A Mathematical Formulation for the Reduction of Iron Oxide Pellets in Moving Beds with Non-uniform Gas and Solids Flow*

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Synopsis

A mathematical formulation is developed describing a moving bed reactor in which iron oxide pellets are reduced with hydrogen, in a counter-current flow arrangement. The model consists of the heat and mass balance equations written for both the gas and the solid streams, and the three interface, shrinking core model is employed for representing the reaction kinetics. The model is thought to be general, in that it allows for non-isothermal behavior and non-uniformity (i.e., maldistribution) of both the solids and the gas streams. A selection of the computed results indicates very strong coupling between the solids flow, gas flow and the rate of reaction. If this coupling is neglected, as was done in previous investigations, very marked distortion of the results may occur.

I. Introduction

The reduction of iron oxide pellets in a moving bed arrangement is a problem of considerable practical importance, because this process plays a major role in both the "direct reduction" systems and in the stack region of the iron blast furnace.

In essence this problem has two components, namely the representation of a reactor where a moving bed is being contacted with a reactant gas and the actual modeling of the non-catalytic gas-solid reaction itself. Considerable work has been done in recent years in both these problem areas.

Yagi, et al.1,2) and Spitzer, et al.3) analyzed a moving bed reactor in which iron oxide pellets are reduced by hydrogen under isothermal condition. Ishida and Wen4) developed a mathematical model of a non-isothermal moving bed for non-catalytic gas-solid reactions. Recently, Hara, et al. completed a study on the reduction of iron oxides in a non-isothermal moving bed and interpreted their results using the three interface model.4-6)

Virtually all these models assumed one dimensional gas flow, thus neglected the possible effect of maldistribution. As a result of the work of Radestock and Jeschar,6,7) Stanek and Szekely,8-10) and Szekely, et al.,11) it has been shown that quite appreciable flow maldistribution may occur in packed beds, provided the distribution of the packing material is non-uniform. It has been shown, furthermore, in preliminary studies by Muchi and Kuwabara12) and Choudhary, Szekely and Weller13) that such flow maldistribution may have a marked effect on the performance of packed bed reactors.

However, all these studies of the effect of flow maldistribution on reactor efficiency were preliminary in that Kuwabara and Muchi uncoupled the fluid flow equations from the conservation of the reactant species, while the work of Choudhry, et al. was confined to isothermal, catalytic systems.

A great deal of work has been done on the other component of the problem, that is the modeling of non-catalytic gas-solid reactions. A thorough survey of this field is available in a recent monograph;14) here the gas-solid reaction models have been proposed by Ishida and Wen,15) Sohn and Szekely,16) and the three interface model of Spitzer, et al.17) and its further development by Hara, et al.18) are particularly noteworthy. Distributed models for non-catalytic reaction have also been proposed among others by Tien and Turkdogan19) and Fujishige.20) The purpose of the work, to be described in this paper, is to combine these previous efforts, by presenting a formulation of the reduction of iron-oxide pellets with hydrogen in a moving bed arrangement, under conditions, when both the gas flow and the flow of the solids are spatially non-uniform. The model thus described should be relevant to a broad range of applications, including direct reduction processes, stack gas injection and to a lesser extent the stack region of the iron blast furnace.

In this paper a general formulation is presented, while a comprehensive set of the computed results will be given in the following paper.21)

II. Formulation of Governing Equations

Let us consider a two-dimensional moving bed in which spherical iron oxide pellets are being reduced (shown in Fig. 1). The solid pellets are charged at the top of the bed and move downward steadily. The reducing gas is introduced at the bottom of the bed and ascends in a counter flow arrangement with the solid particles. The iron oxide pellets containing Fe₂O₃ and Fe₃O₄ are reduced to metallic iron through the intermediates Fe₃O₄ and FeO according to the chemical reactions shown below.

\[
\begin{align*}
3\text{Fe}_2\text{O}_3 + \text{H}_2 &= 2\text{Fe}_3\text{O}_4 + \text{H}_2\text{O} \\
\text{Fe}_3\text{O}_4 + \text{H}_2 &= 3\text{FeO} + \text{H}_2\text{O} \\
\text{FeO} + \text{H}_2 &= \text{Fe} + \text{H}_2\text{O}
\end{align*}
\]

