Mathematical Model for the Onset of Ribbing

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INTRODUCTION

A major problem in the coating industry is the appearance of a ribbing instability under various operating conditions. The objective of this paper is to predict the onset of ribbing in any equal speed two-roll coater in which the fluid is assumed to be Newtonian.

Figure 1 illustrates a coater having contrarotating rollers of radii R_1 and R_2, each of which moves uniformly with speed U through the nip. The appearance of the ribbing instability has previously received attention from both experimentalists and theoreticians. In particular, Pitts and Greiller (1961) used a system of contrarotating rollers of equal radii and speed to obtain experimental data for the onset of ribbing which they fit by a straight line relating critical capillary number $C_a^* = \eta U/T$ and geometry parameter H_o/R,

$$c_a^* = 31\left(\frac{H_o}{R}\right) \quad (1.1)$$

where the error in the slope is ±7.5. Also using rollers of equal speed but varying size, Mill and South (1967) obtained experimental data which they correlated by

$$C_a^* = 17.3\left(\frac{H_o}{R}\right)^{3/4}. \quad (1.2)$$

More recently Greener et al. (1980), with rollers of equal size and speed, obtained a correlation of the form

$$C_a^* = 1{,}875\left(\frac{H_o}{R}\right)^2 \quad (1.3)$$

At first sight it does seem rather strange that each of these three correlations should involve a different power law. However, Cheng (1981) has clearly demonstrated that all the available data falls into separate regions of the $(C_a, H_o/R)$ plane which does suggest, therefore, that the form of the neutral stability curve, defining the onset of the ribbing instability, may be quite different throughout this parameter space.

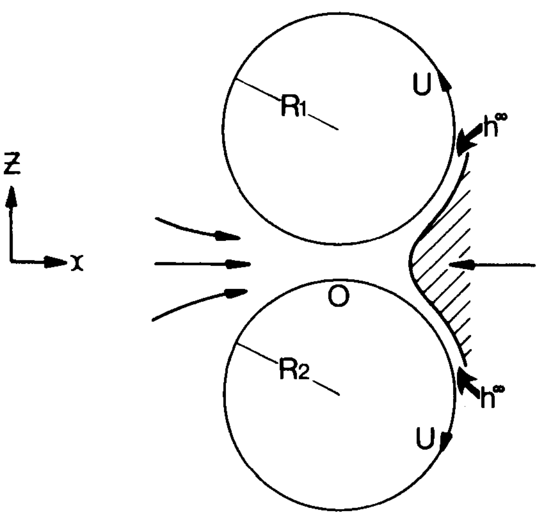

Figure 1. Typical equal-speed two-roll coater.

Theoretical studies were initiated with the work of Pitts and Greiller (1961), Savage (1977), who independently derived a stability criterion for the onset of ribbing. Essentially this stated that a uniform fluid-air interface would remain stable to small disturbances provided $d/dx\,[p + T/r] \leqslant 0$ where p is fluid pressure and r, the radius of curvature of the fluid-air interface. Pitts and Greiller used this criterion together with extra assumptions to derive the equation $C_a^* = 14\,(H_o/R)$ for the stability borderline whereas Savage (1977) and Falle (1976) found reasonable agreement with experiment in the case of the cylinder-plane and wide-angled spreader geometries, respectively.

On close examination it is readily appreciated that the stability criterion is no more than a crude first approximation for the onset of ribbing and hence the aim in this paper is to formulate a more refined mathematical model. In particular we consider the steady and uniform flow of a Newtonian fluid in the narrow gap between the two contrarotating rollers, Figure 1, and then past a uniform, straight line, fluid-air interface. Following a method first introduced by Pearson (1960) and subsequently developed by Savage (1977), a linearized perturbation analysis permits the formulation of a boundary value problem which describes perturbations to the uniform flow. Such perturbations are time-independent and those to the fluid-air interface are represented by a small amplitude harmonic wave of wave number n. An analytic/numerical solution of this boundary value problem yields critical capillary number, C_a^* as a function of (H_o/R) and close agreement between theory and experimental data is observed.

