

**Figure 5**—Average serum concentrations of all-*trans*-retinoic acid following a 5.0-mg dose of a microfine suspension to 11 rats.

peak concentration thus producing the slowly declining serum concentrations. Another possible cause of the slow release of the chemical from the microcapsules was interaction with the rat food in which the microcapsules were tableted. In a preliminary study, it was found that all-*trans*-retinoic acid crystals tableted in rat food were only 73% bioavailable as the solution, possibly indicating that the rat food in which the crystals were tableted was interfering with absorption.

The microfine suspension was formulated as a possible alternative to microencapsulation. It has the advantage of a greater bioavailability, but the disadvantages of being a less stable formulation and less readily

miscible with the rat food as compared to the solid microcapsules. The primary purpose of microencapsulation was to stabilize the retinoids for feeding to rats in their laboratory diets for long-term testing of the compounds for chemo-prevention of tumors.

All-*trans*-retinoic acid was successfully microencapsulated and the bioavailability of the encapsulated and liquid dosage forms compared. Although the microcapsules were only 34% bioavailable as the solution dosage form, this is a good compromise because of the increased stability and ease of handling. Also, all-*trans*-retinoic acid was found to follow dose-dependent kinetics at the doses studied; whereas, 13-*cis*-retinoic acid did not follow dose-dependent kinetics when given at the same dosage levels as the all-*trans*-retinoic acid.

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## Tack Behavior of Coating Solutions I

S. K. CHOPRA \* and R. TAWASHI \*

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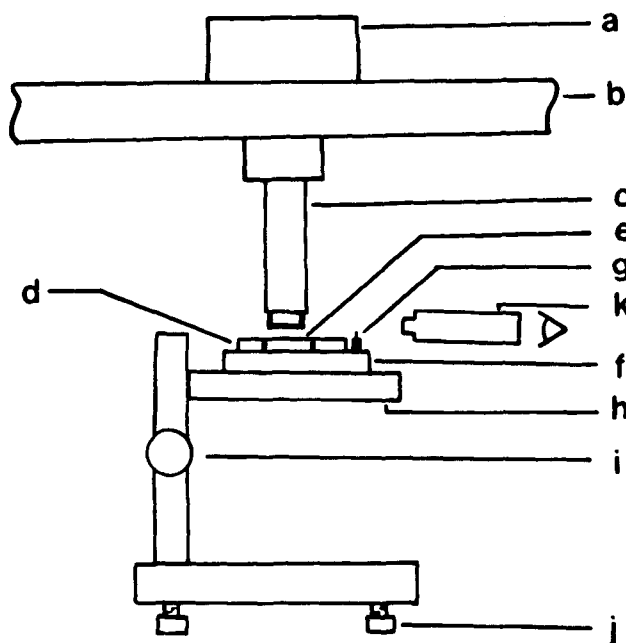
**Abstract** □ The tackiness of various tablet coating solutions was determined using a parallel plate technique with a tensile testing machine in conjunction with an oscilloscope where the separation force was displayed as a function of time. Measurements were made at various rates of separation on liquid films of constant thickness. Results showed that the force required to split a liquid film increases with an increase in rate of separation, and that tackiness increases with an increase in viscosity.

The relation between tack and viscosity was not linear, and a modified Stefan equation was proposed.

