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Homogeneous solidification strategy for freeze-casting-assisted direct ink writing investigated by computational fluid dynamics modeling

Younghoon Son¹, Gyeongwan Lee¹ and Jungwook Choi^{1*}

Abstract

Freeze-casting-assisted direct ink writing (DIW) enables the fabrication of three-dimensional (3D) aerogel architectures with improved geometric versatility. However, multilayer deposition often produces inter-layer microstructural nonuniformity because the pre-frozen underlying layers introduce additional thermal resistance. To address this challenge, we investigated a homogeneous solidification strategy using a computational fluid dynamics (CFD) model. The framework incorporates a moving substrate boundary condition, constant-temperature thermal boundary, and interface tracking, while the nanocomposite ink properties are explicitly modeled as functions of temperature and shear rate. Using the CFD results and a custom digital image-processing algorithm, the freezing time required for the extruded filaments to fully solidify on a cold substrate is quantified. Based on the extracted freezing-time data, we determined the substrate-temperature pairs that yield identical freezing times across sequential layers. The resulting process map provides practical guidelines for mitigating layer-to-layer thermal variations and enables the reliable printing of aerogels with uniform inter-layer microstructures.

Keywords Direct ink writing, Freeze casting, Computational fluid dynamics, Microstructural uniformity

Introduction

Growing energy demand has driven the rapid expansion of research into supercapacitors and a broad range of electrochemical energy storage devices. Device performance is strongly governed by the electrode composition, geometry, microstructural design, and fabrication methodology [1, 2, 3]. Aerogel architectures, which provide high specific surface areas and interconnected three-dimensional porous networks, have attracted attention as

electrode platforms that facilitate efficient ion and electron transport. In particular, aerogel electrodes based on nanomaterials, such as carbon nanotubes (CNTs) and silver nanowires, have been reported to exhibit high electrical conductivity [4, 5]. Owing to their lightweight and high conductivity, aerogel-based electrodes are promising for use in microsensors and wearable devices [6, 7, 8].

However, aerogel fabrication has largely relied on mold-based processes [9, 10]. Such approaches impose limited geometric freedom and make it difficult to fabricate complex 3D architectures reproducibly, thereby motivating process strategies that preserve the structural merits of aerogel electrodes while improving geometric freedom and manufacturing reliability. In this context, direct ink writing (DIW) has emerged as an alternative

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fabrication method. DIW is a material extrusion 3D printing technique in which ink is dispensed through a nozzle and deposited layer-by-layer along a digitally defined toolpath. When integrated with freeze-casting, the ink can be printed onto a cold substrate to induce rapid cooling/solidification during deposition [11]. DIW offers several practical advantages, including (a) digital geometric control, (b) straightforward tunability of ink formulation, and (c) cost-efficient fabrication [12]. In addition, for DIW-processed electrodes, the electrode thickness (or height) can be readily increased by increasing the number of printed layers [13, 14]; for example, a 3D-printed graphene aerogel / MnO_2 electrode has been reported to exhibit a linear increase in areal capacitance with thickness, reaching a high value (e.g., 44.13 F/cm^2).

However, increasing the height of electrodes by stacking additional layers may compromise inter-layer microstructural uniformity, depending on processing conditions. The DIW process comprises a sequence of steps: ink preparation, extrusion, filament deposition, solidification, and post-processing, where the process parameters at each step can directly influence the final structure and device performance [15]. Accordingly, for multilayer architectures, it is critical to quantitatively control the inter-layer solidification conditions. In particular, when a cold substrate is used to induce cooling/solidification, the solidification behavior can vary from layer to layer, leading to inter-layer variations in porosity and microstructure. For example, the first layer is in direct contact with the cold substrate and freezes rapidly. In contrast, the second and subsequent layers are deposited onto an already solidified underlying layer that acts as a thermal barrier, slowing the freezing rate. Because ice crystal size and distribution depend on the freezing kinetics and thermal gradient, layer-dependent freezing can alter the resulting pore architecture of the final aerogel.

However, simulation studies aimed at quantitatively analyzing and designing the DIW process remain limited. In particular, most existing DIW simulations have primarily focused on polymer flow and deposition behavior under isothermal and room-temperature conditions, and therefore fail to capture the key freeze-casting physics under cold substrates, such as phase change, latent heat release, and temperature-dependent material properties. This limitation implies that simulations based on conventional DIW frameworks cannot reliably predict the ice-templating-driven pore formation, which is crucial for aerogel electrode manufacturing.

