Supplementary Notes

Video S1 and Video S2 published by Hrynevich et al.\textsuperscript{1} was chosen as the source of the dataset used in this paper. A Sony Alpha 7 (Sony Corp. Japan) digital camera was used with a Nikon ED 200 mm lens (Nikon Corp. Japan). 1080 p resolution videos of the nozzle, jet and collector were taken at 50 frames per second. Process hyperparameters were set to 8 m s\textsuperscript{-2} and 500 m s\textsuperscript{-3} maximal stage acceleration and jerk, a 22G nozzle was used, polymer temperature was set to 87 °C and the voltage to the collector was set to -1.5kV, while the voltage to the nozzle was set to +5.75kV.

For Video S1 the air pressure feeding the nozzle was set to 1.2 bar and the distance between nozzle and collector was set to 3.5mm with a standard deviation 0.1mm. Collector’s speeds tested in Video S1 were 191.25 mm/min, 212.5 mm/min, 255 mm/min, 340 mm/min, 510 mm/min, 850 mm/min, 1530 mm/min and 2890 mm/min.

For Video S2 the air pressure feeding the nozzle was set to 2.4 bar and the distance between nozzle and collector was set to 4.5mm with a standard deviation 0.1mm. Collector’s speeds tested in Video S2 were 292.5 mm/min, 520 mm/min, 1300 mm/min and 4420 mm/min.

Initially, the videos were split based on the collector speed setting. Then, the video frames were cropped to remove redundant pixels that would result to increased processing time. For real time video processing the user would need to specify the region of interest in the frame, so, as to crop and dispose needless information, as well as the position of the nozzle, the collector, and a factor, which represents the length of the Taylor cone depending on the nozzle’s diameter.

Table S1: Overview of curated dataset that emulates an E-jet printing machine.

| Machine Setting | Air pressure $p$ [bar] | Tip to Collector Distance $Z$ [mm] | Collector Speed $U_c$ [mm/min] | Number of frames | Duration [sec] |
|-----------------|------------------------|-----------------------------------|-------------------------------|------------------|---------------|
| 1               | 1.2                    | 3.5                               | 191.2                         | 1341             | 26.82         |
| 2               | 1.2                    | 3.5                               | 212.5                         | 1672             | 33.44         |
| 3               | 1.2                    | 3.5                               | 255                           | 1437             | 28.74         |
| 4               | 1.2                    | 3.5                               | 340                           | 1343             | 26.86         |
| 5               | 1.2                    | 3.5                               | 510                           | 648              | 12.96         |
| 6               | 1.2                    | 3.5                               | 850                           | 613              | 12.26         |
| 7               | 1.2                    | 3.5                               | 1530                          | 457              | 9.14          |
| 8               | 1.2                    | 3.5                               | 2890                          | 401              | 8.02          |
| 9               | 2.4                    | 4.5                               | 292.5                         | 1108             | 22.16         |
| 10              | 2.4                    | 4.5                               | 520                           | 802              | 16.04         |
| Property                                      | Value             |
|----------------------------------------------|-------------------|
| Zero-shear-rate (at 100°C) (η₀)               | 1900 Pa s         |
| Relaxation time (at 100°C) (λ₀)              | 0.019 s           |
| Activation energy of flow (ΔH/Rᵦg)           | 7938.4 K          |
| Density (ρ)                                  | 1145 kg/m³        |
| Heat capacity (C_p)                          | 1340 J/kgK        |
| Thermal conductivity (k)                     | 0.14 W/mK         |
| Electrical conductivity (K)                  | 9.5 * 10⁻⁹ S/m    |
| Surface tension (γ)                          | 0.0435 N/m        |
| Ratio of solvent to zero-shear-rate viscosity (β) | 0.001             |
| Mobility factor (α)                          | 0.015             |
| Dielectric constant ratio (ε/ε₀)             | 2.9               |

