HYDRODYNAMIC BEHAVIOR OF A LIQUID-SOLID FLUIDIZED-BED REACTOR

FOR THE BIOCONVERSION OF COAL PARTICLES TO LIQUID PRODUCTS*

TIMOTHY C. SCOTT,† ERIC N. KAUFMAN, AND JOHN M. COSGROVE

BIOPROCESSING RESEARCH AND DEVELOPMENT CENTER

OAK RIDGE NATIONAL LABORATORY

P.O. BOX 2008

OAK RIDGE, TENNESSEE 37831-6226 (USA)

TELEPHONE: (615) 574-5962

FAX (615) 574-6442

MOHAMMAD ASIF AND JAMES N. PETERSEN

DEPARTMENT OF CHEMICAL ENGINEERING

WASHINGTON STATE UNIVERSITY

PULLMAN, WASHINGTON 99164 (USA)

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†To whom all correspondence should be addressed.

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ABSTRACT

A predictive mathematical model based on particle convection and dispersion is presented for a liquid fluidized bed of coal particles. The numerical representation can follow transient behavior of liquid fluidized beds that contain a defined particle-size distribution. The calculations exhibited excellent agreement when compared to experimental transient pressure-drop data from a column containing particles of Illinois No. 6 bituminous coal in the size range of 20 to 120 μm . In addition, the model was used to simulate the effect of periodic (hourly) liquid velocity step changes on elutriation of small particles from a given particle-size distribution of 34.5 to 75.5 μm . For the situation tested, the calculations indicate that (a) the column does not reach a steady state between velocity changes and (b) that higher initial particle elutriation rates decay to lower values until no more particles elutriate from the column.

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INTRODUCTION

The solubilization–liquefaction of coal using biocatalysts would enable process operations to be carried out at or near ambient temperature and pressure [1]. The use of either soluble or insoluble biocatalysts in either organic or aqueous media will require a reactor system that allows for efficient mass transport and enables continuous operation with high throughput. The proposed system is that of a continuous liquid–solid fluidized-bed reactor in which coal particles react and are removed from the column by hydrodynamic forces due to changes in size and/or density. In such a system, fresh coal particles of a given size distribution would be continually added to the column. As these particles solubilize, they would move toward the extremities of the bed due to either their increased or decreased terminal velocity.

For the situation in which a constant density is maintained, the smallest particles would eventually elutriate from the top of the bed. In order to achieve optimal reactor design and operation and enable process scaleup, mathematical models of particle fluidization and segregation must be developed. These models must provide predictions of axial pressure drop, dispersion of both the solid and liquid phases, and particle-size distribution as a function of axial position within the reactor. In addition, the model must be able to account for the inherently transient nature of the bed hydrodynamics. In this paper, a numerical model is presented that allows prediction of transient pressure measurements and particle elutriation in a liquid fluidized bed of small coal particles.

DESCRIPTION OF MODEL

The process of biosolubilization of coal particles leads to a situation where coal particles of widely variant sizes are present together in the fluidized bed. The physical forces acting on various particles are a function of the particle size. These force differences will cause the particles of a given size fraction to preferably occupy certain regions of the fluidized bed; specifically, smaller particles will tend to rise toward the upper regions of the bed while larger ones will tend to settle toward the bottom. This process gives rise to a phenomenon known as size segregation.

On the other hand, particle dispersion has a homogenizing effect that tends to uniformly mix all particles throughout the bed. Under the influence of these two competing mechanisms, the particles of a particular size fraction are generally present throughout the bed in varying concentrations. This nonuniform concentration distribution of different-sized particles in the bed leads to a variation in void volume along the bed height. Furthermore, because specific reactivity of particles varies greatly as a function of particle size, both the volume fraction and the particle-size distribution would have pronounced effects on volumetric production rates along the length of the bed.

A review of the literature indicates a certain lack of information in this area. There are a few studies that discuss the transients of fluidized beds; however, they are limited to the evaluation of bed height transients [2,3]. Additionally, most of the reports concerning multisized solid particles in a fluidized bed consider steady state situations and are primarily applicable to situations where only two types of particles are present in the bed.

As a starting point, the transient hydrodynamic behavior of nonreacting particles will be investigated, with that of reacting particles to be studied in future studies. The unsteady state mass balance is expressed as

$$\frac{\partial \rho_i}{\partial t} + \nabla \cdot N_i = 0 \quad , \quad (1)$$

where ρ_i is the mass fraction of solid particles of species i and N_i is the mass flux of this species.

Following Kennedy and Bretton [4], the particle mass flux is given as the sum of mass transport by dispersion and convection, or

$$N_i = -\rho D_i \nabla \omega_i + \rho_i U_i \quad , \quad (2)$$

where ρ is the bulk density, ω_i is the mass fraction of particle species i , D_i its dispersion coefficient, and U_i its velocity.