Previously, a computational fluid dynamics study that implemented nozzle motion to model the material extrusion additive manufacturing process was reported [16]. This work provided a detailed implementation procedure and considered non isothermal flow behavior. However,

the computational cost of this procedure is excessive; for example, simulations with adaptive time-stepping can require on the order of 9–18 h per run. Moreover, it does not directly implement the solidification mechanism that is crucial to the DIW-based freeze-casting of aerogels.

In this study, we developed a transient numerical model for DIW-based freeze casting that maximizes computational efficiency without introducing excessive process complexity. Specifically, instead of explicitly prescribing complex nozzle kinematics, the DIW process was modeled using a moving-boundary condition applied to the substrate. The cold substrate was implemented using a constant-temperature boundary condition, with the substrate temperature ranging from -120°C to 0°C . The temperature-dependent thermophysical properties of the Cellulose Nanofibers (CNF) (3 wt%) and CNT(0.25 wt%) composite ink were modeled using piecewise linear functions, whereas the viscosity was modeled as a shear-thinning property.

To investigate the multilayer deposition, two deposition scenarios were considered: direct writing onto the substrate (first layer writing) and writing onto a pre-solidified first layer (second-layer writing). The solidification time for each layer was quantitatively evaluated. This modeling strategy achieved a reduction in computational time of over 540 times relative to an overset-meshing-based approach (18 h to 2 min) while faithfully accounting for phase change and cold-temperature conditions, thereby offering a practically deployable and physically representative simulation framework.

Subsequently, we computed the solidification time for each layer during the first layer and second-layer writing. We derived a layer-specific substrate temperature to promote inter-layer microstructural uniformity, which we propose as a practical process guide. This study demonstrates the feasibility of designing aerogel electrodes with uniform microstructures, even under multilayer deposition. Moreover, the proposed layer-wise process guide is expected to serve as a practical design guideline for a broad range of applications, including microsensors and energy storage devices.

Methods

Material

In this study, an aqueous nanocomposite ink composed of 3 wt% cellulose nanofibers (CNFs) and 0.25 wt% carbon nanotubes dispersed in deionized water was selected as a representative model system for freeze-casting-assisted DIW. CNFs are nanomaterials that can form an interconnected 3D network during gelation [17], preserve a porous architecture through freeze-drying, provide mechanical support, and serve as a structural template for aerogel formation. The CNTs provide electrical conductivity within the CNF-based network,

thereby activating the electrical functionality of the final electrode. Because this CNF/CNT ink system combines structural stability with electrical conductivity, it is well-suited for aerogel electrodes and microsensor-type applications; therefore, it was adopted as the model ink to analyze the freeze-casting process.

Overview of freeze casting assisted DIW and CFD modeling

Freeze casting-assisted DIW is a patterning process in which ink is deposited onto a cold substrate and rapidly frozen, thereby immediately fixing the printed geometry (Fig. 1a). The steep temperature gradient between the nozzle and substrate induces a solvent phase transition within the deposited filament and promotes ice nucleation and growth. The resulting ice crystals serve as sacrificial templates and are removed by freeze-drying, yielding a porous aerogel network. This process offers high geometric freedom and enables precise control of the electrode structure through digital modeling; the electrode microstructure can be tuned by adjusting the ink composition and freezing conditions.

In this study, we developed a CFD framework to simulate phase-change-coupled flow in cryogenic DIW. To avoid the excessive computational burden associated with resolving the complex transient motion of the nozzle, the nozzle was kept stationary, and a substrate translation of 10 mm/s was imposed as a moving boundary condition to reproduce continuous extrusion and deposition.

A constant temperature boundary condition was applied to the substrate to represent the cryogenic environment for each iteration. This idealization is justified by the small thermal energy delivered to the substrate by an individual filament (~ 0.6 mm in a diameter) relative to the much larger thermal mass of the copper substrate, combined with the high thermal conductivity of copper (~ 400 W/m·K) and active feedback control of the thermoelectric cooler, which together suppress local temperature deviations during deposition. The air-ink interface

was traced using a volume of fluid (VOF) approach, with air treated as the primary phase and ink as the secondary phase, enabling direct visualization of the DIW deposition process (Fig. 1b). This simplified modeling strategy allows the precise tracking of the liquid fraction evolution as the filament contacts the cold substrate and cools, while substantially reducing the computational cost and capturing the key solidification behavior of freeze casting.

