

# **A Review of Gas-Solid Dispersion and Mass Transfer Coefficient Correlations in Circulating Fluidized Beds**

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## **Abstract**

It is increasingly more important to be able to predict the conversion and yield of reactant species given the ever rising cost of the reactants and the ever decreasing acceptable level of effluent contaminants. As such, the development and use of predictive models for the reactors is necessary for most processes today. These models all take into account in some manner the relative movement of the gas and of the solid species as well as the interphase mass transfer. The model developer, unless equipped with specific experimentally based empirical correlations for the reactor system under consideration, is required to go to the open literature to obtain correlations for the axial and radial gas dispersion coefficients, the solids axial and radial dispersion coefficients and the interphase mass transfer coefficient between the solid phase and the gas phase. This is a difficult task at present since these values differ by up to 7 orders of magnitude for the mass transfer coefficient and up to 5 orders of magnitude for dispersion coefficients. This paper presents a summary of the work in these areas as found in the literature dating back to 1949 and provides a critical review to guide the selection of the best correlation(s) in circulating fluidized bed reactors.

## **Background**

A major goal of the system designers and modelers is to be able to simulate the complex behavior in gas-solid chemically reacting flows in circulating fluidized bed systems. To support the mathematical simulation codes, research is being conducted on the hydrodynamics in these advanced gas-solids contactors, namely, circulating fluidized beds and transport reactors under atmospheric and pressurized conditions using various solids. The experimental data on the hydrodynamics of these flows provides crucial information to the modelers, providing them with experimental data on the hydrodynamic behavior of the gas and solids to which they can tune the models. To date, only one part of the simulation is being tuned with the experimental data, that being the overall flow field. The simulation of combustors, gasifiers and the likes requires information on both gas and solids species movements and the interphase transport.

The development of Eulerian-Eulerian simulation codes such as MFIX to predict the performance of circulating fluidized beds and transport reactors to applications ranging from combustion and gasification to pollution control and chemicals production requires the existence of good theoretically based models to predict the interphase mass transport between reacting components and the dispersion of the reactants throughout the flow field within the reactors. MFIX uses the following equations for the gas and solids species continuity.

$$\frac{\partial}{\partial t}(\epsilon_g \rho_g x_{g,n}) + \nabla \cdot (\epsilon_g \rho_g x_{g,n} u_g) = \nabla \cdot \mathcal{D}_{g,n} \nabla x_{g,n} + R_{g,n} \quad \text{Equation 1}$$

$$\frac{\partial}{\partial t}(\epsilon_{s,m} \rho_{s,m} x_{s,m,n}) + \nabla \cdot (\epsilon_{s,m} \rho_{s,m} x_{s,m,n} u_{s,m}) = \nabla \cdot \mathcal{D}_{s,m,n} \nabla x_{s,m,n} + R_{s,m,n} \quad \text{Equation 2}$$

To use the above equations, knowledge of the dispersion coefficients  $\mathcal{D}_{g,n}$  and  $\mathcal{D}_{s,m,n}$  is needed as well as the relationship for the interphase mass transfer coefficient,  $k_d$ , which is buried in the reaction terms,  $R_{g,n}$  and  $R_{s,m,n}$ . The terminology in Equations 1 and 2 is taken directly from the MFIX document (Syamlal). Simulation codes such as MFIX allow for the dispersion coefficients to differ both directionally, spatially and by component. Equations 1 and 2 can be simplified for cylindrical coordinates by neglecting azimuthal contributions. The results of this simplification are presented in Equations 3 and 4.

$$U_{g,i} \frac{\partial C_i}{\partial x} = \mathcal{D}_{ga} \frac{\partial^2 C_i}{\partial x^2} + \mathcal{D}_{gr} \frac{1}{r} \left( \frac{\partial}{\partial r} \left( r \frac{\partial C_i}{\partial r} \right) \right) + R \quad \text{Equation 3}$$

$$U_{s,i} \frac{\partial N_i}{\partial x} = \mathcal{D}_{sa} \frac{\partial^2 N_i}{\partial x^2} + \mathcal{D}_{sr} \frac{1}{r} \left( \frac{\partial}{\partial r} \left( r \frac{\partial N_i}{\partial r} \right) \right) - R \quad \text{Equation 4}$$

Equation 3 is the gas species continuity equation in terms of concentration,  $C$ , and allows for reaction. Equation 4 is the solids species continuity equation in terms of number of particles,  $N$ , and allows for reaction. The values of  $\mathcal{D}_{ga}$ ,  $\mathcal{D}_{gr}$ ,  $\mathcal{D}_{sa}$  and  $\mathcal{D}_{sr}$  are obtained from experimental tests, and correlated to the physical dimensions and operational parameters of the system.

