## Hysteresis and non-uniqueness in the speed at the onset of instability in curtain coating onto a pre-wet substrate

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Experimental observations of an advancing contact line across a solid dry surface have shown that the maximum speed of stable wetting is unique for a plunging tape configuration involving a smooth dry substrate. The same is also true for coating of smooth dry surfaces where inertial effects become important for a given set of flow conditions; Blake *et al.* (1994) demonstrated this for curtain coating of both Newtonian and non-Newtonian fluids, although hysteresis, a difference between the speed at the onset and clearance of air entrainment, was observed only for non-Newtonian fluids.

Most experimental studies of dynamic wetting (Burley & Kennedy 1976; Blake & Ruschak 1979; Gutoff & Kendrick 1982; Burley & Jolly 1984; Ghannam & Esmail 1993; Blake *et al.* 1994; Cohu & Benkreira 1998; Blake *et al.* 1999; Benkreira 2002; Blake *et al.* 2002) determine the speed at the onset of instability (air entrainment) by fixing all parameters and increasing the substrate speed. This procedure not only determines this speed to be unique, but a monotonically decreasing function of the liquid viscosity.

In contrast, Clarke (2002) showed the existence of multiple stable regions for curtain coating of Newtonian liquids onto rough surfaces, whilst Blake *et al.* (2004) noted a similar effect on smooth surfaces using greatly increased curtain heights.

In this work, the maximum speed of stable coating onto a surface pre-wetted with the coating fluid was investigated experimentally by the determination of the onset of air entrainment in curtain coating of Newtonian fluids onto a scraped steel wheel over a broad range of dimensionless parameters (Reynolds number:  $0.14 < Re = \frac{\rho Q}{\mu} < 33.02$ ; Capillary number:  $0.19 < Ca = \frac{\mu U}{\sigma} < 25.07$ ). Aqueous glycerol solutions were used as the working fluid (0.0326 Pa.s  $< \mu < 0.878$  Pa.s). The thickness of the pre-wetted film was  $O(10^{-6} - 10^{-5})$ m.



Figure 1: Horizontal examination of the parameter space for a 0.462Pa.s solution. 1.77 < Ca < 12.86, 0.49 < Re < 2.03.

The usual experimental method to determine maximum speeds of coating (wetting) is to hold all experimental parameters constant and increase the substrate speed until instability occurs, usually manifested by air entrainment. By varying this procedure and by focusing on high liquid viscosities and high impingement velocities, we investigate alternative routes to determine the instability boundaries. In doing so, bifurcations in the stable coating windows are witnessed and, at certain conditions, the speed at the onset of instability is non-unique.

Figure 1 shows the coating window for a high viscosity liquid ( $\mu = 0.462$ Pa.s,  $\sigma = 0.065$ Nm<sup>-1</sup>). The figure clearly shows that the speed at the onset of air entrainment is flow-rate dependent for a pre-wetted surface.

The circle data points mark the onset of air entrainment determined by increasing substrate speed for a number of fixed flow rates. For  $Q < 5.31 \text{cm}^2 \text{s}^{-1}$ , increasing Q leads to sharp increases in the maximum substrate speed. Once this threshold flow rate is passed, there is a dramatic drop in substrate speeds (upper branch) - this is a bifurcation of stability curves. The procedure used to determine these data points is the usual experimental method and forms the *horizontal* examination of the parameter space.

Figure 2 shows data points created in the same fashion for a similarly high viscosity liquid ( $\mu = 0.552$ Pa.s). The data points marked clearance indicate the speeds at which air entrainment ceases and stable coating resumes. It can be seen that for certain intermediate



Figure 2: Horizontal and vertical examinations of the parameter space for a 0.552Pa.s solution. 1.62 < Ca < 19.16, 0.42 < Re < 1.59.

values of Q, there are significant differences between the onset and clearance. This is air entrainment hysteresis, not seen on dry substrates.

Also indicated on the figure are 5 vertical lines. These constitute *vertical* examination of the parameter space. The lines are for fixed substrate speeds and are either solid or dotted to represent either air entrainment or stable coating respectively determined by gradually increasing the flow rate. As can be seen, the stable region here (i.e. dotted sections) do not coincide with that created by the data points, that is the vertical and horizontal analyses do not produce the same coating window.

Given this observation and the enlarged hysteresis region (which may be stable or unstable depending on how it is entered) it was chosen to vary the route through the parameter space by alternatively increasing both the substrate speed and flow rate. Results of this are presented in the remaining figures.

The data points in figure 3 joined by solid lines are those produced by a horizontal analysis, whereas those joined by dotted lines are additional data points which were determined by alternatively increasing both substrate speed and flow rate. Example routes through the parameter space are shown in the figure. It is clear that the 'stable' region can be extended beyond that which is obtained by a purely horizontal examination (which is the usual experimental procedure) and that for certain flow rates, the speed at the onset of instability (air entrainment) is non-unique.



Figure 3: Path-dependent analysis of the parameter space for a 0.597 Pa.s solution. 2.39 < Ca < 21.2, 0.386 < Re < 1.73.



Figure 4: Path-dependent examination of the parameter space for a 0.493Pa.s solution.

Figure 4 shows a repeat of the new procedure applied to a 0.493Pa.s solution. In this case, the data points joined by a dotted line do not indicate observations of air entrainment, but limitations of the equipment, either substrate speed or flow rates. Hence it is entirely plausible that the path-dependent coating window could be extended far beyond that which may be found by a purely horizontal examination of the parameter space.

## **Presentation:** Oral