Gillespie, B. M., and J. J. Carberry, "Influence of Mixing on Isothermal Reactor Yield and Adiabatic Reactor Conversion," *Ind. Eng. Chem. Fundamentals,* **5**, 164 (1966).
Koo, L. C. T., and E. N. Ziegler, "Design of Multistage Reactors for Nonlinear Kinetics," *Chem. Eng. Sci.,* **24**, 217 (1969).
LaRosa, P., and F. W. Manning, "Intensity of Segregation as a Measure of Incomplete Mixing," *Can. J. Chem. Eng.,* **42**, 65 (1964).
Leitman, R., "Effect of Mixing on Chemical Reaction in a Continuous Stirred Tank Reactor," Ph.D. dissertation, Polytechnic Institute of Brooklyn, N.Y. (1970).
———, and E. N. Ziegler, "Stirred Tank Reactor Studies: Part I. Mixing Parameters," *Chem. Eng.,* **2**, 252 (1971).
Manning, F. S., D. Wolf, and D. L. Keairns, "Model Simulation of Stirred Tank Reactors," *AIChE J.,* **11**, 723 (1965).
Marquadt, D. W., "An Algorithm for Least-Squares Estimation of Non-Linear Parameters," *J. Soc. Indust. Appl. Math.,* **11**, 431 (1963).
Nauman, E. B., "The Droplet Diffusion Model for Micromixing," *Chem. Eng. Sci.,* **30**, 1135 (1975).
Rippin, D. W. T., "Segregation in a Two-Environment Model of a Partially Mixed Chemical Reactor," *ibid.,* **22**, 247 (1967).
Spielman, L. A., and O. Levenspiel, "A Monte Carlo Treatment for Reacting and Coalescing Dispersed Phase Systems," *ibid.,* **20**, 247 (1965).
Villermaux, J., and A. Zoulalian, "Etat de mélange du fluide dans un réacteur continu. A proposd'un modéle Weinstein et Adler," *ibid.,* **24**, 1513 (1969).
Wan, C. G., and E. N. Ziegler, "Effect of Mixing on Yield in Isothermal Tubular Reactors," *Ind. Eng. Chem. Fundamctals,* **12**, 55 (1973).
Williams, J. A., R. J. Adler, and W. J. Zolner, III, "Parameter Estimation of Unsteady-State Distributed Models in the Laplace Domain," *Ind. Eng. Chem. Fundamentals,* **9**, 193 (1970).
Worrell, G. R., and L. C. Eagleton, "An Experimental Study of Mixing and Segregation in a Stirred Tank Reactor," *Can. J. Chem. Eng.,* **42**, 254 (1964).
Ziegler, E. N., S. P. Chand, D. R. Goodrich, and R. H. Leitman, "Mixing Models for the CSTR," Paper presented at AIChE Annual Meeting, Reprint 34C, San Francisco, Calif. (1971).
Zwietering, Th. N., "The Degree of Mixing in Continuous Flow Systems," *Chem. Eng. Sci.,* **11**, 1 (1959).

APPENDIX

For first-order reaction with nonideal residence time distribution

$$<Y_{e_{\text{seg}}}> = 4\left(\frac{R_2}{1 + 2R_2}\right)^2$$

which is less than $<Y_{e_{\text{mic}}}> = \dfrac{R_2}{1 + R_2}$

$$<Y_e> = \left[\frac{a R_1(1 + R_1 + 4R_2)}{(1 + R_1 + 2R_2)^2} + (1 - a)\right] Y_{1c} + \frac{4\, a\, R_2^2}{(1 + R_1 + 2R_2)^2}$$

where

$$Y_{1c} = \frac{R_2\,[(1 + R_1 + 2R_2)^2 - a(1 + R_1 + 4R_2 + 4R_2^2)]}{[(1 + R_2)(1 + R_1 + 2R_2)^2 - a(1 + 8R_2^2 + R_1 + 2R_1R_2 + 5R_2 + 4R_2^3)]}$$

Weinstein, H., and R. J. Adler, "Micromixing Effects in Continuous Chemical Reactors," *Chem. Eng. Sci.,* **22**, 65 (1967).

Manuscript received June 28, 1976; revision received January 3, and accepted January 5, 1977.

Nonisothermal Nip Flow in Calendering Operations

An efficient and simple numerical technique is presented for analyzing nonisothermal nip flow of viscous liquids. It has been applied on calendering to calculate the design parameters as well as the interaction effects between roller characteristics, operation conditions, and material properties. Viscous heating is shown to drastically change the mechanics near the nip exit if the rollers rotate at different speeds. Consequences for scaling-up and model experiments are indicated.

FRANS DOBBELS
and
JAN MEWIS
Department of Chemical Engineering
Katholieke Universiteit Leuven
B-3030 Heverlee, Belgium

SCOPE

During their processing, many viscous fluids, including polymer melts, are subjected to flow between rotating rollers. Nip flow can be used to homogenize material with respect to either composition or temperature, for kneading, or to produce continuous layers and sheets. Design, selection of operating conditions, and scaling-up of equipment require an adequate knowledge of the fluid mechanics during the process.

Early analyses assumed isothermal conditions and fluids with constant viscosity (Gaskell, 1950; Torner and Dobroljubov, 1958). McKelvey (1962) and Torner (1972) introduced power law liquids. An analytical solution for asymmetric nip flow of Newtonian liquids was recently obtained by Takserman-Krozer et al. (1975). Non-

Correspondence concerning this paper should be addressed to Jan Mewis.

isothermal effects were considered by Pearson (1966), who estimated the possible temperature rise in the nip. A more complete analysis of the nonisothermal case requires numerical techniques. Torner (1972) has discussed a solution by Petrusanskij et al. (1971) for symmetric and adiabatic conditions.

In the present paper, the real situation has been approached further by taking into account the combined effects of asymmetry and viscous heating. As in earlier publications, the entrance zone or bank region is dealt with in a simplified manner, and elastic effects are neglected. The results should provide improved estimates of the kinematics, the thermal effects, and the dynamic characteristics of calendering as a function of the process variables.

CONCLUSIONS AND SIGNIFICANCE

By means of the method of orthogonal collocation (Finlayson, 1972), a numerical solution has been obtained for the description of the calendering process of viscous liquids. Pressure forces are calculated to be approximately 20% below the isothermal predictions for polymer melts, if there is no slip between the rollers.

If the two rollers rotate at different speeds, as is the case in most real applications, the viscous heating has more drastic effects. The local temperature rise of the fluid near the slower roller can amount to 10 to 20°C. This heating creates locally a lubrication layer and causes important changes in the nip flow kinematics and consequently in the nip flow dynamics. Therefore, only the nonisothermal solution describes adequately the calendering operation with slip between the rollers. The optimal exploitation of the change in properties over the nip depends on the particular process at hand and on the material to be used.

