Towards satellite free drop-on-demand printing of complex fluids

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Abstract

We investigate the influence of fluid properties on jet breakup in the context of drop-on-demand inkjet printing. In drop-on-demand printing, each drop remains temporarily connected to the printhead by a ligament which thins while the drop is in flight. Upon pinch-off the severed ligament may recoil into the leading drop, or it may fragment into 'satellite drops' which reduce printing resolution. A key goal of inkjet research is to prevent or impede the creation of satellite drops while maintaining a high drop speed.

Complex fluids often exhibit enhanced resistance to fragmentation in jetting flows compared to Newtonian fluids of similar viscosity [1]. Indeed, some complex fluids have been found in experiments to produce satellite-free jets even at high drop speeds [2]. In this work we exploit rheological considerations with the aim of eliminating satellite drops when printing at prescribed drop speeds, without any alterations to the driving waveform other than a simple amplification. In explicit terms, we attempt to design the rheology of the fluid in such a way as to calibrate its effective viscosity during the key stages of a drop-on-demand flow cycle.

Using a purely shear-thinning fluid model, we outline the key fluid parameters and dimensionless groups and we use a Lagrangian finite-element numerical method [3] to simulate a drop-on-demand printing flow under realistic industrial inkjet conditions, exploring the parameter space of critical fluid properties for a variety of drop speeds. We show that a shear-thinning fluid model with calibrated viscosity plateaus is able to eliminate satellites without compromising on drop speed and without adjusting the driving waveform.

The present study complements previous work in which we have presented results of drop-on-demand simulations for a viscoelastic fluid model with constant shear-viscosity [4], a generalized Newtonian fluid model (shear-thinning) [5], and the Giesekus fluid model which is both viscoelastic and shear-thinning [6]. In each case we demonstrated the capacity of non-Newtonian fluid properties to reduce the number and net volume of satellite drops. In these works, the elimination or reduction of satellites was considered only as a desirable potential outcome; by contrast, in the current work the elimination of satellites is imposed as a requirement.

Introduction

Among the major challenges in contemporary inkjet research are the enhancements of the speed, resolution, and material diversity of the process in order to broaden the range of industrial and commercial applications. These challenges may be addressed in a number of ways depending on the extent to which each constituent part of the process may be modified, for example, by redesigning the internal structure of the printhead or changing the actuation waveform that drives the jetting, or by adjusting the fundamental fluid properties of the ink (chiefly its viscosity and surface tension). An extension of the latter approach involves the exploitation of non-Newtonian fluid phenomena, and in recent times this has led to the development of several new applications of inkjet technology [7] [8]. Viscoelastic effects may influence jetting behaviour profoundly [9] [10], as may the presence of a particulate phase [11] [12].

Due to the various ingredients necessary to provide the required levels of performance and chemical stability, the majority of fluids used in real inkjet applications are, in essence, colloidal suspensions and consequently they may exhibit some degree of non-Newtonian behaviour when subjected to rheological characterization [13]. The local variation of viscosity in such a fluid can influence the breakup dynamics of a particle-laden jet compared to those of Newtonian jets [12], and may therefore determine whether the fluid is suitable for a particular application. Conversely, the capacity to impose explicit control upon the breakup behaviour, by making appropriate modifications to the rheological properties of the ink, is an important versatility requirement of industrial and commercial printing technologies [14]. A detailed understanding of the effects of rheology on breakup is thus an ongoing ambition of inkjet research.

In a drop-on-demand (DOD) inkjet printer each individual drop is formed by the ejection of a finite ligament of ink from one of an array of nozzles located directly above the target area for deposition. This ligament subsequently either disintegrates into a main drop and a series of smaller 'satellite' drops, or, preferably, contracts to form a single drop, before impacting on the target substrate. The fate of the ligament depends mainly on the speed of printing and on the ink viscosity and surface tension (i.e. the Reynolds, Weber, and Ohnesorge numbers of the flow) which control its rate of capillary thinning [15]. In almost all applications the production of satellite drops in significant number or volume is considered highly undesirable due to reduced resolution in the printed output.

