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NOMENCLATURE

A = rate factor, μmole/g-hr.$^{1/2}$
B = regression parameter in Equation (3*a*), μmole/g
B' = regression parameter in Equation (3*b*), μmole/g-hr.
c = dissolved oxygen concentration, μmole/l
c^* = dissolved oxygen saturation concentration, μmole/l
$\bar{c}$ = oxygen sorbed on activated carbon, μmole/g
Ka = sparger mass transfer coefficient, 1/hr.
Q = flow rate through column, l/hr.
$\bar{r}$ = rate of sorption, μmole/g-hr.
V = volume of solution circulated through column, l
W = weight of activated carbon in column, g

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R & D NOTES

Migration of Two Liquid Phases in Capillary Extrusion: An Energy Interpretation

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In the production of laminated plastic film and certain types of bicomponent fibers, two polymer melts are coextruded by being metered into a die in layered configuration (Figure 1*a*). Such a configuration, however, is known to be unstable; in circular capillary dies there is observed a tendency for mutual phase migration. Downstream, the low-viscosity phase tends to envelope the high-viscosity phase, as depicted in Figure 1*b*. Extensive experimental evidence presented by Lee and White (1974) and by Southern and Ballman (1973) indicates that the relative

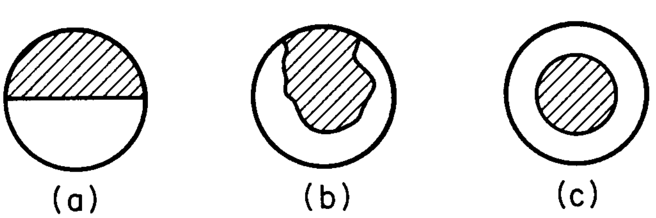

Fig. 1. Cross sections of die with bicomponent flow; dark region has higher viscosity. *a*. Typical inlet condition. *b*. Typical rearrangement downstream, with low-viscosity fingering around circumference of die. *c*. Ultimate result of the process shown in *b*.

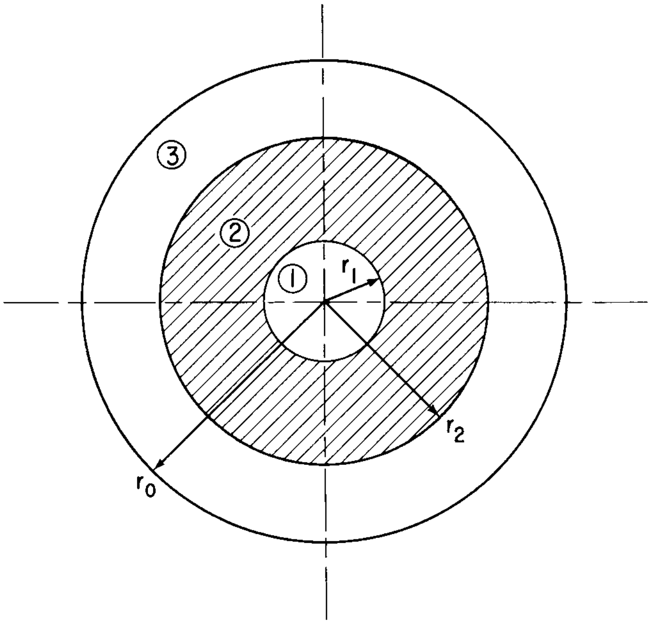

Fig. 2. Geometry of phase positions. Viscosity is η_1 in regions 1 and 3 and η_2 in region 2.

viscosity of the two phases is a dominant factor, with fluid elasticity and surface tension being either irrelevant or negligible features. This is an irreversible and relatively slow phenomenon, as opposed to an initial rapid interface adjustment due to volumetric accommodations analyzed recently by Everage (1975).