With these relationships, the general mass balance equation for a particle species i can be written as

$$\frac{\partial \rho_i}{\partial t} = \nabla \cdot \rho D_i \nabla \omega_i - \nabla \cdot \rho_i U_i . \quad (3)$$

Assuming that radial gradients of mass as well as velocity are generally absent in a liquid fluidized bed, equation (3) can be simplified to yield:

$$\frac{\partial \rho_i}{\partial t} = \frac{\partial}{\partial z} \left[D_i \rho \left(\frac{\partial \omega_i}{\partial z} \right) - U_i \rho_i \right] , \quad (4)$$

where z is the variable representing column length. Since fractional volumetric concentration is more often used, with the following relationship,

$$\rho_i = \rho \omega_i = \rho_{s_i} C_i , \quad (5)$$

equation (4) can be written as

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial z} \left[D_i \left(\frac{\partial C_i}{\partial z} - \frac{C_i}{\rho} \frac{\partial \rho}{\partial z} \right) - U_i C_i \right] , \quad (6)$$

where ρ_{s_i} is the density of solid particles of species i , and C_i is its fractional volumetric concentration.

Hence, equation (6) is the governing mass balance equation for the particles of species i present in the fluidized bed. Its solution requires the evaluation of the particle velocity, U_i , and the dispersion coefficient, D_i , for each type of particle species. In this model, the particle velocity, U_i , has been treated using a modified version of the Richardson and Zaki correlation [5]. Using this approach, the velocity of particle species i in the bed at the location where the bed voidage is ϵ is given by [6]

$$U_i = U_0 + \sum_{i=1}^{n_t} U_{t_i} \epsilon^{n_i - 1} C_i - U_{t_i} \epsilon^{n_i - 1} , \quad (7)$$

where U_0 is the superficial liquid velocity, n_t is the total number of different types of particles, U_{t_i} is the terminal velocity of particle species i , and n_i is the Richardson and Zaki correlation index for particle species i . Upon examination of fluidization behavior of coal size fractions representative of the present work, it has been shown that the correlation index, n , can be represented by the following form [7]:

$$n = 17.78 (d_p)^{0.2} . \quad (8)$$

In order to obtain values for the particle dispersion coefficient, D_p , the following correlation proposed by Asif and Petersen [7] has been used:

$$\frac{Fr}{Pe} = 7.9 \left(\frac{U_0 - U_{mf}}{U_t} \right)^{2.141} , \quad (9)$$

where U_{mf} is the minimum fluidization velocity for the type of particle in question, and the Peclet and Froude numbers are given by

$$Pe = \frac{U_0 d_p}{\epsilon D} \quad Fr = \frac{\rho_f}{\rho_s - \rho_f} \frac{U_0^2}{g d_p} ; \quad (10)$$

where d_p is the particle diameter, g is the gravitational constant, and ρ_f and ρ_p are the fluid and particle densities, respectively.

Substituting the expressions in equations (7), (8), and (9) into the transient mass balance given by equation (6) allows one to solve for the time-dependent particle concentration profiles in the fluidized

bed. Actual solution involves breaking up the particle-size distribution to be investigated into i segments and simultaneously solving i transient partial differential equations of the kind shown in equation (6). From the concentration profiles, one can calculate such aspects as bed height and pressure drop and follow the elutriation of particles from a given total volume of the column. The model will be compared to experimental transient pressure-drop data for verification. It will then be used to numerically examine transient elutriation of smaller particles from a fluidized bed undergoing operations under alternating periods of high and low liquid flow.

EXPERIMENTAL MATERIALS AND METHODS

In order to test the validity of the model, transient pressure-drop data were obtained over two sections of an experimental bed. A glass column of 25.4-mm internal diameter and 1.22-m length was used for carrying out fluidization experiments. More details on the apparatus are given elsewhere [8,9]. The pressure drop along the height of the test section was recorded using differential pressure transducers (Celso Model DP31) which were located 1.1, 21.5, and 31.7 cm above the liquid distributor. Voltage values from these transducers were recorded digitally every 10 s.

During the experiments, 100 g of Illinois No. 6 bituminous coal was used to charge the bed. The particle-size distribution was determined using a Microtrac Full Range Particle Analyzer (version 4.0) and is displayed in Fig. 1. From this plot, the distribution appears to be slightly skewed toward the larger sizes with a mean diameter of about 55 μm and an overall particle-size range of 20 to 120 μm .

During experimentation, the fluidized bed was allowed to reach steady state at a liquid superficial velocity of 0.0057 cm/s. Then, a step change in the liquid velocity was introduced by lowering the flow rate to 0.0037 cm/s. Once the bed again reached a steady state, the velocity was changed by raising it to 0.0109 cm/s. Using this approach, data were obtained for both an increase and decrease in flow rate.

