Fluidized-Bed Reactor Model with Generalized Particle Balances

Part I: Formulation and Solution

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The changes to the solid phase which occur within fluidized-bed gas-solid reactors may have a significant impact on the performance of this class of reactors, particularly in coal conversion applications. In this paper a particle balance model is developed which accommodates particle distributions dependent on both size and density as well as populations consisting of multiple solids. The fast double collocation method used for solving the associated PDE permits this calculation to be performed within the framework of a Davidson-Harrison bubbling-bed model. A modification of the iteration procedure for the emulsion-phase state variables of the D-H model is reported which allows the composite reactor model to be executed reliably and efficiently. A general purpose program has been implemented, whose application is presented in a companion paper.

SCOPE

Fluidized-bed reactors have received considerable attention in processes for the conversion of coal to synthetic fuels and chemical feedstocks because of the many advantages this reactor type offers in carrying out gas-solid reactions. These include: ease in handling large solid flows; enhanced heat and mass transfer rates; and the possibilities for near-isothermal operation. One of the persistent problems with this class of reactors has been the difficulty in predicting reactor performance for purposes of scale-up and design. The difficulty arises because of the complex combination of phenomena which must be considered in developing models of such reactors. These phenomena include:

- Hydrodynamics of gross gas-solid mixing behavior
- Gross solid population changes resulting from reaction and mechanical effects
- Single particle gas-solid reaction-diffusion effects
- Homogeneous gas-phase reactions

Much of the earlier work on fluid-bed modeling was oriented towards catalytic applications, hence, focused primarily on the gross mixing behavior and, thus, led to various forms of two-phase and bubbling-bed models (Horio and Wen, 1977). In coal applications, however, the solid is consumed and the resulting changes in the overall solid phase do significantly influence reactor performance both because the single-particle reaction rate is a function of the particle temperature, size, composition as well as density and because key bed-fluidization characteristics depend on the nature of the solids in the bed. Since the bed solids normally differ substantially from the feed solids, the need for detailed consideration of particle population balances is apparent. However, the incorporation of these calculations into bubbling-bed models leads to a significant computational problem whose general solution has not yet been fully considered (Chen and Saxena, 1976; Gordon and Amundson, 1976).

In this paper a two-phase fluidized-bed model is presented which focuses on the detailed treatment of gas-solid behavior within a generalized two-phase bubbling-bed model. While this paper focuses on model formulation and solution procedures, a companion paper discusses the application of the composite model to char combustion (Overturf and Reklaitis, (1983), and Overturf (1980) considers application to char gasification.

CONCLUSIONS AND SIGNIFICANCE

A generalized particle balance model has been developed which can treat multiple reacting solids distributed in both size and density. The resulting PDE is solved using a double collocation procedure. The basic model can accommodate solids comprised of active and inactive components undergoing uniform reaction, shrinking core reaction with instant attrition of inerts, and shrinking core reaction with nonattriting inert shell. Treatment of attrition of inert solids occurring at a finite rate requires the introduction of a third dimension into the bed-particle distribution function. The particle balance computations are imbedded within the iterative loop of a nonisothermal Davidson-Harrison bubbling-bed model which includes both grid and freeboard region compartments. The key simplifying assumptions of the combined fluid-bed model are that: the particle temperatures are at quasisteady state; all solids are well mixed; the active and inert components of each solid are uniformly distributed; and the grid and dilute-phase compartments undergo only homogeneous gas-phase reactions. A flexible program implementing the model and its solution procedure has been developed. Given the appropriate rate expressions, the program executes the generalized D-H model calculations and thus permits the investigation of a broad range of reactor modeling assumptions.

CONCEPTUAL REACTOR MODEL

The conceptual model of a fluidized-bed reactor employed in this work is shown in Figure 1. Following Davidson and Harrison (1963), we assume that the bed consists of a dilute or bubble phase, composed only of solids-free gas, and a dense or emulsion phase, consisting of the solid particles and of a volume of gas corresponding to the bed-void fraction at minimum fluidization. The solids transfer heat and mass with the emulsion (or interstitial) gas but not directly with the dilute phase. Although as reviewed in

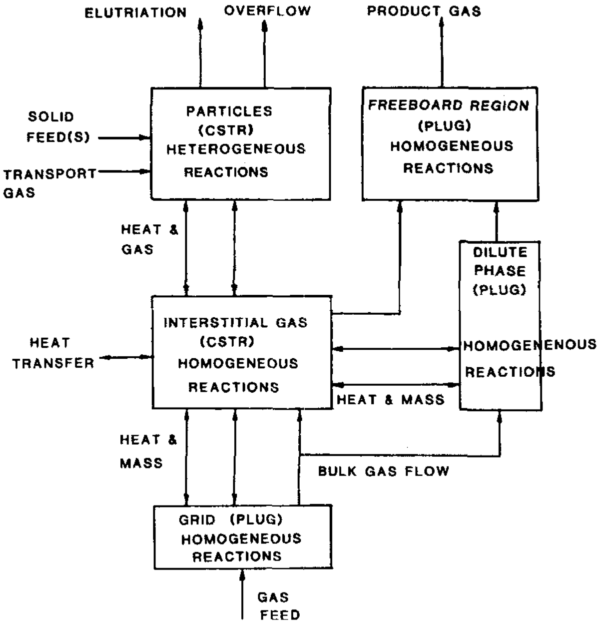

Figure 1. Generalized D-H model.

Horio and Wen (1977), numerous more detailed treatments of bubble growth and interphase heat and mass transfer have been reported; computational studies have not shown any overwhelming superiority of one model over another (Shaw et al., 1974). Moreover, none of these models are able to adequately predict the observed axial concentration profiles (Horio and Wen, 1977). Consequently, we have assumed uniform bubble size, plug flow of the dilute phase, perfectly mixed emulsion phase, and interphase heat and mass transfer following the Kunii-Levenspiel correlations (1969).