Historically there have been few studies of the printing of non-Newtonian fluids, although the subject has undergone substantial development in recent years as the complexity of the fluid dynamics and the rich potential for novel applications have fueled research in this area [7] [8]. Experimental studies of the DOD printing of polymer solutions have revealed a diverse family of viscoelastic jet behaviour depending on the concentration and molecular weight of the polymer [16] [17], which have been reproduced in simulations by the present authors, both qualitatively [4] and quantitatively [5]. Even small amounts of polymer may cause severely different breakup dynamics compared to Newtonian printing, influencing both in-flight fragmentation and detachment from the nozzle, and significant concentrations may also impede jettability due to high elasticity in the ligament.

Shear-thinning fluid models have received somewhat less attention, but have also been shown to exhibit markedly different behaviour in filament stretching flows [18] and jet breakup [19] compared to the purely Newtonian case. Doshi et al. [20] performed a thorough analysis of capillary thinning rates for a Carreau fluid [21] (and a power-law fluid as a limiting case), which complemented contemporary work by Renardy [22] [23]. Drop formation has been addressed, as in computational work by Yıldırım and Basaran [24] who studied the influence of both shear-thinning and shear-thickening rheology on dripping over a range of flow rates, finding a rich family of regular and irregular patterns; parameters conducive to the potential suppression of satellite drops were also discussed. Experiments on the formation of pendant drops of particle suspensions have also suggested the reduction of satellites [11] [12] [25]. A recent experimental survey by Clasen et al. [26] concerning the ejection of a variety of complex fluids through millimetre-sized needles has provided a valuable summary of non-Newtonian effects, including shear-thinning. In a study more specific to high-speed printing, the present authors have shown in simulations of Carreau fluids that shear-thinning behaviour may reduce both the number and sizes of satellites when printing at a prescribed main drop velocity [5], and recent experiments by Hoath et al. [2] have corroborated these overall conclusions.

In this work we consider the influence of non-Newtonian rheology on the suppression of satellite drops in DOD simulations; we use a shear-thinning fluid model and we explore its parameter space with the aim of eliminating satellite drops while maintaining high drop speed.

Numerical method and boundary conditions

The simulations use an axisymmetric Lagrangian finite-element method first developed for the study of creeping flow of dilute polymer solutions [3]. The method has since been extended to inertial flows and has been applied to inkjet printing of Newtonian and complex fluids [27] [4] [5]; details of the computational methods may be found in these references. The shape of the nozzle used in the simulations is identical to that in references [4] and [5], based on a Xaar 126 printhead with nozzle radius $R = 25\,\mu$m. At the nozzle inlet a time-dependent velocity boundary condition is imposed in the form of a 'pull/push/pull' drive waveform of $\approx 30\,\mu$s duration; this waveform is amplified in each simulation in order to establish the maximum satellite-free jet speed for each fluid modelled. Because of this amplification, the total volume of ejected ink is not necessarily constant.

We assume that the jet is axisymmetric, so that it may be fully described by a cylindrical coordinate system $\{r, \theta, z\}$ with all flow variables independent of θ. The origin is taken as the centre of the nozzle outlet, and the fluid is initially at rest. The boundary conditions at the free surface are those of zero shear stress and the pressure jump due to surface curvature,

$$\hat{\mathbf{n}} \cdot \boldsymbol{\sigma} \cdot \hat{\mathbf{t}} = 0, \qquad [\boldsymbol{\sigma} \cdot \hat{\mathbf{n}}]_{\text{air}}^{\text{jet}} = -\frac{1}{\text{We}} \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \hat{\mathbf{n}}, \qquad (1)$$

where $\boldsymbol{\sigma}$ is the dimensionless stress tensor, $\hat{\mathbf{n}}$ is the unit outward normal to the interface, $\hat{\mathbf{t}}$ is the unit tangent in the rz-plane, We is the Weber number (defined in the following section), and R_1 and R_2 are the principal radii of curvature. External air pressure is neglected. Symmetry conditions on the z-axis are $u_r = 0$ and $\sigma_{rz} = 0$, and conditions of no-slip are applied at the rigid interior printhead boundaries. The contact line (meniscus) between the free surface and the printhead is held pinned at the nozzle edge throughout.

The location of the free surface at each time-step is determined implicitly via a kinematic condition. In the simulations this is realized automatically, since the mesh is Lagrangian and the mesh nodes are advected with the local fluid velocity. Drag due to air resistance is neglected, as are variations in temperature.

