

Pore-scale modeling of capillary-driven binder migration during battery electrode drying

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Sodium-ion batteries employing hard carbon electrodes are considered a drop-in technology for lithium-ion batteries. Electrode drying is a critical manufacturing step, as binder migration during pore emptying impacts the mechanical integrity and electrical performance of the electrode. Existing modeling approaches predominantly rely on the film shrinkage phase in a one dimensional way or neglect the capillary transport, resulting in a lack of physically consistent microstructure resolved predictions of binder migration. In this work, a spatially resolved pore scale continuum model is extended to explicitly describe capillary-driven binder transport during pore emptying. The model is applied to hard carbon microstructures with varying mean particle diameters. The simulations reveal that smaller particle sizes lead to a more homogeneous binder distribution, whereas higher evaporation rates and increased surface tension promote stronger binder gradients. Variations in solvent viscosity show only a minor influence on binder migration, as long as no hydrophilic or hydrophobic behavior is present. Finally, the simulations demonstrate that an explicit description of capillary transport and microstructural effects is essential for accurately predicting binder migration and provides a basis for the targeted optimization of electrode drying processes.

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I. INTRODUCTION

The growing demand for high-energy, cost-efficient, and sustainable energy storage systems has intensified research on post-lithium batteries, such as sodium-ion batteries. To fully exploit their electrochemical potential, the manufacturing process of battery electrodes needs to be carefully reviewed. This process involves several steps, including mixing, coating, drying, calendaring, and cutting. Among these, the drying step is particularly critical, as even small variations can significantly alter the resulting microstructure and, consequently, overall cell performance. During this step, the wet coating layer represents a complex multicomponent system. It consists of a solid phase, composed of the active material particles and the conductive carbon black aggregates, and a liquid phase. Crucially, the polymeric binder system does not exist as macroscale solid particles at this stage. Instead, depending on the specific formulation, its constituents are either dissolved within the liquid solvent (such as Carboxymethylcellulose, CMC) or present as a stable colloidal dispersion of submicron polymeric particles

(such as Styrene-Butadiene Rubber, SBR). The drying induced structural evolution directly influences critical electrode properties such as porosity, adhesion to the current collector substrate, and interparticle cohesion, heavily dictated by the phenomenon of binder migration [1]. Specifically, binder accumulation at the electrode surface reduces adhesion to the current collector [2]. Conversely, local binder depletion can compromise particle cohesion. Thus, drying remains a key step in achieving process reliability, as it defines process-structure relations essential for mitigating binder migration.

Physical understanding of solvent drying and binder migration. In the recent decade, there has been a shift in the understanding of the physical processes underlying the drying process and the binder migration. Preliminary studies [3] hypothesized the formation of a dense consolidation layer, at which the active material and the binder accumulate. Consequently, the idea of a top-down consolidation was proposed, suggesting that particle rearrangement and sedimentation dominate the drying behavior. Moreover, the aforementioned hypothesis posits that the binder migration was attributable to convective motion of the solvent, counteracted by molecular diffusion. However, *ex situ* imaging has demonstrated that particle distributions remain homogeneous during the drying process and that no consolidation front develops [4]. Therefore, the fundamental physical mechanism underlying binder migration is capillary-driven transport, which aligns well with experimental works on the optimization of the drying process by multistage drying profiles [5,6]. The binder moves

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in response to capillary pressure gradients between pores of different sizes. Hence, it is evident that binder migration is directly coupled to the capillary solvent flow after reaching a consolidated enough structure [5–7].

Modeling approaches for electrode drying. In addition to experimental approaches, modeling and simulation are key tools for understanding and optimizing different processes as well as determining process-structure relations. In the context of battery electrode drying, the distinct approaches can be categorized into homogenized continuum models (HCM), coarse-grained molecular dynamics (CGMD), discrete element methods (DEM), and spatially resolved continuum models (RCM). HCM models [8,9] reduce the drying process to a set of one-dimensional partial differential equations. Thereby, these approaches establish a connection between macroscopic drying kinetics and process parameters. However, a limitation of such approaches is that they neglect several physical processes, including pore-scale effects, such as capillary flow and wetting behavior. Extensions of these approaches are made in [10–12] by augmenting the governing equations with an additional convection-diffusion equation modeling binder transport. These extended HCMs demonstrate a more pronounced binder gradient, under faster evaporation conditions. Coarse-grained molecular dynamics (CGMD) models describe the binder and carbon black as a single phase, which is termed as CBD phase. The process of solvent removal is modeled by the shrinkage of these phases, while the motion of the solvent is implicitly governed by the phenomenon of Brownian motion. With these CGMD models, the microstructure and the binder distribution are captured [13–15], while pore-emptying physics cannot be modeled. Discrete element method (DEM) simulations focus on the initial drying stage and film shrinkage [16]. There are also works that simulate the subsequent calendaring step using DEM [17]. Furthermore, some studies investigate the final binder distribution employing simplified HCM models [11]. The group of spatially resolved continuum models capture the movement of the fluid phase explicitly by tracking the liquid-gas interface during drying. Examples of this include the volume-of-fluid (VOF) model by Wolf *et al.* [18] and the phase-field approach by Weichel *et al.* [19]. While the first study is restricted to spherical particles with no explicit representation of the surface wetting behavior, the latter is able to incorporate these physical mechanisms. A further class of approaches specifically modeling the pore-emptying process can be grouped into pore network models (PNM) and continuum models (CM). Continuum models describe the pore-emptying process at the macroscopic scale, where volume averaged quantities are employed to represent the underlying pore-scale behavior [20]. In contrast, pore network models rely on statistical representations of the pore network to investigate the influence of different parameters, such as viscosity [21–23].

Modeling approaches for binder migration during drying. Most approaches to model binder migration are based on an early understanding of the drying process. These approaches use a one-dimensional convection-diffusion equation to describe binder transport mostly during film shrinkage, while neglecting all capillary effects during pore emptying [10–12]. In these approaches, film shrinkage drives binder

accumulation at the surface, a process that is partially compensated by diffusion, which transports binder toward the substrate. These effects give rise to a dependence on the macroscopic Péclet number, which correlates with the intensity of binder migration. However, it is important to note that the macroscopic Péclet number does not reflect local capillary velocities, thereby failing to fully capture micromechanistic insights within the porous network. Furthermore, the approach by Lombardo *et al.* [24] which is based on a CGMD model, enables the calculation of the final binder distribution. This model implicitly accounts for capillary forces by simulating interparticle forces of the CBD particles. Although existing modeling frameworks yield valuable insights into drying kinetics and qualitative binder behavior, they generally fail to consistently capture capillary transport at the pore scale, which is the main mechanism for binder migration in porous electrodes. Therefore, the present work aims to investigate drying dynamics and binder migration in wet film coated battery electrodes, using hard carbon (HC) as an example, by means of an extended modeling approach that explicitly resolves the underlying transport mechanisms.

Contribution of the present work. Building on the existing pore emptying model of [19], which directly resolves liquid-air interfaces, capillary forces, and wetting behavior within realistic electrode geometries, the present work introduces an interesting coupled framework. While the previous model is restricted to pure multiphase fluid dynamics with rigid body interaction, this extension captures the full physics of binder migration, irreversible deposition, and global mass conservation by introducing an additional set of partial differential equations.

This extension enables a deeper physical understanding of the governing transport phenomena and supports the identification of improved electrode manufacturing strategies based on deeper insights into the influence of process parameters by the simulation. The analysis links drying conditions, microstructural characteristics, pore-emptying dynamics, and the resulting binder distributions. Therefore, we present a spatially resolved continuum framework, that consistently captures capillary driven binder migration. Based on the previous work [19], the extended system of governing equations is presented in Sec. II, which first summarizes the underlying framework and subsequently introduces the novel binder transport formulation. The model is implemented in the multiphysics simulation framework PACE3D [25], enabling parallelization via the message passing interface (MPI) and allowing physics-based modeling of microstructural evolution driven by multiple physical mechanisms. Section III validates the enhanced model using two representative test cases. Section IV applies the model to experimental hard carbon microstructures to investigate the impact of various process parameters on the final binder distribution. Finally, Sec. V concludes the paper and outlines directions for future research.

II. MATHEMATICAL FORMULATION

To model the drying process in porous media such as battery electrodes, we consider a multiphase problem consisting of two fluids and multiple solid phases. The fluid

region is denoted by Ω_f and the solid region by Ω_s , such that the full computational domain is $\Omega = \Omega_f \cup \Omega_s$. Furthermore, the fluid domain consists of the solvent and air domain as $\Omega_f = \Omega_{\text{air}} \cup \Omega_{\text{solvent}}$. The evaporation model of Weichel *et al.* [19] is employed, which is briefly summarized in Sec. II A. A detailed overview of the underlying drying framework may be found in previous publications [19,26]. To describe binder migration, the governing system of partial differential equations is extended by an additional convection-diffusion equation as well as a deposition model in Sec. II B.

