
Tension-driven three-dimensional printing of free-standing Field's metal structures

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Table S1. Comparison with conventional direct ink writings of conductive materials in the aspect of materials, conductivity, aspect ratio, printing speed, and post treatment. LBL represents layer-by-layer deposition, indicating they cannot print free-standing structures and layer-by-layer deposition is the only way to print out-of-plane structures.

Materials	Conductivity (S/cm)	Aspect ratio	Printing speed (mm/s)	Post treatment	Ref.
Field's metal	2×10^4	750	100	-	This work
Ag, silicone	0.99	LBL	-	-	S1
Ag, PEO	1.4×10^4	LBL	7	-	S2
Ag, PAA	1.9×10^4	7	0.5	Annealing at 150 °C	S3
Ag, carbon nanotube,	2.3×10^4	10	5	Drying out at 80 °C	S4
Ag, PAA	1×10^5	~ 10	0.5	Sintering at 300 °C	S5
Ag, PAA	1.8×10^5	~ 200	2	Laser annealing	S6
Ionic hydrogel	0.2	LBL	6	-	S7
PEDOT:PSS	155	LBL	-	Annealing at 130 °C	S8
Graphene	1.2	LBL	4	Freeze-drying, chemical reduction, annealing	S9
Liquid metal, PDMS	1.4	LBL	-	Curing PDMS	S10
Liquid metal, carbon nanotube	3×10^4	~ 130	-	-	S11
Liquid metal	3.4×10^4	~ 250	3	Reconfiguration	S12
Liquid metal	3.4×10^4	~ 50	0.2	-	S13
Liquid metal	3.4×10^4	56	1	Freeze-Printing	S14
Liquid metal	3.4×10^4	LBL	1	-	S15
Liquid metal	3.4×10^4	0.6	1	-	S16

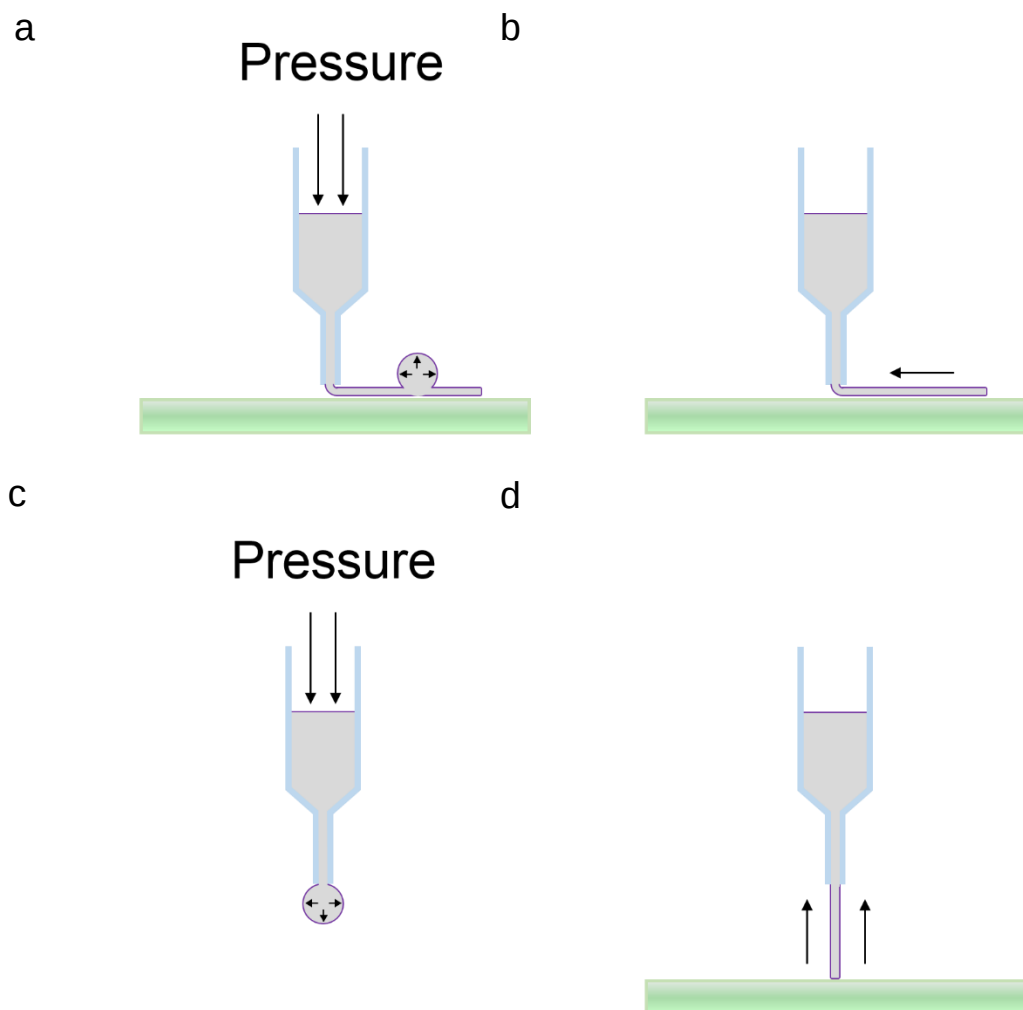


Figure S1. Schematics of press-driven printings and CHARM3D of Field's metal. **a**, Pressure-driven planar printing on substrate. **b**, Shear-driven CHARM3D of planar writing on substrate. **c**, Pressure-driven vertical printing above substrate. **d**, Tensile-driven CHARM3D.

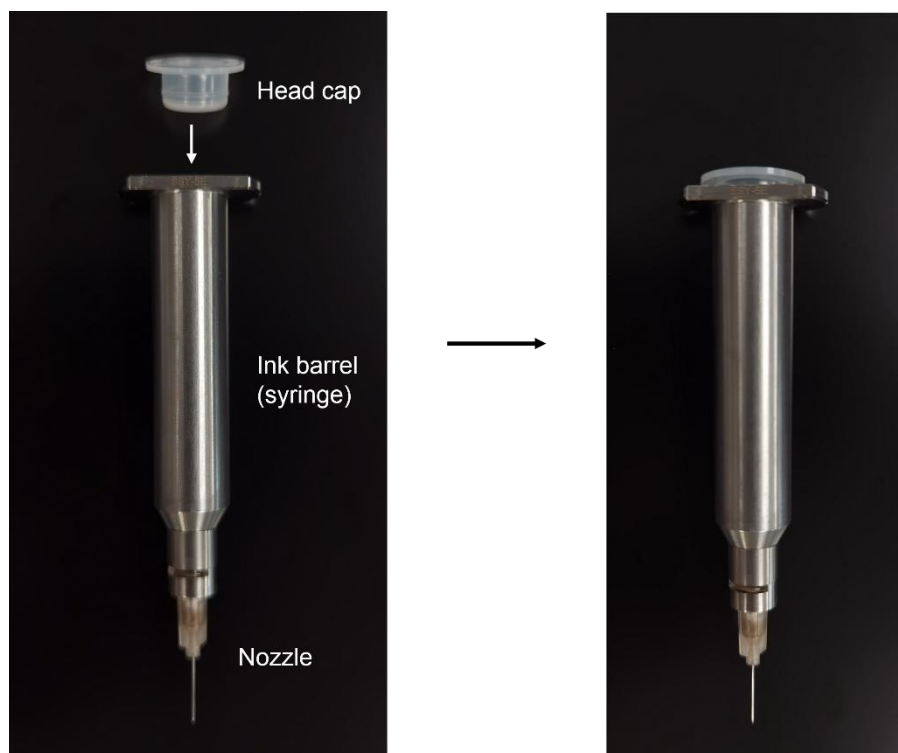


Figure S2. Initiation of CHARM3D. To initiate, the head cap was pressed down to extrude several drops of molten metal. After that, the head cap was removed to make the top end of ink barrel open to air.