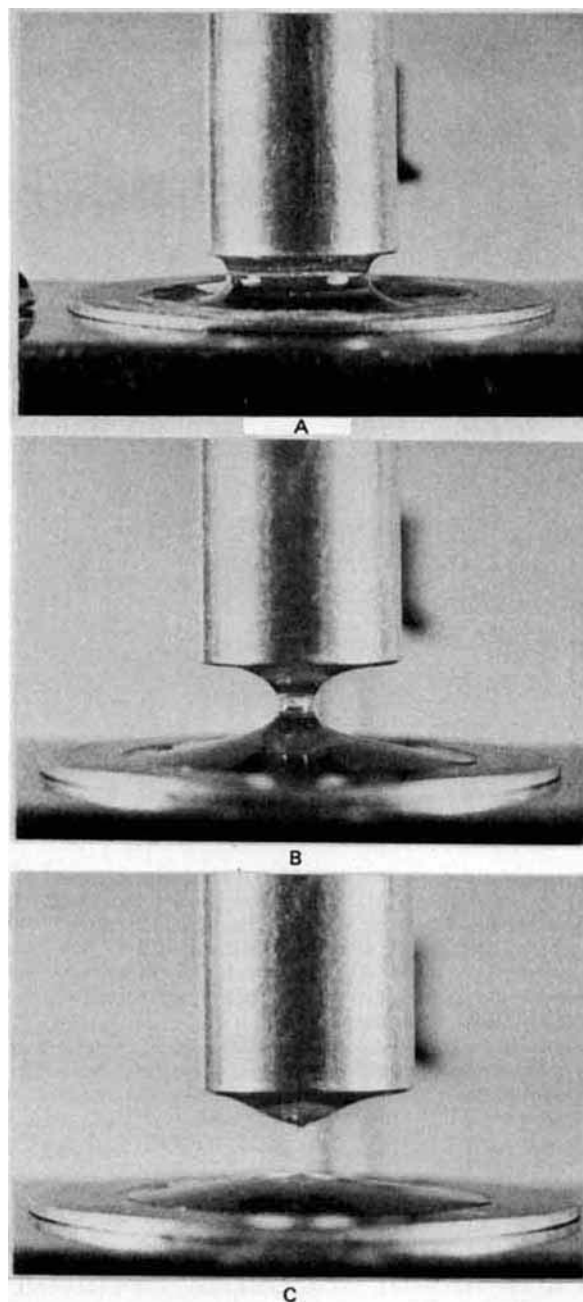
**Keyphrases** □ Tablet coating solutions—tack behavior, viscosity □ Viscosity—tack behavior of tablet coating solutions □ Tackiness—tack behavior of tablet coating solutions

The effect of coating formulations and coating process variables on the appearance of a coated tablet has been the subject of various investigations (1–3). Much effort has been devoted to the study of the theory of adhesion of film-forming materials to the surface of a tablet (4–6).

However, very little is known about the tackiness of coating solutions and its effect on the coating process. Studies on the evaluation of coating formulations have made some reference to the problem of tackiness of hydroxypropyl cellulose solutions during the coating process (7). However,



**Figure 1**—Schematic diagram of tack measuring assembly in conjunction with a tensile tester. Key: a, load cell 500- or 5000-g capacity; b, crosshead bar; c, stainless steel probe,  $r = 0.5642$ ; d, stainless steel ring; e, circular space; f, stainless steel plate; g, pin; h, vertically moving stage; i, knob for moving stage; j, leveling screws; k, telemicroscope.



**Figure 2**—Sequence of liquid film separation. Key: A, column formation; B, necking-down; C, final separation.

to our knowledge, no systematic, quantitative study on the tackiness of tablet coating solutions has been reported.

The objective of this study was to assess the tackiness of some polymer solutions used in tablet coating. Knowledge about the tacky behavior of coating formulations is a prerequisite for solving some of the technical problems encountered in tablet coating.

### BACKGROUND

Tack is defined as the impulse per unit area ( $ft$ ) necessary to separate two planes, initially in contact, through an intervening liquid (8). The basic expression for the separation of two flat circular plates in a viscous Newtonian liquid was given by Stefan (9) as follows:

$$F = \frac{3\pi\eta r^4}{4t} \left( \frac{1}{h_1^2} - \frac{1}{h_2^2} \right) \quad (\text{Eq. 1})$$

Where  $F$  is the force (dynes) of separation,  $\eta$  is the viscosity (poise) of the liquid,  $r$  is the radius (centimeters) of the plates,  $h_1$  and  $h_2$  are the initial and final thicknesses (centimeters) of the liquid film, and  $t$  is the time (seconds) required for separation. The force of separation per square centimeter ( $f$ ) is equal to  $F/\pi r^2$ , where  $\pi r^2$  is the surface area of the plates. Therefore, tack can be expressed as:

$$ft = \frac{3\eta r^2}{4} \left( \frac{1}{h_1^2} - \frac{1}{h_2^2} \right) \quad (\text{Eq. 2})$$

