, facilitating rate capability [23]. However, similar to other studies in this field, they do not include three-dimensional imaging to support their findings. We therefore suggest that our approach, relying on high-resolution 3D tomography data, fills a gap in existing literature by providing insight into the formation of three-dimensional carbon networks. This dimensionality is crucial when evaluating rate-limiting properties like ionic and electronic transport, surface kinetics, and solid-diffusion lengths inside the particle structure.

In a similar study, Mayer et al. studied the influence of carbon black dispersion on the performance of NMC622 electrodes. They combined DEM simulation, 3D imaging with focused-ion-beam scanning-electron-microscopy (FIB-SEM) tomography, and an in-depth investigation of electrode properties to determine optimal carbon black particle sizes. Due to the use of FIB-SEM instead of synchrotron data, they were able to resolve the CBD directly inside the structure and determined an optimal particle size for a 5C discharge [24]. However, due to the lack of a direct correlation between the 3D structure and electrochemical performance and relatively small volumes of the FIB-SEM tomography, they reported difficulties in determining the influence of mixing times on different loss processes during discharge. Griesl et al. studied the influence of mixing duration on electrode tortuosity among other properties. They were able to find subtle differences in tortuosity after mixing for 30 min and 60 min, but provided no additional insight into the resulting cell performance, leaving the question open whether the mixing duration affects the formation of the CBD network in a positive or negative way [25]. Similar findings were reported by Zapata Dominguez et al. who studied the correlation between mixing speed and slurry rheology of NMC-based electrodes and found an increase in tortuosity when slurries were mixed at higher speeds [26]. Both these results suggest a negative impact of mixing duration and speed on ion transport and, therefore, performance at higher current densities, yet a link between process, three-dimensional microstructure,

and performance could not be established. We demonstrate in this study that the addition of highly resolved 3D imaging is a valuable tool because it allows for the analysis of the entire CBD network, its influence on rate-limiting properties, and ultimately, its effect on discharge performance.

While some of the mentioned studies employ 3D imaging, electrochemical testing, or simulation approaches, very few studies exist that leverage all three. Especially FIB-SEM tomography has gained a lot of traction in the battery field since its first introduction in 2010 [27] because it is able to provide high contrast between active material, pore space, and CBD when coupled with suitable preparation [28–30]. Additionally, 3D data from FIB-SEM can be directly used as the domain for structurally resolved simulation in frameworks like BEST [31] or the BatteryDict module in GeoDict [32] to perform studies on electrode structures [33–35]. Leveraging high-resolution FIB-SEM tomography offers two significant advantages over digital-twin generators, as used, for example by Jang et al. [36] It enables a very fine degree of discretization to provide insight into small geometric features of the CBD, allowing for a more precise interpretation of the results. It also inherently complements digital-twin studies by providing real-life electrode structures, which generators can mimic to produce more realistic microstructures. Additionally, FIB-SEM tomography can be directly coupled to the manufacturing process by performing experiments on electrodes with varied processing conditions. In the case of virtually generated digital twins, this link must be established via complex simulation approaches which might introduce errors or deviations. We therefore strongly believe that studies using FIB-SEM tomography are a necessary complement to virtual digital-twin studies by providing real-life structural data and linking it directly to manufactured electrodes.

In a similar approach to investigate the interaction between processing, microstructure, and performance, Lim et al. utilized FIB-SEM tomography and structurally resolved modeling to look at the calendaring process [37]. Similarly to [7], they also found a correlation between electronic conductivity and ionic tortuosity, suggesting that process optimization can only improve one or the other. Laue et al. combined 3D microstructure generation with P2D modeling to come to similar conclusions [38]. They were able to show that a higher CBD content relative to the amount of active material will cause a disproportionate decrease in ionic conductivity and ultimately lower discharge capacity at higher rates. They also suggest that model-based approaches may be used to optimize the electrode structure virtually, arriving at better processing conditions.

Based on previous studies, we suggest that the wet mixing duration acts through the cumulative specific energy introduced into the slurry: increasing shear successively deagglomerates the carbon black structures remaining after dry mixing [39,40], increases the carbon surface available for PVDF adsorption, and thus redistributes the CBD from segregated agglomerates in the pore space towards a thinner phase adhering to the NMC particle surfaces [14,15]. As mixing intensity and duration both scale this energy input, findings obtained by varying shear rate or mixing speed [24,25] are mechanistically transferable. This redistribution acts in two opposing directions: a more continuous CBD lowers its tortuosity and raises the effective electronic conductivity, while the same redistribution increases the covered fraction of the NMC surface and narrows pore constrictions. Quantifying this trade-off on real 3D structures and evaluating its impact on discharge performance is the objective of this work.

This study aims to clarify the relationship between wet mixing duration, electrode microstructure, and electrochemical performance. Our study focuses on investigating large electrode volumes and performing fully 3D-resolved electrochemical simulations directly on the FIB-SEM data using the software GeoDict to understand the influence of mixing time on microstructural properties and, ultimately, on electrode performance. Compared to other studies following similar approaches [6, 41,42], we are able to perform our simulations at significantly finer

discretization (100 nm compared to 178 to 500 nm), allowing us to capture the role of fine CBD structures more accurately. Therefore, we are able to, for the first time, demonstrate how the rearrangement of CBD during longer wet mixing affects not only transport losses in the electrolyte but also kinetic and diffusive losses in the active material. Coupled with electrochemical impedance spectroscopy and rated discharge experiments, we provide valuable insights into the production of different electrode microstructures and their performance caused by changes to the duration of wet mixing. This study underlines the importance of tailoring conductive networks inside porous positive electrode structures for optimal active material utilization and improved electronic and ionic transport, leading to overall improved battery performance.

2. Methods and materials

2.1. Electrode manufacturing

To prepare the positive electrode slurry, N-Methyl-2-pyrrolidone (NMP, 328 634, Merck, Germany) and polyvinylidene fluoride (PVDF Solef 5130, Solvay, Belgium) were mixed in a disperser (Dispermat FM 10, VMA-Getzmann, Germany) at 2000 rpm until a uniform binder solution was achieved. This solution was then combined with $\text{Li}_1\text{Ni}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC622) powder (HED NCM-622 DT011, BASF, Germany), conductive carbon black (C-Nergy Super C65, Imerys, France), and conductive graphite (C-Nergy SFG6L, Imerys, France), which were pre-mixed at 1400 rpm for 1 min in a rotary revolution mixer (Speedmixer DAC 1100.2 VAC-P, Hauschild, Germany). The mixture was added to the binder solution and stirred for 30 min or 60 min at 2000 rpm in the disperser. The resulting slurries had a solid content of approximately 78% according to thermogravimetric analysis after mixing. During the mixing process, the temperature was kept below 25 °C by cooling the mixing container. After dispersing, the slurries were degassed for 5 min at 600 rpm and 250 mbar in the rotary mixer. The solid composition of the slurry is shown in Table 1. Subsequently, the slurries were coated on an aluminum current collector foil (15 µm, 1100-L, Hydro, Norway) with a tape casting lab coater (MSK-AFA-III, MTI, USA), yielding a wet film thickness of 200 µm. The drying was performed in a ventilated oven (Mehrzweck-Heissluftofen, Bartscher, Germany) at a temperature of 80 °C for 45 min. For calendaring, a calender (EA 102, Coatema, Germany) with a roll diameter of 400 mm (and a maximum line load of 1000 N mm⁻¹) was used. The single-sheet electrodes were calendared at a constant roller speed of 0.5 m min⁻¹ using unheated rolls at ambient temperature. All subsequent analysis was performed on calendared electrodes. Apart from the wet mixing duration, all process parameters were kept identical for both electrode variants: both slurries were prepared from the same material batches with the same formulation and solid content, coated in the same coating run at the same wet film thickness, dried under identical conditions, and calendared on the same machine with identical settings. Differences in the resulting microstructure are therefore caused by the wet mixing duration, either directly through the CBD distribution in the slurry or indirectly through its effect on the compaction behavior of the coating.