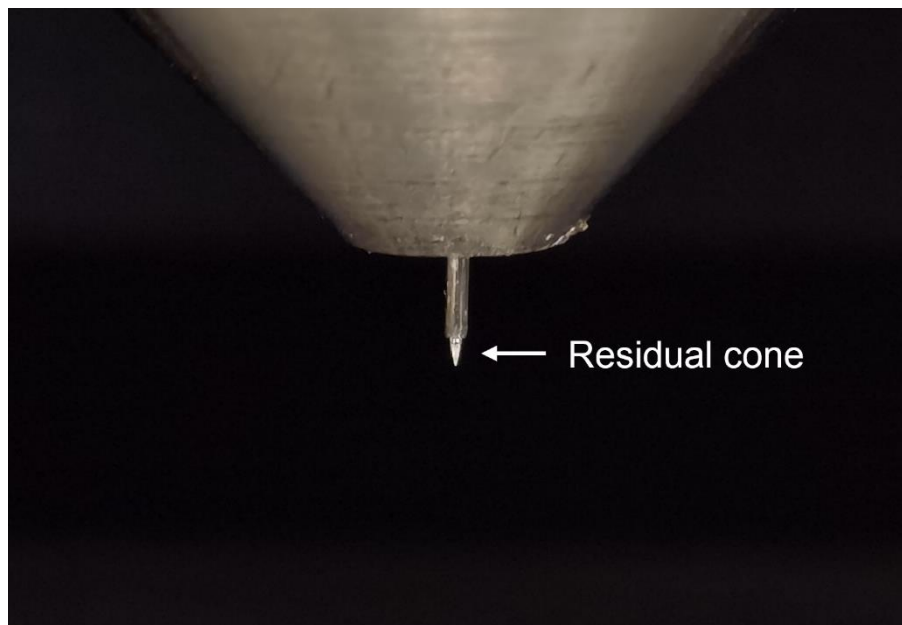


Figure S3. The residual metal cone remaining on the nozzle tip after printing.

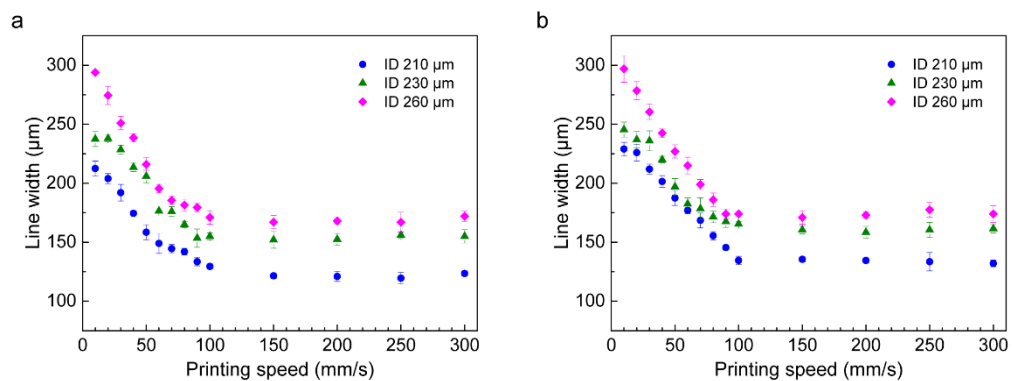


Figure S4. Plot of the line width of printed metal wires versus the printing speed with different inner diameters of nozzles at 90 °C **(a)** and 100 °C **(b)**. Data are presented as mean values $\pm$ standard deviation ($n = 5$).

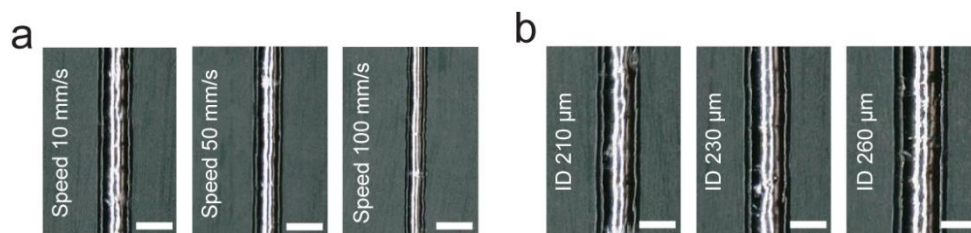


Figure S5. Photographs of printed metal wires under optical microscope. **a**, The wires printed with different speed of 10, 50, and 100 mm s⁻¹, where the nozzle ID was 210 μm and temperature was fixed at 80 °C. **b**, The wires printed with different nozzle ID of 210, 230, and 260 μm, where the printing speed was 10 mm s⁻¹ and temperature was fixed at 80 °C.

