Accurate Prediction of Melt Electrowritten Laydown Patterns from Simple Geometrical Considerations

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Melt electrowriting (MEW), an electrohydrodynamic additive manufacturing technology, can produce polymer structures with micrometer resolution. Unlike conventional direct writing of fluids, the laydown pattern often deviates from the programmed one, due to a lag in the electrified jet. MEW of increasingly complex patterns currently entails a cumbersome trial-and-error procedure of adjusting printing parameters to ensure good accuracy. This issue is addressed with a geometrical model that predicts the expected printed fiber path, based on the coded trajectory, critical translation speed, and the collector speed.

The influence of the process parameters on the jet lag is investigated and a general equation for the lag computation is implemented into the model. The presented model can evaluate the printing result up to ten times more accurately than the designed collector movement path and thus assist in identifying printing defects and adjustment of the printing G-code in advance.

1. Introduction

Additive manufacturing (AM) has profoundly altered the manufacturing landscape in recent decades and expanded the rapid-prototyping boundaries of complexity, resolution, and production speed.[1] Nevertheless, every AM technology has its limits in terms of realizable feature sizes and shapes, restricting its application for various products. A difference between the design and the actual produced part varies from method to method but can be quite noticeable.[2,3] One such AM technology susceptible to this effect is melt electrowriting (MEW), where an electrohydrodynamically stabilized molten polymer jet is direct-written at low flow rates onto a collector.[4] Using repeated layer deposition, these small-diameter fibers can be assembled using AM principles for the fabrication of microscale structures.[5]

While the fiber diameter for optimized MEW can be accurately set and stay constant throughout a print (coefficient of variation <5%),[6] there remains a challenge in precisely predicting the fiber placement that necessitates manual parameter or printing path adjustment to obtain the desired structure.[7] This lack of predictability is, in turn, limiting the research and potential industrial utilization of the technology. The predictive ability is especially compromised when printing curved elements, that are increasingly important for mimicking the morphology and mechanical properties of the living tissues.[8] In this context, most MEW research currently uses linear laydown patterns, as this morphology is least affected by changes in the direct-writing direction.

There is a clear discrepancy between the programmed stage movement and the deposited fiber shape (i.e., laydown pattern) most noticeable with curved MEW structures, as well as at edges where the linear portion of the direct-written fiber ends. The main reason behind this discrepancy is the gap between the nozzle and collector, which is bridged by the viscoelastic jet. Figure 1A schematically shows how the relative movement of the nozzle to the collector results in a jet lag, a line segment between the nozzle position (NP) and the jet contact point (JCP) on the collector (Figure 1A,B). Whereas the NP velocity (VNP) corresponds to the actual collector velocity, the JCP velocity (VCJP), that represents the actual fiber deposition velocity, will often be different from it.[7,8] Due to the MEW jet lag, the changes in direction and speed of the collector movement are not followed by the same changes in the printed fiber pattern (Figure 1C).[9,10] Compounding this issue is that the lag is variable and sensitive to changes in both instrument (i.e., applied pressure/voltage)[11] and environmental (i.e., temperature/humidity) parameters. This is the reason why the elimination of fiber pulsing during MEW is so important for the reproduction of samples.[6]

One approach to this fiber-placement problem is by minimizing the MEW jet lag.[12] Decreasing the collector speed to
just before liquid rope coiling starts, decreases the difference between the $V_{NP}$ and $V_{JCP}$ and improves the predictability of the print. This collector speed value, also known as the critical translation speed (CTS), closely corresponds to the speed of the jet near the contact point, when it is falling on a stationary collector. Since the CTS is sensitive to the process and environmental parameters, it needs to be determined prior to printing in order to avoid entering the liquid-rope coiling region. Even when the speed is properly adjusted, a lag can still exist (Figure 1D). This means that at the corners of the printing path, the jet will bend on the collector at a specific radius, depending on its viscoelastic properties. Furthermore, direct writing at the CTS limits the ability to vary the fiber diameter during a print and increases the printing time.

The first fiber laydown pattern is especially important, since the first layer and further fiber layers provide an “autofocusing effect” where an already printed fiber will attract the next depositing fiber stronger than onto the flat collector surface. Due to curved regions and corners, the fiber path changes from layer to layer, even when the jet lag is minimized by having collector speed close to CTS.

A simplified geometrical representation of an MEW jet, while collector speed is above CTS is a catenary with contact points on the nozzle and collector. This approximation has been described and modeled previously for solution electrohydrodynamic 3D printing. In the case of MEW, where the jet is much thicker and more viscous, it might not be possible to apply the same approach and neglect the resistance to bending. Changing mechanical properties and charge distribution along the length of the jet (through cross-section decrease, heat and charge dissipation through the air and collector surface) also have to be taken into account.