In addition to these two-bed phases, the conceptual model includes a grid region model which accounts for the vigorous gas-solid circulation and transport phenomena which can occur in the space immediately above the distributor (grid) of the bed. Behie and Kehoe (1973) have reported mass transfer rates in the grid region on the order of 50 times greater than the bubble to emulsion transfer rates in the bed itself. Accordingly, they have modelled the region as a plug-flow homogeneous gas-phase reactor of length equal to the jet penetration depth. Errazu and De Lasa (1977), on the other hand, proposed a CSTR grid region model. However, a comparative study (Grace and De Lasa, 1978) showed that there was little difference among alternate grid region models but a large difference among models which did and did not explicitly treat the grid region. In view of the lack of definitive grid region data, we have chosen to adopt the Behie and Kehoe treatment: plug-flow homogeneous reaction of reactor feed gas with heat and mass transfer between jets and the emulsion phase and no solids present in the jets. At the end of the grid region, the remaining gas is divided between the dilute and emulsion phases as in the original D-H model.

The fourth element of our conceptual model consists of the reactor space above the level of the fluidized bed called the freeboard region. This region can be important because of the extra residence time it provides for homogeneous gas-phase reactions as well as (possibly) for heterogeneous reactions between elutriated particles and the surrounding gas. Although treatments of solid reactions in the freeboard region are available (De Lasa and Grace, 1977; Rajan and Wen, 1978), our model simply considers homogeneous reactions occurring in a plug-flow gas because the influence of heterogeneous freeboard reactions on the overall bed performance are normally not significant.

GAS-PHASE BALANCE EQUATIONS

The conceptual model described above can be represented quantitatively by developing the material and enthalpy balance equations for each of the regions. In this section we formulate the dilute-phase, interstitial gas, and grid region equations. In the next section we will consider the balances involving the emulsion-phase solids.

The dilute-phase material balance equations for each gaseous species j is given by

$$U_D \frac{dC_{jD}}{dz} = (K_{BE})_{jB}(C_{jE} - C_{jD}) + \sum_{i=1}^{M} \alpha_{ij} r_i \quad (1)$$

with $\quad C_{jD} = C_{jJ} \text{at } z = z_j \quad (2)$

The enthalpy balance is similarly given by,

$$\bar{\rho}_g \bar{C}_{pg} U_D \frac{dT_D}{dz} = (H_{BE})_B(T_E - T_D) + \sum_{i=1}^{M} (-\Delta H_{rxn,i}) r_i \quad (3)$$

with $\quad T_D = T_J \quad$ at $z = z_J$, the jet region boundary $\quad (4)$

Both equations are derived assuming no solids in the dilute-phase, constant-volume reactions, and properties averaged over the axial reactor length.

The interstitial gas species balances include terms accounting for bulk input and output flow, bubble to emulsion mass transfer, jet to emulsion mass transfer, molar input due to heterogeneous reactions, as well as molar input due to homogeneous reaction.

$$q_E(C_{jf} - C_{jE}) + \frac{1}{L} \int V_D (K_{BE})_{jB}(C_{jD} - C_{jE}) dz$$
$$+ \frac{1}{\delta} \int V_J (K_{JE})_{jJ}(C_{jJ} - C_{jE}) dz + \int_{\text{particles}}^{\text{all}} \left(\sum_{i=1}^{N} \alpha_{ij} r_i^1 \right) dP$$
$$+ V_E \sum_{i=1}^{M} \alpha_{ij} r_i = 0 \quad (5)$$

The interstitial gas species balances are written with the assumption that the volume of gas contained within the solid pores is negligible.

The interstitial gas enthalpy balance includes terms corresponding to those present in the species balances but in addition accounts for heat transfer from reactor walls and from immersed surfaces.

$$q_E \rho_g C_{pg}(T_f - T_E) + \frac{1}{L} \int V_D (H_{BE})_B (T_D - T_E) dz$$
$$+ \frac{1}{\delta} \int V_J (H_{JE})_{jJ} (T_J - T_E) dz + \int h_p A_p (T_p - T_E) dP$$
$$+ h_c A_c (T_c - T_E) + h_w A_w (T_w - T_E)$$
$$+ V_E \sum_{i=1}^{M} (-\Delta H_{rxn,i}) r_i = 0. \quad (6)$$

As a simplifying assumption, the integrals involving heat and mass transfer with the dilute and jet phases are replaced by their average values. Thus,

$$\frac{1}{L} \int V_D (K_{BE})_j (C_{jD} - C_{jE}) dz \simeq V_D (\tilde{K}_{BE})_j (\tilde{C}_{jD} - C_{jE}) \quad (7)$$

$$\frac{1}{L} \int V_D (H_{BE}) (T_D - T_E) dz \simeq V_D (\tilde{H}_{BE}) (\tilde{T}_D - T_E) \quad (8)$$

Similarly,

$$\frac{1}{\delta} \int V_J (K_{JE})_{jJ} (C_{jJ} - C_{jE}) dz \simeq V_J (\tilde{K}_{JE})_j (\tilde{C}_{jJ} - C_{JE}) \quad (9)$$

$$\frac{1}{\delta} \int V_J (H_{JE})_{iJ} (T - T_E) dz \simeq V_J (\tilde{H}_{JE}) (\tilde{T}_J - T_E) \quad (10)$$

where the average values for the dependent variables $\tilde{C}_{jD}, \tilde{C}_{jJ}, \tilde{T}_D, \tilde{T}_J$ and the transfer coefficients $\tilde{K}_{BE}, \tilde{H}_{BE}, \tilde{K}_{JE}, \tilde{H}_{JE}$, are determined independently. This is a significant simplification because in general the coefficients K and H do depend on the gas properties and the bubble size. However, in view of the uncertainties in the predictions of these coefficients and consistent with the uniform bubble-size assumption, this averaging is a reasonable engineering approximation. Clearly, the significance of the averaging effect will vary from system to system and must be individually verified.