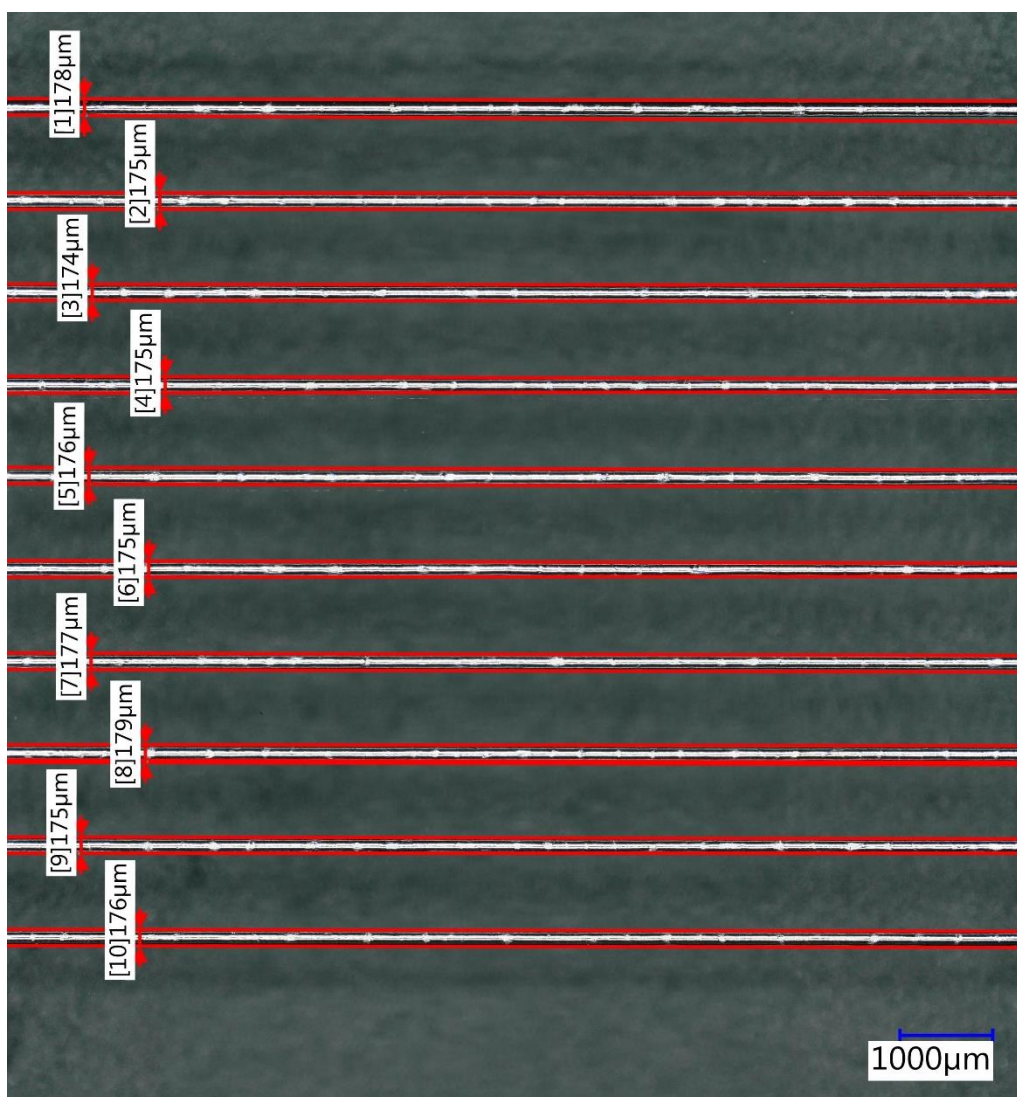


Figure S6. Photograph of printed parallel metal wires under optical microscope. All of them were printed at the same condition, where the nozzle size was 210 μm , printing speed was 30 mm s^{-1} , and temperature was set at 80 $^{\circ}\text{C}$.

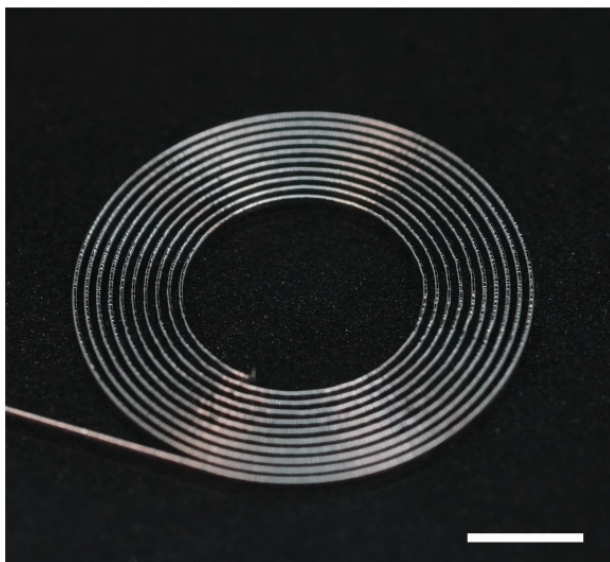


Figure S7. Photograph of a printed 2D inductor. It was printed by a nozzle of ID 210 μm on glass at 80 $^{\circ}\text{C}$. Scale bar, 5 mm.

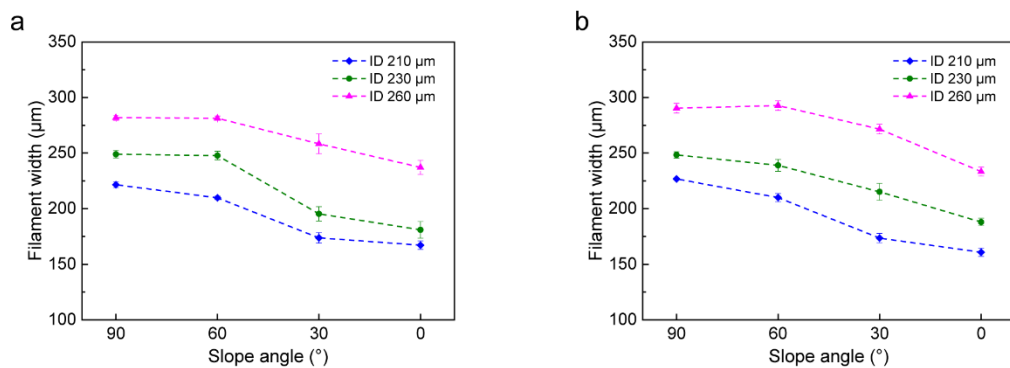
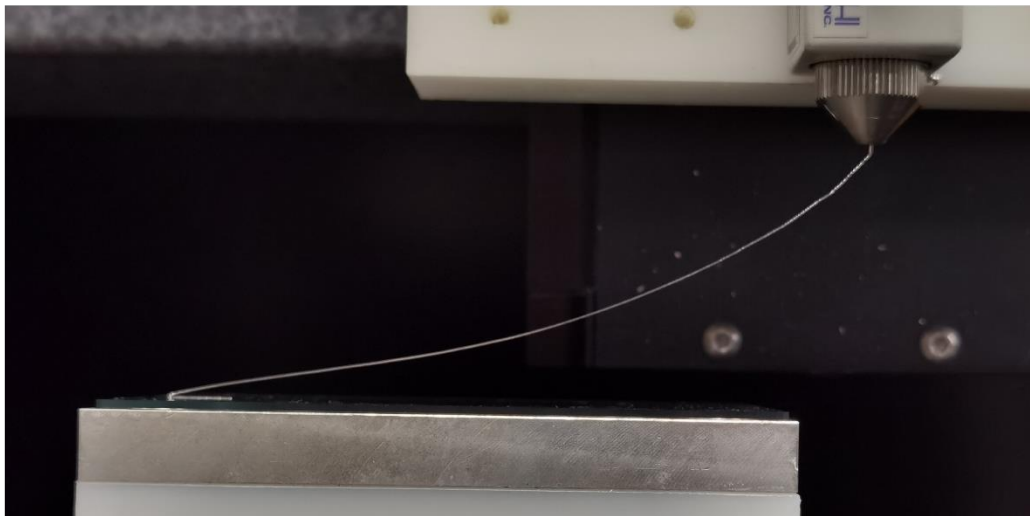


Figure S8. Plot of the widths (diameters) of free-standing filaments versus slope angels printed with nozzles of ID 210, 230, and 260 μm at 90 $^\circ\text{C}$ (a) and 100 $^\circ\text{C}$ (b), respectively. Data are presented as mean values $\pm$ standard deviation ($n = 5$).

a



b

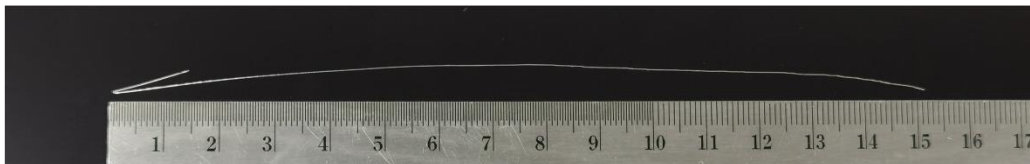


Figure S9. The longest free-standing filament created by CHARM3D. **a**, Photograph taken at the end moment of printing the longest free-standing metal filament. **b**, The longest filament measured with a rule. The longest filament was printed with a printing speed of 0.3 mm s^{-1} by the ID $210 \text{ }\mu\text{m}$ nozzle at $80 \text{ }^{\circ}\text{C}$. It was designed to be a straight free-standing filament with a slope angle of 15° , but the gravity made the middle part sunken a bit.

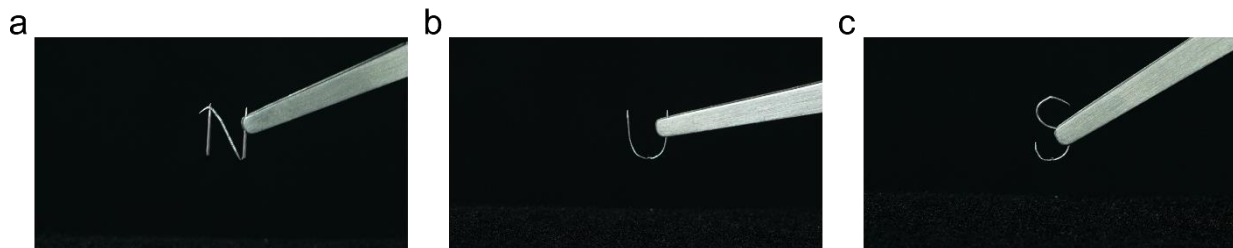


Figure S10. Continuous letters of “NUS”. **a, b, c,** Every individual letter of printed free-standing “NUS” can be lifted up by holding one point with a tweezer, implying that each letter is continuous.

