R & D NOTES

Design Criterion for Radial Flow Fixed-Bed Reactors

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The radial-flow packed-bed reactor (RFBR) is currently being used for a variety of industrial applications. Synthesis of ammonia, nitric oxide conversion, reforming and desulphurization are just a few of the processes in which a fluid must contact solid particles at a high space velocity, thereby suggesting radial flow as a design possibility.

Earlier analytical work on the RFBR concentrated on the effects of radial-flow direction on reaction conversion. Dudokovic and Lamba (1975) used a singular perturbation approach to demonstrate that reactor performance for second- or higher-order reactions is influenced by flow direction. Balakotaiah and Luss (1981) extended the treatment to the general case. Hlavacek and Kubicek (1972) pointed out that the effect of flow direction may be more significant under nonisothermal conditions.

Calo (1978) and Chang and Calo (1978) used a numerical cell model to simulate the difference in reactor performance due to flow direction. They found that although a difference exists, it is not significant. Ponzi and Kaye (1979) were the first to show that imperfect radial-flow profiles have a much larger influence on performance than flow direction. This was confirmed by a more general and analytical approach by Chang and Calo (1981) who also pointed out the large deviation of real systems from the perfect flow profile often assumed. Using the fluid mechanical model developed by Raskin et al. (1968), Genkin et al., (1973) and the experimental data of Kaye (1978), significant maldistribution was found. In general, for most systems a perfect distribution yields the highest conversion. From the numerical study of Chang and Calo (1981), it is apparent that this optimum profile can be achieved by adjusting the reactor dimensions so that the radial pressure drop remains independent of the axial coordinate. Since such an optimum exists, it represents a design criterion for radial-flow reactors.

In this study, asymptotic methods are used to obtain analytical solutions of the model of Genkin et al. (1973). It is then shown that an optimum solution exists and the relationship among the system parameters for the optimum profile is presented. This relationship is then the desired design criterion. However, due to the implicit assumptions of an asymptotic solution, the results are strictly valid only for small values of certain parameters. Numerical integration is then carried out to demonstrate the general validity of the derived criterion even at larger parameter values.

ANALYSIS

Four flow configurations are possible for a RFBR (e.g., Chang and Calo, 1978). The flow direction inward (centripetal) designated "CP" or outward (centrifugal) designated "CF." In addition, the flows in the center pipe and the outer annulus can be in the same (Z) or opposite (π) directions. It was established by Chang and Calo (1978, 1981) that the π-flow configuration is always superior to the z-flow configuration. Consequently, only the CP-π and CF-π configurations will be investigated here. Following the approach of Genkin et al (1973), the dimensionless macroscopic axial velocity in the center pipe must satisfy the dimensionless equation (Chang and Calo, 1978 for derivation and notation):

$$w'w'' + \epsilon w w' \pm \delta w^2 = 0 \tag{1}$$
$$w(0) = 0$$
$$w(1) = 1$$

where w is the dimensionless axial velocity and the $\pm$ term in Eq. 1 denotes CP and CF flow, respectively. The dimensionless parameters are related to the geometry and operating conditions:

$$\epsilon = -\frac{3}{2\sigma}(\xi^2 - 1) \tag{2}$$

$$\delta = \left[\frac{R_o}{R_o^2 - r_2^2}\xi^2 + \frac{1}{r_1}\right] fl/\sigma \tag{3}$$

where

$$\sigma = \frac{b^* r_1^3 (1 - r_1/r_2)}{\psi^3 l^2} \tag{4}$$

and

$$\xi = \frac{r_1^2}{R_o^2 - r_2^2} \tag{5}$$

ξ is the ratio of the center pipe cross-sectional area to the outer annulus cross-sectional area. Qualitatively, σ represents the relative resistance to flow offered by the catalyst basket. The dimensionless parameter ϵ is then the ratio of the difference in flow between the center pipe and the outer annulus to the bed resistance. The parameter δ is the ratio of the center pipe and outer annulus flow channels resistance to the bed resistance.

