# **Confinement effects in dip coating**

## **Onyu Kim** and Jaewook Nam\*

Department of Chemical Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Korea

> Presented at the 18<sup>th</sup> International Coating Science and Technology Symposium September 18-21, 2016 Omni William Penn Hotel, Pittsburg, PA, USA

ISCST shall not be responsible for statements or opinions contained in papers or printed in its publications.

## 1. Introduction

Perhaps the simplest way to produce a thin film is to deposit a layer of liquid onto a plate via withdrawal from a liquid pool. The physics of process is known as the drag-out problem or the Landau-Levich-Derjaguin (LLD) problem, after pioneering studies by Landau & Levich (1942) and Derjaguin (1943). The obtained prototypical dip coating flow is popular because of its simplicity, not only in laboratory experiments, but also in industry. Dip coating and related methods are sufficiently flexible to produce films on various geometries and can be applied to fluids with various properties.

Most previous studies on this topic have assumed that the pool width l is infinitely large. For this "infinite" dip coating flow, most of the pool remains static, except in the small region near the plate, where the flow can be recognized inside the extremely curved meniscus viewed in the far field (Krechetnikov & Homsy 2006).

In this idealized setup, Landau & Levich (1942) have derived the equation without gravitational drainage for slow substrate withdrawal:

$$h_{\rm w} = 0.9457 \, l_{\rm c} \, C a^{2/3},\tag{1.1}$$

where  $h_w$  is the film thickness,  $Ca = \mu u_s / \sigma$  is the capillary number, with  $u_s$  being the plate speed,  $\mu$  being the fluid viscosity, and  $\sigma$  being the surface tension, and  $l_c = (\sigma / \rho g)^{1/2}$  is the capillary length.

Despite the considerable literature on "infinite" dip coating flows, the results of those studies cannot be directly applied to many practical situations in which the pool is confined, such as coating on fiber (Quéré 1999) or laboratory experiments (Brinker et al. 1992). The infinite pool assumption is only valid in the absence of a stationary wall influencing the film thickness  $h_w$  (i.e.  $h_w \ll l$ ). Otherwise, the pool is confined. A confined pool can be achieved either via a small pool width l due to a small container or through large  $h_w$  due to a fast moving plate on which a thick film is deposited. Note that an unconfined pool can become confined as *Ca* increases with increased film thickness. Under these conditions, l becomes the proper characteristic length.



Figure 1. Schematic of confined dip coating system for (a) meniscus-controlled regime and (b) channelcontrolled regime. For the flow in the meniscus-controlled regime, i.e. the region excluding the regions close to the moving plate (I and II), the liquid is virtually stationary (III and IV). For the channel-controlled regime, the static pool disappears, the dynamic region (II) fills the channel, and the viscous stress transferred from the plate at high speed dominates the region inside the channel (IV). The dotted line in (b) represents the meniscus shape at the high-*Ca* limit.

In this study, we investigate two clearly distinct flow regimes for confined dip coating:

(1) the meniscus-controlled regime for low-Ca flow;

(2) the channel-controlled regime for high-Ca flow.

These regimes are shown schematically in Figure 1. Both regimes exhibit distinct scaling behaviors depending on the capillary number *Ca*. These behaviors can be predicted via a theoretical analysis and FE computations of the two-dimensional Navier-Stokes system of the confined dip coating setup. The results can be used to predict the film thickness  $h_w$  entrained from a confined pool, which can be encountered in thin-film production in industry or in laboratory experiments.

#### 2. Dimensionless numbers

The fundamental problem with regard to confined dip coating is determining the dependence of  $h_w$  on the system parameters  $(l, u_s, and g)$ , and the physical properties of the fluid  $(\rho, \mu, and \sigma)$ . According to the Buckingham  $\pi$  theorem, four dimensionless numbers are required in order to describe the flow system uniquely: the dimensionless thickness *T*, the dimensionless pool width *L*, *Ca*, and the material number *m*:

$$T \equiv \frac{h_{\rm w}}{l}, \quad L \equiv \frac{l}{\sqrt{\sigma/\rho g}}, \quad Ca \equiv \frac{\mu u_{\rm s}}{\sigma}, \quad m \equiv \sqrt{\frac{\rho \sigma^3}{g\mu^4}}.$$
 Bookmark  
defined.(2.1)

The Reynolds number *Re* can be deduced via linear combination of the above properties, ( $Re \equiv \rho u_s l/\mu = mLCa$ ). Note that three dimensionless numbers are required in order to define an "infinite" dip coating flow uniquely (Tallmadge & Soroka 1969), and the newly introduced parameter *l* 

generates an additional dimensionless number *L*. In this study, we focus on an inertialess confined dip coating flow ( $m \approx 0$ ), where *T* is solely determined by *Ca* and *L*.