However, some work has been performed on modeling melt electrospinning and various types of electrohydrodynamic printing; those studies have focused on the jet initiation, diameter change in the noncoiling region of the electrified jet, jet buckling, and motionless printing. In this study, we use a universal parameter of MEW (i.e., the CTS), to take many of the confounding parameters out of consideration to provide an accurate method to predict printed shape.

2. Results and Discussion

2.1. Geometrical Model Development

As an alternative to physical modeling, a ready-to-use geometrical model has been developed, that utilizes a programmed path, collector speed and the CTS. The model relies on the jet lag calculation and does not predict the buckling patterns and is therefore valid only when the collector speed is above the CTS. When the collector is moving with a constant speed and $V_{NP}$ and $V_{JCP}$ are equal (Video S1, Supporting Information), the jet can be considered stable and the lag has a characteristic magnitude, further referred to as stable lag ($L_{stable}$). It has been previously shown that with the increasing collector speed, the $L_{stable}$ grows with a rate that depends on other processing parameters. However, in order to be able to readily calculate the lag for any speed, an analytical equation needs to be found. When a collector speed changes, the jet lag quickly approaches its new characteristic length (Video S2, Supporting Information). During this transition, $V_{JCP}$ can be higher (lag is decreasing) or lower (lag is increasing), than the current collector speed.

The complete algorithm of the lag calculation is presented in Figure 2. For the computation of a printed path, the given G-code path is broken into a sequence of equidistant nozzle
positions, represented by NPs, connected by unit movement vectors (Figure 1E). The distance between the points, unit collector movement, is proportional to the collector speed and is covered by the collector within unit step time ($\Delta t$). The density of the points can be arbitrarily chosen but should be high enough to correspond to the expected model accuracy. At the start of the calculation, the coordinates of the starting positions of the nozzle and the jet contact point, NP$_1$ and JCP$_1$ are given. They can either coincide (collector at rest) or be at an arbitrary lag distance (moving collector). In order to calculate the next fiber contact point, JCP$_{i+1}$, a vector connecting the current JCP$_i$ and the next NP$_{i+1}$, termed as ray, is drawn. For a constant linear motion, the length of a ray (e.g., Ray 2 in Figure 1E) is equal to the algebraic sum of the unit collector movement and the current lag. Change of the collector movement direction means that the current lag and the unit NP movement vectors are not collinear, and the ray becomes shorter. The difference between the following ray length and the current lag ($L_i$) magnitude can be divided by the step time to provide a value, termed as moving away speed ($V_{MA}$)

$$V_{MA} = \frac{RL_i - L_i}{\Delta t}$$ (1)

The $V_{MA}$ is the instant speed of the nozzle relative to the JCP (Figure 1E) and, thus, defines the tension in the jet and thereby the lag.

When the chosen unit collector movement is small, compared to the minimal fiber curvature radius, it is assumed that the jet projection on the collector is always a straight line and the next contact point JCP$_{i+1}$ shall lie on the ray connecting JCP$_i$ to NP$_{i+1}$ (Figure 1C,E; Videos S3 and S4, Supporting Information). The new lag length should not exceed the ray length. Its position depends on how strongly the jet is pulled by the collector, i.e., on the value of the $V_{MA}$. To directly determine if the jet is stretched at all or is buckling, the speed ratio (SR) is obtained

$$SR_{i+1} = \frac{V_{MA}}{CTS}$$ (2)

The $L_{stable}$ value can then be calculated, inserting the speed ratio value into an empirical function, which can theoretically be unique for each set of printing parameters.

In general, once the lag is above zero, JCP must be always moving toward the NP, since the viscoelastic jet is constantly being elongated under the tension, caused by electrical and mechanical forces. When a segment of the jet lands on the
collector the current lag and the $V_{MA}$ define whether the new lag value is smaller or bigger than the current one. If a jet is sufficiently stretched horizontally for the lag to appear ($V_{MA} > CTS$), it is increasing or decreasing its length until it reaches the characteristic $L_{stable}$ value for the corresponding speed ratio. After $L_{stable}$ is reached, speed of the jet contact point is equal to the collector speed. Until then, the fiber is deposited with a $V_{JCP}$, that is calculated by substituting the current lag values $L_i$ into Equation (5). The JCP$_{i+1}$ is then positioned on a ray at a distance equal to $V_{JCP} \cdot \Delta t$ from JCP$_i$ and the lag, calculated as follows is termed as “falling” jet lag ($L_{FJ}$)

$$L_{i+1} = L_{FJ} = Ray_{i+1} - V_{JCP} \cdot \Delta t$$

(3)

When $V_{MA}$ is decreasing through either collector deceleration or direction change below the CTS, the tension in the jet is decreasing and lag is getting smaller. The JCP$_{i+1}$ approaches the NP$_{i+1}$ until the jet becomes vertical and lag disappears (minimal lag $\varepsilon$ (Figure 1D) is ignored).