Rheological and thermophysical properties of CNF/CNT ink

The rheological behavior of the ink is a key factor governing the print stability in the DIW process. In this simulation, a non-Newtonian power-law model was employed to represent the viscosity variations [18].

$$\eta = K\dot{\gamma}^{n-1} \quad (1)$$

where η is the apparent viscosity, K is the consistency index, $\dot{\gamma}$ is the shear rate, and n is the power-law exponent. To reflect the flow characteristics of the 3 wt% CNF aqueous suspension used in this study, the power-law parameters reported by Nazari et al. (2016) were adopted and implemented in the numerical model (Fig. 2a). In particular, a low value of $n = 0.15$ indicates strong shear-thinning behavior. As a result, the ink viscosity decreases significantly under high-shear conditions inside the nozzle, which facilitates smooth extrusion without clogging, while recovering after deposition when the shear is removed, thereby mitigating gravity-driven shape collapse and improving shape retention.

To accurately analyze the freezing behavior on a cold substrate, it is essential to account for the sharp changes in the thermophysical properties caused by temperature changes and the phase change of water. Accordingly, the ink density and specific heat were modeled using a mixing rule based on the mass fractions of the constituents (water: 96.75 wt%, CNF: 3 wt%, CNT: 0.25 wt%) (Fig. 2b, c). Because these properties are extensive,

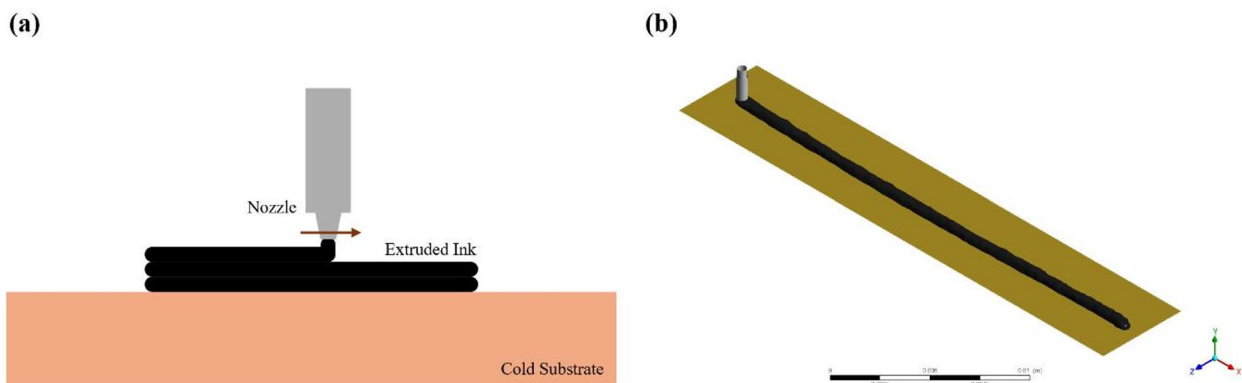


Fig. 1 Overview of DIW freeze casting: **a** Schematic of DIW freeze casting process. **b** CFD modeling of DIW for freeze casting contoured along the volume fraction of ink