Table S2: Material properties of PCL

Table S3: Typical values of dimensionless parameters used for PCL

| Parameter | Description                  | Value               |
|-----------|------------------------------|---------------------|
| Bi        | Biot number                  | 0.424               |
| De        | Deborah number               | 1.14                |
| Bo        | Bond number                  | 0                   |
| Re        | Reynolds number              | 5.785 * 10⁻⁶        |
| Ca        | Capillary number             | 1048.276            |
| Na        | Nahme-Griffith number        | 0.446               |
| Fe        | Electrostatic force number   | 0.0254              |
| Γ         | Temperature factor           | 21.283              |
\( Pe \)  
Peclet number for thermal conductivity \[105.209\]

\( Pe_c \)  
Peclet number for electrical conductivity \[0.1122\]

### Table S4: Types of acquisition functions for Active Learning scheme

|       | \( \sigma^2(x) \) | Purely exploration, makes sure that we learn the function \( f \) everywhere on \( x \) to a similar level of absolute error. |
|-------|------------------|-----------------------------------------------------------------|
| Variance | \( PI(x) = \phi \left( \frac{\mu(x) - f(x^+ - \xi)}{\sigma(x)} \right) \), | Selects the point most likely to offer an improvement of at least \( \xi \) but is extremely sensitive to the choice of the target. |
| Probability of Improvement | \( \phi(\cdot) \) is the normal cumulative distribution function | Similar to PI but takes into account the magnitude of the improvement a point can potentially yield as well. |
| Expected Improvement | \[ EI(x) = \begin{cases} 
\{\mu(x) - f(x^+ - \xi)\phi(Z) + \sigma(x)\phi(Z), & \text{if } \sigma(x) > 0 \\
0, & \text{if } \sigma(x) = 0 \end{cases} \] | Selects points for evaluation based on the lower uncertainty bound. |
| Lower Confidence Bound | \( LCB(x) = \mu(x) - \kappa\sigma(x) \), \( \kappa > 0 \) | |

### Table S5: Concentrated Nomenclature, containing every parameter’s symbol, description, and units.

| Parameters: Symbol – Description – Units |
|------------------------------------------|
| **Process** | \( R_0 \) – Needle tip radius | \( mm \) |
| | \( Z \) – Needle tip to collector distance | \( mm \) |
| | \( t \) – Time | \( s \) |
| Symbol | Description                          | Unit     |
|--------|--------------------------------------|----------|
| $p$    | Pressure                             | $kPa$    |
| $Q$    | Volumetric flowrate                  | $m^3/s$  |
| $V_p$  | Voltage applied to needle tip        | $kV$     |
| $V_c$  | Voltage applied to collector         | $kV$     |
| $T_n$  | Needle temperature                   | °C       |
| $T_c$  | Collector temperature                | °C       |
| $U_c$  | Collector speed                      | $mm/s$   |
| $U_{cr}$ | Critical collector speed            | $mm/s$   |
| $R$    | Jet radius                           | $mm$     |
| $V_j$  | Jet speed in $y$ axis                | $mm/s$   |
| $U_j$  | Jet speed in $x$ axis                | $mm/s$   |
| $V_{jm}$ | Jet speed on impact point         | $mm/s$   |
| $\theta_L$ | Angle left                      | °         |
| $\theta_R$ | Angle right                      | °         |
| $A_j$  | Area                                 | $mm^2$   |
| $L_j$  | Lag distance                         | $mm$     |

