A Comparison of Theoretical and Experimental Results for the Internal Dissolution of Soluble Cylinders by Water in Laminar Flow

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A boundary layer solution applicable to a certain general type of mass transfer operations was developed by Baasel and Smith (*1*). When the solution was adapted to the systems of condensation of a vapor from noncondensing gases and absorption of a gas into a wetted wall column, the temperature and interfacial area could not be completely controlled. To overcome these difficulties, the rate of internal dissolution of a hollow cylinder of benzoic acid was experimentally measured and compared to the theoretically predicted values.

The ratios of the predicted rates to the observed rates fell, with the exception of two runs out of twenty nine, between 0.622 and 1.303. It was therefore concluded that the finite difference techniques used to obtain a mathematical solution were valid. However, it was observed that the assumption of unidirectional diffusion was possibly not valid. It was concluded that for systems in which the ratio of the total molar density of the fluid to the saturation concentration of the material being transferred was large, in the approximate range of 700 to 2,700, the boundary layer approach corresponded to the Leveque approximation of the classical Graetz solution for heat transfer for fluids with a parabolic velocity distribution.

In a previous paper Baasel and Smith (*1*) presented a solution to the problem of the condensation of a vapor from noncondensing gases in laminar flow. The solution uses the fundamental microscopic approach to predict the rate of mass transfer in a vertical cylindrical condenser. Experimental data from a methanol and air system showed good agreement with the results suggested by the mathematical model. Herein is presented the results of applying this same general solution to a different mass transfer system, namely, the internal dissolution of pipes cast from slightly soluble material.

In the condensation system used by Baasel to check his solution, the effect of temperature-induced natural convection currents was unknown. Also, when the rate of gas absorption by a wetted-wall column was compared by Courtney (*2*) with rates predicted using this boundary layer approach, the error introduced by a varying interfacial area, which results from rippling of the wetted-wall surface, was again unknown. It was felt that the system of the dissolution of a cylinder cast from benzoic acid by water flowing longitudinally through the system would eliminate these difficulties and very closely approximate the theoretical model from which the mathematical solution was derived.

The system of the dissolution of a solid cylinder proved to have one primary limitation. In order to maintain a reasonably constant surface area the solubility of the solid material must not be excessive. This in turn results in a small mass transfer rate which means the theoretical model approaches the more familiar classical methods. This will be discussed more fully in the section comparing the theoretical and experimental results.

In adapting the basic equations from which a mass transfer solution may be obtained for this system, that is benzoic acid diffusing into a laminar stream of water, the following assumptions were made:

1. The system operated at steady state conditions.
2. The temperature of the water was constant.
3. There was no diffusion of water into the cylinder of benzoic acid.
4. The flow pattern at the entrance of the benzoic acid section was fully developed laminar flow with a parabolic velocity distribution.
5. The flow pattern and concentration profile in the cylinder was symmetrical with respect to the rotational direction.
6. The concentration of benzoic acid in the water immediately adjacent to the inside wall of the cylinder was equal to the saturation concentration at the particular water temperature.
7. The diffusivity of the benzoic acid in water was a function of water temperature only.
8. The inside surface area of the cylinder remained constant.
9. There were no impurities in the water which would effect the diffusion process.
10. The concentration of the benzoic acid was nowhere large enough to effect the physical properties of the water.

The technique employed by Baasel does not assume any particular velocity profile in the area where mass transfer is occurring. The technique also does not neglect the term in the basic equation which is the product of the velocity perpendicular to the wall and the concentration gradient perpendicular to the wall. While the former velocity term is generally very small, the latter concentration gradient is usually large and their product consequently constitutes a significant term in the equation. This will be seen more clearly as the equations are developed.

The differential molar flux of component *A*, the benzoic acid, across the differential area *dA* can be expressed as:

$$\frac{dN_a}{dA} = -D_{ab}\frac{\partial C_a}{\partial r} + \left[\frac{dN_a}{dA} + \frac{dN_b}{dA}\right]\frac{C_a}{C_t} \quad (1)$$

By assumption 3,

$$dN_b = 0 \quad (2)$$

To obtain the amount of benzoic acid which dissolves and diffuses into the water, Equation (1) must be evaluated at the cylinder wall. Therefore, the differential area *dA* is replaced by $\pi D dz$ and C_a becomes C_{aw}, the molar concentration of benzoic acid at the wall. By rearranging Equation (1) and making the above substitutions gives:

$$dN_a = -\frac{\pi\, C_t\, D_{ab}\, D}{C_t - C_{aw}}\, \frac{\partial C_a}{\partial r}\, dz \tag{3}$$