To demonstrate monotonicity within the investigated mixing times, we include results of an electrode slurry mixed for 45 min in the SI (S10 & SI Table 2).

2.2. Electrochemical experiments

All electrochemical testing was performed with the PAT-Cell system (EL-CELL GmbH, Hamburg, Germany). A glass fiber separator (glass microfiber filter, uncompressed thickness of 0.21 mm) and 150 µL electrolyte (1M LiPF_6 in EC/DMC 1:1 vol., Sigma Aldrich, Germany) were used for all experiments. The investigated electrodes were punched

Table 1
Cathode composition.

NMC622/wt.-%	Carbon black C65/wt.-%	Conductive graphite SFG6L/wt.-%	PVDF/wt.-%
95	2.25	0.75	2

to 18 mm diameter disks from the calendered sheets using the EL-Cut system (EL-CELL GmbH, Hamburg, Germany). For determining the open-circuit voltage (OCV) as well as rate capability of all investigated electrodes, half-cell setups were used with the NMC electrode as the working electrode (WE), lithium metal foil as the counter electrode (CE), and a lithium reference ring (RE). Experiments were performed using a BaSyTec CTS-Lab (BaSyTec GmbH, Germany) inside a climate chamber (ESPEC LU-114, ESPEC Europe GmbH, Germany) set to 25 °C. For OCV measurements, a 0.01 C (0.377 A m^{-2}) discharge was measured between 4.3 V and 3.0 V against the lithium reference. The same upper and lower voltage limits were then used for rated discharge experiments between 3.77 A m^{-2} (0.1 C) and 113.14 A m^{-2} (3 C). Charging was performed with a constant current density of 18.86 A m^{-2} (0.5 C). Measurements were repeated four times per current density to assess reproducibility. Electrochemical impedance spectroscopy (EIS) was performed in symmetrical cells using the same general setup as before but replacing the counter electrode with an identical NMC electrode, both previously set to 50% state-of-charge (3.8 V). All EIS measurements were performed using a Solartron Analytics 1470E Cell Test System with Scribner Multistat Software at 25 °C (Vötsch VT 4002). Spectra were recorded between 0.005 Hz and 10^6 Hz with the high-frequency domain being measured four times to assess reproducibility. Results were then scaled to the surface area of the two electrode disks (18 mm diameter) due to the symmetrical cell setup.

2.3. 3D microstructure reconstruction

The requirements for sample preparation and imaging for FIB-SEM tomography of positive lithium-ion battery electrodes were discussed in great detail in [7]. In this study, an identical preparation was used with vacuum infiltration of a two-component silicon resin (Elastosil R604B, Wacker Chemie, Germany) followed by deposition of the dual-material layer consisting of platinum and a two-component carbon adhesive (M-Bond, Micro Measurements, Germany). FIB-SEM imaging was performed inside a scanning electron microscope (1540 XB, Zeiss Microscopy, Oberkochen, Germany). According to the findings in our previous study [7], the region of interest (ROI) was selected to contain the entire thickness. Milling was performed using a current of 2 nA, realizing a slice thickness of roughly 90 nm. Imaging was performed at an acceleration voltage of 1.3 kV at a pixel size of 80 nm. Three tomographies were performed for each mixing duration, resulting in a total of six datasets. Microstructural parameters were later aggregated, but only one structure for each mixing duration was used for electrochemical simulation. All tomographic datasets were checked against thresholds for representativeness. Datasets used for electrochemical simulation need to contain the entire electrode thickness, as explained in [7]. Additionally, they need to include at least 500 NMC particles and match the threshold for representative volume elements (RVE) suggested by Ghadban et al. [43] ($15 \times 15 \times 15 \mu\text{m}^3$), and Almar et al. [44] ($>1.95 \times 10^4 \mu\text{m}^3$). All requirements are met for the 3D datasets investigated in this study. Our datasets are therefore representative for the investigated electrodes and significantly exceed the thresholds found in the literature.

Image processing in this study has two objectives. First, the image data is converted into segmented 3D datasets suitable for structurally-resolved simulations. Second, microstructural parameters are extracted from these datasets and correlated with electrochemical measurements and simulation results. Most of the processing steps are identical to our previous studies [45,46], and some more details are provided there. The image processing steps were performed in the following

order: alignment, cropping to the final ROI, curtaining removal, cubic voxel scaling, noise removal with anisotropic diffusion, greyvalue gradient adjustments, and contrast enhancement. Finally, three-phase segmentation into active material, carbon-binder-domain, and pore space was performed using a boosted tree classifier trained using the ImportGeo-Vol Module of GeoDict (2024 Release) [32,47].

2.4. Evaluation of the microstructure

Prior to the preparation of the 3D image data for structurally resolved electrochemical simulations, basic microstructural parameters were obtained to allow for a correlation between experimental and simulation results. First, volume fractions of active material, pore space, and carbon-binder-domain were calculated from the voxel counts in the structure. Since segmentation designated each voxel to a specific phase, the volume fractions could easily be obtained from the total voxel count. Electrode density was calculated using bulk densities shown in Table 2 using the MatDict module (2024 Release) (see SI). Specific surface areas for active material and CBD phase were also calculated with the MatDict module and considering the Minkowski parameter approach suggested in [48]. Electronic conductivities were computed using the ConductoDict module in GeoDict, using the parameters shown in Table 2 and assuming a state-of-charge of 50%. The algorithm in the ConductoDict module is based on the work of Wiegmann et al. [49]. Pore space tortuosity factor (τ^2 , sometimes referred to simply as tortuosity) was calculated using the DiffuDict module and based on [50], and geometric CBD tortuosity (τ) was calculated with the MatDict module (see SI). Symmetric boundary conditions were used for all calculations. Median particle diameters were calculated using the Estimate Grain Diameter function in the GrainFind module of GeoDict (see SI), not considering internal and closed pores. The average diffusion length within the NMC particles was determined by calculating the shortest path from each active-material voxel to the nearest electrolyte voxel. Detailed information on this calculation is provided in the SI and [51]. We emphasize that these homogenized values are not required for fully resolved simulations in BatteryDict, as they are inherently encoded in the microstructure.

2.5. Electrochemical simulation

2.5.1. Simulation domain

Because the computational cost of microstructure-resolved simulations scales with the number of voxels, we downsampled the scan by importing the voxelized structure gdt-file with ImportGeo-Vol. Thereby, we selected Image Processing → Image Size → Scale with the scale function *Nearest Neighbor* and re-sampled from a voxel length of 80 nm to 100 nm. This results in a 50% reduction in the number of voxels. A similar rescaling was previously validated in [7]. We created a battery structure based on this electrode data with a lithium anode and the NMC electrode using the *Design Battery* command in BatteryDict [52]. The current collectors and lithium reservoir are defined with a thickness of 3 voxel layers. Reducing the separator from an estimated compressed thickness of 100 μm in the real cell to 10 μm in the simulation domain further lowers the overall voxel count by adjusting the separator tortuosity. A validation for this adjustment is shown in the supporting information of this work (Fig. S2 & S3), where we demonstrate that alterations to simulated overpotentials or the resulting discharge profile are minor. The porosity and tortuosity factors of the separator and carbon-binder-domain (CBD) were defined as listed in Table 2. The total battery structure had a size of $919 \times 224 \times 442$ voxels for the cell with 60 min mixing duration and $919 \times 195 \times 277$ voxels for the cell with 30 min mixing duration. Here, the x-direction (919 voxels) represents the through-direction of the cell.

Table 2
Parameters used for electrochemical simulation.