Upon considering the general case of two-dimensional flow of both the gaseous and the solid streams within a cylindrical coordinate system, the following assumptions are made:

(1) Steady state

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(2) Cylindrical symmetry
(3) Heat of reaction is generated or absorbed at the surface of the solid particles.
(4) The temperature distribution is considered to be uniform within a given solid particle.
(5) The reduction of the iron oxide pellets is represented by the three interface model.
(6) The equation of motion for gas is given by the Ergun equation.21

Within the framework of these assumptions the governing equations have to express the conservation of the gaseous and the solid reactants, the heat balance on the gas stream, the heat balance on the solid stream and the equation of motion for the gas stream. The additional relationships needed to specify the problem are the equation of continuity, i.e., the overall mass balance, written for both the solid and the gas streams, and an appropriate kinetic expression for the reaction of the solids.

The dimensionless form of the governing equations may then be written as follows.

Material balance on the hydrogen gas:

\[ -\alpha_t \cdot (P/R) \cdot (P/T_a) \cdot \frac{\partial^2 X_{Hi}}{\partial z^2} - \alpha_t \cdot (P/R) \cdot (P/T_a) \cdot \left( \frac{\partial X_{Hi}}{\partial z} \right) = 0 \]

\[ \left( \frac{\partial P}{\partial r} \right) = \left( \frac{\partial T_a}{\partial r} \right) \]

\[ \frac{\partial^2 T_a}{\partial z^2} - \frac{\partial T_a}{\partial z} \]

\[ \left( \frac{1 + 2 \cdot \frac{\partial P}{\partial r} - 2 \cdot \frac{\partial T_a}{\partial r} \right) \cdot \frac{\partial X_{Hi}}{\partial r} + \left( F_i + X_{Hi} \right) \]

\[ \frac{\partial G_{es}}{\partial z} \]

Heat balance on the solid particles:

\[ \frac{\partial G_{es}}{\partial z} = \left( \frac{L}{Rr} \right) \cdot \frac{\partial (rG_{es})}{\partial r} - F_{es} R^* = 0 \]

Heat balance on the gas:

\[ \frac{\partial C_y}{\partial z} \]

\[ \left( \frac{C_i - C_i M_i}{\partial z} \right) \cdot \frac{\partial X_{Hi}}{\partial r} \]

where,

\[ \frac{\partial C_y}{\partial z} = \left( \frac{\partial t}{\partial z} \right) \]

Heat balance on the solid particles:

\[ \left( \frac{L}{Rr} \right) \cdot \frac{\partial k_i}{\partial z} + \frac{\partial k_i}{\partial r} \cdot \frac{\partial t}{\partial z} \]

\[ \left( \frac{R}{L} \right) \cdot \frac{\partial C_y}{\partial z} \]

\[ \left( \frac{R}{L} \right) \cdot \frac{\partial C_y}{\partial z} \]

In order to determine the flow field of the gas, Ergun equation* is used for the equation of motion in the z

\[ J_{G_s} = f_1 G_s + f_2 G_e \]

Jeschar and Radestock22,23 pointed out that Eqs. (8) and (9) are not invariant to the transformation of the coordinates. However, Stanek and Szekely8,10 revealed that little error is introduced in using these equations in the case of largely parallel flow field; for this reason in this paper, Eqs. (8) and (9) will be used as the equation of motion for gas.

* Ergun originally proposed the following equation as an empirical relationship between the pressure drop and the mass velocity.

\[ \frac{J_{G_s}}{J_{G_e}} = f_1 G_s + f_2 G_e \]
and the \( r \) directions, respectively.

\[
\frac{\partial P}{\partial z} = f(G_r + f_2G_r | G_r)
\]

The equation of continuity is given as Eq. (10) for the flow field with a chemical reaction

\[
R \frac{\partial (G_r)}{\partial r} + \frac{\partial}{\partial r} \left\{ F_2 \int_0^r R^* r dr \right\} = 0
\]

As discussed in earlier publications it is convenient to work in terms of the stream function, defined as follows:

\[
\frac{\partial \phi}{\partial r} = r G_r R \tag{11}
\]

\[
\frac{\partial \phi}{\partial z} = r G_r - F_2 \int_0^r R^* r dr \tag{12}
\]