MATHEMATICAL MODEL

For the roll coater shown in Figure 1, the gap thickness $h(x)$ is effectively given by

$$h(x) = H_o + \frac{x^2}{R}\cdot \quad (2.1)$$

The origin of coordinates is taken to lie in the nip with the y axis along the axis of the rollers and the z axis vertical such that the surfaces of lower and upper rollers are given by $z = 0$ and $z = h(x)$, respectively. The gap is assumed to be fully flooded with Newtonian fluid both upstream of the nip and downstream as far as a fluid-air interface at $x = c$ where pressure is atmospheric and taken to be zero. Reynolds equation for the pressure distribution $p(x,y)$ is given by

$$\frac{\partial}{\partial x}\left(h^3\frac{\partial p}{\partial x}\right) + \frac{\partial}{\partial y}\left(h^3\frac{\partial p}{\partial y}\right) = 12\eta U\frac{\partial h}{\partial x} \quad (2.2)$$

where the lubrication regime extends over $\infty < x < s$ and $x = s$ marks the onset of the reverse flow region, Figure 2. Following Savage (1982) we shall assume that separation conditions are valid, that fluid pressure is constant throughout the reverse flow region, and that position $x = s$ is both a stagnation point and a point of separation (where both $u = \partial u/\partial z = 0$). Consequently, any basic, uniform flow which is free from perturbations (and hence ribs) has a pressure distribution, $p^o(x)$, which is a function of x only and given by the following boundary value problem

$$\frac{\partial}{\partial x}\left(h^3\frac{\partial p^o}{\partial x}\right) = 12\eta U\frac{\partial h}{\partial x} \quad (2.3)$$

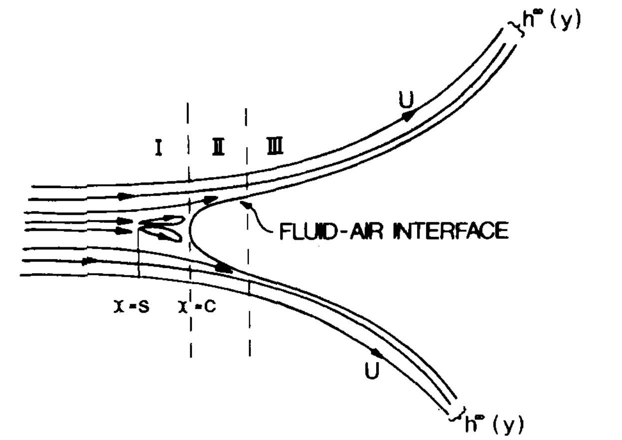

Figure 2. An (x,z) cross section at any station y of a two-roll coater, showing fluid approaching the interface in region I and finally emerging in a uniform layer of thickness $h^\infty(y)$.

$$p^o(-\infty) = 0, \quad p^o(s) = -T/r \qquad (2.4)\ (2.5)$$

where the last condition relates fluid pressure at $x = s$ to surface tension pressure at the fluid-air interface. In addition, the volume flux, Q, past any station x is given by

$$Q = \int_0^{h(x)} u^o dz = 2Uh^\infty \qquad (2.6)$$

where the velocity distribution $u^o(x,z)$ satisfying $u^o = U$ on $z = 0$ and $z = h(x)$ is

$$u^o(x,z) = \frac{p_x^o(z^2 - zh)}{2\eta} + U. \qquad (2.7)$$

Equation 2.6 now yields

$$\left(\frac{\partial p^o}{\partial x}\right)_{x=s} = p_x^o(s) = \frac{12\eta U}{h^2(s)}\left(1 - \frac{2h^\infty}{h(s)}\right). \qquad (2.8)$$