2.2. Jet Lag and Contact Point Speed Measurement

As expected, an increase in collector speed leads to the increase in the $L_{stable}$. This growth is reduced for high speed ratios but is very steep up to $6 \times CTS$, meaning that the lag magnitude is sensitive to collector speed and printing direction changes as well as the changes in the CTS (Figure 3A, B).

During the lag measurement, the possibility of a jet to exceed its $L_{stable}$ values was observed, while accelerating to a speed, higher or equal to $6 \times CTS$ (Video S5, Supporting Information). This observation with a likely deleterious effect was neglected in the model, since the high-speed (above $6 \times CTS$) scenario was not often used in routine MEW research to date.

For the implementation into the model, a power function of the type $a - a \cdot x^b$ was picked to fit the experimental data. It has good fitting behavior in the used speed ratio range (adjusted $R^2 = 0.9811$) but implies the existence of a maximal possible lag value, above the highest $17 \times CTS$ collector speed used here. For default MEW processing conditions, the resulting equation was

$$L_{stable} = 10.02 \cdot \left(1 - SR^{-0.304}\right)$$

(4)

In reverse, for every lag value $L$ a characteristic speed ratio should exist, indicating the corresponding instant contact point speed

$$V_{JCP} = CTS \cdot \left(1 - L / 10.02\right)^{1/0.304}$$

(5)

The calculated/derived $V_{JCP}$ values were compared to those experimentally obtained (Figure 3C,D) during the collector deceleration (Video S6, Supporting Information). In case of a collector full stop, the measured $V_{JCP}$ values (Figure 3C) approximate the derived $V_{JCP}$ curve when a lag is below 1.5 mm and deviate from this for larger lag values up to approximately 5 mm. When the collector speed was decreasing to a nonzero value, from $17 \times CTS$ to $5 \times CTS$, $5 \times CTS$ to $2 \times CTS$, and $2 \times CTS$ to $1.25 \times CTS$ (Video S2, Supporting Information;

![Figure 3](https://www.advancedsciencenews.com)
Figure 3D), the $V_{\text{JCP}}$ values were better represented by the derived $V_{\text{JCP}}$ curve than by the fitted equation to the collector full stop $V_{\text{JCP}}$ data (adjusted $R^2 = 0.9324$ and 0.101, respectively). A possible reason behind the higher $V_{\text{JCP}}$ measured during the full stop in the lag range between 1 and 5 mm is the absence of the tension, caused by the collector movement. This allows the electrostatic force to induce a faster collapse of the jet on the collector with the $V_{\text{JCP}}$ exceeding the values, corresponding to collector speeds, characteristic for $L_{\text{stable}}$ values. For the implementation into the model it was decided to utilize Equation (5), which also minimizes the amount of the necessary input data.

The differences between the contact point speed, obtained for the full stop and speed transitions show the dependence of the jet dynamics on the collector acceleration and deceleration rates. In this study, it was not possible to directly match the instant collector speed and acceleration values to the measured $V_{\text{JCP}}$. The results of the model validation under standard MEW conditions (see the Experimental Section) showed, however, that it was sufficient to use the derived $V_{\text{JCP}}$ Equation (5) without adversely affecting the prediction quality. Nevertheless, for the prints containing multiple stop points the empirical $V_{\text{JCP}}$ fit curve based on the full stop measurements (Figure 3C) can be more applicable or used in combination with the derived equation.

The variety of printing setups and conditions for MEW makes it necessary to investigate whether Equations (4) and (5) are usable at different printing settings. Figure 3E,F demonstrates how the jet lag and $V_{\text{JCP}}$ react to the changes in the key process parameters. A remarkable jet lag increase while printing with a larger collector distance first of all suggests a bigger error between the programmed path and the printed fiber position, thus providing an additional incentive for a prediction model for larger gaps. However, the $L_{\text{stable}}$ in the lower speed range seems to noticeably deviate from the original fit curve only in case of a gap increase. The similarity in the jet fall behavior is even more notable as differences between the $V_{\text{JCP}}$ fit curves can mostly be addressed to the difference in corresponding CTS values (Table S1, Supporting Information). The mutual resemblance between the lag and $V_{\text{JCP}}$ fit curves, obtained at different parameter values suggests that it is possible to use Equations (4) and (5) for any MEW parameter combination by adjusting numerical coefficients of the power function. This procedure can help to avoid optical measurement of the jet but requires further validation.

