Optimal Control of Spin Coating on a Spherical Substrate

Ross G. Shepherd^{*}, Edouard Boujo[†] and Mathieu Sellier^{*} *Department of Mechanical Engineering, University of Canterbury, Christchurch 8140, New Zealand [†]Laboratory of Fluid Mechanics and Instabilities, École Polytechnique Fédérale de Lausanne, CH1015 Lausanne, Switzerland

<u>Summary</u>. We consider the optimal control of spin coating on a convex spherical substrate. We present a lubrication model for the flow a thin fluid film on the surface of a rotating sphere, and derive a corresponding adjoint problem to calculate the effects of changes in spin speed on the thickness profile of films produced by spin coating. This was used to determine an optimal time-varying angular velocity throughout the spin coating process in order to produce a uniform coating over section of the substrate. With this we demonstrate that there are circumstances in which spinning can allow for greater control of coating thickness than gravitational draining, but even the best performance achieved by optimal spinning showed up to a 10% deviation from the desired coating thickness.

Introduction

Spin coating has been used in a wide range of industrial applications since the early 20th century as a technique to deposit thin liquid films onto flat substrates [1]. Today, it is used to apply functional and protective coatings in the manufacturing of printed circuit boards, solar panels, light-emitting diode displays, chemical sensors, and optical components [2]. Current spin coating methods are limited, however, to flat substrate geometries and cannot reliably produce uniform thin films over curved substrates. A novel spin coating process for curved substrates could enable the development of new technologies in consumer electronics, medicine, and optics, among many other fields. Building on the seminal work of Emslie et al. [3], spin coating on flat substrates has been extensively studied, however the problems associated with coating curved surfaces have received relatively little attention despite being identified as early as this original paper. Feng and Sun [4], Chen et al. [5], and Liu et al. [6] have all developed models for spin coating on spherical substrates and validated these against experimental measurements, but the question of how to improve coating performance on curved substrates has yet to be addressed. Here, we investigate whether the spin speed used throughout the process can be manipulated to improve coating performance on a convex spherical substrate.

Model Development

We consider the flow of an axisymmetric thin liquid film on the surface of a rotating spherical substrate, parameterised by the zenith angle ϕ from the top of the sphere. Let *h* be the film thickness measured normal to the substrate, let *R* be the substrate radius, and let Ω be the angular velocity of the substrate around the vertical axis. Choosing the average film thickness \bar{h} and drainage time $t_d = \mu_0 \bar{h}/\varepsilon \gamma$ as characteristic length and time scales (where $\varepsilon = \bar{h}/R$ and μ_0 , γ are the initial viscosity and surface tension of the fluid), we derive a dimensionless 4th-order partial differential equation (PDE) describing the evolution of the film, with hats denoting rescaled dimensionless variables and subscripts denoting partial derivatives:

$$0 = \frac{\partial \hat{h}}{\partial \hat{t}} + \frac{1}{\sin\phi} \left(\frac{\hat{h}^3}{3\hat{\mu}} \sin\phi \left[\varepsilon^2 \hat{\kappa}_{\phi} + Bo \left(\sin\phi - \varepsilon \hat{h}_{\phi} \cos\phi + \varepsilon Ga \hat{\Omega}^2 \left(\sin\phi \cos\phi + \varepsilon \hat{h}_{\phi} \sin^2\phi \right) \right) \right] \right)_{\phi}.$$
 (1)

Here, $Bo = \rho g \bar{h}^2 / \gamma$ is the Bond number, $Ga = \gamma^2 / \mu_0^2 \bar{h}g$ is the Galileo number characterising the relative strength of gravity and centrifugal force, and $\hat{\kappa}_{\phi} = \hat{h}_{\phi\phi\phi} + \hat{h}_{\phi\phi} \cot \phi + \hat{h}_{\phi}(2 - \csc^2 \phi)$ is the derivative of the dimensionless curvature of the free surface. We incorporate the effects of increases in viscosity due to curing or evaporation following the method of Lee et al. [7] by introducing the coefficient:

$$\hat{\mu}(t) = \begin{cases} \exp(\beta t), & t \le t_{\rm c} \\ \hat{\mu}_1 t^{\alpha}, & t > t_{\rm c} \end{cases}$$

$$\tag{2}$$

so that the time-varying viscosity may be written as $\mu = \mu_0 \hat{\mu}(t)$, with a characteristic curing time t_c after which the film becomes essentially solid. Here, $\hat{\mu}_1 = \exp(\beta t_c)t_c^{-\alpha}$ is chosen to ensure continuity. An example film evolution under the effects of gravity alone is shown in figure 1 (left panel) using the same fluid properties and substrate dimensions as [7]. We see that beginning from a non-uniform initial condition concentrated around the top of the sphere, the fluid drains over the entire upper half before hardening, leaving a non-uniform coating with thickness of approximately $\hat{h} \approx 2.5$.