The radial velocity is related to the first derivative of w by a macroscopic momentum balance (Chang and Calo, 1978):

$$v = \pm \frac{r_1}{2\phi_1 l} w' \tag{6}$$

Consequently, for a perfect radial flow profile, w is linearly dependent on z, and from the boundary conditions of Eq. 1:

$$w_{\text{ideal}} = z \tag{7}$$

By inspection, it is clear that the optimum profile is obtained when $\epsilon = \delta = 0$. The qualitative justification is as follows: with no channel resistance and the cross sectional areas equal, the pressure in the feed channel increases in the flow direction (due to the momentum loss through fluid entering the basket) at the same rate as the pressure in the exit channel decreases in its flow direction. Due to the opposed flow configuration of the π-flow, the pressure drop across the bed remains independent of the axial coordinate, thereby giving rise to a uniform radial profile.

The problem arises when the reactor has a finite resistance in the open channels, yielding a nonzero δ. One must then choose an ϵ appropriately to achieve uniformity. Fortunately, however,

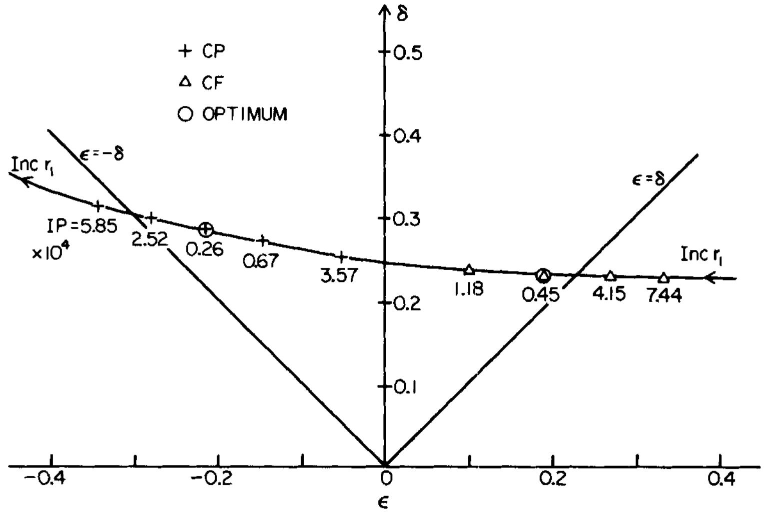

Figure 1. Optimum vs. predicted values for CP and CF flows: standard parameter values, varying r_1.

channel resistance is often small compared to bed resistance [the parameters used by Chang and Calo (1981) yield $\delta = 0.2$]. Since an optimum is achieved when both vanish, one expects that the magnitude of ϵ which yields an optimum for small δ will also be small. This allows an asymptotic approach to the problem based on the order of magnitudes of ϵ and δ.

Expanding w in a two-parameter series in ϵ and δ,

$$w = w_o(z) + \epsilon w_{11}(z) + \epsilon^2 w_{12}(z) + \dots + \delta w_{21}(z) + \delta^2 w_{22}(z) + \dots \quad (7)$$

Substituting the expression into Eq. 1, collecting terms, and applying the boundary conditions, one obtains

$$w \cong z - \frac{z}{6}(z^2 - 1)(\epsilon \pm \delta) \quad (8)$$

where the $\pm$ corresponds to CP flow and CF flow, respectively. The second term in Eq. 8 represents the deviation from uniformity. Consequently, if

$$\begin{aligned} \epsilon &= -\delta \text{ for CP} \\ \epsilon &= \delta \text{ for CF} \end{aligned} \quad (9)$$

Then the flow is uniform up to second order in ϵ and δ, therefore, Eq. 9 represents the desired design criterion. (Note that negative values for ϵ can be obtained, but δ must remain positive-definite).

