

Fluid Mechanics of Multilayer Slide Coating

H. Benkreira and A. Innocente

Advanced Materials Engineering R & KT Centre
School of Engineering, University of Bradford, UK

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Introduction

Multilayer slide coating provides the opportunity to coat in one pass superimposed yet distinct films for a very wide range of applications including products such as photovoltaic cells, pharmaceutical drug release substrates, polymer electrolyte membrane fuel cells, crystal display optical films and many others. Although apparently simple, it is difficult to predict the thicknesses of the films formed on the slide. This is because the flow exiting the slots form banks of fluids which interact with the flow arriving from the slots above. An example of flow profile exiting from a slot is shown in Fig. 1 (slot opening extends from 0 to 7 mm). Clearly, in cases of flow rates $> 4.44 \text{ cm}^2 \text{ s}^{-1}$, the flow arriving from the slot above will have to flow over the 'hump'. Moreover, these banks of fluid that form are not uniform they exhibit recirculation flow regions (see Fig. 2) and these will be detrimental to the stability of the flow, i.e. the formation of ribs on the top surface and undulations at the interface boundaries of the layers. Both of these features make the fluid mechanics of multilayer slide coating exceedingly complex. In this paper, we provide new insight, following from an analysis of the experimental data to inform future directions in the analysis of this type of flows.

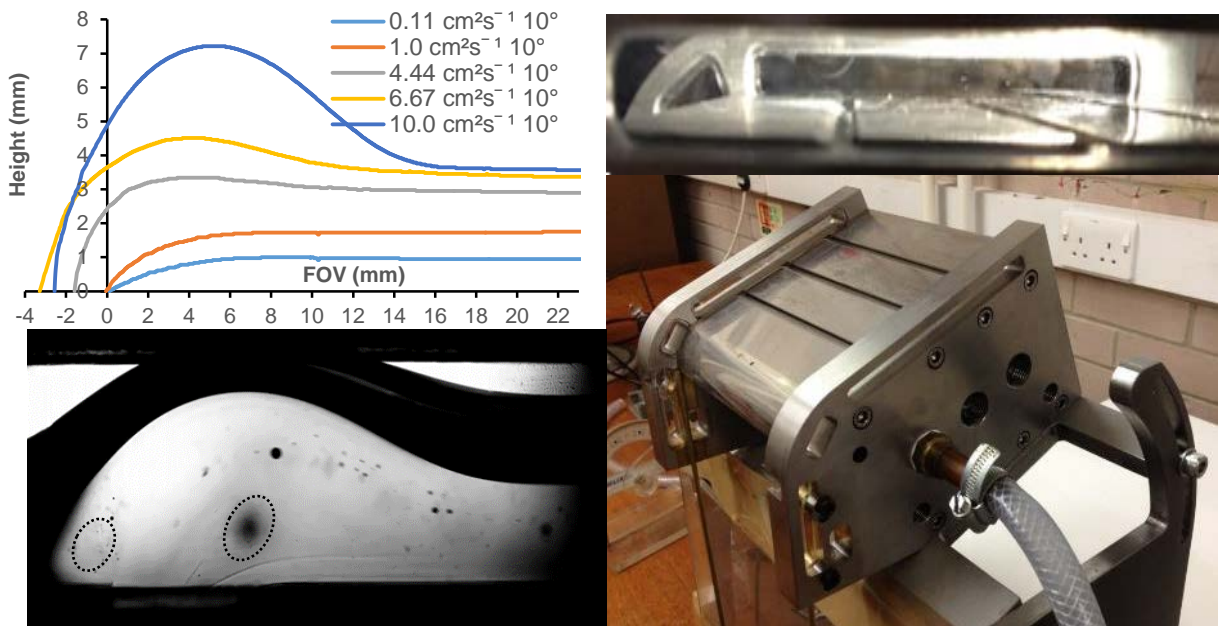


Fig. 1: Effect of 60 mPa.s glycerine solution flowrate on heel formation at slot exit. **Fig.2:** Hump with recirculating zones. **Fig.3:** Viewing window. **Fig 4:** the experimental slot die slide.