The interphase mass transfer coefficient appears in the relationship for the reaction,  $R$ , and is given in Equation 5 for a shrinking particle first order reaction with respect to the gas between gas species,  $j$ , and solid species,  $i$ ,

$$R = \frac{N_i \frac{\pi d_p}{4} C_j}{\left( \frac{1}{k_c} + \frac{1}{k_d} \right)}$$

Equation 5

where  $k_c$  is the chemical reaction rate constant and  $k_d$  is the interphase mass transfer coefficient. In order to predict the performance conversion for a particular reaction in a CFB, values for the dispersion coefficients and the interphase mass transfer coefficient need to be obtained from the literature.

### Literature Review

An extensive literature review has been conducted to obtain the functional relationship of these parameters ( $D_{ga}$ ,  $D_{gr}$ ,  $D_{sa}$ ,  $D_{sr}$  and  $k_d$ ). The review of the-state-of-the-art is divided into a dispersion section and a mass transfer section. Each of these is further divided into a comparative discussion of the experimental facilities and a comparison of the correlations presented.

### Dispersion

A number of the investigators report the dispersion coefficients as a Péclet Number. This number is the product of the Reynolds Number to a modified Schmidt Number based on the dispersion coefficient rather than the diffusion coefficient as shown in Equation 6,

$$Pe = Re * Sc = \frac{\rho U \ell}{\mu} * \frac{\mu}{\rho \mathcal{D}} = \frac{U \ell}{\mathcal{D}}$$

Equation 6

where  $\ell$  represents a characteristic distance, reactor length for axial dispersion and radius or diameter for radial dispersion. Given the variability of the definitions, it is important to use the values defined by the researcher rather than a generic definition. For example, Werther bases his correlation on a definition using a diameter and Rhodes bases his correlation on a definition using radius

### Experimental Facilities Used in Dispersion Investigations

The diversity of the experimental systems, both physical configuration and particle properties is quite broad. The units are hot and cold, rectangular and cylindrical, use Geldart group A, B and D solids (Geldart), have diameter or lateral widths from 0.082 to 1.7m, heights from 2.5 to 13.5 m, particle sizes from 15 to 1600  $\mu\text{m}$ , solids density from 780 to 10000  $\text{kg/m}^3$  – primarily 2450 to 2600  $\text{kg/m}^3$ , solids flux values up to 120  $\text{kg/m}^2\text{s}$  and gas velocities from 0.2 to 8 m/s. The specifics of each facility are summarized in Table 1. Noting the extreme differences in these investigations sheds some light on the range of the dispersion coefficients alluded to in the abstract.

**Table 1 Conditions and Dimensions of Dispersion Investigation Facilities**

Author \ Facility	d (m)	h (m)	T (K)	P (kPa)	dp (um)	ps (kg/m <sup>3</sup> )	Gs (kg/m <sup>2</sup> s)	Ug (m/s)
Werther	0.3 x 1 (rect)	8.5	ambient	ambient	150	2500	20	3
Wei	0.14	10.4	ambient	ambient	15 to 1600	780 to 10000	19	1.5 to 8.5
Wei (hot unit)	0.203	2.5	298 to 710	1 to 3.1 bar	76	1960	0	0.2 to 1.5
Chaoki	0.082	7	ambient	ambient	150, 500 tracer	2600	23 to 75	4
Leckner	.12 x .7	8.5	ambient	ambient	320	2600	1 to 40	1.2 to 4.3
Leckner hot	1.4 x 1.7	13.5	850	ambient	320	2600	0	1.2 to 4.3
Werther	0.4	9	ambient	ambient	130	2600	0 to 70	3 to 6
Adanez	0.1	3.9	ambient	ambient	380 & 710	2600	0 to 115	5 to 8
Rhodes	0.305	6.6	ambient	ambient	71	2450	20 to 120	2.6 to 5

## Dispersion Coefficient Correlations

### Gas Dispersion Coefficients

The results from investigations of the axial and lateral/radial gas dispersion coefficients,  $D_{ga}$  and  $D_{gr}$  respectively, in circulating fluidized bed systems are summarized in Table 2 for eight investigations.