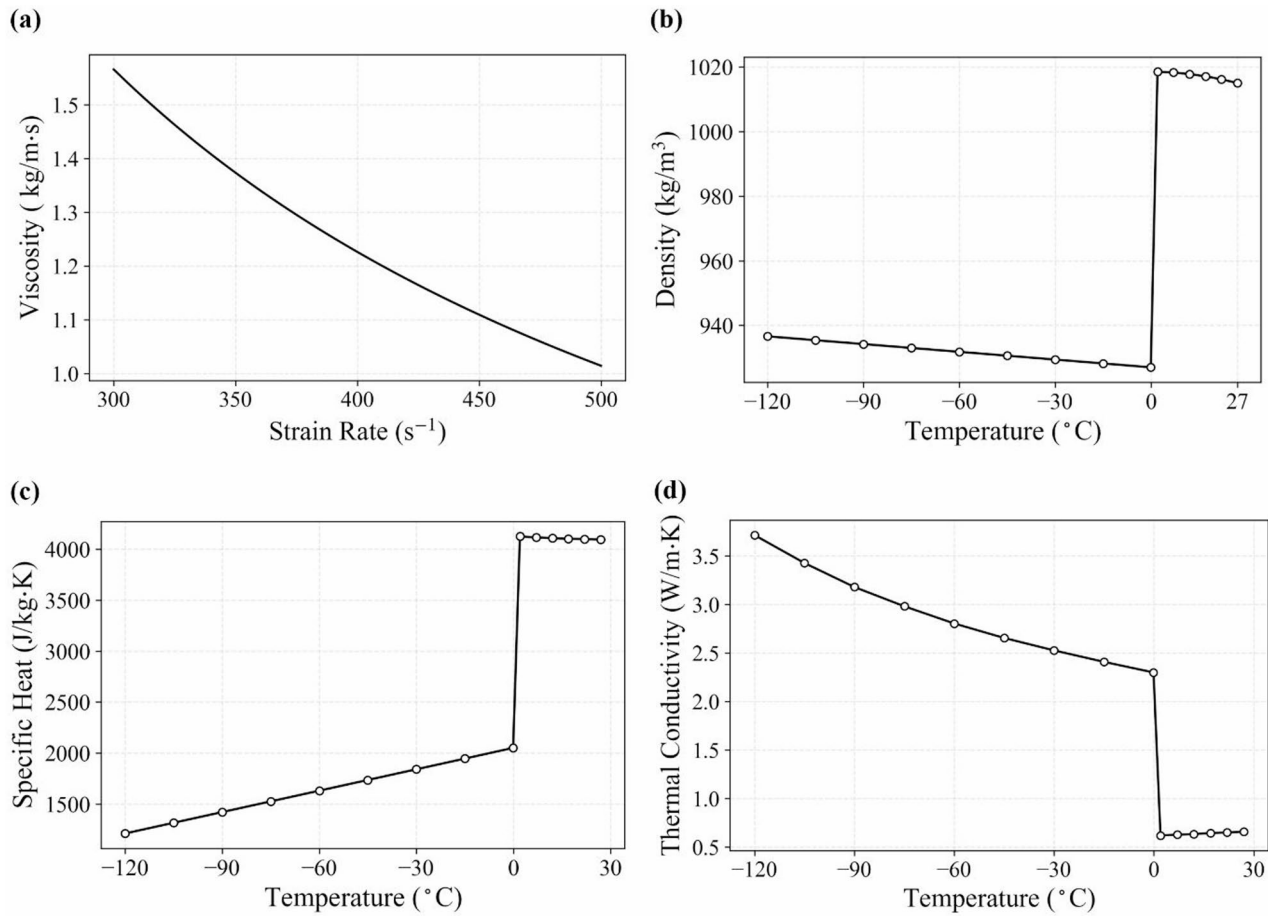


Fig. 2 Properties of CNF/CNT suspension: **a** Viscosity as a function of strain rate, fitted with the non-Newtonian Power-law model ($k=199.7$, $n=0.15$). The suspension exhibits shear-thinning behavior. **b–d** Temperature-dependent thermo-mechanical properties of the CNF/CNT suspension: **b** density, **c** specific heat, and **d** thermal conductivity. Significant transitions in properties are observed near 0 °C due to phase change

the contribution of nano additives can be incorporated by applying the rule of mixtures weighted by the mass fraction of each component [19]. In addition, because the specific heat and density of water and ice vary with temperature, temperature-dependent property tables for ice and liquid water were used [20]. The properties of the composite ink were calculated using the following rule of mixtures:

$$A_{mixture} = 0.9675 A_{water} + 0.03 A_{CNF} + 0.0025 A_{CNT} \quad (2)$$

where A denotes either the density (ρ) or the specific heat (C_p) of the corresponding component.

In contrast, the thermal conductivity was evaluated in Fig. 2d, using the Hamilton–Crosser (H–C) model, which accounts for the behavior of high-aspect-ratio fibers, because the heat transfer network formed by nanoparticles cannot be predicted quantitatively in a straightforward manner. To represent a dispersion containing 0.25 wt% CNT, we first adopted the experimental correlation for the thermal conductivity of a CNT–water nanofluid

reported by Estelle et al. (2015) [21] and defined this as a new base fluid. As the CNTs form a percolated network of interconnected particles within the suspension, their effect had to be incorporated directly from the experimental data. Using this CNT-based base fluid, the contribution of 3 wt% CNF was then combined using the H–C model [22], and the effective thermal conductivity of the ink, k_{eff} , was calculated as follows:

$$k_{eff} = k_{bf} \left[\frac{k_p + (n-1)k_{bf} - (n-1)\phi(k_{bf} - k_p)}{k_p + (n-1)k_{bf} + \phi(k_{bf} - k_p)} \right] \quad (3)$$

where k_{eff} , k_p , and k_{bf} denote the effective thermal conductivity of the ink, the dispersed particles, and the base fluid, respectively, and ϕ is the CNF volume fraction. The H–C model also includes a shape factor n that reflects the particle geometry. In this study, $n=6$ was used to account for high-aspect-ratio CNF structure [23]. Thus, the entangled 3D CNF matrix within the ink represents an effective thermal pathway that promotes heat

