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and A. A. Jonke

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A SLUG FLOW MODEL FOR COAL COMBUSTION AND DESULFURIZATION IN FLUIDIZED BEDS: THEORETICAL FORMULATION

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NOMENCLATURE

A	bed cross-sectional area
C_0	initial oxygen concentration in the fluidizing gas
$\overline{C}_{O_2}$	average concentration of oxygen in the bed, defined by Eq. 15
C_{e,O_2}	concentration of oxygen in the emulsion phase
C_{e,SO_2}	concentration of sulfur dioxide in the emulsion phase
C_{s,O_2}	concentration of oxygen in the bubble or slug phase
C_{s,SO_2}	concentration of sulfur dioxide in the bubble or slug phase
C_{O_2}	oxygen concentration in general
C_{SO_2}	sulfur dioxide concentration in general
$\overline{C}_{SO_2}$	average concentration of sulfur dioxide in the bed, defined by Eq. 16
D	bulk gas diffusion coefficient
D_c	bed diameter
d_b	bubble diameter
d_{bo}	initial bubble diameter at the gas distributor surface
d_{bm}	maximum attainable bubble diameter
$E(t)$	solids residence time distribution function
F	volumetric flow rate of solids through the bed
F_c	volumetric feed rate of coal
F_d	volumetric feed rate of sorbent
f_a	volume fraction of ash in the bed
f_c	volume fraction of coal in the bed
f_d	volume fraction of sulfated sorbent in the bed
g	acceleration of gravity
H	total bed height
h	bed height in general
h_s	the bed height above which slug flow regime prevails
I	an integral along the slug surface
L_s	slug length
M_C	molecular weight of carbon
M_d	molecular weight of the calcium compound in additive
M_H	molecular weight of hydrogen
M_N	molecular weight of nitrogen
M_O	molecular weight of oxygen
M_S	molecular weight of sulfur
n_d	number of orifices in the perforated gas distribution plate
n_{O_2}	moles of oxygen
n_{SO_2}	moles of sulfur dioxide
$P_b(R)$	char particle size distribution function in the bed on volume basis
$P'_b(R)$	char particle size distribution function in the bed on particle number basis
$P_b(X)$	sulfur loading distribution function of sorbent particles in the bed on volume basis
$P'_b(X)$	sulfur loading distribution function of sorbent particles in the bed on particle number basis
Q_{se}	gas-exchange rate between the particulate and slug or bubble phases per slug or bubble
q_c	mass flow rate of coal
q_d	mass flow rate of fresh sorbent
R	coal particle radius
R_c	coal particle radius in the feed
R_d	sorbent particle radius in the feed
$\mathcal{R}_c(R)$	combustion rate constant defined by Eq. 1
$\mathcal{R}_d(X)$	combustion rate constant defined by Eq. 3
t	time
U_o	superficial gas velocity
U_s	bubble or slug velocity
U_{mf}	minimum fluidization velocity
V_s	bubble or slug volume
X	conversion of sorbent

Greek Letters

γ_a	weight fraction of ash in the coal
γ'_a	weight fraction of ash in the char
γ_C	weight fraction of carbon in the coal
γ'_C	weight fraction of carbon in the char
γ''_C	weight fraction of carbon in the volatiles released from coal
γ_d	weight fraction of calcium compound in the sorbent
γ_H	weight fraction of hydrogen in the coal
γ'_H	weight fraction of hydrogen in the char
γ''_H	weight fraction of hydrogen in the volatiles released from coal
γ_N	weight fraction of nitrogen in the coal
γ'_N	weight fraction of nitrogen in the char
γ''_N	weight fraction of nitrogen in the volatiles released from coal
γ_O	weight fraction of oxygen in the coal
γ'_O	weight fraction of oxygen in the char
γ''_O	weight fraction of oxygen in the volatiles released from coal
γ_S	weight fraction of sulfur in the coal
γ'_S	weight fraction of sulfur in the char
γ''_S	weight fraction of sulfur in the volatiles released from coal
γ_v	weight fraction of volatiles in the coal
ρ_c	coal particle density
ρ'_c	char particle density
ρ_d	sorbent particle density
δ	volume fraction of bubbles or slugs in the bed
ε_{mf}	bed voidage at minimum fluidization
η_1	combustion efficiency defined by Eq. 52
η_2	combustion efficiency defined by Eq. 53

A SLUG FLOW MODEL FOR COAL COMBUSTION AND DESULFURIZATION IN FLUIDIZED BEDS: THEORETICAL FORMULATION

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ABSTRACT

A model is developed for coal combustion in fluidized beds including devolatilization under slugging conditions, with sulfur removal effected by a sorbent of limestone or dolomite. The gas-flow analysis is based on the slug flow theory of Hovmand and Davidson with modifications to account for the bubbling region below the point where slug formation starts. Solids are assumed to be back-mixed, and for this condition, char-size distributions and sorbent sulfate loadings in the bed are determined. The gaseous reactant balances are carried out for both oxygen and sulfur dioxide under plug flow conditions, with the reaction rates based on coal size and sorbent sulfate loading. The expressions for the combustion efficiency and sulfur reaction are derived.