The correlations are presented graphically in Figure 1 as  $D_{ga}$  and  $D_{gr}$  versus gas velocity. Values of the dispersion coefficient range from  $0.0001 \text{ m}^2/\text{s}$  to  $10 \text{ m}^2/\text{s}$  with axial dispersion values being greater than radial. Wei found the axial dispersion to be approximately an order of magnitude greater than the radial dispersion at the same conditions. Leckner presents three different correlations for the gas radial dispersion coefficient. One correlation is for a hot unit and the other two for a cold unit. Considering the two correlations for the cold unit, one agrees regionally with the correlation by Wei for the radial dispersion coefficient and the other agrees with Wei's correlation for the axial dispersion coefficient. Closer inspection of the two Leckner correlations reveals that a discrepancy in the correlations. Both are applicable for a gas flow of  $1 \text{ m/s}$  and predict radial gas dispersion coefficients that differ by an order of magnitude. For this case, the Leckner correlations predict that the gas dispersion coefficient could be as low as  $0.0005 \text{ m}^2/\text{s}$  or as high as  $0.005 \text{ m}^2/\text{s}$ . The data from the hot unit show the radial dispersion decreasing with increasing velocity and being approximately an order of magnitude greater than the cold data, being greater than  $0.05 \text{ m}^2/\text{s}$ .

Use of these correlations is cautioned as they are unverified by other investigators. The solids radial dispersion coefficient is reported by all three investigators. The values

**Table 2 Gas Dispersion Investigations**

<b>Author</b>	<b>Model</b>	<b>Constraints</b>
Werther (2003)	$D_{gr} = \frac{d_{hyd} U_o}{Pe} \rightarrow Pe_r \cong 150$	None Identified
Wei (2001a)	$D_{ga} = \frac{LU_o}{Pe} \rightarrow Pe_r \cong 10$	None Identified
	$D_{gr} = \frac{LU_o}{Pe} \rightarrow Pe_a \cong 100$	
Werther (1992a)	$D_{gr} = \frac{2r^* U_c}{Pe} = \frac{2r^* U_c}{465} = 0.00366 r U_c$	Core in Core-Anular Flow
Adanez	$D_{gr}$ between 0.0002 & 0.0007 depending on gas vel & solids flux.	None Identified
	$D_{gr}$ decreases with gas vel & increases with solids flux	
Rhodes (1993)	$D_{gr} = \frac{r U_c}{Pe} = \frac{r U_c}{3.23 Re_m^{0.297}}$ $Re_m = \frac{(G_s + \rho_g U_g) 2r}{\mu}$	$40 < Re_m < 300$ & $150 < Pe < 300$
Grace	$D_{ga} = 7.1 * 9.3 * f(dP)$	None Identified
	$1 < D_{ga} < 11.88$ $Pe = f(dP)$	
Adams	$.004 < D_{ga} < .02$	None Identified
	$D_{ga}$ decrease with gas velocity and increase with solids flow	
Leckner (2000 & 2002)	$D_{gr}$ data table, trends down with increasing velocity	Hot Cold None Identified beyond experimental range
	$D_{gr}$ data table, trends up with increasing velocity	
	$D_{gr}$ data table, has a minimum	

reported differ by a factor of one hundred, Chaouki reporting a constant value of  $1 \text{ m}^2/\text{s}$  compared to the values reported by Wei ranging from 0.005 to 0.7, depending on particle Reynolds number and voidage. At this time, the constant value reported by Werther, of  $D_{s,r}$  equal to  $0.1 \text{ m}^2/\text{s}$  is in the middle of the range and is a better assumption than the value of zero currently being used in MFIX.