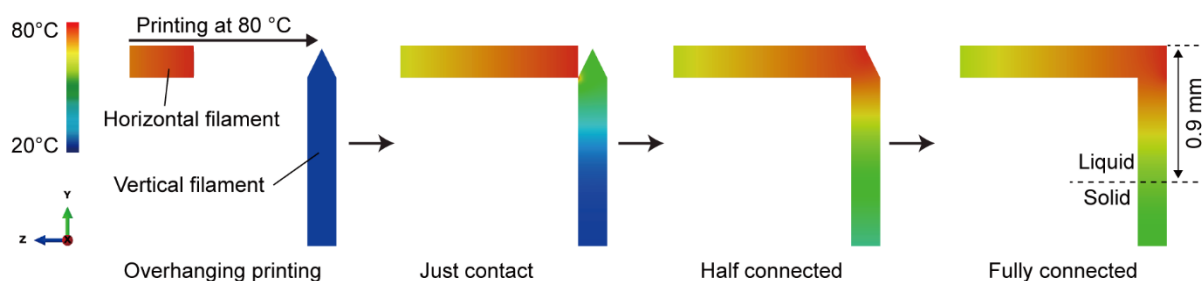


Figure S11. Temperature distribution around the vertex while horizontally printing overhanging filaments on the top of the cubic framework. The horizontal filament is printed at 80 °C with a speed of 1 mm s⁻¹. The heat transfer during the entire process of vertex connection has been simulated through FEA and detail can be found at the end of the supplementary document. When the horizontal filament flits across the vertical filament top in the overhanging printing, the just-printed hot metal ink would transfer heat to the vertical filament quickly from the top downward once they contact. After the horizontal filament fully covers the vertex, a small volume of the vertical filament has been heated up over melting point. The solid liquid interface in the vertical filament is 0.9 mm away from the vertex top. Therefore, the re-melted vertical filament top would fuse with the just-printed horizontal filament, resulting in robust connection at the vertex once solidified.

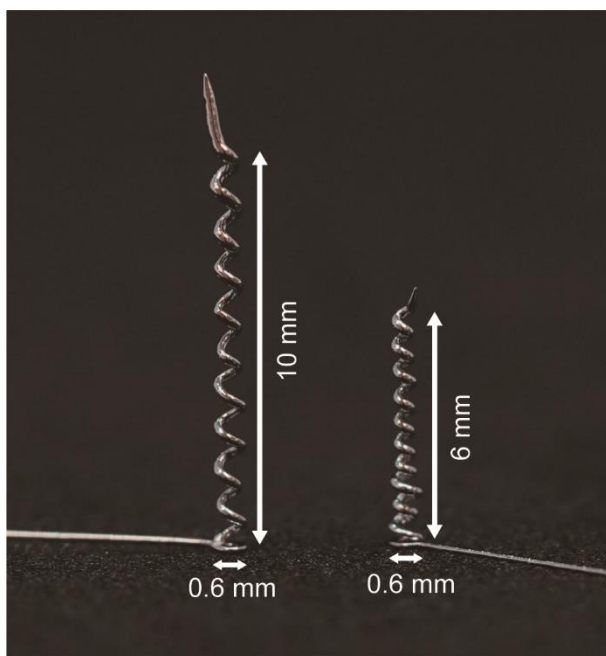


Figure S12. The smallest helical structures that can be printed by CHARM3D. Their diameters were around 600 μm , while the pitches were 600 μm for the left one and 1 mm for the right one. They were printed with the nozzle of ID 210 μm at 80 $^{\circ}\text{C}$.

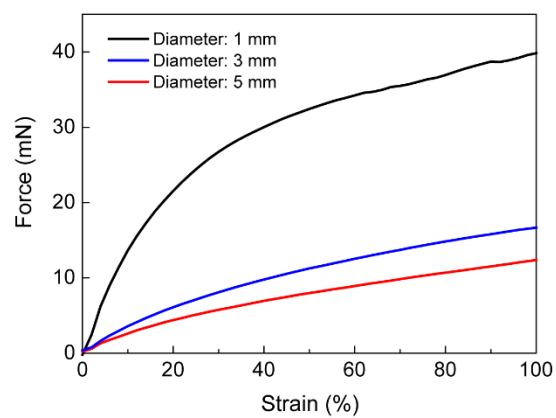


Figure S13. Plot of the force-strain curve for printed helical structures with diameters of 1, 3, and 5 mm. They had the same height of 5 mm with 5 turns of coils and were printed with the nozzle of ID 210 μm at 80 $^{\circ}\text{C}$.

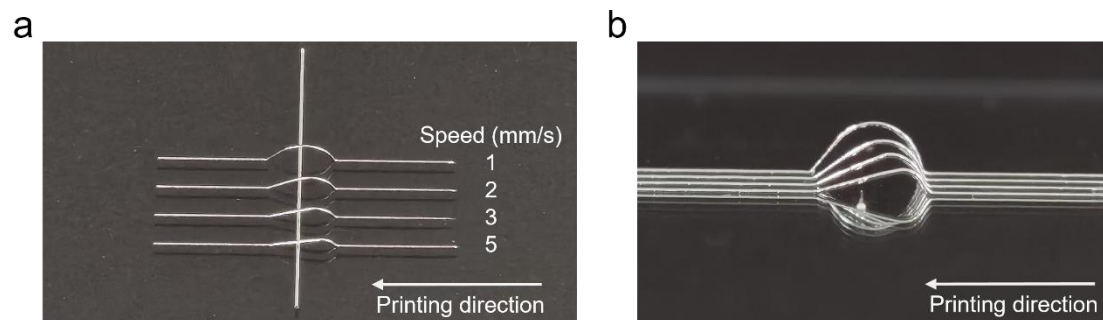


Figure S14. Photographs of printed arc bridges crossing a metal wire from top view **(a)** and front view **(b)**. The printing speeds are 20 mm s^{-1} for wires on glass substrate and $1\text{-}5 \text{ mm s}^{-1}$ for out-of-plane arc bridges. They are printed at $80 \text{ }^{\circ}\text{C}$ with a nozzle of ID $210 \text{ }\mu\text{m}$.



Figure S15. The fabrication process of the 3D printed multilayer circuit of the battery-free temperature sensor system.



Figure S16. A curve needle with a bending angle of 45 °.

Heat transfer Model for Simulation

The heat transfer model used for the simulation consists of model assumptions, parameter selection, the governing equation, and its boundary conditions.

Assumptions:

- a. For density ρ , thermal conductivity k , convective heat transfer coefficient h , and specific heat c_p of Field's metal, the difference between solid and liquid state is neglected.
- b. Since the nozzle moves slowly (<1 mm/s) during printing free-standing metal structures and the pulled out liquid part is quasi-static, the convection inside the liquid part could be ignored.
- c. As the total heat loss Q_t is much larger than the heat loss Q_r by thermal radiation and the difference between the will raise when the printed length increases, the influence caused by thermal radiation could be ignored.
- d. During the 3D printing, the tensile, circumferential and shear force/stress provided by the nozzle is less than the yield force/stress of the oxidized skin, so that the overall structure is uniform and does not deform, and the boundary does not change.
- e. The nozzle and syringe used for 3D printing in the laboratory are wrapped by a heating device and maintained at a constant temperature, the moving upper boundary temperature T_p of the heat transfer model could be set to a constant value.
- f. As the bottom part that contacts the substrate could be cooled to room temperature in a very short time by the metal stage, the temperature T_B of the bottom boundary of the metal structure could also be approximately regarded as a constant value.
- g. Only the main part of the free-standing structure is considered in the heat transfer simulation, and the limited influence of the bottom structure is not considered.