We may now proceed to combine Eqs. (8) and (9) through the use of the stream function to obtain the following single expression:

\[
\left( \frac{L^2}{R} \right) \left\{ f_1 + (2f_2 f_3) \right\} \left\{ \frac{\partial \phi}{\partial r} \right\} \left\{ \frac{\partial \phi}{\partial r^2} - \frac{1}{r} \frac{\partial \phi}{\partial r} \right\} + \frac{\partial f_1}{\partial r}
\]

\[
+ \left( \frac{L}{R^*} \frac{\partial f_2}{\partial r} \right) \left\{ \frac{\partial \phi}{\partial r} \right\} + f_1 \left( \frac{\partial \phi}{\partial r} \right)^2 + \frac{\partial f_1}{\partial r}
\]

\[
+ \frac{\partial f_1}{\partial r} \left( I_B \frac{\partial f_2}{\partial r} + f_2 \frac{\partial f_1}{\partial r} \right) + f_2 \left( \frac{\partial \phi}{\partial r} \right)^2
\]

\[
- \frac{2 \cdot \frac{\partial f_1}{\partial z} \left( I_B \frac{\partial f_2}{\partial z} + f_2 \frac{\partial f_1}{\partial z} \right) + f_2 \left( \frac{\partial \phi}{\partial z} \right)^2
\]

\[
- I_B \left( \frac{\partial f_1}{\partial z} \right) \left( I_B \frac{\partial f_2}{\partial z} + f_2 \frac{\partial f_1}{\partial z} \right) = 0
\]

where,

\[
I_B = \int_0^r F_2 R^* r dr
\]

\( n = 1 \) when \( \frac{\partial \phi}{\partial z} \leq \int_0^r F_2 R^* r dr \)

\( n = 2 \) when \( \frac{\partial \phi}{\partial z} > \int_0^r F_2 R^* r dr \)

The overall pressure drop across the bed may then be calculated from the following equation:

\[
\Delta P = \int_0^r 2dr \int_0^r \left( \frac{\partial P}{\partial z} \right) dz \tag{14}
\]

The pressure field can be calculated from Eq. (15) which is also derived from Eqs. (8) and (9).

\[
\frac{\partial P}{\partial z} + \frac{\partial P}{\partial r} \left( \frac{1}{r} \frac{\partial \phi}{\partial r} - \frac{\partial P}{\partial r} \right) = - \left( \frac{R}{L} \right) \frac{f_1}{f_2 G_r}
\]

\[
+ \frac{f_2 G_r | G_r |}{r} \frac{\partial}{\partial r} \left( \frac{f_1}{f_2 G_r} + f_2 G_r | G_r | \right)
\]

\[
+ \frac{\partial g}{\partial z} \left( f_1 G_r + f_2 G_r | G_r | \right) \tag{15}
\]

### III. Reaction Rate

The three interface kinetic model which was developed by Hara, et al.,\(^{18}\) is used in this paper. The progressive advancement of the three reaction interfaces, \( \text{Fe/FeO, FeO/Fe}_2\text{O}_4 \) and \( \text{Fe}_2\text{O}_3/\text{Fe}_3\text{O}_4 \) may then be calculated from the following expressions:

\[
dX_I = \frac{L}{u_1} \frac{V_1}{4\pi R^*} \cdot \left\{ (V_1 + V_2 + V_3)/R^*_I \right\} \tag{16}
\]

\[
dX_2 = \frac{L}{u_2} \frac{V_2}{4\pi R^*} \cdot \left\{ (V_1 + V_2 + V_3)/R^*_2 \right\} \tag{17}
\]

\[
dX_3 = \frac{L}{u_3} \frac{V_3}{4\pi R^*} \cdot \left\{ (V_1 + V_2 + V_3)/R^*_3 \right\} \tag{18}
\]

The overall reaction rate \( R^* \), the overall heat of reaction \( (-\Delta H_R^+) \) and the reaction rate for each reaction given by Eqs. (1) to (3), \( V_1, V_2, V_3 \) are given below.