Fluid model and governing equations

The governing equations are the conservation of momentum and mass for a generalized Newtonian fluid

$$\rho \frac{\text{D}\mathbf{u}}{\text{D}t} = \nabla \cdot \boldsymbol{\sigma}, \qquad \nabla \cdot \mathbf{u} = 0, \qquad (2)$$

where ρ is the fluid density, t and $\mathbf{u}$ are the time and fluid velocity respectively, and $\boldsymbol{\sigma}$ is the stress tensor which may be expressed as

$$\boldsymbol{\sigma} = -p\mathbf{I} + 2\mu(\dot{\gamma})\mathbf{E}, \qquad (3)$$

where p is pressure and $\mathbf{E} = \frac{1}{2}\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^T\right)$ is the rate of strain tensor.

Shear-thinning effects are incorporated via the response of the viscosity μ to the local shear rate $\dot{\gamma} = \sqrt{2\mathbf{E}:\mathbf{E}}$. In this work we use the Carreau fluid model [21], which describes a smooth transition from a zero shear rate viscosity μ_0 to a limiting infinite shear rate viscosity $\mu_\infty < \mu_0$. The response function is defined as:

$$\mu(\dot{\gamma}) = \mu_\infty + (\mu_0 - \mu_\infty)\left(1 + (\dot{\gamma}/\dot{\gamma}_0)^2\right)^{(n-1)/2}, \qquad (4)$$

where $\dot{\gamma}_0$ is a representative value of the shear rate associated with the onset of shear-thinning, and n is an index which determines the degree of shear-thinning in the transition; throughout this work we set $n = 0.3$.

It should be emphasized that for a generalized Newtonian fluid (GNF) the term "shear-thinning" really means "strain-rate-thinning", in the sense that the viscosity is assumed to decrease as a function of the inner product of the strain-rate tensor. Thus GNF models such as the Carreau fluid exhibit a reduced viscosity both in pure shearing flow and in purely extensional flow, in contrast to the behaviour of most polymeric fluid models which show extension hardening but may have a constant shear viscosity [28].

Using a representative velocity scale $U = 6\,\text{m}\,\text{s}^{-1}$ (based on a typical intended drop speed), we define the Reynolds number $\text{Re} = \rho U R/\mu_0$, the Weber number $\text{We} = \rho U^2 R/\Gamma$ (where Γ is the coefficient of surface tension) and the Ohnesorge number $\text{Oh} = \mu_0/\sqrt{\rho \Gamma R} = \sqrt{\text{We}}/\text{Re}$. Gravity is negligible on the lengthscales considered in this study. Throughout this work we keep ρ and Γ constant, with $\text{We} \approx 14$, and we vary the zero shear rate viscosity μ_0 (hence Re and Oh); typical values considered are $\text{Re} \approx 10$ and $\text{Oh} \approx 0.4$, which are realistic values for many inkjet applications. For flows involving jet breakup it is common to define a capillary time $t_c = \sqrt{\rho R^3/\Gamma}$ as the relevant timescale for inertio-capillary thinning [29].

Rheology and implications

In high-speed printing applications a fluid element experiences extreme shear as it is ejected at a speed of order 6 m/s through a nozzle of diameter 50 μm or less, and the same fluid element may then experience significant extension during the capillary thinning of the ejected ligament. The shear rates in the nozzle and the thinning ligament may differ by an order of magnitude, and so a shear-thinning fluid is likely to undergo a substantial fluctuation in its viscosity during the printing process. Industrial rheologists may be able to exploit this behaviour by producing an ink which has a suitable viscosity to shear-rate response (i.e. high viscosity in the ligament, low viscosity in the nozzle), thereby optimizing the performance of the ink during each stage of the flow.

