

Hydrodynamic Studies of an Advanced Fluidized-Bed Bioreactor for Direct Coal Liquefaction†

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ABSTRACT

Advanced bioreactors can play an important role in coal utilization and conversion. To obtain the high throughput and low inventory required for large-scale bioprocessing of coal, a new family of bioreactors is being developed. The reactors on which this study is focused are continuous columnar bioreactors, which can be operated as three-phase systems. In particular, this study is oriented toward fluidized-bed systems for direct biological interactions with fine (30–150 μm) coal particles. Illinois no. 6 bituminous coal was ball-milled and then suspended in a 0.1% aqueous solution of the surfactant (Tween 80 (polyoxyethylene), which was used to reduce particle aggregation. Experimental fluidized-bed studies were then performed, using size-screened coal fractions. Studies were performed to quantify the extent of particle segregation and to quantify particle mixing effects within the reactor. To do so, pressure drop vs time data has been recorded as a function of axial position along the bed height. To better understand this experimental data, predictive mathematical

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models based on fundamental principles of mass and momentum transfer have been developed. Comparisons of model predictions with experimental data are presented.

Index Entries: Fluidized bed; bioprocessing; coal; liquefaction.

INTRODUCTION

The capability of selected biochemical transformation reactions to convert coal into liquid fuels has generated considerable interest in the literature (1–4). The development of an efficient reaction system suitable for carrying out such biotransformations is key to the successful commercialization of any coal bioprocessing technology. Continuous-flow fluidized-bed bioreactors show great promise in this respect owing to their unique inherent capability of enhancing the global reaction rate by reducing the mass-transport resistances.

The system of ultimate interest will be comprised of fine coal particles that are fluidized by the upflow of liquid media that contains the dissolved biocatalyst. Depending on the operation to be carried out (e.g., solubilization, two-step gasification, beneficiation, and so on), the biocatalyst will be dissolved (or suspended) in either an aqueous-based or organic-based liquid (5). The progressive biotransformation of coal particles in the fluidized-bed bioreactor leads to a situation where the solid phase of the bed consists of coal particles of varying sizes. The effective utilization of such a fluidized-bed reaction system for the bioprocessing of the coal is crucially dependent on the proper characterization of its hydrodynamics.

For a two-phase liquid-solid fluidized-bed bioreactor containing small coal particles, the knowledge of its expansion characteristics and of the concentration distribution of solid particles along the bed height is vital for proper design and scale-up. A significant amount of information is available in the literature about various aspects of the hydrodynamics of liquid fluidized beds containing monosized particles (6). However, the state of current knowledge about the hydrodynamics of fluidized beds containing particles of different sizes is far from satisfactory. As a first step in this direction, the present study concentrates on a binary-solid fluidized bed. The term binary-solid, in the present context, implies solid particles of two different sizes. Such binary-solid systems have been known to reveal interesting hydrodynamic characteristics (7).

It is a commonly known fact that the size of a particle has a great bearing on its fluidization characteristics. A binary-solid mixture, when fluidized with a liquid, tends to manifest a behavior that is different from the one exhibited by either of the solid species if present alone in the fluidized bed. Existing correlations for monosized solid particles will not, therefore, predict the behavior of binary-solid systems. As such, other methods must be developed to predict the behavior of these systems.

The binary-solid fluidization is usually marked by the presence of three different layers in the bed, with the bottom layer mainly consisting of large particles, the top one consisting of small particles, and a transition zone in between the two layers where both components are present in varying concentrations (7). Recently, Vos et al. (8) pointed out that the segregation of solid particles into different layers affects the efficient use of the biocatalyst and could lead to other operational problems in a fluidized-bed bioreactor.

MATERIALS AND METHODS

The fluidized bed used in the experimental portion of this work was contained within a glass column having an internal diameter of 25.4 mm. To ensure a uniform liquid distribution across the cross-section of the column, a distributor made from a coarse glass frit was placed at the column inlet. This distributor was preceded by a calming section that helped to dampen the flow fluctuations induced by the peristaltic pump. The test section of the fluidized bed was 700 mm in length and was followed by a disengagement section having a diameter of 76.2 mm. A schematic of the bed is shown in Fig. 1.

