

STABILITY AND DISTURBANCE OF COATING FILMS

Project 2696-67

Report One A Progress Report to MEMBERS OF THE INSTITUTE OF PAPER CHEMISTRY

March 5, 1980



THE INSTITUTE OF PAPER CHEMISTRY Post Office Box 1039 Appleton, Wisconsin 54912 Phone: 414/734-9251

May 16, 1980

TO: MEMBERS OF THE INSTITUTE OF PAPER CHEMISTRY

Project 2696-67 Final Report

STABILITY AND DISTURBANCE OF COATING FILMS

Enclosed please find a copy of the first and final report on the "Stability and Disturbance of Coating Films". Metering and application systems for adhesives and coatings were studied. Two types of disturbances, namely "ring type" and "Irregular" disturbances were observed and analyzed.

The ring type disturbances were induced and controlled by conditions in the fluid meniscus and the gap between the two rolls. The irregular disturbances are caused by a competition between fluid and aerodynamic stresses on the fluid surface in the gap region between the rolls.

Formulas based on theoretical considerations and experimental findings describe the phenomena and their boundary conditions.

Sincerely,

Douglas Wahren Vice President-Research

DW/el

THE INSTITUTE OF PAPER CHEMISTRY

Appleton, Wisconsin

STABILITY AND DISTURBANCE OF COATING FILMS

Project 2696-67

Report One

A Progress Report

to

MEMBERS OF THE INSTITUTE OF PAPER CHEMISTRY

March 5, 1980

TABLE OF CONTENTS

	Page	
SUMMARY	1	
INTRODUCTION		
THE BASIC DIFFERENTIAL EQUATION		
FILM DISTURBANCE ON A SINGLE ROTATING ROLL		
COUNTER-ROTATING CYLINDERS		
Analytical Model	12	
Experimental Apparatus	19	
Observation of Ring Instability	19	
Dynamics of Menisci for Newtonian Liquids	19	
Rheological Characteristics of Adhesives	20	
Comparison of Theoretical and Experimental Data		
CO-ROTATING APPLICATOR-METERING ROLL SYSTEM		
The Dynamic Meniscus and Types of Disturbances		
Formation of a "Perfect Layer"		
Metering Rate and Disturbances	32	
CONCLUSIONS		
ACKNOWLEDGMENTS		
REFERENCES		
APPENDIX I. A VARIATIONAL APPROACH TO THE PROBLEM OF RING INSTABILITY		
APPENDIX II. FORMULAS FOR ADHESIVES		

THE INSTITUTE OF PAPER CHEMISTRY

Appleton, Wisconsin

STABILITY AND DISTURBANCE OF COATING FILMS

SUMMARY

In the studies of two-roll metering and application systems, two types of disturbances were observed. These were termed "ring type" and "irregular" disturbances. This research established that the physical reason for the appearance of the ring type instability is the competition between surface tension and centrifugal forces at the liquid-air interface. The rings are generated at the surface of the dynamic liquid meniscus, in the gap between the rolls, because of the very large centrifugal forces there. Considering conditions of a constant interfacial pressure difference (pressure jump), one can reduce the problem to a problem with only one free parameter, viz., the radius of the meniscus, and calculate the wavelength of the disturbances.

There is no single formula which will adequately describe the dynamic meniscus. Its curvature depends on the rheological properties of the fluid and on the kinematic conditions in the process. Dimensional analysis is combined with experimental findings to yield a formula for the radius of the meniscus for fluids having a high yield stress for the case of two counter-rotating rolls.

The rheological behavior of the flowing starch adhesive in the dynamical meniscus is analyzed. The theoretical and experimental studies show that systems using two counter-rotating rolls practically always produced ring-type instabilities with all types of fluids.

The picture is more complex for co-rotating roll systems. When non-Newtonian adhesives are used, ring type disturbances are observed in one zone of roll speed ratios, and irregular disturbances are observed in another zone. The two Page 2 Report One

zones are separated by a speed ratio zone (a "speed window") where a more or less perfectly stable fluid layer is observed. When Newtonian oils are used, there are two such speed windows. The first one corresponds to very low metering roll speeds and a minimum of liquid transfer to the applicator roll. The second stable zone occurs at high metering roll speeds and yields a maximum of liquid transfer. The physical reason for the high transfer rate in the high speed "window" is considered and shown to be the thin air layer following the surface of the metering roll. The air pumped into the metering gap returns along the applicator roll and accelerates the film on the applicator roll in the process. Under these conditions the fluidair interface may become unstable, leading to the "irregular" type of disturbance.

INTRODUCTION

The present study was undertaken because of a need in the industry to apply adhesives in a continuous process to corrugating medium. The process of forming a thin liquid layer and its transfer to a moving surface, i.e., the corrugating medium, will be referred to as a metering and transfer process. Similar problems of forming a thin liquid layer occur in the coating of paper and board and in the production of photoemulsion films, various kinds of substrates, thin plastic films, etc. In most cases it is desirable and sometimes necessary to produce a coating film of uniform thickness. In corrugating, disturbance of the adhesive film surface causes nonuniform or even interrupted adhesive contact, which decreases the overall mechanical strength of the board. In the case of photoemulsion films, film nonuniformity causes variability of photographic properties, disturbed color balance, etc.

Theoretical bases for understanding the physics of the instability phenomina and some experimental data are presented in the following problem review.

The simplest type of coater is a single rotating cyliner, such as studied by Yih (1). He used the same mathematical approach to the problem as Taylor (2) did in his classical paper published in 1923. Taylor studied the secondary laminar flow, the so-called Taylor vortex, in the gap between two coaxial cylinders (2). The physical basis for the appearance of secondary laminar flow, disturbing the principal quasi-Couette flow, is the competition between centrifugal forces and the forces of visous flow. To the number of physical parameters which were studied, Yih also added surface tension of the liquid. The rest was a repeat of Taylor's mathematical procedure. Under conditions of neutral stability, Yih determined the wave number of periodic disturbance. Yih also carried out experimental studies and demonstrated that his results were in good agreement with observed wave lengths of ring disturbance of a film on a single rotating roll. In his appendix to Yih's

paper, Kingman $(\underline{3})$ showed that the viscosity of the film had little influence on the wavelength of the disturbance and that Yih's main results could be obtained without considering viscosity in the investigation. The basic formula of Yih and Kingman for the wavelength of periodic disturbance can be represented in the form:

$$\lambda = \sqrt{\frac{2\pi R}{1 + \frac{\rho \omega^2 R^3}{\sigma}}}$$

where λ - wavelength of ring disturbance R - radius of roll

 ω - angular velocity of roll

 ρ - density of liquid

 σ - surface tension of liquid

In view of the fact that the studies of Yih and Kingman were based on farreaching (intense) linearization of the initial equations on movement of a liquid, their results are limited to the wavelength of the disturbance. The experimental report of Karweit and Corrsin (4) should be noted as a relevant publication. The authors observed the periodic system of rings on the inner surface of a hollow rotating cylinder partially filled with a liquid. The periodic disturbance of a liquid surface in a rapidly rotating, partially filled hollow cylinder was studied earlier by Phillips (5). Theoretical investigations were carried out by Phillips when the thickness of the liquid layer spread out on the inner surface of the cylinder had the same order of magnitude as the radius of the cyliner. For this reason, the author did not use surface tension as one of the physical parameters. Surface tension is an important (inherent) part of the energy balance of thin coating films, however.