Name	Symbol	Value	Source
Current collector (negative and positive electrode)			
Electronic conductivity	$\kappa_{s,CC}$	1000 S m ⁻¹	See SI
Lithium reservoir			
Electronic conductivity	$\kappa_{s,n}$	1000 S m ⁻¹	See SI
Maximum exchange current density	$i_{0,max,n}$	12.65 A m ⁻²	Fitted
Separator			
Thickness	d_{sep}	10 μ m	See SI
Porosity	ϕ_{sep}	0.5	Assumed
Tortuosity factor	τ_{sep}	10	See SI
Electrolyte			
Equilibrium concentration	$c_{e,eq}$	1000 mol m ⁻³	Measured
Transference number ²	t_+	Figure S1a	[53]
Ionic conductivity ³	κ_e	Figure S1b	[54]
Ionic diffusivity ³	D_e	Figure S1c	[54]
Active material (NMC622)			
Solid density	ρ_s	4.74 g cm ⁻³	[55]
Ionic diffusivity	D_s	Figure S2a	Adjusted from [22]
Electronic conductivity	$\kappa_{s,p}$	Figure S2b	Adjusted to NMC622 from [7]
Maximum lithium concentration	c_{max}	47 062 mol m ⁻³	Fitted
Maximum exchange current density	$i_{0,max,p}$	0.82 A m ⁻²	Adjusted to NMC622 from [7]
Open-circuit voltage (OCV)	U_0	Figure S3	Measured
Carbon-binder-domain⁵			
Solid density	ρ_{CBD}	1.91 g cm ⁻³	See SI
Electronic conductivity	κ_{CBD}	1000 S m ⁻¹	[7]
Porosity	ϕ_{CBD}	0.5	[7]
Tortuosity factor liquid	$\tau_{CBD,l}$	5	[7]
Tortuosity factor solid	$\tau_{CBD,s}$	5	[7]

2.5.2. Simulation

We simulated the lithiation of the NMC622 electrode which corresponds to discharging the cell, applying constant current density boundary conditions in the through direction and symmetric boundary conditions in the tangential directions. The current densities matched those used in the electrochemical experiments (3.77 A m⁻², 7.54 A m⁻², 18.86 A m⁻², 37.72 A m⁻², 75.43 A m⁻², 113.14 A m⁻²). The cut-off voltage was 3.3 V. We started the lithiation from an equilibrium voltage of 4.3 V.

For performing the simulation, we used a fully resolved model [52, 56–58] that is based on the equations listed in Table 3. The lithium transport is simulated dynamically and the electrochemical potentials are calculated in a stationary way, as the time evolution of the electron reconfiguration is much smaller than the time steps of the simulation and hence can be neglected. We did not consider double-layer formation at the electrode surface, as it is usually formed below one second after switching on the current – much shorter than the time step – and the charge amount stored in the double layer is much smaller than the charge transferred during the lithiation.

The material parameters used in the simulation are listed in Table 2. The CBD parameters in Table 2 were chosen such that the effective conductivity and the effective diffusivity were both 10 times smaller compared to their bulk values, similarly to [7], to account for CBD nanoporosity.

2.5.3. Calculation of overpotentials

The sum of overpotentials in electrode and electrolyte is the difference between the cell voltage during discharging and the equilibrium cell voltage at the respective state-of-charge (SOC). The different contributions to the overpotential show the decrease of cell voltage due to the different physical phenomena inside the cell during the discharge simulations. These contributions are calculated using the equations implemented in BEST [56–58]. The BEST solver uses the 3D equivalents to the 1D equations that were described in [59].

The kinetic overpotential represents the energy losses due to the intercalation of lithium ions from the electrolyte into NMC during lithiation. The total kinetic overpotential η_{Kin} is the mean value of

the local kinetic overpotential $\varphi_{s,p} - \varphi_e - U_0$ weighted with the local Butler-Volmer interface current $i_p \cdot A$. It is calculated via the formula:

$$\eta_{Kin} = -\frac{1}{I} \sum_{all \ interface \ walls} [i_p A (\varphi_{s,p} - \varphi_e - U_0)] \quad (1)$$

where i_p is the local Butler-Volmer interface current density, $\varphi_{s,p}$ the local solid potential, φ_e the local electrolyte potential, U_0 the local open-circuit voltage, A the voxel face area (voxel length²), and I the total current.

Non-homogeneous lithium distributions inside the active material lead to an overpotential contribution at the active surface during the lithiation of NMC. This is represented by the diffusive overpotential for the active material. The total diffusive overpotential for the active material η_{D-AM} is the mean value of the local difference of the open-circuit voltage U_0 and the respective equilibrium open-circuit voltage $\bar{U}_0$ weighted with the local Butler-Volmer interface current $i_p \cdot A$. It is calculated via the formula:

$$\eta_{D-AM} = -\frac{1}{I} \sum_{all \ interface \ walls} [i_p A (U_0 - \bar{U}_0)] \quad (2)$$

where $\bar{U}_0$ is the equilibrium open-circuit voltage corresponding to the electrode's SOC.

Non-homogeneous lithium distributions in the electrolyte also lead to a diffusive overpotential η_{D-EI} . It is the sum over the local power densities, which are the scalar product of the current with the gradient of the chemical potential.

$$\eta_{D-EI} = \frac{1}{I} \sum_{all \ electrolyte \ voxels} \left[\bar{i} \frac{R T}{F c_e} \nabla c_e V (1 - t_+) \right] \quad (3)$$

where $\bar{i}$ is the local current density, R the universal gas constant, T is the temperature, F is the Faraday constant, c_e the local lithium concentration in electrolyte, and V is the voxel volume (voxel length³).

The electronic current in the active material-CBD-network causes the electronic resistive overpotential in the electrode $\eta_{\Omega-Solid}$. The ionic current in the electrolyte causes the migration overpotential in the electrolyte $\eta_{\Omega-EI}$. They are calculated by the sum of the local ohmic power losses divided by the total current via the formulas

$$\eta_{\Omega-Solid} = -\frac{1}{I} \sum_{all \ solid \ voxels} \left[\frac{i^2}{\kappa_{s,k}} V \right] \quad (4)$$

Table 3
Governing equations of the BatteryDict fully resolved model [52,56–58].

Description	Equation
Electrolyte	
Diffusion and migration	$\frac{\partial c_e}{\partial t} = \nabla \cdot (D_e \nabla c_e) - \nabla \cdot \frac{I_+ \vec{j}_e}{F}$
Charge conservation and electroneutrality	$0 = \nabla \cdot \vec{j}_e = -\nabla \cdot (\kappa_e \nabla \varphi_e) + \nabla \cdot \left(\kappa_e (1 - t_+) \frac{RT}{F} \nabla \log c_e \right)$
Active material	
Diffusion	$\frac{\partial c_s}{\partial t} = \nabla \cdot (D_s \nabla c_s)$
Ohm's law	$0 = \nabla \cdot \vec{j}_s = -\nabla \cdot (\kappa_{s,k} \nabla \varphi_{s,k})$
Interface between electrolyte and Active material	
Butler-Volmer interface current density	$i_p = 2 i_{0,\max,p} \sqrt{\left(1 - \frac{c_s}{c_{\max}}\right) \frac{c_s}{c_{\max}}} \sqrt{\frac{c_e}{c_{e,\text{equ}}}} \cdot 2 \sinh\left((\varphi_{s,p} - \varphi_e - U_0) \frac{F}{2RT}\right)$
Interface between electrolyte and Lithium reservoir	
Butler-Volmer interface current density	$i_n = i_{0,\max,n} \sqrt{\frac{c_e}{c_{e,\text{equ}}}} \cdot 2 \sinh\left((\varphi_{s,n} - \varphi_e - U_0) \frac{F}{2RT}\right)$

k = p (pos. electrode), n (neg. electrode), CC (current collector).

Table 4

Microstructural parameters obtained from 3D image data^{a,b} and gravimetric measurements.