Assuming emulsion to jet transfer and only homogeneous reactions, the grid region species balances are

$$U_J \frac{dC_{jJ}}{dz} = (K_{JE})_{jB}(C_{jE} - C_{jJ}) + \sum_{i=1}^{M} \alpha_{ij} r_i \tag{11}$$

with

$$C_{jJ} = C_{jf} \qquad \text{at } z = 0 \tag{12}$$

The grid region enthalpy balance is,

$$\bar{\rho}_g \bar{C}_{pg} U_J \frac{dT_J}{dz} = (H_{JE})(T_E - T_J) + \sum_{i=1}^{M} (-\Delta H_{rxn,i}) r_i \tag{13}$$

with

$$T_J = T_f \qquad \text{at } z = 0 \tag{14}$$

The freeboard region balance equations are similar to those of the grid region but with the emulsion-to-jet transfer terms deleted. This completes the gas-phase equations. In the next section we develop the solid-phase balances.

SOLID-PHASE CALCULATIONS

The preceding gas-phase balance equations contain only two terms which involve the solid particles. These terms are the mass generation integral due to heterogeneous reactions which occurs in the emulsion gas mass balance equations and the corresponding term in the emulsion gas enthalpy balance equation. These integrals are evaluated by integrating the reaction contributions arising from all particles and hence involve as a weighting function the particle distribution function. It is thus necessary to evaluate this distribution function by performing population balance calculations. Such calculations have been reported by Levenspiel et al. (1968) for the shrinking core solid reaction model and by Chen and Saxena (1976) for the uniform reaction, decreasing density reaction model. The dynamic form of the latter population balances has been considered by Weimer and Clough (1980a). The steady-state model presented here encompasses both possibilities.

Balance Formulation

For purposes of the subsequent development, solid particles are assumed to consist of both active and inactive materials uniformly distributed throughout the reactive particle core. Moreover, solid behavior during gas-solid reactions is assumed to be described in terms of three limiting types: shrinking core with attriting shell, shrinking core with nonattriting shell, and uniform reaction. If one or more of these three heterogeneous reaction modes do occur within a fluid-bed reactor, the particle population must be described by particle size and/or density. Hence, the particle balance equations must accommodate a two-dimensional feed population distribution and must predict the bed-particle population distribution expressed as a function of particle size and/or particle density. The density can, of course, be replaced by alternate equivalent variables, for example, concentration of active material in the core or conversion of active material in the particle.

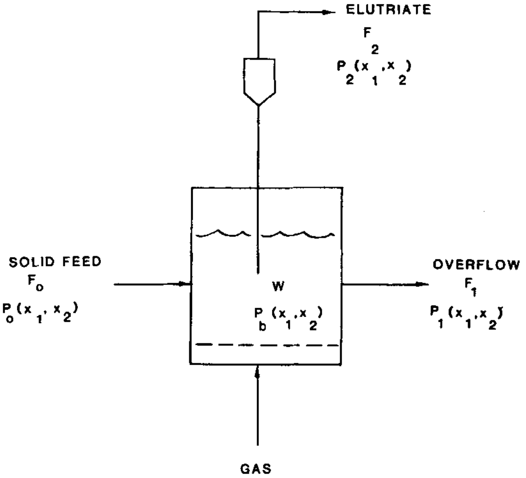

Figure 2. Solid flows for determination of fluidized-bed reactor particle distributions.

Consider the fluidized-bed reactor, Figure 2 in which p_0, p_1, p_2, and p_b are, respectively, the feed, overflow, fines, and bed-particle distribution functions, and W is the total weight of bed solids. Suppose all distribution functions are expressed in terms of size x_1 and density x_2. Over an increment of solid x_1 to $x_1 + \Delta x_1$ and x_2 to $x_2 + \Delta x_2$, the solid mass balances must include the following terms:

- Solid in the increment Δx_1 Δx_2 entering in the feed
- Solid in the increment leaving in the overflow
- Solid elutriated from the bed as fines
- Solid reacting into the increment Δx_1 from the next larger increment via shrinking core reactions
- Solid reacting out of the increment Δx_1 via shrinking core reactions
- Solid reacting into the increment Δx_2 from the next larger density increment via continuous reactions
- Solid reacting out of the increment Δx_2 via continuous reactions
- Solid in the increment Δx_1 Δx_2 which reacts in both modes but does not leave the increment

where

$R(x_1,x_2)$ = total heterogeneous reaction rate for all shrinking core reactions

$R^1(x_1,x_2)$ = total heterogeneous reaction rate for all continuous reaction reactions

The above terms can be assembled in equation form as:

$$\begin{aligned} F_0 p_0(\bar{x}_1,\bar{x}_2)\Delta x_1 \Delta x_2 - F_1 p_1(\bar{x}_1,\bar{x}_2)\Delta x_1\, \Delta x_2 \\ - F_2 p_2(\bar{x}_1,\bar{x}_2)\Delta x_1\, \Delta x_2 + W p_b(x_1,x_2)\, R(x_1,x_2)\Delta x_2 |x_1,\bar{x}_2 \\ - W p_b(x_1,x_2) R(x_1,x_2)\Delta x_2 | x_1 + \Delta x_1,\bar{x}_2 \\ + W p_b(x_1,x_2) R^1(x_1,x_2)\Delta x_1 | \bar{x}_1,x_2 \\ + W p_b(x_1,x_2) R^1(x_1,x_2)\Delta x_1 | \bar{x}_1,x_2 + \Delta x_2 \\ + W p_b(\bar{x}_1,\bar{x}_2)\Delta x_1 \Delta x_2 \left(\frac{3R(\bar{x}_1,\bar{x}_2)}{\bar{x}_1} + R^1(\bar{x}_1,\bar{x}_2)\right) = 0 \end{aligned}$$

where $\bar{x}_i$ is some mean value of x_i used to characterize the cut, $x_i < \bar{x}_i < x_i + \Delta x_i$.