If the final thickness  $h_2 \gg h_1$ , then:

$$ft = \frac{3\eta r^2}{4h_1^2} \quad (\text{Eq. 3})$$

The validity of the relation described by Eq. 3 has been questioned by various investigators because the experimental confirmation of the proposed equality was not satisfactory. Some of the experiments on tack appear to be inadequately designed due to the lack of suitable instrumentation. The deviation of other experimental results from Eq. 3 has been attributed to a variety of factors such as the effect of high rates of separation (10), onset of cavitation and formation of multiple filaments (11, 12), flow properties of highly viscous materials (13), surface roughness, and wettability of the solid surfaces (14).

Based on these considerations, studies were designed to evaluate and quantitate the various physicochemical parameters influencing the tackiness of tablet coating solutions.

### EXPERIMENTAL

**Materials**—Polymers used in this study were hydroxypropyl methylcellulose<sup>1</sup> (5 cps), hydroxypropyl methylcellulose (15 cps), hydroxypropyl methylcellulose<sup>1</sup> (50 cps), hydroxypropyl cellulose<sup>2</sup>, and povidone<sup>3</sup>. Solvents used for making coating solutions were distilled water, ethanol<sup>4</sup> (95%), and methylene chloride<sup>5</sup>. Newtonian liquids used were glycerin<sup>6</sup> (97%) and white mineral oil<sup>7</sup>.

**Measurement of Viscosity**—All measurements were made on a rotational cup and bob viscometer<sup>8</sup> in conjunction with a strip chart recorder<sup>9</sup>. Temperature was held constant at 25° for aqueous test solutions

<sup>1</sup> Methocel E Premium (USP), The Dow Chemical Co., Midland, MI 48640.

<sup>2</sup> Klucel LF, Hercules Inc., Wilmington, DE 19899.

<sup>3</sup> Polyvinylpyrrolidone, plasdone K 29-32, GAF Corp., Linden, NJ.

<sup>4</sup> Les Alcools de Commerce Limitée, Ville St. Laurent, Quebec, Canada H4T 1N3.

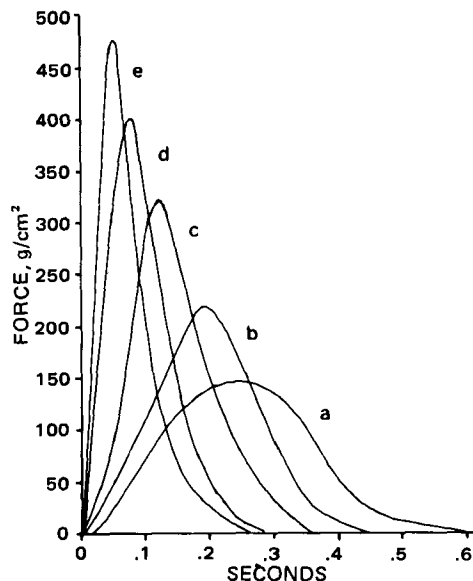
<sup>5</sup> The Dow Chemical Co., Midland, MI 48640.

<sup>6</sup> Emery Industries Ltd., Toronto, Ontario, Canada.

<sup>7</sup> Bate Chemicals, Roydon Road, Montreal, Quebec, Canada.

<sup>8</sup> Rotovisco, Brinkmann Instruments, Inc., Westbury, NY 11590.

<sup>9</sup> Honeywell Elektronik 194 Recorder, Honeywell Ltd., 6277 St. Jacques W., Montreal, Quebec, Canada.



**Figure 3**—Force-time oscillograms at various rates of separation for 10% (w/w) aqueous solution of hydroxypropyl methylcellulose (5 cps). Key: a,  $8.33 \times 10^{-3}$  cm/sec; b,  $1.66 \times 10^{-2}$  cm/sec; c,  $3.33 \times 10^{-2}$  cm/sec; d,  $8.33 \times 10^{-2}$  cm/sec; e,  $1.66 \times 10^{-1}$  cm/sec.

