Supplemental Information for

Depth-controlled laser-induced jet injection for direct three-dimensional liquid delivery

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1. DIMENSIONLESS PRINTABILITY DIAGRAM OF LIFT

1.1. Dimensionless numbers

To analyze the jetting regimes of the laser-assisted device, previous works[1,2] used a dimensionless parametric space based on the Deborah De^* and Ohnesorge Oh^* numbers, modified to account for the specificities of laser-induced forward transfer[1].

$De^* = \lambda\omega_R(\frac{V_c}{V_{jet}})$ represents the ratio of the characteristic internal relaxation time of the liquid λ over the time scale of the deformation applied to the liquid ${\omega_R}^{-1}$. The ratio $(\frac{V_c}{V_{jet}})$ introduced by Turkoz et al.[1] accounts for the stretching of the laser-induced jet filament, where $V_c = \sqrt{\frac{\sigma}{\rho H}}$ is the breakup velocity of the jet (σ is the ink's surface tension, ρ its density and H the thickness of the ink layer), and V_{jet} is the average jet velocity. In other words, De^* is used to predict if the liquid jet relaxes before capillary effects arise to break it up into one or more droplets.

In the same way, $Oh^* = \frac{\eta(\dot{\gamma})}{\sqrt{\rho\sigma H}}$ is the ratio of the characteristic time scales for capillary and viscous effects to respectively appear, where $\eta(\dot{\gamma})$ is the shear-dependent ink's viscosity, ρ the ink's density, σ the ink's surface tension and H the thickness of the ink layer.

Using these dimensionless numbers, previous studies[1,2] defined a predictive semi-empirical model for the jetting regimes of laser-induced forward transfer, as shown in Fig. S1.

Briefly, below a threshold value De^* ~0.2, the liquid relaxes too fast for jetting to take place (see *no jetting:liquid relaxation* region in Fig. S1). In addition, viscous effect will prevent jetting or capillary breakup for large Ohnesorge numbers (Oh^*>~1). On the other hand, capillary effects dominate for small Ohnesorge numbers (Oh^*<~ 0.1), thus resulting in the generation of multiple droplets (see *multiple drops* region in Fig. S1). Finally, for $Oh^* > De^*$ and De^* ~0.2, jetting is possible but viscous effects will dominate the jetting process and prevent breakup of the liquid filament (see *jetting without breakup* region in Fig. S1). All these criteria allow defining a parametric space for single drop generation, as shown in grey in Fig. S1 and further described in previous works[1,2].

1.2. Experimental results

By superimposing our experimental data points with the printability diagram, we observe that the experimental data points lie at the border of the viscosity threshold for breakup (see blue circles and diagonal line $De^* > Oh^*$ in Fig. S1) .

We effectively imaged jetting with breakup for the lowest velocities (see Figs. 2(b)-(d)), and jetting without breakup for the highest velocities as accurately predicted by the diagram. However, as Oh* ∼1 for our ink, we do not expect the filament to breakup and generate multiple drops as experimentally imaged (see Figs. 2(b)-(d)).

Table S1 summarizes the parameters used for plotting the experimental results, as well as their references.

Table S1. Parameters for the dimensionless analysis of the laser-induced jetting regimes

Parameter	Value	Reference
V_{jet} mean jet velocity (s^{-1})	10-81	experimental data
$\dot{\gamma}$ shear rate (s^{-1})	0.35-6.8·10^5	experimental data
σ ink surface tension (N m)	44.6·10^{-3}	[2]
ρ ink density (kg m^{-3})	1010	[2]
$\eta(\dot{\gamma})$ ink viscosity (Pa s)	27·10^{-3}	[3]
V_c breakup speed (m s^{-1})	1.7	[1]
λ characteristic relaxation time (s)	560·10^{-6}	[2]
ω_r capillary instability growth rate (s^{-1})	3.9·10^4	[1,2,4]
De^* modified Deborah number	0.46-3.3	experimental data,[1–3]
Oh^* modified Ohnesorge number	1.0	experimental data,[2,3]

1.3. Predictive jetting regimes with modification of the ink

The ink being composed of water, glycerol and sodium alginate (see section II.A), we study how a change in the concentration of sodium alginate would affect the jetting process.

Not only will a modification of sodium alginate concentration change the characteristic relaxation time of the ink[2], but it will also modify the ink's viscosity[3]. A decreasing sodium alginate concentration c_{alg} will result in a shorter relaxation time λ and therefore in a decreasing Deborah number De^*[5]. Similarly, a decreasing sodium alginate concentration will result in a decreasing viscosity and therefore in a decreasing Ohnesorge number Oh^*.

Assuming that the concentration change of sodium alginate does not affect the jetting

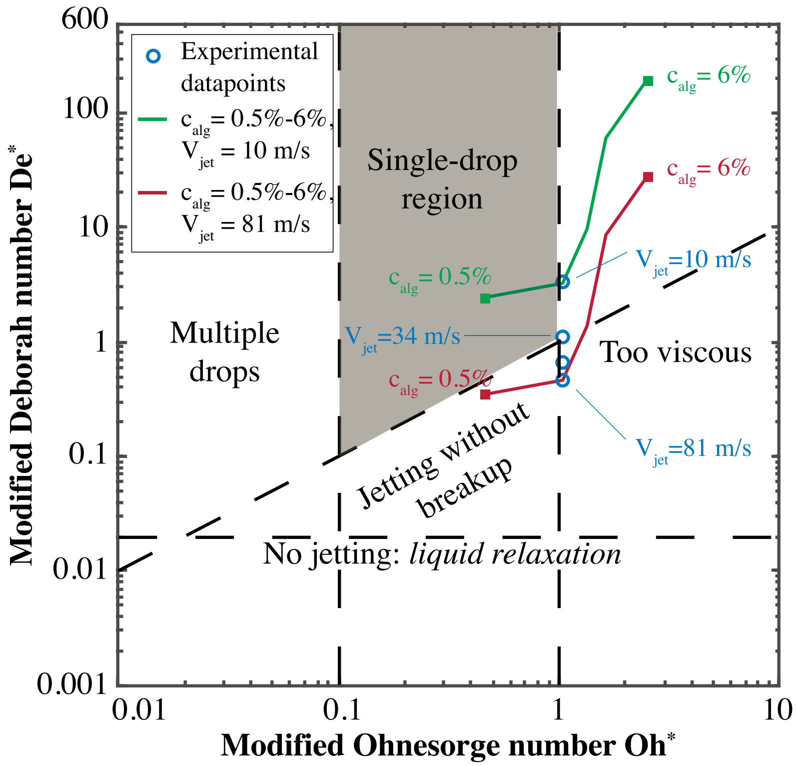

Figure S1. Dimensionless analysis of the laser-induced jetting process

velocity, the parametric curve $Oh^*(c_{alg})$-$De^*(c_{alg})$ for sodium alginate concentration between 0.5% and 6% of the volume and 10-m s^{-1} and 81-m s^{-1} jetting velocities were plotted, as respectively depicted by the green and red curves in Fig. S1.

We observe that stable single-droplet generation is more likely to take place if the sodium alginate concentration is decreased. This physically means that capillary effects are favored by this concentration decrease, which will be studied in future work.

REFERENCES

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[5]It is noteworthy that the ink's surface tension and density will also slightly evolve with the sodium alginate concentration, which we account for plotting the curves of Fig. S1.