Theoretical explanations of the slow-envelopment phenomenon have been based upon minimizing energy dissipation, as suggested originally by Southern and Ballman. MacLean (1973), who considered planar layered flow, and Everage (1973), who studied a cylindrical geometry, both invoked a variational principle to show that a phase configuration with the high-viscosity component located centrally is favored over several other configurations. Those treatments, however, presumed that Δp was constant and the same in all cases and thus cannot be applied directly to the more realistic processing operation, wherein flow rates are controlled and Δp becomes a consequence of the evolving phase configuration. Furthermore, no simple closed-form relationships have ever been presented to permit assessment of the importance of viscosity ratio η_1/η_2.

We present here a very limited treatment of the important case of metered (constant) flow rates for two melts in stratified capillary flow. To illustrate the physics, we ignore the complex cases of evolving configurations (Figure 1*b*; also Everage, 1973) and focus exclusively on classes of concentric cylindrical interfaces which are presumably the steady states. We will show that the complex dependence of Δp on η_1/η_2 and interface position can be eliminated so that energy dissipation can be predicted in a compact exact form which is easily interpretable.

To isolate the essentials here, we will treat only Newtonian fluids and presume they are being metered together with equal flow rates. The problem will be posed initially as having an intermediate layer of viscosity η_2 between an inner core and outer sheath both of viscosity η_1. However, results are presented only for the limiting families of two-layered cases; no third layers have ever been observed.

For the geometry of Figure 2, with fully developed laminar flow $v_z = v(r)$, the z component of the equation of motion can be integrated once to give the shear stress $\tau = Kr$, where $K = \Delta p/2L$. This $\tau(r)$ distribution is independent of stratification, but K itself will vary with viscosity ratio and phase position.

Velocity profiles in regions 1, 2, and 3 can be obtained by a second integration by using boundary conditions of velocity matching at the two fluid interfaces, $v = 0$ at the wall and $dv/dr = 0$ at the center. Results are expressed in terms of dimensionless radius $R \equiv r/r_0$ and viscosity ratio $H \equiv \eta_1/\eta_2$:

$$v_1 = (Kr_0^2/2\eta_1)[1 - R^2 - (1-H)(R_2^2 - R_1^2)] \quad \ldots\ldots 0 \leq R \leq R_1 \quad (1)$$

$$v_2 = (Kr_0^2/2\eta_1)[1 - HR^2 - (1-H)R_2^2] \quad \ldots\ldots R_1 \leq R \leq R_2 \quad (2)$$

$$v_3 = (Kr_0^2/2\eta_1)[1 - R^2] \ldots\ldots R_2 \leq R \leq 1 \quad (3)$$

The integral $\dot{E} = \int \eta (dv/dr)^2 r dr$ is proportional to energy dissipation rate and can be computed for each of the three regions by use of Equations (1) to (3):

$$\dot{E}_1 \equiv \int_0^{r_1} \eta_1 (dv_1/dr)^2 r dr = K^2 R_1^4 r_0^4/4\eta_1 \quad (4)$$

$$\dot{E}_2 \equiv \int_{r_1}^{r_2} \eta_2 (dv_2/dr)^2 r dr = K^2 (R_2^4 - R_1^4) r_0^4 H/4\eta_1 \quad (5)$$

$$\dot{E}_3 \equiv \int_{r_2}^{r_0} \eta_1 (dv_3/dr)^2 r dr = K^2 (1 - R_2^4) r_0^4/4\eta_1 \quad (6)$$

By summing these, the total dissipation is given by

$$\dot{E} = (K^2 r_0^4/4\eta_1)[1 - (1-H)(R_2^4 - R_1^4)] \quad (7a)$$

It will be convenient initially to take $\eta_1 < \eta_2$ so that $0 < H \leq 1$, and to refer all results to the case of homogeneous flow ($H = 1$) of the low-viscosity fluid ($K = K_1$). Thus

$$\dot{E}(H)/\dot{E}_1 = [K(H)/K_1]^2 [1 - (1-H)(R_2^4 - R_1^4)] \quad (7b)$$

Evaluation of K/K_1 requires a statement about constraints on volumetric flow rate Q. Here, for a metered flow, we will simply take Q as a constant and postpone comment on the effect of relative flow rates of the two phases. Using $Q = 2\pi \int v r dr$, we obtain for the three regions