A. Evaporation model

Phase evolution of a two-phase system. The electrode geometry is represented using a phase-field method, where the interfaces between the phases are modeled by a smooth transition layer. For a two-phase fluid system, the temporal evolution of the phase-field variable $\tilde{\varphi}(\mathbf{x}, t)$, representing the local volume fraction of one of the two phases at the spatial position $\mathbf{x} \in \Omega_f$ and time t , can be described by an Allen-Cahn formulation of the form

$$\dot{\tilde{\varphi}} = M(\tilde{\beta} \partial_{\tilde{\varphi}} \psi - \tilde{\alpha} \nabla^2 \tilde{\varphi}), \quad \mathbf{x} \in \Omega_f, \quad (1)$$

where the boundary condition

$$\tilde{\alpha} \nabla \tilde{\varphi} \cdot \mathbf{n}^{\text{fs}} = (\sigma_{2s} - \sigma_{1s}) \partial_{\tilde{\varphi}} h^{\text{ff}}(\tilde{\varphi}), \quad \mathbf{x} \in \partial\Omega_f \quad (2)$$

describes the surface wetting based on solid-fluid surface energies σ_{1s} and σ_{2s} according to Young's law, cf., e.g., [27]. Here, $\dot{\tilde{\varphi}}$ denotes the material derivative, M the mobility, and $\nabla^2 := \nabla \cdot \nabla$ the Laplacian operator. The coefficients $\tilde{\alpha}$ and $\tilde{\beta}$ are defined as $\tilde{\alpha} = \sigma \epsilon$ and $\tilde{\beta} = 18 \frac{\sigma}{\epsilon}$, where σ denotes the surface energy density between the two fluids and ϵ controls the thickness of the diffuse interface. The free-energy density $\psi(\tilde{\varphi})$ is given by the double-well potential $\psi(\tilde{\varphi}) = \tilde{\varphi}^2(1 - \tilde{\varphi})^2$. Since the phase fields satisfy the sum constraint, the second phase is implicitly defined by $1 - \tilde{\varphi}$, and only one evolution equation needs to be solved. In addition, $\mathbf{n}^{\text{fs}}$ denotes the outward-pointing unit normal vector on the boundary of Ω_f , while the interpolation function $h^{\text{ff}}(\tilde{\varphi}) = \tilde{\varphi}^2(3 - 2\tilde{\varphi})$ is used. Following the modifications proposed by Weichel *et al.* [19], the evolution equation is extended to

$$\dot{\tilde{\varphi}} = M \left[\frac{36\sigma}{\epsilon} (2\tilde{\varphi}^3 - 3\tilde{\varphi}^2 + \tilde{\varphi}) - \sigma \epsilon (\nabla^2 \tilde{\varphi} - \|\nabla \tilde{\varphi}\| \nabla \cdot \mathbf{n}) \right] + v^e \|\nabla \tilde{\varphi}\|, \quad \mathbf{x} \in \Omega_f. \quad (3)$$

In this modification, the curvature minimizing dynamics of the Allen-Cahn equation are counteracted through the term $\|\nabla \tilde{\varphi}\| \nabla \cdot \mathbf{n}$, thereby preventing an overestimation of surface tension effects, since these are already considered by a capillary term in the Navier-Stokes equations. In addition, solvent evaporation is represented by the term $v^e \|\nabla \tilde{\varphi}\|$, where the interface velocity v^e is related to experimentally determined evaporation rates through the factor κ (unit s^{-1}) according to Eq. (34). Thereby, v^e denotes the uniform local evaporation velocity applied at the phase boundary. Although the evaporation flux itself is spatially uniform at the interface, the velocity of individual menisci varies dynamically throughout the porous network, as the movement of the fluid is coupled to the locally resolved velocity field $\mathbf{u}$, which accounts for

capillary pressure gradients and local fluid redistribution between different pore sizes.

Fluid motion. To describe the motion of the two fluid phases, the incompressible Navier-Stokes equations of the form

$$\rho \frac{\partial \mathbf{u}}{\partial t} + \rho \nabla \mathbf{u} \cdot \mathbf{u} = -\nabla p + \nabla \cdot [\mu(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)] + \mathbf{K} + \rho \mathbf{f}_V, \quad (4a)$$

$$\nabla \cdot \mathbf{u} = 0 \quad (4b)$$

are employed within the fluid domain Ω_f . Here, $\mathbf{u}$ denotes the velocity field, ρ the mass density, p the pressure, μ the dynamic viscosity, $\mathbf{K}$ the capillary term, and $\mathbf{f}_V$ the external body forces acting on the system. In general, the quantities $\rho = \rho(\tilde{\varphi})$, $\mu = \mu(\tilde{\varphi})$, and $\mathbf{K} = \mathbf{K}(\tilde{\varphi})$ depend on the phase-field variables. Furthermore, the capillary term is expressed as

$$\mathbf{K}(\tilde{\varphi}) = -\tilde{\varphi} \nabla \Phi, \quad \Phi = \tilde{\beta} \partial_{\tilde{\varphi}} \psi - \tilde{\alpha} \nabla^2 \tilde{\varphi}. \quad (5)$$

Extension to multiphase flow with rigid-body coupling. To incorporate rigid-body interactions into the two-phase flow framework, a normalization procedure following Reder *et al.* [26] is employed. This allows for a diffuse representation of the fluid-solid interface and an extension of Eqs. (3) and (4) from the fluid domain Ω_f to the whole computational domain Ω . To this end, a multiphase formulation with N phases $\alpha = 1, \dots, N$ is introduced, employing phase-field variables $\varphi_\alpha(\mathbf{x}, t) \in [0, 1]$ which represent the local volume fraction of phase α . For a number of N^f fluid and N^s solid phases, we introduce the volume fractions of all fluid and solid phases as

$$\varphi^f = \sum_{\alpha=1}^{N^f} \varphi_\alpha^f \quad \text{and} \quad \varphi^s = \sum_{\alpha=1}^{N^s} \varphi_\alpha^s, \quad (6)$$

yielding the summation constraint $\varphi^f + \varphi^s = 1$. The normalization of the fluid phases yields

$$\tilde{\varphi}_\alpha^f := \frac{\varphi_\alpha^f}{\varphi^f}, \quad \text{for } \varphi^f > 0, \quad (7)$$

with the sum condition $\sum_{\alpha=1}^{N^f} \tilde{\varphi}_\alpha^f = 1$. For the case of two fluid phases, the system can be described by the single variable $\tilde{\varphi} := \tilde{\varphi}_1^f$, which is governed by Eq. (3). Therefore, the multiphase problem is split into two-phase subproblems with respect to $\{\varphi^s, \varphi^f\}$ and $\{\tilde{\varphi}_1, \tilde{\varphi}_2\}$ within the fluid, cf. [26]. Furthermore, the boundary conditions at the diffuse fluid-solid interface are incorporated through the approach of Beckermann *et al.* [28] for the Navier-Stokes equations, and the approach of Li [29] for the Allen-Cahn equation. A detailed comparison of these approaches regarding the diffuse application of no-slip boundary conditions can be found in Reder *et al.* [30]. The application of these approaches gives rise to the coupled system of equations

$$\partial_t(\rho h^{\text{fs}} \mathbf{u}) = -\nabla \cdot (\rho h^{\text{fs}} \mathbf{u} \otimes \mathbf{u}) - h^{\text{fs}} \nabla p \quad (8a)$$

$$+ \nabla \cdot (h^{\text{fs}} \mu \nabla \mathbf{u}) + \mathbf{u} \nabla \cdot (\mu \nabla h^{\text{fs}})$$

$$+ \rho h^{\text{fs}} \mathbf{f}_V + h^{\text{fs}} \mathbf{K}$$

$$0 = \nabla \cdot (h^{\text{fs}} \mathbf{u}), \quad (8b)$$

$$\begin{aligned}
h^{\text{fs}} \dot{\tilde{\varphi}} = & M \left[\tilde{\beta} h^{\text{fs}} \partial_{\tilde{\varphi}} \psi - \tilde{\alpha} \left(\nabla \cdot h^{\text{fs}} \nabla \tilde{\varphi} \right. \right. \\
& \left. \left. - h^{\text{fs}} \|\nabla \tilde{\varphi}\| \nabla \cdot \frac{\nabla \tilde{\varphi}}{\|\nabla \tilde{\varphi}\|} \right) \right. \\
& \left. - \partial_{\varphi^{\text{f}}} h^{\text{fs}} \|\nabla \varphi^{\text{f}}\| (\sigma_{2\text{s}} - \sigma_{1\text{s}}) \partial_{\tilde{\varphi}} h^{\text{ff}} \right] \\
& + h^{\text{fs}} v^{\text{e}} \|\nabla \tilde{\varphi}\|
\end{aligned} \quad (8\text{c})$$

which are valid over the entire domain, i.e., $\mathbf{x} \in \Omega$. Herein, $\otimes$ denotes the dyadic product, defined as $(\mathbf{a} \otimes \mathbf{b})_{ij} = a_i b_j$. Furthermore, the interpolation function between the fluid and solid interface is defined as $h^{\text{fs}}(\varphi^{\text{f}}) = (\varphi^{\text{f}})^2(3 - 2\varphi^{\text{f}})$ with φ^{f} according to Eq. (6).