Wet mixing duration/min	Short - 30	Long - 60
Mass loading $m_{\text{Areal}}/\text{mg cm}^{-2}$	22.46 ± 0.72	24.17 ± 0.70
Electrode density $\rho_{\text{Electrode}}/\text{g cm}^{-3}$	3.00 ± 0.19	3.24 ± 0.02
Median particle diameter $D_{50}/\mu\text{m}$	6.11 ± 0.88	5.78 ± 1.22
Porosity $\epsilon/-$	0.31 ± 0.03	0.24 ± 0.02
Active surface area $a_{\text{NMC}}/\text{m}^2 \text{g}^{-1}$	0.14 ± 0.02	0.11 ± 0.01
Pore space tortuosity factor $\tau_{\text{Pore}}^2/-$	2.73 ± 0.49	5.07 ± 1.29
CBD tortuosity $\tau_{\text{CBD}}/-$	1.36 ± 0.20	1.24 ± 0.08
Electronic conductivity $\sigma_{\text{Electrode}}/\text{S m}^{-1}$	0.41 ± 0.40	1.37 ± 1.01

^a Explanation on how parameters are obtained see Section 2.4.

^b Data averaged between three tomographies per mixing duration.

^c Obtained from gravimetric measurements of electrode and current collector foil.

and

$$\eta_{\Omega-\text{El}} = -\frac{1}{I} \sum_{\text{all electrolyte voxels}} \left[\frac{i^2}{\kappa_e} V \right] \quad (5)$$

where i is the local current density, $\kappa_{s,k}$ is the electronic conductivity of the specific component k , and κ_e is the ionic conductivity.

The simulation hardware was equipped with an Intel Xeon E5 2690 V20 with 20 cores and 256 GB RAM running Ubuntu. We used all cores for the parallelization, and the simulation took between 18 h and 80 h (depending on the C-rate) and required up to 208 GB for GeoDict [32] and the BEST solver [31] with the model derived in [56–58].

3. Results and discussion

In a first step to analyze the impact of wet mixing duration on electrode microstructure and electrode performance, we compared observations from 3D imaging to electrochemical impedance spectroscopy in symmetrical cells with different mixing durations. Fig. 1a and b show the change in measured percolation through the CBD for wet mixing durations of 30 and 60 min. We only investigate paths through the CBD here because of the large difference in electronic conductivity between NMC and CBD. While percolation through the CBD is present after the short mixing process with 30 min, most conductive paths concentrate inhomogeneously in one part of the observed volume. Doubling the wet mixing duration from 30 to 60 min vastly increases the amount and homogeneity of conductive paths through the CBD, suggesting an improvement in electronic conductivity and reduced contact resistance between the CBD and active material particles. Quantitative investigation of the CBD shows an increase in connectivity from 0.91 to 0.96 and a decrease in average path length between current collector and separator from 129 to 103 μm for 30 and 60 min of mixing time, respectively. Results from electrochemical impedance spectroscopy of

symmetrical cells are shown in Fig. 1c and confirm this observation. The high-frequency regime with a characteristic frequency of 20 kHz is typically associated with contact losses and ohmic resistance. Here, doubling the mixing time leads to a significant reduction in contact resistance and lower ohmic resistance, again explained by changes in the microstructure. 60 min of wet mixing strongly distributes and homogenizes the CBD, leading to lower tortuosity in the CBD for electrons and a higher coverage of active material particles, adversely affecting active surface area (Table 4). Additionally, the shift in ohmic resistance ($Z'' = 0$) is in line with the results from the percolation analysis and shows an improvement in electronic conductivity after 60 min of wet mixing. The second semi-circle at a characteristic frequency of 7 Hz, often associated with the charge transfer resistance between the active material and the liquid electrolyte, is significantly smaller for electrodes mixed for only 30 min compared to 60 min. This finding is consistent with the analysis of the microstructure, which showed that doubling the mixing time reduced the active surface area, leading to higher charge transfer resistance. The decrease in active surface area can be explained by looking at the distribution of the carbon-binder-domain inside the porous structure. Longer mixing times facilitate the attachment of CBD to active material particles, increasing carbon coverage of the particle surface and limiting lithium-ion access to the active material. 3D image data obtained from FIB-SEM tomography allows a quantification of surface coverage. Here, surface coverage is defined as the relative amount of NMC surface voxels covered by CBD voxels. The mean surface area coverage across three distinct tomographic datasets increases from 21.6% to 29.7% when doubling the mixing time from 30 to 60 min. And while we did not directly investigate the impact of different CBD distributions on wettability, this increase in surface coverage will reduce accessibility of the electrolyte to the NMC surface, impacting the charge transfer reaction. Additionally, the increase in electrode density also leads to a reduction in overall surface area. We suggest that doubling the wet mixing duration from 30 to 60 min causes better attachment of carbon black and graphite on the surface of NMC particles, which leads to better calenderability and therefore higher achievable gravimetric density. This positive effect of graphite flake attachment to NMC particles was previously confirmed by Tran et al [11]. Both electrodes were calendered with identical settings, which means the same machine, unheated rolls at ambient temperature, the same roll speed, and the same gap setting and resulting line load. The higher density after 60 min is therefore not due to a different calendering treatment but to a different response of the coating to the same mechanical load: the more evenly distributed graphite flakes act as a solid lubricant and facilitate particle rearrangement during compaction [11]. Improved calenderability is thus part of the mixing effect rather than an independent process variable. Consequently, mixing duration and final porosity cannot be varied fully independently at a fixed calender setting, and the values in Table 4 represent the combined

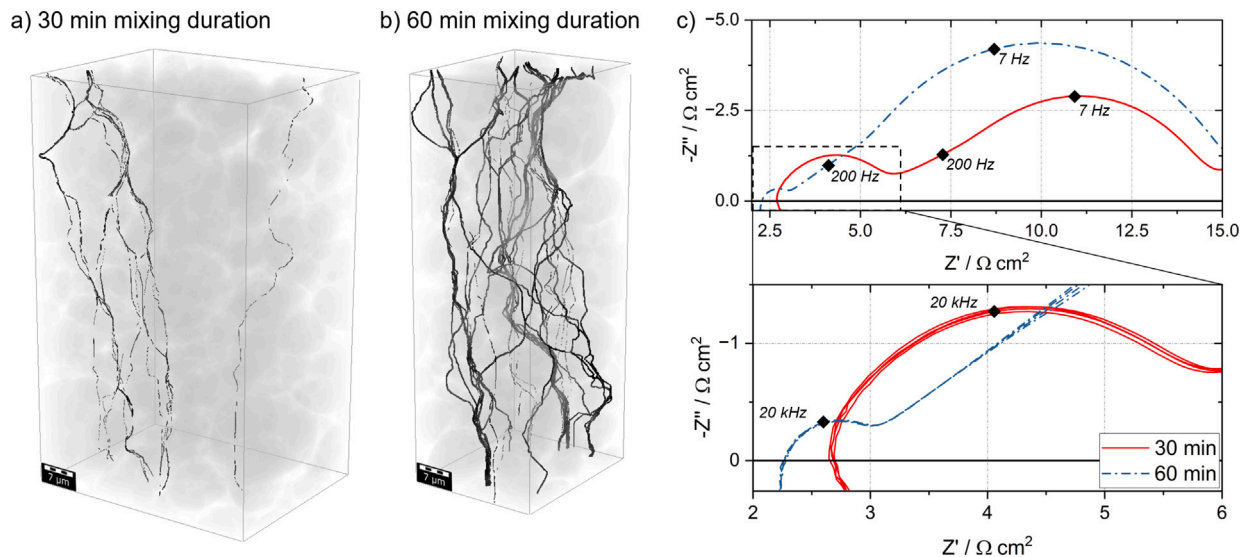


Fig. 1. Experimental observations on electrodes with different wet mixing durations. Percolation analysis of the carbon-binder domain for electrodes with 30 min (a) and 60 min (b) mixing duration. Full percolation paths through the entire electrodes are shown in black; NMC particles are shown as transparent gray. Nyquist plot of full electrochemical impedance spectra for electrodes with 30 min (dashed) and 60 min (solid) mixing duration at ~ 3.8 V (50% SoC) (c). High-frequency domain zoomed in and measured four times.

structural signature of longer mixing. Separating both contributions would require calendering to a matched porosity, which we suggest for future work. In our study, we attempt this by virtually altering one of the structures. Results of this porosity matching in an attempt to disentangle the two mechanisms are provided in the SI and addressed again later in this work. The increase in electrode density was confirmed with gravimetric analysis (Table 4). Additionally, we demonstrate the monotonicity of our findings by adding results from electrodes with a mixing time of only 45 min in the SI. Microstructural parameters obtained from 3D image data change monotonically when increasing the mixing time, supporting our claims made on the effect of mixing time on microstructural changes. We also include simulation results, showing how overpotentials behave with increased mixing time. The total simulated overpotential for the electrode with 45 min mixing time places roughly in the middle between the 30 and 60 min structures. The results are provided in the SI (Figure S10).