Integration of Equation (3) over the length of the cylinder or pipe gives the rate of transfer. Namely

$$N_a = -\frac{\pi\, C_t\, D_{ab}\, D}{C_t - C_{aw}} \int_0^L \frac{\partial C_a}{\partial r_w}\, dz \tag{4}$$

Solution of Equation (4) depends upon expressing the unknown C_a as a function of z and r, the length and radius, respectively, of the cylinder.

The molar flux of benzoic acid can be expressed alternatively as

$$\frac{dN_a}{dA} = C_a v_a \tag{5}$$

Since the velocity of component B, the water, is zero, therefore

$$V_r C_t = v_a C_a \tag{6}$$

and

$$v_r = -\frac{D_{ab}}{C_t - C_a}\, \frac{\partial C_a}{\partial r} \tag{7}$$

The continuity equation, after simplication resulting from assumptions 1, 5, and 10, is written as follows:

$$\frac{\partial (r\, v_r)}{\partial r} + \frac{\partial (r\, v_z)}{\partial z} = 0 \tag{8}$$

Likewise, the component balance equation after appropriate simplication is

$$v_r \frac{\partial C_a}{\partial r} + v_z \frac{\partial C_a}{\partial z} = D_{ab} \left[\frac{\partial^2 C_a}{\partial r^2} + \frac{\partial^2 C_a}{\partial z^2} + \frac{1}{r} \frac{\partial C_a}{\partial r} \right] \tag{9}$$

Equations (7), (8), and (9) must satisfy the following boundary equations for $z = \infty$

$$v_z = v_{mf}\,(1 - r^2/r_w^2), \frac{\partial C_a}{\partial z} = 0, C_a = C_{as}, v_r = 0$$

for $r = r_w$,

$$v_z\, 0, \frac{\partial C_a}{\partial z} = 0, C_a = C_{as}$$

for $r = 0$

$$\frac{\partial v_z}{\partial r} = 0, \frac{\partial C_a}{\partial r} = 0, v_r = 0$$

By following the same steps given in the literature (*1*) the following is obtained:

$$-\left(\frac{\partial C}{\partial y}\right)^2 + \frac{(1 - s^2)\, r_w}{D_{ab}}\, v_{mf}\,(K - 1)\, \frac{\partial C}{\partial y} - (K - C)\, \frac{\partial^2 C}{\partial y^2} = (K - C)\, \frac{\partial^2 C}{\partial s^2} + \frac{K - C}{s}\, \frac{\partial C}{\partial s} + \left(\frac{\partial C}{\partial s}\right)^2 \tag{10}$$

Equation (10) is the desired relationship between the concentration of benzoic acid and the length and diameter of the cast cylinder. The method of finite difference approximation with the aid of an IBM 1620 computer was used to obtain the slope of the concentration gradient at the cylinder wall. This derivative, $\left(\frac{\partial C_a}{\partial r}\right)_w$, is the only unknown in Equation (4) and once it can be determined, the total number of moles of benzoic acid transferred follows. A plot of the concentration gradient, $\left(\frac{\partial C_a}{\partial r}\right)_w$, vs. the length of the cylinder (z) is now possible and the reader is referred to Baasel's paper (*1*) where the general form of this relationship is given.

COMPUTER SOLUTION

Equation (10) is an elliptic partial differential equation which is stable only if the right choice of difference representation is made. Even then the solution becomes unstable as the grid becomes smaller. This instability was first noticed in the digital computer solution and was later verified by using Karplus' (*3*) technique. The point where Karplus' technique predicted instability was essentially the place where the computer solution became unstable. For values of K less than 1.5 the number of intervals required for convergence to a reasonable answer was too large for our computer. However, when K was greater than 10 an approach to within 1% of the true value was possible. This accuracy was estimated by successively reducing the grid size in the stable region.

In the benzoic acid system, the used K always exceeded 300. For this system the length was divided into 40 increments and the width was divided into the largest number of increments which was divisible by forty and was stable. This turned out to be 120, 160, or 200 increments.