$$Q_1(2\eta_1/\pi K r_0^4) = R_1^2 - \frac{1}{2} R_1^4 - (1-H) R_1^2 (R_2^2 - R_1^2) \quad (8)$$

$$Q_2(2\eta_1/\pi K r_0^4) = (R_2^2 - R_1^2)[1 - (1-H)R_2^2] - \frac{1}{2} H (R_2^4 - R_1^4) \quad (9)$$

$$Q_3(2\eta_1/\pi K r_0^4) = 1 - (1-H)(R_2^4 - R_1^4) \quad (10)$$

Once again, this can be referred to single-phase flow (with

η_1), where Q is the same, in order to obtain the relative pressure drops:

$$K_1/K(H) = 1 - (1 - H)(R_2^4 - R_1^4) \qquad (11)$$

This result can be substituted into Equation (7*b*) to yield

$$\dot{E}(H)/\dot{E}_1 = 1/[1 - (1 - H)(R_2^4 - R_1^4)] \qquad (12)$$

Because this is valid for any Q, any relative phase flow rates, and any H, it is also valid for all special cases. Clearly, the relative flow rates of the two phases will influence the interface positions R_2 and R_1. To illustrate, we suppose that equal phase rates exist, so that $Q_2 = Q/2 = Q_1 + Q_3$. This leads to an equation for the position of one phase boundary as a function of the other's position:

$$(R_2^2 - R_1^2)[4 - (3R_2^2 - R_1^2) + H(R_2^2 - 3R_1^2)] - 1 = 0 \qquad (13)$$

It is important to note that this relationship cannot determine either R_1 or R_2 alone; presumably the proper choice would arise from selecting a configuration of minimum energy dissipation. Based on experimental observations, however, we will consider only the limiting cases for which only two layers remain (as in Figures 3*a* and *b*):

(a) Core has higher viscosity $\eta_2(R_1 \rightarrow 0,\ R_2 \rightarrow R_2^\dagger)$.

(b) Core has lower viscosity $\eta_1(R_2 \rightarrow 1,\ R_1 \rightarrow R_1^*)$.

From Equation (13) we obtain for these cases the corresponding interface positions:

$$(R_2^\dagger)^2 = [2 - (1 + H)^{1/2}]/(3 - H) \qquad (13a)$$

$$(R_1^*)^2 = [-2H + (H + H^2)^{1/2}]/(1 - 3H) \qquad (13b)$$

Note that case (a) achieves a limiting boundary position as the core becomes increasingly viscous; as $H \equiv \eta_1/\eta_2 \rightarrow 0$, then $R_2^\dagger \rightarrow 1/\sqrt{3} = 0.577$. This is the perfect plug-flow limit sketched in Figure 3*a*. Case (b), however, collapses in the $H \rightarrow 0$ limit to a situation of infinite channeling of the low-viscosity central core of vanishing diameter; this is represented in Figure 3*b*.

It remains to examine the behavior of $\dot{E}$ from Equation (12):

$$\dot{E}^\dagger/\dot{E}(1) = 1/\{1 - (1 - H) \cdot [5 + H - 4(1 + H)^{1/2}]/(3 - H)^2\} \qquad (14a)$$

$$\dot{E}^*/\dot{E}(1) = 1/\{1 - (1 - H) \cdot [1 - 7H + 4H^2 + 4(H + H^2)^{1/2}]/(1 - 3H)^2\} \qquad (14b)$$

Results for the limit $H \rightarrow 0$ are highly revealing:

$$\dot{E}^\dagger(0)/\dot{E}(1) \rightarrow 9/8 \qquad (15a)$$

$$\dot{E}^*(H)/\dot{E}(1) \rightarrow 0.5/H \rightarrow \infty \qquad (15b)$$

Equation (15*b*) is a consequence of having infinitely high shear rates in the low-viscosity central core which undergoes infinite channeling. Equation (15*a*) shows dramatically why the energetics of this problem favor the more viscous core case. No matter how severe the mismatch of viscosities, the maximum penalty paid by introducing the more viscous component is only 12.5% (for equal phase flow rates).‡ Complete results for cases (b) and (c) are given in Figure 4. For every H, it is clear that case (a) is energetically favored.