To understand the impact of microstructural changes on performance for the two mixing times, we employ rated discharge experiments on electrodes. The resulting discharge curves are shown in Fig. 2a. While the differences in overall capacities are small, a clear trend emerges where electrodes mixed for 60 min show slightly higher capacities at lower discharge currents. This can be attributed to the higher electrode density, which we already discussed in detail. Above 2C, electrodes mixed for only 30 min show higher overall capacities, suggesting smaller loss processes and a microstructure more suitable for higher currents. The same trend can be seen when the electrochemical data is plotted in a Ragone diagram, shown in Fig. 2b. Starting between 1C and 2C, the shorter mixing duration leads to higher power density. In a prior work, we showed the importance of percolation in CBD networks and its influence on electrode performance [7]. When coupling the results of rated discharge experiments to the observations on improved percolation discussed earlier, it appears that improving the percolation of the CBD does not translate to higher power density at discharge rates above 1C. To understand the origin of the reduced power density, we need to deconvolute the different loss contributions using structurally resolved discharge simulations based on the tomography data from electrodes prepared with different mixing times.

Fig. 2c shows simulated cell voltage as a function of discharged capacity for different discharge rates and the two cells. A similar trend emerges compared to the experimental results, with the electrodes mixed for only 30 min showing lower overpotentials at discharge rates

of 2C or higher. The Ragone plot (Fig. 2d) again shows that higher power densities are achieved above 2C for electrodes mixed for 30 min, suggesting that the simulation captures the real electrode behavior quite well. A direct comparison between simulated and experimentally obtained discharge curves is shown in Figure S7. Minor deviations between experimental and simulated results are visible. They are expected because the exchange current density, electronic conductivity, and ionic diffusivity of the active material (NMC622) were parametrized using the 60 min dataset. We want to emphasize again that none of the parameters are changed between simulating different mixing durations, and only the underlying FIB-SEM-based structure was changed between the simulations. The similar trends observed in the experimental fully resolved simulated discharge curves support the conclusion that the simulation provides representative insight into physical processes within the cell that cannot be observed directly in experiments.

The biggest advantage of utilizing simulation results in our discussion is the possibility to extract the overpotential curves of specific loss processes individually. Fig. 3 shows all calculated overpotentials for the positive side of the half cell for a discharge current density of 37.72 A m^{-2} (1C). Similar to the results of the electrochemical impedance spectroscopy, losses attributed to the effective electronic conductivity (Fig. 3a) are lower for electrodes produced with longer mixing times. However, the improvement is only marginal, with a maximum difference of only 3.4 mV, and does not significantly influence the overall overpotential shown in Fig. 3d. This can be explained by previous observations that improving the effective conductivity past the point of CBD percolation does not significantly improve discharge performance, even though effective electronic conductivity increases.

The detrimental effect of increasing wet mixing duration from 30 to 60 min can be seen by looking at the overpotentials caused by lithium diffusion and migration in the electrolyte shown in Fig. 3b. Because 60 min mixing times lead to a higher tortuosity of the pore space (see Table 4), overpotentials rise accordingly, causing a significant loss in power density at higher rates. Ultimately, this leads to a steeper gradient in lithium concentration in the electrolyte and less lithium close to the positive current collector for the electrode with longer mixing time (see Fig. 5c). Similarly, kinetic losses (Fig. 3c) increase with longer mixing times due to a reduction in active surface area (see Table 4), explained by increased CBD coverage of the NMC particles. This observation is again in good agreement with EIS results, which

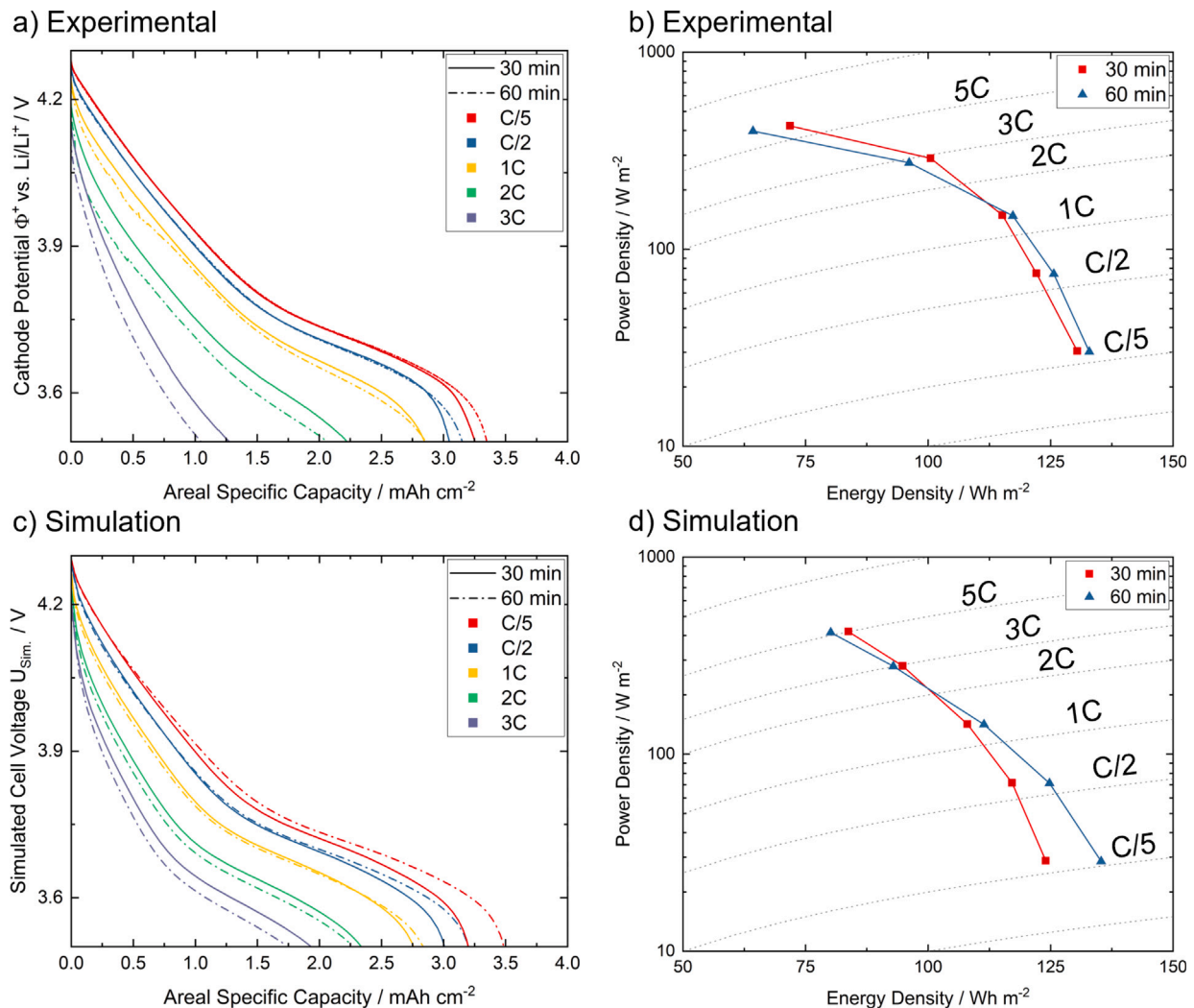


Fig. 2. Comparison between experimental and simulated results for rated discharge capacities, energy, and power density. Experimentally obtained rated positive electrode potentials vs. Li/Li^+ for mixing durations of 30 min and 60 min (a). Experimentally obtained Ragone diagram for electrodes with mixing times of 30 min and 60 min (b). Simulated cell voltage for both mixing durations (c) and simulated Ragone diagram. Dashed lines in the Ragone diagrams indicate the theoretical C-rate performance.

showed a drastic increase in charge transfer resistance after longer mixing times.