The pressure drop along the height of the test section was recorded using six high-sensitivity differential pressure transducers (Celeso model DP31). The pressure transducers were positioned such that the total differential pressure relative to the bottom of the bed could be determined every 101.6 mm (4 in) in the axial dimension. A coarse glass frit was placed at the junction of the column and the pressure taps to avoid clogging of the pressure taps by the coal particles and to minimize flow disruption, which could be caused by the pressure taps. The signals from these transducers were recorded digitally every 5 s and plotted such that, after each change in the column feed rate, steady state was ensured before the reported pressures were recorded.

In the present experimental runs, Illinois no. 6 bituminous coal was ball-milled and then suspended in a 0.1% aqueous solution of the surfactant Tween 80 (polyoxyethylene). This surfactant tends to reduce particle aggregation. It was particularly important that the number of very small particles adhering to larger particles be minimized. Size-screened fractions were then used in the fluidized-bed experiments. In the present study, two different coal samples having average diameters of 49 and 63 μm were used. The distribution of particles sizes within the screened fractions was measured using a Horiba CAPA-700 (Horiba Instruments Inc., Irvine, CA) Particle Size Distribution Analyzer, which is designed to measure the particle size distribution of particulate samples using a non-contact measuring method based on liquid-phase sedimentation (9).

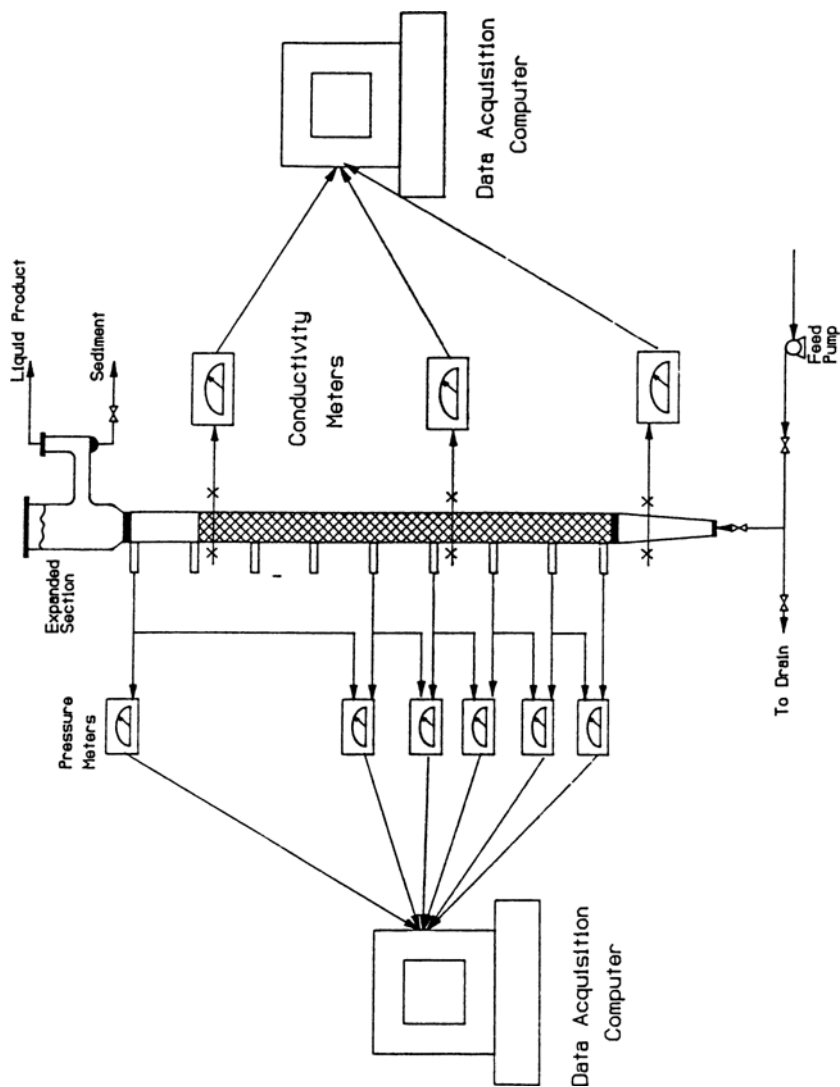


Fig. 1. Schematic of the experimental apparatus.

In this study, the bed was loaded with 36.5 g of the fraction having an average diameter of 49 μm and 46.4 g of the 63 μm fraction. The bed was fluidized at a flow rate sufficient to cover the total height of the test section, i.e., a superficial velocity of 0.0156 cm/s. Once steady state had been established, pressure measurements were recorded. Then, the flow rate was reduced, steady state reestablished, and the pressures again recorded. This procedure was followed for a total of five different flow rates.

