Journal of Non-Newtonian Fluid Mechanics 221 (2015) 55–65

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: http://www.elsevier.com/locate/jnnfm

The effect of shear-thickening on liquid transfer from an idealized gravure cell

Sunilkumar Khandavalli [a], J. Alex Lee [b], Matteo Pasquali [b,c,d], Jonathan P. Rothstein [a,*]

[a] Mechanical and Industrial Engineering, University of Massachusetts Amherst, MA 01003, USA
[b] Chemical and Biomolecular Engineering, Rice University, Houston, TX 77005, USA
[c] Department of Chemistry, Ken Kennedy Institute for Information Technology, Rice University, Houston, TX 77005, USA
[d] Department of Chemistry, The Smalley Institute for Nanoscale Science & Technology, Rice University, Houston, TX 77005, USA

ARTICLE INFO

Article history:
Received 6 January 2015
Received in revised form 25 March 2015
Accepted 29 March 2015
Available online 15 April 2015

Keywords:
Gravure printing
Coating
Shear thickening
Colloids
Extensional rheology
Finite element method

ABSTRACT

Gravure printing is an economical roll-to-roll processing technique with potential to revolutionize the fabrication of nano-patterned thin films at high throughput. In the present study, we investigated the impact of shear-thickening on the liquid transfer from an idealized gravure cell by a combination of experiments and numerical computations. We chose as a model system fumed silica nanoparticles dispersed in polypropylene glycol; these dispersions exhibit shear and extensional thickening as verified by steady shear and filament stretching extensional rheometry. Model gravure printing experiments were conducted using a linear motor to pick out the fluid vertically up from a truncated conical shaped idealized gravure cell cavity; the cell size is large enough that gravity is important, and therefore experiments were also conducted to pickout the fluid vertically down from the cavity-on-top. The amount liquid transfer from the cavity was studied with varying stretch velocities and dispersion concentrations. The filament profile evolution during the pickout process was examined using a high speed camera. Beyond a critical stretch rate, shear-thickening of the fluid, manifested by the formation of long stable filaments, exacerbates gravitational drainage during pickout. Beyond a second critical stretch rate, shear-thinning induces conical profile evolutions that result in pickout insensitive to stretch rate. All of these observations were qualitatively predicted by finite element computations using a generalized Newtonian fluid model, where the shear rheology was modeled explicitly. We showed that under the influence of gravity, wetting/de-wetting may be a critical phenomenon in determining pickout at low stretch rates.

1. Introduction

Roll-to-roll coating and printing of flexible substrates is a technology of great industrial and commercial importance due to its low cost and high throughput [1,2]. This technology enables the fabrication of thin organic, inorganic and mixed organic/inorganic films with nanoscale patterns at high resolution for devices in wide applications such as, solar cells, thin film transistors, organic light emitting diodes, biosensors and biodevices [3–8]. Gravure printing is a roll-to-roll processing technique used to coat/print thin films less than 50 µm for a wide variety of applications in high volumes such as magazines, packaging, flexible electronics, greeting cards and tapes [9–12]. In gravure process, a roller with desired engraving, typically in microns dimensions, is passed through an ink reservoir and the excess is metered off by passing by a doctor blade. The ink from the cavities is then deposited on to the substrate held by another roller at high speeds (up to 15 mm/s).

During the ink transfer process, a liquid bridge is formed and stretched between the gravure cell (the cavity) and web, as the ink is deposited onto the substrate. The liquid bridge experiences complex kinematics; a combination of shear and extension due to relative motion between the gravure cell and web [13,14]. The stability and breakup dynamics of the liquid bridge during the ink transfer can significantly affect the quality of the print or coated film. Partial emptying of the cavities or the formation of satellite drops can negatively impact the quality and the efficiency of the printing process [15]. Including gravure printing, the dynamics of the liquid bridge are strongly relevant in other applications such as contact drop dispensing [16], float-zone crystallization [17] and oil recovery [18]. Therefore, the dynamics, stability and breakup of the liquid bridge have been widely studied

---

* Corresponding author. Tel.: +1 413 577 0110; fax: +1 413 545 1027.
E-mail address: rothstein@ecs.umass.edu (J.P. Rothstein).

http://dx.doi.org/10.1016/j.jnnfm.2015.03.007
0377-0257/© 2015 Elsevier B.V. All rights reserved.

[16,18–22]. Numerous studies, both experimental and numerical, have been performed to better understand the dynamics of a liquid bridge uniaxially stretched between two flat plates [23–25], as well between a flat plate and a cavity [26–29]. There have also been numerical and experimental studies on ink transfer behavior in gravure printing considering pure shear [30–32], a combination of shear and extensional motion of the liquid bridge [33,34], as well as studies which considering the effect of the rotation of the gravure cavity as the liquid is applied to the moving web by the rotating gravure roller [34,13].

Printing fluids often are non-Newtonian, as many inks contain large concentrations of particles and polymer additives [35,7,12]. Only recently have any studied reported on the behavior of non-Newtonian fluids during gravure printing. Huang et al. [29] performed numerical computations using volume of fluid (VOF) method to examine the break up dynamics of shear-thinning fluids. They observed a fast break up of the filament and the formation of more satellite drops for the case of shear-thinning fluids compared to Newtonian fluids. Ahmed et al. [36] employed numerical computations to investigate the ink transfer behavior of shear-thinning fluids between two flat plates. They examined the influence of inertia, surface tension and top plate contact angle on the ink transfer ratio. Numerical computations on a shear-thinning fluid by Ghadiri et al. [37] showed that factors such as groove angle, groove depth and wettability can have significant impact on the filament dynamics and the amount of ink transfer.