Unexpectedly, although the same NMC powder was used to manufacture all electrodes, solid diffusion losses increased significantly with longer mixing times. We propose that the same two effects responsible for the higher kinetic losses also increase losses caused by solid diffusion: Increased particle-to-particle contacts and the reduced surface area for initial lithium intercalation force the same amount of lithium atoms through a smaller active material volume, thereby increasing average diffusion lengths and, consequently, the overpotential. Median particle diameters even slightly decrease from $6.11 \mu\text{m}$ to $5.78 \mu\text{m}$ after doubling the mixing time, as shown in Table 4. The increase in overpotential caused by solid diffusion in the active material is therefore due to higher CBD coverage and more particle–particle contacts, and not an increase in particle diameter. To support our hypothesis, we evaluated the average diffusion length within NMC particles for electrodes with different mixing times and found that it increased by 21% from $1.12 \mu\text{m}$ to $1.36 \mu\text{m}$ when the mixing time was doubled. Because the particle sizes did not increase between 30 and 60 min of mixing, this increase is attributed to the greater CBD coverage of the active material surface. A schematic illustration of how average diffusion lengths are calculated, as well as detailed results, are provided in the SI (Fig. S1). When looking at all overpotentials combined (Fig. 4d), the difference between

30 and 60 min of mixing time becomes obvious, with the shorter mixing time showing smaller losses across the entire discharge process at 1C. In summary, the reduced active surface area caused by doubling the mixing time leads to higher overpotentials.

Fig. 4 visualizes the effect of CBD coverage and particle-to-particle contacts on transport processes and surface kinetics for a small set of particles. For visual clarity, this is demonstrated on 2D cutouts; however, observations are valid in three dimensions. Fig. 4a shows the selected area, containing active material, CBD partly covering active material particles, and pore space that would be filled with electrolyte. As expected, where NMC particles are in contact with CBD, electronic current density (Fig. 4b) is higher because the electronic conductivity of the CBD is multiple orders of magnitude higher than that of the NMC. Conversely, areas with higher amounts of CBD hinder lithium-ion transport through the electrolyte, limiting the number of lithium ions reaching the NMC surface and thereby increasing both the overpotential caused by diffusion and migration (Fig. 4c) and surface kinetics (Fig. 4f). The kinetic overpotential is higher in areas where more CBD is attached to the NMC surface because of the lower local availability of lithium ions. As mentioned previously, solid diffusion is also inhibited by covered surfaces, visualized here as a lithium flux density (Fig. 4e). Whenever access to a surface is partly (CBD) or completely (particle-to-particle contact) blocked, lithium flux density locally decreases, forcing

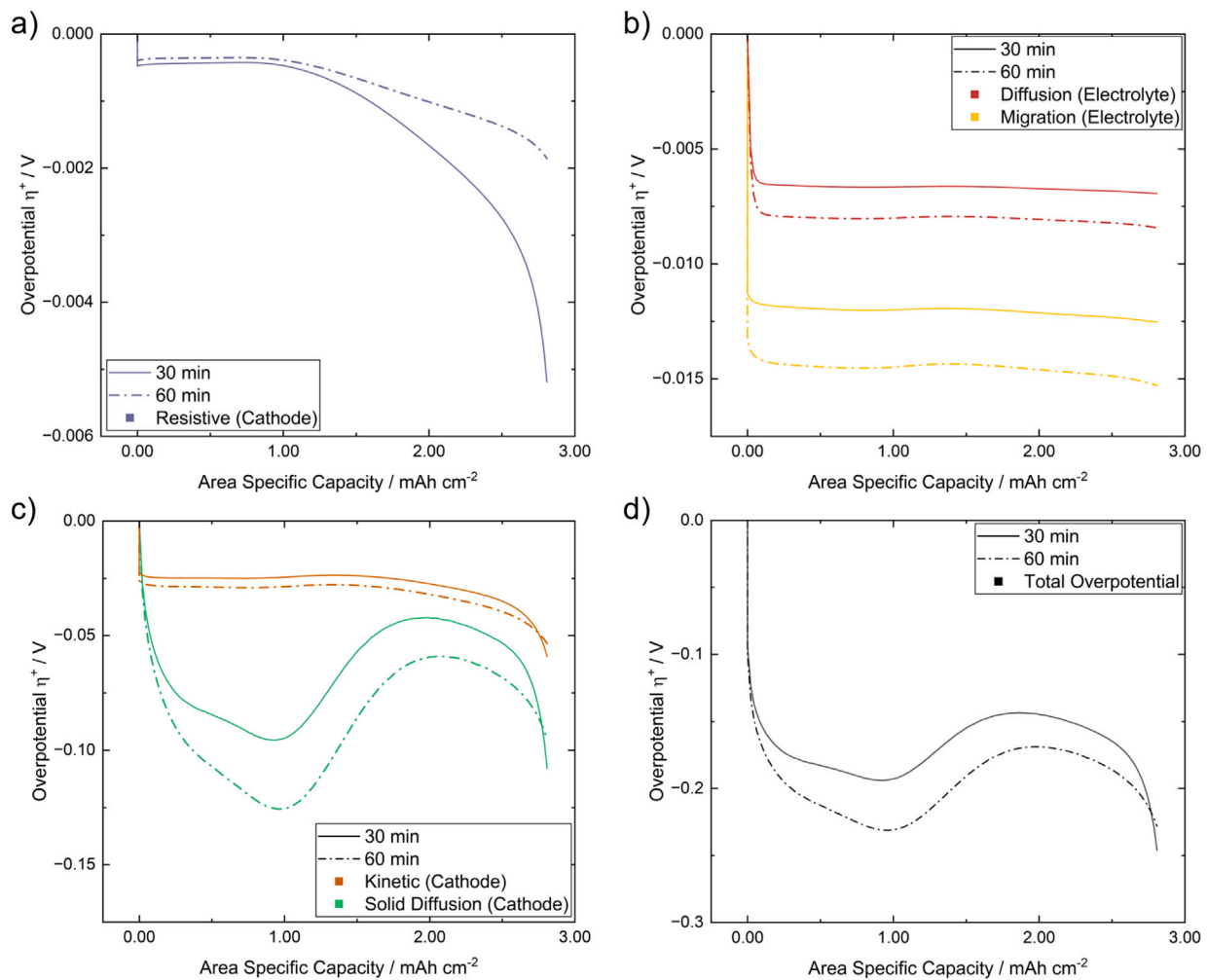


Fig. 3. Predicted positive electrode overpotential contributions simulated for a discharge current density of 37.72 A m^{-2} (1C) in electrode structures with mixing times of 30 min and 60 min. Overpotential caused by ohmic resistance (a), electrolyte diffusion and migration (b), kinetic overpotential at the active NMC622 surface and solid diffusion inside the active material (c), and total overpotential (d).

lithium atoms to follow longer diffusion paths and thereby increasing the overpotential. Because both surface coverage and particle contacts increase with longer mixing times, we conclude that at the particle level, longer mixing times are detrimental to high current density discharge performance. Fig. 4d shows the solid lithium concentration in the 2D cutout after a full 3C discharge. Loss of available surface area and lithium-ion depletion in the electrolyte cause transport and kinetic losses, resulting in strong lithiation gradients.