Material Parameters:

According to the temperature set in the practical 3D printing experiment, the temperature of the moving upper boundary can be $T_p = 70 - 100^\circ\text{C}$, while the temperature of the bottom boundary should be $T_B = 20^\circ\text{C} = 293.15\text{K}$. The convection heat transfer coefficient h at any point on the surface of the 3D printed structure is related to the shape of the overall structure and the height and temperature of the point^{S17, S18}. However, in our simulations, we ignore this series of effects, and regard the average convection heat transfer coefficient h of the whole structure as $25 \text{ W}/(\text{m}^2 \cdot \text{K})$. The rest of the parameter values are shown in the following table.

Table S2. Key parameters of Field's metal.

Physical Properties	Value
Density (kg/m^3) ^{S19}	7.88×10^3
Thermal Conductivity (W/mK) ^{S19}	19
Specific Heat ($\text{J}/\text{kg} \cdot \text{K}$) ^{S19}	285
Solidus Temperature (K) ^{S19}	333.15
Liquidus Temperature (K) ^{S20}	335.15
Latent Heat (J/g) ^{S21}	25.4
Convective heat transfer coefficient ($\text{W}/(\text{m}^2 \cdot \text{K})$)	25

Governing Equation and Boundary conditions:

According to Figure S17 and the assumptions mentioned above, we can obtain the following settings for the governing equation and boundary conditions:

$$\begin{cases} \frac{\partial}{\partial X_1} \left(k \frac{\partial T}{\partial X_1} \right) + \frac{\partial}{\partial X_2} \left(k \frac{\partial T}{\partial X_2} \right) + \frac{\partial}{\partial X_3} \left(k \frac{\partial T}{\partial X_3} \right) + Q - \rho c_p \frac{\partial T}{\partial t} = 0 \\ \text{Top Boundary: } T \left(l = n\delta, t = \frac{n\delta}{v} \right) = T_p \\ \text{Bottom Boundary: } T(l = 0, t) = T_B \\ \text{Convective Heat Transfer Boundary: } q = h(T_s - T_\infty) \end{cases}$$

n is the index number of arbitrary time step, T_p is temperature of top boundary, T_B is the temperature of bottom boundary, h is the convective heat transfer coefficient, T_s is the surface temperature of liquid part, T_∞ is the ambient temperature and q is the heat flux generated by the temperature difference between the surface temperature of liquid part and the ambient temperature.

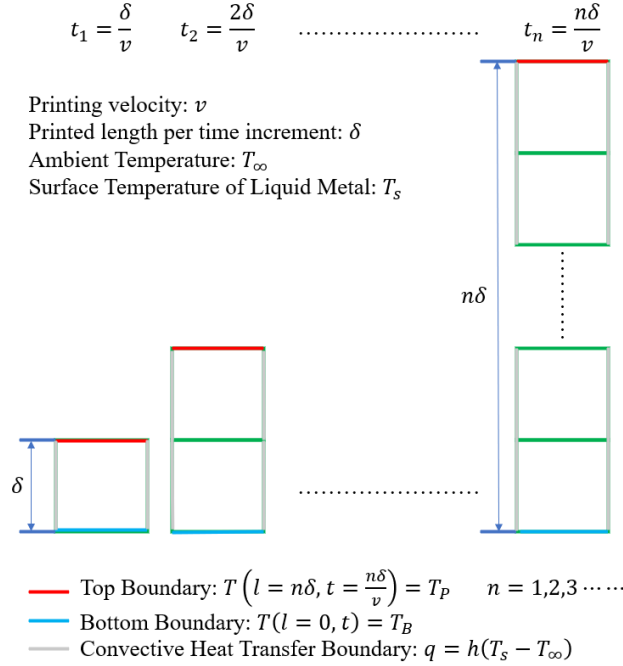


Figure S17. Schematic of heat transfer model with nozzle moving effect

Mechanical Analysis

The purpose of the mechanical analysis is to perform the estimation of suitable printing parameters in combination with the heat transfer simulation. After obtaining the length of the liquid part at different printing speeds and temperatures through heat transfer simulation, we could also determine the critical

value of the maximum allowable length of the liquid part, above which bulging may occur in the liquid part. For continuous 3D printing, the liquid length of the printed structure should be smaller than this critical value. We can use this to predict the maximum allowable printing speeds under different printing temperatures.

The entire mechanical analysis is based on the work reported by Gozen et. al^{S22}. We regard the small liquid part at the end of the printed free-standing structure as the mechanical analysis object. According to the practical 3D printing experiment, the liquid part should always be straightened by tension from the moving nozzle. Hence, the oxidized skin encapsulating the liquid part can be regarded as an inclined cylindrical shell. We mainly analyze the elastic stage of the thin oxide shell to find out when the stress on the oxidized skin reaches the yield stress.

Before conducting the overall mechanical analysis, it is necessary to confirm if the oxide skin shell belongs to the thin shell model or the thick shell model. According to the work from Martin Thuo et al., the thickness δ of the oxidized skin of Fields metal is measured to be about 4.3nm,^{S23} while the diameter d of the transverse section of the metal filament printed in our experiment is about 200 μ m. Obviously, the ratio of diameter to skin thickness is much larger than 20, which satisfies the assumptions of the thin shell model.

For the stress analysis of oxidized skin, the shell is subjected to circumferential stress $\sigma_{\theta\theta}$, longitudinal stress σ_{zz} and radial stress σ_{rr} . Since the oxidized skin is a thin shell, the effects of radial stress σ_{rr} can be ignored.

As shown in Figure S18, circumferential stress $\sigma_{\theta\theta}$ is affected by its internal pressure P . We can calculate the force balance equation through the projected area of the circumferential stress $\sigma_{\theta\theta}$ and the pressure P , to obtain the relationship between the two.

$$N_{\theta\theta} = \sigma_{\theta\theta} \cdot \delta = \frac{Pd}{2}$$

In this equation, $N_{\theta\theta}$ is the circumferential stress with different unit (N/m).

The calculation of longitudinal stress σ_{zz} is different from circumferential stress $\sigma_{\theta\theta}$. In addition to the effect of internal pressure, the gravitational force w of the Field's metal and the reaction force (R_v and R_h) of the solid-liquid interface should also be considered. Its stress formula is as follows:

$$N_{zz} = \sigma_{zz} \cdot \delta = \frac{Pd}{4} + \frac{R_v \sin \psi}{\pi d} + \frac{R_h \cos \psi}{\pi d} + \frac{wL \sin \psi}{\pi d}$$

In this equation, L is the liquid length, N_{zz} is the longitudinal stress with different unit (N/m) and ψ is the inclination angle of the liquid part.

The inclination angle ψ of the metal filaments between the liquid part and the horizontal plane is the printing angle set during the experiments, while the inclination angle ψ of the liquid part of the cylindrical helix depends on the diameter d of the structure and the design of the pitch h_p ($\psi = \tan^{-1} \left(\frac{h_p}{\pi d} \right)$).

