Modelling the Flow of a Thin Liquid Film on a Sphere Rotating around a Varying Axis

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1. Abstract

The versatility of spin coating has made it the preferred method for producing the thin film layers used in the manufacture of common devices, however, one key limitation of the process is the requirement for a flat, rigid substrate. This research outlines the development of a novel fluid mechanics model, linking the flow of a liquid film over a spherical surface to forces induced through rotational motion of the substrate. A 3D Navier-Stokes model was validated using the lubrication approximation where it is observed that the flow is dominated by centrifugal forces, pushing the fluid away from the axis of rotation. The effect of changing the orientation of the rotation axis was then considered, investigating both static and dynamic cases. An effective gravity was determined for a fluid coating on a rotating sphere about an arbitrary axis which combined with the inertial forces producing thickness profiles axisymmetric about the rotation axis, thinning at the poles with a thicker belt close to the axis’ equator. Axis motion perturbed the axisymmetric profile, forming two opposing thick regions of fluid as it was forced away from the path of the rotation axis. Although no uniform coating was developed, the inertial forces induced through complex rotation were shown to be controllable, proposing the investigation into the applicability of spin coating on curved surfaces.

2. Introduction

Spin coating has played a critical part in the development of many common devices we use, from cell phones to solar cells, where thin film coatings are a key aspect of the manufacturing process. Spin coating involves the deposition of a liquid – often a solvent polymer resin – onto a flat substrate which is spun, dispersing the fluid through centrifugal forces. A uniform layer is formed due to a balance of viscous and inertial forces, allowing the solvent to evaporate, resulting in a micro or nanometre thin solid coating. Spin coating models first developed by Emslie et al. (1958) enabled the optimization of the process for planar substrates with various improvements being made to include additional conditions such as varied dynamics, surface tension and evaporation effects. The impact of substrate curvature on coating flows was investigated (Roy & Schwartz, 1997; Schwartz & Weidner, 1995) where it was found that substrate curvature caused surface coating flows to evolve into non-uniform profiles due to surface tension. Studies of thin film coating flows over spherical substrates conducted by Lee et al. (2016) found that drainage effects from gravity also caused the film profiles to be non-uniform, even on surfaces of constant curvature.

Researchers Chen et al. (2009) and Liu et al. (2017) provided the basis for modelling the thin film flow over a rotating convex, axisymmetric substrate through the application of the lubrication approximation. A similar method was also utilized by Kang et al. (2016) to investigate the evolution of a surface film’s thickness profile on the exterior of a sphere rotating about its vertical axis. The effects of surface tension along with gravitational and centrifugal forces were considered, where it was observed that the eventual film profile would exhibit a drop like collection point with no rotation.
but tend towards an equatorial belt at high rotational velocities. Similar findings were observed by Shepherd et al. (2020) where the lubrication model was used to determine the sensitivities of various process parameters, aiming to optimize coating performance. Recently, models using the lubrication approximation have been used to investigate the spin coating of other curved geometries like concave spheres (Feng & Sun, 2005), spheroids (Duruk et al., 2021) and other arbitrary curved surfaces (Weidner, 2018, 2022).

3. Methodology

Rotation of a rigid body causes inertial forces to be experienced by any mass connected to the body, such as a fluid layer on a rotating substrate. These inertial forces: called centrifugal, Coriolis and Euler forces, shown in equations (1-3), are products of the angular velocity $\Omega$, position $R$, and relative velocity $U$, which are vectors defined in a rotating, body fixed reference frame. Figure 1 shows the body forces acting on the system rotating about the centre of the sphere with an arbitrary angular velocity in 3D, described in spherical or Cartesian co-ordinates.

\[ F_{\text{centrifugal}} = -m\Omega \times (\Omega \times R) \]  \hfill (1)
\[ F_{\text{coriolis}} = -2m(\Omega \times U) \]  \hfill (2)
\[ F_{\text{euler}} = -m \frac{d\Omega}{dt} \times R \]  \hfill (3)
\[ F_{\text{gravity}} = -mg(T_r(t) \times \hat{k}) \]  \hfill (4)

Gravity was defined in the global reference frame and therefore rotated relative to the substrate, as shown in equation 4. Where the rotational transform $T_r(t)$, denotes the time dependent rotational transformation matrix between the two reference frames.

Finite element analysis software COMSOL was used to model the flow of a liquid over the exterior of a rotating sphere in 3D, thus removing the requirement for an axisymmetric model. This enabled the consideration of the Coriolis and Euler forces, as well as more complex rotational motion. An O-H structured mesh was used to model the fluid domain in Cartesian co-ordinates as shown in figure 2, where the angular velocity is initially described in spherical co-ordinates.
The laminar flow over the spherical surface was modelled using a moving mesh to apply the Navier-Stokes equations across the deforming fluid domain, where the active body force was a combination of gravity, surface tension and the inertial forces produced through substrate rotation. The internal boundary was modelled as a wall with no slip and thus, only the relative motion of the fluid was captured where the effects of drag on at the fluid/air interface was neglected. A mesh convergence study was then conducted to ensure a sufficiently refined mesh for accurate and efficient results. Using a scaling factor to control the mesh resolution allowed for the comparison of similar solutions from 8 mesh sizes, where the system parameters outlined in table 1, (Shepherd et al., 2020) were used for simulation to compare and validate the new 3D model against published results. The model fluid simulated a curing process with a time dependent viscosity model (Lee et al., 2016) aimed to replicate the thermoset polymer VPS-32.