B. Binder transport model

In previous studies on binder migration during electrode drying [10–12], the redistribution of binder is commonly described using convection-diffusion-type transport equations. In particular, Zihurul *et al.* [11] employ a one-dimensional convection diffusion equation to model binder transport along the electrode thickness,

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - cv, \quad (9)$$

where c denotes the binder concentration, D the diffusion coefficient, and v the effective convective velocity. While such approaches successfully reproduce qualitative experimental trends [10–12], they are restricted to one-dimensional geometries and do not incorporate the capillary forces, which are the main reason for binder migration. To overcome these limitations, we introduce a two field formulation for binder migration, which distinguishes between mobile binder c dissolved in the solvent and deposited binder c_{a} during drying. Binder transport is therefore described by the following three-dimensional system of equations

$$\dot{c} := \frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = \nabla \cdot (D \nabla c) - q_{\text{d}} + \lambda, \quad \mathbf{x} \in \Omega_{\text{solvent}}, \quad (10\text{a})$$

$$\frac{\partial c_{\text{a}}}{\partial t} = q_{\text{d}}, \quad \mathbf{x} \in \Omega_{\text{air}}. \quad (10\text{b})$$

Here, q_{d} denotes a source term

$$q_{\text{d}} = \begin{cases} \geq 0 & \mathbf{x} \in \partial\Omega_{\text{solvent}} \cap \partial\Omega_{\text{air}} \\ 0 & \text{otherwise.} \end{cases} \quad (11)$$

Specifically, the deposition term describes the continuous accumulation and immobilization of the binder at the moving liquid-gas interface. As the solvent evaporates and the fluid domain shrinks, the binder is left behind at the receding front and effectively transfers into the region now occupied by the gas phase. This mathematical formulation represents the consolidation of the binder at the drying front, rather than a physical transfer of the polymer into the gaseous phase. Furthermore, a Lagrange multiplier λ is introduced to enforce global binder mass conservation despite the continuous

reduction of the solvent volume due to evaporation

$$\frac{d}{dt} V_{\text{solvent}} = \frac{d}{dt} \int_{\Omega_{\text{solvent}}} dV \neq 0. \quad (12)$$

Regarding the boundary condition, a homogeneous no-flux boundary condition is applied on the fluid boundary Ω_{solvent} ,

$$(D \nabla c) \cdot \mathbf{n} = 0, \quad \mathbf{x} \in \partial\Omega_{\text{solvent}}. \quad (13)$$

This condition mathematically ensures that the binder is strictly confined within the liquid domain, so that it cannot cross into the solid particles or evaporate into the gas phase when deposition is not permitted. Notably, this constraint does not restrict the hydrodynamic fluid velocity ($\mathbf{u}$), which remains fully resolved and dynamic at the moving liquid-gas interface during capillary recession. Therefore, $\nabla c \cdot \mathbf{n} = 0$ reflects the case where the binder is transported solely alongside the interface velocity (motion + evaporation) and thus remains confined to the solvent phase. Mass transfer of binder over the solvent-air interface is only present in terms of the deposition modeling achieved by the source terms. Since the binder concentration in the air is reflected by the additional field c_{a} , the boundary condition regarding c stays unchanged even for the case with binder transfer over the interface, which is due to the formulation using a splitting of the binder concentration into two fields. Coupling the transport equations (10) with the equation system (8) ensures that capillary-driven binder migration is captured consistently, thereby overcoming earlier approaches that mainly associated binder redistribution with film shrinkage rather than capillary forces [10–12]. In addition, instead of a one-dimensional calculation of binder migration, a three-dimensional approach is employed. To incorporate the no-flux boundary condition in a diffuse interface framework, an indicator function I is introduced to distinguish the solvent domain from the surrounding phases, thereby following the approach of Li *et al.* [29]. Using this indicator function, the binder transport equation (10a) can be reformulated to the whole-domain formulation

$$\frac{\partial c I}{\partial t} = -I \mathbf{u} \cdot \nabla c + \nabla \cdot (I D \nabla c) - I q_{\text{d}} + \lambda + c \frac{\partial I}{\partial t}, \quad \mathbf{x} \in \Omega, \quad (14)$$

where the last term accounts for the motion of the interface between solvent and air. The derivation of Eq. (14) can be found in detail in Appendix. A diffuse interface model for Eq. (14) is obtained by approximating the indicator function as $I \approx \tilde{\varphi}$ using the normalized phase variable $\tilde{\varphi}$ of the solvent. To isolate the individual effects of evaporation and binder deposition, two limiting cases are considered before discussing the fully coupled system. Based on these limiting cases, the corresponding validation setups are defined in Sec. III.

Special case without deposition and evaporation. In the absence of evaporation, the solvent volume remains constant and

$$\frac{d}{dt} V_{\text{solvent}} = \frac{d}{dt} \int_{\Omega_{\text{solvent}}} dV = 0 \quad (15)$$

holds. Consequently, the Lagrange multiplier vanishes, $\lambda = 0$. Furthermore, if binder deposition into the air phase is neglected, the deposition source term can be set to $q_{\text{d}} = 0$. Under these assumptions, the binder transport equation (14) reduces

to a purely convection-diffusion equation in the form of

$$\frac{\partial cI}{\partial t} = -\mathbf{I}u \cdot \nabla c + \nabla \cdot (ID\nabla c) + c \frac{\partial I}{\partial t}. \quad (16)$$

This limiting case is used to validate the implementation by means of the bubble rise benchmark proposed by Reder *et al.* [31] in Sec. III A, where neither evaporation nor binder deposition is present and, therefore, the binder concentration is transported according to the solvent motion and completely remains in the moving solvent domain.

Special case without deposition. In a second step, binder evolution is investigated in the presence of evaporation, while deposition into the air phase is still neglected. Accordingly, the deposition source term is set to $q_d = 0$, whereas the Lagrange multiplier can no longer be omitted, since

$$\frac{d}{dt} V_{\text{solvent}} = \frac{d}{dt} \int_{\Omega_{\text{solvent}}} dV \neq 0 \quad (17)$$

holds due to solvent evaporation. To ensure global binder mass conservation, the constraint

$$\frac{d}{dt} \int_{\Omega} cI dV = 0 \quad (18)$$

must be fulfilled. Therefore, the Lagrange multiplier λ is defined as

$$\lambda = w_c p_c, \quad w_c = \tilde{\varphi}(1 - \tilde{\varphi}), \quad (19)$$

where p_c is a global amplitude obtained through a projection step and w_c a distribution function to redistribute missing binder associated with evaporated solvent over the fluid-fluid interface. In this case, the binder concentration stays inside the solvent while being globally preserved according to Eq. (18). This configuration is investigated using the evaporation of a fluid phase in a capillary geometry, as described in Sec. III B.

General case with deposition. In the general case, the concentration c_a of deposited binder in the air is nonzero and the conservation constraint reads

$$\frac{d}{dt} \left(\int_{\Omega} cI dV + \int_{\Omega} (1 - I)c_a dV \right) = 0. \quad (20)$$

The binder deposition is explicitly modeled by the source term q_d , which is assumed to depend on the local volume fraction and deposited binder concentration $q_d = q_d(\varphi_1, \varphi_2, \dots, \varphi_N, c_a(\mathbf{x}, t))$. It is modeled as

$$q_d = w_d p_d, \quad w_d = \tilde{\varphi}(1 - \tilde{\varphi})(f \varphi^s + (1 - f)), \quad (21)$$

where f is a model parameter that controls the deposition of binder at the triple phase regions, i.e., for $f = 0$, all binder will be deposited at the fluid-fluid interface, while for $f = 1$ the deposition is restricted to triple phase regions. Therefore, the separation parameter f can be used to model increased deposition of binder near the solid particle. Introducing this mechanism to favor deposition near particles is motivated by findings in the literature [2], which hypothesize that binder is preferentially retained close to particle surfaces, either directly on the carbon black surface near the active material particles, or at the fluid-fluid interface, where the binder remains within the carbon binder domain that fills the space between particles and is thereby retained in the electrode. The parameter p_d defines the corresponding deposition amplitude.

Solving method. A split-step approach is used to solve the binder transport equations, which separates transport, deposition, and mass-conservation correction into consecutive substeps. All model equations are discretized using a finite difference method on a Cartesian grid. The corresponding algorithm to update the binder field, which is executed for each time step n , is subsequently described. Therefore, the information about I^{n+1} is known *a priori* to the concentration calculation.

(i) Solving the transport equation (14) without the source term to get the interim solution:

$$(cI)^* = (cI)^n + \Delta t [-\mathbf{I}u \cdot \nabla c + \nabla \cdot (ID\nabla c)]^n + c(I^{n+1} - I^n). \quad (22)$$

(ii) Calculate the difference of initial binder mass in the solvent and the binder mass in the solvent of the interim solution

$$\Delta_{cl} = \int_{\Omega} cI(\mathbf{x}, t^0) dV - \int_{\Omega} (cI)^*(\mathbf{x}, t^n) dV. \quad (23)$$

(iii) Update the binder concentration c_a in the air phase to model the binder deposition according to

$$q_d = w_d p_d, \quad (24)$$

$$w_d = f \tilde{\varphi}(1 - \tilde{\varphi}) \varphi^s + (1 - f) \tilde{\varphi}(1 - \tilde{\varphi}), \quad (25)$$

$$p_d = \frac{\Delta_{cl} - \int_{\Omega} (1 - I^n) c_a(\mathbf{x}, t^n) dV}{\int_{\Omega} w_d(\mathbf{x}, t) dV \Delta t}, \quad (26)$$

$$(c_a)^{n+1} = (c_a)^n + \Delta t q_d. \quad (27)$$

(iv) Ensure mass conservation by the correction step to obtain the solution for cI :

$$\lambda = w_c p_c, \quad (28)$$

$$w_c = \tilde{\varphi}(1 - \tilde{\varphi}), \quad (29)$$

$$p_c = \frac{\Delta_{cl} - \int_{\Omega} (1 - I^{n+1}) c_a(\mathbf{x}, t^{n+1}) dV}{\int_{\Omega} w_c(\mathbf{x}, t) dV \Delta t}, \quad (30)$$

$$(cI)^{n+1} = (cI)^* + \Delta t \lambda. \quad (31)$$

III. VALIDATION

In this section, the model is validated using two examples without deposition showing conservation of mass in the fluid despite movement of phase boundaries and evaporation. In the initial example, a liquid droplet is transported in a gravitational field to ensure that the initial amount of binder remains within the upwards moving liquid. In the second example, a fluid undergoes evaporation, while the initial amount of binder should remain within the fluid. Additionally, the gradient can be balanced by diffusion of the concentration. The results are evaluated based on the interpolated concentration

$$\tilde{c} = cI + (1 - I)c_a. \quad (32)$$

A. Bubble rise problem

Setup and parameters. In order to investigate the conservation of mass in a moving fluid without evaporation, case 2

TABLE I. Overview of the simulation parameter for bubble rise problem, adopted from Reder *et al.* [31].