### Physics Jet Model

| Symbol | Description                          | Unit     |
|--------|--------------------------------------|----------|
| $Re$   | Reynolds number                      | –        |
| $Pe$   | Peclet number                        | –        |
| $Ca$   | Capillary number                     | –        |
| $Fe$   | Electrostatic force parameter        | –        |
| $Bo$   | Bond number                          | –        |
| $Na$   | Nahme-Griffith number                | –        |
| $De$   | Deborah number                       | –        |
| $Bi_L$ | Local Biot number                    | –        |
| $\Theta$ | Dimensionless temperature           | –        |
| $\Gamma$ | Temperature factor                  | –        |
| $\chi$ | Aspect ratio                         | –        |
| $\alpha$ | Mobility factor                     | –        |
| $\beta$ | Ratio of solvent to zero-shear-rate viscosity | – |
| $\beta_E$ | Dielectric constant ratio          | –        |
| $\sigma$ | Surface charge density              | $C/m^2$ |
| $E_T$  | The tangential component of the electric field to the jet surface | $V/m$ |
| $f(\Theta)$ | Temperature dependence of the zero-shear-rate viscosity | – |
| $\Delta H$ | Activation energy                  | $K$      |
| $R_{ig}$ | Ideal gas constant                  | –        |
| $\Delta T_{Rh}$ | Temperature change necessary to substantially alter the rheological properties of the fluid | $K$ |
| $\tau_{zz}$ | Total axial normal stress           | $Pa$     |
| $\tau_{rr}$ | Total radial normal stress         | $Pa$     |
\[ \tau_{p,zz} \quad \text{Axial polymeric stress} \quad Pa \]
\[ \tau_{p,rr} \quad \text{Radial polymeric stress} \quad Pa \]

**Geometrical model**

- \( R_c \): Steady coiling radius \( mm \)
- \( q \): Jet’s trace on the collector \( mm \)
- \( r \): The contact point \( mm \)
- \( s \): Deposited jet’s arc length \( mm \)
- \( r \): Polar radius coordinate \( mm \)
- \( \psi \): Polar angle coordinate \( (°) \)
- \( \theta \): Curvature at the bottom of the jet \( (°) \)

**CV Metrology**

- \( t_{CV} \): Processing time \( s \)
- \( fps \): Frames per second \( s^{-1} \)
- \( \text{cf} \): Calibration factor \( mm/pix \)
- \( \text{stride} \): Indicating every how many pixels along the z-axis we perform computations \( pix \)
- \( \text{re} \): Jet’s Right Edge at a specific point across the needle tip to collector distance (type: integer). \( pix \)
- \( \text{le} \): Jet’s Left Edge at a specific point across the needle tip to collector distance. (type: integer). \( pix \)
- \( \text{re}_{\text{previous}} \): Jet’s Right Edge at last accessed point before the currently processed point across the needle tip to collector distance (type: integer). \( pix \)
- \( \text{le}_{\text{previous}} \): Jet’s Right Edge at last accessed point before the currently processed point across the needle tip to collector distance (type: integer). \( pix \)
- \( \text{re}_{\text{current}_{\text{frame}}} \): Jet’s Right Edge at current frame at all processed points across the needle tip to collector distance (type: array). \( pix \)
- \( \text{re}_{\text{previous}_{\text{frame}}} \): Jet’s Right Edge at previous frame at all processed points across the needle tip to collector distance (type: array). \( pix \)

**Gaussian Processes**

- \( D \): Dataset, available input-output pair of observation data
- \( f(\cdot) \): Unknown function to be approximated
- \( m(\cdot) \): Mean function determining the unknown function
- \( K(\cdot) \): Covariance matrix determining the unknown function
- \( k(\cdot) \): Kernel reflecting the prior available knowledge on the unknown function
$$\theta$$  Kernels hyperparameters to be trained
$$\mu(\cdot)$$  Mean prediction of GP model
$$\sigma^2(\cdot)$$  Variance prediction of GP model
$$n$$  Number of available data

### Multifidelity Modeling

- $$f_{\text{high}}(\cdot)$$  High fidelity GP model
- $$f_{\text{low}}(\cdot)$$  Low fidelity GP model
- $$\rho$$  A scaling constant quantifying the correlation between the two models
- $$f_{\text{err}}(\cdot)$$  Another GP modeling the bias term for the high-fidelity data
- $$n_{\text{low}}$$  Number of low-fidelity data available
- $$n_{\text{high}}$$  Number of high-fidelity data available

### Active Learning and Bayesian Optimization

- $$u(\cdot)$$  Acquisition function
- $$\xi$$  Parameter specifying the least required improvement
- $$\Phi(\cdot)$$  The normal cumulative distribution function
- $$\phi(\cdot)$$  The normal probability distribution function
- $$\kappa$$  Parameter specifying reliability of confidence intervals

### Performance Metrics

- **RMSE**  The Root Mean Squared Error
  - Performance Metric measures the average magnitude of the error between the predicted values and the true values.