I. INTRODUCTION

A new combustion process for coal is currently under extensive study by various organizations. It involves a fluidized-bed operation with *in situ* sulfur removal by limestone or dolomite sorbent. The feasibility of this process for reducing sulfur dioxide emission to an acceptable level, as well as for maintaining a sufficiently high combustion efficiency, has been demonstrated by several pilot-plant operations such as those discussed by Jonke *et al.*,[1] Ehrlich,[2] Hammons and Skopp,[3] Hoke *et al.*,[4] Rice and Coates,[5] Combustion Power Company,[6] and National Research and Development Corporation.[7] The advantages of this fluidized-bed combustion process as opposed to the conventional pulverized-coal boiler have been discussed by Squires.[8]

Models have been proposed by Bethell *et al.*,[9] O'Neill and Keairns,[10] Jonke *et al.*,[11-13] Horio and Wen[14] for sulfur removal by this process, assuming that the bed is composed entirely of sorbent particles. However, it is known that the bed may also contain a significant amount of coal-derived ash and uncombusted coal particles. Also, since these models do not consider simultaneous coal combustion, the sulfur dioxide generation patterns they assume remain open to question. A complete treatment of the reactions in the bed is given by Chen and Saxena[15] and shall be applied here.

The effects of gas bypassing through bubbles on the reaction performance of the combustor have been investigated by Horio and Wen[14] and Chen and Saxena.[15-16] However, these analyses were limited to uniformly bubbling beds.

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Because of the high gas-solids feed ratio required for coal combustion, the beds may frequently be slugging. Furthermore, Matsen and Tarmy[17] pointed out that scale-up of slugging beds tends to be more successful and reliable than scale-up of uniformly bubbling beds.

A pertinent slug flow theory has been given by Hovmand and Davidson.[18] Stewart and Davidson[19] have discussed two types of slug flow, and the present work is confined to what they called type A, in which slugs rise through the bed of particles like large bubbles in a liquid. According to Stewart,[20] this type of slug flow is of more general interest and is likely to occur in gas-fluidized beds with $H/D_c > 1$ when $(U_o - U_{mf})/0.35(gD_c)^{1/2}$ is greater than about 0.2. In the application of slug flow theory to this work, a modification is made to account for the bubbling region below the slug flow region. Since the gas-exchange rate of the bubbles with the emulsion phase is relatively large as compared to that of the slugs with the emulsion phase, a considerable difference in the rate of coal combustion will be observed in the two cases. The effects of physico-chemical changes of the coal and sorbent particles on the reaction rates of combustion and sulfation are also noted in this work.

II. DESCRIPTION OF THE SYSTEM

Typically the coal combustor is a fluidized bed, with coal and sulfur-removing sorbent fed continuously at the bed base at mass flow rates of q_c and q_d, respectively. The bed is fluidized by atmospheric or pressurized air at a velocity U_o and oxygen concentration C_0. The sorbents are usually limestone and/or dolomite, which differ from each other in relative concentrations of calcium carbonate and magnesium carbonate. They can be used either directly or pretreated to the half-calcined and fully calcined forms before being fed into the bed. Only the calcium compound in the sorbents is reactive toward sulfation, and its weight fraction is denoted here as γ_d.

Various grades of coal, ranging from lignite to bituminous coal, have been successfully combusted in this type of reactor. The weight fractions of volatile matter, ash, carbon, hydrogen, oxygen, sulfur, and nitrogen in the coal, as determined from proximate and ultimate analyses, are denoted γ_v, γ_a, γ_c, γ_H, γ_O, γ_S and γ_N, respectively. For most types of coal burned in fluidized beds, ash will strip off after its formation at the coal surface, resulting in shrinkage of the coal particle size from an initial radius, R_c. The sorbents, on the other hand, retain approximately the same particle size, R_d, during sulfation in the bed.

In many fluidized bed combustors, coal will quickly lose its volatile matter at the bottom region of the bed. The char so generated will become sparsely dispersed among the ash and sorbent particles in the bed because of its rapid consumption by combustion. This combustion environment is very similar to the ash bed investigated by Avedesian and Davidson.[21] Their results give the molar consumption rate of oxygen by a coal particle of radius R as:

$$-\frac{dn_{O_2}}{dt} = C_{O_2}\mathcal{R}_c(R)$$

$$= 8\pi DRC_{O_2} \qquad (1)$$

The combustion of volatile matter released from coal in the gas phase proceeds much faster than that of char and therefore is assumed to be completed in a region close to the coal feed point. The char obtained from this devolatilization process is further assumed to retain the same particle size as the coal feed, and its content of ash, carbon, hydrogen, oxygen, sulfur, and nitrogen by weight fraction will be denoted by γ_a', γ_C', γ_H', γ_D', γ_S', and γ_N', respectively. Further, if the weight fractions of carbon, hydrogen, oxygen, sulfur, and nitrogen in the volatile matter released from coal are represented by γ_C''; γ_H''; γ_O''; γ_S''; and γ_N''; respectively, we have the following relations:

$$\begin{aligned}
\gamma_a' &= \gamma_a/(1 - \gamma_v) \\
\gamma_C' &= (\gamma_C - \gamma_v\gamma_C'')/(1 - \gamma_v) \\
\gamma_H' &= (\gamma_H - \gamma_v\gamma_H'')/(1 - \gamma_v) \\
\gamma_O' &= (\gamma_O - \gamma_v\gamma_O'')/(1 - \gamma_v) \\
\gamma_S' &= (\gamma_S - \gamma_v\gamma_S'')/(1 - \gamma_v)
\end{aligned} \tag{2}$$

and

$$\gamma_N' = (\gamma_N - \gamma_v\gamma_N'')/(1 - \gamma_v)$$

Numerous experimental studies have been made to investigate the sulfation kinetics. Some of these are Borgwardt,[22] Keairns *et al.*,[23] Hubble *et al.*,[24] O'Neill *et al.*,[25] Yang *et al.*,[26] and Wen and Ishida.[27] The reaction is found to be first order with respect to the sulfur dioxide concentration, C_{SO_2}. Therefore, the molar consumption rate of sulfur dioxide for a sorbent particle of conversion level X can be written in the following form:

$$-\frac{dn_{SO_2}}{dt} = C_{SO_2} R_d(X) \tag{3}$$

A closed form is not given for the $R_d(X)$ in Eq. 3 because it has been observed to be largely dependent on the type of sorbent used. A good correlation of $R_d(X)$ with the sorbent chemical structure is not presently available. However, the value of $R_d(X)$ for a particular sorbent can be easily determined from the weight-change measurement of a given sample in a thermogravimetric analyzer. Although the gas-flow conditions encountered by the sorbent sample in such a measurement are usually not the same as in the combustor, the use of thermogravimetric analysis for finding the $R_d(X)$ is justified by the experimental evidence that the gas film diffusion is not the rate controlling step of the reaction.[22]

III. THE MODEL

A. The Gas-Flow Analysis

Bubbles formed at the gas distribution surface coalesce and grow while rising through the bed. The bubble diameter, d_b, according to Mori and Wen,[28] varies with the bed height, h as:

$$d_b = d_{bm} - (d_{bm} - d_{bo}) \exp\left[-0.15h/\sqrt{(A/\pi)}\right] \tag{4}$$

Here A is the bed cross-sectional area, and d_{bo} and d_{bm} are the initial and maximum attainable bubble diameters, respectively, as given below:

$$d_{bo} = 0.347[A(U_o - U_{mf})/n_d]^{2/5} \tag{5}$$

for a perforated gas distributor,

$$d_{bo} = 0.00376\ (U_o - U_{mf})^2 \tag{6}$$

for a porous gas distributor, and

$$d_{bm}' = 0.652[A(U_o - U_{mf})]^{2/5} \tag{7}$$

where all quantities are expressed in the cgs unit system. U_{mf} and n_d are the minimum fluidization velocity and the number of orifices on the perforated gas distributor plate, respectively. Mori and Wen[28] made a simplification that $U_o - U_{mf} << 0.711\sqrt{gd_{bm}}$ while expressing d_{bm} in the closed form given by Eq. 7. This simplification will not usually be valid in most fluidized-bed combustion operations because U_o is normally very large. Following their derivation, but without making this simplifying assumption, the following relation is derived for the determination of d_{bm}:

$$22.3d_{bm}^{2.5} + (U_o - U_{mf})d_{bm}^2 = 7.64\ A(U_o - U_{mf}) \tag{8}$$

For small-diameter columns or high gas velocities, the bubble may grow to a size comparable to the column diameter D_c at a bed height h_s above the gas distributor. The resulting slug flow will then persist in the remaining upper portion of the bed. According to Davidson and Harrison,[29] h_s may be evaluated from Eq. 4 as that value of h which gives a value for d_b approximately equal to 1/3 D_c. Therefore

$$h_s = 3.76\sqrt{A}\ \ln\left\{(d_{bm} - d_{bo})/[d_{bm} - (D_c/3)]\right\} \tag{9}$$

A schematic representation of these two different gas-flow patterns is shown in Fig. 1.