The mathematical model is then complete once h^∞ and r are specified. This is accomplished by means of the separation condition $\partial u/\partial z = 0$, $u = 0$ at $(x,z) = (s,h/2)$ as a result of which there emerges a second condition on pressure gradient

$$p_x^o(s) = \frac{8\eta U}{h^2(s)}. \qquad (2.9)$$

This together with Eq. 2.8, yields

$$\frac{h^\infty}{h(s)} = \frac{1}{6} \qquad (2.10)$$

Also an estimate of the radius of curvature, r, follows from Figure 2; $r = h/2 - h^\infty$ which via Eq. 2.10 yields

$$\frac{r}{h(s)} = \frac{1}{3}. \qquad (2.11)$$

We now seek steady perturbations to the basic flow which we shall assume to vary harmonically with y, to have magnitude ϵ and wave number n such that $x = c + \epsilon \sin ny$ represents the new location of the fluid-air interface, Figure 3, and $x = s + \epsilon \sin ny$ the termination of the lubrication regime. The pressure distribution $p(x,y)$ and velocity distribution (u,v) satisfying boundary conditions $u = U$, $v = 0$ on both $z = 0$ and $z = h(x)$ are given by

$$p(x,y) = p^o(x) + \epsilon G(x) \sin ny \qquad (2.12)$$

$$u(x,y,z) = \frac{p_x^o(z^2 - zh)}{2\eta} + U + \frac{\epsilon G_x(z^2 - zh)}{2\eta} \sin ny \qquad (2.13)$$

$$v(x,y,z) = \frac{G(z^2 - zh)}{2\eta} n \cos ny. \qquad (2.14)$$

The governing differential equation for $G(x)$ is found by substituting Eq. 2.12 into Eq. 2.2 as a result of which terms of order ϵ yield

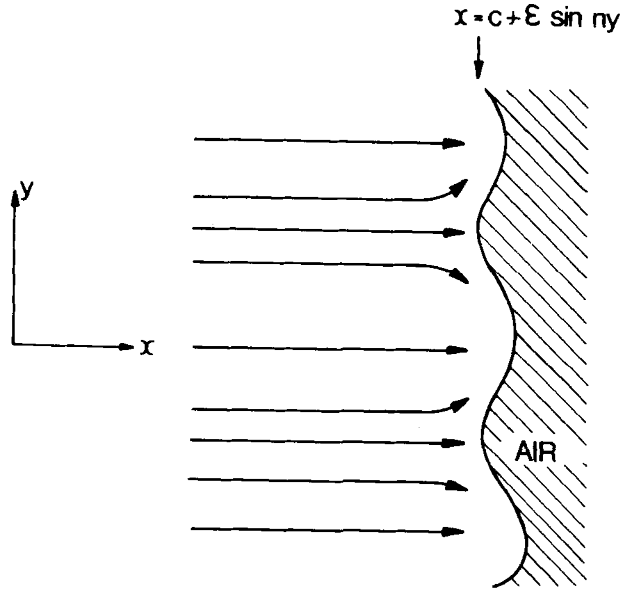

Figure 3. Ribbed fluid-air interface positioned at $x = c + \epsilon \sin ny$.

$$G_{xx} + \left(\frac{3h_x}{h}\right) G_x - n^2 g = 0. \qquad (2.15)$$

For conditions on $G(x)$; these include $G(-\infty) = 0$ (2.16)

together with the pressure condition at the fluid-air interface which balances fluid pressure, surface tension, and atmospheric pressures. On linearizing this condition yields

$$G(s) = \frac{3Th_x(s)}{h^2(s)} - p_x^o(s) + n^2 T \qquad (2.17)$$

where it is recalled that fluid pressure is assumed to be constant over $s \leqslant x \leqslant c$. A third condition is obtained via flow continuity. Figure 2 shows an (x,z) cross section through any station y of the coater in which fluid approaches in the interface in Region I and subsequently moves through a transition region, Region II, before emerging into Region III in a uniform layer of thickness, $h^\infty(y)$. Following Savage (1977) we shall assume that there is negligible lateral diffusion in the transition region such that on linearization flow continuity yields

$$G_x(s) = -p_{xx}^o(s) - \frac{16\eta U h_x(s)}{h^3(s)} \qquad (2.18)$$

SOLUTION FOR THE ONSET OF RIBBING

Uniform flow is described by the boundary value problem (Eqs. 2.3 − 2.5), the solution to which is readily obtained by means of the substitution

$$s = \sqrt{RH_o}\tan\gamma$$

$$-\frac{1}{8C_a}\left(\frac{H_o}{R}\right)^{1/2}\cos^2\gamma$$

$$= \left(\frac{\gamma}{2} + \frac{\pi}{4} + \frac{\sin 2\gamma}{4}\right) - \frac{\sec^2\gamma}{3} \times$$

$$\left(\frac{3\gamma}{8} + \frac{3\pi}{16} + \frac{\sin 2\gamma}{4} + \frac{\sin 4\gamma}{32}\right) \qquad (3.1)$$