It is worth mentioning that according to the work from Gozen et al, the magnitude of the measured reaction force ranges from about 20~150 μ N.^{S22} Therefore, in our mechanical analysis, the sum of the components of the reaction force acting on the longitudinal direction was set to 150 μ N for an estimation. The calculation of the internal pressure P should consider the pressure loss P_{loss} when the Field's metal is pulled from the syringe.

$$P = \rho g \Delta h - P_{loss}$$

$$\Delta h = z + a + L \sin \psi$$

In these two equations, z is the height of liquid volume in the syringe (Figure S20), a is the length of the commercial needle (3 mm), ρ is the density of the Field's metal and g is the gravitational acceleration.

The calculation of the pressure loss P_{loss} can directly refer to the work from Gozen et al^{S14}.

After above mechanical analysis, the relationship between the above stress and the length of the liquid part L /the printing velocity v / the liquid level z in the syringe can be obtained through Matlab calculation.

According to the surface yield stress of Field's metal found by Félix Faura et al. (417mN/m), we could thus determine the maximum allowable length of the liquid part^{S24}.

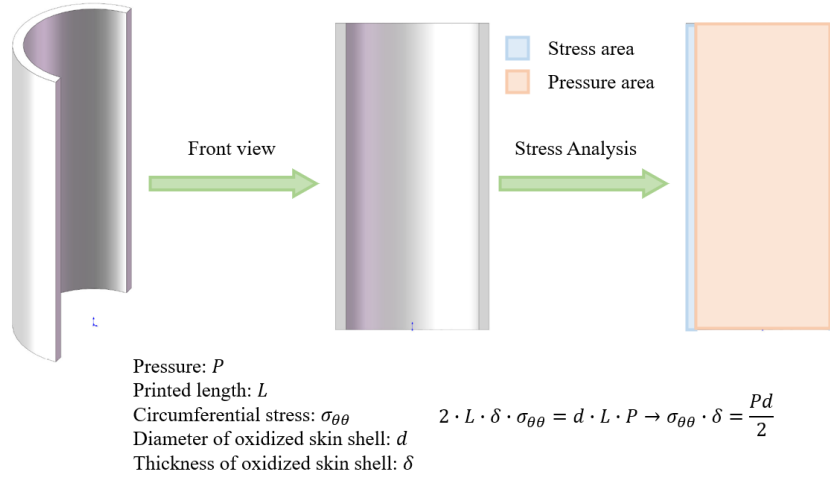


Figure S18. The schematic of circumferential stress

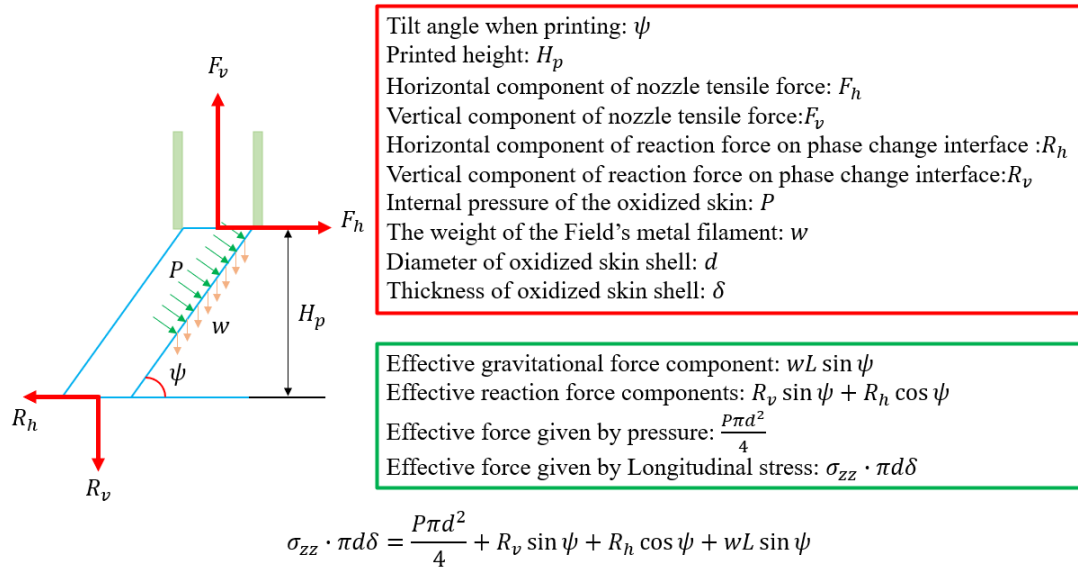


Figure S19. The schematic of longitudinal stress

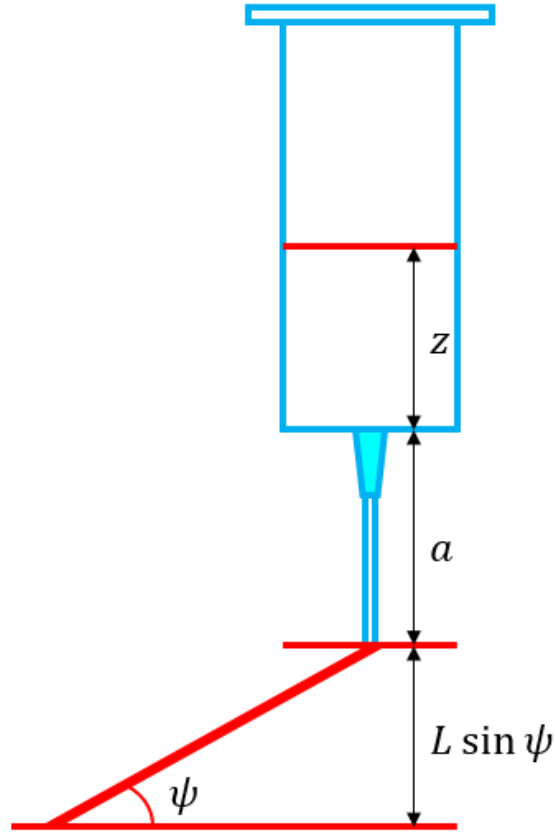


Figure S20. The schematic of the calculation of Δh

Vertex connection

To simulate the heat transfer during the vertex connection process, we use the same assumptions, parameters, governing equation, and boundary conditions as above mentioned. An ideal vertex connection between horizontal and vertical filaments should be the geometry shown in the left subfigure in Figure S21. To simplify the simulation process, we only consider the heat transfer analysis near the vertex as highlighted in the right subfigure in Figure S21. As shown in Figure S22, there are two geometry parts in this simulation, one is the connector part (horizontal filament), and the other is the vertical filament with the tip. The connector part is approaching and will flit across the vertical filament top with a speed of 1 mm s^{-1} , representing the real overhanging printing process. As shown in Figure S23, the connector part is first sliced into small segments, then applied with the convective heat transfer condition, and the moving

temperature boundary conditions. The model change technique in ABAQUS is used to mimic the printing process.

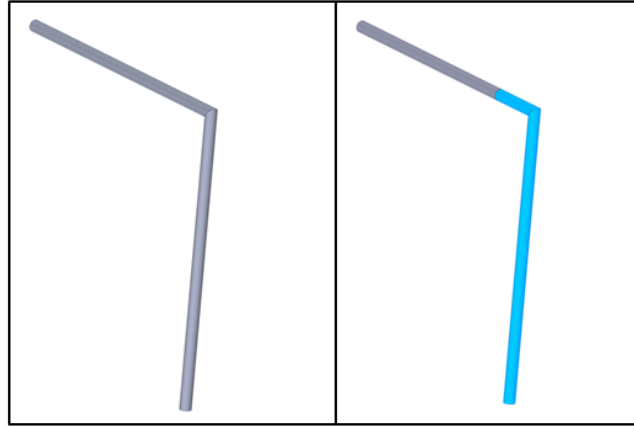


Figure S21. Schematic diagram of the vertex connection.