Figure 2. Dimensionless film thickness *T* versus capillary number *Ca* for  $\theta_c = 30^\circ$  and various *L*. The curves of *T* overlap at  $L \leq 0.5$ .

## 3. Meniscus-controlled regime

In the low-*Ca* limit, the entrained film is extremely thin, and  $l_d \sim (h_w l)^{1/2}$  is small. Therefore, the meniscus is predominantly covered by a static pool, which can be described by the Young-Laplace equation, and only a small part of the meniscus, close to the moving plate, deviates from the static meniscus. Unlike an "infinite" pool, the curvature of a static meniscus at the center point cannot vanish. Instead, the curvature has a finite value because of the confined geometry.

Following Derjaguin (1943), we use the geometrical matching condition. For low Ca, the matching condition yields

$$T = \frac{(0.9457/\sqrt{2}\cos\theta_{\rm c}) \ Ca^{2/3}}{1 + (0.9457/\sqrt{2}\cos\theta_{\rm c}) \ Ca^{2/3}}.$$
Bookmark  
not  
defined.(3.1)

Frent

where  $\theta_c$  is the contact angle. The predictions from the FE computations support the above equations, as shown in Figure 2.

## 4. Channel-controlled regime

When  $u_s$  increases, the viscous force from the moving plate surpasses the capillary force in this high-*Ca* regime. In addition, the pressure force becomes minuscule compared to the viscous force without an external source, such as a pump. The momentum is transferred from the plate to the stationary container wall through the liquid inside the channel, and is completely absorbed.

The flow inside the channel at high Ca resembles the coating flow under a knife or blade. The well-developed theory for the knife coating case shows that the film thickness is half the coating gap for a blade parallel to a moving plate under negligible surface tension (Ruschak 1985; Coyle 1997). Here, the gap is the distance between the moving plate and the blade face, which is essentially identical to l in the confined dip coating case. The knife coating theory predicts that the pressure gradient vanishes, which yields  $T \rightarrow 1/2$  in the high-Ca limit. FE computations support this prediction. T asymptotically approaches 1/2 as shown in Figure 2.

## 5. Final remarks

In this study, we analyzed the behavior of confined dip coating flows. Unlike the "infinite" dip coating case, the role of gravity in the force balance diminishes as the container wall is approached by a moving plate. Consequently, the characteristic length is no longer determined by the material properties, but by the pool width l. The confinement effects for a dip coating flow are strongly associated with the increasing importance of the newly-introduced characteristic length l in relation to the force balance.

It is difficult to determine the Ca criteria for both regimes, because they depend on the dimensionless pool width *L*, as shown in Figure 2. However, when the gravitational effect vanishes ( $L \leq 0.5$ ), ranges of  $Ca \leq 0.1$  and  $Ca \geq 10$  can be established for the meniscus- and channel-controlled regimes, respectively.

## References

Brinker, C. J., Hurd, A. J., Schunk, P. R., Frye, G. C. & Ashley, C. S. 1992 Review of sol-gel thin film formation. *J. Non-Cryst. Solids* **147-148**, 424–436.

Coyle, D. J. 1997 Knife and Roll Coating. In *Liquid Film Coating*, pp. 539–571. Springer Netherlands.

Derjaguin, B. V. 1943 Thickness of liquid layer adhering to walls of vessels on their emptying and the theory of photo- and motion-picture film coating. *C. R. Acad. Sci. URSS* **39**, 13–16.

Krechetnikov, R. & Homsy, G. M. 2006 Surfactant effects in the Landau–Levich problem. *J. Fluid Mech.* **559**, 429–450.

Landau, L. & Levich, V. G. 1942 Dragging of a liquid by a moving plate. *Acta Physiochim. USSR* **17**, 42–54.

Quéré, D. 1999 Fluid coating on a fiber. Annu. Rev. Fluid Mech. 31, 347-384.

Ruschak, K. J. 1985 Coating Flows. Annu. Rev. Fluid Mech. 17, 65–89.

Tallmadge, J. A. & Soroka, A. J. 1969 The additional parameter in withdrawal. *Chem. Eng. Sci.* 24, 377–383.