By introducing nondimensional variables of the form

$$x = \sqrt{RH_o}X,\ s = \sqrt{RH_o}S,\ G(x) = \frac{12\eta U}{h^2(s)} g(X),\ n\sqrt{RH_o} = N$$

the following boundary value problem for $g(X)$ and N arises;

$$g_{XX} + \frac{6X}{1 + X^2} g_X - N^2 g = 0, \ -\infty < X < S \qquad (3.2)$$

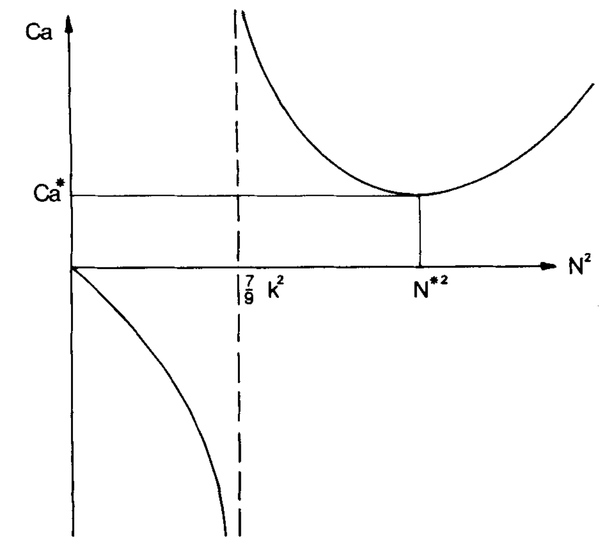

Figure 4. Capillary number C_a against N^2.

$$g(S) = -\frac{2}{3} + \frac{1}{4C_a}\left(\frac{H_o}{R}\right)^{1/2} S + \frac{1}{12C_a}\left(\frac{H_o}{R}\right) N^2(1 + S^2)^2 \quad (3.3)$$

$$g(-\infty) = 0,\ g_X(S) = -\frac{2S}{3(1 + S^2)} \quad (3.4), (3.5)$$

Rather than seek a numerical solution to Eq. 3.2, we shall follow Savage (1977) and obtain an approximate analytic solution since more insight together with a sufficient degree of accuracy can be gained.

Approximate Analytic Solution

In Eq. 3.2 it is the coefficient of g_X, namely $6X/(1 + X^2)$ which hinders the determination of an exact solution. This coefficient which is associated with the geometry of the gap close to the nip, also arises in Eq. 2.3 where it is of crucial importance in determining the location of the separation point. Its significance in Eq. 3.2 is less crucial since it affects only the decay of perturbations arising at the fluid-air interface. Experimental evidence confirms that such perturbations do decay rapidly and therefore a convenient approximation is to replace $6X/(1 + X^2)$ by a local value—its value at $X = S$, for example. The solution to Eq. 3.2 which satisfies Eqs. 3.3 and 3.4 is therefore

$$g(X) = g(S)e^{\omega(X-S)} \quad (3.6)$$

where $\omega = -k + [k^2 + N^2]^{1/2}$ and $k = 3S/(1 + S^2)$ (3.7), (3.8)

Substituting Eq. 3.6 into Eq. 3.5 yields C_a as a function of N/k and d where $d = 3S(H_o/R)^{1/2}$.

$$C_a = \frac{\frac{d^2}{8}\left[\left(\frac{N}{k}\right)^2 + \frac{1}{d}\right]\left(\left[1 + \left(\frac{N}{k}\right)^2\right]^{1/2} - 1\right)}{\left[1 + \left(\frac{N}{k}\right)^2\right]^{1/2} - \frac{4}{3}} \quad (3.9)$$

The graph of C_a against N^2, Figure 4, consists of two branches with an asymptote where $N^2 = 7/(9)k^2$. The lefthand branch is of no physical interest whereas the right branch exhibits a minimum at a critical point (C_a^*, N^{*2}) such that for $C_a < C_a^*$ the flow is stable with no ribs appearing on the fluid-air interface, for $C_a = C_a^*$ the flow is neutrally stable with a disturbance of wave number N^*, and for $C_a > C_a^*$ the flow is unstable.