**Overall reaction rate:**

\[
R^* = \frac{6(1 - \varepsilon)}{\pi d_B^2} \cdot \left\{ (V_1 + V_2 + V_3)/R^*_I \right\} \tag{19}
\]

**Overall heat of reaction:**

\[
(-\Delta H_R^+) = \frac{6(1 - \varepsilon)}{\pi d_B^2} \cdot \left\{ (-\Delta H_R^+) \cdot V_1 \right\}
\]

\[
+ \left\{ (-\Delta H_R^+) \cdot V_2 + (-\Delta H_R^+) \cdot V_3 \right\} \tag{20}
\]

The rate of the individual chemical reactions (1) to (3) given by the following Eqs. (21) to (23), respectively.

\[
V_1 = 4\pi r_0 \left\{ [(A_1 + B_2 + B_3 + F) + (A_2 + B_2 + B_3 + F)] \cdot (C_r - C_{a3}) - (B_1 + B_2 + B_3 + F) \right\}
\]

\[
+ (A_1 + B_1 + B_2)(A_2 + B_2 + B_3 + F)
\]

\[
+ A_2 \cdot (B_2 + B_3 + F) \cdot (C_r - C_{a3}) - (B_2 + (A_1 + B_1 + B_2) + (B_1 + B_2 + B_3 + F)) \cdot (C_r - C_{a3}) \right\} \tag{21}
\]

\[
V_2 = 4\pi r_0 \left\{ [(A_1 + B_1 + B_2) + (A_3 + B_3 + F)] \cdot (C_r - C_{a3}) - (B_1 + B_2 + B_3 + F) \right\}
\]

\[
+ (A_1 + B_1 + B_2)(A_2 + B_2 + B_3 + F)
\]

\[
+ A_2 \cdot (B_2 + B_3 + F) \cdot (C_r - C_{a3}) - (B_2 + (A_1 + B_1 + B_2) + (B_1 + B_2 + B_3 + F)) \cdot (C_r - C_{a3}) \right\} \tag{22}
\]

\[
V_3 = 4\pi r_0 \left\{ [(A_1 + B_1)(A_2 + B_2 + B_3 + F) + A_2\]

\[
\cdot (B_2 + B_3 + F) \cdot (C_r - C_{a3}) - (A_2 + (B_1 + B_2) + (B_1 + B_2 + B_3 + F)) \cdot (C_r - C_{a3}) \right\} \tag{23}
\]

where,

\[
W_1 = (A_1 + B_1) \cdot [(A_2 + (A_2 + B_1 + B_2 + F) + (A_2 + B_2) + (B_1 + B_2 + B_3 + F)]
\]

\[
\cdot (B_2 + B_3 + F)
\]

\[
+ A_2 \cdot (B_2 + B_3 + F) \cdot (C_r - C_{a3}) - (A_2 + (B_1 + B_2) + (B_1 + B_2 + B_3 + F)) \cdot (C_r - C_{a3}) \right\} \tag{24}
\]

\[
W_2 = 4\pi r_0 \left\{ [(A_1 + B_1 + B_2) + (A_3 + B_3 + F)] \cdot (C_r - C_{a3}) - (B_1 + B_2 + B_3 + F)
\]

\[
+ A_2 \cdot (B_2 + B_3 + F) \cdot (C_r - C_{a3}) - (B_2 + (A_1 + B_1 + B_2) + (B_1 + B_2 + B_3 + F)) \cdot (C_r - C_{a3}) \right\} \tag{25}
\]

\[
W_3 = 4\pi r_0 \left\{ [(A_1 + B_1)(A_2 + B_2 + B_3 + F) + A_2 \cdot (B_2 + B_3 + F) \cdot (C_r - C_{a3}) - (A_2 + (B_1 + B_2) + (B_1 + B_2 + B_3 + F)) \cdot (C_r - C_{a3}) \right\} \tag{26}
\]

where,
\[ W_2 = (A_2 + B_2) \cdot \left( A_3 + B_3 + F \right) + A_3 \cdot (B_3 + F) \]

Finally when the reduction of magnetite is completed, only one reaction interface remains between FeO and Fe. At this stage we have:

\[ V_1 = 0 \] (27)
\[ V_2 = 0 \] (28)
\[ V_3 = 4r \frac{T_2(C_p - C_{eq})}{W_3} \] (29)

where,

\[ W_3 = A_3 + B_3 + F \]
\[ A_1 = 1 \cdot \frac{X^2}{k_2(1 + K_3)} \]
\[ A_2 = 1 \cdot \frac{X^2}{k_3(1 + K_3)} \]
\[ A_3 = 1 \cdot \frac{X^2}{k_4(1 + K_3)} \]
\[ B_1 = (X_2 - X_1) \frac{D_{12} X_1 X_2}{D_{13} X_1 X_2} \]
\[ B_2 = (X_3 - X_2) \frac{D_{12} X_2 X_3}{D_{13} X_1 X_3} \]
\[ B_3 = (1 - X_3) \frac{D_{12} X_2 X_3}{D_{13} X_1 X_3} \]
\[ F = 1 / k_1 \]

