g = gravitational acceleration, m/s^2
H = height of bed expansion, m
$\bar{H}$ = H/H_I
H_I = initial bed height, m
k = reaction rate constant based on the volume of catalysts, 1/s for the first-order reaction and m^3/s/kg-mole for the second-order reaction
n = order of chemical reaction, —
N_d = number of orifice openings on the distributor, —
Pe_i = Peclet number in phase i, $U_{i0}H/E_{i0}$
Pe_{i0} = $U_{i0}H_I/E_{i0}$
t = time, s
U_i = linear gas velocity in phase i, m/s
$\bar{U}_i$ = U_i/U_{i0}
U_{i0} = linear gas velocity in phase i evaluated just above the distributor, m/s
U_{is} = superficial gas velocity in phase i, m/s
U_{mf} = minimum fluidization velocity, m
U_0 = superficial gas velocity, m/s
x = axial distance from the distributor, m
$\bar{X}$ = overall conversion of the gaseous reactant, —

Greek Letters

δ_i = volume fraction of the bed occupied by phase i, —
ϵ_{mf} = void fraction in the bed at U_{mf}, —
ϵ_{ig} = volume fraction of gas in phase i, based on δ_i, —
ϵ_{is} = volume fraction of solid in phase i, based on δ_i, —
ϵ_I = void fraction at the initial condition, —
ϕ = $\dfrac{kHC_0^{n-1}}{U_0}$
ξ = x/H
ϕ = tU_0/H

Subscripts

i = 1 for the bubble phase, 2 for the cloud wake phase, 3 for the emulsion phase
g = gas
s = solid

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The Effect of an Expanded Section on Slugging

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Slugging experiments were performed in a semicircular Plexiglas column 5.6 m in height to study the effect of an expanded section on slugging. The bottom section of the column (3.05 m long) is 28.6 cm in inside diameter and the expanded section 40.6 cm. Both the bed height and the fluidizing velocity were varied during the study, and the results were successfully correlated with a theoretically derived equation. The implications in designing a bed with an expanded section to reduce the slugging effectively are also discussed.

The slugging phenomenon in fluidized beds has been extensively studied and is described in detail by Davidson and Harrison (1971). A slugging bed is characterized by gas slugs of sizes close to reactor cross section that rise at regular intervals and divide the main part of the fluidized bed into alternate regions of dense and lean phases. The passage of these gas slugs produces large pressure fluctuations inside the fluidized bed. The occurrence of slugging is usually accompanied by deterioration in quality of bed mixing and gas solids contacting. According to Stewart (1965), slug flow will occur in gas fluidized beds when $(U - U_{mf})/0.35\sqrt{gD}$ is greater than about 0.2 with $H/D > 1$. Thus, slugging generally occurs in reactors of laboratory and pilot plant scale. An effective way to suppress the slugging and to reduce the maximum slugging bed height is to expand the bed cross-sectional area toward the top of a fluidized-bed reactor. No information is available in the literature, however, on how to design an expanded section for this purpose. This problem was studied both theoretically and experimentally. The theoretical model and initial slugging results are presented here.

DEVELOPMENT OF A PREDICTIVE MODEL

The development of this model follows closely that of Matsen et al. (1969) for expansion of fluidized beds in slug flow. Before any slugs reach the bed surface, the bed height will increase at different velocities in the bottom section and in the expanded section at different stages of development. The bed height will reach the maximum when the first slug breaks the surface. Thereafter, the bed height will fluctuate between this maximum and a minimum position that depends on the size of the gas slug. The expansion of a slugging fluidized bed with an expanded section is divided into three different stages (as shown in Figure 1) in the present development. For the time being, the static bed height is assumed to be lower than the height of the bed transition; that is, $H_{mf} < h$.

First-Stage Development (See Figures 1*a* and *b*)

The first-stage development starts from the time the first gas slug is formed and lasts until the bed surface reaches the height of the bed transition. In this stage, the fluidized-bed expansion is similar to that in a bed of uniform cross section. The time required for the bed surface to reach h is $(h - H_{mf})/(U_1 - U_{mf})$. During this time, the first gas slug will travel over a distance of

$$[(U_1 - U_{mf}) + U_{B1}]\cdot\frac{(h - H_{mf})}{(U_1 - U_{mf})} = (h - H_{mf})\left[1 + \frac{U_{B1}}{(U_1 - U_{mf})}\right] \quad (1)$$

where $(U_1 - U_{mf})$ is the velocity of bed height increase in the bottom section, and U_{B1} is the relative velocity between the gas slug and the surrounding solids. The bed surface is now at h.