If the solids in the fluid bed are assumed to be well mixed, it follows that

$$p_1(x_1,x_2) = p_b(x_1,x_2)$$

The solids elutriated from the bed can be quantified in terms of an elutriation constant $K(x_1,x_2)$ defined as,

$$K(x_1,x_2) = \frac{F_2 p_2(x_1,x_2)}{W p_b(x_1,x_2)}$$

for which correlations are available. Substituting these relations and taking the limit as $\Delta x_1 \to 0$, $\Delta x_2 \to 0$, the balance takes the form,

$$\begin{aligned} \frac{\partial}{\partial x_1}[p_1(x_1,x_2)R(x_1,x_2)] + \frac{\partial}{\partial x_2}[p_1(x_1,x_2)R(x_1,x_2)] \\ = P_1(x_1,x_2)\left[\frac{3R(x_1,x_2)}{x_1} + R^1(x_1,x_2)\right. \\ \left. - \frac{F_1}{W} - K(x_1,x_2)\right] + F_0 \frac{p_0(x_1,x_1)}{W} \end{aligned} \tag{15}$$

For simplicity in notation, define

$$\alpha_1(x_1,x_2) = \frac{3R(x_1,x_2)}{x_1} + R^1(x_1,x_2) - \frac{F_1}{W} - K(x_1,x_2) - \frac{\partial R(x_1,x_2)}{\partial x_1} - \frac{\partial R^1(x_1,x_2)}{\partial x_2}$$

$$\alpha_2(x_1,x_2) = \frac{F_0 p_0(x_1,x_2)}{W}$$

Thus, the balance equation reduces to the partial differential equation,

$$R(x_1,x_2)\frac{\partial p_1(x_1,x_2)}{\partial x_1} + R^1(x_1,x_2)\frac{\partial p_1(x_1,x_2)}{\partial x_2} = \alpha_1(x_1,x_2)p_1(x_1,x_2) + \alpha_2(x_1,x_2)$$

with boundary conditions

$$\begin{aligned} p_1(x_1,x_2) &= 0, \quad \text{for } x_1 = x_{1f}^+ \\ \text{and } p_1(x_1,x_2) &= 0, \quad \text{for } x_2 = x_{2f}^+ \end{aligned} \tag{16}$$

where x_{1f} and x_{2f} are the feed conditions.

An analytical expression can be obtained for the solution of this equation by using the method of characteristics. Let,

$$\frac{dx_1}{ds} = R(x_1,x_2) \qquad \text{and } \frac{dx_2}{ds} = R^1(x_1,x_2)$$

where s is the characteristic variable. Then it can be shown that the solution will be given by:

$$p_1(s) = \frac{F_0}{W}\frac{1}{R(s)}\frac{1}{R^1(s)}x_1^3\exp(x_2)\cdot\left\{\exp\int_{s0}^{s} -(F_1/W + K(s)ds\right\} \int_{s0}^{s}\frac{p_0(s^*)R(s_0^*)R^1(s_0^*)}{x_1^*\exp(x_2^*)}\left\{\exp\int_{s0}^{s} -(F_1/W + K(s))\,ds\right\}ds^* \tag{17}$$

This solution must of course satisfy the normalization condition

$$\int_{s0}^{s_{\max}} p_1(s)\,ds = 1 \tag{18}$$

Satisfaction of this condition is achieved by iterating on the unknown overflow rate F_1 or the bed weight W.

Note that the analytical solution reduces to the Kunii and Levenspiel (1969) derivation when there is no change in the particle density x_2.

Solution Approach

While the analytical solution can be evaluated by using quadrature formulas to calculate the multiple integrals, a direct numerical solution of the PDE is preferable because of the repeated evaluations required to meet the normalization condition. In selecting the solution approach it is important to note that the actual distribution is never required explicitly; rather it is only needed to evaluate several integrals, that is the heterogeneous term in the emulsion balances and the above normalization condition. Consequently, it proves quite efficient to solve the PDE by using a collocation method (Villadsen and Michelsen, 1978). In the present instance, since a two-dimensional solution function is required, a double collocation technique is appropriate (Birnbaum and Lapidus, 1978).

Thus, the solution of the PDE (Eq. 15) is assumed to be approximated by a double sum of Lagrange interpolating polynomials $1(x)$.

$$p_N(x_1,x_2) = \sum_{i=1}^{NTC}\sum_{i=1}^{NTD} p_{ij}1_i(x_1)1_j(x_2)$$

where

NTC = total number of size collocation points
NTD = total number of density collocation points

The polynomial coefficients are evaluated by solving the residual equations at collocation points whose locations are determined as roots of an orthogonal polynomial using well-known procedures (Villadsen and Michelsen, 1978). In the present case the residuals will take the form,

$$R = \mathcal{R}_{ij}\sum_{k=1}^{NTC} B_{jk}p_{ik} + \mathcal{R}'_{ij}\sum_{k=1}^{NTD} A_{ik}p_{kj} - \alpha_{1ij}p_{ij} - \alpha_{2ij} = 0 \tag{19}$$

at each of the ixj collocation points. The arguments for $\mathcal{R}$, $\mathcal{R}'$, α_1, and α_2 have, for convenience, been dropped. The entities B_{jk}, A_{ik} are constant coefficients arising from the differentiation of the Lagrange polynomials. If we define the corresponding matrices, $\bar{\bar{B}} = (B_{jk})$, $\bar{\bar{A}} = (A_{ik})$, $\bar{\bar{P}} = (p_{ij})$ and for $\bar{\bar{x}} = \bar{\bar{\mathcal{R}}}, \bar{\bar{\mathcal{R}}}', \bar{\bar{\alpha}}_1, \bar{\bar{\alpha}}_2$ the matrix $\bar{\bar{x}}$ is a diagonal matrix with diagonal elements $(x_{11},x_{12},\ldots,x_{ij},\ldots x_{1j},\ldots x_{1,NTC},x_{21},\ldots,x_{NTD,NTC})$, following Birnbaum and Lapidus (1978), the set of $NTD \times NTC$ residual equations can be written in the compact form,