The low particle-to-column diameter ratio (approx 0.002) in the present experimental setup rendered the wall effects to be insignificant. No wall effects were observed for a much higher particle-to-column diameter ratios (0.008–0.035) by Van der Meer et al. (10) in their study.

To understand the data obtained in the above experimental program better, a mathematical description of the problem was developed and is presented in the next section of this article.

MODEL DEVELOPMENT

In this portion of this article, necessary background information will be first provided so that our approach to the development of a mathematical model that describes the fluidized bed can be better understood. Then, the details of our approach to the mathematical description of this system will be described.

Background

This study will first examine the existing models for representing the mixing and segregation characteristics of the binary-solid fluidized beds and briefly discuss their important features. Then, in the next section, modifications to these descriptions that improve the description of the fluidized-bed system will be proposed and justified.

Kennedy and Bretton (11) proposed that the solid mixing in binary-solid fluidization is caused by the combined effects of diffusive and convective mass-transport mechanisms, and can be represented as:

$$D_i (dC_i / dz) - U_{si}C_i = 0 \quad (1)$$

where C_i is the concentration of component i , D_i is the axial dispersion coefficient, and U_{si} is the segregation velocity of the i^{th} particle species. The contribution to the diffusive flux comes from the random motion of fluidized coal particles. On the other hand, it is the segregation velocity that gives rise to the convective flux. The segregation velocity is the mean rate at which particles tend to segregate based on differences in properties. More precisely, the segregation velocity of species i is the difference between the interstitial velocity of the liquid and the interstitial velocity

required to produce a bed of the same voidage when that species is fluidized on its own. The required interstitial velocity can be evaluated with the help of available correlations. Kennedy and Bretton used the well-established correlation of Richardson and Zaki (12) for this purpose. Other correlations could also be used for this purpose, such as that reported recently by Shippey and Watson (13).

This approach was followed by several other investigators with minor or no variations. For example, Juma and Richardson (7), based on the results of their experimental investigations, proposed a linear z -dependence of U_{si} whereas a quadratic dependence was recommended by Dutta et al. (14) to make the model valid for solid particles that differ in their sizes as well as densities. The model of Dutta et al., however, required the concentration profiles to intersect at a point in the transition zone for the underlying mathematical analysis to be valid. It is worthwhile to note here that contrary to the model of Kennedy and Bretton, these models (7,10,14) do not employ the empirical correlation of Richardson-Zaki, but use an assumed profile instead. When an assumed profile is used, a few additional parameters are required to describe the z -dependence of the segregation velocity, but the data can be represented quite well.

Fan et al. (15) essentially extended the approach of Juma and Richardson to model the solid concentration profile in three-phase gas-liquid solid fluidized beds of binary-solids. The comparison of model predictions with the experimental results were found to be satisfactory. Using a correlation, based on force balance considerations, for the prediction of the pressure drop in a liquid fluidized bed, Felice et al. (16) were able to represent the mixing and segregation phenomena in a binary-solid fluidized bed. However, these authors assumed a linear profile of the total particle concentration without any apparent physical justification. Also, they followed the approach of Van der Meer et al. (10) for evaluating the segregation flux, which is based on the assumption that the particle segregation velocities are a linear function of their concentrations.

It is evident from the foregoing discussion that most of the studies used the basic model of Kennedy and Bretton, but relied on either introducing additional parameters or using correlations to predict solid particle concentration profile in the bed. In addition, several investigators forced the distribution to follow prescribed forms with no apparent fundamental reason for using the forms given.

Increasing the number of parameters in the model does help to improve the prediction that is, however, obtained only at the expense of more tedious mathematical calculations. On the other hand, the use of available correlations introduces limitations on the application of the model that are inherent with the correlation itself.

It is worthwhile to point out at this stage that these empirical correlations contain a vast amount of information of the fluid mechanics of the present multiparticle fluid-solid system of fluidized beds and have been

found to represent its important hydrodynamic characteristics extremely well. Therefore, the evaluation of the segregation velocity based on a well-established correlation will reflect more closely the actual hydrodynamics of the system. Using the Richardson-Zaki (12) correlation, the segregation velocity of species i , in terms of measurable variables, is given by:

$$U_{si} = [(u_o / \epsilon) - u_{ti} \epsilon^{n_i - 1}] \quad (2)$$

where, u_o is the liquid superficial velocity, u_{ti} is the terminal velocity of i , ϵ is the bed voidage or the volumetric fluid concentration, and n_i is the constant in Richardson-Zaki correlation (12). The objective of the present study is to develop a mathematical model capable of realistically describing the concentration profiles of coal particles of two different sizes in a fluidized bed. The proposed model has only two parameters and uses the well-established correlation of Richardson-Zaki for the evaluation of particle segregation velocities.