Second-Stage Development (See Figures 1*b* and *c*)

The second-stage development starts at the end of the first-stage development and ends when the first gas slug reaches the bed transition height h. At the end of the first stage, the first gas slug is at a height expressed by Equation (1). The distance of the first gas slug from the transition is then

$$h - (h - H_{mf})\left[1 + \frac{U_{B1}}{(U_1 - U_{mf})}\right] = H_{mf} - (h - H_{mf})\frac{U_{B1}}{(U_1 - U_{mf})} \quad (2)$$

Since the absolute gas slug velocity is known to be $U_{B1} + (U_1 - U_{mf})$, the time required for the first slug to reach the transition can be calculated as

$$\frac{H_{mf}}{U_{B1} + (U_1 - U_{mf})} - \frac{(h - H_{mf})}{U_{B1} + (U_1 - U_{mf})}\cdot\frac{U_{B1}}{(U_1 - U_{mf})} = \frac{H_{mf}\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right] - h}{\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right](U_1 - U_{mf})} \quad (3)$$

During this time, the bed will expand into the expanded section. Two assumptions can be made here about the rate of increase of bed height in the expanded section. On the basis of purely physical considerations, the bed height can be expected to increase at a rate of $(D_1/D_2)^2 (U_1 - U_{mf})$ in the expanded section as opposed to $(U_1 - U_{mf})$ in the bottom section. In this case, the gas leakage from the gas slug to the surrounding emulsion phase in the expanded section is assumed to be negligible (case I). On the other hand, the gas leakage from the gas slug to the surrounding emulsion phase in the expanded section can be assumed to be instantaneous so that the emulsion phase in the expanded section is always minimally fluidized. In this case (case II), the rate of increase in bed height will be $(U_2 - U_{mf})$. The advancement of bed height during this time period can be calculated for both cases to be

$$\frac{H_{mf}\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right] - h}{\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right]}\cdot\left(\frac{D_1}{D^2}\right)^2 \quad \text{for case I} \quad (4)$$

$$\frac{H_{mf}\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right] - h}{\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right]}\cdot\frac{(U_2 - U_{mf})}{(U_1 - U_{mf})} \quad \text{for case II} \quad (5)$$

Third-Stage Development (See Figures 1c and *d*)

The final stage of development will proceed until the first gas slug breaks the bed surface. The gas slug will travel with a relative velocity of U_{B2} with respect to the surrounding solids through a bed height in the expanded section expressed in Equations (4) and (5). The advancement of bed height during this time period can be determined to be

$$\frac{H_{mf}\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right] - h}{\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right]}\cdot\left(\frac{D_1}{D_2}\right)^4\cdot\frac{(U_1 - U_{mf})}{U_{B2}} \quad \text{for case I} \quad (6)$$

$$\frac{H_{mf}\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right] - h}{\left[1 + \dfrac{(U_1 - U_{mf})}{U_{B1}}\right]}\cdot\frac{(U_2 - U_{mf})^2}{U_{B2}(U_1 - U_{mf})} \quad \text{for case II} \quad (7)$$

Maximum Bed Height

The maximum bed height is thus the addition of three separate bed heights advanced during the three stages