To understand the importance of possible gradients in the electrodes on performance losses, we now outline how increasing wet mixing time from 30 to 60 min influences lithium-ion transport in the electrolyte through the entire electrode structure. As reported in our virtual electrode study [7], continuously lowering CBD tortuosity by either adding more CBD or redistributing existing CBD will lead to an increase in ionic pore space/electrolyte tortuosity. Fig. 5a shows the impact of mixing time on the spatial distribution of lithium ions in the electrolyte between separator and current collector for a 3C discharge simulation at $\sim 3.5 \text{ V}$ (identical conditions as in Fig. 4). A steeper gradient and stronger depletion of lithium ions close to the current collector for the electrode mixed for 60 min can be explained by an increase in pore constrictivity caused by CBD blocking potential lithium-ion pathways. Median pore throat diameters decrease from $1.02 \mu\text{m}$ to $0.87 \mu\text{m}$ when doubling the mixing time, and the full pore throat diameter distributions are provided in the SI S11. Fig. 5b and c show the corresponding 3D distribution of lithium-ion concentration. Again, a clear difference

is visible for the differently mixed electrodes, with stronger depletion of lithium ions in the electrode mixed for 60 min. If we combine this observation with the previously discussed effect on local lithium depletion, we can conclude that the redistribution of CBD and the attachment of more CBD on active material causes a distinct increase in transport and kinetic loss processes on both the particle and the electrode scale.

Since we observed a more homogeneously distributed CBD with lower tortuosity after longer mixing times, the question arises whether this has a positive effect on electron transport and therefore on performance. We take a look at the electronic current density on an electrode scale in Fig. 6. No clear improvement in homogeneity or distribution of the electronic current density can be observed, both in the active material (Fig. 6a&b) and the CBD (Fig. 6c&d). Moreover, even though the CBD tortuosity is lowered from 1.53 to 1.27 and the effective electronic conductivity of the electrode is almost doubled from 2.66 to 5.00 S m^{-1} by increasing the mixing time from 30 to 60 min, this improvement does not translate to overall lower overpotentials. In discharge simulations performed at 3C, the resistive cathode overpotential was only 3.4 mV lower after doubling the mixing time from 30 to 60 min. This apparent improvement only translates to 1% of the total simulated overpotential. We therefore conclude that an improvement to the electronic conductive network past the point of initial percolation plays no significant role in determining electrode performance. This is presumably the case because the electrode recipe contains enough

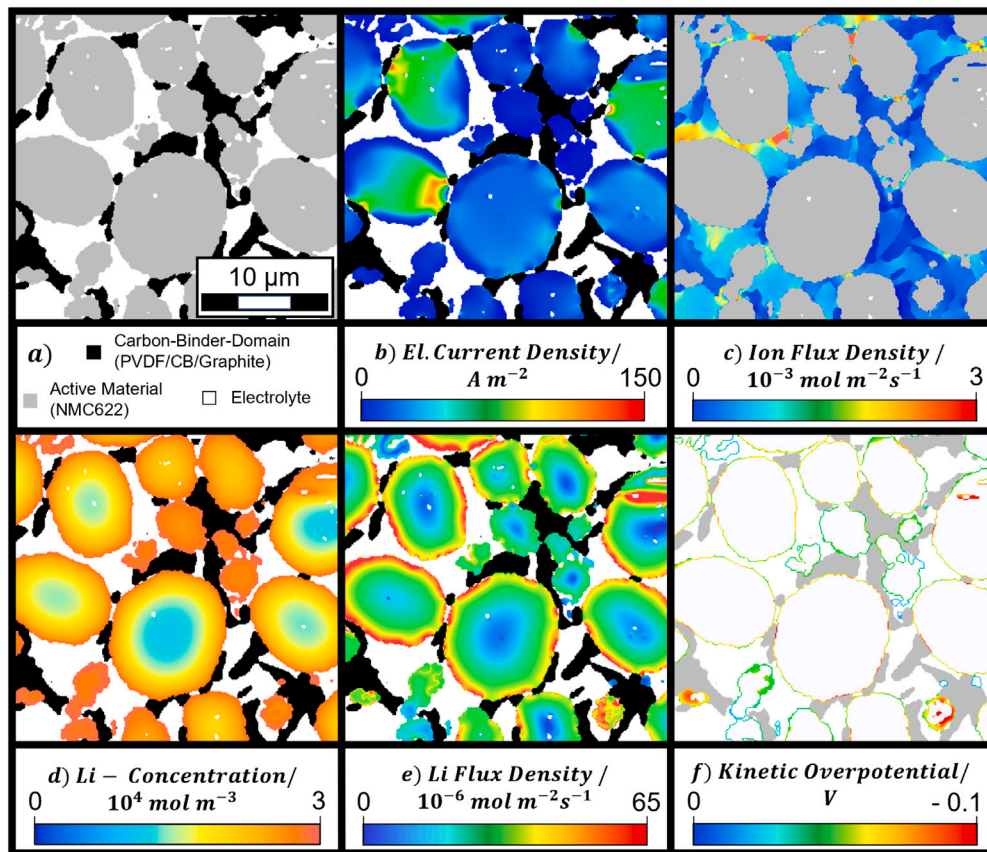


Fig. 4. Impact of CBD coverage and distribution as well as particle-to-particle contacts on simulation results at the particle level. 2D profiles obtained from 3C discharge simulation on the electrode with 60 min mixing duration at ~ 3.5 V. Segmented FIB-SEM data (a); electronic current density in the NMC (b); Lithium-ion flux in the electrolyte and homogenized porous CBD (c); Lithium concentration in the NMC (d); Lithium flux density in the CBD during lithiation (e); local kinetic overpotential at the NMC surface (f).

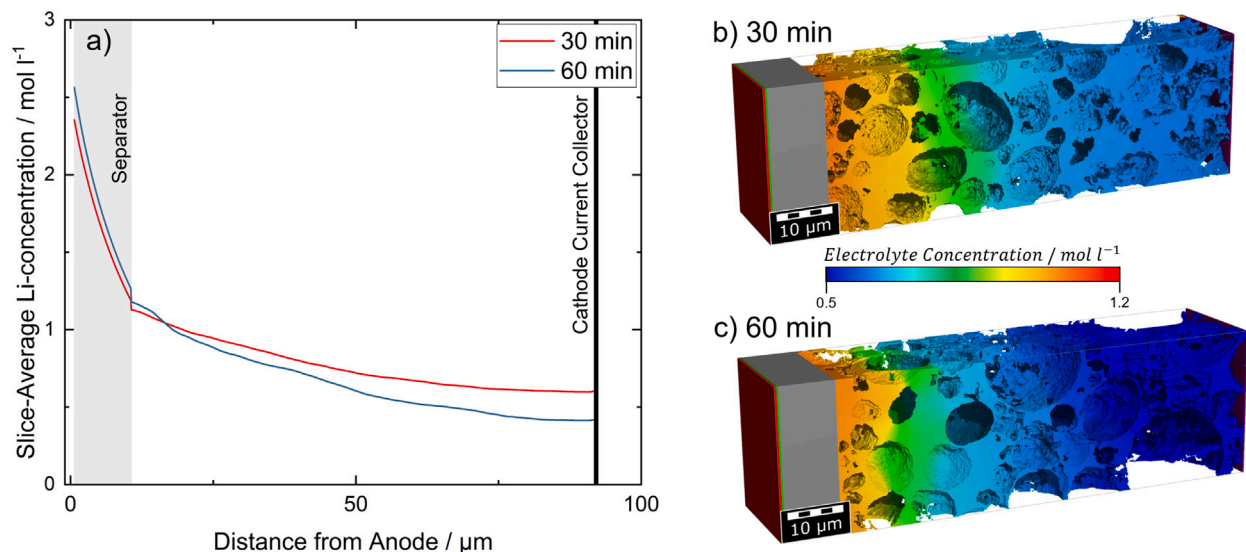


Fig. 5. Visualization of effects of mixing time variation on concentration distribution in the electrolyte. Simulated slice-averaged lithium concentration in the electrolyte during 113.14 A m^{-2} (3C) discharge at ~ 3.5 V (a) and corresponding 3D concentration profiles for electrodes with 30 min (b) and 60 min (c) mixing time.

additives that percolation is achieved even at shorter mixing durations, as shown in Fig. 1. A combination of longer mixing and less inactive materials (carbon black and/or conductive graphite) might lead to enhanced performance.