The nip pressure is very sensitive to minor changes in roller distance. Hence laboratory experiments, meant for scaling-up or for verifying theoretical work, require a very accurate measurement of the expansion ratio (final over minimal layer thickness between the rollers) in order to be significant. The improved analysis, presented here, can be utilized to increase the efficiency in design and operation of roller systems in polymer processing. The results can be adapted to other applications of nip flow.

During calendering, a polymer melt is forced to flow between two rotating cylinders. Mechanically speaking, this operation constitutes an example of a larger class of processes based on nip flow. Apparently, Reynolds (1886) was the first to propose an analysis of this type of flow, namely, for lubrication. His solution is based on isothermal, symmetric, and Newtonian flow conditions. It has been applied in calendering by Gaskell (1950) and Torner and Dobroljubov (1958).

Analytical solutions do not allow elimination of the major simplifications introduced in the basic solution (Pearson, 1966). The introduction of a mean effective viscosity (Torner and Dobroljubov, 1958) or of bipolar coordinates (Ehrlich and Slattery, 1968; Takserman-Krozer et al., 1975) extends somewhat the range of the analytical solutions. McKelvey (1962), Pearson (1966), and Torner (1972) discussed partial solutions for power law fluids. Renert (1966) provided an approximate formula for the pressure profile in the nip for the same liquid model.

In order to combine non-Newtonian behavior with more realistic boundary conditions, numerical techniques are required (Reher and Grader, 1971; Alston and Astill, 1973). Owing to the transient pattern of the flow, a satisfactory way to incorporate the viscoelastic nature of the liquid has not yet been found. Preliminary attempts have been made by Paslay (1957) and by Tanner (1960). Perturbation around the first-order solution (Walters, 1972; Davies and Walters, 1972) provides insight in the problem but is not amenable to extrapolation for strongly nonlinear fluids. Dimensional analysis could eventually be used if sufficient insight and data were available (Tokita and White, 1966; White and Tokita, 1967; Chong, 1968).

Petrusanskij et al. (1971) seem to have been the first to tackle the nonisothermal case. Their numerical solution is based on a finite-difference scheme which limits its application to symmetric geometries (Kiparissides and Vlachopoulos, 1975). The method of finite elements (Zienkiewicz, 1971; Oden, 1971) offers better possibilities for solving general problems in fluid mechanics (for example, Booker and Huebner, 1972; Bilgen and Too, 1973-74; Tanner et al., 1975; Eidelberg and Booker, 1976; Yagawa et al., 1976). An attempted application on calendering, both symmetric and asymmetric cases, was presented by Kiparissides and Vlachopoulos (1975).

For highly viscous fluids, one expects the isothermal solution to lead to serious errors. The present study attempts to incorporate viscous heating in the nip flow of non-Newtonian liquids with asymmetric flow geometry. As in previous analyses, viscoelastic effects and, to some extent entrance effects, are not taken into account. This approximation is considered meaningful in many design calculations.

ANALYSIS OF GENERAL NIP FLOW

The general problem requires a numerical solution method. The method of finite elements (Zienkiewicz, 1971; Oden, 1971) suggests itself as a possibility. It was considered, however, whether other numerical techniques might be more suitable with respect to computing time and clarity of development. Orthogonal collocation (Finlayson, 1972), reducing an ordinary differential equation into a system of algebraic equations, has been shown to be an elegant and powerful tool in chemical engineering. The original applications in chemical reactor engineering (Villadsen and Stewart, 1967) have been extended to heat transfer (Chawla et al., 1975) and to fluid mechanics (Nirschl, 1972; Serth, 1974).

Orthogonal collocation is often applied on linear differential equations, resulting in linear algebraic equations. In the problem at hand, strongly nonlinear behavior must be expected. For the solution of moderately large systems of equations without strong nonlinearities, suitable subroutines are available (Marquardt, 1963). In the present case, a different route is followed.

The flow geometry of the present problem is represented in Figure 1. The rollers can be assumed to be rigid, contrary to some other applications of nip flow, where either the pressure is much higher or the rollers are more deformable.

The rheological behavior of the fluid will be described by means of the Ellis model in order to comprise the power law region as well as deviations towards Newtonian behavior at small shear rates

$$\frac{\tau_{ij}}{A} + \left(\frac{\tau_{ij}}{B}\right)^m = \dot{\gamma}_{ij} \quad (1)$$

In the calendering of highly viscous liquids, inertia can be neglected, and the usual creeping flow approximation will be used (Pearson, 1966). Then, the dynamic equation reduces to

$$\frac{\partial \tau_{xy}}{\partial y} = \frac{\partial p}{\partial x} \quad (2)$$

with boundary conditions

$$y = h_1(x), \quad v_x = U_1$$

$$y = h_2(x), \quad v_x = U_2$$

Equation (2) will be used in an integrated form

$$\tau_{xy} = f(x) \cdot y + g(x) \quad (3)$$

together with an integrated form of the continuity equation

$$Q = \int_{h_1(x)}^{h_2(x)} v_x dy \quad (4)$$

Dimensionless expressions are obtained by defining the following terms

$$V = \frac{v_x}{U_1}$$

$$Y = \frac{y - h_1}{h} \quad (5)$$

where $h = h_2 - h_1$

$$C = \frac{A^m U_1^{m-1}}{B^m h^{m-1}}$$

$$F(X) = \frac{f(x) \cdot h^2}{A \cdot U_1}$$

$$G(X) = \frac{g(x) \cdot h}{A \cdot U_1}$$

$$X = \frac{x + x_1}{2x_1}$$

From Equations (1) through (5), the dimensionless velocity gradient can be derived

$$\frac{\partial V}{\partial Y} = \left[F(X)\left(Y + \frac{h_1}{h}\right) + G(X) \right]$$

$$\left\{ 1 + C\left|F(X)\left(Y + \frac{h_1}{h}\right) + G(X)\right|^{m-1} \right\} \quad (6)$$

The differential equation is solved by applying orthogonal collocation in the points $Y = Y_i$

$$\sum_{k=1}^{n} E_{ik} V_k = \left[F(X)\left(Y_i + \frac{h_1}{h}\right) + G(X) \right]$$

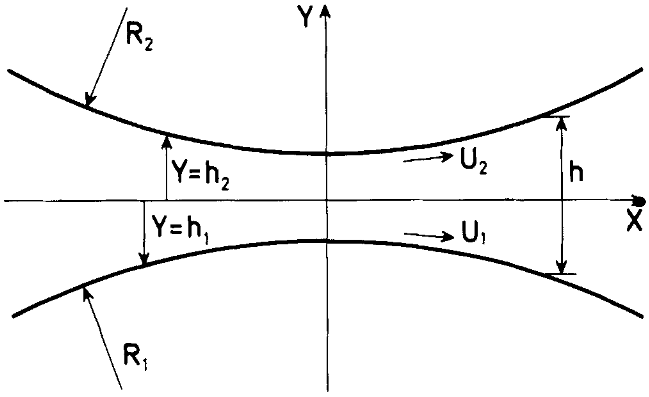

Fig. 1. Nip flow geometry.