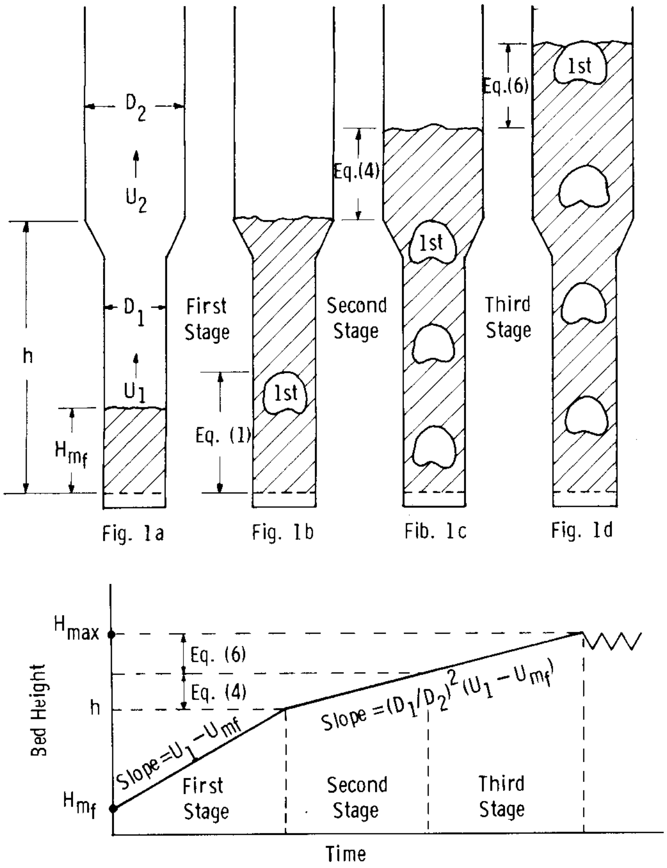

Figure 1. Three-stage development of maximum slugging bed height with an expanded section.

of development [that is, h plus Equations (4) and (6) for case I and h plus Equations (5) and (7) for case II]. The resulting equations are

$$\frac{H_{\max}}{H_{mf}} = \frac{h}{H_{mf}} + \left(\frac{D_1}{D_2}\right)^2 \left[1 + \left(\frac{D_1}{D_2}\right)^2 \frac{(U_1 - U_{mf})}{U_{B2}}\right] \cdot \left\{1 - \frac{h}{H_{mf}} \cdot \frac{1}{\left[1 + \frac{(U_1 - U_{mf})}{U_{B1}}\right]}\right\} \quad \text{for case I} \quad (8)$$

$$\frac{H_{\max}}{H_{mf}} = \frac{h}{H_{mf}} + \frac{(U_2 - U_{mf})}{(U_1 - U_{mf})} \left[1 + \frac{(U_2 - U_{mf})}{U_{B2}}\right] \cdot \left\{1 - \frac{h}{H_{mf}} \cdot \frac{1}{\left[1 + \frac{(U_1 - U_{mf})}{U_{B1}}\right]}\right\} \quad \text{for case II} \quad (9)$$

When $D_1 = D_2$, $U_1 = U_2$, and $U_{B1} = U_{B2}$, Equations (8) and (9) reduce to Equation (10), the slugging bed height equation for a bed with uniform cross section:

$$\frac{H_{\max}}{H_{mf}} = 1 + \frac{(U_1 - U_{mf})}{U_{B1}} \quad (10)$$

For the situation where $H_{mf} > h$, similar equations can be derived as follows:

For case I

$$H_{\max} = H_{mf} + \frac{h}{U_{B1}} \cdot \left(\frac{D_1}{D_2}\right)^2 (U_1 - U_{mf}) + \frac{(H_{mf} - h)}{U_{B2}} \left(\frac{D_1}{D_2}\right)^2 (U_1 - U_{mf}) \quad (11)$$

or

$$\frac{H_{\max}}{H_{mf}} = 1 + \left(\frac{D_1}{D_2}\right)^2 \left[\frac{h}{H_{mf}} \cdot \frac{(U_1 - U_{mf})}{U_{B1}} + \left(1 - \frac{h}{H_{mf}}\right) \cdot \frac{(U_1 - U_{mf})}{U_{B2}}\right] \quad (12)$$

For case II

$$\frac{H_{\max}}{H_{mf}} = 1 + \left[\frac{h}{H_{mf}} \cdot \frac{(U_2 - U_{mf})}{U_{B1}} + \left(1 - \frac{h}{H_{mf}}\right) \cdot \frac{(U_2 - U_{mf})}{U_{B2}}\right] \quad (13)$$

EXPERIMENTAL APPARATUS

The experiments were conducted in a semicircular Plexiglas column 5.6 m high, as shown in Figure 2. The bottom section (3 m long) was 28.6 cm in inside diameter and the expanded section 40.6 cm in inside diameter. The transition between the two sections was a conical section with a 60 deg included angle. The distributor plate was located so that h = 2.8 m. The distributor plate used