To sum up the above-mentioned reports, we want to point out that they have one thing in common — each is a theoretical description or experimental observation

Page 4 Report One

(1)

of the periodic regular type of disturbance of a liquid layer in a field of centrifugal forces.

In spite of the fact that Yih's research was initiated at the request of the paper industry for the purpose of studying instability of coating films, the single roll studied by him does not constitute even the simplest coater which can be used in practice for continuously forming and transferring a liquid film to substrates or corrugating medium.

There are several papers on investigating instability of a liquid film in a system of two <u>counter</u>-rotating cylinders (Fig. 1). Such a system can and has been used in coating processes. Pearson (6), Pitts and Greiller (7) and Savage (8, <u>9</u>) attempted to describe the movement of a liquid in the vicinity of the nip in an approximation to the theory of lubrication from the point of view of stability to small disturbances. Such an approach results in partial or complete loss of the effect of inertia terms, depending on the model. As we shall show later, the centrifugal effect, reflected in the inertia terms, is the principal destabilizing factor in the meniscus zone of a two-roll coater.

Moreover, the above authors investigated only the movement of a Newtonian liquid, while in most cases the coating medium is a non-Newtonian body. In two articles by Mill and South (10) and Greener and Middleman (11) are experimental studies of ring formation on counter-rotating cylinders. Greener and Middleman pointed out that visco-elasticity of a coating medium is an additional destabilizing factor. Instability in applicator-metering roll systems (two co-rotating cylinders) which are widely used for coating paper, is not described in the literature, even though the occurrence of undesirable instabilities is recognized. Page 6 Report One Members of The Institute of Paper Chemistry Project 2696-67



Figure 1. A System of Two Counter-Rotating Rolls. The Fluid Forms a Thin Coating on the Surface of the Rolls. The Formation and Instability of This Coating is Dependent Upon the Shape of the Meniscus. R is the Radius of Undisturbed Meniscus. The Variable Radius of Disturbed Meniscus Surface is Expressed as $R = F(z, \Theta)$

In dealing with the disturbances at the air-fluid interface, we study the problem from the point of view of interfacial science rather than hydrodynamic instability. This approach makes it possible to determine the physical mechanism and to qualitatively describe the disturbances in fluids used as a coatings and adhesives. The results of this type of approach are less sensitive to many details of the arrangement of the process. Page 8 Report One Members of The Institute of Paper Chemistry Project 2696-67

THE BASIC DIFFERENTIAL EQUATION

The differential equation is derived for a disturbed cylindrical liquid surface. The result can be applied to various problems in coating and adhesive metering. As can be shown by a variational approach (see Appendix I) the physical reason for disturbances of a rotating cylindrical surface is a competition between surface tension and centrifugal force. The basic differential equation can also be derived more simply.

We will start with the so-called Young-Laplace equation, which reflects the difference between surface tension and centrifugal force as a pressure difference, ΔP , at the liquid-air interface, a so-called interfacial pressure jump, (<u>12</u>):

$$\Delta P = \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) - \frac{\rho \omega^2 R^2}{2}$$
(2)

Here σ - surface tension of the liquid
ρ - density of the liquid
R₁ and R₂ - principal radii of curvature at a point of the liquid surface
R - instantaneous radius of rotation for a surface point
ω - instantaneous angular velocity for the same surface point

The problem of a permanent angular velocity vector of rotation for all liquid points is considered. If the radius $R = R_0$ is the same for all surface points, the cylindrical surface is nondisturbed, and R_0 is the only radius of curvature. Then Eq. (2) can be rewritten as follows:

$$\Delta P_{\rm o} = \frac{\sigma}{R_{\rm o}} - \frac{\rho \omega^2 R_{\rm o}}{2}$$

(3)

If the surface is weakly disturbed, the difference $R-R_0 = f$ is a small disturbance under condition $f/R_0 << 1$. Moreover, ΔP should be the same at all surface points if ω =constant and if the disturbance is stationary. This can be the case only if there is (inter)compensation of pressure between the competing surface tension and centrifugal pressure terms of Eq. (2). Then, ΔP should have the same value on a disturbed and on a nondisturbed interface. Equating Eq. (2) and (3), we obtain:

$$\sigma\left(\frac{1}{R_1} + \frac{1}{R_2}\right) - \frac{\rho\omega^2 R^2}{2} = \frac{\sigma}{R_0} - \frac{\rho\omega^2 R_0^2}{2}$$
(4)

The curvature of disturbed surfaces may be expressed in cylindrical coordinates (z, R, θ) by the following formula (<u>13</u>):

$$\frac{\frac{1}{R_{1}} + \frac{1}{R_{2}}}{R_{1}} = \frac{\sqrt{\frac{1 + \frac{1}{R'} \left(\frac{\partial R}{\partial \theta}\right)^{2} + \left(\frac{\partial R}{\partial z'}\right)^{2}}{R}} - \frac{1}{R} \frac{\partial}{\partial z}}{R} \left[\frac{R \frac{\partial R}{\partial z}}{R \sqrt{1 + \frac{1}{R^{2}} \left(\frac{\partial R}{\partial \theta}\right)^{2} + \left(\frac{\partial R}{\partial z}\right)^{2}}}{\frac{\partial R}{\partial \theta} \left(\frac{R \frac{\partial R}{\partial \theta}}{R \sqrt{1 + \frac{1}{R^{2}} \left(\frac{\partial R}{\partial \theta}\right)^{2} + \left(\frac{\partial R}{\partial z}\right)^{2}}}{R \sqrt{1 + \frac{1}{R^{2}} \left(\frac{\partial R}{\partial \theta}\right)^{2} + \left(\frac{\partial R}{\partial z}\right)^{2}}} \right]}$$
(5)

Taking into account that $R(z, \theta) = R_0 + f(z, \theta)$ and $\left(\frac{\partial f}{\partial z}\right)^2 << 1; \left(\frac{\partial f}{\partial \theta}\right)^2 << 1$, because f is a small disturbance, Eq. (5) can be linearized to the formula:

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{1}{R_0} - \frac{f}{R_0^2} - \frac{\partial^2 f}{\partial z^2} - \frac{1}{R_0^2} \frac{\partial^2 f}{\partial \theta^2}$$
(6)