More detailed analysis of all mechanical forces will be required (for example, White and Lee, 1975) to describe the time evolution of these systems to the postulated steady states. Still, the present analysis suggests that phase migration can be thought of as driven by the search to minimize energy dissipation; if so, the kinetics of the phase rearrangement might be represented by $dE/dt = -k(\dot{E} - \dot{E}^\dagger)$, where $k(H)$ increases as H decreases. This would predict an exponentiallike approach to the stable configuration, accelerated as viscosity mismatch becomes more severe. Indeed, data of Lee and White (1974) seem consistent with this, although direct comparison is impossible because of differing basic conditions. They show an exponentiallike approach to the state of complete encapsulation as L/D (that is, time measured for a fluid element) increases, with the approach more rapid as H decreases (~100% complete for $H = 0.024$ but only ~50% complete for $H = 0.29$, at $L/D = 20$).

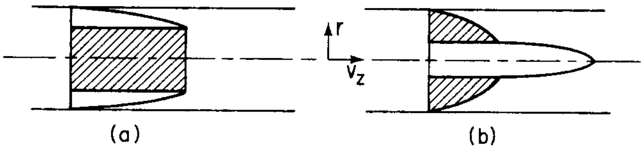

Fig. 3. Limiting two-region cases of hypothetical three-region flow. *a*. Higher-viscosity fluid in the center, shown as the plug-flow limit H → 0. *b*. Lower-viscosity fluid in the center.

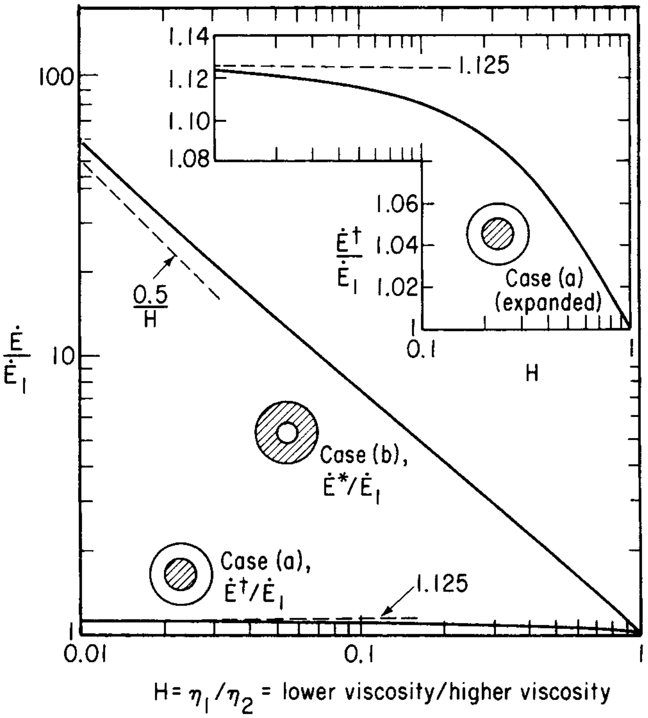

Fig. 4. Dependence of energy dissipation rate on phase viscosity ratio $H = \eta_1/\eta_2$, for both limiting phase arrangements. The semi-log inset figure expands $\dot{E}^\dagger/\dot{E}_1$.