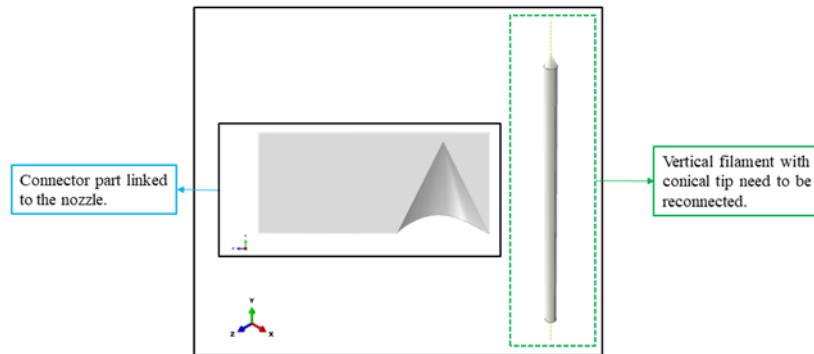


Figure S22. Schematic diagram of two geometry parts in the simulation

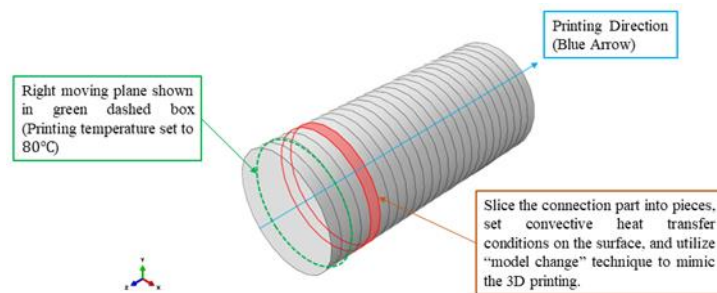


Figure S23. Schematic diagram of simulation settings

Viscosity Thresholds for Tension-Driven 3D Printing

In the tension-driven free-standing 3D printing, the viscosity of materials plays a crucial role in enabling smooth printing. High viscosity of the ink would significantly increase the resistance and induce excessive pressure loss when ink flows through the nozzle. As a result, tension could not overcome the resistance and fail to pull out the ink continuously. Therefore, CHARM3D prefers low-viscosity materials. To quantitatively analyze the viscosity range for successful printing, we introduce the Darcy–Weisbach equation to describe the pressure loss of fluids in a narrow tube^{S25}. The pressure loss due to the viscosity appears in three segments, including syringe, nozzle, and unsolidified wire after printing.

The Darcy–Weisbach equation could be written as^{S25}

$$\Delta P_{nl} = f_D \cdot \Delta l \cdot \frac{\rho}{2} \cdot \frac{v_{print}^2}{D}$$

In which,

ΔP_{nl} stands for the pressure loss due to the viscosity;

f_D stands for the Darcy friction coefficient;

Δl stands for the length of different segments, which could be the length (height) z of liquid volume in the syringe, the length a of the nozzle, and the length L of the unsolidified wire;

ρ stands for the density of the metal in liquid state;

v_{print} stands for the mean velocity of the flow rate of metal inside the nozzle, which approximates the printing velocity;

D stands for the diameter of different segments, which could be the diameter d_s of the syringe, the diameter d_n of the nozzle, and the diameter d_o of the oxidized skin on the unsolidified wire.

During the 3D printing process, the flowing metal inside the syringe, nozzle, and oxidized skin has a low flow rate so that it could be considered as laminar flow. Therefore, the Darcy friction factor formula for fluids in laminar flow could be employed, which is $f_D = \frac{64}{Re}$ (Ref^{S26}), where Re is the Reynolds number.

The formula of the Reynolds number could be written as $Re = \frac{\rho u L_{char}}{\mu}$ (Ref^{S27}).

In which,

ρ stands for the density of the metal in liquid state.

u stands for the flow velocity, which approximates the printing velocity v_{print} .

L_{char} stands for the characteristic length of each segment. In the 3D printing case, as materials exhibit the cylindrical shape in the syringe, nozzle, and oxidized skin, the characteristic length could be regarded as the diameter d_s of the syringe, the diameter d_n of the nozzle, and the diameter d_o of the oxidized skin respectively.

μ stands for the dynamic viscosity of the fluid materials.

After substituting $Re = \frac{\rho u L_{char}}{\mu}$ and $f_D = \frac{64}{Re}$ into the Darcy–Weisbach equation, the total pressure loss due to viscosity over the three segments would be written as

$$\Delta P_{nl} = \left(\frac{32z}{d_s^2} + \frac{32a}{d_n^2} + \frac{32L}{d_o^2} \right) v_{print} \cdot \mu$$

For the convenience of writing, we set $k_{nl} = \frac{32z}{d_s^2} + \frac{32a}{d_n^2} + \frac{32L}{d_o^2}$.

Then, $\Delta P_{nl} = k_{nl} v_{print} \mu$.

This equation describes the total pressure loss ΔP_{nl} inside the syringe, nozzle, and oxidized skin due to the viscosity of the materials. This term is added to the pressure formula $P = \rho g \Delta h - P_{loss}$ mentioned in the supplementary section “Mechanical Analysis” as a correction term to derive the viscosity thresholds for successful printing. To clarify, P_{loss} is caused by the diameter change at the interface of syringe and nozzle, while ΔP_{nl} is induced by the viscosity. The specific calculation method of P_{loss} is mentioned in the reference^{S14}.

Then, the pressure given by the molten metal inside the printer could be written as

$$\begin{cases} P = \rho g \Delta h - P_{loss} - \Delta P_{nl} \\ \Delta P_{nl} = k_{nl} v_{print} \mu \end{cases}$$

The upper threshold of viscosity is defined when the gravity of the ink could overcome the pressure loss caused by the viscosity and the diameter change at the interface, so that the material could flow out of the nozzle. In this case, the inequality for determining the upper threshold of viscosity could be written as

$$\rho g \Delta h - P_{loss} - \Delta P_{nl} > 0$$

After substituting the formula of ΔP_{nl} into the inequality, the upper viscosity threshold for successful printing could be determined as below.

$$\mu < \frac{\rho g \Delta h - P_{loss}}{k_{nl} v_{print}}$$

The lower threshold of viscosity mainly depends on the yield stress σ_{yield} of the oxidized skin. Figure S18 and Figure S19 illustrate the stress analysis of the failure of the oxidized skin in the cylindrical coordinate system. To realize smooth 3D printing, the longitudinal stress σ_{zz} should be smaller than the yield stress σ_{yield} to avoid necking or breaking, enabling smooth elongation of metal wires in longitudinal direction^{S22}. Simultaneously, the circumferential stress $\sigma_{\theta\theta}$ should be always smaller than the oxide yield stress to maintain the uniform feature and prevent bulge in the printed filament. According to the formulas of longitudinal stress σ_{zz} , circumferential stress $\sigma_{\theta\theta}$, and the maximum principal stress theory^{S22}, we could obtain following inequality.

$$\begin{cases} \sigma_{\theta\theta} < \sigma_{yield} \\ \sigma_{\theta\theta} = \frac{P d_o}{2} \\ \sigma_{zz} < \sigma_{yield} \\ \sigma_{zz} = \frac{P \pi d_o^2}{4} + R_v \sin \psi + R_h \cos \psi + w L \sin \psi \end{cases}$$

In which,

R_v and R_h stand for the vertical and horizontal components of reaction force on the phase change interface, respectively. They could be obtained through experiments.

ψ stands for the tilt angle during the printing.

w stands for the weight force distribution of the metal filament along the printing direction.