Combining Eq. (4) and (6):

$$\sigma \left(\frac{1}{R_0} - \frac{f}{R_0^2} - \frac{\partial^2 f}{\partial z^2} - \frac{1}{R_0} \frac{\partial^2 f}{\partial \theta^2}\right) - \frac{\rho \omega^2 (R_0 + f)^2}{2} = \frac{\sigma}{R_0} - \frac{\rho \omega^2 R_0^2}{2}$$
(7)

Simplifying Eq. (7), and neglecting term of order f^2 , the differential equation for small disturbances of a rotating cylindrical liquid surface is finally obtained:

 $\frac{\partial^2 f}{\partial z^2} + \frac{1}{R_0^2} \frac{\partial^2 f}{\partial \theta^2} + \left(\frac{1}{R_0^2} + \frac{\rho \omega^2 R_0}{\sigma}\right) \cdot f = 0$ (8)

FILM DISTURBANCE ON A SINGLE ROTATING ROLL

This problem was considered by Yih and Kingman $(\underline{1},\underline{3})$. Let R_0 be the radius of the nondisturbed liquid surface and ω the angular velocity of the roll. The ring disturbance is sought in the form:

$$f = \varepsilon \cos kz \tag{9}$$

where ε - the small amplitude

k - wave number of disturbance

Substituting Eq. (9) in Eq. (8), the wave number is obtained:

$$k = \frac{1}{R_0} \sqrt{1 + \frac{\rho \omega^2 R_0^3}{\sigma}}$$
(10)

The wavelength of the disturbances is:

$$\lambda = \frac{2\pi}{k} = \frac{2\pi R_0}{1 + \frac{\rho \omega^2 R_0^3}{\sigma}}$$
(11)

This result is the same as that obtained by Yih and Kingman $(\underline{1,3})$. The variational approach, used in Appendix I, also yields the same expression.

COUNTER-ROTATING CYLINDERS

The simplest system is a system of two identical rolls separated by a narrow gap. The rolls rotate with the same surface velocity and direction in the gap, i.e., in opposite angular directions. Such a system has been considered in the literature (6-9,11). The method described above is now used to analyze this system. The results are then qualitatively generalized to the applicator-metering roll system.

ANALYTICAL MODEL

Figure 1 shows a schematic of the system being considered. Figure 2 illustrates the disturbances to be considered. Figure 3 shows the experimental apparatus used. Figures 4 and 5 show actual photographs of disturbances. We will study the disturbances of the liquid-air interface in the meniscus zone and consider the undisturbed meniscus surface as a free cylindrical surface with radius R_0 . According to the Landau-Levich theory of coating transfer processes (14,15), the cylindrical meniscus surface has to have an asymptotic contact line with a drawing surface. In our case, with two roll surfaces, each having the radius ro, the contact lines are at points A and A' (Fig. 1). Generally speaking, this is not exact since the film thickness is disregarded. Moreover, Laudau and Levich (14, 15)neglected the movement of the liquid in the meniscus. The movement of liquid is taken into account in Eq. (8). However, we assume that such a mathematical inconsistency makes it possible to correctly describe the qualitative effect of the formation of disturbances. This seems to be permissible at least at low speeds, because the shape of the undisturbed meniscus is then only weakly distorted from being a free circular cylindrical surface with the radius R_0 .

1



Figure 2. Parameters of the Two Counter-rotating Roll System and of the Disturbances Being Generated. λ is the Wavelength of Ring Disturbance. f is the Small Disturbance

The fluid forms a thin coating on the surface of the rolls. The formation and instability of this coating is dependent upon the shape of the meniscus. R_0 is the radius of undisturbed meniscus.

The variable radius of disturbed meniscus surface is expressed as $R = F(z,\theta) = R_0 + f(z,\theta).$



Figure 3. Experimental Metering Roll System

As we noted by reference to the Landau-Levich method, the free surface of a meniscus with a radius R_0 must make asymptotic contact with the surface of the rotating roll. We will extend this requirement to the case of the disturbed meniscus as well. Thus, a small disturbance is equal to zero along the asymptotic contact line. (This asymptotic contact line is placed on the cylindrical roll surface under the physical liquid layer.) The last requirement gives the following boundary condition:

$$f(\theta_0, z) = 0 \tag{12}$$

where θ_0 - the sectorial meniscus angle (see Fig. 1).



Figure 4. Side View of Meniscus and Ring Disturbances. Speed $V_0 \simeq 50$ cm/s. Gap $d \simeq 0.4$ mm

Let us rewrite Eq. (8) in the form:

$$\frac{\partial^2 f}{\partial z^2} + \frac{1}{R_0^2} + \frac{\partial^2 f}{\partial \theta^2} + \left(\frac{1}{R_0^2} + \frac{\rho V^2}{\sigma R_0}\right) f = 0$$
(13)

The method of solving Eq. (13) under the boundary condition in Eq. (12) is to reduce V^2 to the constant average value. We will suppose that (linear hypothesis):

$$V = V_0 \frac{\theta}{\theta_0} \tag{14}$$



Figure 5. Side View of Meniscus and Ring Disturbances. Speed $V_0 \simeq 50$ cm/s. Gap $d \simeq 1$ mm

Then,

$$V^{2} \rightarrow \left\langle V^{2} \right\rangle_{\theta} = \frac{1}{\theta_{0}} \int_{0}^{\theta_{0}} V^{2}_{0} \frac{\theta^{2}}{\theta^{2}_{0}} d\theta = \frac{1}{3} V^{2}_{0}$$
(15)

By substituting $\langle V^2 \rangle_{\theta}$ from Eq. (15) in place of V^2 in Eq. (13) we get:

$$\frac{\partial^2 f}{\partial z^2} + \frac{1}{R_0^2} \frac{\partial^2 f}{\partial \theta^2} + \left(\frac{\rho V_0^2}{3\sigma R_0} + \frac{1}{R_0^2} \right) \cdot f = 0$$
(16)

Eq. (16) is a partial differential equation of the second order with constant coefficients.