<table>
<thead>
<tr>
<th>Physical Parameter</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rotational velocity</td>
<td>30</td>
<td>(rad/s)</td>
</tr>
<tr>
<td>Substrate radius</td>
<td>38</td>
<td>mm</td>
</tr>
<tr>
<td>Initial film thickness</td>
<td>0.1</td>
<td>mm</td>
</tr>
<tr>
<td>Density</td>
<td>1160</td>
<td>(kg/m^3)</td>
</tr>
<tr>
<td>Surface tension</td>
<td>0.02</td>
<td>(N/m)</td>
</tr>
<tr>
<td>Initial viscosity</td>
<td>7.1</td>
<td>(Pa \cdot s)</td>
</tr>
<tr>
<td>Characteristic curing time</td>
<td>574</td>
<td>s</td>
</tr>
<tr>
<td>Viscosity curing power</td>
<td>5.3</td>
<td>-</td>
</tr>
<tr>
<td>Viscosity curing coefficient</td>
<td>2.06e-3</td>
<td>1/s</td>
</tr>
<tr>
<td>Drainage time</td>
<td>2.37e3</td>
<td>s</td>
</tr>
</tbody>
</table>

Table 1. Model parameters used for 3D numerical simulations of liquid coating flows on a rotating sphere.

This 3D model was then used to investigate the effects of varying the rotational axis orientation, analysing the impact of gravity and complex substrate kinematics. First, a series of simulations were completed with a stationary tilted rotation axis, varying the zenith angle from 0\( rad \) (parallel with gravity, Z-axis aligned) to \( \frac{\pi}{2} \)\( rad \) (perpendicular to gravity, X-axis aligned). Angular velocities ranging from 10 – 30\( rad/s \) were modelled utilizing a time-weighted average gravity force to reduce model complexity. A dynamic rotation axis was then modelled using a time dependant orientation with constant rate of change, keeping gravity aligned with the axis of rotation. Two different rotation rates were compared using an angular velocity of 30\( rad/s \) - an order of magnitude greater - which ensured that the additional rotation had negligible contribution to the inertial forces.
3. Results and Discussion

Vertical axis spinning provided an ideal test case to validate the 3D model as published analytic lubrication solutions enable a comparison of resulting film profiles. Meshes with degrees of freedom (DoF) ranging from 13,000 to 500,000 were tested in a mesh convergence study, shown in figure 3, which compared the resulting film profile after two characteristic curing cycles \((t = 1,148s)\) to the analytic lubrication profile. Using a mesh scaling factor of 5 provided a balance of model accuracy and efficiency, resulting in a mesh with 184,790 total DoF’s, where the final surface film profile is shown in figure 4 with a non-dimensional error of \(\varepsilon = 0.004\). The thickness profile showed an initially uniform 0.1\text{mm} film evolve into a band with peak thickness of 0.127\text{mm} at approximately 100° from the vertical rotation axis.

![Figure 3](image-url)  
Figure 3. Solution error for O-H structured mesh resolutions used to model spin coating on a sphere.

![Figure 4](image-url)  
Figure 4. Film thickness profile and flow velocity at \(t = 1,148s\) with uniform initial coating on a sphere rotating at \(\Omega = 30\text{rad/s}\), validated using published analytic and numeric results (Shepherd et al., 2020).

The axisymmetric thin film profile matched results in the literature, where the final shape was dependent on the substrate’s angular velocity. Inertial forces generated at low rotational speeds were insufficient and thus, gravity forced drainage flow was observed, producing a gradual increase from the minimum thickness at the top of the sphere to a drop-like bulge at the bottom. Increasing the rotational speed causes the centrifugal force to grow proportional to the angular velocity squared \((F_{cent} \propto \Omega^2)\), making it the dominant forcing mechanism. This produces film thickness profiles similar to what is shown in figure 4, forming a thick belt below the equator and thinning at the poles. The sharpness of the peak and distance from the equator were found to vary depending on both the angular velocity and fluid properties, however, it was evident that given these parameters and an
initial condition, the analytical lubrication models could accurately predict the eventual coating profile where uniform thickness is unachievable. In this case, the other inertial forces can be neglected as the low surface velocities and constant angular velocity render the Coriolis and Euler forces insignificant.