$$\left\{ 1 + C\left|F(X)\left(Y_i + \frac{h_1}{h}\right) + G(X)\right|^{m-1} \right\} \quad (7)$$

together with an expression for the flow rate, based on a Lobatto quadrature (Villadsen, 1970)

$$\sum_{k=1}^{n} V_k W_k = \frac{Q}{2U_1 h} = Q' \quad (8)$$

and the dimensionless boundary conditions

$$Y = 0 \quad V_1 = 1$$

$$Y = 1 \quad V_n = \frac{U_2}{U_1} \quad (9)$$

Equations (7), for $i = 1, 2 \ldots n - 1$, together with Equation (8) constitute a system of n algebraic equations with n unknowns $V_2 \ldots V_{n-1}$, $F(X)$, and $G(X)$, only nonlinear in the last two. A double iteration scheme, based on the known values of V_n [Equation (9)] and Q' [Equation (8)], provides values for $G(X)$ and $F(X)$, respectively. The starting value of $F(X)$ is based on an approximate solution for the pressure gradient, which becomes exact for weak asymmetry. $G(X)$ is derived initially from the corresponding function for Newtonian fluids. The viscosity is derived from a suitable expression, based on the maximum shear rate of the Newtonian velocity profile. The regula falsi of second order has been used. The outlined procedure eliminates the nonlinearity. It makes an efficient use of existing subroutines for the solution of systems of linear algebraic equations.

The thus computed velocity and pressure profiles were compared with corresponding results from the analytical solution (McKelvey, 1962) and from a numerical solution, based on finite elements (Kiparissides and Vlachopoulos, 1975). The comparison shows the present method to be sufficiently accurate and in most cases more accurate than the other available numerical solution, even after an error in the pressure profile for the latter was corrected (Dobbels, 1977). A comparison with available experimental work (Bergen and Scott, 1951) is difficult, as the necessary rheological information about the liquids used is lacking.

In order to calculate the dynamic characteristics of the calender, the limiting values for the pressure and the contribution of the bank region must be taken into account. In one way or another, all authors have extended the creeping flow analysis to the bank region (for example, Myers and Hoffman, 1961; Brazinsky et al., 1970). In the present analysis, a stepwise estimation of the pressure has been obtained, if we assume creeping flow and power law behavior. If we start at the peak value,

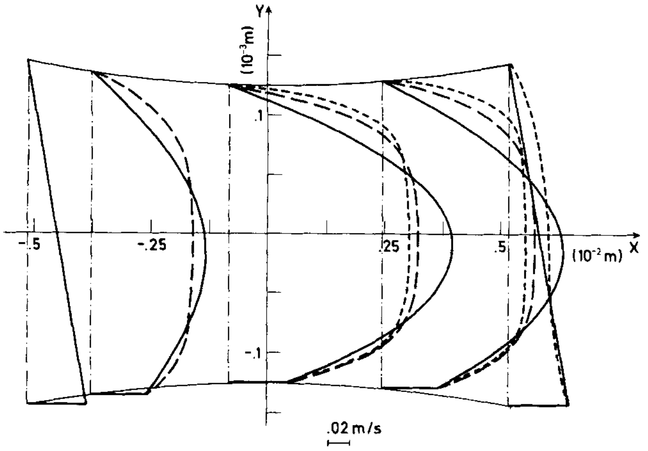

Fig. 2. Velocity distribution ($v_x - U_1$) for asymmetric nip flow.
———— Newtonian model, isothermal
—— —— Ellis model, isothermal
- - - - - - Ellis model, nonisothermal
$U_1 = 0.75$ m/s; $U_2 = 0.70$ m/s
$R_1 = 0.72$ m; $R_2 = 0.62$ m
$Q = 2.1 \times 10^{-4}$ m³/s,m; $H = 1.159$
other conditions: see Table 1

TABLE 1. MATERIAL PROPERTIES USED IN DIMENSIONAL COMPUTATIONS

A	=	180 000 $Pa \cdot s$
B	=	122 000 $Pa \cdot s^{1/m}$
m	=	3.7
c_A	=	0.05 °C^{-1}
c_B	=	0.046 °C^{-1}
k	=	0.1382 $J/m, s$, °C
c_v	=	1 716 J/kg, °C
ρ	=	1 250 kg/m^3
T_o	=	180°C

computation is stopped at the position where $p = o$, or otherwise at $x = -4x_1$. In this manner the complex flow pattern in the bank region is ignored (Stara et al., 1973), which is acceptable in design calculations but not in quality considerations. The neglect of elastic contributions for polymers leads to similar restrictions. Hence both simplifications are compatible.

In order to take into account nonisothermal effects, the energy equation and the temperature dependence of the rheological characteristics must be included. For the latter, the following expressions will be used

$$A = A_o \exp c_A(T_o - T)$$
$$B = B_o \exp c_A(T_o - T) \tag{10}$$
$$m = \text{constant}$$

For nip flow, the energy equation can be written as

$$\rho c_v v_x \frac{\partial T}{\partial x} = k \frac{\partial^2 T}{\partial y^2} + \eta(\dot{\gamma})\,\dot{\gamma}^2 \tag{11}$$

Again, a more suitable expression results if dimensionless terms are defined

$$Pe = \frac{\rho c_v U_1 h^2}{2kx_1}$$
$$Br = \frac{\eta U_1^2}{kT_o}$$
$$T' = \frac{T}{T_o}$$
$$X = \frac{x + x_1}{2x_1} \tag{12}$$

This leads to

$$PeV \frac{\partial T'}{\partial X} = \frac{\partial^2 T'}{\partial Y^2} + Br\left(\frac{\partial V}{\partial Y}\right)^2 \tag{13}$$

If we apply orthogonal collocation for $Y = Y_i$ and $X = X_j$, Equation (13) becomes

$$PeV_{ij} \sum_{k=1}^{n} T'_{ik}E_{kj} = \sum_{k=1}^{n} D_{ik}T'_{kj} + Br\left(\sum_{k=1}^{n} E_{ik}V_{kj}\right)^2 \tag{14}$$

As an initial condition, the liquid is assumed to be at constant temperature at the entrance of the nip. In addition, the heat flux in liquid and calender are put equal at the roller surface. The thermal effects must be taken into account while the velocity gradient is calculated. Equation (6) thus becomes

$$\frac{\partial V}{\partial Y} = \left[F(X)\left(Y + \frac{h_1}{h}\right) + G(X)\right] e^{-c_A(T_o - T)}$$
$$\left\{1 + Ce^{(c_A - mc_B)(T_o - T)}\left|F(X)\left(Y + \frac{h_1}{h}\right) + G(X)\right|^{m-1}\right\} \tag{15}$$

Equations (13) and (15) constitute a system of differential equations describing the nonisothermal process. A discretization of the whole problem leads to a very complicated and huge set of nonlinear algebraic equations. A decomposition is proposed which proves to converge sufficiently. It is based on a separate solution of Equation (14) and the collocation form of Equation (15), followed by an iteration between the two.