We need a solution of Eq. (16) as a wave along the axis z:

$$f = A\cos n\theta \cdot \cos kz \tag{17}$$

Then substituting Eq. (17) into Eq. (16), we find the relationship for the wave number k:

$$-k^{2} - \frac{n^{2}}{R_{0}^{2}} + \left(\frac{p_{1}^{\prime} r_{0}^{2}}{3 \sigma R_{0}} + \frac{1}{R_{0}^{2}}\right) = 0$$
 (18)

$$k = \sqrt{\frac{\rho V_0^2}{3\sigma R_0} + \frac{1}{R^2} - \frac{n^2}{R_0^2}}$$
(19)

From the boundary condition, Eq. (12):

$$f(\theta_0, z) = A\cos n\theta_0 \cos kz = 0$$
⁽²⁰⁾

or:

$$\cos n\theta_0 = 0 \tag{21}$$

$$n\theta_0 = \pi/2 \tag{22}$$

or:

$$n = \pi/2\theta_0 \tag{23}$$

Taking Eq. (23) into account Eq. (19) can be rewritten and we get the formula for the wave number:

$$k = \sqrt{\frac{\rho V_0^2}{3\sigma R_0} + \frac{1}{R_0^2} - \frac{\pi^2}{4\theta_0^2 R_0^2}}$$
(24)

Using geometrical relationships (see Fig. 1);

$$\theta_0 = \arcsin \frac{r_0}{R_0 + r_0} = \arcsin \frac{1}{1 + R_0/r_0}$$
(25)

Page 18 Report One

we get from Eq. (24) and (25):

$$k = \sqrt{\frac{\rho V_0^2}{3\sigma R_0} + \frac{1}{R_0^2}} - \frac{\pi}{2R_0 \arctan \frac{1}{1 + R_0/r_0}}^2$$
(26)

and for wavelength:

$$\lambda = 2\pi/k \tag{27}$$

Equation (22) has a physical sense only if the inequality

$$\frac{\rho V_0^2}{3\sigma R_0} > \frac{\pi^2}{4R_0^2 \operatorname{arc} \sin^2 \left(\frac{1}{1+R_0/r_0}\right)} - \frac{1}{R_0^2}$$
(28)

is satisfied. Then:

$$V_{0} > V_{cr} = \sqrt{\frac{3\sigma R_{0}}{\rho}} \left[\frac{\pi^{2}}{4R_{0}^{2} \cdot \arcsin^{2}\left(\frac{1}{1+R_{0}/r_{0}}\right)} - \frac{1}{R_{0}^{2}} \right]$$
(29)

the critical (minimum) speed of the rolls at which the ring disturbances can exist. Obviously, V_{cr} is a small value, because $R_0 << r_0$ in practical cases.

The same reason permits us to neglect the two last terms in Eq. (26), which leads to a simplified formula for the wavelength:

$$\lambda \simeq 2\pi \sqrt{\frac{3\sigma R_0}{\rho V_0^2}}$$
(30)

Figure 2 illustrates the expected ring disturbances for two counter-rotating cylinders.

EXPERIMENTAL APPARATUS

The experimental equipment had two rolls with independently controllable electrical motors (Fig. 3). The gap between rolls could be accurately controlled. The smaller roll could be rotated in both directions. The equipment was essentially the same as that described by Jurewicz (<u>16</u>).

OBSERVATION OF RING INSTABILITY

Figures 4 and 5 illustrate some experimental observations of ring disturbances of a liquid adhesive film. The surface velocities V_0 of the two rolls were equal. The conditions corresponding to Fig. 4 were: $V_0 = 50$ cm/sec; gap-thickness, d = 0.04 cm. Conditions for Fig. 5 were: $V_0 = 100$ cm/s; d = 0.1 cm. The radius of the disturbed meniscus depends on V_0 as well as on d. This is partially illustrated by the photographs.

The pictures also show that the disturbances arise in the meniscus zone and that they are caused by the centrifugal forces. Particles close to the surface of the liquid are in a much stronger field of centrifugal force in the meniscus zone than on the surface of rolls. The reason is that $R_0 << r_0$. There is full observational agreement between the model used (Fig. 2) for the computations-and the pictures of the disturbances (Fig. 4 and 5) in the experimental apparatus.

It should be noted that the flange on the front plane (Fig. 4 and 5) has a larger radius than the main part of the dynamic meniscus, for which calculations have been done. But this is just an edge effect.

DYNAMICS OF MENISCI FOR NEWTONIAN LIQUIDS

Equation (30) was derived on the basis of a balance of interfacial and body forces, without considering the rheological properties of the liquid. As a result,

the radius of the dynamic meniscus, R_0 , remained undetermined. The simplifying assumption was made that this radius remains constant and does not depend on the angle θ . The shape of the dynamic meniscus remains one of the unsolved problems in fluid mechanics and interfacial science. The equation of motion for a Newtonian surface fluid in its general form was investigated by Scriven (17). Unfortunately, this work was not pursued enough to be applied to concrete problems dealing with the shape of dynamic menisci. We are not concerned with studies dealing with small deviations of the shape from a circular meniscus, since the deviations are of the same order as the disturbances under consideration. Such an approach can lead to only small deviations from the radius of a circular cylindrical meniscus which can be determined within the quasi-static approach of Landau and Levich (12,13,15). This radius is expressed by the equation:

$$R_0 \simeq h (\sigma/\mu V_0)^{2/3}$$
 (31)

where h - film thickness on the roll

μ - dynamic viscosity

This formula was derived with the condition that $\mu V_0/\sigma \ll 1$. The following dynamic condition exists for typical industrial roll adhesive applications: $\mu V_0/\sigma > 1$. The rheological conditions should be considered in more detail, and Eq. (31) cannot be generally applied to the technology of adhesives.

RHEOLOGICAL CHARACTERISTICS OF ADHESIVES

We will try to estimate the rheological characteristics of the adhesive in the dynamic meniscus. The linear velocity of the roll surfaces is approximately 1 m/sec. Observed radii of meniscii, as illustrated in Fig. 4 and 5, are between 0.2 and 1 cm. Then, the rate of deformation in the meniscus zone is approximately $\mathring{\gamma} \simeq V_0/R_0 \simeq 100$ to 500 sec⁻¹. This part of the rheological curve is approximated by the Schwendoff-Binghamm equation of state, as suggested in ref. (18):

$$\tau = \eta \dot{\gamma} + \tau_0 \tag{32}$$

where τ - shear stress

η - plastic viscosity

 τ_0 - yield stress

A typical formula for a starch based "carrier" type of adhesive as used is given in Appendix II. The rheological characteristics of the adhesive were measured with a Hercules cone and plate type viscosimeter.

Typically, the results were: $\eta \simeq 2p$, $\tau_0 \simeq 650 \text{ d/cm}^2$. We can then estimate the viscous part of the shear stress $\tau = \eta \dot{\gamma} \simeq 200$ to 1000 d/cm^2 , which is of the same order of magnitude as the yield stress τ_0 . Therefore, the "carrier" adhesive behaves essentially as a visco-plastic rheological body in this application.

COMPARISON OF THEORETICAL AND EXPERIMENTAL DATA

If the theory of dynamic menisci for Newtonian liquids had very little development, the corresponding theory for non-Newtonian liquids has not been touched at all. However, the rheological analysis that has been done above for the adhesive in the meniscus shows that an additional parameter, the yield stress τ_0 , is present and must be taken into account.