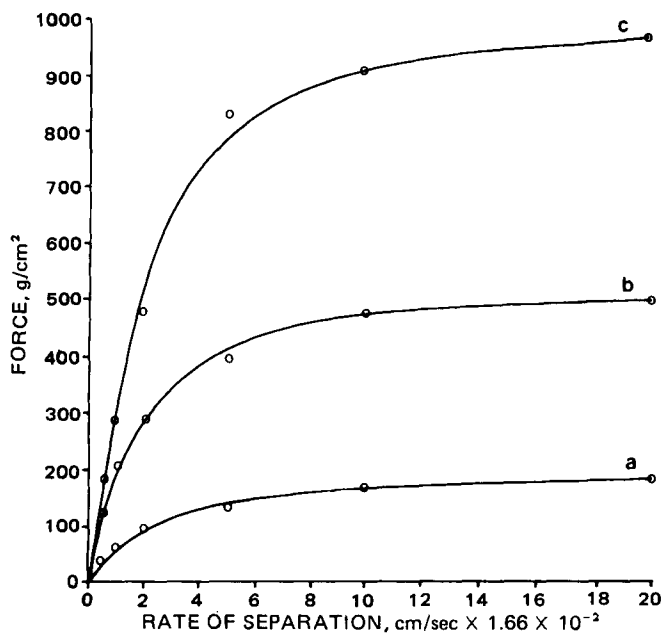
and at 15° for polymer solutions in organic solvents. The cup and bob geometry was varied according to the viscosity of the sample.

**Tack Measurement Assembly**—Tack measurements were made using a tensile testing machine<sup>10</sup>. A schematic diagram of the assembly used is shown in Fig. 1.

The probe (Fig. 1c) and plate (Fig. 1f) are of stainless steel, type 302. The end of the probe was machined and lapped. The working area of the probe and plate were polished using techniques similar to that described previously (14).

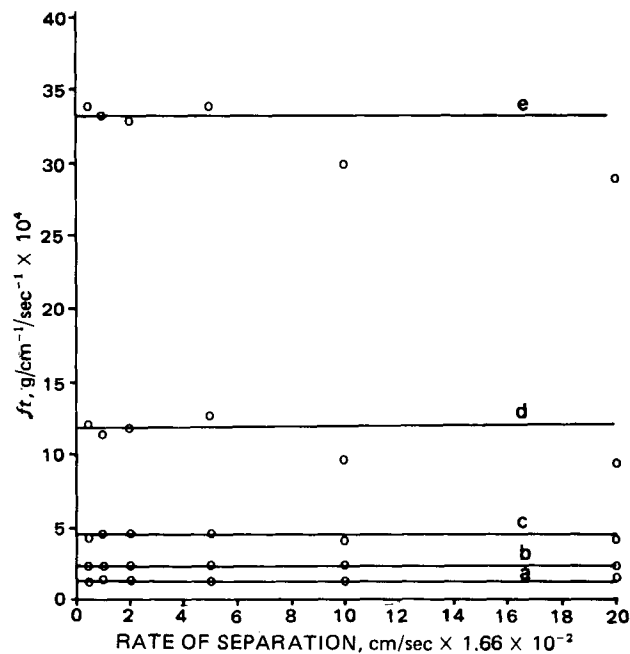
The horizontal position of the bottom plate (Fig. 1f) was checked using a spirit level. Prior to the measurement of tack, it was ascertained that the probe was perfectly parallel to the plate.

**Measurement of Film Thickness**—The bottom plate (Fig. 1f) was brought into contact with the probe by raising the vertically moving stage



**Figure 4**—Dependence of peak force on rates of separation for aqueous solutions of hydroxypropyl methylcellulose (5 cps). Key: a, 5% (w/w); b, 10% (w/w); c, 15% (w/w).

<sup>10</sup> Instron Tensile Tester model 1130, Instron, Canada, Burlington, Ontario, Canada.



