Wetting at High Capillary Numbers

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Extended Abstract

Introduction. The coating of liquids onto solids is an important industrial process. A prerequisite for successful coating is that the liquid dynamically wet the surface of the solid. One of the limits to high-speed coating is the onset of dynamic wetting failure, which leads to air entrainment. In simple experiments in which a tape or fiber plunges vertically into a pool of liquid, air entrainment usually occurs at capillary numbers Ca < 1. However this limit is not immutable. Indeed, the term "hydrodynamic assist" has been coined to emphasize the fact that coating flows may be manipulated to promote wetting and so postpone air entrainment [1]. Commercial curtain coating typically operates in the range 0.5 < Ca < 10. Flow visualization has shown that hydrodynamic assist leads to a reduction in the dynamic contact angle for a given wetting speed, and it is this that permits the higher coating speeds by postponing contact angles approaching 180° [2]. Methods of coating optical fibers have evolved to the point where comparatively viscous liquids can be coated successfully at very high speeds with Ca of order 1000. In a recent paper Jacqmin [3] has suggested that, in this case, the high speeds are possible because any air entrained into the coating dissolves under the high fluid pressures found in the fiber-coating die.

Here we report successful curtain coating with a viscous Newtonian liquid up to $Ca \sim 50$. With tall curtains, coating speeds up to 1000 cm s⁻¹ were achieved without air entrainment provided the wetting line was located beneath the curtain. The new data and their evolution with curtain height suggest that the high speeds are possible because of intense hydrodynamic assist. The possibility that the effect is caused by entrained air dissolving under the high pressures generated by the coating process seems less likely. The work will be presented and discussed in more detail in a forthcoming publication [4].

Experimental. The coating apparatus has been described previously [1]. The substrate was smooth ($R_z \sim 0.6 \,\mu$ m) poly(ethyleneterephthalate) (PET) tape, 175 μ m thick and 35 mm wide. The liquid was a 92% solution of glycerol in deionized water, having a viscosity of 0.32 Pa s, a surface tension γ = 65 mN m⁻¹, and a density ρ = 1238 Kg m⁻³. Maps of coating speed U (cm s⁻¹) were determined as a function of volumetric flow rate per unit coated width Q (cm²s⁻¹) at curtain heights of H = 10.2, 15.2, 17.8, 20.1 and 25.2 cm, respectively. At each flow rate, the coating speed was progressively increased until air entrain-

ment was detected; the speed was then decreased until air entrainment ceased. For the Newtonian liquid used here, the coating speeds at the onset and cessation of air-entrainment were essentially the same; there was no evidence of the hysteresis seen with shear-thinning polymer solutions. The main results are shown in Figs. 1-3. For completeness, boundaries delimiting recirculation phenomena are also included in Figs. 2 and 3

Results and Discussion. Fig. 1 shows data obtained with curtain heights of 10.2, 15.2 and 17.8 cm. At each height, air entrainment occurs to the right of the data, uniform coating to the left. The symbols mark the onset of air entrainment (AE) as the coating speed is increased. The map obtained at 10.2 cm is typical of that for a Newtonian liquid [1,5]. The curtain is unstable at low flow rates where surface-tension forces dominate inertial forces. At sufficiently high flow rates (~1 cm² s⁻¹) stable coating is possible, but the wetting line tends to be in front of the curtain (i.e. downstream) and air entrainment occurs at low coating speeds. As the flow rate is increased, the wetting line moves upstream and the air-entrainment speed increases, attaining a maximum value U_{max} at some optimum flow rate Q_{max} when the wetting line is directly beneath the curtain. At still higher flow rates, the wetting line moves behind the curtain and a "heel" of liquid develops at its base. Air entrainment now occurs at progressively lower speeds until at some point recirculating viscous eddies appear in the heel [6]. Thereafter, the wetting line is isolated from the main flow in the curtain and air-entrainment speed attains a limiting value U_l that is independent of flow rate (and curtain height) and approximates the immersion speed at which air entrainment commences in plunging tape experiments with the same materials.



With liquids of moderate viscosity (of order 0.1 Pa s), as the curtain height is increased, U_{max} typically moves to higher speeds and lower flow rates. However, with the relatively viscous liquid used in the present study, although U_{max} still increased with curtain height, Q_{max} also increased slightly, so that the upper

boundary defining the air-entrainment limit of good coating became progressively flatter, until at a curtain height of 17.8 cm the onset of air entrainment became extremely sensitive to flow rate.

For the data in Fig. 2, the curtain height was increased to 20.1 cm, and it was immediately apparent that a qualitative change had taken place. The upper and lower air-entrainment boundaries appeared to have separated, becoming roughly parallel and extending to much higher speeds and flow rates. More significantly, there was no longer a clearly defined maximum coating speed. Instead, coating without air entrainment was possible within the upper and lower boundaries up to a speed of about 75 cm s⁻¹. At higher speeds, a few entrained air bubbles could be detected, but these were small and infrequent. Above the upper boundary and below the lower one, air entrainment was as well defined as before.



The trends seen in Fig. 2 became more marked when the curtain height was increased still further to 25.2 cm, as shown in Fig. 3. The upper and lower air-entrainment boundaries now diverged and the separation between them increased substantially, giving a broad region in which air entrainment was no-where detectable up to speeds of 1000 cm s⁻¹ (the maximum web-conveyance speed). Examination of samples under the microscope using dark field illumination confirmed the absence of air bubbles in the coated layer. Crucially, careful observation of the base of the curtain indicated that between the upper and lower air-entrainment boundaries and to the right of the recirculation boundary, the wetting line was always located beneath the curtain, migrating slowly in the downstream direction as coating speed was increased. Whether or not the two air-entrainment boundaries would eventually join is not clear. The answer must await the outcome of experiments at still higher speeds. It is obviously an interesting question, with both practical and theoretical implications.