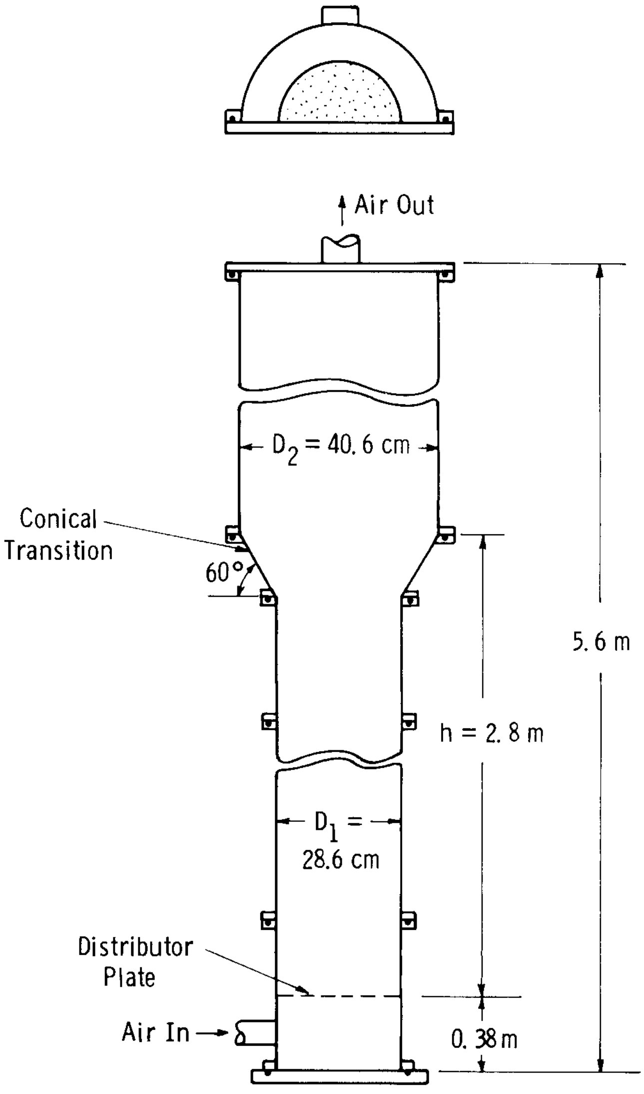

Figure 2. Schematic of the semicircular Plexiglas column.

was an 1.3 cm thick steel plate with 128 holes, 1.57 mm in diameter. The bed material was −20 +50 mesh Ottawa sand with an average particle size of 750 μm.

EXPERIMENTAL RESULTS

Seven different bed heights were used, ranging from an H_{mf}/D ratio of 4.5 to 8.8. The cross-sectional area of the expanded section in the present configuration is twice the area of the bottom section. The experiments were carried out by increasing the gas flow slowly to determine the minimum fluidization point and the maximum slugging bed height at each flow rate. Both the slugging bed heights in the bottom section and in the expanded section were recorded visually and by high speed movies.

The maximum slugging bed heights in the bottom section (when $H_{max} < h$) could be successfully correlated with the equation for slugging bed heights in a bed of uniform cross-sectional area [Equation (10)]; if the bubble velocity is taken to be $0.35\sqrt{2gD_1}$, the bubble velocity corresponds to a wall slug rather than to an ideal slug. The gas bubbles rising against the wall (wall slug) rather than in the middle of the column (ideal slug) were experimentally observed:

$$\frac{H_{max}}{H_{mf}} = 1 + \left(\frac{U_1 - U_{mf}}{0.35\sqrt{2gD_1}}\right) \quad H_{max} < h \quad (14)$$

The data are shown in Figure 3. Note that the diameter of the semicircular column rather than the hydraulic diameter is employed here.

The maximum slugging bed heights in the expanded section where $H_{max} > h$ and $h > H_{mf}$ were correlated with both Equations (8) and (9) for different values of U_{B1} and U_{B2}. The best correlation was obtained by using Equation (8) and by assuming $U_{B1} = U_{B2} = 0.35\sqrt{gD_1}$. The comparison with the experimental data is shown in Figure 4. Slightly worse results were obtained with Equation (8) and $U_{B1} = 0.35\sqrt{gD_1}$, $U_{B2} = 0.35\sqrt{gD_2}$. The assumptions that $U_{B1} = 0.35\sqrt{gD_1}$ and $U_{B2} = 0.711\ g^{1/2}V^{1/6}$ [where $V = (\pi/6)D_1^3$] or $U_{B1} = 0.35\sqrt{2gD_1}$ and $U_{B2} = 0.711\ g^{1/2}V^{1/6}$ gave much poorer results. This led to the conclusion that the gas leakage from the gas slug to the emulsion phase at the expanded section is very small and the emulsion phase at the expanded section is actually less than minimally fluidized. By the time the bed surface expands into the expanded section, the bed tends to defluidize and causes the bridge formation at the transition section. On the basis of experimental observation, the bed in the expanded section tends to bridge at the conical transition between the bottom section and the expanded section, especially at high bed heights. The gas slugs rising up in the bottom section tend to be trapped under the bridge and are periodically released as big ideal bubbles.