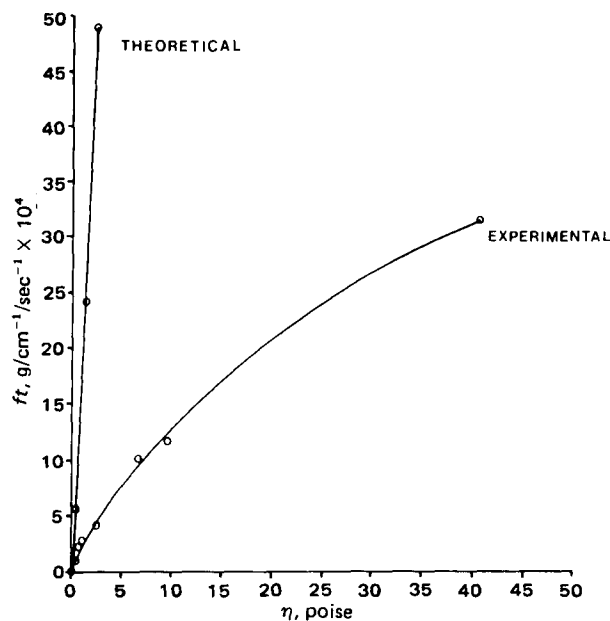
**Figure 5**—Dependence of tack on rates of separation for aqueous solutions of hydroxypropyl methylcellulose (5 cps). Key: a, 5% (w/w),  $\eta = 0.29$  poise; b, 7.5% (w/w),  $\eta = 0.8$  poise; c, 10% (w/w),  $\eta = 2.44$  poise; d, 15% (w/w),  $\eta = 9.5$  poise; e, 20% (w/w),  $\eta = 41$  poise.

(Fig. 1h). The pointed end of the pin (Fig. 1g) was then aligned with the crosshair line of the ocular micrometer attached to the telemicroscope<sup>11</sup> to mark the zero clearance between the probe and the plate.

The bottom plate was then lowered and the circular space (Fig. 1e) within the ring (Fig. 1d) was filled with ~0.5 ml of the test liquid. Utmost care was taken to avoid the introduction of air into the liquid during this transfer.

The bottom plate was then brought closer to the probe leaving a desired gap and any excess liquid was squeezed out. The difference in number of divisions between the new position and the zero position of the pointed end of the pin was used to calculate the film thickness of the liquid.

The resistance to necking-down takes place under the probe and not around it; therefore, the size of the reservoir is not of serious importance. The amount of the liquid should not be so small that the reservoir is



**Figure 6**—Experimental and theoretical tack versus viscosity of aqueous solutions of hydroxypropyl methylcellulose (5 cps).

<sup>11</sup> Telemicroscope, model M101T, Gaertner Scientific Corp., Chicago, Ill.

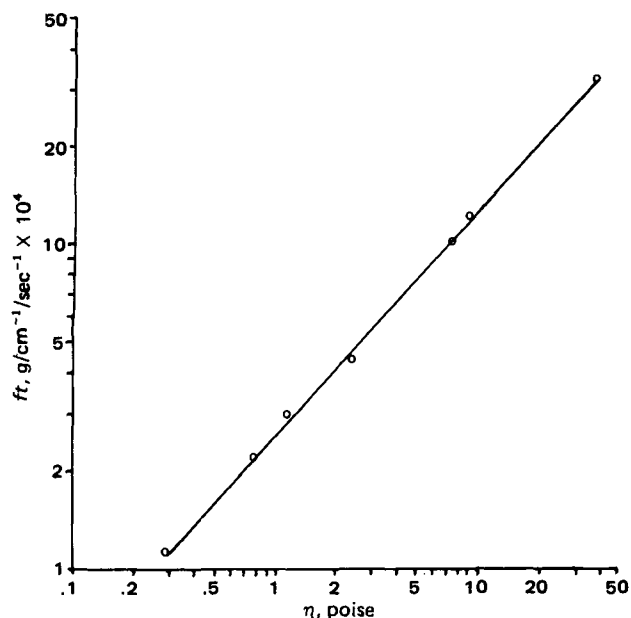


Figure 7—Logarithmic relationship (tack versus viscosity) of experimental data shown in Fig. 6.

completely used up at the very start of the necking-down process. When the volume of the test liquid is properly adjusted, a variation in volume of as much as 50% does not affect the tack measurement.