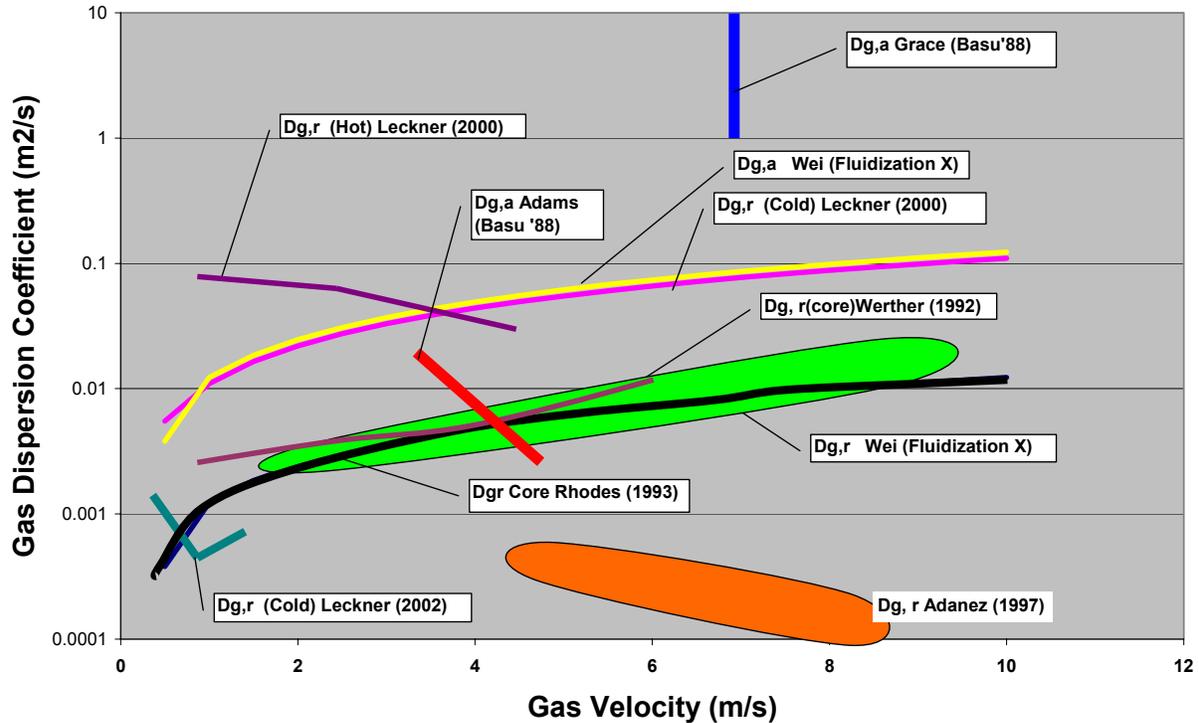


Figure 1 Gas Dispersion Coefficients

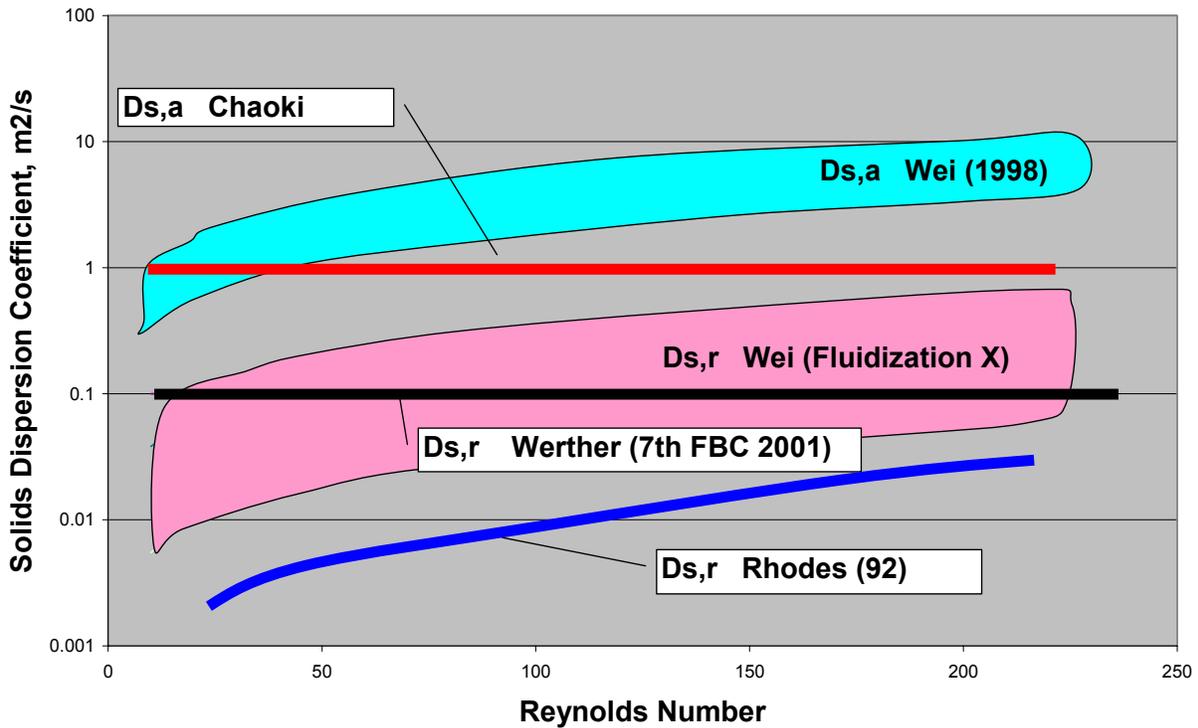
### Solids Dispersion Coefficients

The correlations for solids dispersion are presented in Table 3. The dispersion coefficients presented span more than 4 orders of magnitude. Closer inspection of Figure 2 reveals that one correlation for the solids axial dispersion is given and three correlations of the solids radial dispersion are given. The two correlations by Wei each vary by more than an order of magnitude over the expected particle Reynolds number. The correlations by Werther and Chaouki state that the solids dispersion is essentially constant. Werther identifies the radial dispersion coefficient as being  $0.1 \text{ m}^2/\text{s}$  while Chaouki identifies the axial dispersion coefficient as  $1.0 \text{ m}^2/\text{s}$ .

Three different techniques were used to determine the solids dispersion values presented in Figure 1. Wei used fluorescing particles, a flash system and photographic image analysis of the fluorescing particles. Werther conducted his experiments using dry ice as the tracer. Werther injected trace particle of dry ice looking at both  $\text{CO}_2$  concentration and temperature. Chaouki used radioactive gold particles as the tracer.

**Table 3 Solids Dispersion Coefficient Correlations**

Author	Model	Constraints
Werther (2002)	$D_{sr} \sim 0.1 \text{ m}^2/\text{s}$	None Identified