Importantly, the obtained results are relevant for the specific poly($\varepsilon$-caprolactone) (PCL) only, which is also the current “gold-standard” polymer used for MEW to date. Other materials, that can be processed by MEW, have different viscoelastic and electrical properties that might affect the jet behavior, making the power model in Equations (4) and (5) less accurate. However, it is possible to build on this study to incorporate the increasing number of polymers used for MEW in the future.

2.3. Model Validation

2.3.1. Single Layer Print

The proposed geometrical model allows the trajectory prediction of a first layer with an average $x$–$y$ placement error below $70 \, \mu$m and maximal deviation below $200 \, \mu$m (Figure 4A,B). Although the measurements were done on a limited set of a test shapes, it covers the basic geometrical elements of the majority of MEW scaffolds. With increasing collector speed, the maximal error increases whereas the average prediction error does not grow significantly. In contrast, the average error between the G-coded path and the printed fiber is already higher than the model one at $1.5 \times \text{CTS}$ and grows rapidly with the increasing collector speed. The maximum error of the G-coded path is higher than the model prediction for all speed values. Those maxima correspond to the corner points of the square, where already a small lag can cause noticeable deviations (Figure 4B,C). Evaluating the prediction error by the area between the printed fiber (Video S7, Supporting Information), programmed and predicted pattern, the model provides up to ten times more accurate prediction of the printed fiber position than the G-code programmed trajectory (Figure 4C,D).

The prediction was found applicable for all tested shapes, but the differences in prediction quality were observed between them. The model gives more accurate result for “simple” shapes, such as circles and squares. For the sinusoidal lines, printed at a speed below or equal to $1.5 \times \text{CTS}$, the average error between the predicted and printed path is comparable to the difference between the printed path and the G-code. Taking the maximal error into the account (Figure S1A,B, Supporting Information) the model result can be useful already at $1.25 \times \text{CTS}$. The biggest difference between the actual and the programmed fiber position normally occurs near the points where collector movement direction abruptly changes. Therefore, for printing of a smooth curve at a low collector speed, the error caused by a jet lag might be comparable to the error of the model, caused by the algorithm imperfection and validation image aberration. It is necessary to mention that the amplitude and the wavelength of the “sinusoidal” lines were calculated correctly, and the larger part of the error came from a phase mismatch (Figure 4G,H), partially caused by the image distortion from microscopy.

2.3.2. Translation to Multiple Layers

The results presented above are only valid for the first layer, printed on the flat collector surface. In case of a curved surface or an already deposited fibers, changes in the electrical field and effects which appear due to fiber’s attachment lead to the path distortion with each layer. This fiber position change depends on both printing parameters and path shape.

The fiber trajectory at increasing build height suggests that the lag is increasing with the number of layers. Indeed, it was found that the jet lag, measured at the same speed ratios, can be different when it is printed on top of eight vertically stacked fibers ($178.2 \pm 2.8 \, \mu$m total height) compared to printing onto the collector surface (Figure 5A). However, the CTS is also changing with the wall height (Figure 5B). When the jet lag is measured, corresponding to the corrected speed ratio value, calculated relative to the CTS (decreasing with the build height), its values are close to the flat collector ones for low speed ratio values (<2) but are predominantly below them at a higher speed.
We speculate that it is possible to adjust the input parameters of the model in order to predict the $L_{\text{stable}}$ and $V_{\text{JCP}}$ for each layer. It can be done either by updating the coefficients in Equation (4) or by changing the CTS value for every layer. The first approach might give more precise results but requires the collection of a large amount of data, whereas the latter one is easier to implement. Figure 5A suggests that it would be possible to use the original fit (Equation (4) for a limited range of speed ratio values.

The fiber paths, predicted using the original Equations (4) and (5) and updating the CTS with a power fit function are compared to the printed structures in Figure 5C. A significant prediction error can be observed on the larger test shapes (Figure S1C, Supporting Information). This result could be expected since the model does not take into account the electrostatic attraction[29] that makes the fibers to be deposited on top of each other as well as mechanical properties of the jet. The consideration of these interactions gains even higher importance for the prediction of fiber deposition in MEW scaffolds, which contain numerous fiber intersections, which make the scaffold height uneven.