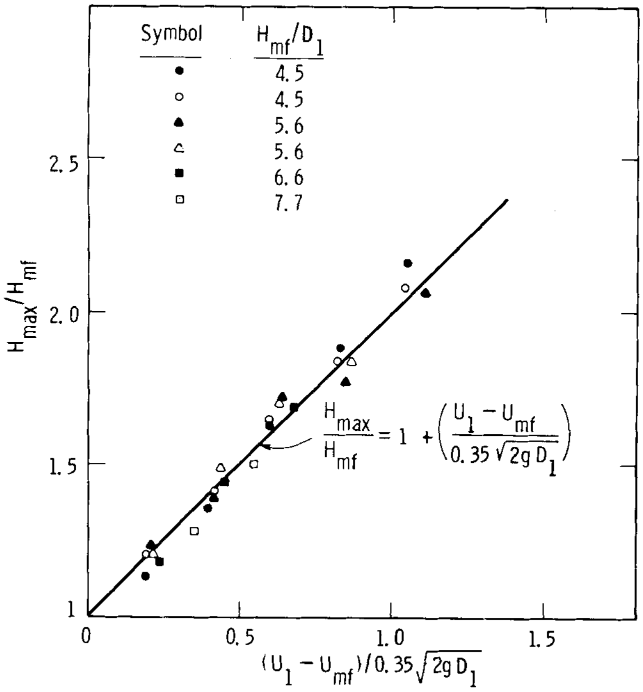

Figure 3. Maximum slugging bed height in a semicircular column ($H_{max} < h$).

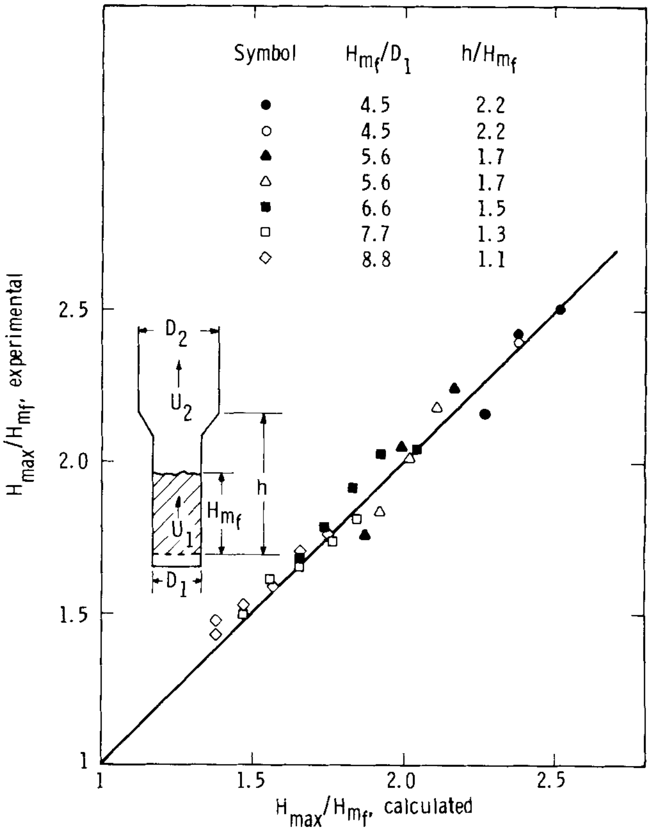

Figure 4. Comparison of experimental and calculated slugging bed heights using Equation (8) and $U_{B1} = U_{B2} = 0.35\sqrt{gD_1}$.

CONCLUSIONS

On the basis of initial experimental evidence, the expanded section will be effective in reducing slugging. Care must be exercised, however, in locating the expanded section at a proper distance from the gas distributor plate or to provide aeration at the transition to reduce the bridging tendency at the conical transition. A predictive model was developed that successfully projects the maximum slugging bed heights observed in the expanded section. In correlating the data obtained in a semicircular column, the diameter of the semicircular column rather than its hydraulic diameter should be used.