Starting from the isothermal velocity distribution, one calculates the temperature profile by iteration on $\partial^2 T'/\partial Y^2$. This profile results in improved values for pressure and velocity distribution. The whole procedure is repeated until the desired accuracy is achieved, giving the nonisothermal temperature, velocity, and pressure profiles. The number of steps in each iteration remains relatively small. The outlined procedure provides an efficient and flexible method for the modeling and analysis of nip flow processes with various degrees of complexity.

APPLICATION TO THE ANALYSIS OF NONISOTHERMAL CALENDERING

The previous analysis has been applied to investigate nonisothermal calendering. The dimensional results are based on material properties as collected in Table 1.

The kinematics provide the basis for the discussion of nip flow. Therefore, the velocity profiles for the isothermal Newtonian, the isothermal and nonisothermal Ellis cases are compared in Figure 2. As could be expected, viscous heating has, qualitatively, a similar effect as shear thinning. Only shear thinning is an immediately reversible effect, resulting in a velocity profile symmetric with respect to the y axis. Viscous heating, on the other hand, proceeds throughout the nip.

Some important conclusions can be drawn from the results. If compared with shear thinning, the effect of viscous heating is not negligible. It is also concentrated at the exit and near the slower roller, if the rollers rotate at different speeds. In this manner a layer develops which more or less acts as a lubricant for the other liquid layers. These particular kinematics can be beneficial through the corresponding reduction in forces and through some gain in mixing. However, local temperature peaks and distortions in the flow pattern will put an upper limit to the possible asymmetry as they affect the quality of the product.

Further insight in nonisothermal calendering is gained from the flow dynamics. The forces are derived from the pressure profile, represented in Figure 3. The isothermal peak pressure is taken as reference pressure. It can be seen that the Ellis liquid shows a somewhat more linear change of pressure. Viscous heating destroys the symmetry with respect to the nip center. In addition, the local values of the pressure are reduced by 25% in this particular case. With respect to the isothermal Newtonian solution ($\eta_o = A$), this means a reduction by a factor of 200 . . . 300. Peak pressures of 50 MPa and total pressure forces of about 500 $\times$ 10^3 N/m are reached quite easily. In that range, the maximum elastic deformation with steel rollers amounts only to 4 μm (Love, 1952). With industrial minimum nip clearances of 250 . . . 500 μm, the decrease in pressure buildup due to deformation of the rollers is of only minor importance.

If we combine the evidence from Figures 2 and 3, we can conclude that a consistent approximation of viscous calendering, which takes into account shear thinning, should also take into account viscous heating. Figure 2 seems to reveal a particular nonisothermal effect due to the flow asymmetry. As most industrial nip flow processes work under asymmetric conditions, this point is investigated in more detail. The total normal pressure force between the cylinders is used as a characteristic of the dynamics. In Figure 4, the effects of radius ratio and roller speed ratio are represented.

A change in geometry can be approached in several ways. An increase of the radius ratio by reducing R_2 amounts to lowering the resistance in the nip. Consequently, the total pressure force is also reduced. The reduction is nearly identical for Newtonian and Ellis fluids. In Figure 4, the radius ratio has been changed, keeping the sum of the radii constant in order to minimize the effect of variable cross section. If required, this effect could be further eliminated by keeping the sum of the inverses of the radii constant. If the nonisothermal solution is used, the total pressure drops below the isothermal case in a proportion that could be predicted from Figure 3. The relative effect of radius ratio remains the same as in the isothermal case. Although the asymmetric geometry has some mathematical interest, it is of no direct importance in practical design calculations. The symmetric calculations can be extrapolated adequately to the asymmetric case. With a lubrication theory, assumming slowly changing functions $h_1(x)$ and $h_2(x)$, it is quite evident that asymmetry in the boundaries, keeping other parameters constant, is of minor importance. This conclusion is only valid for the calendering of viscous fluids. Viscoelastic phenomena might result in divergent conclusions (Metzner, 1968).

A change in the velocity ratio, with the sum $U_1 + U_2$ constant, leads to more specific effects. For Newtonian liquids the roller speeds enter the equations only through their sum. Hence, Figure 4 shows no change in pressure if U_2/U_1 is altered. If shear thinning liquids are considered, a speed difference between the rollers induces a distorted velocity profile with larger shear rates. The corresponding drop in apparent viscosity explains the decreasing pressure in Figure 4. For the strongly nonlinear material of Table 1, the global decrease can be important.

For the nonisothermal solution, the reduction is amplified further. In this manner, an interesting interaction between shear thinning, viscous heating, and velocity asymmetry is proved to exist. The drop in pressure force is in agreement with the lubrication effect detected earlier in the kinematics (Figure 2).

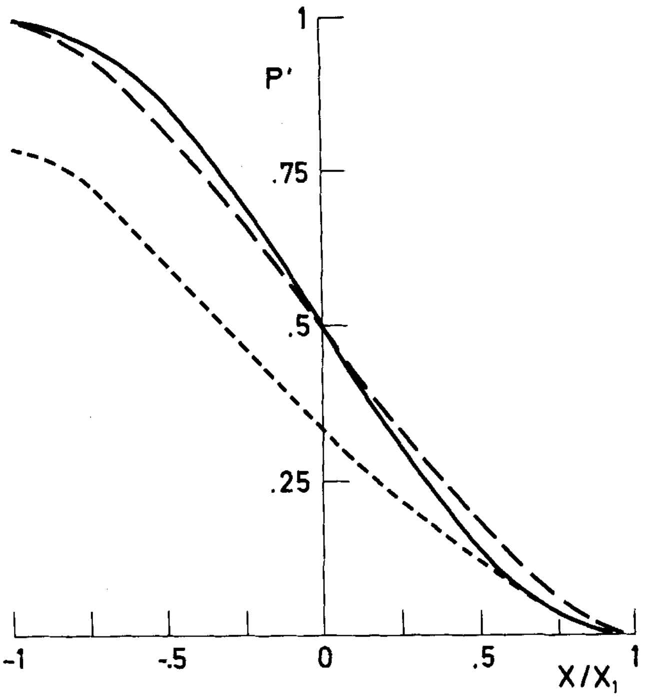

Fig. 3. Pressure profiles in the creeping flow region of the nip, effect of shear thinning and viscous heating. (Legend, see Figure 2.)