The finite difference approximations used were

$$\frac{\partial C}{\partial y} = \frac{Cy + 1, s - Cy, s}{\Delta y}$$

$$\frac{\partial^2 C}{\partial y^2} = \frac{Cy + 1, s - 2\, Cy, s + Cy - 1, s}{(\Delta y)^2}$$

$$\frac{\partial C}{\partial s} = \frac{4\, Cy, s + 1 - 3\, Cy, s - Cy, s + 2}{2 \Delta s}$$

$$\frac{\partial^2 C}{\partial s^2} = \frac{Cy, s + 1 - 2\, Cy, s + 1}{(\Delta s)^2}$$

This computer program written in Fortran for the experimental input is available from the authors.

EXPERIMENTAL PROCEDURE

An experimental procedure was now worked out to test the transfer rates predicted by the mathematical model.

To obtain cylinders of benzoic acid, the acid was first melted to a liquid state. A section of phenolic plastic pipe 1½ in. in diameter and approximately 6 in. long served as a mold. This mold was placed in a steel base section which supported the mold in a vertical position. A steel core 1 in. in diameter and 6 in. long was precisely centered and supported in the mold by the base section. The liquid benzoic acid was then poured into the mold and allowed to cool for several hours. The base section was then easily removed. The steel core section was provided with a ⅜ in. diameter longitudinal hole into which an electrical heating rod could be inserted. Suspension of the cylinder in midair and rapid heating of electrical rod resulted in the steel core sliding out of the benzoic acid under its own weight. This produced a 6 in. section of plastic pipe with a very smooth and continuous ¼ in. thick liner of benzoic acid.

After allowing the cylinder to cool to room temperature, flanges which had been machined from plexiglass were cemented to each end of the cylinder. A test cylinder and a control cylinder were used for each experimental run. Both cylinders were weighed with an analytic balance and the length and diameter of the test cylinder were accurately measured. The test cylinder was then placed as a section in a 1 in. copper line through which water was to be pumped.

The 1 in. copper line consisted chiefly of a 13 ft. calming section which is sufficient to insure complete development of laminar flow (*4*). At one end of the vertically supported calming section was a copper flange with a groove machined in it. A rubber O-ring was fitted into this groove. The test cylinder could then be bolted vertically to the end of the calming section with the O-ring giving a water tight seal. A shorter

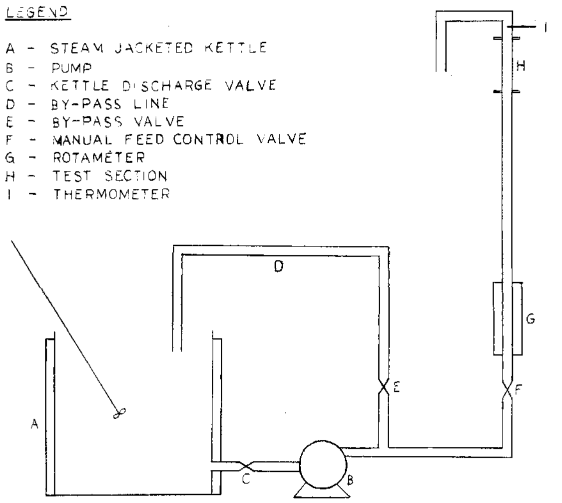

Fig. 1.

section of copper pipe including a hose connection and copper flange with O-ring assembly was bolted to the top or downstream end of the test cylinder.

A 100 gal. steam jacketed kettle supplied preheated water which could be maintained at a constant temperature throughout a run. A small centrifugal pump discharged water from the kettle through a calibrated flowmeter which was just upstream of the calming section. A by-pass line from the pump discharge back into the kettle enabled a constant flow rate to be maintained during a test run. A schematic outline of the test setup is shown in Figure 1.

At the conclusion of a test run which lasted from one to three hours the test cylinder and the control cylinder were placed in an oven at 125 to 130°F. This drying period lasted about eight hours. The cylinders were then placed in a calcium sulfate desiccator and periodically weighed until their respective weight changes were approximately equal. The percent weight change of the control cylinder was then used to correct the weight loss of the test cylinder. This corrected weight loss was then the number of moles of benzoic acid transferred to the water over a measured length of time.