Wei (2001b)	$D_{sr} = 0.018 \text{Re}^{0.74} \exp(-19(1-\varepsilon)) \left(\frac{d_p}{d_{cir.sol.}}\right)^{-0.53} \left(\frac{\rho_p}{\rho_{cir.sol.}}\right)^{-0.72} \phi^{2.32}$	None Identified
Wei (1998)	$Pe_{sa} = 71.86(1-\varepsilon)^{0.67} \text{Re}^{0.23}$ $Pe_{sr} = 225.7(1-\varepsilon)^{-0.29} \text{Re}^{0.43}$	None Identified
Chaoki	$D_{sa} \sim 1 \text{ m}^2/\text{s}$	



**Figure 2 Solids Dispersion Coefficients**

**Mass Transfer Coefficients**

**Experimental Facilities Used in Dispersion Investigations**

A description of the operating conditions and experimental facilities for the review of the mass transfer data is presented in Table 4.

**Table 4 Conditions and Dimensions for Mass Transfer Investigations**

Author \ Facility	d (m)	h (m)	T (K)	P (kPa)	dp (um)	ps (kg/m <sup>3</sup> )	Gs (kg/m <sup>2</sup> s)	Ug (m/s)
Li	0.072	3	ambient	ambient	300	1200	6 & 12	1.86 & 2.12
Subbarao	0.025	1.05	ambient	ambient	196 & 390	2500	~15 to 60	4, 5 & 6
Bolland	0.411	8.5	333	ambient	134	2500	31 to 53	5.6 to 7.2
Kalil	0.09525	0.3	ambient	ambient	685 to 2000	800 to 10000	N/A	N/A
Resnick	.022 to .044	0	ambient	ambient	3 to 35 mesh	0	N/A	N/A
Venderborsch	0.015	1.17	750	ambient	65	1375	5 to 40	2.5 to 4.5

### Mass Transfer Coefficient Correlations

Six investigators have reported correlations for the interphase mass transfer coefficient in fluidized systems from research spanning 50 years. These are presented in Figure 3. There is little agreement between the reported results with values of the Sherwood number ranging from a low of  $10^{-5}$  reported by Bolland to a high of 200 as reported by Kalil.