In order to choose appropriate values of the parameters in the Carreau fluid model, we consider how the viscosity of the fluid should vary during the three key phases of a drop-on-demand cycle. Firstly there is high shear during the 'push' stage of the driving waveform as fluid is ejected rapidly through the nozzle; in this phase the viscosity should be low for ease of ejection without excessive amplification (i.e. voltage). Secondly there is extension during the 'pull' stage of the driving which draws out a ligament behind the leading drop; the viscosity should remain quite low in this phase to allow prompt detachment of the ligament from the printhead, but not so low that the capillary instability develops fully along the ligament while it is still attached. Thirdly there is the thinning of the detached ligament, during which phase the viscosity should be high in order to delay breakup and maximize the proportion of ink which ultimately ends up within the main drop, ideally 100% (no satellites). We therefore wish to find values for the Carreau model parameters which optimize the fluid properties in each of these key phases.

An example case of the Carreau response function $\mu(\dot{\gamma})$, as defined in Eq. 4, is shown in Fig. 1; for this case the model parameters were $\mu_0 = 50\,\text{mPa s}$, $\mu_\infty = 3\,\text{mPa s}$, and $\dot{\gamma}_0 = 10^5\,\text{s}^{-1}$.

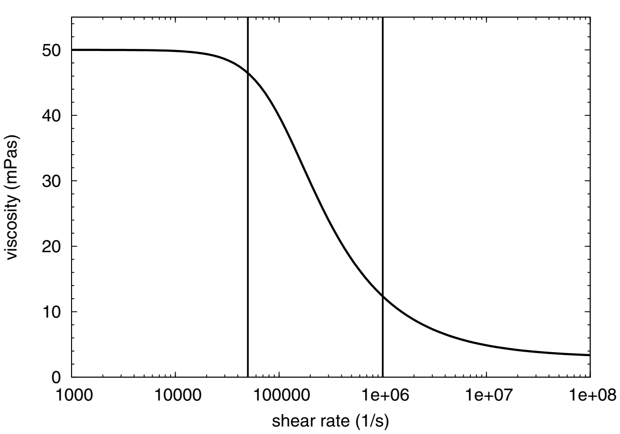

Figure 1. *The Carreau fluid model response function* $\mu(\dot{\gamma})$.

The two vertical lines plotted in Fig. 1 represent the estimated range of deformation rates most relevant to the key DOD flow phases discussed above. The right-hand line ($\dot{\gamma} = 10^6\,\text{s}^{-1}$) is an approximate maximum shear rate in the nozzle (achieved during the 'push' phase of the ejection), and the left-hand line ($\dot{\gamma} \approx 1/t_c \approx 5\times10^4\,\text{s}^{-1}$) is an estimate of the relevant deformation rate during the capillary thinning of the ejected ligament. The interval between the two lines corresponds to the range of shear rate variation during the critical stages of ligament ejection and breakup, and thus our value of $\dot{\gamma}_0$ has been chosen in order that the transition between the upper and lower viscosity plateaus (μ_0 and μ_∞) occurs precisely in this range.

Satellite-free jet speeds

In this study we keep $\dot{\gamma}_0$ fixed (equal to $10^5\,\text{s}^{-1}$) and consider the effects of varying μ_0 and μ_∞. The results of varying $\dot{\gamma}_0$ have been considered in a previous study [5]. For each case, simulations were repeated iteratively with increasing amplitudes of the driving waveform in order to determine the maximum satellite-free jetting speed for each fluid. In general, as the amplitude increases so the jet speed increases until a certain critical speed beyond which the ligament no longer contracts fully and instead satellite drops are formed. Fig. 2 shows the influence of rheology on the maximum satellite-free jetting speed. Fluids with $\mu_\infty = 10\,\text{mPa s}$ are superior to Newtonian cases whereas fluids with lower values of μ_∞ are superior only if μ_0 is above 60 mPa s, in which case is it possible to print without satellites at significantly higher jet speeds.

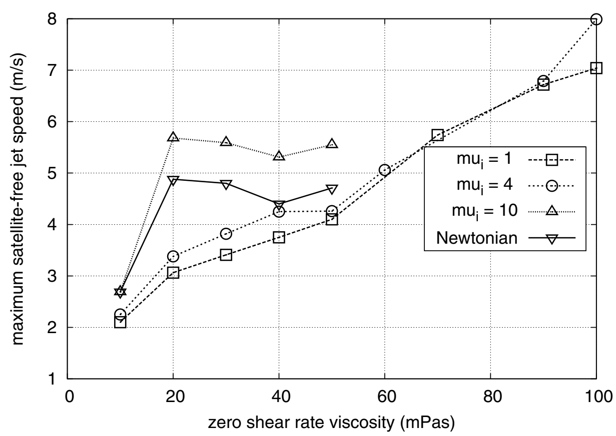

Figure 2. *Maximum satellite-free jet speeds as a function of zero shear rate viscosity* μ_0 *and infinite shear rate viscosity* μ_∞*. Each data-point represents a different fluid.*