Proposed Model

In this section, the steps involved in the derivation of the final modeling equation will be briefly outlined. The total mass flux, N_i , of particle species " i " as a result of dispersion and segregation mass-transport mechanisms is given by:

$$N_i = - \rho D_i [(d\omega_i / dz) + \rho U_s \omega_i] \quad (3)$$

where, ρ is the bulk density, ω_i is the mass fraction of species i , D_i is the dispersion coefficient of i , and U_s is the overall segregation velocity. Unlike previous models where the segregation velocity of component i was assumed to be responsible for the particle convective flux, the segregation velocity in the present model is the sum of the individual segregation velocities of both particle species, given by:

$$U_s = \omega_a U_{sa} + \omega_b U_{sb} \quad (4)$$

Here, subscripts a and b represent the two particle species present in the bed, and U_{sa} and U_{sb} are the segregation velocities of particle species a and b , respectively.

In order to calculate the value of the individual segregation velocities, one of the relative velocity correlations available in the literature could be employed. In terms of the Richardson-Zaki correlation:

$$U_{ri} = U_f - U_{si} = U_{ti} \epsilon^{n_i - 1} \quad (5)$$

where U_{ri} is the particle relative velocity, U_{ti} is the particle terminal velocity, U_f is the fluid velocity, ϵ is the bed voidage fraction, and n_i is the Richardson-Zaki index.

The interstitial fluid velocity, U_f , in the above equation can be evaluated from overall mass-balance considerations. At steady state:

$$(d/dz)(\rho_{sa}\epsilon_{sa}U_{sa} + \rho_{sb}\epsilon_{sb}U_{sb} + \rho_f\epsilon U_f) = 0 \quad (6)$$

where, ρ_{si} is the solid density of component i , ϵ_{si} is the volumetric concentration of i , and ϵ is the fluid volumetric concentration. At steady state:

$$(\rho_{sa}\epsilon_{sa}U_{sa} + \rho_{sb}\epsilon_{sb}U_{sb} + \rho_f\epsilon U_f) = \rho_f U_o \quad (7)$$

In terms of relative velocities, the above expression can be written as

$$U_f = (\rho_f / \rho) U_o + \omega_a U_{ra} + \omega_b U_{rb} \quad (8)$$

Here, the bulk density, ρ , is defined as:

$$\rho = \rho_{sa}\epsilon_{sa} + \rho_{sb}\epsilon_{sb} + \rho_f\epsilon \quad (9)$$

Therefore, the segregation velocity of particle species i can now be written as:

$$U_{si} = U_f - U_{ti}\epsilon_i^{n_i-1} \quad (10)$$

At steady state:

$$(dN_i/dz) = 0 \quad (11)$$

Substituting the expression for N_i in the above equation from Eq. (3) gives:

$$(d/dz)[\rho D_i(d\omega_i/dz) - \rho U_s \omega_i] = 0 \quad (12)$$

The above equation governing the concentration distribution of individual particle species in the fluidized bed comes in the form of second-order linear differential equations that are coupled via the presence of bulk flow term. Its solution will require specification of proper boundary conditions at both ends of the solution domain, which should preferably be determined by the physics of the problem. In the present case, coal particles of different sizes are present in the bed. The larger coal particles will predominantly occupy the lower region of the bed, whereas the smaller ones will be present in the upper region for the bed. Between these two regions of the relatively pure components, a gradual variation of the concentration of the two coal species is observed. This intermediate zone is the result of particle dispersion. In the ideal case of no dispersion, it would be logical that two zones, each having essentially pure large and small particles, would develop within the bed. However, in the actual case, because of particle dispersion effects, a region exists where the two particles are interspersed. This behavior is also evident from the void fraction profile of the fluidized bed, which shows a gradual increase along the length of the intermediate zone. This region, usually called the mixing region, is of interest for the present modeling. If the height of this region is " L ," then the following set of boundary conditions, as pointed out by Juma and Richardson (7), will appropriately describe the present system: For large particles a , at $z=0$, $\omega_a = \omega_{a0}$, and at $z=L$, $\omega_a = 0$. For small particles b , at $z=0$, $\omega_b = 0$, and at $z=L$, $\omega_b = \omega_{b0}$.