ρ_1 in kgm^{-3}	ρ_2 in kgm^{-3}	μ_1 in Pas	μ_2 in Pas	g in ms^{-2}	σ in Nm^{-1}	κ in s^{-1}	D in m^2/s
1000	1	10	0.1	0.98	1.96	0	0

of the bubble rise problem [31,32] is utilized. The simulation parameters are summarized in Table I. For the purpose of this study, a spherical bubble of fluid 2 (φ_2) with a radius of $R = 0.25$ and an initial concentration of $c_0 = 0.01$ is placed within the simulation domain.

Results. Considering the dimensionless time $t^* = t\sqrt{g/R}$, Fig. 1 shows the results of the phase field variable φ_2 (top) and the concentration distribution $\tilde{c}$ (bottom) as a function of the geometric dimensions for the three points in time $t^* = 0$, $t^* = 1.46$, and $t^* = 2.83$. φ_2 undergoes a transformation over time, evolving from a spherical to a nonconvex shape. This behavior is in accordance with the findings reported in Reder *et al.* [31]. In addition, the concentration distribution $\tilde{c}$ exhibits a similar tendency, manifesting analogous shapes over time. Furthermore, the total amount of binder, quantified by the volume integral of the concentration distribution $\tilde{c}$,

$$\int_{\Omega} \tilde{c}(\mathbf{x}, t) dV = 1.86 \times 10^{-3},$$

is conserved over time. Thus, the model demonstrates its ability to transport the binder within a moving fluid, with no artificial mass loss or nonphysical concentration drift.

B. Binder migration during evaporation in a two fluid system

Setup and parameters. In order to investigate the behavior of binder migration in the presence of evaporation, the validation case contact angle at a moving contact line from [19] is employed, with slightly modified parameters. In contrast to the original setup, the contact angle is set to $\theta = 90^\circ$ instead of $\theta = 60^\circ$, which results in a horizontal drying front. A schematic sketch of the present simulation setup is shown in Fig. 2, while the corresponding simulation parameters are summarized in Table II. The dimensionless binder concentration is implemented as a relative scalar field representing the local solute fraction. This decoupled approach is physically justified since the binder concentration has no backcoupling to other constitutive properties in the system, such as the viscosity. At the beginning of the simulation ($t = 0$), this field is initialized uniformly with a baseline scalar value of $c_0 = 0.01$ within the liquid solvent domain φ_2 and 0.0 within the gas phase. The rates of evaporation are determined by the parameter κ . Furthermore, the system is characterized by the Péclet numbers $\text{Pe} = 12.5 \times 10^{-2}$ and $\text{Pe} = 12.5$, respectively. The Péclet number is thereby defined through

$$\text{Pe} = u_c l_c / D = \kappa l_y^2 / D. \quad (33)$$

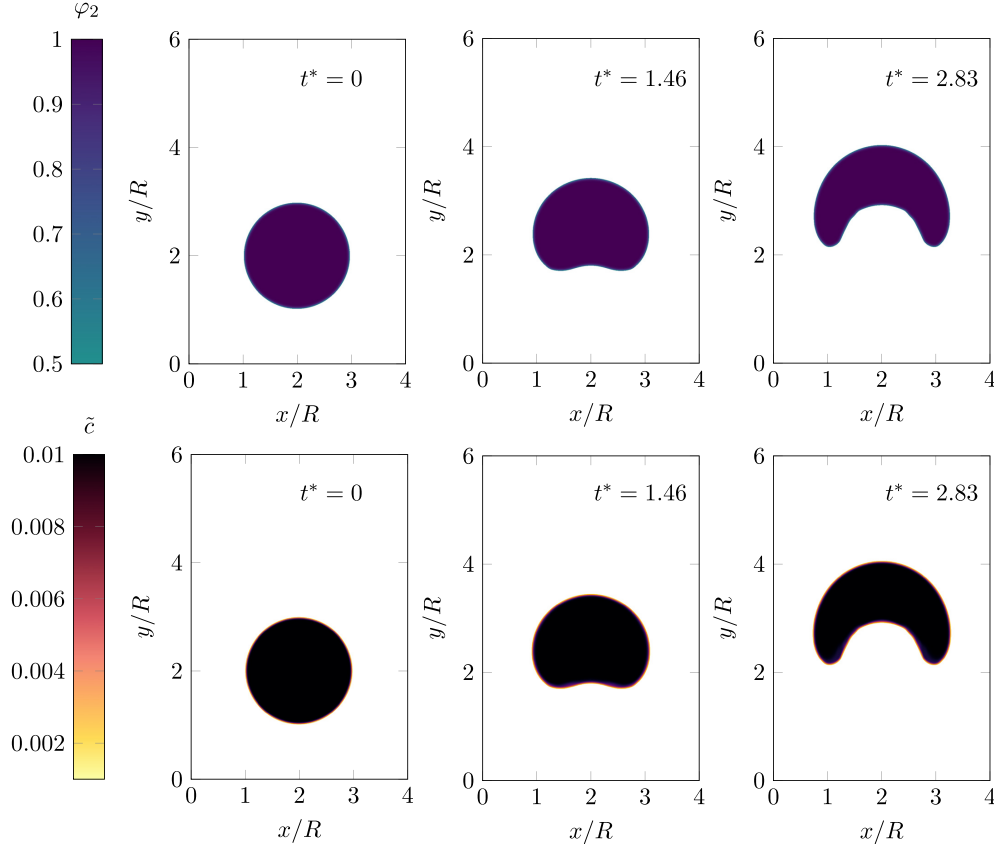


FIG. 1. Spatial representation of the phase-field variable φ_2 (top) and the concentration $\tilde{c}$ (bottom) for the three dimensionless time points ($t^* = t\sqrt{g/R}$): $t^* = 0$, $t^* = 1.46$, and $t^* = 2.83$.

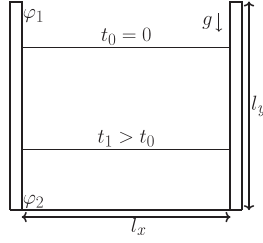


FIG. 2. Schematic representation of the validation setup for the binder evolution in a drying fluid.

This setup closely resembles the simulation setup used by Li *et al.* [12], who have employed the same Péclet numbers. Therefore, the diffusivity is fitted according to the characteristic velocity.

Results. Figure 3 displays the normalized concentration profiles $\tilde{c}/\tilde{c}^0$ as a function of the normalized height $\hat{y}$ at three dimensionless times $t_1 = 0.2719$, $t_2 = 0.625$, and $t_3 = 0.8103$. The outcomes for $Pe = 12.5 \times 10^{-2}$ are shown on the left of Fig. 3, while those for $Pe = 12.5$ are shown on the right. Utilizing a diffuse interface leads to the formation of a transition zone subsequent to the physical liquid front, thereby elucidating the decline in the concentration curves depicted in Fig. 3. It can be seen that as the Péclet number increases, the rate of evaporation of the liquid accelerates. Furthermore, the differences between the maxima and minima are more pronounced for a high Péclet number than for a low Péclet number. This outcome is anticipated, given that convective transport dominates at a Péclet number of $Pe = 12.5$, while diffusive transport prevails at a Péclet number of $Pe = 12.5 \times 10^{-2}$. Furthermore, a qualitative comparison with the studies by Font *et al.* [10] and Li *et al.* [12] reveals behavior consistent with the present results. Besides correctly capturing the dominant transport phenomena, these findings demonstrate that, if no deposition of binder into the air phase is considered, the proposed model reproduces the given results from literature.

IV. APPLICATION

In this section, the proposed model is applied to investigate capillary induced binder migration in battery electrodes. The influence of key process and material parameters, like the solvent viscosity, evaporation rate, and surface tension is systematically analyzed by varying these parameters. In addition, the impact of wettability is assessed by considering different contact angle conditions. The resulting binder migration is quantified using a normalized vertical binder gradient, enabling a direct comparison between microstructures and process parameters.