At high velocity ratios, the present analysis is bound to break down; the increasing contributions from the bank region and the splitting zone make the creeping flow approximation finally unreliable. For the same reasons and owing to the increasing distortion in velocity and temperature profiles, resulting in quality loss of the final product, the calendering operation must become less effective at high velocity ratios. Therefore, a roller speed ratio different from unity is found to be advantageous, but the ratio should probably remain below 1.2 to 1.3 or even lower. This conclusion is in agreement with industrial experience. Hence, the nonisothermal effects are essential in evaluating speed ratios.

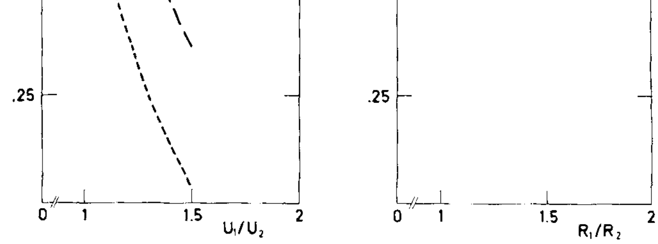

Fig. 4. Effect of radius and velocity ratios on the total pressure forces.
$R_1 + R_2 = 1.44$ m
$U_1 + U_2 = 1.4\ 5$m
(other conditions: see Figure 2.)

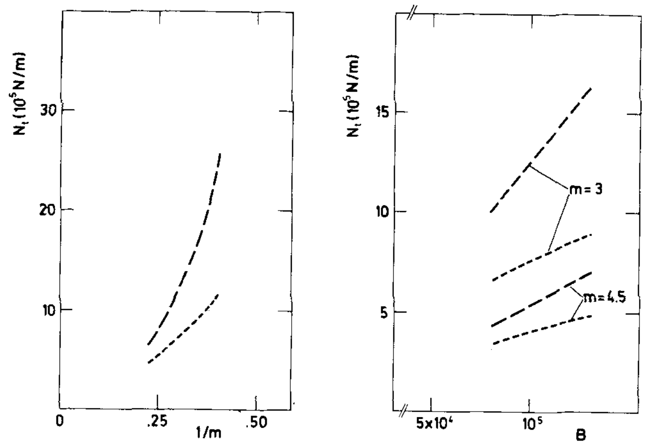

Fig. 5. Effect of rheological parameters on total pressure forces. (Conditions: see Figure 2.)

The sensitivity of nip flow dynamics to the rheological parameters under isothermal and nonisothermal conditions has been compared in Figure 5. For the range of model parameters used, the low shear Newtonian viscosity turns out to be unimportant; the power law behavior dominates nip flow. A change in B then corresponds to an overall shift of the viscosity curve. This

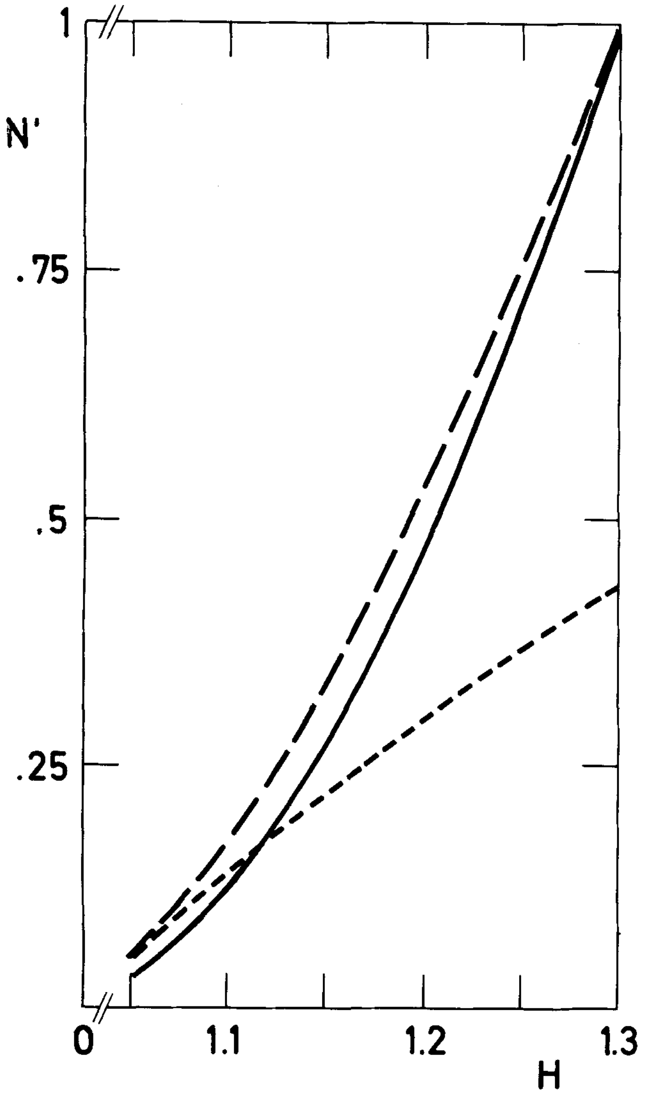

Fig. 6. Dependence of nip flow dynamics on the expansion ratio *H*. (Conditions: see Figure 2.)

TABLE 2. SENSITIVITY OF DYNAMIC CHARACTERISTICS AND TEMPERATURE TO THE THERMAL PROPERTIES
($p_{\max,T=cte} = 778 \times 10^5\ Pa$; $N_{T=cte} = 936 \times 10^3\ N/m$)

c_v, J/kg, °C	k, J/s, m, °C	$c_A = c_B$, °C^{-1}	$T_{\max}$, °C	$P_{\max,T=cte}$, $10^5\ Pa$	$N_{T=cte}$, $10^3\ N/m$
1 716	0.1382	0.046	192	615	615
1 716	0.1382	0.040	193	630	641
1 716	0.1382	0.035	193	646	668
1 716	0.1382	0.050	192	604	596
1 716	0.1382	0.055	192	593	577
1 716	0.0921	0.046	194	606	598
1 716	0.1842	0.046	191	622	628
1 144	0.1382	0.046	194	575	548
2 289	0.1382	0.046	191	642	660

explains the nearly linear relationship between N_t and B. Clearly, the proportionality factor will depend on the power law index $1/m$ (Hellinckx and Mewis, 1969).

The sensitivity to the latter parameter depends on the way the comparison is made. Figure 5 is based on independent variations of m and B, and therefore a considerable effect of $1/m$ on N_t is found. Material constants could also be compared, keeping the apparent viscosity constant at a given, arbitrary value of shear rate. The results will depend on the chosen value of $\dot{\gamma}$ and therefore are not discussed further. One expects the viscous heating to reduce the force more when the viscosity levels are higher. In the range of parameters investigated here, the resulting drop in pressure force changes from a few percent up to 30%. The latter value depends on the thermal properties. The extent to which these parameters can alter the process is illustrated in Table 2.