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Experimental Method

The key consideration here is measuring from visualisation of the flow on the slide both the film thicknesses of the layers formed and their ribbing intensity (amplitude and wave length of the ondulations on the top surface and at the interface between layers). To achieve this, sapphire glass windows were built on the edge of the slide (see Fig. 3 and 4) with the flow viewed using telecentric lenses (greater depth of field than conventional lenses) in conjunction with a high speed camera linked to a pc with a high performance frame grabber card and LABVIEW software to capture image sequences of 3000 images each. These were then processed using Adobe Photoshop, FIJI-ImageJ and MATLAB 7.0 to measure the height of the fluid layers at various location along the slide, the peak to peak amplitude and frequency of the instability waves forming on the free surface and on the interfaces. The approach described was used to analyse 1, 2 and 3-layers flows in a wide range of conditions (10 to 35° slide angles, 0.15 to 10L/m.min flow rates in each layer and 3 different viscosities, 10, 60 and 200 mPa.s). A glycerol-water solution was used throughout the experimental programme so the effect of changes in density and surface tension were not considered.

As for the stable coating window, prior work (Weinstein and Kurz, 1991; Martinez, 2011) showed that for a given viscosity stratification, maximum stability is achieved when the first layer < 22% of the total film thickness and the second layer >18% of the total film thickness. Satisfying this criterion is difficult unless the individual layer thicknesses can be accurately predicted. Following on these theoretical observations, the experimental programme is empirically split into four ranges (Table 1) by manipulating flow rates. Thus the assessment is based on flow rates (that obeyed these rules), which can be controlled.

Flow Stability	Layer 1	Layer 2
Stable	< 22	> 18
Unstable Lower Layer	> 22	> 18
Unstable Middle Layer	< 22	< 18
Unstable Both Layers	> 22	< 18

Table 1: Depth percentages required, in comparison to the total film thickness, for the first and second layers of 3-layer flow to obtain prescribed theoretical stability (Alpin,

Analysis

As explained in the Introduction, this is a complex flow problem, usually oversimplified by extending the one-layer solution to the multilayer case using one constant viscosity (the bottom layer viscosity) across the layer (Nusselt's solution). The full lubrication model must take into consideration the physical properties of each individual layer. Adopting such an approach, the flow in each layer is assumed to be fully developed, balancing gravity and shear stress forces down the slide inclined at an angle θ :

$$\text{x-component: } 0 = \mu_n \left[\frac{d^2 v_{x,n}}{dy^2} \right] + \rho_n g \sin \theta \quad , \quad \text{y-component: } 0 = -\frac{dp_n}{dy} - \rho_n g \cos \theta \quad (1)$$

With the no slip boundary condition on the slide wall, continuity of shear stresses across the layers boundaries and zero shear on the top layer, a solution to the problem is permissible, albeit numerical. Fig. 5a shows that, compared with the Nusselt's solution (which ignores variation in viscosity across the film), the theoretical model described by Eq. (1) is significantly better at predicting the thickness of a 3-layer stack, displaying a closer fit with the ideal line. Figs. 5b-d show some typical results for a 3-layer flow using this approach compared with measured data at various viscosity stratifications. The fit is poor, particularly for increasing viscosity stratification (10-60-200 mPa.s) where the lower bottom layer (which exits the slot close to the end of the slide) experiences less shear on the slide die surface and does not have sufficient time to reach terminal velocity. This suggests that Eq. (1) do not hold and the need to include the variation of velocity in the x-direction, i.e. modifying Eq. (1) to:

$$\text{x-component: } v_{x,n} \frac{dv_{x,n}}{dx} = \mu_n \left[\frac{d^2 v_{x,n}}{dy^2} \right] + \rho_n g \sin \theta \quad (2)$$

Solving Eq. (2) now requires further boundary conditions, i.e. *how the liquid is fed to the slide*, the key determinant not grabbed by the simple model. How to organise these additional boundary conditions is a challenge and a recommendation for future work.