A. Preprocessing and simulation setup

Microstructures. The microstructures used in this study are obtained from the real hard carbon electrodes published by Klemens *et al.* [2] and were subsequently processed following the workflow described by Weichel *et al.* [19]. In the present work, the two considered microstructures are denoted as HC-B and HC-C, consistent with the terminology used in previous studies [2,19]. Compared to Weichel *et al.* [19], the image segmentation parameters were slightly adjusted, leading to minor changes in the resulting pore size distributions. More specifically, the image segmentation thresholds were recalibrated to capture particle contact junctions and capillary throats with greater accuracy, as these geometric features are highly critical for the local pinning mechanisms of the binder transport model. These modifications do not alter the qualitative microstructural characteristics but influence quantitative measures such as breakthrough times. HC-C generally exhibits a broader pore size distribution and a larger mean particle diameter compared to HC-B [2]. In the segmented simulation input, the hard carbon particles are treated as solid bodies, since resolving internal porosity [33–35] would be computationally prohibitive. Both microstructures are characterized by the solid volume fractions χ and the area-specific weight of the dry coating M . The solid volume fraction χ is obtained directly from the segmented simulation input, while the area-specific mass M is taken from experimental coating data. For HC-B, these values are $\chi_{\text{solid}}^{\text{HC-B}} = 0.465$ and $M_{\text{S}}^{\text{HC-B}} = 79.5 \times 10^{-3} \text{ kg/m}^2$, whereas HC-C is defined by $\chi_{\text{solid}}^{\text{HC-C}} = 0.549$ and $M_{\text{S}}^{\text{HC-C}} = 78.4 \times 10^{-3} \text{ kg/m}^2$. It is to be noted, that the value for κ is sensitive to the choice of χ , which is influenced by the segmentation process. The simulation domain represents a physical section of the microstructure with a width of $l_x = 55.0 \mu\text{m}$ and a dry film thickness of $l_y = 37.5 \mu\text{m}$. A total physical simulation height of $l_y = 40.07 \mu\text{m}$ is employed to account for the air phase. The initial binder concentration c is set to $c = 0.01$ as a dimensionless concentration, and uniformly initialized within the solvent filled regions. Across the diffuse interface, the concentration is smoothly interpolated following the variable $\tilde{\phi}$. Figure 4 illustrates the original SEM pictures of the electrodes and the segmented simulation input.

Parameters. Table III provides an overview of all physical parameters utilized in the simulations, including fluid densities, viscosities, surface tension, gravity, mobility, and the binder diffusion coefficient in the solvent. To obtain the velocity of the receding liquid surface v^e [see Eq. (3)], we employ the parameter

$$\kappa = \frac{\rho_{\text{solid}} \chi_{\text{solid}}}{\rho_{\text{solvent}} M_{\text{S}}} \dot{r}_0, \quad (34)$$

which links the prescribed evaporation rate of $\dot{r}_0 = 9 \text{ gm}^{-2} \text{ s}^{-1}$ to the simulation framework [19]. Using this relation and the information about the microstructures, we

TABLE II. Overview of the simulation parameter for the validation case contact angle at a moving contact line.

l_x in μm	l_y in μm	ρ_1 in kgm^{-3}	ρ_2 in kgm^{-3}	μ_1 in Pas	μ_2 in Pas	g in ms^{-2}	σ in N m^{-1}
56	56	1.225	997	1.72×10^{-5}	1×10^{-3}	9.81	7.3×10^{-2}

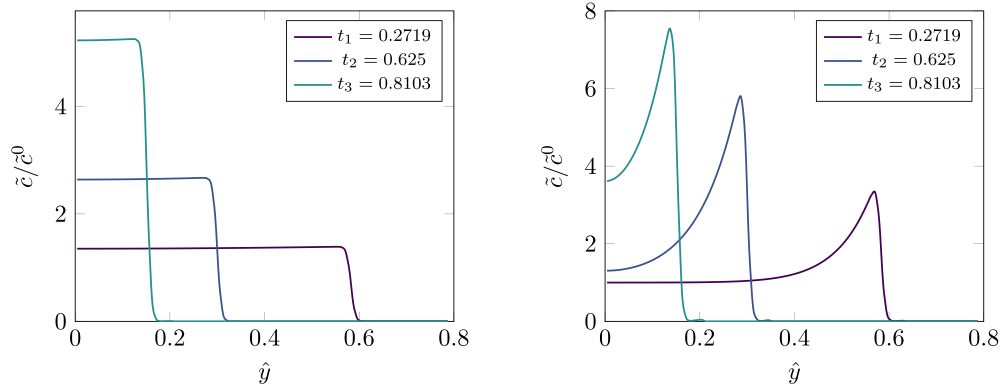


FIG. 3. Normalized concentration profiles $\tilde{c}/\tilde{c}^0$ as a function of the height $\hat{y}$ at $x = l_x/2$ at three dimensionless times: $t_1 = 0.2719$, $t_2 = 0.625$, and $t_3 = 0.8103$, and the Péclet numbers $Pe = 12.5 \times 10^{-2}$ (left) and $Pe = 12.5$ (right).

obtain $\kappa^{\text{HC-B}} = 0.1089 \text{ s}^{-1}$ and $\kappa^{\text{HC-C}} = 0.1303 \text{ s}^{-1}$ for HC-B and HC-C, respectively. Binder separation is modeled by the separation factor $f = 0.9$. Furthermore, the grid consists of $n_x = 608$ cells in the x direction and $n_y = 448$ cells in the y direction, which directly follows from the underlying scanning electron microscopy (SEM) data. Mapping the physical domain onto the computational domain requires fulfillment of two physical criteria while ensuring computational feasibility. First, the computational domain must be sufficiently fine to resolve the diffuse liquid-gas interface. Second, the grid resolution must be fine enough to resolve the narrowest geometric pore throats in order to correctly map the pore emptying. Using the characteristic length $l_c = l_x = 55.0 \mu\text{m}$, the

dimensionless spatial step sizes are $\Delta x^* = \Delta x/l_c = 1.64 \times 10^{-3}$ and $\Delta y^* = \Delta y/l_c = 1.65 \times 10^{-3}$. Moreover, the interface widths parameter ϵ is chosen, such that the fluid-solid interface and fluid-fluid interface are resolved using a grid-dependent thickness corresponding to five and ten grid cells, respectively. The numerical time-step size Δt is controlled dynamically throughout the simulation, adhering to a standard Courant-Friedrichs-Lewy (CFL) stability criterion based on the local fluid velocity combined with the diffusive stability limits imposed by the binder diffusion coefficient [26]. Regarding computational performance, the two-dimensional phase-field drying simulations were performed using parallel computing on a high-performance cluster with 128 AMD

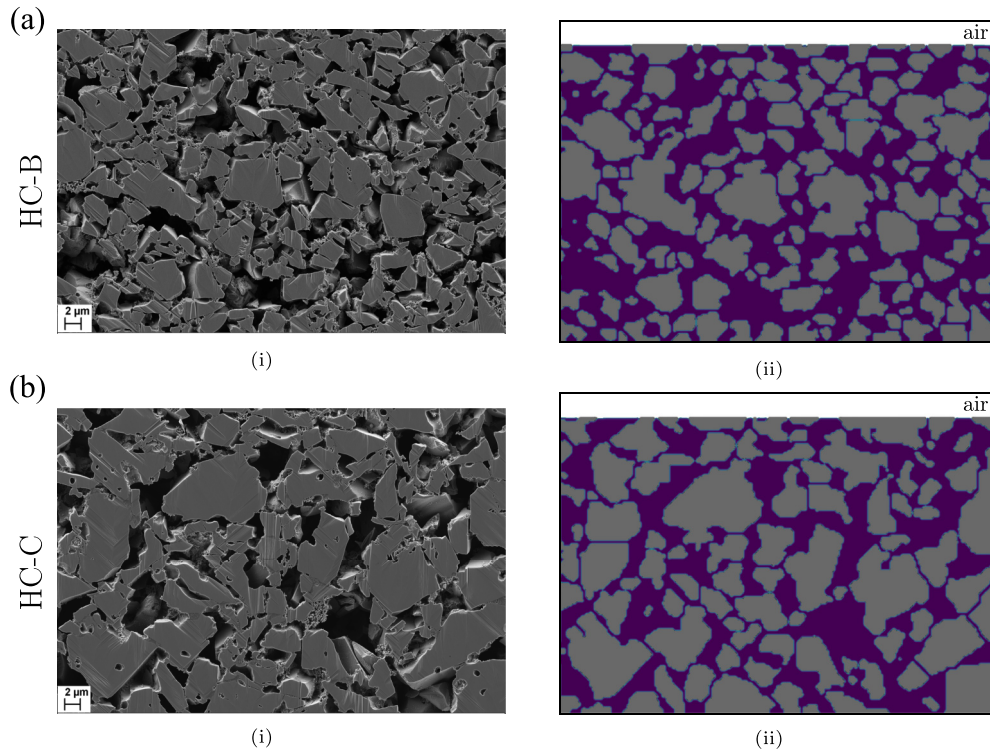


FIG. 4. SEM images of dried microstructures (i) with different pore size distributions and particle diameters, labeled (a) HC-B and (b) HC-C [2]. The corresponding digital representation of the initial three-phase microstructure (ii) composed of air (white), particles (gray), and the solvent phase (blue) containing the dissolved mobile binder.

TABLE III. Overview of the physical and numerical simulation parameters for the hard carbon microstructures.

Parameter	Symbol	Physical value	Unit	Reference/justification
Physical length	l_x	55.0	μm	SEM Image Domain
Physical height	l_y	40.07	μm	SEM Image Domain
Density hard carbon	ρ_{HC}	2062.0	kgm^{-3}	[2]
Density solvent phase	ρ_{solvent}	997.0	kgm^{-3}	Standard value (Water, 25 °C)
Density air phase	ρ_{air}	1.225	kgm^{-3}	Standard value (Air, 25 °C)
Viscosity solution	$\mu_{\text{solvent},0}$	1.0	Pas	Concentrated polymer solution [2]
Viscosity air phase	μ_{air}	1.72×10^{-5}	Pas	Standard value (Air, 25 °C)
Surface tension	σ_0	72.4×10^{-3}	N m^{-1}	Standard value (Water/Air)
Gravity	g	9.81	ms^{-2}	Standard value
Phase-field mobility	M	30	ms/kg	Numerical calibration
Binder diffusion coeff.	D	1.12×10^{-16}	m^2/s	[36]

EPYC cores of third generation. A typical simulation run for a single parameter set required approximately 48 h to reach the fully dried state.