At present we have considered only those cases for which the driving amplitude for the maximum satellite-free jet was within a factor of 1.5 times its default value (defined as the amplitude required to jet a particular Newtonian fluid at a speed of 6 m/s), as excessive amplification without altering the shape or period of the waveform would be considered unrealistic in some applications. However, depending on the priorities or limitations of a particular application, shear-thinning rheology may improve the capacity to jet faster without forming satellites. In particular, in Fig. 3 the values of the amplification factor are plotted together with the corresponding jet speeds, and these results show that shear-thinning fluids jet faster at the same drive amplitude compared to Newtonian fluids.

It should also be noted that in this study we do not incorporate any collisions or coalescence of droplets after ligament breakup has occurred. In some cases it is apparent that such coalescence does occur, which, if taken into account, would result in a redefinition of the maximum satellite-free jet speed and cor-

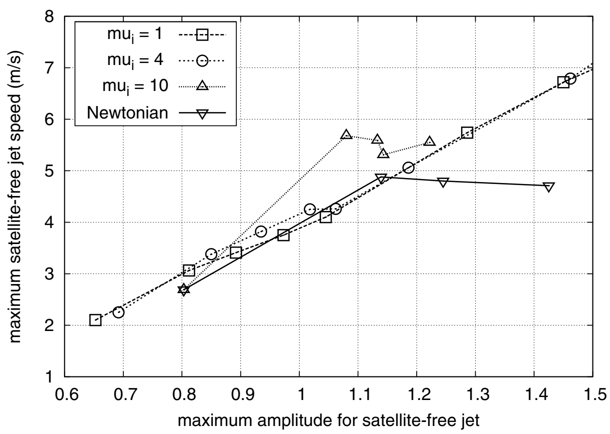

Figure 3. *Comparison of jet speeds and waveform amplitudes in maximal satellite-free cases. Each data-point represents a different fluid.*

responding adjustment of the results. However, as our aim is to consider non-Newtonian effects on the flow dynamics *prior* to ligament breakup, we choose not to incorporate such adjustments.

The preliminary results shown here demonstrate the clear potential of shear-thinning rheology to enable inkjet printing at increased jet speeds without satellite drops and without altering the driving waveform beyond a simple amplification.

Acknowledgments

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References

[1] J. Eggers and E. Villermaux, "*Physics of liquid jets*," Rep. Prog. Phys., **71** 036601 (2008).

[2] S.D. Hoath, S. Jung, W.-K. Hsiao, and I.M. Hutchings, "*How PEDOT:PSS solutions produce satellite-free inkjets*," Org. Electron., **13 (12)** 3259 (2012).

[3] O.G. Harlen, J.M. Rallison, and P. Szabó, "*A split Lagrangian-Eulerian method for simulating transient viscoelastic flows*," J. non-Newtonian Fluid Mech., **60** 81 (1995).

[4] N.F. Morrison and O.G. Harlen, "*Viscoelasticity in Inkjet Printing*," Rheo. Acta, **49** 619 (2010).

[5] N.F. Morrison and O.G. Harlen, "*Inkjet printing of non-Newtonian fluids*," Proc. NIP27, pg. 360 (2011).

[6] N.F. Morrison and O.G. Harlen, "*Drop-on-demand printing of complex liquids*," Proc. NIP29, pg. 271 (2013).

[7] O.A. Basaran, H. Gao, and P.P. Bhat, "*Nonstandard inkjets*", Annu. Rev. Fluid Mech., **45** 85 (2013).

[8] B.-J. de Gans, P.C. Duineveld, and U.S. Schubert, "*Inkjet printing of polymers: state of the art and future developments*," Adv. Mater., **16** 203 (2004).