According to Davidson and Harrison[29] and Stewart and Davidson,[19] the bubble and slug velocities, both expressed as U_s, are given by Eqs. 10 and 11, respectively:

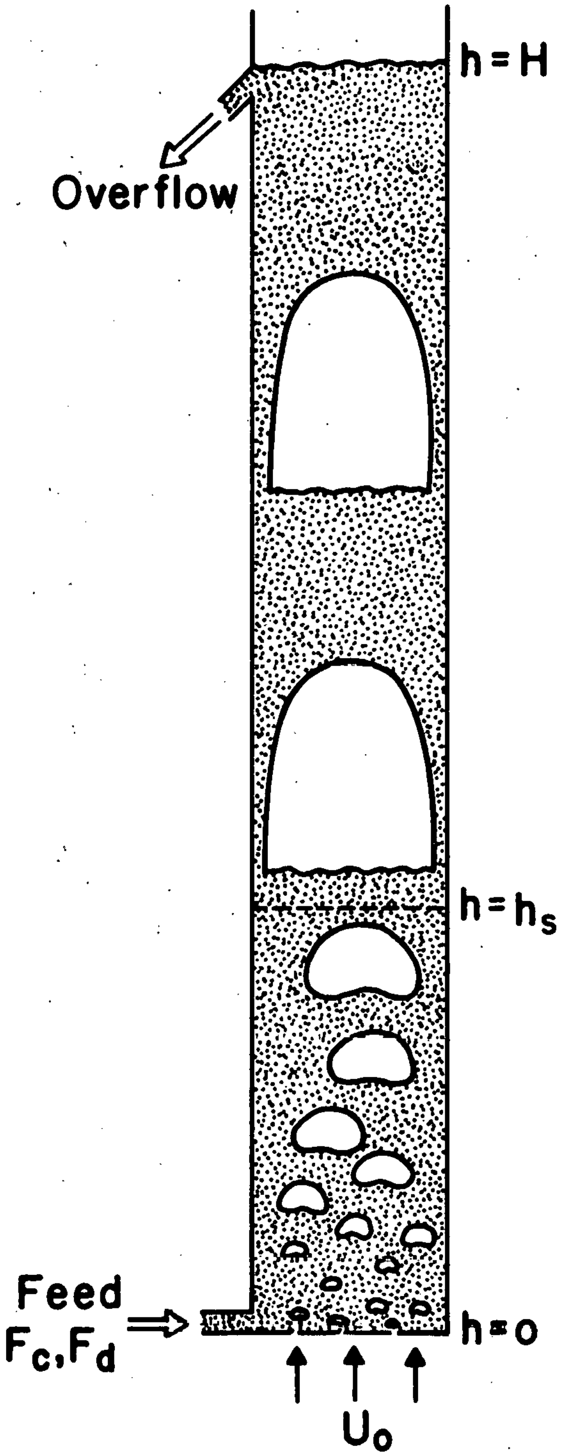

Fig. 1. Bubbling and Slugging Sections in a Fluidized Bed

$$U_s = U_o - U_{mf} + 0.711(gd_b)^{1/2} \tag{10}$$

for $h < h_s$ or $d_b < (D_c/3)$

and

$$U_s = U_o - U_{mf} + 0.35(gD_c)^{1/2} \text{ for } h > h_s \text{ or } d_b > (D_c/3) \tag{11}$$

Following the two-phase theory[29] which asserts that gas in excess of that required for minimum fluidization passes through the bed as gas pockets, the volume fraction of bubbles or slug in the bed is:

$$\delta = (U_o - U_{mf})/U_s \tag{12}$$

B. The Gas and Solids Reactant Balance

Since gas mixing in fluidized-bed combustors which have internal components is not a very efficient process, it is assumed here that plug flow is a good representation for gases in both the emulsion and bubble/slug phases. The gaseous concentrations of oxygen and sulfur dioxide in these two phases then become dependent on bed height. On the other hand, for solids, a back-mix pattern is assumed for all particles in the bed, in view of the high frequency of rising bubbles and slugs at the high gas throughput of the coal combustor.

1. The Solids Reactant Balance

Three kinds of solids, *viz.*, the ash, unburned coal, and sorbent particles, exist in the bed, and their volume fractions at steady state may be denoted by f_a, f_c and f_d, respectively. Further,

$$f_a + f_c + f_d = 1 \tag{13}$$

Since these solids are in back-mix condition in the bed, each one has a residence-time distribution given by:

$$E(t) = \frac{1}{(AH/F)} \exp[-t/(AH/F)] \tag{14}$$

where F is the volumetric flow rate of solids through the bed.