COMPARISON OF THEORETICAL AND EXPERIMENTAL RESULTS

The experimental results showed a mass transfer rate ranging from 1.023×10^{-4} to 9.646×10^{-4} g./sec. The corresponding theoretical results ranged from 0.753×10^{-4} to 6.144×10^{-4} g./sec. The Reynolds number was varied from 628 to 2,374 and the Schmidt number from 305 to 1,619. The water temperature included values from 66 to 131°F.

Table 1 presents the results in terms of the Reynolds number, the Schmidt number, and the concentration ratio K. The modified mass transfer Peclet number is readily calculated from these data. However, the mass transfer is a function of both the Peclet number and K, so by presenting the results in terms of the Peclet number alone is of little value. At constant values of K the mass transfer is proportional to the Peclet number and at constant Peclet numbers the mass transfer is inversely proportional to K.

As a method of comparing the theoretical and experimental results, a ratio of the predicted or theoretically calculated mass transfer rate divided by the experimental rate was determined for each experimental run. In a total of 29 runs this mass transfer ratio varied from 1.49 to 0.588. The mean value of the mass transfer ratio was 0.889 with a variance of 0.065 and a standard deviation of 0.255. This mass transfer ratio can be considered as the fraction of the experimental rate which the mathematical solution predicted. Thus, with the mean value of the ratio, as a basis, the theoretical solution gave a mass transfer rate which was 88.9% of the experimental rate.

Table 1. Data and Results

	$\frac{Dv_{mf}\rho}{\mu}$	$\frac{\mu}{D_{AB}\rho}$	K	Exp. mass trans. rate $\times 10^4$ g./sec.	Calc. mass trans. rate $\times 10^4$ g./sec.
3	1857	352	848	5.724	4.577
4	1282	352	848	4.163	3.973
5	2138	310	730	5.980	5.690
6	2424	305	714	8.909	6.144
7	1545	310	729	5.747	5.043
8	1531	493	1170	2.010	2.987
9	1580	352	827	6.790	4.473
10	1872	347	817	5.668	4.869
11	1313	364	859	3.956	4.005
12	1540	352	848	4.614	4.313
13	1798	310	730	7.386	5.464
14	762	922	1938	1.213	1.239
15	628	1025	2189	1.023	1.039
16	899	1169	2261	1.061	1.046
17	1979	310	730	8.812	5.480
18	1274	347	817	4.928	4.072
19	1264	352	848	5.080	3.940
20	1264	352	848	4.468	3.907
21	2107	310	730	9.646	5.668
22	1535	353	848	5.294	4.250
23	1535	352	848	6.537	4.286
24	1501	310	730	7.450	4.990
25	1857	352	848	6.045	4.347
26	700	1618	2714	0.928	0.753
27	1568	483	1150	2.723	2.987
28	1725	466	1094	2.677	3.188
29	1793	466	1094	3.209	3.241
30	1432	503	1190	2.143	2.794
31	1272	350	848	4.358	3.907

An attempt was made to quantify the experimental errors that could be expected to occur in the system. The variables which were measured for each run were the temperature and flow rate of the water and the mass and physical dimensions of the test cylinder and the control cylinder. A numerical value for the maximum error which could reasonably be thought to occur was determined for each variable and the effect of the error on the results calculated. The maximum percent error in the mass transfer ratio caused by an error in one variable was approximately 8% while a maximum of 12% occurred for the unlikely but possible case of a combination of experimental errors. The value obtained for the saturation concentration of benzoic acid in water from the available data was accurate to only ± 0.05 g. acid/100 g. water. The results of a ± error of this magnitude was a maximum error in the mass transfer ratio of 16%. Generally speaking this type of error analysis showed that the maximum values of the errors in the ratio resulting from incorrect determinations of the system variables ranged from 10 to 16%.

One large departure of the experimental system from the theoretical model, in the benzoic acid-water system studied, was the existence of counterdiffusion of water into the cast cylinders. This is a direct contradiction of assumption 3. Tests were made with a cylinder which consisted of benzoic acid only, that is, the plastic mold was removed leaving just the benzoic acid shell. The exact experimental run conditions could not be duplicated with the pure cylinder. However, contacting this cylinder with water indicated that the mass rate of water absorption could possibly be as great as the mass rate of benzoic acid dissolution. Consequently, the molar flux of water could conceivably be seven times as great as the molar flux of the acid.