For any (H_o/R) the critical capillary number is found by solving $\partial C_a/\partial N = 0$ and using Eq. 3.9. This procedure then yields the following cubic equation

$$d^2(11C_a^* + d^2 - d)\left(21C_a^* + d^2 - \frac{3d}{2}\right) = (8C_a^* + d^2 - d)\left(6C_a^* + d^2 - \frac{3d}{4}\right)^2 \quad (3.10)$$

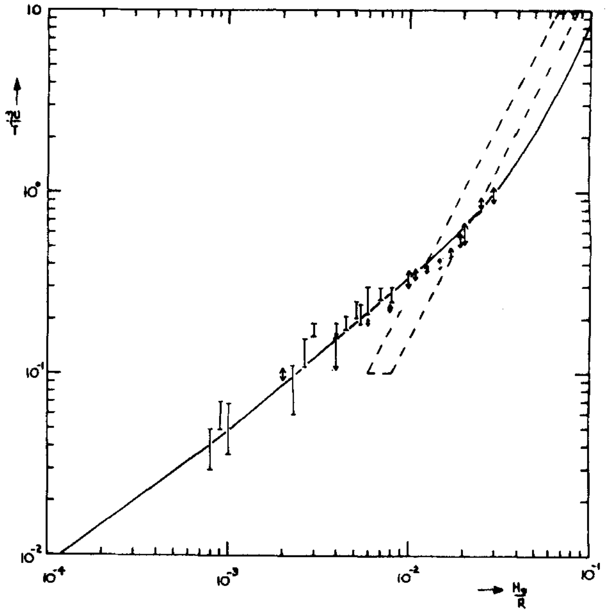

Figure 5. Neutral stability curves representing the onset of ribbing in a two-roll coater together with experimental data from Pitts and Greiller, Mill and South I, Grener et al.

The solution of the above equation together with Eq. 3.2 enables C_a^* to be found for any (H_o/R) and yields the neutral stability curve shown in Figure 5. In particular we observe that over the range $10^{-2} < C_a < 10^{\circ}$ there is very good agreement with experimental data provided by Mill and South, (1967) and Pitts and Greiller (1961). Beyond $C_a = 10^{\circ}$ the neutral curve steepens considerably with slope between 1 and 2, and runs parallel to, yet is slightly displaced from the set of data due to Greener et al. (1981).

In spite of the many simplifying assumptions it is clear that the mathematical model considered does provide reasonably accurate predictions for the onset of ribbing in an equal-speed two-roll coater and moreover offers extension to the general case of a varible speed coater.

NOTATION

C	= position of the fluid-air interface
$G(x)$	= function giving the x variation of the perturbation pressure
$h(x), h(s)$	= gap thickness at stations x and s
h^∞	= thickness of uniform layers on upper and lower rollers
H_o	= nip width
k	= constant $k = 3S/(1 + S^2)$
n	= wave number of the disturbance
$p^o(x)$, $P(x,y)$	= Pressure in uniform flow and perturbed flow respectively
Q	= volume flux
r	= radius of curvative of fluid-air interface
R_1, R_2	= Radii of upper and lower rollers
R	= average radius $2/R = 1/R_1 + 1/R_2$
T	= surface tension
U	= roller speed
(u,v)	= velocity components in the x and y directions
(x,y,z)	= Cartesian coordinates
ϵ	= magnitude of the disturbance
η	= Newtonian fluid viscosity
ω	= $\omega = -k + [N^2 + k^2]^{1/2}$
γ	= $C = \tan\gamma$

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Testing the Relative Importance of Temperature and Concentration Gradients in Catalyst Pellets

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The intrapellet concentration and temperature gradients within a catalyst pellet cause the reaction rate to vary with position. To predict the average rates, simultaneous solution of the differential equations for the temperature and concentrations is required. The temperature and the concentration dependence of the rate expressions are generally nonlinear. Also, for many systems more than one chemical reaction has to be considered. For such systems, it is difficult to predict the observed rates by the solution of the energy and species conservation equations. The mathematical theory of diffusion and reaction in porous catalysts is discussed in detail by Aris (1975). Treatment of nonisothermal conditions within a pellet is also reviewed by Petersen (1965a), Smith (1970), Froment and Bischoff (1979), and Carberry (1979).