For converting the residence-time distribution into the distributions of char-particle size and sorbent sulfate loading, relations of t with R, as well as with X, have to be determined from Eqs. 1 and 3. Since the combustion and sulfation reactions are carried out only in the emulsion phase, the linearity of Eqs. 1 and 3 in C_{O_2} and C_{SO_2}, respectively, leads to a convenient way of defining mean values of these quantities, *viz.*,

$$\bar{C}_{O_2} = \int_0^H C_{e,O_2} \frac{dh}{H} \qquad (15)$$

and

$$\bar{C}_{SO_2} = \int_0^H C_{e,SO_2} \frac{dh}{H} \qquad (16)$$

It can be shown that the terms on the left side of Eqs. 1 and 3 can be expressed in the following forms, respectively,

$$-\frac{dn_{O_2}}{dt} = \frac{d}{dt}\left[\frac{4\pi}{3} R^3 \rho'_c \left(\frac{\gamma'_C}{M_C} + \frac{\gamma'_S}{M_S} + \frac{\gamma'_H}{2M_H} - \frac{\gamma'_O}{M_O}\right)\right] \qquad (17)$$

and

$$-\frac{dn_{SO_2}}{dt} = \frac{d}{dt}\left[4\pi R_d^3 \rho_d \gamma_d \; X/3M_d\right] \qquad (18)$$

Here, ρ' and ρ_d are the densities of char and sorbent particles, respectively. The quantities M_C, M_S, M_H, M_O and M_d are the molecular weights of carbon, sulfur, hydrogen, oxygen, and calcium compound, respectively. Equations 1 and 3 after substitution from Eqs. 15 through 18 assume the following forms:

$$\frac{dR}{dt} = -2D\bar{C}_{O_2} \Big/ \left[R \rho'_c \left(\frac{\gamma'_C}{M_C} + \frac{\gamma'_S}{M_S} + \frac{\gamma'_H}{2M_H} - \frac{\gamma'_O}{M_O}\right)\right] \qquad (19)$$

$$\frac{dX}{dt} = 3\bar{C}_{SO_2} \mathcal{R}_d(X) M_d \Big/ (4\pi R_d^3 \rho_d \gamma_d) \qquad (20)$$

On integration and rearrangement, the above two relations yield:

$$t(R) = \int_{R_c}^{R} \frac{R\rho'_c}{2D\bar{C}_{O_2}} \left(\frac{\gamma'_C}{M_C} + \frac{\gamma'_S}{M_S} + \frac{\gamma'_H}{2M_H} - \frac{\gamma'_O}{M_O}\right) dR \qquad (21)$$

and

$$t(X) = \int_0^X \frac{4\pi R_d^3 \rho_d \gamma_d}{3\bar{C}_{SO_2} \mathcal{R}_d(X) M_d}\, dX \qquad (22)$$

Let $P_b'(R)\,dR$ and $P_b'(X)\,dX$ represent, respectively, the number fractions of char and sorbent particles in the bed over a size increment dR and a conversion increment dX. These probability distribution fractions can be obtained using Eqs. 14 and 19 through 22 and are as follows:

$$P_b'(R) = E[t = t(R)] \left|\frac{dt}{dR}\right|$$

$$= \frac{RF\rho_c'}{2AHD\bar{C}_{O_2}} \left(\frac{\gamma_C'}{M_C} + \frac{\gamma_S'}{M_S} + \frac{\gamma_H'}{2M_H} - \frac{\gamma_O'}{M_O}\right)$$

$$\times \exp\left[\int_{R_c}^{R} \frac{RF\rho_c'}{2AHD\bar{C}_{O_2}} \left(\frac{\gamma_C'}{M_C} + \frac{\gamma_S'}{M_S} + \frac{\gamma_H'}{2M_H} - \frac{\gamma_O'}{M_O}\right) dR\right] \qquad (23)$$

$$P_b'(X) = E[t = t(X)] \left|\frac{dt}{dX}\right|$$

$$= \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AH\bar{C}_{SO_2} \mathcal{R}_d(X) M_d} \exp\left[-\int_0^X \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AH\bar{C}_{SO_2} \mathcal{R}_d(X) M_d}\, dX\right] \qquad (24)$$

$P_b'(X)$ becomes a delta function at X = 1 because the corresponding residence time could extend from t(X = 1) to infinity. This delta function can be obtained from the following:

$$\delta'(X = 1) = \int_{t(X=1)}^{\infty} E(t)dt = \exp\left[\frac{-Ft(X = 1)}{AH}\right]$$

$$= \exp\left[-\int_0^1 \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AH\bar{C}_{SO_2} \mathcal{R}_d(X) M_d}\, dX\right] \qquad (25)$$

Let $P_b(R)$, $P_b(X)$ and $\delta(X = 1)$ represent, respectively, the distribution functions $P_b'(R)$, $P_b'(X)$, and $\delta'(X = 1)$ evaluated on a solids volume basis. The following interrelations are valid for these two types of distribution functions:

$$\frac{Ff_cP_b(R)\ dR}{(4/3)\pi R^3} = \frac{q_cP_b'(R)\ dR}{(4/3)\pi R_c^3\rho_c} \tag{26}$$

$$\frac{Ff_dP_b(X)\ dX}{(4/3)\pi R_d^3} = \frac{q_dP_b'(X)\ dX}{(4/3)\pi R_d^3\rho_d} \tag{27}$$

$$\frac{Ff_d\delta(X = 1)}{(4/3)\pi R_d^3} = \frac{q_d\delta'(X = 1)}{(4/3)\pi R_d^3\rho_d} \tag{28}$$