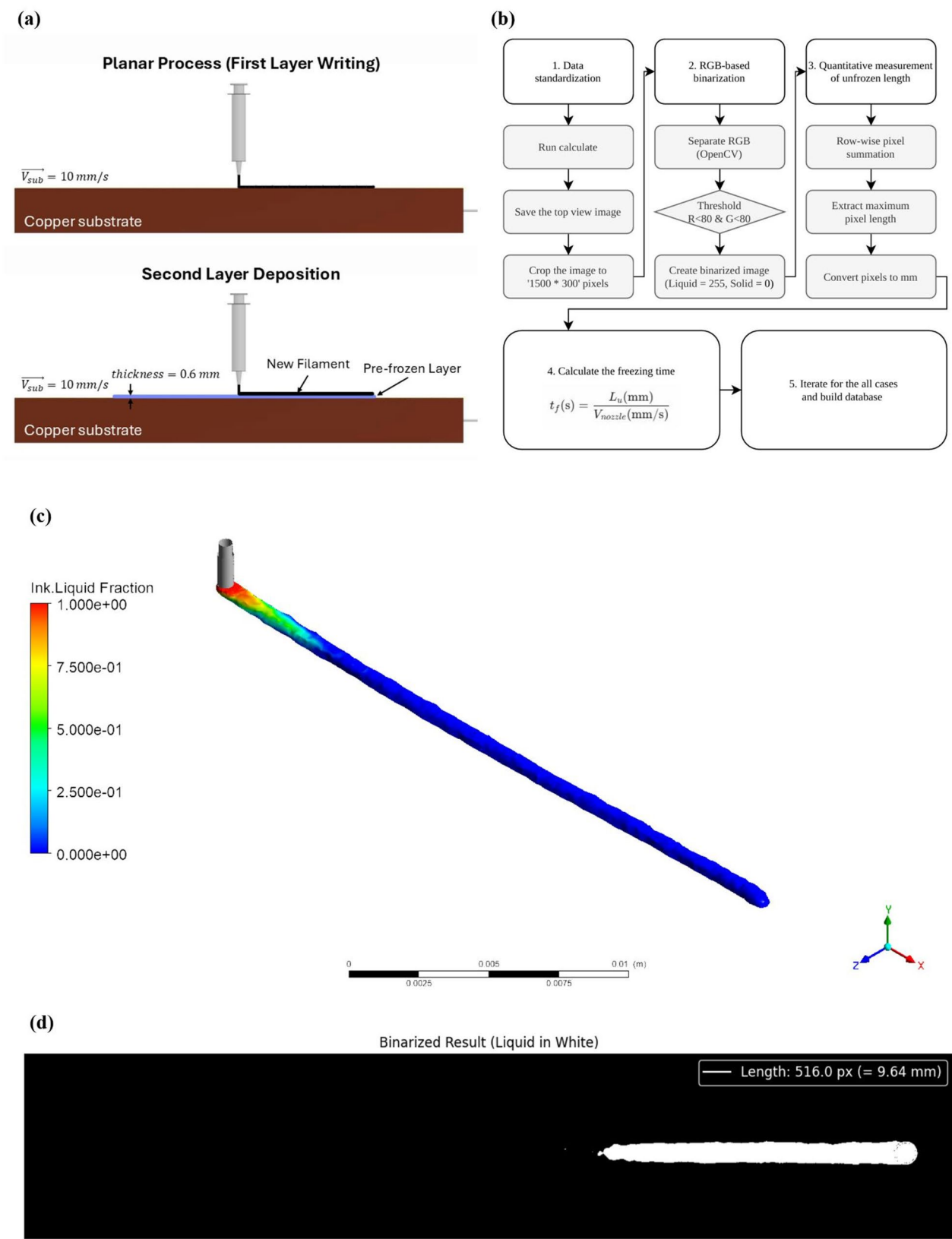


Fig. 3 (See legend on next page.)

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Fig. 3 Simplified numerical modeling and digital image-based analysis for the DIW process: **(a)** Schematic illustration of the modeling strategies. The planar patterning (one layer writing) directly on the copper substrate and the second-layer writing utilizing a 0.6 mm-thick “wall thickness” condition to simplify the thermal resistance of the pre-frozen layer. **(b)** Image processing flow chart: An algorithm implemented using Python’s OpenCV(CV2) library. **(c)** 3D overview of the CFD simulation: Representative liquid fraction contour showing the phase transition of the extruded filament. The blue region indicates the completion of the solidification process, where the liquid fraction reaches zero. **(d)** Post-processed binarized result: A binary image where white pixels represent the remaining liquid phase. The measured length of the unfrozen line is automatically calculated and displayed in the upper-right corner to determine the final freezing time

transfer, thereby increasing the overall thermal conductivity of the ink.