An interplay of those factors can lead to such an unexpected deposition behavior as formation of a “heel” (Video S8, Supporting Information) near the JCP.[13] The calculated lag would need to have a negative value for a correct JCP position determination. An accurate prediction of the fiber trajectory in these conditions can be problematic since the fiber can be deposited strictly on top, on the side of the structure and next to it (Figure S2, Supporting Information). While this effect can be tamed and used to position fibers in a virtually arbitrary way,[7,30] its mathematical description lies beyond the scope of this study.

2.3.3. Diameter Prediction

Despite the direct impact of the collector speed on the fiber diameter, its prediction proved to be quite challenging. The viscoelastic jet here is acting as a buffer, that does not allow instant diameter change, proportional to the move away speed. Measurement of fiber diameters after a collector speed change shows that the transition between diameters can take a significant amount of time and be distributed over a fiber segment up to 10 mm long (Figure S3, Supporting Information). In a situation when the $V_{\text{MA}}$ is constantly changing, the prediction of the fiber diameter is therefore problematic and technically impossible for the points where $V_{\text{MA}}$ takes negative values (NP, the moving toward JCP, occurs in patterns with rapid direction changes, e.g., sinusoidal lines). Nevertheless, qualitatively, it is possible to correlate the segments with the lower $V_{\text{JCP}}$ and the higher fiber diameter (Figure 5D,E). When the predicted $V_{\text{JCP}}$ stays relatively constant or quickly oscillates, e.g., on the stacked loops or sinusoidal lines, an averaged diameter calculation...
might be considered with the equation from the literature.\cite{9} For the sinusoidal lines with both amplitudes the measured fiber diameter was constant along the fiber, yet different for each amplitude and exceeded the characteristic value for $2 \times \text{CTS}$ ($18.03 \pm 0.97 \mu m$). For both amplitudes though it was lower than the calculated values, $23.78$ and $24.70 \mu m$ for $0.75$ and $1.5 \text{ mm}$ amplitude respectively (Figure 5F). When such elements are incorporated into a scaffold, which contains fiber intersections\cite{31} or sagging fibers,\cite{32} local diameter deviations caused by the fiber flattening\cite{33} might make the estimation less reliable.

Printing with a small gap and higher stage accelerations were reported to allow more rapid diameter transitions than presented here.\cite{34} Applying the model in those conditions could make the diameter calculation more feasible also for the segments with nonperiodical speed ratio changes. A concept of fiber diameter planning for straight fibers, based on a linear lag model, was proposed by the same group;\cite{35} however, no details on the lag and diameter computation were presented as well as the prediction quality.

### 2.4. General Implications for Printing

The geometrical model and the validation experiments further demonstrated the limited applicability of the collector speed decrease approach for predictability improvement during MEW. Although the mean error for printing at $1.125 \times \text{CTS}$ was equal to the model error, the maximal error was still bigger. Further speed decrease would bring the jet within the CTS variance range, meaning that some of the samples or even separate fibers within one sample can be coiled, e.g., due to a fluctuation of the environment conditions or a collector flatness deviation. Additionally, maintaining the collector speed equal to the CTS is practically unattainable for increasing fiber layers containing corner points, where the collector speed can fall below the CTS due to the stage dynamic limitations. The smaller the curvature radius of a nonlinear segment or the angle of a corner, the higher those deviations from a set speed can be. Nevertheless, the printing of such features is the most affected by the presence of a jet lag and, therefore maximally deviates from the G-coded path. In the present study, it was not possible to maintain the constant movement speed near the corners of a square pattern. Therefore, in those locations the actual lag could be smaller than the calculated one, and the actual positions of the NP points differed from the ones in the model.

There exists a compromise solution for accuracy improvement that is often applied while printing above CTS. Reduced speed segments\cite{36} or even pause G-code blocks can be introduced between the linear movement blocks, in order to let the jet lag to decrease temporarily. This method requires a precise evaluation of the waiting time for each printing parameter combination and is only applicable for the patterns consisting of the straight segments. Also, the fiber diameter equity can be sacrificed in favor of higher precision. For this case, the proposed model can be useful to calculate the required speed decrease or pause durations. Specifically, it can be beneficial for structures that are highly sensitive to the accurate jet positioning, such as fiber grids with small fiber spacing, where a slight deviation from the designed fiber placement toward an already deposited fiber can cause its attraction and unwanted stacking.\cite{37}