An attempt was made to combine the relevant set of parameters in such a way that the combination would satisfy the theory of dimensional analysis as well as match the experimentally determined dependence of the wavelength, λ , of the ring disturbances on the surface velocity, V_0 , of the rolls. The following expression for average radius of the dynamic meniscus was obtained:

$$R_0 \simeq 2(\eta V_0/\sigma)^{2/3} (\eta V_0/\tau_0)^{1/2} \sqrt{h}$$
(33)

The parameter $\eta V_0/\sigma$ is the capillary number of the process, η/τ_0 is the characteristic relaxation time of the rheological fluid, and $\eta V_0/\tau_0$ then expresses a characteristic length relating to the relaxation process. The coefficient 2 is the experimental coefficient.

Formula 33 gives the same order of magnitude for the radius, $R_0 \simeq 0.2$ to 1 cm, as we could observe experimentally. Substitution of Eq. (33) into Eq. (30) yields satisfactory agreement with experimental data. This is shown in Fig. 6.



Figure 6. The Wavelength of Ring Type Disturbances as a Function of the Surface Velocity of the Two-roll System. The Curve Drawn Through the Experimental Points Was Computed From Eq. (30) and (33) for h = 0.2 mm, $\eta = 220$ cP, $\tau_0 = 650$ dyn/cm²; $\sigma = 50$ dyn/cm and $\rho = 1$ g/cm³

CO-ROTATING, APPLICATOR-METERING ROLL SYSTEM

THE DYNAMIC MENISCUS AND TYPES OF DISTURBANCES

Experiments were carried out with the equipment shown in Fig. 3 but with co-rotating rolls. The bigger roll represents the applicator roll and the small one the metering (or doctor) roll. The doctor removes the liquid layer from the surface of the metering roll. The layout is shown in Fig. 7.

The experiments with carrier and noncarrier adhesives show that for a given velocity, V_a , of the applicator roll the region of the metering roll velocity, V_m , can be divided into three zones:

- Zone 1: $0 < V_m < V_{m_1}$ (see Fig. 8, 9). In this zone the contact line is placed above mid-nip. The meniscus radius is finite, and ring disturbances are observed.
- Zone 2: $V_{m_1} < V_m < V_{m_2}$ (see Fig. 10, 11). In this zone the meniscus is practically absent. The contact line is placed approximately at midnip, and a perfect nondisturbed layer is observed.
- Zone 3: $V_{m} > V_{m}$ (see Fig. 12). In this zone irregular disturbances, "blotching," are observed. There is no stationary contact line.

The wave number of the ring disturbances, Eq. (19), depends neither on the position of the asymptotic contact line nor on the position of the zero velocity point in the meniscus. Further study leads to the conclusion that the second and third terms under the square root, Eq. (19), almost cancel out. Because $V_{\rm cr}$ is so

Page 24 Report One







Figure 8. Ring Disturbances When $0 < V_m < V_{m_1}$ (Zone 1)



Figure 9. A Close-up of the Nip Zone of the Applicator – Metering Roll System Shows the Curved Meniscus When $0 < V_m < V_m_1$



Figure 10. Perfect Layer When $V_{m_1} < V_m < V_m$ (Zone 2)

small, we were unable to establish values of V_{cr} in our experiments with applicator-metering roll systems. Therefore, we can suggest that, in theoretical consideration of this system, the same terms in Eq. (19) will nearly cancel. This suggestion leads to the same simplified formula for wavelength in the applicator-metering rolls system as Eq. (30):

$$\lambda \simeq 2\pi \sqrt{\frac{3\sigma R_0}{\rho V_0^2}}$$

(34)

Here, V_0 is the velocity of the applicator roll and R_0 is the effective radius of the dynamic meniscus. This radius should correspond to a zone of the dynamic meniscus where vector projections of surface velocities on a vertical plane (\vec{V}_a in midnip is vertical) have the same direction as vector projections of the applicator roll speed in the area of the nip zone.



Figure 11. A close-up of the Nip Zone in the Applicator-Metering Roll System When $V_{m_1} < V_m < V_m$ (Zone 2)



Figure 12. Irregular Disturbances When $V_m \ge V_{m_2}$ (Zone 3)

FORMATION OF A "PERFECT LAYER"

As pointed out above there exists a "speed window," $V_{m_1} < V_m < V_{m_2}$, in which a nearly perfect layer of the fluid is formed. The explanation of the existence of such a speed window starts with a consideration of Eq. (34). Obviously, if R_0 tends to either zero or infinity, the ring type disturbances will disappear. This appears to be what actually happens in the transition from ring disturbances into the speed window where a "perfect layer" is formed.

The exact mechanism of the transition is not quite clear. From experimental observations it appears, however, that for Newtonian fluids the radius of the meniscus tends toward zero. This can be seen as a gradual diminishing of the wavelength as the speed of the metering roll is gradually diminished.

For carrier-type adhesives, which exhibit a substantial yield stress, the opposite appears to be true. It has not been possible to thoroughly document this transition, nor to fully evaluate the basic physical reasons, but the observations to date indicate that the radius of the meniscus tends to infinity for liquids having an appreciable yield stress.

The reasons for the transition from zone 2, the "perfect layer" speed window into zone 3, where "irregular" disturbances occur, is more complex and is discussed in the two following sections.

The zone $V_{m_1} < V_m < V_{m_2}$ is the "speed window" in which a "perfect layer" is formed. Experiments were made with two carrier and two noncarrier types of adhesives. The results are presented in Fig. 13 through 19. Various sizes of rolls were employed, and some of the experiments were actually made on the IPC pilot paper-coating machine.



Figure 13. Perfect Layer "Speed Window" for No-carrier Adhesive, Gap 100 µm



Figure 14. Perfect Layer "Speed Window" for No-carrier Adhesive, Gap 200 µm

.















Figure 19. Perfect Layer for No-carrier Adhesive on Pilot Coating Machine. $V_m = 150 \text{ cm/sec}; V_m/V_a = 0.61; \text{ gap } 200 \text{ }\mu\text{m}$

METERING RATE AND DISTURBANCES

It is desirable to establish a functional relationship between the metering rate of the liquid in applicator-metering roll systems and the stability of the liquid film. In the past, Jurewicz (<u>16</u>) investigated the transfer of adhesives and liquids in the same system. Comparison of his and our results lead to the conjectural conclusion that a nondisturbed perfect layer is observed when there is a condition of minimum film thickness on the applicator roll.