As a result, the present property model captures both the contribution of nano additives and the temperature- and phase-change-dependent behavior of the matrix, thereby overcoming the limitations of conventional room-temperature polymer-based simulations. Beyond the specific CNF/CNT/water system examined here, the same procedure—rule-of-mixtures with solvent-specific thermophysical tables (Eq. 2), Hamilton–Crosser modeling for high-aspect-ratio fillers (Eq. 3), and Power-law fitting of measured rheology (Eq. 1)—is expected to enable recalibration of the framework for other aqueous or solvent-based nanocomposite inks used in freeze-casting-assisted DIW.

Method for analyzing solidification behavior

We performed simulations by varying the substrate temperature from $-120\text{ }^{\circ}\text{C}$ to $0\text{ }^{\circ}\text{C}$ in $15\text{ }^{\circ}\text{C}$ increments. These simulations were conducted for two cases: (i) extrusion of the first layer directly onto a copper substrate and (ii) extrusion of the second-layer onto a pre-frozen layer. In multilayer DIW processes, newly extruded ink is inevitably deposited on top of an already frozen underlying layer. To reproduce this in our numerical model, the second-layer simulation assumed a pre-existing frozen ink layer with a thickness of 0.6 mm (Fig. 3a). The use of a fixed thickness reflects the underlying geometry of DIW: filament height is set by the nozzle inner diameter and the prescribed layer height (both 0.6 mm here), whereas the substrate temperature affects only the freezing kinetics. This was confirmed by additional CFD simulations of filament cross-sections at substrate temperatures of -15 , -30 , -45 , and $-60\text{ }^{\circ}\text{C}$, which yielded heights of 0.561, 0.607, 0.586, and 0.612 mm, respectively (0.591 ± 0.020 mm; Fig. 4). This pre-frozen layer acts as a thermal resistance that impedes heat transfer between the newly deposited ink and the cold substrate. Therefore, even under the same substrate temperature conditions, the freezing behaviors observed in the first and second simulations exhibited completely different characteristics.

To analyze the freezing behavior of the extruded ink quantitatively, we introduced a digital image-based measurement algorithm. Once extruded from the nozzle, the ink filament undergoes a gradual phase change over time owing to transient heat transfer on the substrate. The

region near the nozzle remained mostly liquid because it had minimal time to cool after the extrusion. Farther downstream, the filament is in contact with the cold substrate for a longer period; thus, a larger portion becomes frozen. This spatiotemporal behavior can be interpreted from the 3D CFD results by tracking the location where the liquid fraction converges from 1 to 0 (i.e., the progression of the solidification front). Accordingly, Fig. 3c shows the solidification propagation as liquid-fraction contours.

From a process-design perspective, these two quantities are important. The first is the duration for which the filament remains in the liquid. The other is how long (in time) the liquid region remains unfrozen. To measure this, we built a digital image-processing algorithm using the Python OpenCV library, as shown in Fig. 3b. For each simulation condition, a top-view image was first extracted. We then resized all the images to the same resolution of 1500×300 pixels. Next, we performed RGB-based binarization to isolate the unfrozen regions from the liquid fraction contours. In the contour (Fig. 3c), the solid region (liquid fraction close to zero) appears dark purple. Therefore, the threshold was tuned accordingly. The liquid region was classified as white (255), while the other regions were converted to black (0).

Figure 3d shows an example of a binarized image obtained using this procedure, where the white region corresponds to the unfrozen region. The algorithm scans each row of the image and sums the number of white pixels in each row. It then finds the row with the maximum white pixel count and defines this value as the pixel length of the longest remaining liquid segment. Next, we converted the pixels to real physical lengths using a scale bar (e.g., $1\text{ mm} = 53.5$ pixels). Finally, we divided this unfrozen length by the nozzle’s relative travel speed (10 mm/s) to estimate the freezing time, that is, the time from deposition until the location became fully solidified. Using this digital image-based method, we transformed the 3D CFD results from a simple visualization into quantitative data, enabling a direct comparison of freezing times across different substrate temperatures and stacking conditions. This analysis provides the key basis for deriving (in the following sections) the substrate-temperature-dependent freezing behavior, freezing-delay effect caused by a pre-frozen layer, and process guidelines for achieving a uniform microstructure between layers. Although direct experimental validation is reserved for future work, the