**Figure 5.** Applicability of the model for multilayer patterns and diameter prediction. A,B) Influence of the construct height on the lag and critical translation speed (CTS), respectively. C) Image overlay of test patterns with eight stacked fiber layers. D–F) Fiber diameter measurement points and respective measurement results, compared to the typical fiber diameter at the given collector speed (red line) and predicted values, based on the calculated contact point speed (blue lines).
The presence of a lag also limits the feature size in the curved or segmented linear scaffolds that can be adequately reproduced. Thus, although the distance between the adjacent points on a simulated path can be chosen arbitrarily small, it is reasonable both to ignore the predicted features with the sizes below the realistic bending radius (e.g., the pinnacles of acute angles or circles with a smaller radius) as well as to include them into the MEW scaffolds design in general. Next to the material and processing parameters it is expected that this specific bending radius of the fiber also depends on the collector movement. An easy way to evaluate the minimal feature size would be to measure fiber coiling radius of a fiber, falling on a stationary[38] or a slowly (below the CTS) moving collector.[13]

When periodic curved shapes are to be produced, it is possible to experimentally adjust the G-code programmed path (e.g., amplitude or phase of the viscoelastic fiber) and thus achieve the required pattern geometry. Those G-code adjustments can also be done in layer by layer manner, thus giving extraordinary possibilities for nearly free-form scaffold creation.[7] At the moment, this method is indispensable for printing of highly complex structures. The application of the model here could decrease the necessary amount of experimentation data and thus accelerate the manufacturing process.

In the future it might be possible to combine the principle of a laydown pattern prediction with a multi-physical mathematical model. For that, the differential equations governing the fall of the liquid on a moving collector[44] must be complemented with the mathematical description of the changing viscoelastic jet properties due to the cooling and flow induced crystallization.[40]

3. Conclusion

The model described here provides a simple tool for the prediction of a MEW fiber laydown pattern, given that it is printed on a flat surface, at constant collector speed, and with a known CTS. Its applicability was successfully tested on different shapes and collector speed values. This model is expected to be relevant for different printing parameter combinations, and thus, printing setups, by adjustments of the lag equation coefficients. It is recognized that both the transition to a 3D scaffold and fiber diameter prediction remains limited, but the model is useful for a qualitative assessment of the complex printed structures. The proposed geometrical model can assist in MEW scaffold design and manufacturing by providing the user with expected first layer print results, and thus aid in either adjusting programmed pattern or printing settings to obtain the designed melt-electrowritten object. The same geometrical approach could be adopted and extended to generate the G-code for the collector movement, based on the required scaffold design.

4. Experimental Section

MEW Setup: Printing was performed on a MEW device, described previously.[46] The maximal stage acceleration and jerk were set to 5 m s\(^{-2}\) and 500 m s\(^{-3}\), respectively. Standard printing parameters for the model validation were: 87 °C set polymer temperature, 1.2 bar air pressure, 22G nozzle, 3.5 ± 0.1 mm gap, –1.5 kV collector voltage and +5.75 kV nozzle voltage. The samples were printed on 1.1 mm thick glass slides (VWR, Germany), put on a stainless-steel collector. Polycaprolactone (Purac PCL, Corbion, The Netherlands) was used as a polymer for printing. Due to the observation that CTS can vary, depending on the environmental conditions and the position on the collector, it was measured, as previously described,[6] before each print and this measured value was used to calculate the actual speed ratio.

Jet Imaging and Lag Measurement: The jet was imaged by a Sony Alpha 7 (Sony Corp., Japan) digital camera with a Nikon ED 200 mm lens (Nikon Corp., Japan). Videos of the jet with 1080p resolution were taken at 50 frames s\(^{-1}\). Stills were captured in Blackmagic Resolve 16 (Blackmagic Design, USA) and then analyzed in ImageJ[41] to measure the distance between the contact point and the nozzle center projection on the collector. \(L_{\text{stable}}\) values were measured in the middle of a 50 mm path at collector speed equal to 1.125, 1.25, 1.5, 2, 5, 9, and 17 × CTS. For the measurement of the \(V_{\text{CP}}\) the falling jet was filmed during both collector speed drop from 17 × CTS to zero and collector speed transitions between 1.125 and 2, 2 and 5, and 5 and 17 × CTS. Lag values were measured starting from the movie frame, preceding the one, where the maximal lag decrease was observed. \(V_{\text{CP}}\) values were calculated by adding the collector displacement relative to the nozzle to the difference between the current and the following lag (Figure S4, Supporting Information) and dividing the result by the frame time (0.02 s).