where ρ_c is the particle density of coal feed. Based on Eqs. 26-28 and other relations given above, we have

$$P_b(R) = \frac{q_cR^3}{f_cF\rho_cR_c^3}P_b'(R)$$

$$= \frac{R^4q_c\rho_c'\left[\left(\gamma_C'/M_C\right) + \left(\gamma_S'/M_S\right) + \left(\gamma_H'/2M_H\right) - \left(\gamma_O'/M_O\right)\right]}{2Df_cAH\bar{C}_{O_2}R_c^3\rho_c}$$

$$\times \exp\left\{\int_{R_c}^{R}\frac{RF\rho_c'\left[\left(\gamma_C'/M_C\right) + \left(\gamma_S'/M_S\right) + \left(\gamma_H'/2M_H\right) - \left(\gamma_O'/M_O\right)\right]}{2AHD\bar{C}_{O_2}}dR\right\} \tag{29}$$

$$P_b(X) = \frac{q_d}{f_dF\rho_d}P_b'(X)$$

$$= \frac{4\pi R_d^3\gamma_dq_d}{3AH\bar{C}_{SO_2}f_d\mathcal{R}_d(X)M_d}\exp\left[-\int_0^X\frac{4\pi R_d^3\rho_d\gamma_dF}{3AH\bar{C}_{SO_2}\mathcal{R}_d(X)M_d}dX\right] \tag{30}$$

$$\delta(X = 1) = \frac{q_d}{f_dF\rho_d}\delta'(X = 1)$$

$$= \frac{q_d}{f_dF\rho_d}\exp\left[-\int_0^1\frac{4\pi R_d^3\rho_d\gamma_dF}{3AH\bar{C}_{SO_2}\mathcal{R}_d(X)M_d}dX\right] \tag{31}$$

Further, it is known that

$$\int_0^{R_c} P_b(R)\, dR = 1$$

and

$$\delta(X = 1) + \int_0^1 P_b(X)\, dX = 1 \tag{33}$$

Therefore, from Eqs. 29 and 32, we obtain

$$f_c = \int_0^{R_c} \frac{R^4 q_c \rho_c' \left[\left(\gamma_C'/M_C\right) + \left(\gamma_S'/M_S\right) + \left(\gamma_H'/2M_H\right) - \left(\gamma_O'/M_O\right)\right]}{2DAH\overline{C}_{O_2} R_c^3 \rho_c}$$

$$\times \exp\left\{\int_{R_c}^{R} \frac{RF\rho_c'\left[\left(\gamma_C'/M_C\right) + \left(\gamma_S'/M_S\right) + \left(\gamma_H'/2M_H\right) - \left(\gamma_O'/M_O\right)\right]}{2HAD\overline{C}_{O_2}}\, dR\right\} dR \tag{34}$$

Similarly, from Eqs. 30, 31, and 33, we obtain

$$f_d = \frac{q_d}{F\rho_d} \exp\left[-\int_0^1 \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AH\overline{C}_{SO_2} \mathcal{R}_d(X) M_d}\, dX\right] + \int_0^1 \frac{4\pi R_d^3 \gamma_d q_d}{3AH\overline{C}_{SO_2} \mathcal{R}_d(X) M_d}$$

$$\times \exp\left[-\int_0^X \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AH\overline{C}_{SO_2} \mathcal{R}_d(X) M_d}\, dX\right] dX \tag{35}$$

The ash residue stripped from the coal surface after combustion is much more compact than the coal feed. Assuming this compaction ratio to be about 1/6, we can relate the ash collected in the overflow stream to that generated in the bed as follows:

$$Ff_a = -\int_0^{R_c} \frac{AHf_c P_b(R)\ dR}{(4/3)\pi R^3} \cdot \frac{1}{6} \cdot \frac{d(4\pi R^3/3)}{dt}$$

$$= -\int_0^{R_c} \frac{AHf_c P_b(R) dR}{2R} \frac{dR}{dt} \qquad (36)$$

Substituting $\frac{dR}{dt}$ from Eq. 19 into Eq. 36, we obtain

$$Ff_a = \int_0^{R_c} \frac{D\overline{C}_{O_2} AHf_c P_b(R)}{R^2 \rho'_C \left[\left(\gamma'_C/M_C\right) + \left(\gamma'_S/M_S\right) + \left(\gamma'_H/2M_H\right) - \left(\gamma'_C/M_O\right)\right]}\ dR \qquad (37)$$