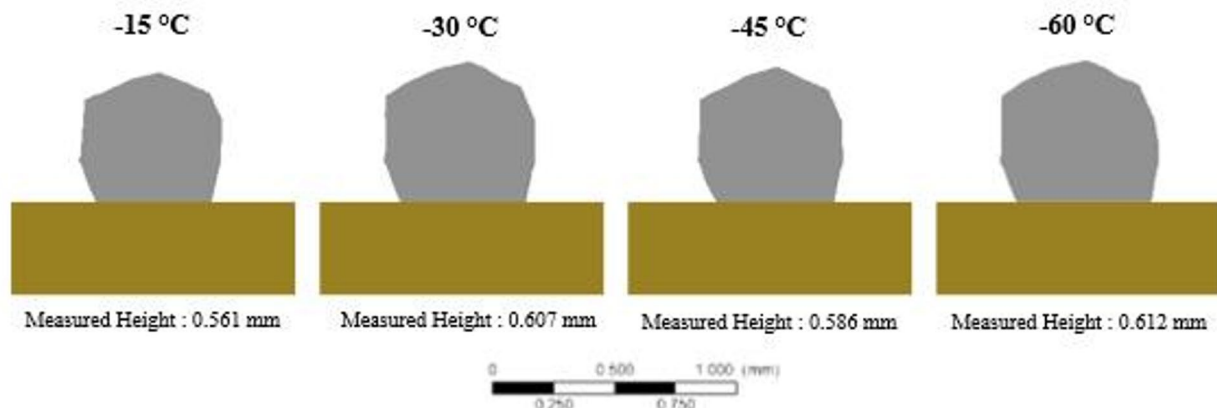


Fig. 4 CFD-predicted cross-sectional contours of the deposited filament at substrate temperatures of $-15\text{ }^{\circ}\text{C}$, $-30\text{ }^{\circ}\text{C}$, $-45\text{ }^{\circ}\text{C}$, and $-60\text{ }^{\circ}\text{C}$

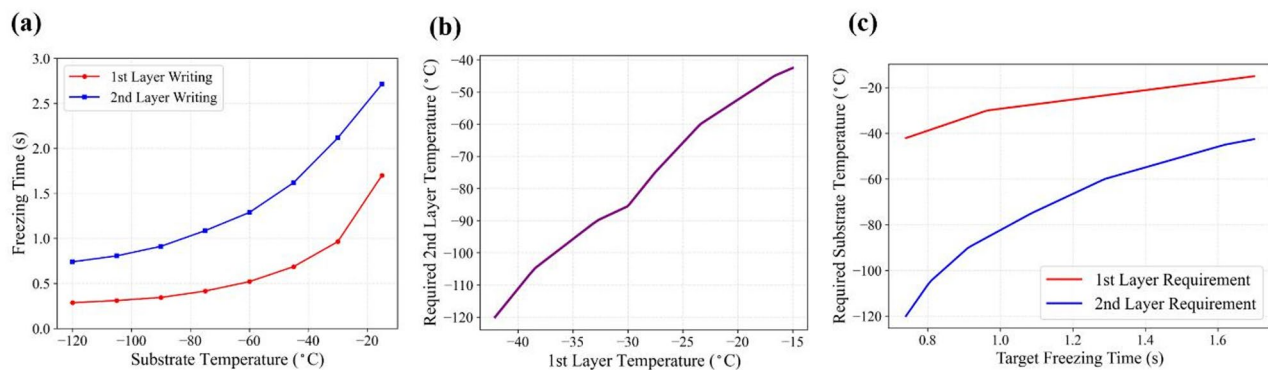


Fig. 5 Process guidelines for achieving homogeneous microstructures in double-layer DIW: **a** Substrate temperature-dependent solidification kinetics. The measured freezing time profiles for the first and second-layer. **b** Layer-wise temperature calibration curve. A process guide derived from the iso-solidification strategy. This curve indicates the necessary substrate temperature adjustment for the second-layer corresponding to the first layer's processing temperature to maintain consistent solidification dynamics throughout the multilayer DIW. **c** Optimized process parameter guidelines. The required substrate temperatures for both layers plotted as a function of the desired target freezing time. This chart serves as a practical guide for controlling solidification dynamics; by selecting a specific cooling rate, the exact required temperatures for each layer can be determined

reliability of the present framework is supported by the prior experimental validation of its constituent sub-models—the VOF-based filament deposition [24], the Power-law shear-thinning flow [25], and the enthalpy–porosity solidification [26]—each previously benchmarked against experimental data.