Reference to Equation (1) shows the effect which the violation of assumption 3 could have on the predicted results. If dN_b in Equation (1) was not zero, and yet subsequent solution of Equation (1) upon the premise it was zero gave approximately correct results, it must be concluded that the entire second term on the right side of Equation (1) is very small relative to the remaining terms. Examination of the ratio C_a/C_t show that this is possibly the case. The saturation concentration of benzoic acid is in the range of $2.5 - 75 \times 10^{-5}$ g. mole/cc. for the temperatures involved, therefore, the value of C_a would be less than this. The value of C_t, the total concentration, was approximately 0.05 g./cc. Thus, only a large value for dN_a, or dN_b would give the second term any significance. The low solubility of benzoic acid in water precludes a large dN_a and the maximum value of dN_b was estimated to be seven times dN_a.

As a further check of the magnitude of this term in the equation, the following substitution was made for dN_b:

$$dN_b = -7\, dN_a \tag{11}$$

When this factor was introduced into the equations and the calculations repeated, the predicted results showed practically no change from the initial results.

After establishing the relatively small magnitudes of dN_b and dN_a it can readily be seen that v_r, the radial velocity component, is likewise a small quantity, since

$$\frac{dN_a}{dA} + \frac{dN_b}{dA} = v_r C_t \tag{12}$$

The classical Graetz solution for heat transfer and adapted for mass transfer as shown by Pigford and Sherwood (*6*) assumes a zero velocity in the radial direction. This solution should therefore provide an accurate description of the physical process of a slightly soluble material diffusing into a laminar stream. Actually the Leveque equation [Linton and Sherwood (*5*)], which is an approximation to the Graetz equation, is appropriate when a material is diffusing only a short distance from the conduit wall. This equation assumes a linear velocity gradient in the fluid and is written

$$\frac{C - C_o}{C_s - C_o} = 5.5 \left(\frac{w}{D_a b\, L'} \right)^{-2/3} \tag{13}$$

The results given by the equation compare very well with the concentrations predicted by the original method. Table 2 shows the experimental cylinder exit concentrations, the original predicted exit concentrations, and the exit concentrations predicted by the Leveque equation. The good agreement of this work with the Leveque solution indicates the validity of the approximation techniques which were used in connection with the mathematical model.

TABLE 2. COMPARISON OF EXPERIMENTAL AND ANALYTICAL RESULTS

Run no.	Experimental exit concentration $\times 10^5$ g./cc.	Predicted exit concentration $\times 10^5$ g./cc.	Leveque exit concentration $\times 10^5$ g./cc.
3	2.613	2.146	2.107
4	2.794	2.778	2.726
5	2.527	2.468	2.422
6	3.301	2.315	2.281
7	3.361	3.078	3.017
8	0.931	1.415	1.385
9	3.644	2.461	2.441
10	2.588	2.284	2.246
11	2.514	2.650	2.598
12	2.476	2.390	2.352
13	3.605	2.763	2.708
14	0.856	0.887	0.878
15	0.803	0.833	0.822
16	0.573	0.559	0.577
17	4.023	2.570	2.529
18	3.307	2.856	2.803
19	3.409	2.755	2.702
20	2.998	2.731	2.677
21	4.075	2.460	2.412
22	2.841	2.352	2.306
23	3.508	2.374	2.327
24	4.357	3.044	2.983
25	2.760	2.080	2.054
26	0.546	0.430	0.450
27	1.261	1.386	1.383
28	1.108	1.369	1.376
29	1.285	1.322	1.335
30	1.067	1.405	1.395
31	3.045	2.740	2.688

CONCLUSIONS

After evaluating the experimental data it was concluded that, for Reynolds numbers in the general range of 600 to 2,000 for water flowing through a slightly soluble cylinder, the boundary layer approach to the solution of the microscopic mass transfer equations provided a reasonably accurate means of predicting the dissolution rate of the cylinder.

It was further concluded that for systems in which the ratio of the total molar density of the fluid to the saturation concentration of the material being transferred was large, in the approximate range 700 to 2,700, the boundary layer approach corresponded to the Leveque approximation of the classical Graetz solution for heat transfer for fluids with a parabolic velocity distribution.