IV. Rate Constants, Equilibrium Constant and Heat of Reaction

Rate constants and equilibrium constants used in the reaction model were taken from Hara, et al.\(^{18}\) These expressions take the following forms:

\[ k_1 = 1.44 \times 10^6 \exp \left[ -6.650 / (t' + 273) \right] \] (30)
\[ k_2 = 2.88 \times 10^6 \exp \left[ -8.000 / (t' + 273) \right] \] (31)
\[ k_3 = 2.43 \times 10^6 \exp \left[ -14.000 / (t' + 273) \right] \] (32)

The effective diffusivity of hydrogen through the porous intermediate solid phases was estimated from the following:

\[ D_{c1} = 0.13 D_H \] (33)
\[ D_{c2} = 0.20 D_H \] (34)
\[ D_{c3} = 0.35 D_H \] (35)

The pore diffusion coefficients \( D_{c1}, D_{c2} \) and \( D_{c3} \) depend on the solid structure. The numerical values selected were quoted from Hara's work.\(^{15,18}\)

The mass transfer coefficient for the pellet was taken from Ranz's equation\(^{23}\) for packed beds.

\[ Sh = 2.0 + 0.6 (9 Re_p)^{1/2} (Sc)^{1/3} \] (36)

The equilibrium constants for the reactions (1) to (3) are given as follows:

\[ K_1 = \exp \left[ 362 / (t' + 273) + 10.32 \right] \] (37)
\[ K_2 = \exp \left[ -8.580 / (t' + 273) + 8.98 \right] \] (38)
\[ K_3 = \exp \left[ -2.070 / (t' + 273) + 1.30 \right] \] (39)

These equations were derived by Hara, et al.\(^{18}\) from data observed by Emmett and Schultz.\(^{24}\) The equations for the heat of reaction were derived from the data given by von Bogdandy and Engell.\(^{23}\)

\[
(- \Delta H_2^0) = \begin{cases} 
2.60 t' + 2220 & 100 \leq t' \leq 300^\circ C \\
3000 & 300 \leq t' \leq 800^\circ C \\
3.61 t' + 112 & 800 \leq t' \leq 1300^\circ C 
\end{cases} 
\] (40)

\[
(- \Delta H_3^0) = \begin{cases} 
10.37 t' - 21090 & 100 \leq t' \leq 911^\circ C \\
-11640 & 911 \leq t' \leq 1300^\circ C 
\end{cases} 
\] (41)

\[
(- \Delta H_3^0) = \begin{cases} 
3.38 t' - 5810 & 100 \leq t' \leq 500^\circ C \\
-3870 & 500 \leq t' \leq 927^\circ C \\
3.37 t' - 6990 & 927 \leq t' \leq 1300^\circ C 
\end{cases} 
\] (42)

V. Appropriateness of the Assumptions Made in the Development of the Model

The previously given Eqs. (4) to (42) represent the complete statement of the problem. However, in order to render the solution of these equations a manageable task, it is helpful to make a number of simplifying assumptions. The following assumptions were made:

1. Axial dispersion of heat and matter in the gas phase were neglected. This assumption is reasonable, because of the relatively high linear gas velocities encountered in most shaft furnaces.
2. The viscous term was neglected in the Ergun equation. The appropriateness of this assumption for high linear gas velocities has been discussed by Stanek and Szekely.\(^{8,10}\)
3. The solids were assumed to pass through the system in plug flow, i.e., the radial solids flow component was assumed to be zero.

This assumption was made because of the lack of information on solids flow patterns in shaft furnaces. It is noted that the form adopted for the solids flow equation still allows for a radially varying axial solids mass velocity, because both the void fraction and the density of the solid particles are allowed to vary both with the radial and with the axial coordinates.