**Measurement of Tack**—After adjustment of the thickness of the test liquid, the crosshead bar (Fig. 1b) was allowed to move upwards at a specified speed. The test liquid first formed a column, necked-down, and then finally separated. The separation process was photographed and is shown in Fig. 2.

The events were recorded on a storage type oscilloscope<sup>12</sup> where the separation force ( $F$ ) was displayed as a function of time. The measurements were made at 25° for aqueous test solutions and at 15° for polymer solutions prepared in organic solvents.

A minimum of six measurements were taken for every test solution at each separation rate. The test solution was changed if the presence of air bubbles was noticed.

## RESULTS AND DISCUSSION

**Effect of Rate of Separation on Force of Splitting Liquid Film**—Figure 3 shows the force-time oscillograms obtained for a 10% aqueous solution of hydroxypropyl methylcellulose (5 cps) at various rates of separation. The traces show that as the time of separation decreases, the peak force increases.

The relation between peak force and the rate of separation for 5, 10, and 15% (w/w) aqueous solutions of hydroxypropyl methylcellulose (5 cps) is shown in Fig. 4. These plots demonstrate that the force required to split a liquid film increases with an increase in the rate of separation. The plots obtained are not linear and results are consistent with the experimental data presented in the study of tackiness of inks by Strasburger (15). Nevertheless, that author tried to fit the experimental data points to a theoretical straight line based on a modified Stefan equation derived previously (12):

$$F = \frac{3}{2} \pi \eta u \frac{r^4}{h_2^3} \quad (\text{Eq. 4})$$

where  $u$  is the rate of separation.

**Effect of Rate of Separation on Tack**—The plots of rate of separation versus tack of aqueous solutions of hydroxypropyl methylcellulose (5 cps) for concentrations ranging from 5 to 20% are shown in Fig. 5. The tack was calculated by dividing the area under the curves of the force-time oscillograms by the surface area of the probe.

Figure 5 illustrates that the tack is independent of the rate of separation for 5, 7.5, and 10% (w/w) aqueous solutions of hydroxypropyl methylcellulose (5 cps) solutions. These results are in agreement with Eq. 3. The tack values of 15 and 20% (w/w) solutions for rates of separa-

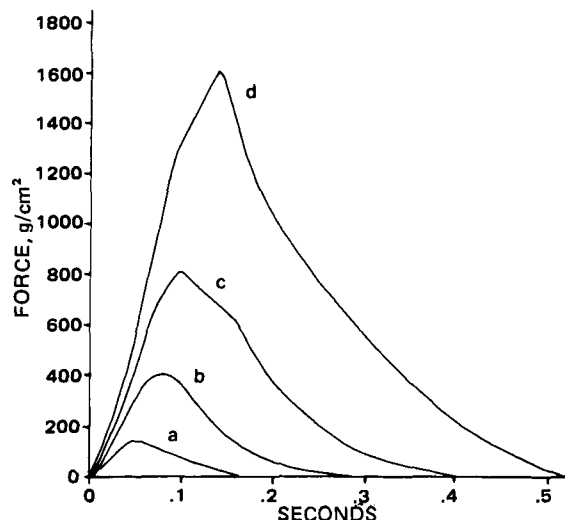


Figure 8—Force-time oscillograms at rate of separation of  $8.33 \times 10^{-2}$  cm/sec for aqueous solutions of hydroxypropyl methylcellulose (5 cps). Key: a,  $\eta = 0.29$  poise; b,  $\eta = 2.44$  poise; c,  $\eta = 9.5$  poise; d,  $\eta = 41$  poise.

tion,  $\geq 1.677 \times 10^{-1}$  cm/sec, fell below the average tack values obtained at lower rates of separation.