NOTATION

A = area, sq. cm.
C = dimensionless concentration, C_a/C_{as}
C_a = concentration of species A, g. moles/cc.
C_{as} = saturation concentration of benzoic acid, g. moles/cc.
C_{aw} = molar concentration of A at the cylinder wall, g. moles/cc.
C_o = concentration of inlet stream, g./cc.
C_t = total concentration, g. moles/cc.
Cy, s = dimensionless concentration at a point in space y, s
$Cy + 1, s$ = dimensionless concentration at a point in space $y + \Delta y,\ s$
$Cy - 1, s$ = dimensionless concentration at a point in space $y - \Delta y,\ s$
$Cy, s + 1$ = dimensionless concentration at a point in space $y,\ s + \Delta s$
$Cy, s + 2$ = dimensionless concentration at a point in space $y, s + 2\Delta s$
$Cy, s - 1$ = dimensionless concentration at a point in space $y,\ s - \Delta s$
D = diameter, cm.
D_{ab} = diffusivity of species A in B, sq. cm./sec.
K = C_t/C_{as}
L = length of cylinder, cm.
L' = thickness of region in which molecular transfer is controlling, cm.
N_a = flow rate of A, g. moles/sec.
N_b = flow rate of B, g. moles/sec.
r = radial distance from center of cylinder, cm.
r_w = radius of cylinder, cm.
s = dimensionless radial coordinate, r/r_w
u = dimensionless radial velocity, v_r/v_{mf}
v = dimensionless longitudinal velocity, v_z/v_{mf}
v_a = molar velocity of A, cm./sec.
v_{mf} = maximum final velocity in the longitudinal direc-

tion, cm./sec.

v_r = molar average velocity perpendicular to the cylinder wall, cm./sec.

v_z = molar average velocity parallel to the cylinder wall, cm./sec.

w = mass flow rate, g./sec.

y = dimensionless longitudinal coordinate, z/r_w

z = length dimension, cm.

Δy = distance between consecutive points in the y direction in the finite difference grid

Δs = distance between consecutive points in the s direction in the finite difference grid

μ = viscosity, g.cm./sec.

ρ = density, g./cc.

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Methods for Solving the Boundary Layer Equations for Moving Continuous Flat Surfaces with Suction and Injection

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Several methods that can be used to obtain solutions to the laminar boundary layer momentum, energy, and diffusion differential equations for moving continuous flat surfaces with suction and injection are presented. Results are obtained for a wide range of the injection parameter, $f(0)$, at Prandtl and Schmidt numbers of 1, 10, and 100. Those methods which permit hand calculation of the properties of interest are compared using the numerical solutions of the boundary layer differential equations as the exact solutions. The new integral method of Hanson and Richardson which gives results for the momentum thickness that deviate less than 2.2% from the exact values is recommended for predicting values of the momentum boundary layer parameters. The Von Karman-Pohlhausen method, which was modified to account for suction and injection, is most generally valid. This method gives acceptable values of the transfer coefficients for heat, mass and momentum transfer for most of the values considered.

Several new and modified methods for solving differential equations describing the laminar boundary layer behavior on a moving continuous flat sheet with suction and injection are presented. Emphasis is placed on those methods which permit hand calculation of the properties of interest.

The boundary layer adjacent to continuous moving solid surfaces was first investigated by Sakiadis (1). In his pioneering paper the boundary layer differential equations for continuous moving flat and continuous moving cylindrical surfaces were presented. The differential equations describing boundary layer behavior on these continuous surfaces were shown to be the same as the equations describing the behavior of a boundary layer next to a geometrically similar object of finite length. However, the boundary conditions for these two different classes of problems are different. Sakiadis' paper also contained the development of the integral momentum equation for both flat and cylindrical continuous surfaces.

In a subsequent publication Sakiadis (1) presented an exact and an approximate solution for the case of laminar flow and flat geometry. An approximate solution thought to be valid for turbulent flow and flat geometry was also given. Later, Sakiadis (1) treated the case of a laminar and a turbulent boundary layer on a continuous circular cylinder by an integral method.

Koldenhof (2) working independently used an integral technique somewhat different from that used by Sakiadis to treat the problem of a laminar boundary layer on a continuous moving cylinder. The equations of Koldenhof are essentially the same as Sakiadis. Koldenhof also presented the results of an experimental investigation of drag on continuous cylinders and good agreement was obtained between the theoretical and the experimental results.

Griffith (3) presented experimental data for shear stress and velocity profiles in the boundary layer adjacent to continuous moving cylindrical surfaces. These data qualitatively agreed with the data of Koldenhof. Griffith also reasoned that for the boundary layer associated with polymer spinning, the boundary layer equation of energy could

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