RESULTS AND DISCUSSION

Figures 2a and 2b contain plots of the experimental transient pressure profiles and model

predictions for the bed range of 1.1 to 21.5 cm and 21.5 to 31.7 cm, respectively. These curves indicate that as the liquid velocity is lowered, the pressure drop between points in the fluidized bed increases, the result of a decrease in total bed height. Conversely, an increase in velocity expands the bed and the pressure drop between points decreases. The figure also reveals that the response of the bed to a step change in velocity is faster in the lower regions of the bed. In addition, it is clear that it takes considerably more than an hour for such a bed to achieve a new steady state.

From these graphs, it is evident that the overall agreement between the model predictions and experimental data is excellent. For these simulations, the particle-size distribution was broken up into 18 size regimes ($i=18$). The model does seem to predict a faster response to the step change of liquid velocity from 0.0037 cm/s to 0.0109 cm/s than was observed between the 1.1 and 21.5 cm heights; however, this trend is not indicated in the predictions for the behavior between the 21.5 and 31.7 cm points. From these results, it is evident that the model should serve as a useful tool for examining transient behavior in these types of fluidized beds.

The ultimate goal of the modeling effort is to predict elutriation of reacting coal particles under various flow conditions in the reactor. In order to begin to test this type of behavior, the model was used to predict elutriation of nonreacting particles under conditions where the liquid flow is alternately increased and decreased. Table 1 contains the parameters used in this simulation.

The results of the simulation are shown in Figs. 3 and 4. In Fig. 3, the time evolution of total-particle concentration (volume fraction) profile along the bed is shown. The effect of the periodic changes in the fluid velocity upon the particle concentration in the bed can be clearly seen. During the periodic operation, smaller particles elutriate from the bed. Figure 4 shows the mass of coal particles elutriated from the bed as the time progresses. It is evident from the plot that there is no elutriation of coal particles at the lower velocity (0.008 cm/s); however, when the velocity is increased to 0.03 cm/s, the coal particles begin to elutriate from the bed. It is also evident from Figs. 3 and 4 that 1 h is not enough time for the system to obtain a steady state even though a significant fraction of the smaller particles elutriate during the first hour of operation at the higher flow rate. As particle elutriation continues during

subsequent hours of high-velocity operation, the mass of coal particles elutriated slowly decreases until a time is reached when no more particles leave the bed.

CONCLUSIONS

A predictive mathematical model based on particle convection and dispersion has been developed for a liquid fluidized bed of coal particles. The numerical representation can follow transient behavior of liquid fluidized beds which contain a defined particle-size distribution. The calculations exhibited excellent agreement when compared to experimental transient pressure-drop data from a column containing particles of Illinois No. 6 bituminous coal in the size range of 20 to 120 μm . In addition, the model was used to simulate the effect of periodic liquid velocity step changes on elutriation of small particles from a given particle-size distribution of 34.5 to 75.5 μm . For the situation tested, the calculations indicate that the column does not reach a steady state between velocity changes and that higher initial particle elutriation rates decay to lower values until no more particles elutriate from the column.

The model appears to render a reliable approach to predicting the transient hydrodynamic behavior of liquid fluidized beds of small nonreacting coal particles. In addition to variances in size, the numerical system can accommodate circumstances in which particle-density distributions arise. The next stage in development of this predictive technique will be to include the additional complication of particles that change size and/or density due to reactions with microbes or dissolved catalysts such as enzymes. This will most likely entail the coupling of a population-balance-type model with the transient hydrodynamics in order to be able to predict conditions which will allow both steady addition of fresh coal into and steady elutriation of spent particles out of the column.

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TABLE 1

Parameters used in the particle elutriation simulation

Parameter	Value
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Particle-size range (μm)	34.5 - 75.5 μm
Particle density (g/cm^3)	1.3
Fluid density (g/cm^3)	1.0
Fluid viscosity (cP)	1.0
Column height (cm)	100
High liquid velocity (cm/s)	0.03
Low liquid velocity (cm/s)	0.008
Period of step change (h)	1

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FIGURE CAPTIONS

Fig. 1. Size distribution of coal particles used in transient pressure-drop experiments.

Figs. 2a and 2b. Pressure transients due to step changes in liquid velocity. The liquid velocity was first lowered from 0.0057 cm/s to 0.0037 cm/s at 14,500 s then raised to 0.0109 cm/s at 28,900 s.

Fig. 3. Response of the fluidized bed to a periodic step change in liquid velocity (0.008 cm/s and 0.03 cm/s; period of step change = 1 h).

Fig. 4. Elutriation of coal particles from the fluidized bed in response to a periodic step change in liquid velocity (0.008 cm/s and 0.03 cm/s; period of step change = 1 h).

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