It must be noted that it was not possible to accurately determine the corresponding collector speed for each lag measurement point and the assumption was made that the speed decreased linearly over the deceleration time. Based on the measurement results, fit functions were chosen, and their coefficients were found in Matlab 2018R Curve fitting toolbox (MathWorks Inc., USA). To determine out how \(L_{\text{stable}}\) and \(V_{\text{CP}}\) depend on the changes in selected processing parameters, pressure, nozzle voltage and gap were independently changed from their default values, described above to 2.4 bar, 5.75 kV, and 4.5 mm, respectively.

Model Implementation and Validation: The path prediction model was implemented with Matlab 2018R, using the “curvspace” function.[40] The validation of the model under the standard printing settings was done with a set of primitives (squares and circles with three different sizes: 2, 4, and 6 mm and two sinusoidal lines with the same wavelength (1 mm) and two different amplitudes (0.75 and 1.5 mm)) using five collector speed values: 1.125, 1.25, 1.5, 2, and 3 × CTS. The predictive ability for multiple layer structures was assessed for the same pattern at the 1.5 × CTS collector speed for samples with eight fiber layers. The imaging of the printed fiber patterns was made with a stereomicroscope (Discovery V20, Carl Zeiss Microscopy GmbH, Germany) in a polarized light, processed by an ImageJ Skeletonize plugin[41] and analyzed in MATLAB 2018R.

The ability of the model to predict fiber diameters was examined with single layer samples printed at 2 × CTS. A series of SEM images in selected points of a 2 mm circle, sinusoidal lines (Figure S5D) were taken with a Crossbeam 340 SEM (Carl Zeiss Microscopy GmbH, Germany) and analyzed in ImageJ. Fiber diameter, characteristic for 2 × CTS collector speed for samples with eight fiber layers. The imaging of the printed fiber patterns was made with a stereomicroscope (Discovery V20, Carl Zeiss Microscopy GmbH, Germany) in a polarized light, processed by an ImageJ Skeletonize plugin[41] and analyzed in MATLAB 2018R.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

3D printing, additive manufacturing, electrohydrodynamic, melt electrospinning writing, melt electrowriting, modeling