Now with regard to instabilities, the experimental results show that flow stratification is also critical. For the 10-60-200 mPa.s viscosity stratification, maximum theoretical stability is achieved when $q_1 < q_2 < q_3$, which is validated by the uniform free-surface profile (Figs. 6). We can also observe from these figures that moderately stable flow is even achievable for the 60-10-200 mPa.s viscosity stratification as long as $q_1 < q_2 < q_3$. A plausible explanation is that the i^{th} layer can easily overcome the vortex at the $(i-1)^{\text{th}}$ slot exit when $q_{(i-1)} < q_i$, leading to improved stability. On the contrary, flow perturbations appear as the condition deviates from the criteria shown in Table 1. For the 10-60-200 mPa.s viscosity stratification, maximum theoretical instability is achieved when $q_1 > q_2 < q_3$, which is validated by the sinusoidal free-surface profile. The 10-60-200 mPa.s viscosity stratification is naturally the most stable so the wavelength is relatively long and the peak-to-peak amplitude is relatively small. For the 60-10-200 mPa.s viscosity stratification, maximum theoretical instability is achieved when $q_1 = q_2 = q_3$, which is validated by the free-surface profile shown in Figs. 6, displaying a relatively short wavelength and relatively large peak-to-peak amplitude. A range of inequalities, relating 3-layer flow stability with flow rate stratification, is produced for different viscosity stratifications.

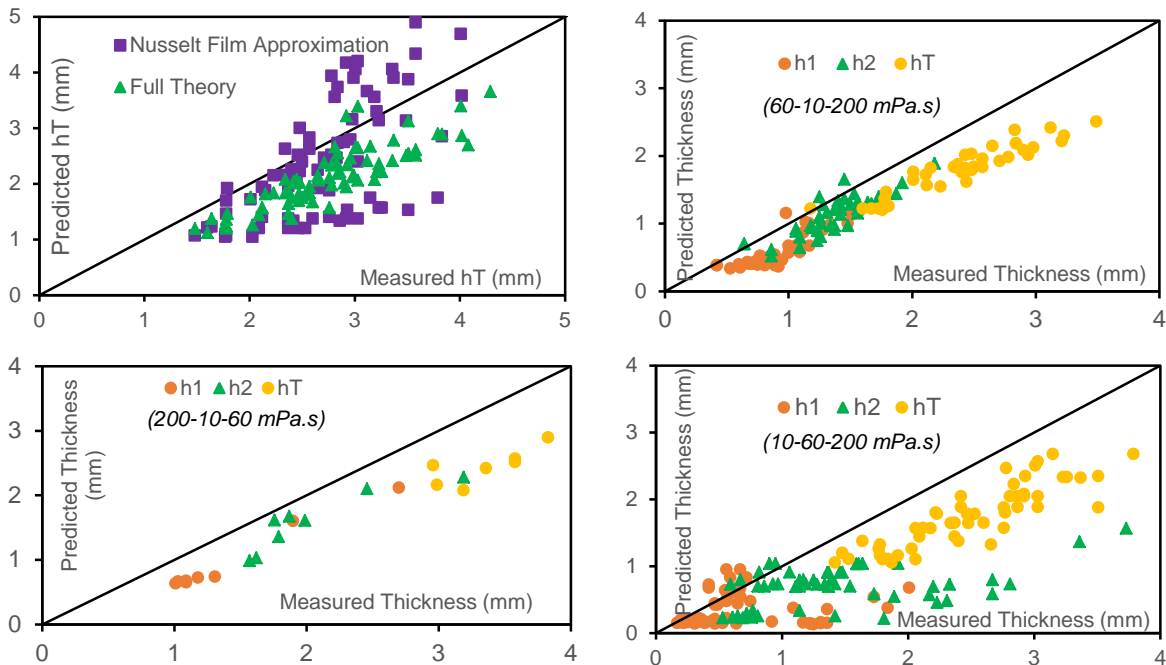


Fig. 5: Comparison of full theoretical model predictions with Nusselt film extension for 3-layer flow and comparison between model and data at various viscosity stratifications.

Conclusions

Multilayer slide coating is a high-speed continuous process with good prospects. The current challenge is being able to achieve a stable 3-layer flow. Weinstein and Kurz (1991) and Martinez (2011) related flow stability to depth ratio, which is only useful if we can accurately predict the individual layer thicknesses formed at the Film Flow Zone. A full theoretical model, which takes into consideration the die inclination angle, density, flow rate and viscosity stratifications of each individual layer, was developed. Critical analysis of the model validation has revealed that the assumption $\frac{dv_{x,n}}{dx} = 0$ is likely a major source of error, leading to the proposal of a more comprehensive model.