Under the conditions used throughout the calculations, the local temperatures increase up to 15°C. Under a change of conductivity, of specific heat, or of the viscosity-temperature coefficients within reasonable limits, that is, for a given class of materials, no relevant effects can be detected. A variation of 3% in thermal properties causes a change in peak pressure and total pressure force of about 1%. The latter is somewhat more sensitive to variations in c_i, c_v, and k due to the contribution of the bank region. Rather large deviations (20 . . . 30%) in the said properties are required to alter the local maximum for temperature rise by a few degrees. Nevertheless, the total change in temperature profile is sufficient to cause a substantial difference in the calender dynamics, as already indicated above.

The calendering operation can be influenced during the process by changing the nip geometry. Either the total pressure force or the distance between the rollers can be varied. This procedure is one of the possible methods to collect information about the operation. For the purpose of comparison, the roller distance, expressed as the ratio h_{exit}/h_{min}, was changed, keeping the other variables constant. The isothermal results at $H = 1.3$ are taken as reference values. Figure 6 proves the sensitivity of the dynamics to H, even with the partially compensating effect of viscous heating. An increase in H from 1.1 to 1.2 doubles the pressure. Elastic deformation of the rollers, which is said to be unimportant in normal operating conditions, should slightly decrease this dependence on H.

This sensitivity hampers the experimental verification of calendering calculations. To be meaningful, the values of h_{min} and h_{exit} in any experiment have to be available with an accuracy which is often difficult or impossible

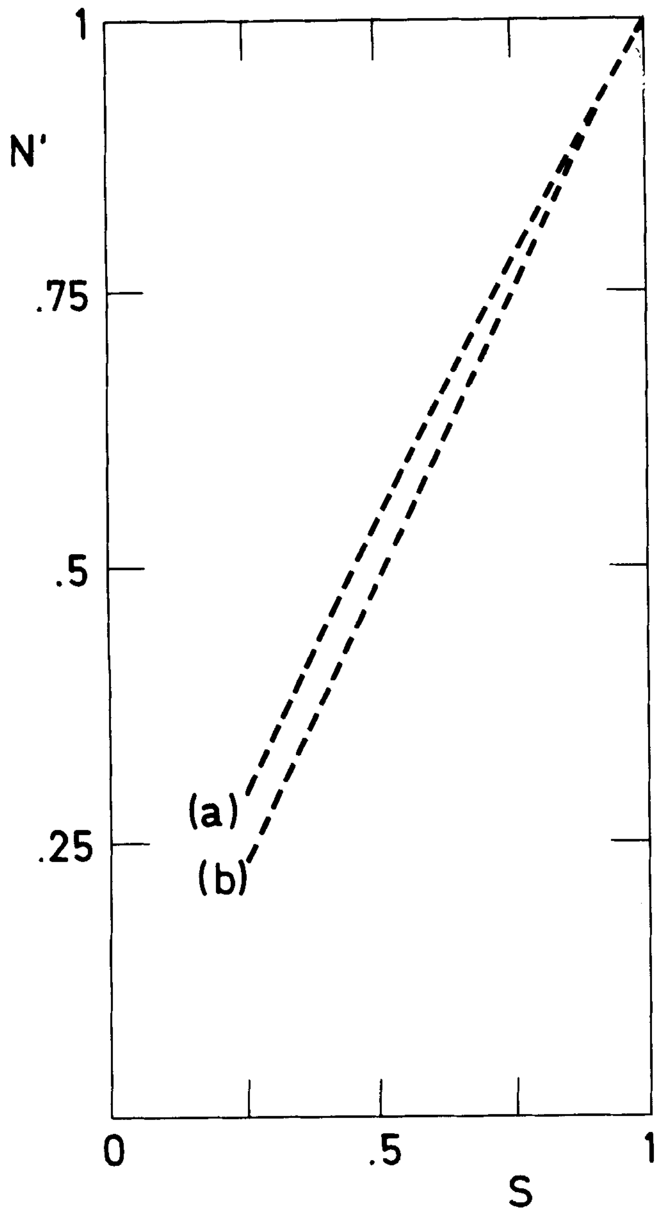

Fig. 7. Scaling-up for total pressure force in nonisothermal calendering (conditions: see Figure 2).
(a) $R_{is} = S \cdot R_{if}$
(b) $R_{is} = S \cdot R_{if}$
$U_{is} = S \cdot U_{if}$

to reach. These difficulties should be considered when calendering experiments are planned. Finally, the calculations can be applied on the problem of scaling-up. The total pressure force N_t is used as a characteristic design parameter. The conclusions can be extended to the other dynamic characteristics. Two specific scaling-up procedures are distinguished. In the first one, the roller radii are scaled up by a factor S, while all other geometric and kinematic parameters are kept identical:

$$R_{is} = SR_{if}$$

As a result, corresponding sections, having the same height, are given by $x_s = x_f\sqrt{S}$

From Equation (15) it can be seen that the pressure gradient at corresponding sections remains unchanged, at least for isothermal conditions. Integration then shows that the values for local pressure are related by $\sqrt{S}$. Hence, the total pressure forces are governed by the same scale factor as the radii.

Viscous heating alters the picture, as the local Peclet numbers at corresponding positions are related by a scale factor of $1/\sqrt{S}$, giving a lower temperature gradient $\partial T'/\partial X$ [Equation (13)]. Therefore, a scale model shows less heating and higher total pressure forces than expected from an isothermal analysis. With the material properties of Table 1, the nonisothermal scaling-up results in deviations to less than 20% with respect to the isothermal case, if $S > 0.25$ (Figure 7). In order to avoid very short residence times on the scale model, one can reduce the roller velocities in the same way as the radii. With this second procedure, no simple scaling factor can be derived for N_t. The isothermal solution leads to values for N_{ts}, which now drop below SN_{tf}. However, the viscous heating, if taken into account, more or less compensates these deviations (Figure 7), making S again an approximate scale factor.

This result, together with the discussion on model experiments, suggests that scaling-up of calender dynamics is possible, provided the geometric parameters can be very accurately measured in a model experiment.

ACKNOWLEDGMENT

Professor Vl. Hlavacek, Prague, and J. C. Chauffoureaux, Solvay, Belgium, are thanked for stimulating discussions on the numerical technique and on calendering, respectively.