We studied the metering rate curve and observed the disturbances at the same time. Polybutane oil, viscosity $\mu = 40$ cP, was used as the liquid. Figure 20 illustrates the dependence of the film thickness on the surface of the applicator roll on the speed of the metering roll V_m . The applicator roll speed,

 $V_{\rm a}$, was 90 cm/sec. A perfect layer was observed in two zones. The first zone $(V_{\rm m} = 15 \text{ to } 30 \text{ cm/sec})$ corresponds to a minimum of h. In the second zone, in the range $V_{\rm m} = 140$ to 240 cm/sec, the film thickness, h, is larger than the gap thickness, d. This has also been observed by Nikkel (19), Wahren (20) and Kartsounes (21). So, it is unlikely that this effect is a result of an experimental error. An explanation of this effect is given below.

The results shown in Fig. 20 can be divided into five zones. Zones 1 and 2 are analogous to the corresponding zones in the previously described experiments with adhesives. At higher metering roll velocities, the Newtonian oil develops some peculiarities compared to the behavior of the non-Newtonian adhesive. Most remarkable is the existence of a wide interval of velocities, $V_{\rm m}$ = 140 to 240 cm/sec, where a perfect layer is observed. This zone corresponds to a maximum film thickness, h, on the applicator roll (Fig. 20).

We suggest that the contact line, A, is then positioned below the center of the gap, and that the effective radius of the dynamic meniscus, R_0 , tends to infinity (Fig. 21). Then, according to Eq. (30), a condition for the existence of a perfect layer is satisfied. It is important to note that, when the contact line is located below the center of the gap, the surface of the liquid layer is separated from the surface of the metering roll by a layer of air. The air corpuscles adjacent to the metering roll are entrained deep into the air wedge, and the developing pressure gradient causes the air corpuscles which are farther away from the metering roll to move in an opposite direction, i.e., out of the wedge. The net flow rate of air through the gap between the rolls is zero, and hence the velocity profile has a complex form. In a certain interval of roll velocities, the air generates an additional shear stress on the surface of the liquid layer so that $V_1 > V_a$, i.e., the surface velocity of the liquid film on the applicator roll becomes larger

Page 34 Report One

than the surface velocity of the roll. In this case the flow rate of the liquid on the applicator roll is:

$$Q_{\ell} = \frac{V_{a} + V_{1}}{2} h_{1}$$
(35)

where h_1 is the thickness of the liquid in the nip zone. Well past the nip zone, where the influence of the surface friction of the air wedge against the liquid can be disregarded:

$$Q_{l} = V_{a} h \tag{36}$$

so that:

$$h = \frac{h_1}{2} \left(1 + \frac{V_1}{V_a} \right) \tag{37}$$

From Eq. (37) the surface velocity of a liquid as:

$$V_1 = V_2 (2h/h_1 - 1)$$
(38)

If
$$h > h_1$$
, then $|V_1| > |V_a|$ (39)

Let us determine the air layer thickness when the inequality Eq. (39) is satisfied.

The following parameters will be used:

U(y) is the velocity of the air; the y axis is normal to the surfaces of the rolls at midnip; $2h_a = d_a$ is the thickness of the air gap; μ_a is the viscosity of air; $\overline{\nabla P}$ is the scalar projection along the nip of the pressure gradient.



Layers are Indicated. The realistic Estimate of Actual Gap Thickness

Layers are indicies 200 to 240 µm



Page 36 Report One

Members of The Institute of Paper Chemistry Project 2696-67

Because the viscosity of air is independent of pressure over a wide range of pressures and because the problem is one of steady state, the Navier-Stokes equation for a one-dimensional case is applicable:

$$\frac{d^2 U}{dy^2} = \frac{\nabla P}{\mu_a} \tag{40}$$

with boundary conditions:

$$y = -h_{a}; U = V_{1}$$

$$y = +h_{a}; U = V_{m}$$
(41)

The point y = 0 is placed in the middle of the air gap.

The solution of Eq. (40) with conditions as in Eq. (41) leads to the following expression for the velocity profile:

$$U(y) = -\frac{\nabla P}{2\mu_{a}} (h_{a}^{2} - y^{2}) + \frac{1}{2} \frac{y}{h_{a}} (V_{m} - V_{1}) + \frac{1}{2} (V + V_{1})$$
(42)

From the condition that the net flow rate is zero:

$$\begin{aligned} h_{a} \\ Q &= \int U(y) \, dy = 0 \\ -h_{a} \end{aligned} \tag{43}$$

we find the pressure gradient

$$\nabla P = \frac{3\mu_{a} (V + V_{1})}{2 h_{a}^{2}}$$
(44)

The shear stress on the liquid-air interface is:

$$\tau (y = -h_{a}) = -\overline{\nabla P} h_{a} + \frac{1}{2} \frac{\mu a}{h} (V_{m} - V_{1})$$
(45)

Combining this with Eq. (44) and remembering that $2h_a = d_a$, one can rewrite Eq. (45) in the form:

$$\tau (y = -h_{a}) = -\frac{2\mu_{a}}{d_{a}} (2V_{1} + V_{m})$$
(46)

Page 38 Report One Members of The Institute of Paper Chemistry Project 2696-67

Let us denote the direction of $\vec{V}_{\rm m}$ as the positive direction. Then, $V_1 \ge V_a$, if $\tau(y = -h_a) \le 0$, which leads to the conclusion:

$$V_{\rm m} \ge -2 V_1 \tag{47}$$

In the case of our experiment (see Fig. 21), $V_{\rm m} = 2|V_1| \simeq 2|V_a| \simeq 180$ cm/sec.

Keeping in mind the unavoidable experimental errors, we see that this velocity does not differ greatly from the experimental velocity of $V_m = 140$ cm/sec during which an abrupt change is observed in the character of the transfer curve and disappearance of disturbance. From these considerations, noting the curve in Fig. 21, the thickness of the gap can be assumed to be d $\simeq 240 \ \mu m$.

The shear stress on the liquid-air interface can also be expressed through the liquid flow parameters:

$$\tau (y = -h_a) = \mu_l \frac{dV}{dy} = \mu_l \frac{V_1 - V_a}{d - d_a}$$
 (48)

Combining Eq. (46) and (48) and accounting that $d_a << d$, the following formula for the thickness of the air gap can be found:

$$d_{a} = -\frac{\mu_{a} (V_{m} + 2 V_{1})}{\mu_{l} V_{a} (\frac{h}{d} - 1)} d$$
(49)

where V_1 is expressed by:

$$V_1 = V_a \left(\frac{2h}{d} - 1\right) \tag{50}$$

The minus sign appears ahead of the expression [Eq. (49)] because the velocities V_1 and V_a have negative values.

