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## Effect of viscoelasticity on liquid transfer during gravure printing

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### ABSTRACT

Roll-to-roll patterning of small-scale features on a rapidly moving web is an industrially important process with a wide array of commercial applications both old and new. Examples include magazine printing and more recently the patterning of flexible electronics. Among the many existing web coating techniques for large-scale fabrication, slit die and gravure coating are the most commonly used. In gravure coating, an engraved roller with a regular array of shallow cavities/cells is used to pick up fluid from a bath. It is then passed through a flexible doctoring blade in order to meter off excess fluid before printing the fluid onto a flexible substrate. Here we present an experimental investigation into the effect that viscoelasticity has on the dynamics of liquid transfer from an idealized gravure cell to a flat rigid substrate. Although the dynamics of the actual gravure coating process is quite complex, we chose to study a simplified process by imposing an extensional flow using a modified filament stretching rheometer in which one of the endplates is replaced by a cell containing a single truncated conical gravure cell. The deformation and stretching of the resulting liquid bridges, the motion of the contact line within the gravure cell and the total amount of fluid removed from the gravure cell are studied as a function of the imposed stretch rate, the fluid rheology, and the geometry of the gravure cell. Two different viscoelastic solutions of high molecular weight polyethylene oxide in water were studied and compared to a series of Newtonian fluids. The results show that the primary impact of viscoelasticity is the addition of an elastic stress which increases the tension along the liquid bridge and significantly increases the bridge lifetime. For stretches where the gravure cell was placed on the bottom and the top plate moved vertically, viscoelasticity was found to significantly reduce the amount of fluid transferred to the top plate. However, by placing the gravure cell on top and reversing the relative direction of the inertial and gravitational stresses, viscoelasticity was found to significantly increase the amount of fluid transferred. Increasing the stretch rate was found to amplify these observations. Finally, increasing the contact angle between the fluid and the gravure cell and decreasing the aspect ratio of the gravure cell were both found to increase the amount of fluid transferred.

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### 1. Introduction

The rapid printing and patterning of small features and the deposition of uniform thin films through roll-to-roll processing is of great importance to a variety of both old and new industries [1,2]. Classically, these printing techniques have been used to produce newspapers and magazines, but more recently they have been extended to manufacture solar panel, electronics, opto-electronics and functional materials. Among the many existing web coating and printing techniques for large-scale fabrication such as slit, roll, dip and gravure are the most commonly used. Each of these techniques has a unique set of advantages and disadvantages. The gravure process has been used widely for printing magazines, tapes and packaging [3]. The primary advantage of roll-to-roll processing that is large areas of coated/printed films can be processed quickly at low manufacturing costs. This has

been known for quite some time in traditional printing industries, but has now become of particular interest to the electronics industry who have begun to use an offset gravure printing mechanism to print electronic components [4]. In addition, a host of researchers have been investigating the adaptation of a number of water-based processing techniques like nano-inprint lithography and optical lithography techniques to web-based platforms [1].

A gravure roll-to-roll coater employs two rollers and a doctoring blade. An engraved roller is used to pick up fluid from a bath which passes through a flexible doctoring blade which meters off excess fluid. The engraved roller deposits the fluid from the cells onto the flexible substrate which is held in tension by another roller. An offset gravure process has an intermediate roller between the engraved roller and the substrate. Typical gravure cell sizes have a width ranging from 50  $\mu\text{m}$  to several hundred microns and depths in similar ranges [5,6]. A number of these cells are placed in an array on the engraved roller to print patterned arrays or coat films.

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During the gravure printing process a liquid bridge is formed between the cell and the substrate that is being coated or printed on. This bridge plays an important role in the liquid transfer process from the cell on to the substrate. The physics of the pickout process from a cell is extremely complex resulting in the stretching of a liquid bridge with strong shear and elongational flow components [7]. It is also strongly influenced by the surface properties of the cell and substrate, the limiting parameters being the contact angle between the liquid bridge and both the substrates and moving contact lines that are often pinned by the geometry of the cell [8]. There has been a many studies looking into the dynamics of liquid bridge stretching in detail [9]. The main forces that act on the bridge arise from viscosity and surface tension. The former is stabilizing and latter destabilizing. Due to high curvature of the bridge near the gravure cell and the substrate, the bridge tends to break up at the ends rather than the middle. This results in the formation of two sessile drops on either surfaces and one dynamic drop as the liquid bridge pinches off. The dynamic drop will land on either the substrate or the gravure cell depending upon the orientation of the printing system. If it were to land on the substrate it would cause a hazy print known as misting thereby reducing the quality of the print. In most cases, not all the ink is transferred from the engraved roller on to the substrate. This can in turn become a major problem because residual inks can evaporate leaving behind particulates and dissolved polymers which accumulate within the cell over time and require frequent cleaning. Clogged cells can also cause non-uniform film thickness and reduce print quality.