Aligning gravity with the rotation axis greatly simplifies the fluid mechanics system, however, modelling the flow numerically in 3D provided the opportunity to explore the effects of varying the orientation of the rotational axis. Due to the choice of body fixed reference frame, accurately implementing gravity becomes more complex as the observed gravitational force rotates inversely around the arbitrary axis. The path of the forcing vector forms a cone (or disk in the perpendicular case) with a central axis aligned with the tilted axis of rotation and point at the origin. This dynamic force rotates with the same angular velocity as the substrate and is therefore changing with a faster timescale than the liquid film dynamics, adding unnecessary complexity and extending simulation solve time. To overcome this, an effective gravitational force was used where the time weighted average is modelled as the component of gravity aligned with the rotation axis, due to the perpendicular component theoretically averaging to null over time. To ensure this simplification was valid, both methods were used to model rotation around a tilted axis at the lowest speed of $10 \text{rad/s}$, as this provides the ‘worst case’ where the slower rotation creates the smallest difference in dynamic timescales and increases the proportional effect of gravity. Comparing the results from the two methods after one curing cycle ($t = 574 \text{s}$) showed the static effective gravity method was found to agree within $\pm 10^{-4} \text{mm}$ of the true film thickness profile for all cases of axis tilt. Therefore, the assumption of a time weighted, effective gravity was found to be valid when considering rotation around an arbitrary axis with unit vector $\hat{n}$, as shown in equation 5.

$$F_{grav,eff} = -mg \times \cos(\beta) \times \hat{n} \quad (5)$$

Using the effective gravity force, a series of tilted axis simulations were run for one curing cycle at increasing angular velocities as shown in figure 5.

![Figure 5. Cross sectional film thickness profiles produced after $t = 574 \text{s}$ from an initially uniform coating on a sphere rotating about a tilted axis.](image-url)
These film thickness profiles reveal similar features to the vertical axis case, showing a threshold angular velocity between $10 - 20\, rad/s$ at which point centrifugal force becomes predominant over gravity. Comparing the different axis tilt angles provides insight into the system’s sensitivity to gravity as the effective gravitational force is directly linked to the rotation axis’ orientation. It is shown that gravity elongates the peaks caused by the centrifugal force which reduces the maximum and minimum thickness, softening the surface gradient. As the rotation axis was kept in the X-Z plane, this cross section best shows the impact of altering the axis orientation, where a phase shift observed in the thickness peaks is directly proportional to the axis tilt. Therefore, the rotation axis always passes through the point of minimum thickness due to both gravitational and centrifugal forces pulling the fluid away from this pole. The other inertial forces were again negligible, resulting in film profiles that were axisymmetric around the rotation axis, thus, analytic methods could be employed to model this thin film flow.

Complex dynamic rotation was then investigated where gravity was kept aligned with the dominant rotation axis, where the axis orientation varied with time. The angular velocity vector was rotated and oscillated, using two different rates of change as shown in figure 6. The same forcing mechanisms dictated the film evolution, where centrifugal force pushed fluid away from the axis of rotation, leading to the build up of fluid at the poles normal to the plane of axial motion. The two rates of change produced similar film thickness profiles, where the difference was in order of $10^{-5}\, mm$. These small differences can be attributed to the Coriolis and Euler forces, showing that for relatively small rotation rates these forces are negligible. Therefore, the final profile was determined by the path of the angular velocity vector, where magnitude controlled the surface gradient. This was supported by results produced with a non-constant and oscillating axis orientation. Small oscillations perturbed the axisymmetric profiles created from a rotation about a stationary axis, breaking the thick belt into two isolated zones at opposing poles. Whereas large oscillations of the rotation axis over $\pi\, rad$ developed similar profiles to those shown in figure 6, with the two thick zones consolidated, becoming axisymmetric around the minor rotation axis, normal to the plane of dominant axial motion.

![Figure 6. Film thickness profiles at $t = 550s$ from coating a sphere spinning with $\Omega = 30\, rad/s$ about a dynamic axis or rotation.](image)
Conclusions

A 3D Navier-Stokes model has been developed to simulate the thin film flow of a liquid over the convex surface of a rotating sphere. The effects of gravity and surface tension were considered alongside the inertial forces induced through substrate motion, particularly centrifugal force. Analytic lubrication fluid mechanics models were utilized to validate the new model using the case of vertical rotation before investigating the effects of varying the orientation of the rotation axis. It was found that when considering an arbitrary constant rotation axis, the effective gravitational force could be modelled as a time-averaged constant force proportional to the rotation axis’ zenith angle. Once above the threshold angular velocity to overcome drainage, the liquid film developed into an axisymmetric profile about the substrate’s rotational axis, thinning at the axis poles and thickening towards the equator. A dynamic axis of rotation was then analysed, showing that the centrifugal force could be manipulated by changing the orientation of the rotation axis without producing significant Coriolis or Euler forces. This enables the control of the inertial forces acting on the thin film where the orientation of the rotational axis controls the direction of force, and the angular velocity determines the magnitude. Therefore, the flow mechanisms were utilized to create previously unseen film coating profiles through the implementation of complex substrate motion.

The modelling tools described in this research provide a gateway into exploring the flow of a thin liquid film over a curved surface which is a key aspect of many relevant manufacturing processes. The ability to manipulate centrifugal force on a flat surface has made spin coating a critical technique for many of our devices, and this new model has enabled the investigation into the applicability of these coating mechanisms for curved surfaces. Future goals look to combine various motion sequences to develop a technique for optimized coating performance whilst developing a prototype rotational manipulator to validate this model through experimental trials.

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References