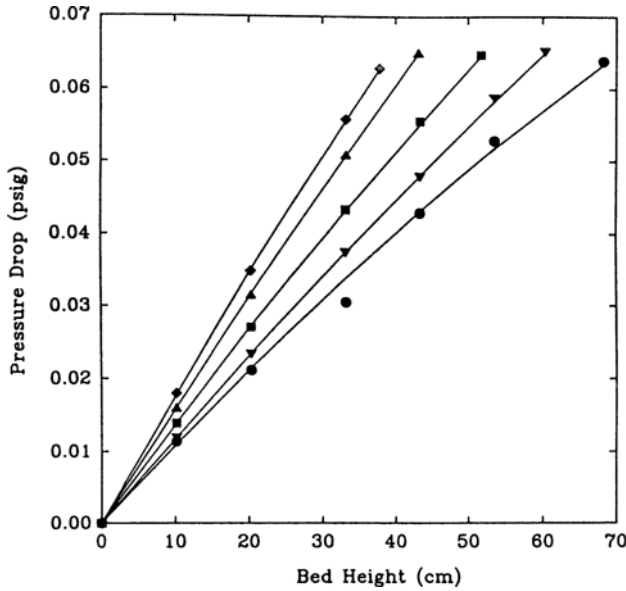


Fig. 2. Influence of liquid superficial velocity on the pressure drop along the bed height (solid lines represent the model predictions). Liquid velocity: ● 0.0156 cm/s, ▼ 0.0122 cm/s, ■ 0.0080 cm/s, ▲ 0.0045 cm/s, ◆ 0.0028 cm/s.

DATA ANALYSIS

As shown in Fig. 2, the pressure drop along the bed height in a liquid fluidized bed containing 44 and 63- μm coal particles was recorded for five different flow rates. The pressure drop vs bed height data were converted into the total particle concentration vs bed height data using the following formula:

$$(1 - \epsilon) = - (dp / dh) \{ [1 / (\rho_s - \rho_f)g] \} \quad (13)$$

where ϵ is the bed void fraction, ρ_s is the density of the solid particles, ρ_f is the density of the fluidizing liquid, g is the acceleration owing to gravity, and dp/dh is the pressure drop along the bed height. Based on these calculations, the total particle concentration was calculated as a function of axial position and is shown in Fig. 3.

The average diameter of small coal particles was 49 μm and that of large particles was 63 μm . A sphericity factor of 0.7 was introduced in the calculations of the equivalent diameter of the particles to account for the departure of the coal particles from the actual spherical shape. In the present case, the value of Richardson-Zaki correlation index, n , in Eq. (2) was found to be 4.65. Relevant variables of interest are as shown in Table 1.

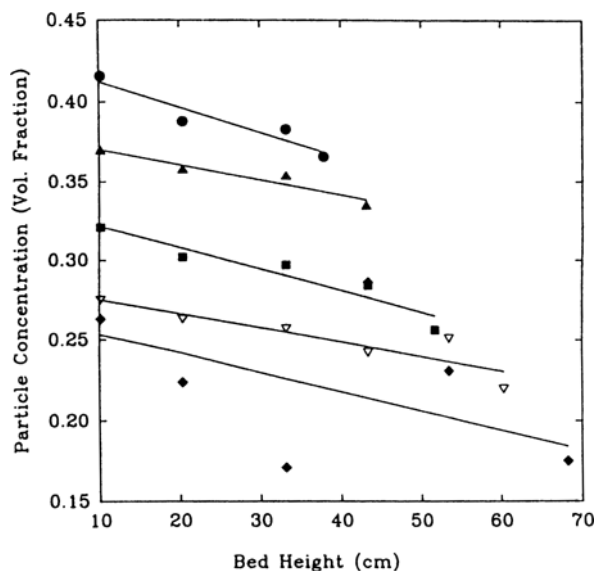


Fig. 3. Influence of liquid velocity on the total particle concentration profile in the bed (solid lines represent the model predictions). Liquid velocity: ● 0.0028 cm/s, ▲ 0.0045 cm/s, ■ 0.0080 cm/s, ▽ 0.0122 cm/s, ◆ 0.0156 cm/s.