Within the framework of the above assumptions the governing equations may be written as:

Material balance on the hydrogen:

\[
-\alpha_1 \cdot (P_1 / P_0) \cdot (P / T_a) \cdot \frac{\partial^2 X_{H_2}}{\partial r^2} + \left[ F_i \cdot G_i - \alpha_1 \cdot (P_1 / P_0) \cdot \frac{\partial X_{H_2}}{\partial r} \right] = 0 
\] (43)

Material balance on the solids:

\[
\frac{\partial G_{si}}{\partial z} - F_s R^* = 0 
\] (44)

Heat balance on the gas:

\[
\frac{R}{L} \cdot G_t \cdot \left[ \frac{\partial T}{\partial z} + T \cdot \frac{\partial G_t}{\partial z} \right] + \left[ G_s \cdot T \cdot \frac{\partial C_s}{\partial r} + \left( G_t \cdot C_t \right) \right] - \beta \cdot \left( \frac{\partial k_0 + k_3}{\partial r} \right) \cdot \frac{\partial T}{\partial r} - \beta \cdot \frac{\partial^2 T}{\partial r^2} = \gamma \cdot (T - t) + F_2 \cdot C_{eq} \cdot TR^* = 0 
\] (45)
Heat balance on the solids:

\[
\begin{align*}
\left(\frac{R}{L}\right)\frac{\partial}{\partial t}k_x\frac{\partial T}{\partial z} + \frac{R}{L}\left[G_s, \left\{ G_s, \frac{\partial T}{\partial z} + t, \frac{\partial C_s}{\partial z} \right\} \right. & \\
+ \frac{\partial}{\partial z}\left(k_x\frac{\partial T}{\partial z}\right) + \frac{\partial k_x}{\partial z}\frac{\partial T}{\partial z} + \frac{\partial}{\partial r}\left(k_x\frac{\partial T}{\partial r} + \frac{k_x}{r}\frac{\partial T}{\partial r}\right) & \\
+ C_iF_sR^* + (-\Delta H)\cdot C^* + r_s\cdot(T - t) = 0 \quad \cdots \quad (7')
\end{align*}
\]

The equation for the stream function:

\[
\left(\frac{L}{R}\right)\nabla \cdot \left[ \frac{\nabla \phi}{\partial z} \right] + (-1)^{n}\nabla \cdot \left[ \frac{\nabla \phi}{\partial z} \right] - 2\nabla \cdot \left( \frac{\partial \phi}{\partial z} \right) I_R = 0 \quad \cdots \quad (13')
\]

The axial and radial distributions of process variables such as \(X_{hi}, G_s, G_r, G_{sr}, X_1, X_2, X_n, T\) and \(t\) can be obtained by solving Eqs. (4') to (7'), (11), (12), (13') and (16) to (18) simultaneously with the aid of proper boundary conditions to be discussed subsequently and reaction rate which has been described.

**VI. Boundary Conditions**

The reducing gas is introduced and the solid product is discharged at the bottom of the moving bed. Upon assuming a uniform gas flow at the inlet, the boundary conditions at \(z=0\) are given as follows:

\[
\begin{align*}
G_s &= 1 \\
G_r &= 0 \\
X_{hi,0} &= \text{const.} \\
X_{hi,0} &= \text{const.} \\
X_{hi,o} &= \text{const.} \\
\frac{\partial T}{\partial z} &= 0 \\
T &= 1 \\
P_0 &= \text{const.}
\end{align*}
\]

The boundary conditions at the top of the bed are given as follows:

\[
\begin{align*}
G_r &= 0 \\
\frac{\partial X_{hi}}{\partial z} &= 0 \\
G_{hi} &= u_0\rho_s(1-\varepsilon) \\
X_1 &= \frac{\varepsilon}{1 - \varepsilon} \left[ X_1 - \left( \frac{5.88}{71.85} \right) \left( \frac{3C_{F,10}}{C_F} \right) \right] \\
X_2 &= 1 \\
X_3 &= 1
\end{align*}
\]

\[
\frac{\partial T}{\partial z} = 0 \\
\frac{\partial P}{\partial z} = 0
\]

Symmetry at the central axis is expressed as:

\[
\begin{align*}
G_r &= 0 \\
\frac{\partial X_{hi}}{\partial r} &= 0 \\
\frac{\partial t}{\partial r} &= 0 \\
\frac{\partial T}{\partial r} &= 0 \\
\frac{\partial P}{\partial r} &= 0
\end{align*}
\]