Parameter studies. To enable computationally feasible simulation times, the evaporation rate is artificially scaled by a factor of 5.0×10^4 , following the approach proposed by Wolf [18] and adopted in our previous work [19]. To preserve the capillary number, the solvent viscosity is reduced by the same factor. Through this approach, the computational cost can be significantly reduced. Furthermore, the diffusion coefficient is scaled by the same factor to maintain the Péclet number constant, thereby preventing an under representation of diffusion. The binder distribution is evaluated using the previously defined binder concentration field $\tilde{c}$ [see Eq. (32)]. To quantify the binder distribution after electrode drying, the concentration $\tilde{c}$ is evaluated using the following procedure. First, horizontal averaging over the x direction is applied

$$\langle \tilde{c} \rangle_x = \frac{1}{l_x} \int_0^{l_x} \tilde{c} dx, \quad (35)$$

yielding a one dimensional vertical concentration profile along the y direction at the reference time t_0 and the final time t_{end} . The resulting profiles are truncated at the last location where the concentration exceeds a negligible threshold. Subsequently, the relative changes in the binder distribution

$$R(y) = \frac{\langle \tilde{c} \rangle_x(y, t_{\text{end}})}{\langle \tilde{c} \rangle_x(y, t_0)} \quad (36)$$

are computed. A linear regression of $R(y)$ yields the slope m , which defines the normalized vertical binder gradient. This scalar quantity is used as the primary metric for comparing binder migration across different simulations. In a first study, the impact of the parameters contributing to the capillary number,

$$Ca = \frac{\mu_{\text{solvent}} u_c}{\sigma}, \quad (37)$$

on the final binder distribution is examined by varying the solvent viscosity μ_{solvent} , the evaporation rate $\dot{r}$, and the surface tension σ . An increased evaporation rate leads to a steeper binder gradient, whereas a higher viscosity counteracts this effect. Furthermore, the effect of changing the contact angle is analyzed while changing the viscosity. Finally, the breakthrough time $t^{\text{breakthrough}}$, defined as the moment when the air

phase first reaches the substrate, is analyzed as an additional characteristic parameter.

B. Temporal evolution of drying

Figure 5 depicts the distribution of the solvent and the binder $\tilde{c}$ for the two microstructures at selected nondimensionalized times for a capillary number of $Ca = 6.23 \times 10^{-3}$. HC-B is illustrated in the upper row and HC-C in the lower row. The first two columns correspond to the nondimensionalized times $t_1 = 0.158$ and $t_2 = 0.359$, while the last column shows the state at the end of drying ($t_{\text{HC-B}}^{\text{end}} = 0.789$ and $t_{\text{HC-C}}^{\text{end}} = 0.555$). Only regions exceeding the initial value of $c = 0.01$ are displayed for the binder distribution. The simulations for both microstructures demonstrate that binder accumulates in areas where the interparticle distance is relatively small and capillary channels are narrow. Such narrow capillary channels are preferential pathways for solvent flow, as larger pores drain prior to smaller ones due to differences in Laplace pressure, thereby promoting enhanced binder accumulation. A comparison of the two microstructures reveals that the microstructure HC-B exhibits a more homogeneous binder distribution over the electrode height, whereas the binder distribution in the HC-C microstructure is predominantly concentrated in the upper region. This trend is also reflected in the evaluation of the normalized vertical binder gradient m . For comparison, HC-B exhibits a normalized vertical binder gradient of $m = 0.61$, whereas HC-C shows a higher value of $m = 1.45$. These findings are consistent with experimental observations [37], which report reduced binder migration in microstructures composed of smaller particles and narrower pore networks.

C. Modification of the solvent viscosity

To further investigate the impact of the solvent viscosity on the final binder distribution, this section examines the effect of increasing solvent viscosity under constant evaporation rate and surface tension. Thereby, the reference viscosity $\mu_{\text{solvent},0}$ is varied by the factor $k^{\text{viscosity}}$, with $k^{\text{viscosity}} \in \{1, 2, 4, 6, 8, 10, 15, 20, 30, 40, 50\}$. Figure 6 shows the nondimensionalized breakthrough time $t^{\text{breakthrough}}$ as a function of the solvent viscosity, while Fig. 7 presents the vertical binder gradient m as a function of solvent viscosity. The results are shown in blue and red for the microstructures

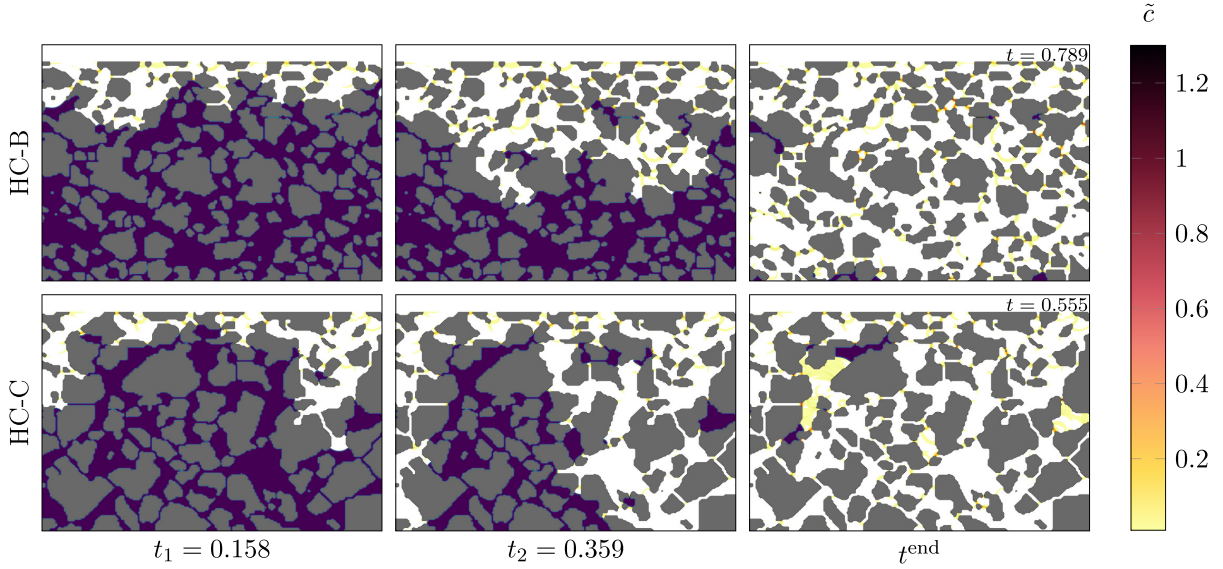


FIG. 5. Temporal evolution of the solvent and binder concentrations for HC-B (top row) and HC-C (bottom row). The first two temporal sequences correspond to the identical nondimensionalized times $t_1 = 0.158$ and $t_2 = 0.359$, whereas the last pictures correspond to the end of drying for the nondimensionalized times $t_{\text{HC-B}}^{\text{end}} = 0.789$ and $t_{\text{HC-C}}^{\text{end}} = 0.555$. The normalized vertical binder gradients are $m = 0.61$ and $m = 1.45$ for HC-B and HC-C, respectively. The solvent is depicted in blue and the particles in gray. For clarity, the binder distribution is displayed only for concentrations larger than $c > 0.01$.

HC-B and HC-C, respectively. For both microstructures, the breakthrough time increases with increasing viscosity, exhibiting a behavior consistent with previous studies [19,23]. With respect to binder migration, microstructure HC-C exhibits a larger vertical gradient than microstructure HC-B for all considered viscosities, which can be explained by the smaller particle size of HC-B [2]. Furthermore, neither microstructure shows a systematic increase in the binder gradient with increasing viscosity, indicating that viscosity alone does not constitute a dominant control parameter for binder migration under the investigated drying conditions. It should be noted that real slurries exhibit shear-thinning behavior [2] and sometimes yield point behavior [38], which is not captured by the present model [19], but can have an influence.

D. Modification of the evaporation rate

To increase the throughput of dried electrodes, the evaporation rate can be enhanced. However, higher drying rates lead

to stronger binder gradients across the electrode thickness, which can deteriorate the chemical and mechanical properties of the electrode [2,38]. This section investigates the influence of the evaporation rate and the microstructure on the gradients in the final binder distribution. For this purpose, the evaporation rate is varied according to $\dot{r} = k^{\text{evap}} \dot{r}_0$ with $k^{\text{evap}} \in \{3, 4, 5, 6, 7, 8, 9\}$ and $\dot{r}_0 = 1 \text{ gm}^{-2} \text{ s}^{-1}$. The viscosity is fixed at $\mu = 10\mu_0$ for all simulations. Figure 8 displays the results for the microstructures HC-B and HC-C in blue and red, respectively. The black circles indicate the cases marked in black from the previous section corresponding to a factor of $k^{\text{evap}} = 9$. For all simulations, the normalized gradient m is higher for the microstructure HC-C. This can be attributed to the larger particle diameter of HC-C, which is consistent with findings reported in the literature [2] and with the discussion in the previous section. Furthermore, it is observed that the binder gradient increases with increasing

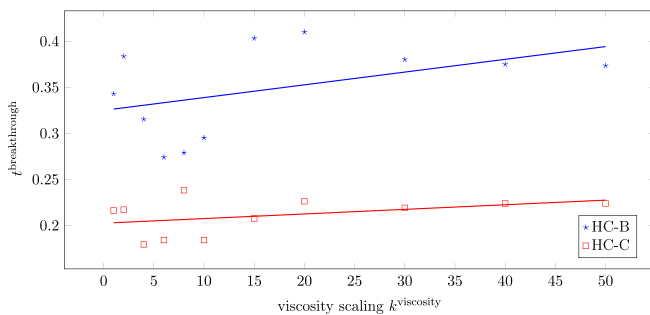


FIG. 6. Nondimensionalized breakthrough times as a function of the solvent viscosity $\mu_{\text{solvent}} = k^{\text{viscosity}} \mu_{\text{solvent},0}$. The results for microstructure HC-B and HC-C are shown in blue and red, respectively.