A similar behavior was also noticed for pure liquids such as glycerin and liquid paraffin, where the tack values at the rate of separation of  $3.33 \times 10^{-1}$  cm/sec are lower than those obtained at a lower rate of separation (Table I). Thus, for a liquid of given viscosity, there is an upper limit of the rate of separation beyond which the equation described by Stefan is not applicable.

The reason for obtaining low tack values at higher rates of separation may be due to viscoelastic effects (9, 16). When the probe is pulled away from the plate, the liquid flows towards the axis of the plate at the rate governed by  $h_1$ ,  $t$ , and  $\eta$ . When stress is applied slowly, i.e., when  $t$  is long, the liquid responds by flow. If  $t$  is too short, the mechanism of rupture of the liquid film is quite different from that envisaged by Stefan. The liquid gets no chance to flow and the elastic forces become more important for the breakdown of the film. Under such circumstances, the tack values obtained are lower than that predicted by Eq. 3.

**Effect of Viscosity on Tack**—The relation between tack and viscosity is shown in Fig. 6. The theoretical  $ft$  was calculated by substituting the values of  $r$ ,  $h$ , and  $\eta$  in Eq. 3. The experimental curve was plotted using the average of tack values obtained at various rates of separation for a number of solutions of hydroxypropyl methylcellulose (5 cps) in varying concentrations and hence viscosities.

Figure 6 shows that the experimental curve falls below the theoretical curve and also deviates from linearity as the viscosity of the test solution increases. The same experimental data plotted on a log-log scale is shown in Fig. 7 with a correlation coefficient of 0.9994, indicating the validity of the relationship  $ft = k \eta^c$ ;  $k$  and  $c$  are constants with respective values of 24,862 and 0.6737. The value of  $k$  depends on the instrumental factor,  $k'$  and  $3r^2/4h_1^2$ . The value of  $k'$  was found to be  $1.215 \times 10^{-1}$ . The equation for the experimental conditions described then becomes:

$$ft = \frac{3}{4} r^2 k' \eta^{0.67} \frac{1}{h_1^2} \quad (\text{Eq. 5})$$

To confirm the relationship described in Eq. 5, aqueous solutions of three different molecular weight grades of hydroxypropyl methylcellu-

Table I—Tack Values of Glycerin and Liquid Paraffin at Various Rates of Separation<sup>a</sup>

Rate of Separation, cm/sec	$ft$ (g/cm <sup>-1</sup> /sec <sup>-1</sup> )	
	Liquid Paraffin ( $\eta = 1.20$ poise)	Glycerin, 97% ( $\eta = 5.17$ poise)
$8.33 \times 10^{-3}$	$2.99 \times 10^4$	$7.57 \times 10^4$
$1.66 \times 10^{-2}$	$2.83 \times 10^4$	$7.32 \times 10^4$
$3.33 \times 10^{-2}$	$2.81 \times 10^4$	$7.44 \times 10^4$
$8.33 \times 10^{-2}$	$2.99 \times 10^4$	$7.47 \times 10^4$
$1.66 \times 10^{-1}$	$3.03 \times 10^4$	$6.38 \times 10^4$
$3.33 \times 10^{-1}$	$2.45 \times 10^4$	$5.98 \times 10^4$

<sup>a</sup>  $h_1 = 0.00108$  cm.

<sup>12</sup> Model 5113 with 5B12N time base and two 5A26 dual differential amplifiers, Tektronix, Inc., Beaverton, Ore.