The solution presented is approximate because it does not take into account the parabolic term of the liquid layer velocity profile under the action

of pressure gradient $\overline{\Delta P}$. The shear stress at the liquid-air interface is a function of the flow of the liquid layer; we get:

$$\tau_{\rm LV} = \overline{\nabla P} \, (d - h_{\rm a}) + \frac{1}{2} \, \frac{\mu_{\ell}}{d - h_{\rm a}} \, (V_1 - V_{\rm a}) \tag{51}$$

The precision of Eq. (49) and (50) is better when the second term of Eq. (51) is considerably greater than the first one. In other words, Eq. (49) and (50) are well applicable when the following inequality is satisfied:

$$|V_1 - V_a| \gg |\overline{\nabla P}| (d - h_a) \frac{2}{\mu_{\ell}}$$
(52)

When $h = h_1$ and velocities V_1 and V_a are equal, inequality [Eq. (52)] is not satisfied, and Eq. (49) makes no sense.

The values $\mu_a = 2.10^{-4}$ P, $\mu_l = 40.10^{-2}$ P, $V_m = 240$ cm/sec, $V_a = -90$ cm/sec, $d = 240 \ \mu$ m, $h = 250 \ \mu$ m yield the air wedge thickness $d_a \simeq 10^{-4}$ cm = 1 μ m.

We tried to adjust the nominal gap, d to 200 µm. This results in an air wedge thickness of $d_a = 2.10^{-5}$ cm = 0.2 µm. The mean free path of air molecules at atmospheric pressure is 7.10^{-6} cm. An air wedge thickness of 2.10^{-5} cm is therefore in a borderline region where the air phase may not properly be considered as a continuum. Kinetic slipping may also occur, but is very difficult to calculate. If the air-liquid interface becomes unstable, the position of the contact line will also become unstable. We may then observe this as an irregular disturbance of the liquid film on the applicator roll (see Fig. 12).

CONCLUSIONS

 In the studies of two-roll metering and application systems, two types of disturbances were observed. These were termed "ring type" and "irregular" disturbances.

2. The physical reason for the appearance of the ring type instability is the competition between surface tension and centrifugal forces at the liquid-air interface.

3. The rings are generated at the surface of the dynamic liquid meniscus, in the gap between the rolls, because of the very large centrifugal forces there.

4. Considering conditions of a constant interfacial pressure difference (pressure jump), we could reduce the problem to one with only one free parameter, viz., the radius of the meniscus, and to calculate the wavelength of the disturbances.

5. There is no single formula which will adequately describe the dynamic meniscus. Its curvature depends on the rheological properties of the fluid and on the kinematic conditions in the process.

6. Dimensional analysis was combined with experimental findings to yield a formula for the radius of the meniscus for fluids having a high yield stress in the case of two counter-rotating rolls. Starch-based "carrier" type adhesives typically have yield stress values of 500 d/cm.

7. Systems using two counter-rotating rolls always produced ring-type instabilities with all types of fluids.

8. The picture is more complex for co-rotating roll systems. When "carrier" type adhesives are used, ring type disturbances are observed in one zone.

of roll speed ratios, and irregular disturbances are observed in another zone. The two zones are separated by a speed ratio zone (a "speed window") where a more or less perfectly stable fluid layer is observed. When Newtonian oils are used, there are two such speed windows. The first one corresponds to very low metering roll speeds and a minimum of liquid transfer to the applicator roll. The second stable zone occurs at high metering roll speeds and yields a maximum of liquid transfer.

9. The physical reason for the high transfer rate in the high speed "window" is shown to be the thin air layer following the surface of the metering roll. The air pumped into the metering gap returns along the applicator roll and accelerates the film on the applicator roll in the process.

10. The fluid-air interface may become unstable, leading to the "irregular" type of disturbance. Page 42 Report One

Members of The Institute of Paper Chemistry Project 2696-67

ACKNOWLEDGMENT

The authors thank Dr. D. Wahren, Vice President-Research, for his assistance in the final phase of this work. We also thank and acknowledge the valuable assistance of Dr. Gr. Sivashinsky, Associate Professor of Mathematics, Tel-Aviv University, in the linearization of mathematical equations and general discussion of the problem.

REFERENCES

- 1. Yih, C. S., Proc. Roy. Soc. A258:63(1960).
- 2. Taylor, G. I., Phil. Trans. A223:289(1923).
- 3. Kingman, J. F. C., Proc. Roy. Soc. A258:98(1960).
- 4. Karweit, M. J. and Corrsin, S., Phys. Fluids. 18:111(1975).
- 5. Phillips, O. M., J. Fluid Mech. 7:340(1960).
- 6. Pearson, J. R. A., J. Fluid Mech. 7:481(1960).
- 7. Pitts, E. and Greiller, J., J. Fluid Mech. 11:33(1961).
- 8. Savage, M. D., J. Fluid Mech. 80:743(1977).
- 9. Savage, M. D., J. Fluid Mech. 80:757(1977).
- 10. Mill, C. C. and South, G. R., J. Fluid Mech. 28:523(1967).
- 11. Greener, J. and Middleman, S., Preprint, AIChE 71st Annual Meeting, Miami. November 1978.
- Scriven, L. E., Preprint, Physicochemical Hydrodynamics Conference, Washington, D.C. November 1978.
- 13. Finikoff, G., Differential Geometry, Moscow, 1962.
- 14. Landau, L. D. and Levich, V. G., Acta Physiochemical USSR, 17:42(1942).
- 15. Levich, V. G., Physicochemical Hydrodynamics, Prentice-Hall, 1962.
- 16. Jurewicz, J. T., Project 2696-17, Report Two, A Progress Report to the Fourdrinier Kraft Board Group of The American Paper Institute, November, 1978.
- 17. Scriven, L. E., Chem. Eng. Sci. 12:98(1960).
- 18. Babchin, A. J. and Raevsky, V. G., Adhesives Age, p. 25. December 1975.
- 19. Nikkel, W. A. private communication.
- 20. Wahren, D. private communication.
- 21. Kartsounes, G. T. The Institute of Paper Chemistry, Project 3331-14, Status Report to RAC Container and Board Subcommittee, September 25, 1979.

Page 44 Report One

THE INSTITUTE OF PAPER CHEMISTRY

Alexander J. Babchin Research Associate Engineering Division

Robert J. Ollsh Research Assistant Engineering Division

Approved by:

Douglas Wahren Vice President-Research

APPENDIX I

A VARIATIONAL APPROACH TO THE PROBLEM OF RING INSTABILITY

A thin liquid layer on the surface of a single rotating cylinder will be considered (Fig. 1A). The conditions for minimizing the functional:

$$= E - T \tag{1}$$

are sought. Here;

E =free surface energy

T = kinetic energy (of rotation) of the liquid film.

U



Figure 1A. Ring Phenomenon on a Single Roll

Page 46 Report One Members of The Institute of Paper Chemistry Project 2696-67

The functional, Eq. (1), reflects the competition between surface tension and centrifugal forces. The effect of gravity is neglected. It is also assumed that no fluid is thrown from the roll, and that the system is constrained to maintain a constant fluid volume.

The individual terms of Eq. (1) are written in terms of the parameters shown in Fig. 1, where ω is the angular velocity of rotation.