[9] M. Goldin, J. Yerushalmi, R. Pfeffer, and R. Shinnar, "*Breakup of a laminar capillary jet of a viscoelastic fluid*," J. Fluid Mech., **38** 689 (1969).

[10] Y. Christanti and L.M. Walker, "*Effect of fluid relaxation time of dilute polymer solutions on jet breakup due to a forced disturbance*," J. Rheol., **46** 733 (2002).

[11] R.J. Furbank and J.F. Morris, "*An experimental study of particle effects on drop formation*," Phys. Fluids, **16** 1777 (2004).

[12] R.J. Furbank and J.F. Morris, "*Pendant drop thread dynamics of particle-laden liquids*," Int. J. Multiphase Flow, **33** 448 (2007).

[13] D.C. Vadillo, A.C. Mulji, and M.R. Mackley, "*The rheological characterization of linear viscoelasticity for ink jet fluids using piezo axial vibrator and torsion resonator rheometers*," J. Rheol., **54** 781 (2010).

[14] B. Derby, "*Inkjet printing of functional and structural materials: fluid property requirements, feature stability, and resolution*," Annu. Rev. Mater. Res., **40** 395 (2010).

[15] J. Eggers, "*Nonlinear dynamics and breakup of free-surface flows*," Rev. Mod. Phys., **69** 865 (1997).

[16] A.V. Bazilevskiy, J.D. Meyer, and A.N. Rozhkov, "*Dynamics and breakup of pulse microjets of polymeric liquids*," Fluid Dynamics, **40** 376 (2005).

[17] S.D. Hoath, G.D. Martin, J.R. Castrejón-Pita, and I.M. Hutchings, "*Satellite formation in drop-on-demand printing of polymer solutions*," Proc. NIP23, pg. 331 (2007).

[18] O.E. Yildirim and O.A. Basaran, "*Deformation and breakup of stretching bridges of Newtonian and shear-thinning liquids: comparison of one- and two-dimensional models*," Chem. Eng. Sci., **56** 211 (2001).

[19] Z. Gao and K. Ng, "*Temporal analysis of power law liquid jets*," Comput. Fluids, **39** 820 (2010).

[20] P. Doshi, R. Suryo, Ö.E. Yıldırım, G.H. McKinley, and O.A. Basaran, "*Scaling in pinch-off of generalized Newtonian fluids*," J. non-Newtonian Fluid Mech., **113** 1 (2003).

[21] P.J. Carreau, D. De Kee, and M. Daroux, "*An analysis of the viscous behaviour of polymeric solutions*," Can. J. Chem. Eng., **57** 135 (1979).

[22] M. Renardy, "*Self-similar jet breakup for a generalized PTT model*," J. non-Newtonian Fluid Mech., **103** 261 (2002).

[23] M. Renardy and Y. Renardy, "*Similarity solutions for breakup of jets of power law fluids*," J. non-Newtonian Fluid Mech., **122** 303 (2004).

[24] Ö.E. Yıldırım and O.A. Basaran, "*Dynamics of formation and dripping of drops of deformation-rate-thinning and -thickening liquids from capillary tubes*," J. non-Newtonian Fluid Mech., **136** 17 (2006).

[25] M.S. van Deen, T. Bertrand, N. Vu, D. Quéré, E. Clément, and A. Lindner, "*Particles accelerate the detachment of viscous liquids*", Rheol. Acta, **52** 403 (2013).

[26] C. Clasen, P.M. Phillips, L. Palangetić, and J. Vermant, "*Dispensing of rheologically complex fluids: the map of misery*," AIChE Journal, **58** 3242 (2012).

[27] J.R. Castrejón-Pita, N.F. Morrison, O.G. Harlen, G.D. Martin, and I.M. Hutchings, "*Experiments and Lagrangian simulations on the formation of droplets in drop-on-demand mode*," Phys. Rev. E, **83** 036306 (2011).

[28] M.D. Chilcott and J.M. Rallison, "*Creeping flow of dilute polymer solutions past cylinders and spheres*," J. non-Newtonian Fluid Mech., **29** 381 (1988).

[29] C. Clasen, J.Eggers, M.A. Fontelos, J. Li, and G.H. McKinley, "*The beads-on-string structure of viscoelastic threads*", J. Fluid Mech., **556** 283 (2006).

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