\(T\) = 1 (49)

\[
\begin{align*}
\text{The wall is impervious to gas; however, heat can be transferred through the wall; thus we have:
\}
\]

\[
\begin{align*}
G_r &= 0 \\
\frac{\partial X_{hi}}{\partial r} &= 0 \\
-k_s\frac{\partial t}{\partial r} &= (R/\kappa_s)h_s(T - T_{wa}) \\
-k_s\frac{\partial T}{\partial r} &= (R/\kappa_s)h_s(T - T_{wa}) \\
\frac{\partial P}{\partial r} &= 0
\end{align*}
\]

**VII. Transport Coefficients**

The viscosity of the gas mixture was determined using a procedure suggested by Bird, et al.\(^{26}\) and Licht, et al.\(^{27}\)

The effective thermal conductivity of the gas was estimated from Eq. (47)\(^{28}\)

\[
k_g' = \mu(1 + h_s\rho_p/\kappa_p) + 1.5(1 - \varepsilon)\rho_p/\mu \quad \cdots \quad (47)
\]

where,

\[
h_{re} = \left[ \frac{0.1592}{2(1 - \varepsilon)} \right] \left( \frac{1}{1 + \varepsilon} \right) \left( T' + 273 \right)^3
\]

The thermal conductivity of the gas mixture was obtained from the procedure described by Bird, et al.\(^{30}\)

The effective thermal conductivity of the solids was evaluated from Eq. (48) proposed by Lahiri and Seshadri\(^{30}\) on the basis of the equation given by Schotte.\(^{30}\)

\[
k_s' = 1.972 \times 10^{-7} \varepsilon_d \rho_d(T' + 273)^3 \quad \cdots \quad (48)
\]

The effective diffusivity of the reducing gas in radial direction was calculated from Eq. (49) which was obtained experimentally by Bernard and Wilhelm.\(^{30}\)

\[
P_{cr} = d_{\mu}/D_{hi} = 11 \quad \cdots \quad (49)
\]

The diffusivity of hydrogen in the gas mixture was determined by using a procedure proposed by Wilke\(^{31}\) and Andrusow.\(^{32}\)

**VIII. Numerical Computation**

The mathematical model developed in this paper consists of a set of nonlinear partial and ordinary differential equations. Therefore, machine computation has to be used to obtain the solution. The successive over-relaxation method was applied to solve the second order elliptic and parabolic type partial differential equations while simple difference equa-
tions were used for the first order ordinary differential equations.

The results of the computation will be presented in the following paper. Here we note that three specific cases were considered, namely:

1) Uniform gas flow and isothermal conditions
2) Distributed gas flow and isothermal conditions
3) Distributed gas flow and non-isothermal conditions.

The compilation of the program required some 12 to 16 sec of computer time, while the actual execution of the program needed some 900 to 2 000 sec on a CDC 6 400 digital computer.

Most of the calculations were carried out using a 101 x 11 grid and the convergence criterion employed is defined in Eq. (50):

$$\sum_{j=1}^{n} \left| \frac{Y_{i+1,j} - Y_{i,j}}{Y_{i,j}} \right| < \varepsilon \quad \text{(50)}$$

Here $\varepsilon$ ranged from 0.02 to 0.0001, $Y$ is a dependent variable such as $\psi$, $X_{mol}$, $P$, $t$ and $T$. Suffixes $l$, $i$, $j$ designate the iteration number and the mesh points in the axial and the radial direction, respectively.

**IX. Conclusions**

A generalized mathematical formulation is developed describing a moving bed reactor in which iron oxide pellets are reduced by hydrogen under steady-state conditions. This model consists of heat and mass balance equations written for the gas and the solid particles, of the equation of motion for gas, and of expressions representing the reduction kinetics of the iron oxide pellets, using the three interface model.

This formulation provides, for the first time, a comprehensive representation of the reduction of iron oxides with hydrogen in a countercflow arrangement, where the system is non-isothermal and where both the gaseous and the solids streams may be maldistributed. While the governing equations are complex, the model presented here is thought to be much more realistic than previous models of moving bed reactors systems, where maldistribution had been neglected.