L stands for the liquid length of the printed but unsolidified wire.

d_o stands for the diameter of the oxidized skin.

After substituting all the equations into the inequality, the lower viscosity threshold for successful printing could be determined as below. The two inequalities should be fulfilled simultaneously.

$$\begin{cases} \mu > \frac{1}{k_{nl}v_{print}} \left(\rho g \Delta h - P_{loss} - \frac{2\sigma_{yield}}{d_o} \right) \\ \mu > \frac{1}{k_{nl}v_{print}} \left[\rho g \Delta h - P_{loss} - \frac{4(\sigma_{yield} - R_v \sin \psi - R_h \cos \psi - wL \sin \psi)}{\pi d_o^2} \right] \end{cases}$$

To conclude, the viscosity range for successful printing can be determined as below.

$$\begin{cases} \mu < \frac{\rho g \Delta h - P_{loss}}{k_{nl}v_{print}} \\ \mu > \frac{1}{k_{nl}v_{print}} \left(\rho g \Delta h - P_{loss} - \frac{2\sigma_{yield}}{d_o} \right) \\ \mu > \frac{1}{k_{nl}v_{print}} \left[\rho g \Delta h - P_{loss} - \frac{4(\sigma_{yield} - R_v \sin \psi - R_h \cos \psi - wL \sin \psi)}{\pi d_o^2} \right] \end{cases}$$

Furthermore, the viscosity of the metals in the liquid state could also be influenced by the shear rate $\dot{\gamma}$ and temperature T , which could be defined as $\mu(T, \dot{\gamma})$. The formula of $\mu(T, \dot{\gamma})$ could be determined by the curve fitting of rheology experiment data.

Therefore, when generalizing tension-driven 3D printing to different metals, the above inequality could be used to determine the upper and lower viscosity thresholds for smooth printing. The printable viscosity range is mainly regulated by nozzle size, printing speed, liquid volume in syringe, and yield stress of the oxide skin.

References:

- S1. Guo SZ, Qiu K, Meng F, Park SH, McAlpine MC. 3D Printed Stretchable Tactile Sensors. *Adv Mater* 2017, **29**(27).
- S2. Zhu Z, Guo SZ, Hirdler T, Eide C, Fan X, Tolar J, *et al.* 3D Printed Functional and Biological Materials on Moving Freeform Surfaces. *Adv Mater* 2018, **30**(23): e1707495.
- S3. Ahn BY, Duoss EB, Motala MJ, Guo X, Park SI, Xiong Y, *et al.* Omnidirectional printing of flexible, stretchable, and spanning silver microelectrodes. *Science* 2009, **323**(5921): 1590-1593.
- S4. Jo Y, Kim JY, Kim SY, Seo YH, Jang KS, Lee SY, *et al.* 3D-printable, highly conductive hybrid composites employing chemically-reinforced, complex dimensional fillers and thermoplastic triblock copolymers. *Nanoscale* 2017, **9**(16): 5072-5084.
- S5. Zhou N, Liu C, Lewis JA, Ham D. Gigahertz Electromagnetic Structures via Direct Ink Writing for Radio-Frequency Oscillator and Transmitter Applications. *Adv Mater* 2017, **29**(15).
- S6. Skylar-Scott MA, Gunasekaran S, Lewis JA. Laser-assisted direct ink writing of planar and 3D metal architectures. *Proc Natl Acad Sci U S A* 2016, **113**(22): 6137-6142.
- S7. Zhu Z, Park HS, McAlpine MC. 3D printed deformable sensors. *Sci Adv* 2020, **6**(25): eaba5575.
- S8. Yuk H, Lu B, Lin S, Qu K, Xu J, Luo J, *et al.* 3D printing of conducting polymers. *Nat Commun* 2020, **11**(1): 1604.
- S9. Tang X, Zhou H, Cai Z, Cheng D, He P, Xie P, *et al.* Generalized 3D Printing of Graphene-Based Mixed-Dimensional Hybrid Aerogels. *ACS Nano* 2018, **12**(4): 3502-3511.
- S10. Neumann TV, Facchine EG, Leonardo B, Khan S, Dickey MD. Direct write printing of a self-encapsulating liquid metal-silicone composite. *Soft Matter* 2020, **16**(28): 6608-6618.
- S11. Park YG, Min H, Kim H, Zhexembekova A, Lee CY, Park JU. Three-Dimensional, High-Resolution Printing of Carbon Nanotube/Liquid Metal Composites with Mechanical and Electrical Reinforcement. *Nano Lett* 2019, **19**(8): 4866-4872.
- S12. Park YG, An HS, Kim JY, Park JU. High-resolution, reconfigurable printing of liquid metals with three-dimensional structures. *Sci Adv* 2019, **5**(6): eaaw2844.
- S13. Ladd C, So JH, Muth J, Dickey MD. 3D printing of free standing liquid metal microstructures. *Adv Mater* 2013, **25**(36): 5081-5085.
- S14. Gannarapu A, Gozen BA. Freeze-Printing of Liquid Metal Alloys for Manufacturing of 3D, Conductive, and Flexible Networks. *Advanced Materials Technologies* 2016, **1**(4).
- S15. Andrews JB, Mondal K, Neumann TV, Cardenas JA, Wang J, Parekh DP, *et al.* Patterned Liquid Metal Contacts for Printed Carbon Nanotube Transistors. *ACS Nano* 2018, **12**(6): 5482-5488.
- S16. Boley JW, White EL, Chiu GTC, Kramer RK. Direct Writing of Gallium-Indium Alloy for Stretchable Electronics. *Advanced Functional Materials* 2014, **24**(23): 3501-3507.
- S17. Gannarapu A, Dutta P, Gozen BA. Prediction of steady-state freeze front position during 3D printing of microstructures. *International journal of heat and mass transfer* 2017, **115**: 743-753.
- S18. Bejan A. *Convection heat transfer*. John wiley & sons, 2013.

- S19. Lipchitz A. Characterization of Eutectic In-Bi-Sn Alloy (Field's Metal) for Use in Single and Two-phase Liquid Metal Flow in Natural Circulation Systems. UOIT, 2015.
- S20. Acton QA. Heavy Metals—Advances in Research and Application: 2013 Edition. 2013.
- S21. Manasijević I, Balanović L, Grgurić TH, Minić D, Gorgievski M. Study of microstructure and thermal properties of the low melting Bi-In-Sn eutectic alloys. *Materials Research* 2018, **21**.
- S22. Gannarapu A, Gozen BA. Micro-extrusion-based additive manufacturing with liquid metals and alloys: Flow and deposition driven by oxide skin mechanics. *Extreme Mechanics Letters* 2019, **33**: 100554.
- S23. Martin A, Chang BS, Pauls AM, Du C, Thuo M. Stabilization of undercooled metals via passivating oxide layers. *Angewandte Chemie International Edition* 2021, **60**(11): 5928-5935.
- S24. Zamora R, Martínez-Pastor J, Faura F. Thermal, viscoelastic and surface properties of oxidized Field's metal for additive microfabrication. *Materials* 2021, **14**(23): 7392.
- S25. Gorissen, B. et al. Hardware Sequencing of Inflatable Nonlinear Actuators for Autonomous Soft Robots. *Advanced Materials* 2019, **31**, 1804598.
- S26. Kim, S. K. Darcy friction factor and Nusselt number in laminar tube flow of Carreau fluid. *Rheol Acta* 2022, **61**, 243–255.
- S27. Cheng, Z. et al. Investigation of drag reduction by slurry-like drag-reducing agent in microtube flow using response surface methodology (RSM). *Sci Rep* 2023, **13**, 22433.