NUMERICAL VERIFICATION

In this section, the results of the previous analysis are verified numerically. To this end, the following measure is defined to gauge the deviation from optimality,

$$\mathrm{IP} = \int_0^1 (w' - 1)^2 dz \quad (10)$$

Instead of integrating Eq. 1 over the entire $\epsilon - \delta$ parameter spece, the radius of the center pipe (r_1) was chosen as a suitable design parameter. Since ϵ and δ are both functions of r_1, a locus in the $\epsilon - \delta$ plane is described if one varies r_1 while holding all other parameters constant. The following values (Chang and Calo, 1981) are used here:

$r_2 = 0.10$ m	$\psi = 0.42$
$R_o = 0.12$ m	$f = 0.004$
$l = 1.0$ m	$b^* = 1{,}327\ \mathrm{m}^{-1}$

Equation 1 is then solved numerically using Euler's method and IP is evaluated by Eq. 10 at each point. The locus is shown in Figure 1 with the optimal location for both CP and CF flow indicated. The agreement with predicted values is quite good. In Figure 2, the same procedure is carried out for four different values of porosity (ψ), for the CP flow. Note that agreement worsens with increasingly large ϵ and δ, as expected from the asymptotic origin of Eq. 9. However, even for values approaching unity, Eq. 9 is still a good approximation for the optimum condition. In Figure 3, the same procedure is carried out while the diameter of the outer annulus is varied. Again, very good agreement with theory is noted.

CONCLUSION

A design criterion for radial flow fixed bed reactors is obtained by solving the relevant fluid mechanics by asymptotic methods.

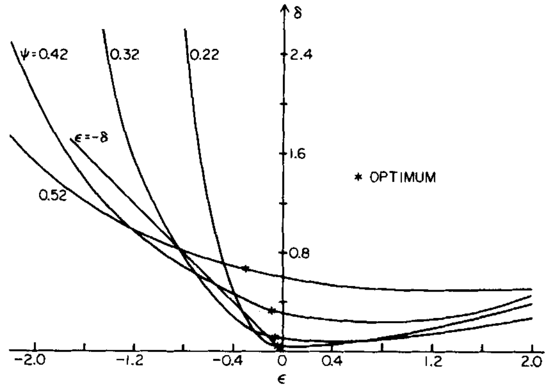

Figure 2. Optimum vs. predicted values for CP flow for various values of porosity.

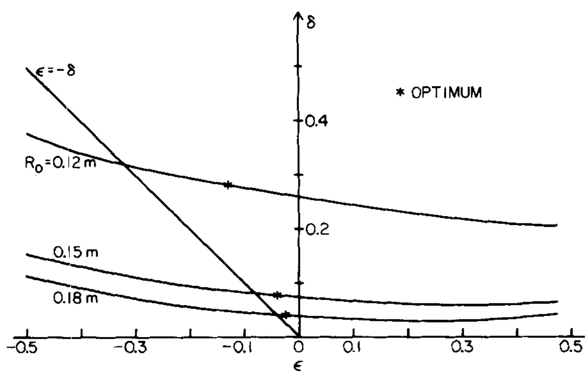

Figure 3. Optimum vs. predicted values for CP flow for various radius of outer annulus.

The criterion is shown to be valid for small ϵ and γ, which is common in practice. Given the constraints of a particular reactor, Eq. 9 allows one to optimize one or more free parameters to achieve a uniform radial flow distribution.

NOTATION

b^* = friction coefficient in bed
f = friction factor in flow channels
l = height of reactor
R_o = outer radius of outer annulus
r_2 = outer radius of catalyst basket
r_1 = inner radius of catalyst basket
v = dimensionless radial velocity
w = dimensionless axial velocity
ψ = catalyst porosity
ϕ_1 = fraction free surface area of screen of catalyst basket

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Method for Estimation of Second Virial Coefficients from Pressure Measurements Alone

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INTRODUCTION

The compressibility factor may be expressed in terms of the virial expansion

$$z = 1 + nB/V + n^2C/V^2 + \dots \tag{1}$$

In moderate pressure operations virial coefficients after the second may be neglected.