$$[\bar{\bar{\mathcal{R}}}(\bar{\bar{A}} \otimes \bar{\bar{I}}_{NTC}) + \bar{\bar{\mathcal{R}}}'(\bar{\bar{I}}_{NTD} \otimes \bar{\bar{B}}^T) - \bar{\bar{\alpha}}_1] \cdot (rs\bar{\bar{P}})^T - (rs\bar{\bar{\alpha}}_2)^T = 0 \tag{20}$$

where $rs\,\bar{\bar{x}}$ *is* the row string of $\bar{\bar{x}}$—the catenation of the rows of $\bar{\bar{x}}$ into a single vector—and ⊗ indicates a Kronecker product. The probability density, $p_{NTD,NTC}$, for the largest particles in a solid feed which is consumed by reaction can be computed from the reaction rate for the solid feed. When this value is substituted the expressions for $p_{NTD,j}$, $j = 1, NTC - 1$ and $p_{i,NTC}$, $i = 1, NTD - 1$ must be adjusted accordingly. The resulting set of $NTD \times NTC - 1$ linear equations can be solved using Gaussian elimination to determine the solid probability density function.

Particle Temperature Calculation

The reaction rates used in the development of the particle balance equations were expressed as functions of particle size and density only. Strictly speaking, there is, however, a further dependence on the particle temperature. Since particles of different size and density will experience different reaction and heat transfer rates, it is to be expected that different particles will follow different temperature trajectories. Inclusion of the single-particle balances in the bed model is therefore necessary.

Consider a single solid particle of diameter d_p, volume V_s and density ρ_s reacting according to S shrinking core and C continuous mode reactions. The mass balance simply is

$$\frac{d(\rho_s V_s)}{d\theta} = -\sum_{i=1}^{S} R_i(d_p,\rho_s,T_p) - \sum_{j=1}^{C} R_j^1(d_p,\rho_s,T_p)$$

where T_p is the particle temperature.
The enthalpy balance which includes the heat of reaction terms, the convective heat transfer term, and the radiation term, takes the form,

$$\frac{d}{d\theta}(\rho_s C_{p_s} V_s T_s) = \sum_{i=1}^{S} R_i(-\Delta H_{rxn,i}) + \sum_{j=1}^{C} R_j^1(-\Delta H_{rxn,j}) + h_p A_p(T_E - T_p) + \epsilon_p \sigma(T_E^4 - T_p^4)$$

In this formulation particle-to-wall and particle-to-particle radiative heat transfer are neglected. The latter assumption affects particle temperature and will be discussed further in the next section. Separating the enthalpy derivative into two parts and using the material balance to eliminate the $d(\rho_s V_s)/d\theta$ term, the following equation for the particle temperature can be obtained, assuming constant heat capacity,

$$C_{p_s}\rho_s V_s \frac{dTs}{d\theta} = -C_{p_s}T_s\left[-\sum_{i=1}^{S} R_i(d_p,\rho_s,T_p) - \sum_{i=1}^{C} R_j^1(d_p,\rho_s,T_p)\right] + \sum_{i=1}^{S} R_i(-\Delta H_{rxn,i}) + \sum_{j=1}^{C} R_j^1(-\Delta H_{rxn,j}) + h_p A_p(T_E - T_p) + \epsilon_p \sigma A_p(T_E^4 - T_p^4) \tag{21}$$

On the basis of numerical studies (Weimer and Clough, 1980b; Overturf, 1980) it can be shown that with particle sizes in the normal range used in fluid-bed applications (up to 10 mm), the particle temperature response is quite rapid compared to the normal residence time of solids in a well-fluidized bed. Consequently, we assume that the particle temperature response is instantaneous; i.e., particle temperature is at quasisteady state. Thus, $dT_p/d\theta \simeq 0$ and the particle enthalpy balance is reduced to an algebraic equation in T_p. For given particle size and density and emulsion-phase conditions, the particle temperature therefore can be determined from the above equation. Consequently, since particle temperature is an implicit function of d_p and ρ_s, the temperature dependence of the heterogeneous rates R_i and R_j^1 need not be explicitly considered in the solids population balance equations. The reader will note that the algebraic equation for T_p can not be explicitly solved for T_p and thus does require an iterative calculation.

Complete Solids Balance Calculations

The solution of the solids balance equation for a single solid feed typically will proceed as follows:

1) The stoichiometry and nature of the solid reactions are specified. (The nature of the solid reaction, i.e., whether it behaves according to a shrinking core or continuous reaction model, need not be specified *a priori*. This decision can be made for each particle size and may depend upon some computed value such as the Thiele modulus.)

2) Location of collocation points and values of quadrature weights are computed for the Gauss-Jacobi formula.

3) Particle temperatures and thereby reaction rates are computed at the collocation points based on the quasisteady-state temperature assumption.

4) The bed weight, W, is estimated from the specified solid volume and the best guess of solid density.

5) The nature of the solid feed distribution, either mono- or polydispersed, is used to determine the value of the probability density function for the unreacted particles.

6) A value for the overflow rate, F_1, for the solid is estimated.

7) The set of linear equations for the residuals is solved to determine the probability density function.

8) The normalizing condition is checked using the quadrature formula,

$$\iint p_1(x_1,x_2)dx_1dx_2 = \cong \sum_{i=1}^{NTD} \sum_{j=1}^{NTC} w_i w_j p_1(x_1,x_2)\Big|_{i,j} = 1. \quad (22)$$

9) The value of the solid overflow rate, F_1, is adjusted until the normalizing condition is satisfied.

Updated estimates for F_1 are generated using a bounded Newton-Raphson method. Overall convergence is well-behaved since $\iint p_1(x_1,x_2)dx_1dx_2$ is a monotonic function of F_1.

OVERALL REACTOR MODEL SOLUTION

The set of gas and solid-phase balance equations consists of algebraic, ordinary differential, and partial differential equations which must be solved simultaneously to simulate the entire reactor. Fortunately by taking advantage of the structure of the model, a reasonably efficient iterative calculation scheme can be developed.