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[1] I. Campbell, D. Bourell, I. Gibson, Rapid Prototyping J. 2012, 18, 255.
[2] A. Bauhofer, C. Daraio, Int. J. Adv. Manuf. Technol. 2020, 107, 2563.
[3] F. P. W. Melchels, K. Bertoldi, R. Gabbrielli, A. H. Velders, J. Feijen, D. W. Grijpma, Biomaterials 2010, 31, 6909.
[4] P. D. Dalton, Curr. Opin. Biomed. Eng. 2017, 2, 49.
[5] T. M. Robinson, D. W. Hutmacher, P. D. Dalton, Adv. Funct. Mater. 2019, 29, 1904664.
[6] G. Hochleitner, A. Youssef, A. Hrynevich, J. N. Haigh, T Jungst, J. Groll, P. D. Dalton, BioNanoMaterials 2016, 17, 159.
[7] I. Liashenko, A. Hrynevich, P. D. Dalton, Adv. Mater. 2020, 32, 2001874.
[8] O. Bas, D. D’Angella, J. G. Baldwin, N. J. Castro, F. M. Wunner, N. T. Saidy, S. Kollmannsberger, A. Reali, E. Rank, E. M. De-Juan-Pardo, D. W. Hutmacher, ACS Appl. Mater. Interfaces 2017, 9, 29430.
[9] A. Hrynevich, B. S. Elci, J. N. Haigh, R. McMaster, A. Youssef, C. Blum, T. Blunk, G. Hochleitner, J. Groll, P. D. Dalton, Small 2018, 14, e1800232.
[10] D. Shin, J. Kim, J. Chang, J. Manuf. Processes 2018, 36, 231.
[11] F. M. Wunner, P. Miezczanek, O. Bas, S. Eggert, J. Maartens, P. D. Dalton, E. M. De-Juan-Pardo, D. W. Hutmacher, Biofabrication 2019, 11, 025004.
[12] N. T. Saidy, F. Wolf, O. Bas, H. Keijdener, D. W. Hutmacher, P. Mela, E. M. De-Juan-Pardo, Small 2019, 15, e1900873.
[13] T. D. Brown, P. D. Dalton, D. W. Hutmacher, Adv. Mater. 2011, 23, 5651.
[14] N. M. Ribe, J. R. Lister, S. Chiu-Webster, Phys. Fluids 2006, 18, 124105.
[15] S. W. Morris, J. H. Dawes, N. M. Ribe, J. R. Lister, Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys. 2008, 77, 066218.
[16] N. M. Ribe, M. Habibi, D. Bonn, Annu. Rev. Fluid Mech. 2012, 44, 249.
[17] M. K. Jawed, P.-T. Brun, P. M. Reis, J. Appl. Mech. 2015, 82, 121007.
[18] Z. Yin, Y. Huang, Y. Duan, H. Zhang, in Electrohydrodynamic Direct-Writing for Flexible Electronic Manufacturing (Eds: Y. Duan, H. Zhang), Springer, Singapore 2018, pp. 31–65.
[19] E. Zhmayev, H. Zhou, Y. L. Joo, J. Non-Newtonian Fluid Mech. 2008, 153, 95.
[20] E. Zhmayev, D. Cho, Y. L. Joo, Polymer 2010, 51, 274.
[21] M. J. Divvela, Y. L. Joo, J. Appl. Phys. 2017, 121, 134306.
[22] M. J. Divvela, L. M. Shepherd, M. W. Frey, Y. L. Joo, 3D Print. Addit. Manuf. 2018, 5, 248.
[23] M. J. Divvela, Y. L. Joo, J. Manuf. Processes 2020, 54, 413.
[24] C. P. Carroll, Y. L. Joo, J. Appl. Phys. 2011, 109, 094315.
[25] C. Ru, J. Chen, Z. Shao, M. Pang, J. Luo, AIP Adv. 2014, 4, 017108.
[26] G. Zheng, W. Li, X. Wang, H. Wang, D. Sun, L. Lin, in 2010 IEEE 5th Int. Conf. on Nano/Micro Engineered and Molecular Systems, IEEE, Piscataway, NJ 2010, pp. 284–288.
[27] P. T. Brun, N. M. Ribe, B. Audoly, Phys. Fluids 2012, 24, 043102.
[28] J. C. Kade, P. D. Dalton, Adv. Healthcare Mater. 2020, 2001232, https://doi.org/10.1002/adhm.202001232.
[29] H. Ding, K. Cao, F. Zhang, W. Boettcher, R. C. Chang, Mater. Des. 2019, 178, 107857.
[30] M. de Ruijter, A. Hrynevich, J. N. Haigh, G. Hochleitner, M. Castillo, J. Groll, J. Malda, P. D. Dalton, Small 2018, 14, 1702773.
[31] G. Hochleitner, T. Jungst, T. D. Brown, K. Hahn, C. Moseke, F. Jakob, P. D. Dalton, J. Groll, Biofabrication 2015, 7, 035002.
[32] N. T. Nguyen, J. H. Kim, Y. H. Jeong, Mater. Sci. Eng., C 2019, 103, 109785.
[33] J. N. Haigh, T. R. Dargaville, P. D. Dalton, Mater. Sci. Eng., C 2017, 77, 883.
[34] C. Xie, Q. Gao, P. Wang, L. Shao, H. Yuan, J. Fu, W. Chen, Y. He, Mater. Des. 2019, 181, 108092.
[35] Y. Jin, Q. Gao, C. Xie, G. Li, J. Du, J. Fu, Y. He, Mater. Des. 2020, 185, 108274.
[36] M. Castillo, A. van Mil, M. Maher, C. H. G. Metz, G. Hochleitner, J. Groll, P. A. Dovendans, K. Ito, J. P. G. Sluijter, J. Malda, Adv. Funct. Mater. 2018, 28, 1803151.
[37] T. Tylek, C. Blum, A. Hrynevich, K. Schlegelmilch, T. Schilling, P. D. Dalton, J. Groll, Biofabrication 2020, 12, 025007.
[38] H. Y. Kim, M. Lee, K. J. Park, S. Kim, L. Mahadevan, Nano Lett. 2010, 10, 2138.
[39] J. Schindelin, I. Arganda-Carreras, E. Frise, V. Kaynig, M. Longair, T. Pietzsch, S. Preibisch, C. Rueden, S. Saalfeld, B. Schmid, J. Y. Tinevez, D. J. White, V. Hartenstein, K. Eliceiri, P. Tomancak, A. Cardona, Nat. Methods 2012, 9, 676.
[40] Y. Fukushima, curvspace, https://www.mathworks.com/matlabcentral/fileexchange/7233-curvspace (accessed: May 2020).
[41] I. Arganda-Carreras, R. Fernandez-Gonzalez, A. Munoz-Barrutia, C. Ortiz-De-Solorzano, Micros. Res. Tech. 2010, 73, 1019.