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## A simple model for viscoplastic thin film formation for coating flows



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#### ABSTRACT

A simple model of viscoplastic thin film formation flow, which is designed to describe a prototype down-stream slot-coating problem, is proposed. Initially, a coating material, typically a dense suspension, is completely yielded due to an imposed shear stress caused by the stationary coating die lip and the moving substrate. After the die, the material experiences a sudden absence of shear stress at the gas/liquid interface on the top, while the bottom maintains a still shear-dominant flow. The asymptotic analysis of this film formation flow reveals that the zeroth order terms alone cannot properly explain stress evolution, hence predicting the shape and location of the yield plane inside this flow. When higher-order terms are included, an augmented pressure jump across the interface is observed in the normal stress balance due to yield stress. The results are compared with solutions of two-dimensional mass and momentum balance equations with the same constitutive equation using the finite volume method. Implications for desirable operating conditions for slot coating are discussed.

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#### 1. Introduction

Continuous liquid coating is used to produce various films or sheet-like products, such as adhesive tapes, optical films, display panels, etc. It is also a strong candidate for nanoparticle assembly film production [1,2]. Slot coating is a popular high-precision coating method, because the film thickness is directly controlled by the flow rate and substrate speed rather than the properties of the coating liquid. In slot coating, which is classified as a pre-metered method, a liquid pumped through a feed slot forms a bridge surrounded by gas/liquid interfaces, a moving substrate and a slot die lip; this bridge is called the coating bead. When forces inside the bead, such as viscous drag, surface tension and pressure, are balanced, the coating flow is stable and produces a thin liquid layer with a high degree of uniformity.

When the force balance is disrupted, the flow becomes unstable and leads to coating defects. The upstream meniscus location is one of the important indicators for defects, which can be determined by the force balance. When it reaches one of the upstream die lip corners, the flow become unstable and fail to control the thickness due to the loss of the coating liquid through the upstream meniscus (weeping), or may create dry lanes in the film due to an air layer poking through the meniscus (bead breakup). When the film thickness is fixed, i.e., given flow rate and substrate

speed, the meniscus location is controlled by the pressure gradient inside the coating bead, which can be adjusted by applying a vacuum near the meniscus.

Many studies on slot coating have focused on the desirable range of vacuum pressure at a given film thickness, usually called the coating window. After the pioneering works by Ruschak [3] and Higgins and Scriven [4], a simplified lubrication-type flow model considering viscous drag, pressure and capillary effect was developed to construct such windows by predicting the pressure profile inside the coating bead, and hence the upstream meniscus location. This model is typically referred to as a visco-capillary model and can provide a quick but reasonably accurate coating window prediction in many industrially relevant flow conditions. However, most models are designed for Newtonian coating liquids, and there are only few examples for non-Newtonian liquids, e.g., power-law fluids [5].

Many high-performance multi-functional films for electronics and optical devices require nano- or micro-sized particles inside a coating liquid. Furthermore, environmental regulations and high energy costs associated with a solvent removal process, or drying step, recommend the use of less solvent, which eventually leads to a highly-loaded coating liquid or dense suspension. Consequently, the liquid shows complex non-Newtonian behavior including viscoplasticity, i.e., exhibiting little or no deformation up to a certain level of stress, called the yield stress, and then flowing above this stress [6]. This yield stress is due to the micro structure in the suspension [7]. Although the existence of the true yield stress is uncertain [8,9], yield stress is considered in many engineering flow

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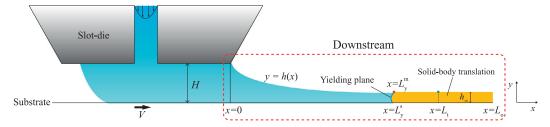


Fig. 1. A schematic of the slot coating flow. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article).

systems, especially when the viscosity of a given material is greater than a value measured for an engineering purpose under proper time and length scales [10].

After the influential works by Hohenemser and Prager [11] and Oldroyd [12], various viscoplastic flows, such as flows inside a narrow slowly varying channel [13] or along an inclined plane [14,15], have been systematically analyzed. However, a lubrication analysis of such fluid flows, even in the simplest Bingham fluid, may lead to an inconsistent or paradoxical situation due to the presence of a plug flow region, where the plug speed varies slowly in the principal flow direction. This problem was first analyzed in depth by Lipscomb and Denn [16], who called this region a 'pseudo plug'. The region cannot be captured by the leading order term in an asymptotic analysis expansion, and is weakly yielded with higher order-terms [14]. Therefore, it is typical to modify viscosity in a lubrication-type analysis to overcome such difficulties.