Let C_E be the vector consisting of the interstitial gas compositions and the gas temperature, $C_D(z)$ be the vector of dilute-phase composition and temperature (a function of axial position), and $C_J(z)$ be the vector of grid region compositions and temperatures. Furthermore, let $\overline{C}_D$ and $\overline{C}_J$ represent the average values of C_D and C_J over the axial length and denote by I_j the solid integral term in the jth emulsion-phase balance. Then, the model equations can be grouped into the following sets of vector equations:

1. The grid region balances:

$$\frac{dC_J}{dz} = g(C_J,C_E) \qquad \text{with } C_J(0) = C_f, \text{ the gas feed conditions}$$

2. The dilute-phase balances

$$\frac{dC_D}{dz} = f(C_D,C_E) \qquad \text{with } C_D(\delta) = C_J(\delta)$$

3. The interstitial gas balances

$$\phi((\overline{C}_D,\overline{C}_J)C_E,C_J(\delta),I) = 0$$

4. The particle temperature equations

$$h_{ij}(C_E,T_{ij}) = 0, \qquad \text{at each collocation point } (i,j)$$

5. The particle balance PDE which is directly dependent on C_E and T_p through the reaction rates,

$$p(x_1,x_2) = r(C_E,T_p)$$

6. The integrals I_j which involve the particle distribution function and thus depend on C_E and T_p.

7. The definition of $\overline{C}_D$ and $\overline{C}_J$

$$\text{e.g., } \overline{C}_D = \frac{\int C_D(z)dz}{\int dz}$$

8. The freeboard ODE's which depend on C_E and C_D.

9. The fluidization correlations (Appendix A).

The C_E variables are common to all balances and thus are the natural iteration variables for the model. With an initial estimate of C_E, the jet and bubble balances can be integrated to determine C_D, C_J and their average values $\overline{C}_D$ and $\overline{C}_J$. Next, the particle temperature equations can be solved at the selected particle-size and density collocation points. The heterogeneous rates can be evaluated at the collocation points and then the particle balance equation can be solved as described earlier. Recall this will have to be done iteratively since the overflow rate, F_1, must be adjusted so that the normalization condition is satisfied. Next, the integrals (quadrature formulas) for the I's must be evaluated. Finally, with $\overline{C}_B$, $\overline{C}_J$, $C_J(\delta)$, and the I's fixed, the interstitial gas balances must be solved to update C_E. Strictly speaking the dependence of I on C_E would require that this variation be reflected in the Newton-Raphson iteration formula when solving the ϕ equations. In fact, this was attempted by Gordon and Amundson (1976) for the special case of a fluid-bed combustor with monosized feed and shrinking core reaction and was found to require prohibitive computation times. In the present work, the updated I's are assumed fixed during the Newton-Raphson iterations. This leads to considerable computational savings without noticeable deterioration of the convergence rate. When the ϕ equation solution has converged, the resulting new estimate of C_E is used to start another pass of calculations. To speed convergence of C_E, Henrici's extension of Aitken's algorithm (Henrici, 1965) is applied to the generated C_E vector prior to reinitiating the jet balance integration.

Finally, when the calculations have converged, the resulting value of C_E and C_D are used in the integration of the freeboard region balances.

In implementing these calculations, it was found expedient to carry out integration of the plug-flow models using Gear's method (1971) with Jacobians evaluated by numerical differencing. To enhance the reliability of the calculations for the solid temperatures and the interstitial gas-state variables C_E, the following fallback procedures proved useful. If the Newton-Raphson iterations for the particle temperatures failed to converge, the initial estimates of each T_p were first increased to seek a solution for the diffusion-limited branch of the heat generation curve. If this failed, the initial estimates of each T_p were reduced to check for a kinetically-limited solution. If the Newton-Raphson iterations for C_E failed to converge or if negative concentrations were encountered, the unsteady-state form of the balances were integrated to find a better estimate of the steady-state C_E.

EXTENSIONS OF BASIC MODEL

To simply the exposition, the development of the particle balance equation and overall calculation strategy were given in terms of particles which were comprised solely of active material of a single-solid type. Extensions to treat multiple solids and solids composed of both active and inactive material will be briefly outlined in this section.

To extend the development to particles containing inactive material two cases must be considered:

i) The inert material attrites instantaneously as the particle undergoes a shrinking core solid reaction.

ii) An inert shell remains intact while the active material is consumed.

The first case can be accommodated by merely modifying the basic equation. (Eq. 15) to reflect the fact that change in particle mass is due both to consumption of active solid and attrition of inert. Thus, in Eq. 15 the term,

$$\frac{3\mathcal{R}(x_1,x_2)}{x_1} \text{ is replaced by } \frac{3\mathcal{R}(x_1,x_2)}{x_1 f_a}$$

$$\text{with } f_a (=) \frac{\text{amount of active solid in particle}}{\text{total amount of solid in particle}}$$

In addition to this modification, the amount of attrited material must be computed. All solid particles generated by attrition are treated as monodispersed fines which exit the reactor instantaneously at the emulsion-phase temperature. The flow rate of inert fines leaving the reactor can be determined from a balance on the inert material.

$$\begin{aligned}\text{FINES} = F_0 \iint (1 - f_a(x_1,x_2))p_0(x_1,x_2)dx_1dx_2 \\ - \iint (1 - f_a(x_1,x_2))p_1(x_1,x_2)dx_1dx_2 \\ - F_2 \iint (1 - f_a(x_1,x_2))p_2(x_1,x_2)dx_1dx_2\end{aligned}$$