Table 1
Experimental Parameters
Also Used in the Mathematical Model

Parameter	Value
Diameter of particle <i>a</i> , d_{pa}	0.0063 cm
Diameter of particle <i>b</i> , d_{pb}	0.0049 cm
Sphericity, ϕ	0.7
Particle density, ρ	1.3 g/cm ³
Fluid density, ρ_f	1.0 g/cm ³
Fluid viscosity, μ	0.87 cP

The proposed model contains two adjustable parameters, D_a and D_b , the dispersion coefficients of the two particle species present in the bed. The best fit with the experimental data was obtained using an generalized reduced-gradient optimization code (17,18). The first and the last data points were treated as the boundary conditions in the solution of the differential equation.

RESULTS AND DISCUSSION

The total particle concentration profiles experimentally obtained are compared with the prediction of the model in Fig. 3. The comparison is

satisfactory for most liquid superficial velocities. However, significant scattering of the experimental data is observed at the intermediate bed heights for a liquid velocity of 0.0156 cm/s. Since the primary set of the experimental data in the present study is the pressure drop along the height of the bed, it is worthwhile to compare the model pressure drop predictions with the experimental data. The model pressure drop predictions were computed from the model predictions of the concentration profile. The comparison of the predicted and computed pressure drops in the bed is shown in Fig. 2. In this case, the comparison is excellent except for the velocity of 0.0156 cm/s and the pressure transducer located at the bed height of about 35 cm. It is interesting to notice how a slight scattering of the pressure drop data at a point could have a pronounced effect on the experimental concentration profile in the bed. It is tempting at this stage to smooth the pressure drop data by a curve-fitting method. This could, however, lead to erroneous conclusions. For example, if the pressure drop data are fitted with a second-order function of the bed height, this will lead to a linear total particle concentration dependence with the bed height, which may not always be true.

The absence of any significant curvature in the model prediction in Fig. 3 indicates that the primary mechanism of the mass transport in the bed is the dispersion of the coal particles. This could be attributed to the fact that the two particle samples used here are of close size fractions. The large particles, when carried to the upper regions of the bed as a result of dispersion, show a downward or negative segregation velocity. On the other hand, the small particles present in the lower bed regions tend to show an upward or positive segregation velocity. In the present case, this renders an overall segregation flux that is substantially smaller than the dispersion flux.

In Fig. 4, the gradient of the total particle concentration profile in the bed (i.e., the slope of the lines in Fig. 3) is plotted as a function of the liquid superficial velocity. This gradient exists because of the mixing and segregation tendencies of the fluidization of the different sized particles. Although it was expected that this gradient would decrease as the superficial velocity increased owing to the fact that the dispersion coefficient should increase with velocity, this was not clearly demonstrated by these results. Apparently, the liquid superficial velocity has little influence on the gradient of the particle concentration in the present case of two closely sized fractions of coal particles.

CONCLUSIONS

An experimental system has been developed that will allow the pressure drop as a function of axial position to be determined for a system containing finely divided coal particles. Further, modifications to established mathematical descriptions of fluidized-bed contractors have been

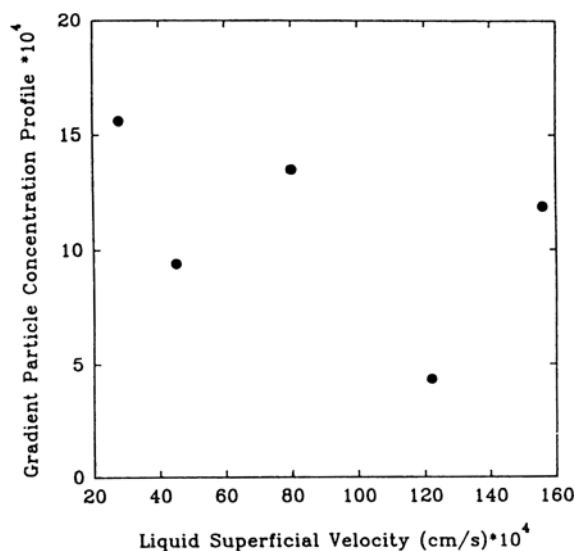


Fig. 4. Effect of liquid velocity on the gradient of the total particle concentration in the bed.

developed to describe these data. These mathematical models are based on fundamental principles of mass and momentum transfer. This mathematical model has been shown to provide an excellent description of the experimental pressure drop vs position data. For this system composed of finely divided coal particles, dispersion has been found to be the dominant driving force in the system.

ACKNOWLEDGMENTS

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