The results presented above allow us to draw conclusions about the macroscopic dynamic wetting behavior. First, the maximum speed achieved with the 25.2 cm curtain is approximately 100 times that observed previously in plunging tape experiments with the same solid/liquid combination [7,8]. Evidently, the effect we are dealing with is a major one. Moreover, it appears to operate in the opposite sense to that expected for a Newtonian liquid. Such liquids usually give lower air-entrainment speeds with increasing viscosity (roughly as the 0.7 - 0.8 power) [9]. Here we see an effect that appears to be enabled by a high liquid viscosity.

Perhaps the next most significant observation is that the dynamic wetting behavior evolved smoothly with curtain height. There was no abrupt change in character. The upper and lower air-entrainment boundaries first expanded, until the upper one became more-or-less flat, then separated and progressively pulled apart.

From flow visualization experiments [2] with comparatively low curtains (H = 3 cm and 7 cm) and low viscosity liquids (25 and 57 mPa s) we know that changes in the flow field directly affect the dynamic contact angle at constant wetting-line speed. This effect has been termed "hydrodynamic assist" [1,2]. Variation by as much as 20° was found on simply changing the liquid flow rate or curtain height. Significantly, the effect was more marked for the taller curtain and the higher viscosity liquid. At the same time, the maximum speed before air entrainment increased from 40 cm s⁻¹ for the plunging tape, to about 50 cm s⁻¹ at H = 3 cm and 75 cm s⁻¹ at H = 7 cm (57 mPa s liquid). These effects were seen when the wetting line was directly beneath the falling curtain, which is precisely the configuration found to give the very high coating speeds in the work reported here. It therefore seems entirely reasonable that we are dealing with the same phenomenon, specifically a substantial reduction in the dynamic contact angle arising from

interactions with the flow; in other words hydrodynamic assist. Furthermore, because the speeds and viscosities are much higher in the present work, we may expect that the effect on the dynamic contact angle is correspondingly greater.

We have, of course, no direct evidence about the magnitude of the dynamic contact angle in our experiments. It is possible that beneath the curtain the dynamic contact angle increases to a value approaching 180°, even though no air is apparently entrained. Coating would then be a kind of lamination process. Nevertheless, it seems reasonable to suppose that at the high capillary numbers achieved in these experiments, the contact angle is very largely dependent on the global hydrodynamics of the flow [10]. This is not the case during wetting at low to moderate capillary numbers, where interactions across the solid/liquid interface have a marked influence upon wetting dynamics [7,9,11]. Experiments to determine whether or not the nature of the solid surface affects dynamic wetting under the global conditions described here would be helpful in confirming the mechanisms at work. A negative result would indicate that the information flow is predominately inwards, towards the wetting line. Such a result might be expected, because in curtain coating inertia dominates surfaces tension. For the 25.2 cm curtain the Weber number, $We = \rho Q U_c / \gamma$, varied from 5 to 30 over the range of flow rates investigated, and disturbances propagate down the curtain only. At the same time, the inertial force was comparable with the viscous force. In fiber coating, inertial forces are much smaller and viscous forces dominate surface tension, so in this respect the two techniques differ.

Although the Reynolds number in the present experiments was always close to unity, the impingement speed of the curtain U_c increased from about 140cm s⁻¹ for H = 10.2 cm to 222 cm s⁻¹ for H = 25.2 cm. Hence, the inertial pressure at the foot of the curtain, ρU_c^{2} , increased from about 2.5 kPa to slightly more than 6 kPa. This increase could be significant, but the absolute values are much smaller than the lubrication pressures generated in a fiber-coating die, which can be of order 1 MPa [3,12]. While sound arguments can be advanced for entrained gasses dissolving in the later case, this outcome must be less likely at the pressures found at the foot of the curtain, which are more than 100 times smaller. Furthermore, in fiber coating we know that flow rates and pressures must be carefully adjusted to keep the upstream meniscus in the optimum position to avoid air entrainment [13-15]. A parallel may therefore be drawn with the need in curtain coating to maintain the wetting line beneath the curtain. Thus, it is possible that the very high speeds found in fiber coating are also the result of hydrodynamic assist and are not dependent on gas solubility.

At large curtain heights the curtain becomes extremely thin (< 0.1 mm for the 25.2 cm curtain). Indeed, a feature of curtain coating is that the scale of the flow is very small and so may approach the magnitude of the scale of the physical processes of dynamic wetting, thus maximizing the potential for hydrodynamic assist. The sensitivity of the dynamic contact angle to the flow conditions appears to confirm this [2]. Fiber coating flows share this very small scale, which is determined by the gap between the fiber and the cylindrical coating die. Similar mechanisms might therefore be expected to operate. **Conclusions**. The new results support the idea that at high capillary numbers the dynamic contact angle becomes very largely dependent on the global hydrodynamics of the flow and that it is this that permits the very high coating speeds obtained. The possibility that the effect is due to entrained air dissolving seems less likely. However, more work, both experimental and theoretical, is required to understand this phenomenon and to determine the extent to which the properties of the solid/liquid interface and the molecular mechanisms that must operate in the vicinity of the wetting line can still influence dynamic wetting at high capillary numbers. The theoretical treatment of wetting proposed by Shikhmurzaev would appear to offer a starting point [2]. Further work is also required to determine the limits of high-speed coating in the global regime.

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