**Table II—Predicted and Experimental Tack Values of Various Polymer Solutions**

Polymer	Concentrations, % (w/w)		Solvent	Viscosity, poise	$ft$ ( $g/cm^{-1}/sec^{-1} \times 10^4$ )	
					Predicted	Experimental
Hydroxypropyl methylcellulose (15 cps)	5	Water		1.26	2.90	2.91
	8	Water		8.4	10.34	10.9
Hydroxypropyl methylcellulose (50 cps)	3	Water		1.19	2.80	2.74
	5	Water		7.45	9.55	9.70
Hydroxypropyl cellulose	5	Water		0.85	2.23	2.01
	10	Water		9.54	11.27	11.4
Povidone	30	Water		.93	2.37	2.44
	43.5	Water		5.97	8.23	7.95
Hydroxypropyl methylcellulose (5 cps)	5	Ethanol-Methylene Chloride (50:50, w/w)		0.41	1.37	1.41
Hydroxypropyl methylcellulose (5 cps)	5	Ethanol-Methylene Chloride (75:25, w/w)		0.95	2.4	2.51
Hydroxypropyl methylcellulose (5 cps)	5	Ethanol-Water (50:50, w/w)		0.68	1.92	2.01
Hydroxypropyl methylcellulose (5 cps)	5	Ethanol-Water (75:25, w/w)		0.84	2.21	2.13

lose, a single grade of hydroxypropyl cellulose and of povidone, all in varying concentrations, were prepared. Solutions of hydroxypropyl methylcellulose (5 cps) were also prepared in mixtures of varying proportions of ethanol and methylene chloride, and ethanol and water.

The viscosities and the corresponding tack values of the above solutions were determined and the results are presented in Table II. This table illustrates that the experimental tack values, within a margin of error of  $\pm 5\%$ , match the predicted tack values calculated using Eq. 5.

In the present study, the experimental tack values obtained for various coating solutions were lower than those predicted by Stefan's equation. Also, the relationship obtained between tack and viscosity was not linear. The low experimental tack values obtained in Fig. 6 could be attributed to the instrumental factors such as the surface roughness of the probe and plate. The nonlinearity of the relationship between the tackiness and viscosity cannot be attributed to the experimental conditions, as these were kept constant throughout the study.

One of the factors that can influence the values of tack is the initial lag in force. This is due to the finite time of acceleration of the crosshead bar before it attains a constant speed. The force-time oscillograms in Fig. 8 were obtained at the same rate of separation. The shape of the traces obtained for lower viscosity solutions (Fig. 8, Curves a and b) are different than those for higher viscosity solutions (Fig. 8, Curves c and d). This suggests that the mode of liquid film separation is different for the higher viscosity solutions. It has been suggested (9) that when the ratio  $\eta/t$  increases to a point where the flow pattern of the liquid towards the axis of the plate is not uniform, the resulting  $ft$  is a fraction of that calculated by Eq. 3. This may account for the nonlinearity between tack and viscosity.

**Tack Effects in Tablet Coating**—The fact that a force is required to separate two objects joined by a thin liquid film (Fig. 4) could explain the phenomenon of picking and sticking in the pan-coating process.

At any given moment during the process of pan coating, a thin film of coating solution is present between two tablets, or between a tablet and the wall of a coating pan. When the pan is allowed to rotate, the tablets are carried upwards by the combined effects of centrifugal and frictional forces until gravity overcomes the effects and the tablets cascade down. The force exerted by these moving tablets splits the liquid film before the complete evaporation of solvent occurs.

If the force required to split the liquid film is greater than the forces of adhesion between the polymer film and the tablets, then picking may occur. The picking here is defined as the transference of portions of polymer film from the weakly adhered parts of one tablet to another or to the wall of a coating pan. However, if the force exerted by the moving tablets is not sufficient to split the liquid film present between the wet tablets, then sticking may occur.

The fact that the resistance of the liquid film to splitting increases with the rate of separation may further explain why the picking is more pronounced in a larger pan than in a smaller one. In the larger coating pan, the tablets cascade down at higher velocity than in a small coating pan, and therefore, the stress exerted on the polymer film during the separation of wet tablets will be higher in the former than in the latter.

In the tablet coating process, coating solutions when applied are dilute solutions of high polymers. As the polymer solution dries during the coating process, the concentration of the polymer increases from  $\sim 5\%$  to nearly 100%. Consequently, the flow behavior of the polymer solution changes from Newtonian to non-Newtonian and finally to an elastic solid. The tacky behavior of these solutions during the coating process is a target for future investigation.

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