Another type of difficulty appears when the stress-deformation behavior constitutive relation is not modified. In numerical computations of the conservation equation with an unmodified constitutive equation, yield planes in a flow field must be tracked [17], because only yielded parts can be treated as a viscous liquid. Therefore, it is common to use an approximated viscosity or a viscosity regularized model in many practical situations [18]. In numerical computations, smoothly regularized viscosity models [17,19] are widely used; however, in a thin-film or lubrication model, it is acceptable to use a simple biviscous model [16,20].

In this study, a simple viscoplastic thin film formation flow model of the downstream section of slot coating flow is proposed and analyzed. The simple model is designed to predict the downstream meniscus shape and the pressure profile inside a biviscous Bingham fluid flow. The validity and limitations of such a simple model are examined in a full-scale two-dimensional (2D) model based on finite volume method using OpenFOAM [21]. Using the simple model, the effects of the yield stress on yield plane, meniscus shape, and pressure field will be examined in detail.

#### 2. A simple viscoplastic model of film formation flow

#### 2.1. Description of the flow system

Fig. 1 shows a schematic diagram of the two-dimensional film formation flow in a slot coating bead. The length of the downstream die lip is 1 mm, and the gap height, the distance between the die lip and the moving substrate, is  $100\,\mu\text{m}$ . The flow domain of interest is depicted inside the red dotted box in Fig. 1. This downstream coating flow under the downstream meniscus spanned from the die lip corner (x=0) to the outflow boundary  $(x=L_0)$ , where the meniscus is essentially flat. This area is called the film formation region.

The coating materials, which contain a high solid content, additives and solvents, pass through a region called the coating gap, which is surrounded by stationary die lips and the moving substrate. The solution is subjected to strong shear stress inside this narrow coating gap when the substrate speed is large enough, and this stress causes the solution to yield. Once yielded, the materials

behave like a liquid and flow through the gap. However, the magnitude of stress rapidly decreases when the materials pass through the downstream corner of the die lip and form the downstream meniscus. Eventually, the stress decreases to a value below the yield stress, which causes the material to behave like a solid and become a part of the thin-layer film region. Note that in either viscoplasite or viscous materials, the motion in this thin-film region is a solid-body translation. Due to the lack of relative motion, the shape of the meniscus in this region is completely flat, and the beginning of this region can be determined by the point of tangency, where the slope of the meniscus becomes exactly parallel to the moving substrate. For viscoplastic materials, this tangency point on the meniscus also coincides with the yield point on the meniscus, i.e.,  $L_v^m = L_t$ , where  $L_v^m$  and  $L_t$  are the location of the yield point and the point of tangency, respectively. A similar film formation flow is observed in the blade coating of viscoplastic materials, which was computationally analyzed by Loest et al. [23].

The boundary that demarcates a transition from liquid-like to solid-like behavior is called the yield plane. The shape and location of the plane depends on the state of stress inside the film formation region. As previously discussed, the stress under the die lip mainly consists of shear stress. However, because of the comparably low viscosity of gas, the shear stress vanishes on the downstream meniscus. There is an enormous transition in the state of stress in this region.

The stress evolution behaviors are different near the meniscus and near the moving substrate. Near the meniscus, normal stress is a dominant component. Because the fluid is stationary near the die lip due to the no-slip condition and its velocity reaching that of the moving substrate after the point of tangency, it is accelerated along the meniscus after exiting the die lip. The normal stress is large at the lip exit, but rapidly decays along the meniscus. Therefore, the location of the yield plan on the meniscus strongly depends on the change of normal stress.

Near the moving substrate, shear stress is the dominant component. The momentum transferred from the substrate moves faster than the coating materials, and this momentum flux is the shear stress. Unlike the flow under the die lip, where the no-slip condition dictates the materials to stop, the film formation flow does not have any mechanism to slow the fluid except for the action of viscosity. The velocity of the materials approaches that of the moving substrate, similar to a Sakiadis-type boundary layer [24]; the difference between fluid velocities across the flow direction decreases in the downstream direction, and hence the shear stress decreases.

Therefore the shape and location of the yield plane are dictated by the normal stress near the meniscus and the shear stress near the substrate. Quantitative analysis of this stress and the shape of the yield plane near the meniscus and the substrate will be discussed later.

#### 2.2. Bingham viscoplastic fluid with biviscous approximation

The simplest constitutive equation for the description of a yielding material is that of a *Bingham material*. Following Oldroyds

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