A number of recent articles have investigated both experimentally and computationally the gravure printing process [4,5,8–22]. In some of the earliest computational work, Powell et al. [9] simulating the printing process for a Newtonian liquid from a trapezoidal cavity. The configuration of their simulation is similar to the initial experiments reported in this paper. The gravure cell is placed upside down and a downward velocity is imposed on the substrate to simulate the printing process. In this particular configuration of the cell, gravity assists the emptying process. The authors assumed that the contact lines move down along the side-walls of the gravure cell and pinned at the bottom internal corner of the cell. They found that by increasing the substrate speed, a longer-lived filament between the cavity and the substrate was formed which in turn facilitated more removal of the fluid from the cavity. Schwartz et al. [19] presented a 2D numerical model for liquid withdrawal from cavities in gravure coating. They restricted the model to the fluid beneath the moving contact line and studied the effect of gravitational drainage for both a Newtonian and a shear thinning fluid. Small gravure cells were found to retain a large volume of liquid for extended periods of time. Schwartz [20] further investigated the effect of gravure cell shape and patterns of gravure cells on the pickout. Yin and Kumar later [21,22] studied the gravure printing process by simulating a single isolated gravure cell. They assumed the lubrication approximation for a flow between the cavity and a flexible wall and used a 1D model to investigate the effect of web flexibility on the pressure profiles inside the gravure cell. They conclude that some amount of flexibility is needed to effectively remove fluid from the cell.

Previous experimental studies have separated the process into touchdown, shear and pickout [18,22]. In this paper and many others, the emphasis has been on the pickout process. In their recent experiments, Yin and Kumar [22] used a glass top plate moving over a scaled up gravure cell to look into the flow dynamics that occur during the emptying of the cell. They measured the amount of fluid removed from the gravure cell as a function of capillary number by using different water/glycerol fluids. The amount of fluid removed, or pickout, was found to decrease with increasing capillary number for a trapezoidal cavity of aspect ratio greater than one. Later, Hoda and Kumar [14] simulated the removal of a

Newtonian liquid from a model gravure cell, they also observe that volume of liquid left in the cell behind decreases as the capillary number decreases. They were also the first to point out the importance of moving contact lines, contact angles and the normal velocity component to the cell on the amount of liquid removed. They demonstrated that imposing a normal velocity enhances the rate of emptying and removal of liquid from the cavity. Above a critical normal velocity the cells were found to completely empty. This result highlights the importance of strong elongational flow during the emptying process. A later work from Kumar's group [11] numerically investigated the slipping of contact lines in stretching of liquid bridges from an axisymmetric cavity. The fluid removal was found to increase as contact angle of the gravure cell increases and the contact angle made with the substrate decreases. They also pointed out the significance of the position of the contact line inside the gravure cell. Wider cavities were found to be capable of transferring more liquid from the gravure cells. This is because they facilitated contact line slipping down the wall of the cell before the contact line eventually pinned at the bottom of the cell. The effect of inertia and contact angle on the amount of liquid transfer from trapezoidal cavity and flat plates was investigated by Dodds et al. [12]. Variation of contact angle and inertia were found to affect the formation of a satellite drop in the presence of cavity or a flat plate. As discussed later in this paper, the breakup of the liquid bridge is accelerated in the presence of the cavity due to higher curvature; hence the symmetry of breakup is very much dependent on the contact angles and inertia. A number of other studies have also investigated the role of cell geometry and wettability on printing and coating. Huang et al. [15] studied the amount of liquid transferred from a trapezoidal cavity onto a moving plate by varying the contact angles, initial distance and cavity shape. They used a freely moving contact line and demonstrated that the evolution of the contact line in the cell is strongly dependent on the contact angle between the liquid and the cell. Kang et al. [16] studied the effect of contact angle on the liquid transfer between two separating flat plates. They found that by increasing the contact angle of the bottom and reducing the contact angle on the top plate more liquid can be transferred onto the moving substrate.

Inks, color pigments and other organic compounds commonly exhibit a non-Newtonian flow behavior due to the presence of colloidal particles and high molecular weight additives. Even though non-Newtonian fluids should respond differently in the complex flows involved in gravure printing, very little is known about how the rheology of the printing liquid affects pickout as all the studies available are all numerical simulations [10,13,15]. Huang et al. [15] showed that a shear thinning accelerates the breakup of the liquid bridge and concluded that viscoelasticity in inks plays a major role in the transfer process. Ahn et al. [10] modeled an elastic fluid using an Oldroyd-B model at a Weissenberg number of  $Wi = \lambda\dot{\gamma} = 0.15$ . The Weissenberg number is defined as the product of the fluid relaxation time,  $\lambda$ , and shear rate imposed,  $\dot{\gamma}$ . They computed velocity and pressure fields inside the gravure cell, even at low Weissenberg numbers the effect of elasticity was found to be significant as the pressure gradient between the gravure roll and the web increases by a factor of three. This work highlights the need to consider elasticity in the ink transfer from gravure cells. Ghadiri et al. [13] used a Carreau model to represent a non-Newtonian ink and optimized the geometry of the gravure cell. The optimized geometry yielded over 90% liquid removal from the cavity. Side wall angle and the depth of the cavity was found to play a crucial role in the emptying process, a shallow cavity removed more fluid, whereas the side wall angle showed symmetry about 45° [10].

Due to the primarily extensional nature of these flows, the elongational rheology of the coating fluids is a critical parameter to be

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