A selection of the computed results will be given in the following paper, which will indicate a very strong coupling between the solids flow, gas flow, and the reaction kinetics. If this coupling is neglected, as was done in previous investigations, very marked distortion of the results may occur.

**Nomenclature**

- $C_{Fe}$: Total Fe content of iron oxide pellet
- $C_{FeO}$: FeO content of iron oxide pellet
- $C_a$: Molar density of gas at inlet condition (kmol/m$^3$)
- $C_r$: Molar concentration of reducing gas (H$_2$) in bulk (kmol/m$^3$)
- $C_{e1}$, $C_{e2}$, $C_{e3}$: Equilibrium concentration of gas for the reaction of Fe$_2$O$_3$ → Fe$_3$O$_4$, Fe$_3$O$_4$ → FeO and FeO → Fe (kmol/m$^3$)
- $C_{g}$, $C_{g}^*$: Dimensionless and dimensional specific heat of gas mixture (kcal/kg·°C)
- $C_{f}$, $C_{f}^*$: Dimensionless and dimensional molar specific heat of i-th species of gas (kcal/kgmol (°C))
- $C_{g0}$: $\sum (X_{ story }

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$k_1, k_2$: Effective thermal conductivity of solid pellets, dimensionless, dimensional and at the inlet condition (kcal/m·hr·°C)

$L$: Axial length of reactor (m)

$M_i$: Molar weight of $i$-th species of gas (kg/kg mol)

$\bar{M}$: Average molar weight of gas (kg/kgmol)

$P, P_c, P_b$: Dimensionless pressure, pressures at outlet and at inlet condition (kg/m²)

$P_r$: Prandtl number

$P_{ci}$: Critical pressure of $i$-th species of gas (atm)

$R$: Radius of reactor (m)

$R^*, R^b$: Dimensionless overall reaction rate and at inlet condition (kgmol (H₂)/m³ (bed) hr)

$r$: Dimensionless radial distance from the center in the reactor

$r_0$: Radius of the pellet (m)

$Re_p$: Reynolds number

$Sh$: Sherwood number

$Sc$: Schmidt number

$T, T^*, T_0$: Dimensionless and dimensional gas temperature and it at inlet condition (°C)

$T_a$: Dimensionless gas temperature $(T'/+273)/(T_o+273)$

$T_{am}$: Dimensionless ambient temperature

$T_{ci}$: Critical temperature of $i$-th species of gas (°K)

$T_r$: Reduced temperature $(T'+273)/(T_{ci})$

$t, t', t_0$: Dimensionless and dimensional solid temperatures and it at inlet condition (°C)

$u_i$: Velocity of solid (m/hr)

$V_1, V_2, V_3$: Reaction rate from Fe₂O₃ to Fe₃O₄, Fe₃O₄ to FeO, and FeO to Fe (kg·mol(H₂)/hr·particle)

$v_{ci}$: Molar volume of $i$-th species of gas (ml/mol)

$X_1, X_2, X_3$: Dimensionless radial distance of reaction interface from the center of the pellet

$X_i$: Molar fraction of $i$-th species of gas

$z$: Dimensionless axial distance from the bottom of the reactor

$\alpha_1, \alpha_2$: $M_{21}D_{21}C_{20}G_{20}R, M_{21}D_{21}C_{20}G_{20}R$

$\beta, \beta_i$: $k_2^v/RG_{20}C_{20}, k_2^v/RG_{20}C_{20}$

$\gamma, \gamma_i$: $6(1-\varepsilon_i)h_pR/G_{20}G_{20}d_{20}d_{20}$, $6(1-\varepsilon)h_pR/G_{20}G_{20}d_{20}d_{20}$

$\delta, \delta_i$: $k_2^vC_{20}G_{20}L, k_2^vC_{20}G_{20}L$

$\varepsilon$: Voidage in the reactor

$\zeta$: $RR^b(\Delta H_{f20}^\circ)/C_{20}d_{20}d_{20}$

$\mu_{mix}$: Viscosity of gas mixture (kg/m·hr)

$\mu_i$: Viscosity of $i$-th species of gas (kg/m·hr)

$\rho$: Density of gas (kg/m³)

$\rho_a$: Apparent density of solid pellet (kg/m³ (solid))

$\phi$: Shape factor

$\phi$: Stream function

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**REFERENCES**