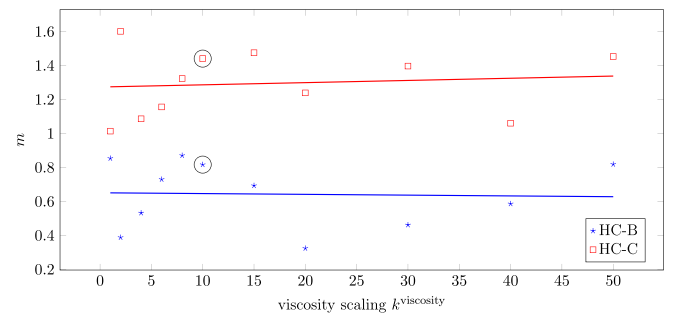


FIG. 7. Normalized binder gradient m as a function of the solvent viscosity $\mu_{\text{solvent}} = k^{\text{viscosity}} \mu_{\text{solvent},0}$. Black circles indicate the reference case for the further simulation studies, displayed in Figs. 8 and 9 with varying evaporation rate and surface tension, respectively.

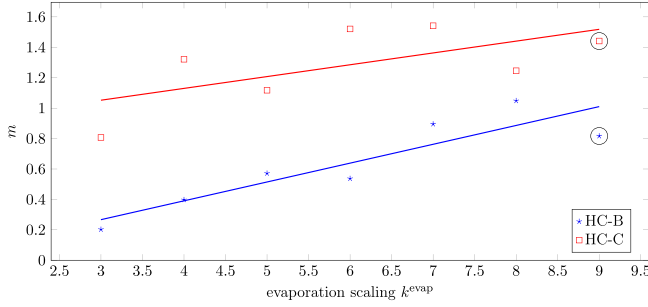


FIG. 8. Normalized slope of the binder gradient m as a function of the evaporation rate. The evaporation rate is given by $\dot{r} = k^{\text{evap}} \dot{r}_0$ and $\dot{r}_0 = 1 \text{ gm}^{-2} \text{ s}^{-1}$. The results for microstructures HC-B and HC-C are shown in blue and red, respectively. A viscosity of $\mu = 10\mu_0$ is used in the simulation study. The black circles indicate the marked cases from the previous section.

evaporation rate for all investigated drying conditions. For the microstructure HC-C, this trend is in good agreement with previous experimental studies [2,5,38]. A similar qualitative trend is observed for the HC-B microstructure in the present work. However, this finding is not fully consistent with the results reported by Klemens *et al.* [2]. In their study, hard carbon electrodes with smaller particle sizes exhibit a constant or even increased adhesion force with increasing evaporation rate, indicating a less pronounced binder migration. This behavior is attributed to a reduced amount of binder that can be transported by capillary driven flow in systems with smaller particles. In such microstructures, the slurry rheology exhibits a gel-like CMC/SBR network, which immobilizes a significant fraction of the binder and counteracts the capillary forces during the pore emptying process. In contrast, the present model predicts an increasing binder gradient for the HC-B microstructure with increasing drying rate. This result is consistent with the underlying physics of the model, in which higher evaporation rates lead to stronger capillary forces, while rheological effects associated with shear-thinning are neglected. Consequently, although the model correctly captures the fundamental capillary driven mechanism of binder migration, it overestimates binder mobility for microstructures characterized by small particle sizes. This discrepancy highlights the importance of incorporating shear-dependent viscosity into future model extensions in order to accurately represent binder transport in battery electrodes across different particle size regimes.

E. Variation of the surface tension

In this section the capillary number Ca is varied by modifying the surface tension σ according to $\sigma = k^{\text{surf}} \sigma_0$. The viscosity is set to $\mu = 10\mu_0$ for all simulations and an evaporation rate of $\dot{r} = 9 \text{ gm}^{-2} \text{ s}^{-1}$ is chosen. Figure 9 presents the results of the simulation study for $k^{\text{surf}} \in \{1/50, 1/40, 1/30, 1/20, 1/15, 1/10, 1/8, 1/6, 1/4, 1/2, 1\}$. The results for microstructure HC-B are shown in blue, while those for microstructure HC-C are shown in red. For both microstructures, the normalized binder gradient m increases with increasing factor k^{surf} , which indicates that a reduced surface tension leads to a weaker binder accumulation at the surface

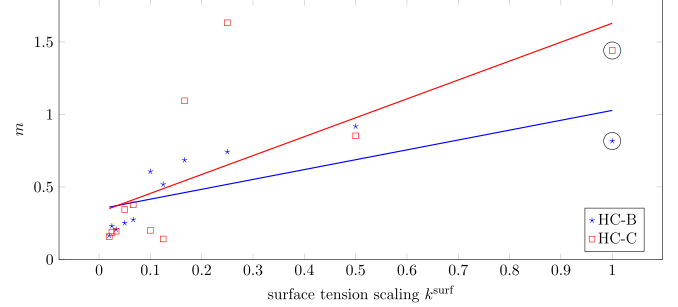


FIG. 9. Resulting normalized binder gradient m for both microstructures. The HC-B microstructure is shown in blue, while HC-C is shown in red. The surface tension σ_0 is varied by a factor of k^{surf} according to $\sigma = k^{\text{surf}} \sigma_0$. The viscosity is set to $\mu = 10\mu_0$ for all simulations with the reference case marked with black circles.

of the electrode. The impact of the surface tension variation is more pronounced for microstructure HC-C, whereas for smaller values of k^{surf} the gradients of both microstructures become nearly identical, suggesting a reduced influence of the chosen microstructure in this regime. Furthermore, for microstructure HC-C, a pronounced drop in the normalized binder gradient m is observed between $k^{\text{surf}} = 1/4$ and $k^{\text{surf}} = 1/8$. The overall decrease in the normalized binder gradient with a lower surface tension reduction factor k^{surf} can be attributed to weakened capillary forces, and thus a more homogeneous drying front. As a result, upward solvent flow by capillary driven convection and thus binder transport toward the electrode surface are suppressed. The sharp increase of the normalized binder gradient observed for HC-C between $k^{\text{surf}} = 1/8$ and $k^{\text{surf}} = 1/4$ can be explained by differences in the solvent front at the time of breakthrough to the substrate. These patterns are displayed in Fig. 10. On the left, the solvent front for a factor of $k^{\text{surf}} = 1/8$ is illustrated, while the result for a factor of $k^{\text{surf}} = 1/4$ is shown on the right. For the lower surface tension ($k^{\text{surf}} = 1/8$), the drying front appears homogeneous compared with the case of higher surface tension ($k^{\text{surf}} = 1/4$), where pronounced pinning at elevated positions within the microstructure is observed. Due to this pinning, the binder is transported toward the electrode surface over a longer period, driven by sustained solvent flow. These findings highlight the importance of physically consistent modeling approaches for binder migration. In particular, a simple diffusion-convection equation that neglects capillary effects is insufficient to accurately describe drying induced binder migration. In addition, the study shows that binder migration can be influenced by a change in surface tension.

F. Variation of the contact angle

In this section the impact of the contact angles, 90° , 60° , and 120° , on the microstructure HC-C is investigated. The simulations are performed using the reference parameter set listed in Table III. In this study, a contact angle of 60° represents hydrophobic behavior, whereas a contact angle of 120° corresponds to hydrophilic behavior [19]. Figure 11 illustrates the influence of solvent viscosity on the binder gradient m for the different contact angles. The results show that, for both hydrophilic and hydrophobic wetting conditions, the

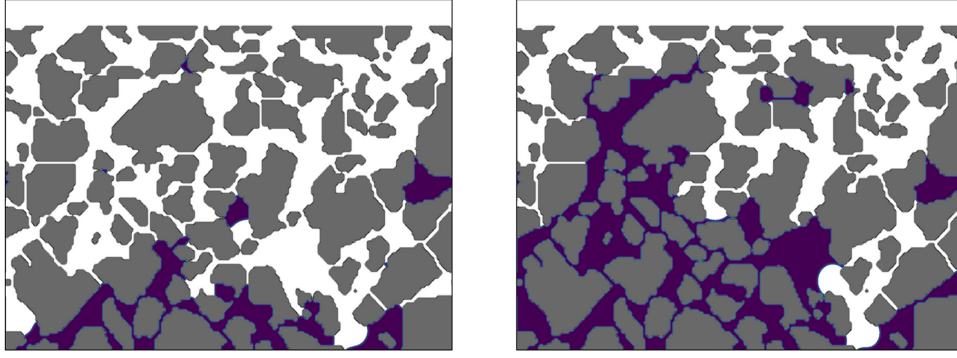


FIG. 10. Solvent distribution at the time of breakthrough for $k^{\text{surf}} = 1/8$ (left) and $k^{\text{surf}} = 1/4$ (right) for microstructure HC-C. The resulting slopes of the binder gradient m are $m = 1.63$ and $m = 0.14$ for $k^{\text{surf}} = 1/4$ and $k^{\text{surf}} = 1/8$, respectively.

sensitivity of the binder gradient to changes in viscosity is more pronounced compared with the reference case of 90° . In the hydrophilic case, the binder gradient increases more strongly with increasing viscosity, although it does not necessarily reach the highest absolute value for all viscosities considered. In contrast, hydrophobic wetting behavior exhibits a different trend, where the binder gradient remains less pronounced with an increasing viscosity. In summary, deviations of the contact angle from 90° significantly amplify the sensitivity of binder migration to solvent viscosity. This demonstrates that wettability of the active material constitutes a control parameter for tailoring binder migration in battery electrodes.