Optimal Control Methodology

We now wish to determine how the angular velocity $\hat{\Omega}$ can be changed as a function of time throughout the spin-coating process in order to achieve a more even coating. We begin by choosing (as a proof of concept) the region of the substrate surface $\phi \in D = [0, \pi/4]$ over which we want a produce a uniform coating. We also choose $\hat{h}_{opt} = 2$ as a desired coating



Figure 1: (Left panel) Example evolution of a film under gravitational draining alone with $\hat{\Omega} = 0$, $Bo = 5.7 \times 10^{-3}$, $Ga = 6.9 \times 10^{-10}$, $\varepsilon = 2.6 \times 10^{-3}$, and $\hat{t}_c = 0.2421$ in order to match [7]. (Right panel) The optimal angular velocity distribution over time by the adjoint method with the same parameters, compared with an optimal constant angular velocity with comparable performance.

thickness, corresponding to the thickness of a uniform coating over the entire upper-half sphere. The uniformity of the final film $\hat{h}_{\rm f} = \hat{h}(\phi, \hat{t}_{\rm f})$ at the time $\hat{t}_{\rm f} = 2\hat{t}_{\rm c}$ can then be characterised by:

$$\mathcal{J}(\hat{h}, \hat{\Omega}^2) = \int_D (\hat{h}_{\rm f} - \hat{h}_{\rm opt})^2 \sin \phi \, \mathrm{d}\phi, \tag{3}$$

where small values of \mathcal{J} correspond to good coating performance. We determine the optimal $\hat{\Omega}(\hat{t})$ using the same method as Boujo and Sellier [8] by considering the constrained minimisation of (3) subject to (1). We then introduce an adjoint variable λ as a Lagrange multiplier for \hat{h} and derive a terminal value PDE which can be solved for λ . The adjoint variable then allows for the calculation of the gradient $d\mathcal{J}/d\hat{\Omega}$ and the use of a simple gradient-descent optimisation algorithm.

Results and Conclusions

The optimal control methodology described above was implemented using COMSOL Multiphysics and MATLAB for two cases: (i) with no restrictions on the angular velocity distribution over time $\hat{\Omega}(\hat{t})$, and (ii) with the additional constraint that $\hat{\Omega}$ is constant over time, similar a typical speed profile currently used in spin coating. The angular velocity profiles resulting from the optimisation are shown in figure 1 (right panel). We see that the optimal angular velocity is greatest early in the coating process, then decreases as the viscosity increases and the film hardens. The final film uniformities produced using adjoint optimised and constant angular velocities are $\mathcal{J}_{adjoint} = 7.55 \times 10^{-4}$ and $\mathcal{J}_{constant} = 8.17 \times 10^{-4}$, respectively, compared to $\mathcal{J}_0 = 0.0159$ if the film drains under gravity alone ($\hat{\Omega} = 0$). However, following optimal spinning the final film thickness still varied by up to 10% from \hat{h}_{opt} over the domain $D = [0, \pi/4]$. This shows that spin coating offers significantly improved control of film thickness over simple gravitational draining, but a constant angular velocity is able to achieve almost the same coating performance as the optimal time-varying velocity profile. We can conclude from this that spin coating cannot, in general, be used to uniformly coat a curved substrate even with optimised angular velocity. We also see that the use of an adjoint-optimised time-varying angular velocity cannot produce a significantly more uniform coating than the constant speeds typically used in spin coating. This suggests that new coating processes designed specifically for curved surfaces will be required if we wish to produce uniform spin-coated films on non-planar substrates.

References

- [1] Norrman, K., Ghanbari-Siahkali, A., and Larsen, N. B. (2005) Studies of spin-coated polymer films. *Annual Reports on the Progress of Chemistry Section C* 101:174–201.
- [2] Cohen, E. and Lightfoot, E. J. (2011) "Coating Processes" in Kirk–Othmer Encyclopedia of Chemical Technology. Wiley, NJ.
- [3] Emslie, A. G., Bonner, F. T., and Peck, L. G. (1958) Flow of a Viscous Liquid on a Rotating Disk. J. App. Phys. 29(5):858–862.
- [4] Feng, X.-g. and Sun, L.-c. (2005) Mathematical model of spin-coated photoresist on a spherical substrate. Optics Express 13(18):7070–7075.
- [5] Chen, L. J., Liang, Y. Y., Luo, J. B., Zhang, C. H., and Yang, G. G. (2009) Mathematical modeling and experimental study on photoresist whirlcoating in convex-surface laser lithography. *Journal of Optics A: Pure and Applied Optics* 11(10).
- [6] Liu, H., Fang, X., Meng, L., and Wang, S. (2017) Spin Coating on Spherical Substrate with Large Central Angles. Coatings 7(8).
- [7] Lee, A., Brun, P. T., Marthelot, J., Balestra, G., Gallaire, F., and Reis, P. M. (2016) Fabrication of slender elastic shells by the coating of curved surfaces. *Nat. Comm.* 7.
- [8] Boujo, E. and Sellier, M. (2019) Pancake making and surface coating: Optimal control of a gravity-driven liquid film. Phys. Rev. Fluids 4(6).