Looking at the specific research that was conducted, it is possible to reduce this span by a couple orders of magnitude. Bolland conducted his research utilizing an ozone decomposition reaction. Ozone, upon first thought seems to be a good tracer to use, after all it decomposes to oxygen. Ozone decomposition is also highly dependent upon the moisture concentration in that gas and as no moisture data was taken and no effort made to control the moisture content during Bolland's test, his reported correlation for the Sherwood number is highly suspect. Eliminating this correlation from the analysis tightens the data range substantially, now being 0.01 to 200. The work of Kalil was conducted in a bubbling fluidized bed. He reports that the bed voidage is an important factor in the correlation of the Sherwood number. However, the application of this correlation to the riser of a CFB where voidages are on the order of 0.8 and higher likely introduces significant error in the estimate of the Sherwood number as his the voidage in his work varied about 0.6. Thus, it can be expected for the Sherwood number to fall between 0.01 and 10. This is still a significant difference, but no other factors could be identified which suggest that any of the other correlations should be discarded.

**Table 5 Mass Transfer Coefficient Correlations**

Li	$Sh = 2\varepsilon + 0.69\left(\frac{Re_{sl}}{\varepsilon}\right)^{0.5} Sc^{0.3}$
Subbarao	$Sh = 8.314 * 10^{-5} \left(\frac{U_o \rho_p}{G_s}\right)^{1.43}$
Bolland	$10^{-5} < Sh < 10^{-3}$
Zenenhoven	$Sh = 2\varepsilon + 0.89 Re_p^{0.5} Sc^{0.33}$
Kalil	$Sh = 1.77 \left(\frac{Re \varepsilon}{1 - \varepsilon}\right)^{0.56} Sc^{0.33}$
Resnick	Graphical J factor
Zenenhoven	$Sh = 2\varepsilon + 0.89 Re_p^{0.5} Sc^{0.33}$
Venderborsch	Sh=Sh(Uo, solids fraction) Sh decreases with solids fraction and increase with Uo

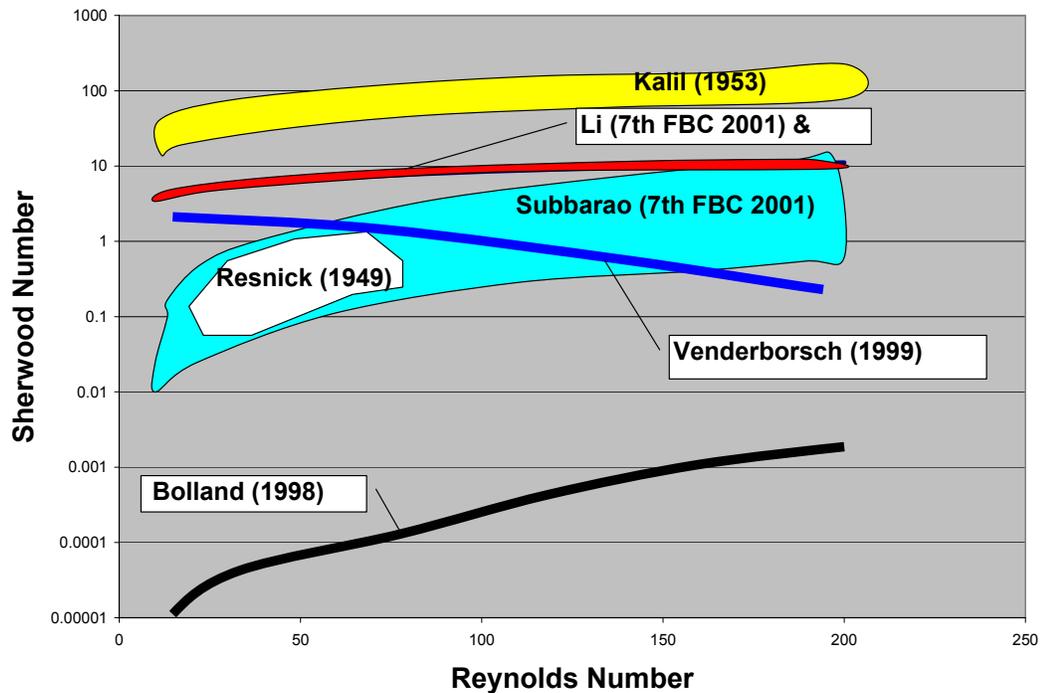


Figure 3 Mass Transfer Coefficients

## Discussion

The first two questions that arise with respect to understanding the wide level of discrepancies in the reported literature values for the dispersion and mass transfer coefficients are:

- Were all the researchers working in the same flow regime as the solids and gas behavior is significantly different in the various regimes?
- Were all the test sections in a region of the riser well away from the effects of the entrance and exit, that is, was the flow regime fully developed?

The answers to these questions coupled to answering similar questions by the design/application engineer will help to eliminate some of the correlations presented in the literature and provide smaller ranges for these important transport properties.

Flow regime definitions have been developed to both qualify and quantify the solids and gas behavior in the risers of circulating fluidized beds and transport reactors. Due to the complexity of the system, different definitions have been developed by different investigators. Review papers from Grace, Kato, Mei, Rhodes and Shadle, as well as discussions with Monazam have been analyzed to develop a qualitative picture of the potential flow regimes that the mass transfer and dispersion researchers presented above were likely to have encountered. This qualitative picture is presented in Figure 4.

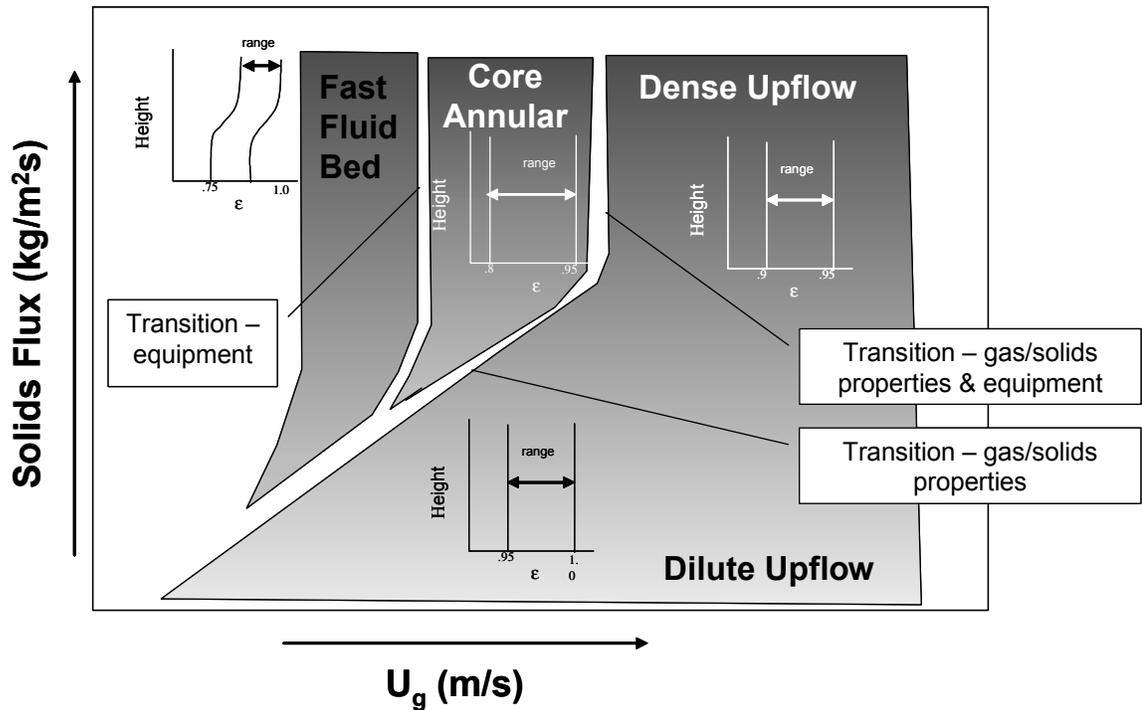


Figure 4 Flow Regimes in Circulating Fluidized Systems

The definition of a flow regime takes, at a minimum, three variables; solids flux, gas velocity and voidage (pressure) profile. Four regimes are presented in the figure: Fast fluid bed, Core annular, Dilute upflow and Dense upflow, although there is controversy as to whether or not the fast fluid bed regime and the core annual regime are the same as one definition simply states the co-existence of a dilute region and a dense region with the dilute region being above the dense region (denoted fast fluid bed in figure) or the dilute region being surrounded by a dense region (denoted core annular in figure). The transition from dilute up flow to either the fast fluid bed regime or the core annular regime depends upon the gas and solids properties. It is in these regions of transition that most researchers work because of the physical limitations of their equipment. Therefore, depending upon what voidage profile existed in each of the experimental investigations, the flow regime may differ significantly.

Dilute upflow is the simplest of the flow regimes. It consists of solids uniformly dispersed across the riser moving upward. At a particular gas velocity, increasing the solids flux decreases the voidage from 1 to about 0.95. Now depending upon the particle type and equipment a transition may be reached. At high gas velocities, no transition is reached and there is a gradual transformation from dilute upflow to dense upflow. At lower velocities, however, a transition is reached in which the riser solids profile significantly changes into one of two distinct patterns, fast fluid bed or core annular. Which of these

two regimes is reached depends not only on the gas velocity, but also on the equipment.