A procedure for estimation of the second virial coefficient described by Couldwell et al. (1978) has the advantage that three pressures only need be measured but requires quantitative transfer of material by condensation as well as requiring estimation of the number of moles of material. The procedure described here requires pressure measurement only and is not restricted to condensible substances.

EXPERIMENTAL METHOD

In Figure 1, schematic representation of the apparatus, the quantity of gas to be studied, n moles, is first loaded into volume V_1 at the selected pressure, p_1. This pressure is measured via the null differential pressure gauge using a manometer or dead weight gauge. The gas is then expanded into the previously evacuated vessel V_2, and the pressure p_{12} measured. Tap T_2 is closed and the gas remaining, n', is allowed to expand into previously evacuated vessel V_3. The pressure p_{13} is measured, and then tap T_2 opened allowing the total quantity of gas access to volume $V_1 + V_2 + V_3$ at a pressure of p_{123}.

Employing the pressure series virial expansion truncated after the third virial coefficient, we have for each of the four pressures the following relations:

$$\frac{V_1}{n} = \frac{RT}{p_1} + B + C'p_1 \tag{2}$$

$$\frac{V_1 + V_2}{n} = \frac{RT}{p_{12}} + B + C'p_{12} \tag{3}$$

$$\frac{V_1 + V_2 + V_3}{n} = \frac{RT}{p_{123}} + B + C'p_{123} \tag{4}$$

$$\frac{V_1 + V_3}{n'} = \frac{RT}{p_{13}} + B + C'p_{13} \tag{5}$$

Now since the ratio of the numbers of moles n' and n is equal to the ratios of the volumes V_1 and $V_1 + V_2$, we have

$$n' = n\frac{V_1}{V_1 + V_2} \tag{6}$$

and Eq. 5 becomes

$$\frac{(V_1 + V_3)(V_1 + V_2)}{nV_1} = \frac{RT}{p_{13}} + B + C'p_{13} \tag{7}$$

The relation which results from substitution of the volumes V_1, V_2 and V_3 in Eq. 7 with the volumes given by Eqs. 2, 3 and 4 may be arranged to give the following expression for B.

$$\begin{aligned}\frac{B}{RT} = &\left\{\frac{1}{p_{12}}\left(\frac{1}{p_{123}} - \frac{1}{p_{12}}\right) - \frac{1}{p_1}\left(\frac{1}{p_{13}} - \frac{1}{p_{12}}\right)\right\}\Big/\left\{\frac{1}{p_{13}} - \frac{1}{p_{123}}\right\} \\ &+ \frac{C'}{RT}\left\{p_1\left(\frac{1}{p_{13}} - \frac{1}{p_{12}}\right) - p_{12}\left(\frac{1}{p_{123}} + \frac{1}{p_1}\right)\right. \\ &\left. + \frac{p_{13}}{p_1} - \frac{p_{123}}{p_{12}} + 2\right\}\Big/\left\{\frac{1}{p_{123}} - \frac{1}{p_{13}}\right\} \\ &+ \frac{BC'}{(RT)^2}\{p_{13} - p_{123}\} + \text{a term in}\left(\frac{C'}{RT}\right)^2\end{aligned} \tag{8}$$

At moderate pressures, the terms containing the third virial coefficient may be neglected and the expression reduces to:

$$B = RT\left\{\frac{1}{p_{12}}\left(\frac{1}{p_{123}} - \frac{1}{p_{12}}\right) - \frac{1}{p_1}\left(\frac{1}{p_{13}} - \frac{1}{p_{12}}\right)\right\}\Big/\left\{\frac{1}{p_{13}} - \frac{1}{p_{123}}\right\} \tag{9}$$