NOTATION

$\dot{E}$ = $\int \eta (dv/dr)^2 r\,dr$, proportional to within the volume rate of viscous energy dissipation
H = η_1/η_2, ratio of viscosities of two liquid phases
K = $\Delta p/2L$
k = effective rate coefficient, characterizing a first-order process seeking a minimum energy dissipation state
L = length of die over which Δp is measured
p = pressure (Δp = pressure drop over length L)
Q = volumetric flow rate

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‡ The same physics explains why the introduction of water into an oil pipeline is repaid by such great reductions in ΔP for oil transport.

r = radial position
R = r/r_0, dimensionless position
v = velocity
γ = dv_z/dr, velocity gradient
η = Newtonian viscosity
τ = τ_{rz}, shear stress

Subscripts

1, 2, 3 = regions of flow, identified in Figure 2 (used as v_i, Q_i, $\dot{E}_i$), or defining interfaces (as R_i), or identifying a phase (as η_i or K_1)
0 = outer boundary, wall of cylinder die

Superscripts

† = associated with limiting case having only two regions, with higher viscosity (η_2) in central core
* = associated with limiting case having only two regions, with lower viscosity (η_1) in central core

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Light Distribution in Cylindrical Photoreactors

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In a communication published in the *AIChE Journal*, Matsuura and Smith (1970) proposed a partially diffused-light model in which they derived the radial distribution of light intensities. Because of the interest in photochemical reactors during recent years and of the scarcity of fundamental studies in photochemical reaction engineering, the paper has been cited as a reference (Markl and Vortmeyer, 1975).

Unfortunately, it appears that their derivation is ambiguous because of the choice of a reference intensity at the wall $I_{w,\lambda}$ depending on the way light rays are distributed within the reactor.

The purpose of the present communication is to give a more rigorous derivation of the radial intensity distribution in the partially diffuse light model and to point out which results of Matsuura and Smith (MS) have to be revised.

It must first be noticed that a regrettable confusion is prevailing about the exact meaning of the word intensity in the photochemical literature. We recall below the more useful definitions, conserving MS's notations as far as possible (following their assumptions, we consider a two-dimensional problem in a plane perpendicular to the cylindrical reactor axis):

J = photometric intensity = einsteins/s/radian or W/radian. Let $d\phi$ be the light power received within an angle $d\omega$; then $J = d\phi/d\omega$. In the case of an isotropic light emission, the total power output of the lamp is $\Phi = 2\pi J$ (J = intensity in the absence of adsorption).

I = intensity at a point P within the reactor : einsteins/s/cm^2 or W/cm^2. I is the total photon flux received at P per unit surface area normal to the rays whatever the direction of the incident rays (MS).

I_0 = intensity at the wall of the reactor = einsteins/s/cm^2 or W/cm^2. I_0 is the photon flux which passes through a fixed unit area of the reactor's wall. It must be noticed that I and I_0 do not have the same meaning.

I_a = photochemical absorbed intensity = einsteins/s/cm^3 or W/cm^3. I_a is in fact an absorbed power per unit volume at P (MS). As a consequence of Beer's law, $I_a = \mu I$.

$I_{w,\lambda}$ = intensity at the wall in MS' text is such that in the absence of absorption, $I = 2\, I_{w,\lambda}$ at the wall. As will be shown below, this definition is ambiguous.

We may now pass on to the correct derivation of the partially diffuse light model. The notations are those of MS' paper to which the reader is asked to refer.

According to the basic assumption of the model, the reactor is irradiated by isotropic light beams having a certain breadth $2R_2$ ($R_2 < R_1$) and centered on the axis of the reactor (see Figure 1). Assume the photometric intensity J to be constant in the incident beams.

In the absence of absorption, the intensity at P per unit length of reactor would be, in the direction (ϕ, $\phi + d\phi$)

$$dI = J d\phi / 2R_2 \qquad (1)$$

The denominator takes into account the fact that the power is displayed into a beam of section $2R_2$.

We may now follow MS's treatment. Owing to absorption according to Beer's law, the intensity at P is in fact

$$dI = \frac{J d\phi}{2R_2}\left(e^{-\mu x'} + e^{-\mu x''}\right) \qquad (2)$$

where

$$x' = -r\cos\phi + (R^2 - r^2\sin^2\phi)^{1/2} \qquad (3)$$