The second case, in which the inert material remains intact, requires that the distribution function be expanded to include a third variable, namely the initial particle size x_3. However, computation of the distribution of particle sizes $p_1(\ , \ ,x_3)$ (not active core size) is relatively simple since particle size is not altered by reaction. In fact, in the absence of elutriation, the particle-size distribution in the bed will be identical to the particle-size distribution of the solid feed. If elutriation is included, differences between the bed and feed distributions will reflect the influence of the particle characteristics on the elutriation rates. Since particle size is not altered by reaction, the double collocation solution technique for characterization of the active core can be applied to each of the discrete particle sizes independently. The mean reaction rates will be revised prior to each iteration to reflect the updated estimates of the bed distribution. The distribution function is still expressed in terms of the total bed weight. Now, however, a further possibility can exist, namely the presence of solid particles in the reactor which, because the active material has been completely consumed, cannot undergo further reaction. One means of determining the quantity of inert particles present in the reactor is by a mass balance for the inert material in all particles of a specified size. The balance assumes the form:

$$\begin{aligned}F_0 \{\iint f_i(x_1,x_2,x_3)p_0(x_1,x_2,x_3)dx_1dx_2 + p_{x_i}(0,0,x_3)\} \\ + F_1 \{\iint f_i(x_1,x_2,x_3)p_1(x_1,x_2,x_3)dx_1dx_2 + p_{x_b}(0,0,x_3)\} \\ + W\{\iint f_i(x_i,x_2,x_3)\mathrm{K}(x_1,x_2,x_3)p_1(x_1,x_2,x_3)dx_1dx_2 \\ + \mathrm{K}(0,0,x_3)\, p_{x_b}(0,0,3\} = 0\end{aligned}$$

where

$$f_i(x_1,x_2,x_3) = 1 - f_a(x_1,x_2,x_3)$$

$$(=) \frac{\text{amount of inert in particle}}{\text{total amount of particle}}$$

$p_{x_i}(0,0,x_3)$ and $p_{x_b}(0,0,x_3)$ are probability densities for feed and bed solid particles respectively comprised entirely of inert material while $K(0,0,x_3)$ is the elutriation constant for these particles. The normalization condition for the solid probability density function becomes

$$\int \{\iint p_1(x_1,x_2,x_3)dx_1dx_2 + p_{x_b}(0,0,x_3)\}dx_3 = 1$$

The case of multiple solid feeds to a fluidized-bed reactor are of particular interest in coal conversion applications and can readily be accomodated within our basic model. In fact, the solution procedure for multiple solid feeds is identical to that described previously for a single type of solid feed of distributed size comprised, in part, of inert material which remains intact. In the case of multiple solids the solid probability density function will refer to all of the solid particles present in the bed, not just a single solid type. As a result, assuming perfectly mixed solids, the relation,

$$\left(\frac{F_1}{W}\right) \text{solid } 1 = \left(\frac{F_1}{W}\right) \text{solid } 2 = \left(\frac{F_1}{W}\right) \qquad \text{(I-86)}$$

must hold for each type of solid. Thus the solid balance calculations can be carried out by iterating on the total solid overflow rate, F_1, as is done for the single solid feed type. However, unlike before, the bed weight W, which is estimated at the beginning of each interation, reflects the differences in solid type as well as size and extent of conversion. The updated bed weight will now be based on the solid weight fractions as determined by the solids balance calculations during the previous iteration.

With the high degree of solid circulation that is characteristic of fluidized-bed operations particle attrition is frequently a problem. In addition to the cases of no solid attrition and instantaneous attrition of inert solids, which have already been discussed, the attrition of solid which contains both active and inert material is possible. Such attrited particles would probably be characterized by both very short residence times and very high reaction rates. Unfortunately the behavior of the attrited particles in the bed would not conform to either normal fluidized bed or entrained flow patterns. Although the particles would be entrained, their residence time would be increased by collisions with nonentrained solids. Moreover, the reaction rates for these smaller particles could well be limited by factors different from those affecting the remainder of the particles in the bed. Thus, even though a term could be included in the solid balance calculations to account for attrited solids with a distribution of active solid content, it would be difficult to provide an estimate of the extent to which these particles react while being elutriated from the bed. Of course, particles for which the attrition of active material is quite high are probably not well suited for use in fluidized beds.

A final possibility is the attrition of inert solids at a finite rate. Although quite easy to handle conceptually, implementation would require extension of the double collocation technique to a third coordinate dimension since the active core balance calculations could no longer be performed independently for each initial particle size. The more limited situation in which the active solid is consumed by a shrinking core type of reaction while the inert solid attrites at a finite rate can be accommodated without an increase in dimensionality.

COMPUTER PROGRAM

A general purpose computer program BED (Overturf and Reklaitis, 1980) implementing the model and solution strategy of this paper has been developed. The program incorporates the fluidization correlations given in Appendix A, uses the physical properties estimation package PPROPS (Overturf, 1979) and the IMSL Gear integrator. Detailed case study results obtained using this program are discussed in a companion paper (Overturf and Reklaitis, 1983) and in Overturf (1980).

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APPENDIX A

The fluidization parameters employed in the model are estimated from widely accepted correlations available in the literature.

1. For *minimum fluidization velocity*, the IGT modification of the Ergun equation is used (IGT, 1976).
2. For *void fraction at minimum fluidization*, available correlations are not reliable. Therefore, the value of ϵ_{mf} is treated as model parameter.
3. For *bubble rise velocity and bed expansion*, the Davidson-Harrison (1963) treatment is used.
4. For *interphase mass and heat transfer coefficients*, the Kunii-Levenspiel (1969) correlations are used. The cloud to emulsion transfer is assumed to take place solely by diffusion.
5. *Solid-Gas Mass and Heat Transfer Coefficients.* The solid and gas are assumed to be stationary with respect to each other. Thus, the Sherwood and Nusselt numbers are each assumed to be equal to 2.0.
6. For *jet penetration depth*, the correlation due to Zenz (1968) is used. The number and diameter of orifices are specified.
7. *Bubble size* can either be specified as a model parameter or be estimated using the Mori and Wen (1975) correlations. A check is made to insure maximum predicted bubble size does not exceed the maximum allowable bubble size.
8. *Elutriation Constant.* The Merrick and Highley (1974) correlation was used since it is at least qualitatively consistent for polydispersed solids.
9. For *heat transfer coefficients*, bed-to-tubes and emulsion-to-wall convective heat transfer coefficients were estimated using the correlations recommended by Kunii and Levenspiel (1969).