- **MCIW**  The Mean Confidence Interval Width
  - Performance Metric measures the average width of a confidence interval.

- **Min. Regret**  The Minimum Regret Performance Metric
  - quantifies how well a method works at finding the optimum.
Figure S1: Features Extracted from Computer Vision Module. a) Normalized jet radius ($R_j/R_o$) obtained from the computer vision metrology module of the GPJet framework plotted against the normalized jet length ($Z/R_o$) and the normalized time ($t/t_{max}$). b) Normalized jet area ($A_j/A_o$) obtained from the computer vision metrology module of the GPJet framework plotted against the normalized jet length ($Z/R_o$) and the normalized time ($t/t_{max}$). c) Jet angles ($\theta$) obtained from the computer vision metrology module of the GPJet framework plotted against the normalized jet length ($Z/R_o$) and the normalized time ($t/t_{max}$). d) Jet velocities ($U_j$) obtained from the computer vision metrology module of the GPJet framework plotted against the normalized jet length ($Z/R_o$) and the normalized time ($t/t_{max}$).
Figure S2: Performance Metrics Evolution for Active Learning Tasks presented in Figure 7 and Figure 8. 

a) Root Mean Squared Error (RMSE) evolution after each iteration, regarding the normalized jet radius ($R_j/R_o$). 
b) Root Mean Squared Error (RMSE) evolution after each iteration, regarding the lag distance ($L_j$). 
c) Mean Confidence Interval Width (MCIW) evolution after each iteration, regarding the normalized jet radius ($R_j/R_o$). 
d) Mean Confidence Interval Width (MCIW) evolution after each iteration, regarding the lag distance ($L_j$).
Figure S3: Collective Performance Metrics for Regression and Active Learning Tasks. a) Root Mean Squared Error (RMSE) for every case, regarding the normalized jet radius ($R_j/R_o$). b) Root Mean Squared Error (RMSE) performance metric, for every case, regarding the lag distance ($L_j$). c) Mean Confidence Interval Width (MCIW) performance metric, for every case, regarding the normalized jet radius ($R_j/R_o$). d) Mean Confidence Interval Width (MCIW) performance metric, for every case, regarding the lag distance ($L_j$).
Figure S4: Performance Metrics. **a)** Minimum Regret Performance Metric evolution, after each iteration, regarding the Bayesian Optimization Task to find the minimum lag-distance ($L_j$). The termination criterion is set to 0.5 mm (red dashed line). **b)** Minimum Regret Performance Metric evolution, after each iteration, regarding the Active Learning Task to explore the design space of lag-distance ($L_j$) for specific speed ratios ($U_c/V_{jm}$).
Figure S5: Results of Active Learning process on Multifidelity Modeling Task (column a) versus on High Fidelity Modeling Task (column b). a) exploring the design space using Active Learning to fit a Multifidelity Gaussian Process to normalized high fidelity observation data (red color) of jet radius \( R_j/R_o \) and low fidelity model data obtained from the computer vision metrology module of the GPJet framework and from the multi-physics model, respectively, at specific z axis coordinates along the normalized jet length \( Z/R_o \), \( i - vi \) denote the iterations of the active learning algorithm until it meets its termination criteria. b) exploring the design space using Active Learning to fit a Gaussian Process to normalized high fidelity observation data (red color) of jet radius \( R_j/R_o \) obtained from the computer vision metrology module of the GPJet framework at specific z axis coordinates along the normalized jet length \( Z/R_o \), \( i - vi \) denote the iterations of the active learning algorithm until it meets its termination criteria. In every case, the observation point chosen at each iteration is denoted with a black dashed line box pointed by a black arrow.
Supplementary References

1. Hrynevich, A., Liashenko, I. & Dalton, P. D. Accurate Prediction of Melt Electrowritten Laydown Patterns from Simple Geometrical Considerations. *Adv Mater Technologies* 5, 2000772 (2020).