NOTATION

A = parameter of the Ellis model (Pa.s)
A_o = $(A)_{T=To}$
B = parameter of the Ellis model (Pa.s$^{1/m}$)
B_o = $(B)_{T=To}$
Br = Brinkmann number, defined for calendering by Equation (12)
C = dimensionless function, defined by Equation (5)
c_A,c_B = constants of the temperature dependency of A and B, Equation (10) (°C^{-1})
c_v = specific heat of the polymer melt at constant volume (J/kg,°C)
D_{ik} = component ik of the collocation matrix $||D||$, Equation (14)
E_{ik} = component ik of the collocation matrix $||E||$, Equation (7)
f = index, refers to the full scale model
$f(x)$ = function, defined by Equation (3), (Pa/m)
$F(X)$ = dimensionless form of $f(x)$, Equation (5)
$g(x)$ = function, defined by Equation (3), (Pa)
$G(X)$ = dimensionless form of $g(x)$, Equation (5)
h = local nip height (m)
H = expansion ratio = h_{exit}/h_{min}
h_{exit} = nip height at the exit, $h(x_1)$ (m)
h_i = ordinate of the wall of calender i (m)
h_{min} = minimum nip height in the center (m)
k = thermal conductivitv of the melt (J/m, s, °C)
m = parameter of the Ellis model
N_t = total pressure force per unit width (N/m)
N' = dimensionless total pressure force
$N_{T\neq cte}$ = isothermal total pressure force per unit width (N/m)
$N_{T=cte}$ = nonisothermal total pressure force per unit width (N/m)
p = hydrodynamic pressure (Pa)
P' = dimensionless hydrodynamic pressure
Pe = Peclet number, defined for calendering by Equation (12)
$p_{max,T=cte}$ = isothermal hydrodynamic peak pressure (Pa)

$p_{max,T \neq cte}$ = nonisothermal hydrodynamic peak pressure (Pa)
Q = flow rate per unit width (m^3/m, s)
Q' = dimensionless flow rate, defined by Equation (8)
R_i = radius of the calender i (m)
s = index, refers to the scale model
S = scaling factor, Figure 7
T = temperature of the melt (°C)
T' = dimensionless temperature, Equation (12)
T_{max} = peak temperature of the polymer melt (°C)
T_o = temperature of the polymer melt at the entry (°C)
U_i = velocity of the calender i (m/s)
V = dimensionless velocity, Equation (5)
V_i = value of the dimensionless velocity for $Y = Y_i$
v_x = velocity of the polymer melt in the flow direction x(m/s)
W_k = weights of the Lobatto quadrature, Equation (8)
x = abscissa, in the direction of the one-dimensional creeping flow (m)
X = dimensionless abscissa, normalized to the interval [0, 1], Equation (5)
X_i = element i of the vector of collocation points $\underline{X}$
x_1 = abscissa of the exit, defined from $Q = 0.5\, \overline{h}(x_1)\, (U_1 + U_2)$ (m)
y = ordinate, perpendicular to the flow direction x (m)
Y = dimensionless ordinate, normalized to the interval [0, 1], Equation (5)
Y_i = element i of the vector of collocation points $\underline{Y}$

Greek Letters

$\dot{\gamma}$ = shear rate (s^{-1})
$\dot{\gamma}_{ij}$ = component ij of the deformation rate tensor $\underline{\underline{\dot{\gamma}}}$ (s^{-1})
η = non-Newtonian viscosity (Pa.s)
η_o = Newtonian viscosity (Pa.s)
ρ = specific weight of the polymer melt (Kg/m^3)
τ_{ij} = component ij of the stress tensor $\underline{\underline{\tau}}$ (Pa)

LITERATURE CITED

Alston, W. W., Jr., and K. N. Astill, "An Analysis for the Calendering of Non-Newtonian Fluids," *J. Appl. Polymer Sci.*, 17, 3157 (1973).
Bergen, J. T., and G. W. Scott, Jr., "Pressure Distribution in the Calendering of Plastic Materials," *J. Appl. Mech.*, **18**, 101 (1951).
Bilgen, E., and J. J. M. Too, "On the Finite Element Formulation of Navier-Stokes Equations," *Trans. CSME*, **2**, No. 4, 205 (1973-74).
Booker, J. F., and K. H. Huebner, "Application of Finite Element Methods to Lubrication: An Engineering Approach," *Trans. ASME(F), J. Lubr. Techn.*, **94**, No. 4, 313 (1972).
Brazinsky, I., H. F. Cosway, C. F. Valle, Jr., R. Clark Jones, and V. Story, "A theoretical Study of Liquid-Film Spread Heights in the Calendering of Newtonian and Power Law Fluids," *J. Appl. Polymer Sci.*, **14**, 2771 (1970).
Chawla, T. C., G. Leat, W. L. Chen, and M. A. Grolmes, "The Application of the Collocation Method using Hermite Cubic Splines to Non-Linear Transient One-Dimensional Heat Conduction Problems," *Trans. ASME(C), J. Heat Transfer*, **97**, 562 (1975).
Chong, J. S., "Calendering Thermoplastic Materials," *J. Appl. Polymer Sci.*, **12**, 191 (1968).
Davies, J. M., and K. Walters, "The Behaviour of Non-Newtonian Lubricants in Journal Bearings—A theoretical Study," Confer. Rheol. Lubr., Nottingham (July 6-7, 1972).
Dobbels, F., PH.D. thesis, in preparation, K. U. Leuven (1977).
Ehrlich, R., and J. C. Slattery, "Evaluation of Power-Model Lubricants in an infinite Journal Bearing," *Ind. Eng. Chem. Fundamentals*, **7**, No. 2, 239 (1968).
Eidelberg, B. E., and J. F. Booker, "Application of Finite Element Methods to Lubrication: Squeeze Films between Porous Surfaces," *Trans. ASME(F), J. Lubr. Techn.*, **98**, No. 1, 175 (1976).
Finlayson, B. A., *The Method of Weighted Residuals and Variational Principles*, Academic Press, London and New York (1972).
Gaskell, R. E., "The Calendering of Plastic Materials," *J. Appl. Mech.*, **17**, 334 (1950).
Hellinckx, L., and J. Mewis, "Rheological Behaviour of Pigment Dispersions as related to Roller Passage," *Rheol. Acta*, **8**, No. 4, 519 (1969).
Kiparissides, C., and J. Vlachopoulos, "Finite Element Analysis of Calendering," 46th Annual Meeting, Soc. Rheol., St. Louis, Mo. (Oct., 1975).
Love, A. E., *Mathematical Theory of Elasticity*, 4 ed., p. 193, Cambridge Univ. Press, England (1952).
Marquardt, D. W., "An Algorithm for Least Squares Estimation of Non-Linear Parameters," *J. of SIAM*, **11**, 431 (1963).
McKelvey, J. M., *Polymer Processing*, Wiley, New York (1962).
Metzner, A. B., "The significant rheological Characteristics of Lubricants," *Trans. ASME(F), J. Lubr. Techn.*, **90**, 531 (1968).
Myers, R. R., and R. D. Hoffman, "The Distribution of Pressures in the Roll Application of Newtonian Fluids," *Trans. Soc. Rheol.*, **5**, 317 (1961).
Nirschl, J. P., "Orthogonal Collocation Analysis of Viscoelastic Fluid Flow in the Disc and Cylinder System," Ph.D. thesis, Univ. Wisc., Madison (1972).
Oden, J. T., *Finite Elements of Non-Linear Continua* McGraw-Hill, New York (1971).
Paslay, P. R., "Calendering of a viscoelastic Material," *J. Appl. Mech.*, **24**, 602 (1957).
Pearson, J. R. A., *Mechanical Principles of Polymer Melt Processing*, Pergamon Press, New York (1966)).
Petrusanskij, V. Ju., et al., "Masiny i Technologija Pererabotki Polimerov," isd. LTI im. Lensoveta (1971); ref. by Torner, R. V., Grundprozesse der Verarbeitung von Polymeren, VEB Deutscher Verlag für Grundstoffindustrie, Leipzig (1974).
Reher, E. O., and L. Grader, "Zur Berechnung einer isothermen Doppel-Walzen-Kalanderströmung nichtlinear-plastischer Medien," *Plaste und Kautschuk*, **18**, No. 8, 597 (1971).
Renert, M., "Asupra Teoriei Procesului de Calandrare a Materialelor Plastice," *Materiale Plastice*, **3**, No. 3, 132 (1966).
Reynolds, O., "On the Theory of Lubrication and its Application to Mr. Beauchamp Tower's Experiments, including an experimental Determination of the Viscosity of Olive Oil," *Phil. Trans. Part I*, **177**, 157 (1886).
Serth, R. W., "Solution of a Viscoelastic Boundary Layer Equation by Orthogonal Collocation," *J. Eng. Math.*, **8**, No. 2, 89 (1974).
Stara, D., V. Krizek, and M. Hastik, "Studium Proudeni Taveniny PVC Mezivalci, *Plasticke Hnioty & Kaucuk*, **7**, 196 (1973).
Takserman-Krozer, R., G. Schenkel, and G. Ehrmann, "Fluid Flow Between Rotating Cylinders," *Rheol. Acta*, **14**, 1066 (1975).
Tanner, R. I., "Full-Film Lubrication Theory for a Maxwell Liquid," *Int. J. Mech. Sci.*, **1**, 206 (1960).
———, R. E. Nickell, and R. W. Bilger, "Finite Element Methods for the Solution of Some Incompressible Non-Newtonian Fluid Mechanics Problems with Free Surfaces," *Comput. Meth. Appl. Mech. Eng.*, **6**, No. 2, 155 (1975).
Tokita, N., and J. L. White, "Milling Behaviour of Gum Elastomers: Experiment and Theory," *J. Appl. Polymer Sci.*, **10**, 1011 (1966).
Torner, R. V., Grundprozesse der Verarbeitung von Polymeren, VEB Deutscher Verlag für Grundstoffindustrie, Leipzig (1974) (published in Russian, 1972).
———, and G. V. Dobroljubov, *Kaucuk i Resina*, **4**, 6 (1958); ref. by Torner, R. V., Grundprozesse der Verarbeitung von Polymeren, VEB Deutscher Verlag für Grundstoffindustrie, Leipzig (1974).
Villadsen, J. V., *Selected Approximation Methods for Chemical Engineering Problems*, p. 123, Danmarks Tekniske Højskole, Lyngby (1970).
———, and W. E. Stewart, "Solution of Boundary—Value Problems by Orthogonal Collocation," *Chem. Eng. Sci.*, **22**, 1483 (1967).