V. SUMMARY AND OUTLOOK

Most studies investigating binder migration during the drying of battery electrodes rely on simplified one-dimensional (1D) models. In these approaches, binder migration is commonly attributed to film shrinkage. However, according to the current state of knowledge, this assumption is incomplete, as binder migration predominantly occurs due to capillary transport during pore emptying. While our previous work [19] successfully established a phase-field based framework to simulate the pore-emptying stage during battery electrode drying, it was unable to predict binder transport within the porous structure. To close this scientific gap, the present work introduces a whole-domain transport equation coupled with an irreversible pinning model to capture, for the first time, microstructure-resolved, capillary-driven binder migra-

tion. The developed model is validated using two validation cases. First, the concentration distribution in a bubble-rise benchmark is analyzed, demonstrating that, in the absence of deposition, the concentration is transported by convection within the correct phase. Second, drying without deposition is investigated using a simplified setup by evaluating two Péclet numbers. The results are in good agreement with previously reported findings on binder migration. Subsequently, the model is applied to the drying process of real hard carbon electrodes. The influence of microstructure, viscosity, evaporation rate, surface tension, and contact angle is investigated. To this end, the considered microstructures differ in their pore size distributions and mean particle diameters. The simulations reveal that microstructures with smaller particle diameters generally exhibit a more homogeneous binder distribution. This behavior is consistent with observations reported in the literature [2]. With respect to variations in viscosity, the simulations indicate only a comparatively minor influence on binder distribution when a neutral contact angle of 90° is considered. However, a more pronounced impact regarding the viscosity can be seen by altering the wetting behavior. In contrast, increasing evaporation rates lead to a more pronounced binder inhomogeneity. A similar trend is observed with increasing surface tension. Overall, these results indicate that binder homogeneity can be effectively optimized by adjusting the contact angle and surface tension, thereby enabling an improved control of the drying process as a whole. In addition, this study demonstrates that models aiming to predict binder migration during battery electrode drying should explicitly incorporate capillary forces and microstructural information. It is important to emphasize that while the present 2D simulations provide valuable mechanistic insights into the pore-emptying process, they inherently approximate the complex transport pathways of a realistic 3D microstructure. Due to the reduction in dimensionality, 2D porous structures are more susceptible to pore blocking, as the fluid phase lacks the alternative geometric bypasses available in a true 3D network. Although this dimensional constraint does not alter the fundamental physical mechanisms and qualitative trends disclosed in this work, transitioning to fully resolved 3D microstructures remains a necessary step to evaluate the precise quantitative impact of these features on the overall transport dynamics. Furthermore, it should be noted that the evaporation velocity is imposed as spatially uniform at the

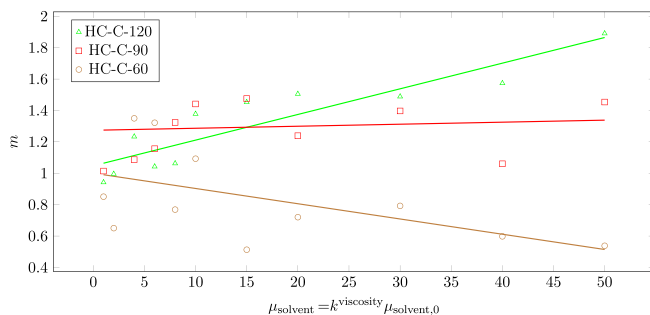


FIG. 11. Normalized gradient m as functions of the viscosity for HC-C with different angles (120° , 90° , and 60°).

interface, thereby neglecting local variations that may impact the binder movement and hence the final binder distribution.

In future works, the model is planned to be refined further to depict transport processes during pore emptying more accurately. The most prominent next step involves the transitioning to fully resolved three-dimensional microstructures. While this would increase the computational complexity, it also demands suitable 3D datasets, which are currently limited. In addition, the influence of mass transport resistance in the gas phase will be integrated into the model, since saturation of the gas phase in the pores may amplify capillary-driven transport toward the top by mitigating evaporation inside the pore structure. This is especially true for thicker electrodes, for which the assumption of a linear drying rate also becomes increasingly inaccurate [39]. To achieve this, the framework will be extended to incorporate the coupled differential equations for local humidity and temperature, define the corresponding transport boundary conditions, link these fields to the fluid flow, and account for the thermodynamic effects of latent heat phase transformations. By introducing this approach, the evaporation velocity will no longer be uniform at the interface but will instead be locally resolved. Furthermore, the model will be extended to account for shear-thinning solvent behavior, enabling a more realistic representation of non-Newtonian rheology during the drying process. Beyond rheological refinements, a promising direction for future model extensions lies in incorporating structural mechanics to capture particle mobility and deformation. Coupling the current phase-field transport framework with equations of particle motion [40] or continuum solid mechanics would enable a consistent description of capillary-driven compaction and rearrangement during the critical transition from initial film shrinkage to the pore-emptying regime. Moreover, this model extension will allow the simulation of the first drying stage, which is a critical step in the overall process and can create better initial conditions for binder distribution, thereby overcoming the limitations of the present study. To additionally account for internal porosity without explicitly resolving the small internal pores, the framework could be augmented by using homogenized approaches, such as Darcy-type approximations within the particle regions, cf., e.g., [41–43]. However, such multiscale model extensions are highly nontrivial and remain out of scope for the present work.

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M.W.: Methodology, software, validation, investigation, visualization, writing of the original draft, and review and

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DATA AVAILABILITY

The raw data underlying the plots, as well as additional data, are available from the corresponding author upon reasonable request. The related simulation software is commercially available [44].

APPENDIX: DERIVATION OF THE BINDER EQUATION

Consider the boundary value problem

$$\dot{c} := \frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = \nabla \cdot (D \nabla c) + q, \quad \mathbf{x} \in V, \quad (\text{A1})$$

$$(D \nabla c) \cdot \mathbf{n} = g, \quad \mathbf{x} \in \partial V. \quad (\text{A2})$$

Multiplying Eq. (A1) with test function ψ and integration over the volume V yields

$$0 = \int_V -\psi \dot{c} + \psi \nabla \cdot (D \nabla c) + \psi q \, dV \quad (\text{A3})$$

$$= \int_V -\psi \dot{c} + \nabla \cdot (\psi D \nabla c) - D \nabla c \cdot \nabla \psi + \psi q \, dV \quad (\text{A4})$$

$$= \int_V -\psi \dot{c} - D \nabla c \cdot \nabla \psi + \psi q \, dV + \int_{\partial V} \psi D \nabla c \cdot \mathbf{n} \, dS \quad (\text{A5})$$

$$= \int_V -\psi \dot{c} - D \nabla c \cdot \nabla \psi + \psi q \, dV + \int_{\partial V} \psi g \, dS. \quad (\text{A6})$$

This was done exploiting the Gauss divergence theorem and assuming the Neumann boundary conditions everywhere on ∂V . Consider a domain Ω with $V \subset \Omega$ we exploit

$$\int_V (\cdot) \, dV = \int_{\Omega} I(\cdot) \, dV \quad \text{and} \quad \int_{\partial V} (\cdot) \, dS = \int_{\Omega} \delta_S(\cdot) \, dV \quad (\text{A7})$$

with the indicator function I and the surface Dirac distribution δ_S , with $\nabla I = -\delta_S \mathbf{n}$. The weak form (A6) becomes

$$0 = \int_{\Omega} -\psi I \dot{c} - ID \nabla c \cdot \nabla \psi + \psi I q + \psi g \delta_S \, dV. \quad (\text{A8})$$

Looking at the second summand, we obtain

$$\begin{aligned} & \int_{\Omega} -ID \nabla c \cdot \nabla \psi \, dV \\ &= \int_{\Omega} -\nabla \cdot (\psi ID \nabla c) + \psi \nabla \cdot (ID \nabla c) \, dV \end{aligned} \quad (\text{A9})$$

$$= \int_{\Omega} \psi \nabla \cdot (ID \nabla c) \, dV. \quad (\text{A10})$$

The last step uses the divergence theorem, exploiting $\forall \mathbf{x} \in \partial\Omega : I = 0$. Therefore, we get

$$0 = \int_{\Omega} -\psi I\dot{c} + \psi \nabla \cdot (ID\nabla c) + \psi Iq + \psi g \delta_S \, dV \quad (\text{A11})$$

$$= \int_{\Omega} \psi [-I\dot{c} + \nabla \cdot (ID\nabla c) + Iq + g \delta_S] \, dV. \quad (\text{A12})$$

Localizing this gives

$$I\dot{c} = \nabla \cdot (ID\nabla c) + Iq + g \delta_S \quad (\text{A13})$$

using

$$I\dot{c} = \frac{\partial cI}{\partial t} + \mathbf{I}\mathbf{u} \cdot \nabla c - c \frac{\partial I}{\partial t}, \quad (\text{A14})$$

and we can rewrite Eq. (A13) as

$$\frac{\partial cI}{\partial t} = -\mathbf{I}\mathbf{u} \cdot \nabla c + \nabla \cdot (ID\nabla c) + Iq + g \delta_S + c \frac{\partial I}{\partial t}. \quad (\text{A15})$$

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