To find an optimized electrode recipe and provide practical advice, we use our findings to calculate microstructural indicators that should be targeted and then obtain the resulting changes to the recipe. To achieve this, we first determine the percolation threshold for both mix-

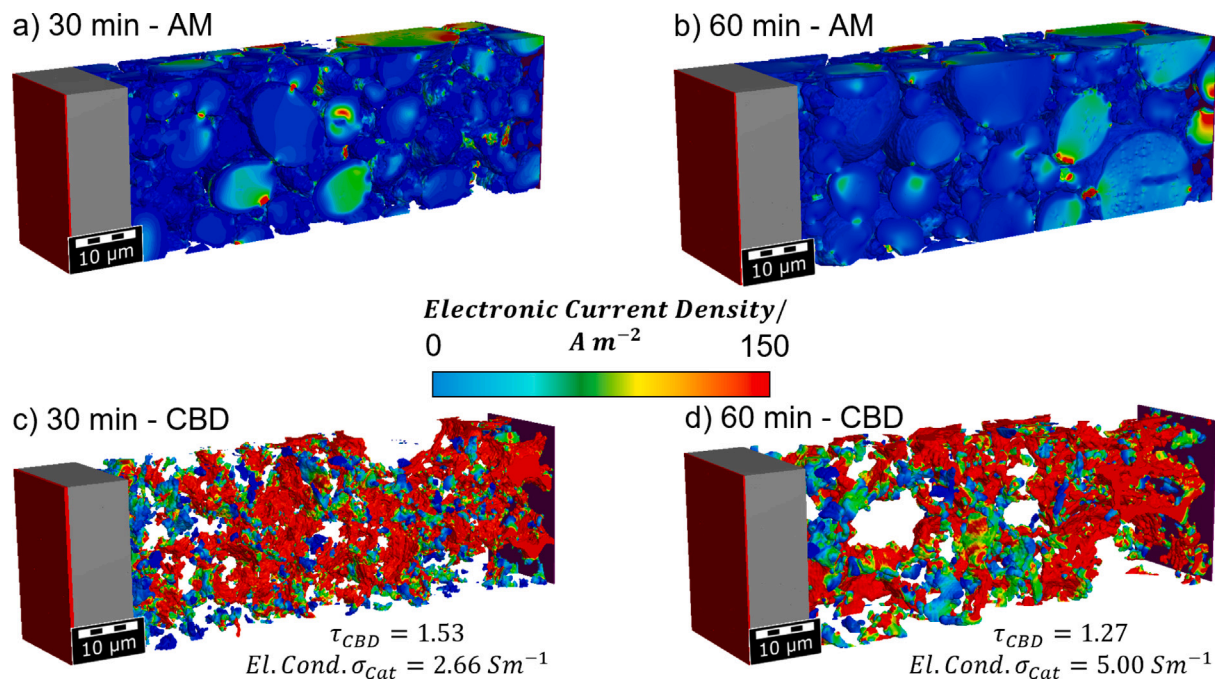


Fig. 6. 3D profiles of simulated electronic current density inside the active material (a&b) and the CBD (c&d) during $113.14 A m^{-2}$ (3C) discharge at $\sim 3.5 V$ for a electrode with 30 min (a&c) and 60 min (b&d) mixing time. It is clearly visible that for both 30 and 60 min of mixing time, almost all of the electronic current is conducted in the CBD and not in the active material.

ing times. Results of this calculation are provided in the SI (S12). We then use the electrode just above the percolation threshold, mixed for 60 min, and calculate our target indicators, which are also provided in the SI (SI Table 4). Overall, we found that (virtually) increasing active material weight percentage from 95 to 97% still maintains percolation but significantly lowers pore space tortuosity (-32%) and increases available surface area ($+17\%$). As a guideline for this processing route, we determine a CBD connectivity Index of 0.86 and 5 vol.-% CBD to be sufficient to achieve percolation. Our investigation revealed how CBD rearrangement and an increased densification occur when doubling the wet mixing time from 30 to 60 min. In order to disentangle these two processes and understand which of the two effects contributes to the observed increase in overpotential during discharge, we performed additional simulations on a virtually compressed microstructure with matched porosity. Starting from our 30 min mixing time simulation domain, we compressed the microstructure until we matched the porosity of the 60 min electrode. After simulating discharge at 1C, we compared the results to the two original microstructures to assess how increased densification and CBD rearrangement interact. Results of the comparison are provided in the supporting information of this study (S13). We can see that overpotential slightly increases after the virtual compression. However, the total overpotential is still considerably lower compared to the electrode mixed for 60 min. Quantitatively speaking, the simulated maximum overpotential during 1C discharge increases from $-0.194 V$ to $-0.231 V$ when doubling the mixing time from 30 to 60 min. After virtual compression, maximum overpotential reaches $-0.198 V$. We are therefore confident that our reported mixing-duration effect is mostly the result of CBD rearrangement, while the increased densification only plays a minor role at the investigated discharge rates.

4. Conclusion

Understanding how electrode processing impacts the resulting properties and performance of the cells is critical for establishing an efficient battery manufacturing value chain. In this work, we investigated the complex interactions occurring during wet mixing, sometimes referred

to as the dispersion process. To this end, we combined high-resolution FIB-SEM tomography of positive electrodes manufactured with different wet mixing times, electrochemical testing, and structurally resolved simulations with GeoDict to understand the relationship between wet mixing duration, electrode microstructure, and electrochemical performance. Our analysis shows that increasing the mixing duration improves the conductive network through homogenization of the carbon-binder-domain, resulting in lower contact resistance and higher electronic conductivity. However, this more homogeneous distribution of CBD also reduces the active surface area by increasing CBD coverage of active material, resulting in higher kinetic losses and charge transfer resistance. For the first time, this mixing-induced CBD rearrangement could also be coupled to increased diffusive losses inside the NMC particles. By increasing the amount of NMC surface area covered in CBD, mean solid diffusion paths are 21% longer after doubling the mixing time, leading to increased diffusive losses. Additionally, the direct correlation between tortuosity of the CBD and ionic transport in the electrolyte was confirmed. Although a more connected and homogeneous CBD improves electronic transport, it also impedes ionic transport by reducing the available pore space and transport pathways, leading to local electrolyte depletion at the particle level and near the current collector. While longer mixing durations improve the conductive network, extending the mixing duration beyond the point of percolation does not appear to provide additional benefits for electrode performance. The findings presented in this work suggest that the mixing procedure should be optimized in conjunction with the recipe formulation. Specifically, the amount of inactive additives like carbon black or graphite may be reduced by optimizing the interplay between CBD content and mixing conditions. By using virtual CBD adjustments inside the longer mixed electrode structure, we found that active material weight percentages could be increased from 95 to 97% without sacrificing CBD percolation when doubling the mixing time from 30 to 60 min. By removing excess inactive material, pore space tortuosity could be lowered by 32%, and electrolyte accessible surface area was increased by 17%. Future studies should aim to validate whether CBD percolation can be achieved with substantially lower amounts of inactive material through extended mixing while maintaining favorable surface kinetics, solid diffusion, and ionic transport in the electrolyte.

CRedit authorship contribution statement

Adrian Lindner: Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Roman Buchheit:** Writing – review & editing, Software, Methodology, Investigation. **Maximilian Lechner:** Methodology, Investigation. **Fabian Biebl:** Writing – review & editing, Software, Methodology, Investigation. **Wolfgang Menesklou:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Ulrike Krewer:** Writing – review & editing, Supervision.

Declaration of Generative AI Usage

Image Generation based on Google Gemini Flash 3.6 was used to enhance parts of the graphical abstract. The resulting schematic was checked carefully for accuracy and originality.

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 material related to this article can be found online at <https://doi.org/10.1016/j.jpowsour.2026.241651>.

Data availability

Research data is available via the KITopen repository: [10.35097/kg2m8j742cc65kyr](https://nbn-resolving.org/urn:nbn:de:hebis:30-1035097-kq2m8j742cc65kyr).

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