Replacing $P_b(R)$ in Eq. 37 by Eq. 29, we obtain

$$f_a = \frac{1}{F}\int_0^{R_c} \frac{R^2 q_c}{2R_c^3 \rho_c} \exp\left\{\int_{R_c}^{R} \frac{RF\rho'_c \left[\left(\gamma'_C/M_C\right) + \left(\gamma'_S/M_S\right) + \left(\gamma'_H/2M_H\right) - \left(\gamma'_O/M_O\right)\right]}{2AHD\ \overline{C}_{O_2}} dR\right\} dR \qquad (38)$$

substituting f_a, f_d, and f_c from Eqs. 38, 35, and 34 respectively, into Eq. 13, we obtain

$$1 = \int_0^{R_c} \frac{R^2 q_c}{2R_c^3 P_c} \left\{\frac{1}{F} + \frac{R^2 \rho'_c \left[\left(\gamma'_C/M_C\right) + \left(\gamma'_S/M_S\right) + \left(\gamma'_H/2M_H\right) - \left(\gamma'_O/M_O\right)\right]}{D\Lambda H\overline{C}_{O_2}}\right\}$$

$$\times \exp\left\{\int_{R_c}^{R} \frac{RF\rho'_c \left[\left(\gamma'_C/M_C\right) + \left(\gamma'_S/M_S\right) + \left(\gamma'_H/2M_H\right) - \left(\gamma'_O/M_O\right)\right]}{2AHD\overline{C}_{O_2}} dR\right\} dR$$

$$+ \frac{q_d}{F\rho_d} \exp\left[-\int_0^1 \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AHC_{SO_2} R_d(X) M_d}\ dX\right]$$

$$+ \int_0^1 \frac{4\pi R_d^3 \gamma_d \rho_d}{3AH\overline{C}_{SO_2} R_d(X) M_d} \exp\left[-\int_0^X \frac{4\pi R_d^3 \rho_d \gamma_d F}{3AH\overline{C}_{SO_2} R_d(X) M_d}\ dX\right] dX \qquad (39)$$

The only unknown in Eq. 39 is F, which can therefore be determined by trial and error. The f_a, f_c, f_d, $P_b(R)$, $P_b(X)$, and $\delta(X = 1)$ can then be determined on the basis of the F value so obtained.

2. The Gas Reactant Balance

The gas exchange between the slugs and the particulate phase plays an important role in the redistribution of gaseous reactants in the bed. For the present process, the transfer of oxygen from slugs to the particulate phase can be expected to be a limiting step for the coal combustion since the combustion rate is usually greater than the gas-exchange rate. On the other hand, the poorer gas exchange generally observed in a slugging bed may impede the transfer of sulfur dioxide from the particulate phase into the bypassing gas stream in the slug phase, resulting in higher sulfur retention. The gas-exchange rate is known to be a function of the slug dimensions. Equations have been derived by Hovmand and Davidson[18] for the slug length, L_s, and then the slug volume, V_s. These are:

$$\frac{L_s}{D_c} - 0.495\left(\frac{L_s}{D_c}\right)^{1/2}\left[1 + \frac{(U_o - U_{mf})}{0.35(gD_c)^{1/2}}\right] + 0.061 - \frac{1.939(U_o - U_{mf})}{0.35(gD_c)^{1/2}} = 0 \qquad (40)$$

$$V_s = \frac{\pi D_c^3}{4}\left[\frac{L_s}{D_c} - 0.495\left(\frac{L_s}{D_c}\right)^{1/2} + 0.061\right] \quad \text{for } h > h_s \qquad (41)$$

The gas-exchange rate per slug, Q_{se}, based on the L_s obtained from Eq. 11, is now:[18]

$$Q_{se} = \frac{\pi D_c^2 U_s (1 - \delta)}{1.4(gD_c)^{1/2}}\left[U_{mf} + \frac{16\varepsilon_{mf} I}{1 + \varepsilon_{mf}}\left(\frac{D}{\pi}\right)^{1/2}\left(\frac{d}{D_c}\right)^{1/4}\right] \quad \text{for } h > h_s \qquad (42)$$

where I is a function of L_s/D_c, representing the surface integral along the slug surface. The values of I for various L_s/D_c have been tabulated by Hovmand and Davidson.[18]

The bubble volume and gas-exchange rate related to the bubbling stage below the point where slug formation starts, denoted as V_s and Q_{se}, respectfully, are:[30]

$$V_s = (\pi/6)d_b^3 \text{ for } h < h_s \qquad (43)$$

$$Q_{se} = (3/4)\pi d_b^2\, U_{mf} + 0.975\, \pi D^{1/2} g^{1/4} d_b^{\,7/4} \text{ for } h < h_s \qquad (44)$$

Since gas mixing is usually poor in fluidized beds, the gas in either the particulate phase or the slug or bubble phase is assumed to be in plug flow. Therefore, the oxygen and sulfur dioxide concentrations in the slug or bubble phase, C_{s,O_2} and C_{s,SO_2} respectively, due to the gas exchange with the particulate phase would vary along the bed height as:

$$V_s U_s \frac{dC_{s,O_2}}{dh} = Q_{se}(C_{e,O_2} - C_{s,O_2}) \tag{45}$$

$$V_s U_s \frac{dC_{s,SO_2}}{dh} = Q_{se}(C_{e,SO_2} - C_{s,SO_2}) \tag{46}$$

where C_{e,O_2} and C_{e,SO_2} are the oxygen and sulfur dioxide concentrations in the particulate phase.