NOTATION

D = diameter of the vessel
g = gravitational acceleration

h = height of the conical transition from the distributor plate
H = bed height
H_{max} = maximum slugging bed height
U = superficial gas velocity
U_{mf} = superficial minimum fluidization velocity
U_B = bubble velocity

Subscripts

1 = bottom section
2 = expanded section

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Determination of Diffusion Properties of Impervious Layer of a Double Layered Catalyst Pellet

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Deposition of contaminants on the outer surface of a catalyst pellet sometimes results in the formation of impervious or near impervious outer layer. Examples are given in an article by Chou and Hegedus (1978). Characteristics of this impervious layer of the double layered catalyst pellet are such that the thickness of the impervious layer is very small and the porosity very low. As pointed out by Chou and Hegedus, it is very difficult to accurately measure the thickness and porosity of this impervious layer. In order to determine the diffusivities of two layers of a catalyst pellet, Chou and Hegedus (1978) solved on the Laplace domain mass balance equations for each of the two layers of catalyst pellet and a conservation equation for a pellet string reactor. The diffusivities were then determined by minimizing the error between the theoretical frequency response and the experimental frequency response obtained through pulse tests. The thickness and porosity of the near impervious layer independently determined were used for accurate determination of the diffusivities. However, they found that the results are insensitive to the porosity.

In this note, it is shown that the single-pellet chromatography of Smith and co-workers (Suzuki and Smith, 1972; Dogu and Smith, 1975) leads to straightforward determination of the diffusion properties of the impervious layer, such as diffusivity and porosity, and the thickness of the impervious layer. It is also shown that diffusivity of the impervious layer can be determined without the knowledge of thickness and porosity of the impervious layer. We will treat the general case where the retention time for the impervious layer is of the same order of magnitude as that for the unobstructed layer. This general case can be reduced to that of Chou and Hegedus (1978), where the retention time for the near impervious layer is much smaller than that for the unobstructed layer. Diffusion in catalysts with a bipore distribution was studied using transient response by Haynes and Sarma (1973) and Ma and Lee (1976).

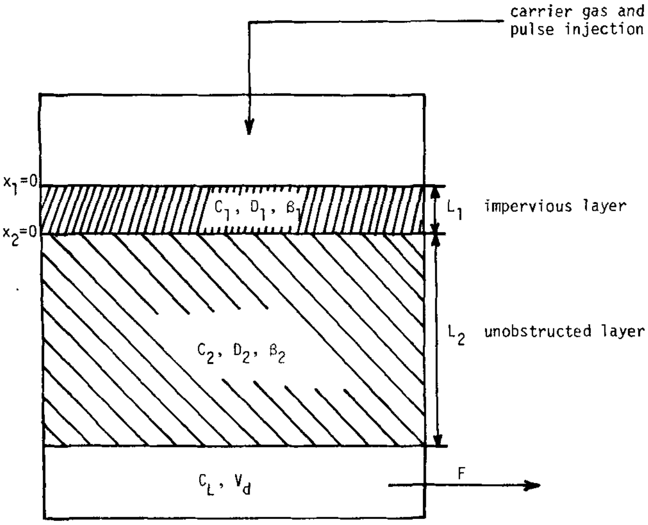

Figure 1. Dimensions and conditions of diffusive cell.

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SINGLE-PELLET CHROMATOGRAPHY FOR DOUBLE LAYERED CATALYST PELLETS

Dimension and conditions of a double-layered, single-pellet, diffusion cell are shown in Figure 1. For a non-adsorbed gas, the conservation equation applied to each of the layers can be written in terms of dimensionless variables as follows:

$$\frac{\partial^2 C_1}{\partial z_1^2} = \phi_1^2 \frac{\partial C_1}{\partial t} \tag{1}$$

$$\frac{\partial^2 C_2}{\partial z_2^2} = \phi_2^2 \frac{\partial C_2}{\partial t} \tag{2}$$

where

$$z_i = x_i/L_i \tag{3}$$

$$i = 1, 2$$

$$\phi_i^2 = \beta_i L_i^2/D_i \tag{4}$$

The boundary and initial conditions are (see Figure 1)

$$C_1(0, t) = M\delta(t) \tag{5}$$

$$C_1(1, t) = C_2(0, t) \tag{6}$$