When the activation energy of the rate constants is not small, the impact of thermal gradients on the activity and selectivity of catalysts is much more important than the concentration gradients. For an exothermic reaction, the increase of the rate due to temperature rise within the pellet can more than offset the decrease in rate due to drop in reactant concentration. From the practical point of view it is quite important to know the relative importance of diffusion and heat transfer on the observed rates and the conditions under which the effects of temperature and concentration gradients can be neglected.

Anderson (1963) derived a criterion to test the isothermal behavior of a catalyst pellet. The Anderson criterion

$$R_{(i)_a} \left|-\Delta H_r\right| \frac{r_o^2}{T_s \lambda_e} < \frac{3}{4} \frac{T_s R_g}{E_a}, \tag{1}$$

assumes that the rate of reaction depends on temperature in the Arrhenius fashion and does not consider the effect of temperature rise on the concentration gradients and diffusion limitations. Weisz and Hicks (1962) also considered the nonisothermal behavior of the catalyst pellet, and a set of computations were reported for the prediction of effectiveness factors under nonisothermal conditions. Weisz and Prater (1954) proposed a criterion to test the importance of diffusional limitations on the observed rates for isothermal pellets. This criterion is later generalized by Petersen (1965), Hutchings and Carberry (1966), Schneider and Mitschka (1966), Bischof (1967), Hudgins (1968), and Narshimhan and Guha (1972). The criterion developed by Hudgins is also applicable for reactions having other than power-type rate expressions.

$$R_{(i)_a} \frac{r_o^2}{C_{d,s} D_j} < \frac{1}{C_{j,s}} \frac{R_{(i)_s}}{\left(\frac{\partial R_{(i)}}{\partial C_j}\right)_s} \tag{2}$$

Doğu and Doğu (1980, 1982) generalized the Hudgins criterion to bidisperse porous catalysts and to multiple reaction systems. In this work, a general criterion is developed to test the relative importance of diffusion and heat transfer limitations on the observed rates of reactions catalyzed by porous solids. The criterion is applicable to reactions conforming to any rate law. It can be used even if there are more than one temperature-dependent parameters in the rate expression, and it is derived for the general case of a multiple reaction system.

For a multiple reaction system with n-independent reactions and m species, the intrinsic rate of reaction i can be given by a Taylor expansion about the surface temperature and concentrations. The terms higher than the first are neglected considering small deviations of these variables from the surface values.

$$R_{(i)} = R_{(i)_s} + \sum_{j=1}^{m} \left(\frac{\partial R_{(i)}}{\partial C_j}\right)_s (C_j - C_{js}) + \left(\frac{\partial R_{(i)}}{\partial(1/T)}\right)_s \left(\frac{1}{T} - \frac{1}{T_s}\right) \tag{3}$$

For such systems temperature and concentration profiles can be approximated as

$$\frac{1}{T} - \frac{1}{T_s} = \delta \left(1 - \frac{r^2}{r_o^2}\right) \tag{4}$$

and

$$C_j - C_{j,s} = -\beta_j \left(1 - \frac{r^2}{r_o^2}\right) \tag{5}$$

The effectiveness factor,

$$\eta = \frac{R_{(i)_a}}{R_{(i)_s}} = 3 \int_0^1 \frac{R_{(i)}}{R_{(i)_s}} \rho^2 d\rho \tag{6}$$

is then obtained by substituting Eqs. 3, 4 and 5 into Eq. 6 and carrying out the integration

$$\eta = 1 - \frac{2}{5R_{(i)_s}} \sum_{j=1}^{m} \beta_j \left(\frac{\partial R_{(i)}}{\partial C_j}\right)_s + \frac{2}{5R_{(i)_s}} \delta \left(\frac{\partial R_{(i)}}{\partial \left(\frac{1}{T}\right)}\right)_s \tag{7}$$