Walters, K., "New Concepts in Theoretical and Experimental Rheology," Confer. Rheol. Lubr., Nottingham, (July 6-7, 1972).
White, J. L., and N. Tokita, "Elastomer Processing and Application of Rheological Fundamentals," *J. Appl. Polymer Sci.*, 11, 321 (1967).
Yagawa, G., Y. Ishida, and Y. Ando, "The Finite Element Method Applied to the Navier-Stokes Equation, *Int. Chem. Eng.*, 16, No. 2, 253 (1976).
Zienkiewicz, O. C., *Finite Element Method in Engineering Science*, McGraw-Hill, New York (1971).

Manuscript received November 2, 1976; and accepted December 15, 1976.

Flow of Slurries in Pipelines

RAFFI M. TURIAN
and
TRAN-FU YUAN
Department of Chemical Engineering and Materials Science
Syracuse University
Syracuse, New York 13210

Pressure drop correlations for flow of slurries in pipelines were developed for each of the following four flow regimes: flow with a stationary bed, saltation flow, heterogeneous flow, and homogeneous flow. A total number of 2 848 data points, comprised of 1 912 collected from the published literature together with 936 taken using our own test pipelines and relating to ranges of the pertinent variables extensive enough to span all four flow regimes were used as the basis of these correlations. Also, these data were used in developing an associated quantitative regime delineation scheme. The correlations provide an improved predictive capability over previously available procedures and are also broader in scope. The delineation procedure developed here permits straightforward classification of the data according to the flow regime prevailing, and it is moreover inclusive of all the data and is self-consistent.

SCOPE

The principal objective of this study was to explore possible effective schemes for correlating data relating to flow of slurries in pipelines. The immense practical implications of this mode of solid transport have motivated a resurgence of activity which is continuing to contribute to an expanding storehouse of raw data. Some unifying framework capable of enforcing order in this vast maze of data, and hopefully of providing a quantitative means for predicting the prevailing flow regime, is needed. A number of different flow regimes, depending upon the dynamic conditions of the flow and the nature of the slurry, are known to occur in such flows, and therefore we have developed pressure drop correlations pertaining to each of the following four regimes: flow with a stationary bed, saltation flow, heterogeneous flow, and homogeneous flow. Quantitative criteria for ascertaining the flow regime prevailing under given conditions of flow were also developed, and these are used in conjunction with the pressure drop correlations. The estimation of pressure drop in slurry pipeline design is an important practical problem which, owing to the complexity of the process involved, has been subject to considerable uncertainty. A comparison of the few available slurry flow correlations with the present results was made using the entire body of data collected for establishing our correlations as a basis. Such a comparison, besides establishing the improved predictive ability of our new correlations, serves also to provide the proper perspective needed in the selection of the most suitable design procedure in any given situation. In some of the previous empirical correlations, significant portions of the data have been excluded on the grounds that they do not belong to flow regimes being considered. However, the criteria used for such exclusion can not definitively identify the demarcation between flow regimes since, at the very least, in reality the transition from one flow regime to another must take place gradually and not in the abrupt fashion implied by the use of a transition number. Indeed, the descriptive designation of the various regimes is an essentially subjective procedure. Nonetheless, it is important to emphasize that the criteria developed in the present work for ascertaining the appropriate flow regimes are not used to exclude portions of the experimental data but to insure that the most nearly suitable correlation is used in pressure drop estimation. Furthermore, these quantitative criteria are consistent with the expected progressive transition from one flow regime to the next, and, in fact, even in those very few cases in which the flow regime number configuration appears to be anomalous, these criteria do provide a fully satisfactory explanation of the observed behavior.

R. M. Turian is with the National Science Foundation on leave from Syracuse University. T-F. Yuan is with Bristol-Myers Industrial Division, P.O. Box 657, Syracuse, New York 13201.