For a given viscosity stratification, flow rate stratification is critical in achieving the maximum potential flow stability. Until we can accurately predict the individual layer thicknesses formed at the Film Flow Zone, relating 3-layer flow stability to flow rate inequalities is much more practical and convenient for operation. The limitation associated with each set of inequalities is that they are specific to one viscosity stratification. Nevertheless, the experimental method used for this investigation is applicable to determine the optimum flow stratification for any given viscosity stratification.

References

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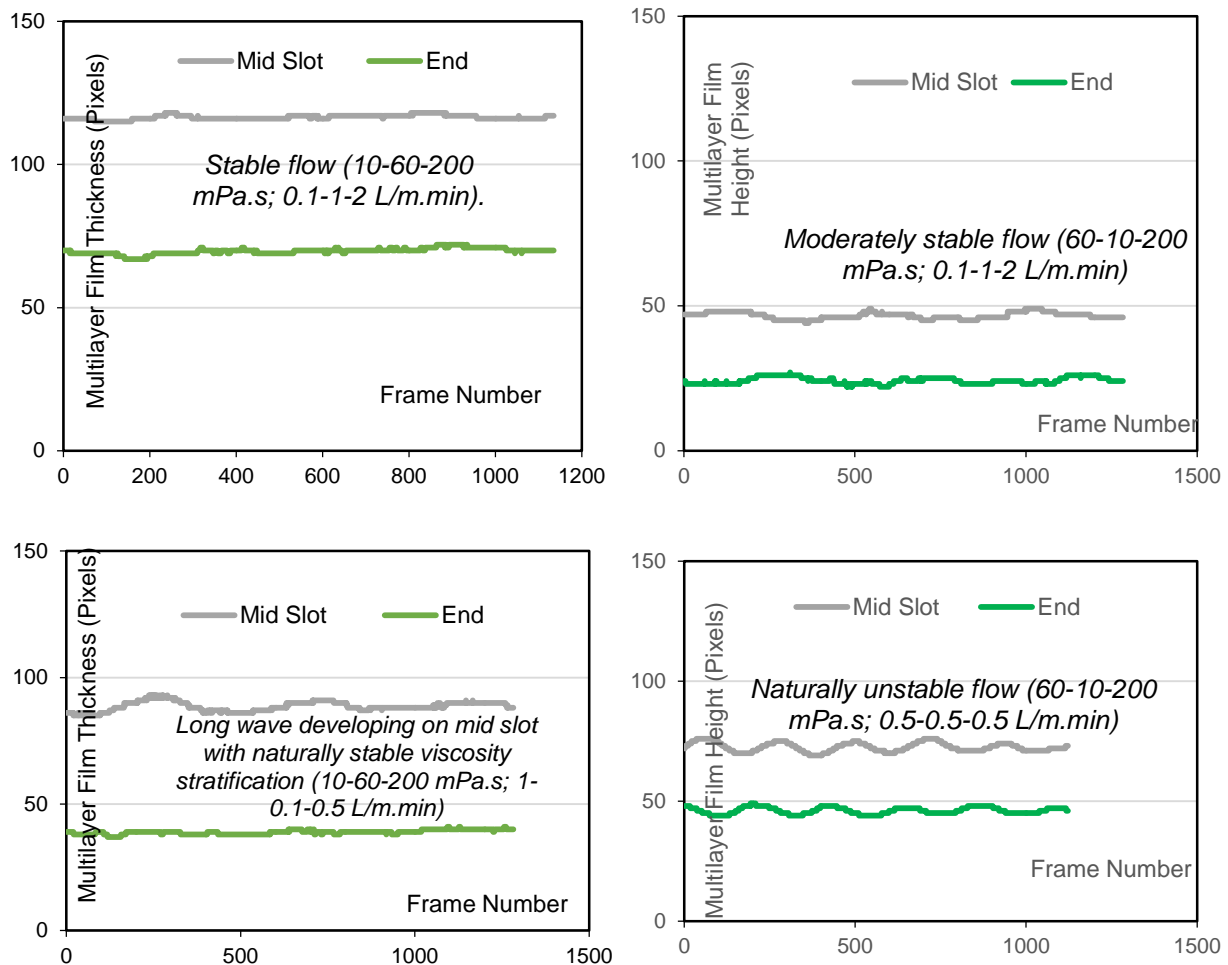


Fig. 6: Stability window for varying flow rate and viscosity stratifications. [35.962 Pixels/mm; $\theta = 23^\circ$; Mid Slot signifies halfway along slide; End signifies end of slide.]