The variations of the two gas concentrations in the particulate phase along the bed height result not only from the gas exchange but also from related reactions. The molar consumption rate of oxygen per unit volume of the expanded bed can be given as

$$-\int_0^{R_c} \frac{3(1-\delta)(1-\varepsilon_{mf})f_c}{4\pi R^3}\, C_{e,O_2}\mathcal{R}_c(R)P_b(R)\, dR - \frac{3(1-\delta)(1-\varepsilon_{mf})f_d}{4\pi R_d^3}\,\frac{1}{2}\, C_{e,SO_2}\int_0^X \mathcal{R}_d(X)P_b(X)\, dX \tag{47}$$

The first term is the combustion contribution, and the second term is the sulfation reaction contribution. The numerical factor 1/2 corresponds to the stoichiometric relation that one-half mole of oxygen and one mole of sulfur dioxide are required for conversion of one mole of the additive to the sulfate product. The gaseous reactant balance for the oxygen in the particulate phase is then of the following form:

$$U_{mf}\frac{dC_{e,O_2}}{dH} = \frac{\delta}{V_s} Q_{se}(C_{s,O_2} - C_{e,O_2}) - \int_0^{R_c} \frac{3(1-\delta)(1-\varepsilon_{mf})f_c}{4\pi R^3}\, C_{e,O_2}\mathcal{R}_c(R)P_b(R)\, dR - \frac{3(1-\delta)(1-\varepsilon_{mf})f_d}{4\pi R_d^3}\,\frac{1}{2}\, C_{e,SO_2}\int_0^X \mathcal{R}_d(X)P_b(X)\, dX \tag{48}$$

A similar formulation can be derived for C_{e,SO_2}, noting that the sulfur dioxide is generated from combustion and absorbed by the sorbent. Therefore,

$$U_{mf} \frac{dC_{e,SO_2}}{dh} = \frac{\delta}{V_s} Q_{se} (C_{s,O_2} - C_{e,O_2})$$

$$+ \frac{3(1 - \delta)(1 - \varepsilon_{mf}) f_c}{4\pi R^3} \left[\frac{\gamma_0/M_S}{(\gamma_S/M_S) + (\gamma_C/M_C) + (\gamma_H/2M_H) - (\gamma_0/M_0)} \right]$$

$$\times C_{e,O_2} R_c(R) P_b(R) dR - \frac{3(1 - \delta)(1 - \varepsilon_{mf}) f_d}{4\pi R_d^3} C_{e,SO_2} \int_0^X R_d(X) P_b(X)\, dX \qquad (49)$$

The boundary conditions of Eqs. 45-48, respectively, are as follows:

$$\text{at } h = 0,\ C_{s,O_2} = C_{e,O_2} = C_0 - \frac{q_c\ \gamma_v \frac{\gamma_C''}{M_C} + \frac{\gamma_S''}{M_S} + \frac{\gamma_H''}{2M_H} - \frac{\gamma_0'}{M_0}}{AU_0} \qquad (50)$$

and

$$C_{s,SO_2} = C_{e,SO_2} = \frac{q_c \gamma_v \gamma_S''}{M_S AU_o} \qquad (51)$$

If the combustion efficiency is defined as the conversion of non-ash material in the coal, it can then be calculated from the following expression:

$$\eta_1 = 1 - \frac{F f_c \rho_c' (1 - \gamma_a')}{q_c (1 - \gamma_a)} \qquad (52)$$

On the other hand, the sulfur retention, defined as the amount of sulfur dioxide absorbed by the sorbent as a percentage of that released during combustion, can be obtained from the following:

$$\eta_2 = \left[\frac{(q_d \gamma_d X)}{M_d} \Big/ \frac{(q_c \gamma_S - F f_c \rho_c' \ \gamma_S')}{M_S} \right] \qquad (53)$$

IV. GENERAL COMMENTS

The above formulation allows calculation of coal combustion efficiency and sulfur retention for a fluidized-bed combustor operating in a combined bubbling and slugging mode under realistic assumptions for gas-flow and solids mixing. The phenomenon of coal devolatilization, usually neglected in modeling efforts, is included in this work as is also the char-particle size distribution function and the sulfur loading distribution function of sorbent particles in the bed. Numerical calculations are being planned to determine the predictions from this model for such quantities as: (1) the combustion efficiency and sulfur retention as a function of coal particle size and Ca/S ratio, (2) the concentration profiles of oxygen and sulfur dioxide in the particulate and slug phases, and (3) the volumetric fractions of ash, carbon, and sorbent along the bed height. Comparison of the calculated combustion efficiency and sulfur retention with the experimentally measured values at the 15.2 cm dia ANL fluidized-bed combustor will also be made as a check of the appropriateness of the theoretical model described above and other models which are being developed at MIT.[31-33]

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