There have also been studies both experiments and computations dedicated to the influence of viscoelasticity on gravure printing [38,10,39]. Ahn et al. [38] examined the influence of elasticity through numerical computations using Oldroyd-B model and observed significant differences in the velocity field and pressure distribution in the cavity between viscoelastic and Newtonian fluids. Sankaran and Rothstein [10] conducted experimental investigation using polyethylene oxide (PEO) fluids to study the impact of viscoelasticity, gravity and gravure cell design on the fluid transfer in gravure printing. Filament stabilization due to elasticity was found to enhance or worsen the fluid removal, depending on whether the fluid removal direction is aligned with or opposite to the direction of gravity. Computational studies later by Lee et al. [39] on the influence of viscoelasticity in gravure printing using FENE-P constitutive model were in excellent agreement with experiments [10]. Their computations were able to extend beyond the parameters of the experiments to investigate a larger viscoelastic parameter space.

In the current work, we investigated the impact of shear-thickening—namely the magnitude and critical shear rate for onset of shear-thickening—on liquid transfer from an idealized gravure cell. The model experimental setup for the gravure printing study was the same as those adopted by Sankaran and Rothstein [10]. The model fluids used in the study were shear-thickening fumed silica nanoparticle dispersions in polypropylene glycol. The shear and extensional rheology of this system has been well characterized in literature [40,41]. The role of shear-thickening on the amount of liquid transfer from an idealized gravure cell was studied by conducting experiments on nanoparticle dispersions at various particle concentrations. The experimental liquid transfer behavior was further examined by transient finite element computations using a generalized Newtonian fluid model.

2. Methods and materials

2.1. Gravure experimental setup

In our experiments, a modified version of a filament stretching extensional rheometer was used to model the pickout process for an idealized gravure cell or cavity [10]. The experimental setup, shown in Fig. 1, consists of a computer-controlled linear motor attached to a aluminum plate which is in-line with the cavity. The initially cylindrical fluid bridge was stretched vertically from the cavity with constant velocity. In real gravure processes, the fluid bridge experiences a combination of shear and extensional deformation, with the latter being dominant at later times near the mid-plane and the former being important at earlier times and near the end plate/cell. For simplicity, the touch down process that would occur in the real roll-to-roll coating is not modeled—instead the initial fluid bridge has a finite aspect ratio. A truncated cone shaped cavity was fabricated by casting PDMS (Silgard 184) onto a negative mold of the cavity machined into aluminum. The cavity has a sidewall angle of $\alpha = 75^\circ$ from horizontal, radius of $R = 2.5$ mm and a depth of $h = 1$ mm. The real gravure rollers have cavity features that are typically ten to hundred microns in order. Here the cavity dimensions were scaled for the convenience of a lab scale study. The effect of cavity size was probed numerically. A cylindrical fluid filament was held between the plate and cavity at an initial aspect ratio of $L_i/R = 0.3$, where L_i is the separation. The top plate was separated at different velocities ranging from 0.1 to 200 mm/s to a final aspect ratio of $L_f/R \sim 32$, up to which the filaments survived in only a few cases. The stretching speed corresponded roughly to printing speed. The inertia effects were negligible in these tests as the Reynolds numbers were all very low, $\mathrm{Re} < 5 \times 10^{-2}$. Reynolds number is the ratio of inertial to viscous forces given by, $\mathrm{Re} = D_0 U \rho / \eta_0$. Here D_0 and U are the characteristic diameter and velocity, while ρ and η_0 are the fluid density and viscosity. Because gravity is significant at these dimensions (the initial Bond number is greater than one, $Bo \sim 2$), tests were performed with cavity on both top and bottom configurations. Bond number quantifies the relative importance of gravity to

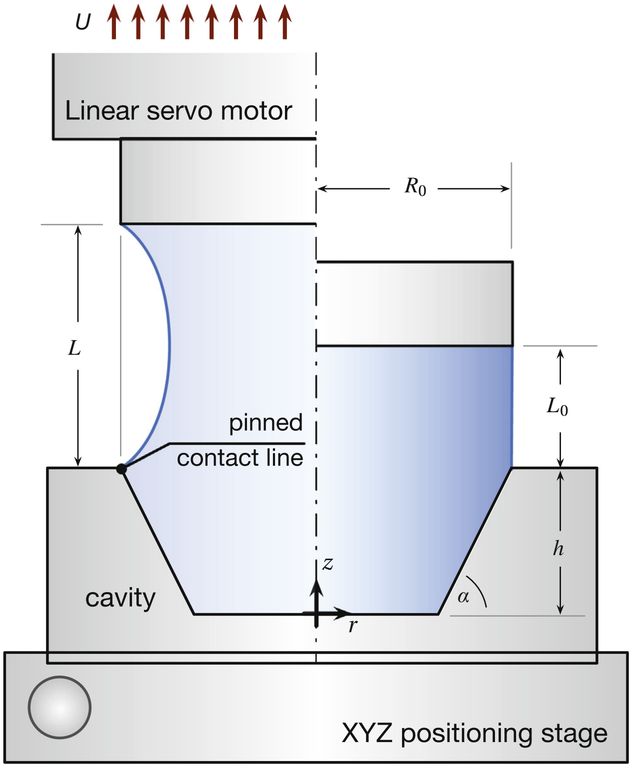

Fig. 1. Schematic diagram of experimental setup used for idealized gravure cell study.