The fast fluid bed regime, as stated above, has a dense bottom zone and a dilute upper zone. By changing the solids flux, the solids in the upper region have collapsed into a dense lower region. In the dense region, solids flow downward as well as upward. Now clearly, the dispersion and the mass transfer are likely to be significantly different in these two regimes and significantly different with height. So depending on where the experimental measurements were taken is also likely to significantly affect the results. A similar phenomenon is obtained at slightly larger gas velocities with a transition to core annular flow, providing the equipment supports a core annular flow regime. In this circumstance, the uniformly distributed solids collapse to the wall and provide significant downflow. Again, this is significantly different behavior than dilute upflow and is likely to have significantly different values for dispersion coefficients and mass transfer coefficients.

As noted above, whether or not the tests were conducted in a fully developed flow field is also likely to affect the results of the work and contribute to the wide spread in the reported literature values for the mass transfer coefficient and the dispersion coefficients. Recently, Monazam has developed a correlation to predict the length of the acceleration zone. This work was conducted in a 0.3 m facility with a height of 16 m using 65  $\mu$ m glass beads, 806  $\mu$ m cork particles and 230  $\mu$ m coke particles. Using the Monazam correlation to predict the acceleration length and comparing that length to the reported testing height reveals that about 50% of the data reported in the literature surveyed was obtained within the acceleration zone.

## Conclusion

Clearly the correlations presented by the researchers herein are not universally applicable. At present, there is no basis for recommending any of these correlations in favor of the others. The reasons for the wide discrepancy are likely due to the fluidization flow regime in which the researcher was working. Research needs to be conducted in a systematic manner across the operating regime map to determine these crucial transport properties. Studies conducted within the acceleration zone and in the fully developed zone need to be conducted and reported for use within the respective zones. In some hardware configurations, the acceleration zone may be in excess of 50 percent of the bed riser height.

## Nomenclature

C	Concentration, mol/m <sup>3</sup>
d	diameter, m
G	Gas flux, kg/m <sup>2</sup> s
G <sub>s</sub>	Solids flux, kg/m <sup>2</sup> s
k	rate constant or coefficient, m/s
N	particle count

Pe	Péclet
R	Reaction Rate, mol/s
$r^*$	Core radius, m
Re	Reynolds Number
Sc	Schmidt Number
Sh	Sherwood Number
t	time, s
U	Velocity, m/s
x	mole fraction

#### Greek

$D$	Diffusivity, $m^2/s$
$\varepsilon$	Voidage
$l$	Length in Pe number, m
$\mu$	Viscosity, $kg/ms^2$
$\pi$	Pi
$\rho$	Density, $kg/m^3$

#### Subscripts

0	initial
a	axial
c	chemical
d	mass transfer
g	gas
i	index
hyd	hydraulic
m	index
n	index
p	particle
r	radial
s	solid
sl	slip

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## **Acknowledgement**

This research was performed while the author held a National Research Council Research Associateship Award at the Department of Energy National Energy Technology Laboratory (Morgantown).