Results and discussion

To fundamentally resolve the microstructural nonuniformity caused by the thermal resistance of the underlying layer during multilayer stacking, we proposed a homogeneous solidification strategy that ensures the same solidification time for each layer. Figure 5a shows a visual and quantitative analysis of the minimum solidification time required as a function of the substrate temperature for (i) first-layer writing and (ii) second-layer writing. By cross-analyzing these two plots, we can inversely derive the operation of the substrate temperature combination that causes the two layers to solidify at the same rate.

For example, when the first layer is printed at a substrate temperature of $-30\text{ }^{\circ}\text{C}$, the ink takes about 0.9 s to completely freeze. When the second-layer is deposited on top of a pre-solidified first layer (0.6 mm thick), the lower layer adds thermal resistance. To overcome this resistance and achieve the same freezing time of 0.9 s, the substrate temperature must be reduced to approximately $-80\text{ }^{\circ}\text{C}$ to compensate for the reduced heat removal. The temperature curve in Fig. 5b maps the thermodynamic correction relationship. Using this curve, the required corrected temperature for the second-layer can be directly determined from the processing temperature of the first layer.

Furthermore, we can derive process guidelines that specify the substrate temperatures for the first and second-layers based on the user-defined target freezing time (Fig. 5c). These guidelines can serve as practical process maps for manufacturing. Moreover, they can be used when an operator wishes to control the pore size of a multilayer aerogel electrode to be uniform in the

thickness direction. Although the present map is derived for a double-layer configuration, the required substrate temperature correction is expected to scale approximately linearly with layer count, as each additional frozen layer of identical thickness contributes an equal increment of thermal resistance. In practice, however, the process is limited to a small number of layers because cumulative weight and reduced heat removal beyond approximately three layers lead to structural sagging and loss of shape fidelity before complete solidification.

The physical significance of matching solidification times across layers can be understood through the well-established relationship between freezing kinetics and pore architecture in freeze casting: the freezing front velocity v scales inversely with solidification time, and the resulting pore size λ follows a power-law dependence $\lambda \propto v^{-n}$ [27, 28]. Consequently, shorter solidification times produce finer pores after freeze-drying, while longer times yield coarser ones. Layer-dependent solidification times in multilayer DIW would therefore translate directly to layer-dependent pore sizes—and to layer-dependent electrochemical behavior, since pore size simultaneously governs the specific surface area available for ion adsorption and the ion-transport pathway through the electrode [29]. By matching solidification times across layers, the proposed process map matches the pore size distribution along the electrode thickness, providing a physically grounded route toward consistent electrochemical performance and a solution to the long-standing challenge of quality variation with build thickness in freeze-casting-assisted DIW. It is noted, however, that matched solidification times ensure pore size uniformity but do not fully eliminate residual differences in thermal gradient between the directly cooled first layer and the thermally insulated second layer, which may introduce secondary effects on pore wall morphology. In aqueous CNF/CNT systems, such effects are expected to be limited because the lamellar pore architecture is predominantly governed by the intrinsic anisotropic growth of ice crystals rather than by the thermal gradient magnitude [28, 30], and pore orientation is preserved in both layers as the heat flux direction remains identical; nevertheless, a quantitative assessment is left as future work.

Conclusion

To model the transient phase change in the freeze-casting-assisted DIW process, we developed a CFD framework using a moving boundary condition for the substrate, a constant-temperature boundary condition, and VOF-based interface tracking. We incorporated the power-law rheology and temperature-dependent properties of CNF (3 wt%)/CNT (0.25 wt%) ink over a substrate temperature ranging from $-120\text{ }^{\circ}\text{C}$ to $0\text{ }^{\circ}\text{C}$.

Using the simulation and our image processing algorithm, we determined that a 0.6 mm pre-frozen layer acts as a thermal resistance and delays the solidification time of the second-layer. To address this issue, we proposed a homogeneous solidification strategy (Fig. 5). The proposed process map can be used as a guideline for the quantitative design of the microstructural homogeneity of double-layer aerogel electrodes. Finally, the simulation approach and process map construction methods in this study can be applied to supercapacitor and transducer research employing multilayer electrodes.

Abbreviations

3D	Three-dimensional
DIW	Direct ink writing
CNF	Cellulose nanofiber
CNT	Carbon nanotube
wt%	Weight%
CFD	Computational fluid dynamics
VOF	Volume of fluid
H-C	Hamilton-Crosser

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Author contributions

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Data availability

The datasets used and/or analyzed during the current study are available from the corresponding author on request.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare no competing interests.

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