Free Surface Energy

$$E = \sigma S = 2\pi \sigma \int R \left[1 + (dR/dz)^2 \right]^{1/2} dz$$
 (2)

(3)

(5)

Kinetic Energy

$$T = 2\pi \int_{0}^{\ell} \frac{R}{r} (\frac{1}{2} \rho \omega^{2} r^{2}) r dr dz = \frac{1}{4} \rho \omega^{2} \pi \int_{0}^{\ell} (R^{4} - r_{0}^{4}) dz,$$

where $\sigma = surface$ tension

 ρ = fluid density

The volume constraint is expressed by:

$$V = 2\pi \int_{0}^{k} \int_{r_{0}} r \cdot dr \cdot dz$$

$$= \pi \int_{0}^{k} (R^{2} - r_{0}^{2}) \cdot dz = \text{constant}$$
(4)

Using variational methods for finding the minimum energy state, and including Eq. (4), requires that Eq. (1) be restated as:

$$\delta \mathbf{U} = \delta (\mathbf{E} - \mathbf{T} + \overline{\lambda} \mathbf{V}) = \mathbf{0}$$

where λ is the Lagrange multiplier and δ is the variational operator. Substituting Eq. (2), (3) and (4) into Eq. (5) and reversing the order between variation and integration yields:

$$\delta U = \int_{0}^{\ell} \delta \left\{ 2\pi \sigma R \left[1 + (dR/dZ)^{2} \right]^{1/2} - \frac{1}{4} \pi \omega^{2} \rho \left(R^{4} - r_{0}^{4} \right) + \bar{\lambda} \pi \left(R^{2} - r_{0}^{2} \right) \right\} dz = 0$$
(6)

The general condition of variational analysis for the minimum of functional is:

$$\delta U = \int_{0}^{\chi} \delta L dz = 0 \tag{7}$$

where L is the Lagrangian.

So, the Lagrangian in the case under consideration will be equal to the expression in the brackets in Eq. (6). The condition for $\delta U = 0$ is that the Lagrangian satisfies Euler's equation.

$$(d/dz) (\partial L/\partial R') - \partial L/\partial R = 0$$
(8)

where R' = dR/dz

Substituting the Lagrangian into Eq. (8) produces a differential equation for the film surface:

$$2\pi\sigma\left\{\left[1+(R')^{2}\right]^{1/2}-\frac{d}{dz}\left[\frac{RR'}{\sqrt{1+(R')^{2}}}\right]\right\}-\pi\omega^{2}\rho R^{3}+2\pi\bar{\lambda}R=0$$
(9)

The trivial solution is $R = \overline{R}$ where \overline{R} is the average film thickness. Substitution into Eq. (9) yields:

$$2\pi\sigma - \pi\rho\omega^2 \overline{R}^3 + 2\pi\overline{\lambda}\overline{R} = 0$$
(10)

or

$$\overline{\overline{\lambda}} = (\rho \omega^2 \overline{R}^3 - 2\sigma)/2\overline{R}$$
(11)

Page 48 Report One Members of The Institute of Paper Chemistry Project 2696-67

After a partial linearization,

$$(R')^2 << 1,$$
 (12)

which assumes that the amplitude of variations in the surface film are small compared to the wavelength of the disturbances, and accounting for Eq. (11) yields:

 $2\pi\sigma(R \cdot R'' - 1) + \pi\omega^2 \rho R^3 - 2\pi[(\omega^2 \rho \overline{R}^3 - 2\sigma)/2\overline{R}] R = 0$ (13)

Assuming that the surface can be described with a periodic function,

$$R = \overline{R} + \varepsilon \cos kx \tag{14}$$

where k is the wave number, and assuming that also

$$\varepsilon \ll \overline{R}$$
, (15)

such that ϵ^2 terms can be neglected, Eq. (13) reduces to:

$$-k^2 \sin kx + (1/\overline{R}^2 + \sigma \omega^2 R/\sigma) \sin kx = 0$$
 (16)

or the wave number k is:

$$k = \frac{1}{R} \sqrt{1 + \rho \omega^2 R^3 / \sigma}$$
 (17)

and the corresponding wavelength:

$$\lambda = \frac{2\pi}{k} = \frac{2\pi \overline{R}}{\sqrt{1 + \rho \omega^2 \overline{R}^3 / \sigma}}$$
(18)

This is identical to the relationship found by Yih and Kingman $(\underline{1,3})$, but it was arrived at from a completely different viewpoint. It is physically more informative, since it describes the mechanisms of the instability as being associated with a competition between the centrifugal and surface tension forces under conditions of constant liquid volume.

APPENDIX II

FORMULAS FOR ADHESIVES

A. "Carrier" adhesive used in experiments (chapter 6)

 $T = 73^{\circ}F$

Mix the solution several min after each step below.

Heat water Add starch Add sodium Add sodium	to 135°F borate hydroxide solution	10.2 liters 1 kg 13.6 g 157 g
Mix 20 min Add 100°F v	vater	15.5 liters
Add sodium Add starch	borate	86.2 g 5.8 kg
Add sodium	borate	68 g

B. "No-carrier" (controlled swelling) used in experiments (chapter 7)

Caustic dilution tank (130°F)

Water - 2.3 liters Liquid caustic (50%) - 0.12 liter

Mixing tank

Water - 8.86 liters Starch - 2.85 kg Boric acid - 54 g

After the starch is mixed thoroughly, the dilute caustic soda is metered into the starch slurry at a slow rate to prevent gelatinization of the starch. When the desired viscosity has been reached, the boric acid is added to stop the reaction and to stabilize the viscosity. Yield stress $\simeq 100 \text{ d/cm}^2$.

C. CARRIER TYPE ADHESIVE (TWO TANK) Used in Experiments (Chapter 7)

"Carrier" type adhesive used for experiments

Carrier-cooked starch - upper mixer:

```
Water - 2.88 liters (110°F)

Starch - 728 g

Caustic soda (50%) - 107 g

Water - 1.82 liters (cold)

Raw starch - lower mixer:

Water - 12.1 liters (90°F)

Borax decahydrate - 116 g

Starch - 3.63 kg
```

After the starch in the upper tank is mixed thoroughly, the caustic soda is added, the temperature is increased to $160^{\circ}F$ (71°C), and the mixture is agitated for 15 min. The cold water is added to reduce the temperature of the cooked starch to about 130°F (54°C). The amount of starch in the upper tank will determine the viscosity of the finished adhesive.

The contents of the upper tank are metered at a slow rate into the lower tank after the starch and borax decahydrate in the lower tank have been thoroughly dispersed. The mixture of the carrier and the raw starch slurry is agitated until a uniform viscosity is reached. Yield stress $\simeq 600 \text{ d/cm}^2$.