NOTATION

A = coefficients in polynomial approximation to the solids distribution function
A_c = external surface area of internal heat transfer coil
A_p = external surface area of particle
A_w = area of bed wall
B = coefficients in polynomial approximation to the solid-distribution function
C = number of continuous mode reactions
C_{jD} = molar concentration of species j in bubble
C_{jE} = emulsion phase
C_{jf} = feed phase
C_{jJ} = jet region phase
Cp_g = gas molar heat capacity
Cp_s = solid heat capacity
$\bar{D}_B$ = mean bubble size
d_p = particle diameter
$\bar{d}_p$ = mean particle diameter
f_a = fraction of active material in particle
F_0 = solid feed rate to fluidized bed
F_1 = solid overflow rate
F_2 = solid elutriate rate
H_{BE} = bubble-to-emulsion heat transfer coefficient based on bubble volume
h_c = bed-to-internals heat transfer coefficient
H_{JE} = jet-to-emulsion heat transfer coefficient based on jet volume
h_p = single particle convective heat transfer coefficient
$\Delta H_{rxn},i$ = heat of reaction for reaction i
h_w = bed-to-wall heat transfer coefficient
$(K_{BE})_{jB}$ = bubble-to-emulsion mass transfer coefficient for species j based on bubble volume
$(K_{JE})_{jJ}$ = jet-to-emulsion mass transfer coefficient for species j based on jet volume
$K(x_1, ..)$ = elutriation constant
L = expanded length of bed
NTC = total number of discretization points used to represent density of active core
NTD = total number of discretization points used to represent size of active core
$p_b(x, , , ...)$ = probability density for solids in bed
p_0 = probability density for solids in feed
p_1 = probability density for solids in overflow
p_2 = probability density for solids in elutriate
q_E = volumetric flow of gas through emulsion phase
r_i = rate of homogeneous reaction i
r_i^1 = rate of heterogeneous reaction i
$R(x_1, ..)$ = rate of shrinking core heterogeneous
$R^1(x_1, ..)$ = rate of continuous particle heterogeneous reactions
S = number of shrinking core reactions
T_c = coolant temperature
T_D = bubble phase temperature
T_E = emulsion phase temperature
T_f = gas feed temperture
T_J = jet temperature
T_p = particle temperature
T_w = exterior wall temperature
U_D = bubble phase gas velocity
U_J = jet gas velocity
V_D = volume of bubble phase
V_E = volume of gas in emulsion phase
V_J = volume of jets
V_s = particle volume
W = weight of solids in bed
z = axial position
α_{ij} = stoichiometric coefficient for species j of reaction i
δ = jet penetration depth
ϵ_p = emissivity of solid
σ = Stefan-Boltzman constant
ρ_a = density of active material
ρ_g = gas density
ρ_s = particle density
Θ = time

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Part II: Coal Combustion Application

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The model described in Part I is applied to study an atmospheric fluidized-bed coal combustor. Extensive case studies are investigated on the effects of: enhanced grid region heat and mass transfer; bed void fraction; emulsion phase temperature; elutriation rates; reaction rate parameters; feed particle-size distribution; and particle superheat. Comparison is made with experimental results reported by Babcock and Wilcox. Proper representation of the grid region and use of actual feed distributions are shown to be essential to predicting combustor performance. Better particle elutriation and single-particle combustion submodels are found to be key requirements for improved combustor modeling.

SCOPE

Atmospheric fluidized-bed coal combustors (AFBC) have received considerable attention in recent years for use in commercial power generation and hence have also been subjected to extensive experimental and modeling studies. Typical operating conditions for such reactors are (Babcock and Wilcox, 1976): bed temperature, 760–1,100°C; particle size range, 150–2,500 μm; superficial gas velocity, 0.3–5 m/s; expanded bed height, 0.3 to 2m; and bed char fraction, 3 wt. %. The oxidizing medium for the bed is air and, as evident from the low-bed char fraction, the largest portion of the bed is noncombustible material: coal ash, limestone or dolomite. Important concerns in the utilization of AFBC's are the coal carbon losses which occur through elutriation and with bed solids withdrawal (Beer, 1976). Elutriation can, of course, be controlled by choosing larger particle sizes and lower gas velocities. However, small particles and large velocities favor higher particle combustion rates. Efficient design clearly requires a careful balance of these factors. A model which can accommodate polydispersed feeds and includes an adequate representation of the grid region can serve a very useful function in permitting studies of elutriation vs. combustion rate tradeoffs. Models reported in the literature do not incorporate these features. The model described in Part I does accomplish this and is applied in the present work to study the sensitivity of a simulated AFBC to key model parameters. Comparison is made to experimental results obtained by Babcock and Wilcox (1976) on a pilot unit.

CONCLUSIONS AND SIGNIFICANCE

On the basis of an extensive collection of simulation case studies, it can be demonstrated that the flue gas compositions as well as fixed carbon and oxygen conversions of an experimental fluidized-bed combustor can be predicted by suitable tuning of model parameters. More importantly, however, it is evident that

- Proper representation of the grid region is essential in combustor modeling. If, as is likely the case, most dilute to emulsion-phase transfer occurs in the grid region, bubbling-bed models may be of limited value in predicting reactor performance.
- Representation of reactive solids by an average size and density is adequate for the prediction of combustor performance. The key factor seems to be the sensitivity of particle elutriation rates to particle size (and density).
- Satisfactory modeling of combustors requires a more reliable prediction of single-particle elutriation rates and a more detailed description of single-particle combustion than is offered by the conventional single-film shrinking core model.

The overall reactor model used in this work forms a satisfactory tool for combustor studies and should remain useful when better elutriation and single-particle reaction submodels become available.