Mass Transfer Resistance in Downer Reactors

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Abstract

Conversions in downer reactors are lower than what is expected from a pseudo homogeneous plug flow reactor model. This was attributed to particles moving as clusters, axial gas mixing and gas to particle mass transfer. In the present paper, a two phase tanks in series model is developed for the downer reactors considering gas to particle mass transfer. The model is compared with recent experimental observations on a downer reactor reported in the literature to estimate mass transfer coefficients. Estimated Sherwood Numbers are observed to increase as a function of the ratio of gas to particle velocities. This result is in line with the recently reported direct observations on gas to particle mass transfer in downers.

Introduction

Downers are receiving attention as a possible efficient gas solid contactors [1]. Talman et al [2] and Talman and Reh [3] reported experimental observations on the gas-solid catalytic reactions in downers reactors. Conversions were observed to be lower than the expected from a pseudo homogeneous plug flow model at higher solid loadings. This was attributed to particles moving as clusters. Fan et al [4] investigated downer reactor performance by catalytic ozone decomposition and observed that gas to particle mass transfer may be affecting the conversions.

In the present paper, a two phase tanks in series model is developed for the downer reactor performance considering gas to particle mass transfer. The model parameters are evaluated in the light of the experimental observations of Fan et al [4].

The Model

In a downer reactor, reactant gas reacts at the surface of catalyst particles as gas and particles flow concurrently downward through a vertical pipe. Along the length of flow axially, the reactor is visualized to consist "n tanks" in series through which gas and particles flow down. Each tank consists of gas phase and particle phase, each well mixed with in itself, with mass transfer of reactants and products between the two phases. All the tanks are assumed to be of equal size with a height of Δz.

A schematic of the structure of the model is shown in fig.1. Processes taking place in "ith compartment" are shown in fig.2. The model is applied to develop equations for decomposition of ozone by 1^{st} order irreversible catalytic reaction. Component material balance for each phase in "i"th compartment can be written as

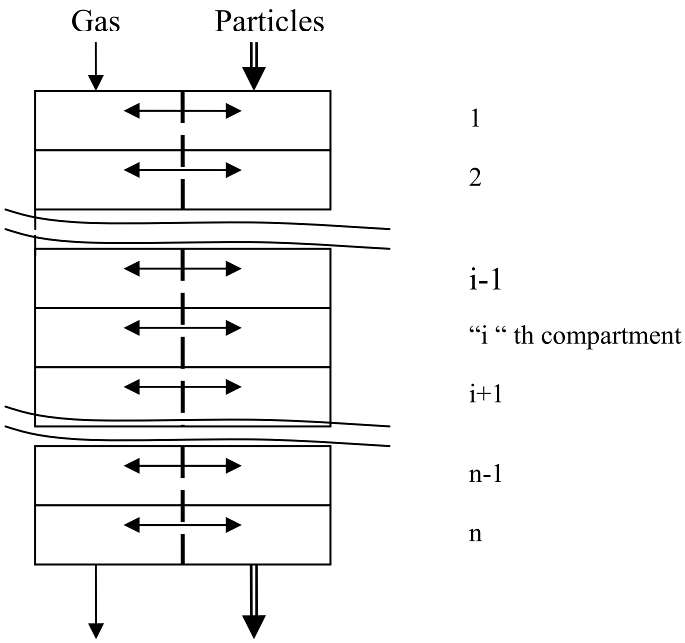

Fig.1. Structure of the two phase tanks-in-series model for a downer reactor.

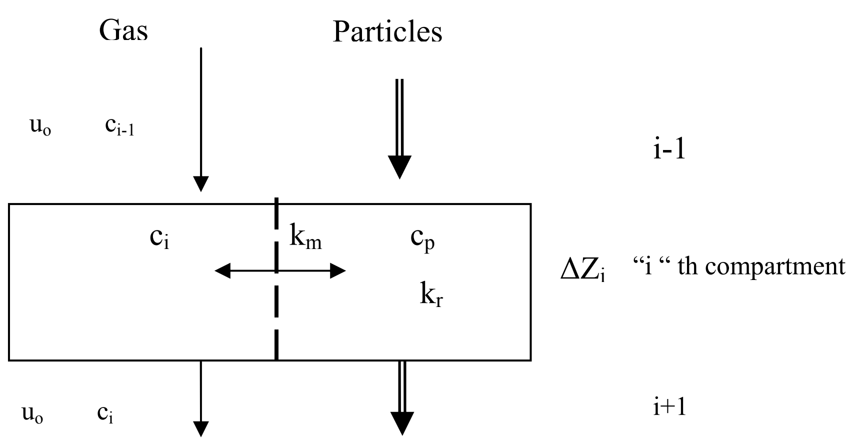

Fig. 2. Mass Transfer and reaction processes between gas phase and particle phase in ith compartment

For gas phase

input rate = output rate + mass transfer rate

$$u_0 c_{i-1} = u_0 c_i + k_m \Delta z (1-\varepsilon) \frac{s_p}{v_p} (c_i - c_p) \tag{1}$$

for particle phase

mass transfer rate = reaction rate

$$k_m \Delta z (1-\varepsilon) \frac{s_p}{v_p} (c_i - c_p) = k_r \Delta z (1-\varepsilon) \rho_p c_p$$

Eliminating particle phase concentration between these equations

$$\frac{c_i}{c_{i-1}} = \left[1 + \frac{\Delta Z (1-\varepsilon)}{u_o} \frac{1}{\dfrac{1}{k_r \rho_p} + \dfrac{1}{k_m \, s_p / v_p}} \right]^{-1} \tag{2}$$

Number of tanks in the downer can be obtained as

$$n = \frac{L}{\Delta z} \tag{3}$$

Then, fraction of reactant not converted in the reactor can be obtained as

$$(1-X) = \frac{c_{out}}{c_{in}} = \left[1 + \frac{L(1-\varepsilon)}{n\, u_o} \frac{1}{\dfrac{1}{k_r \rho_p} + \dfrac{1}{k_m \, s_p / v_p}} \right]^{-n} \tag{4}$$

This formulation explicitly brings out the importance of mass transfer and degree of axial mixing in heterogeneous catalytic reactors. With zero mass transfer coefficient no conversion is possible; with infinitely large mass transfer coefficient, the reaction can be considered as homogeneous in nature. The reactor behaves as a CSTR for n=1 and as a PFR for extremely high values of n.

Model Evaluation

Fan et al [4] measured Ozone conversion (X) by catalytic decomposition along the reactor length (L) in a downer reactor (column height of 8.5 m and diameter of 0.09 m) as a function of gas velocity (2.2 m/s to 3.7 m/s) and particle flux (8.4 to 26.8 kg/m^2/s. FCC catalyst particle of 62 micron size with density of 1747 kg/m^3 impregnated with Ferric Oxide were used for the study. The equations developed for two phase tanks in series model for the downer reactor can be used to evaluate this data.

Equation (4) can be rearranged as

$$\frac{L(1-\varepsilon)}{n\,u_o\left\{(1-X)^{-1/n}-1\right\}} = \frac{1}{k_r\rho_p} + \frac{1}{k_m\, s_p/v_p} \qquad (5)$$

Downer reactors consist of three zones along the axial length; in the first zone particles accelerate due to gas drag force and gravity; in the second zone particles accelerate due to gravity while drag force retards the acceleration; in the third zone particles will be moving at terminal velocity as the gravity and drag force balance each other. Particle holdup in the particle accelerating zone can be slightly more than the terminal zone. Particle holdup is estimated for each operating gas velocity and particle flux by the equation proposed by Xiao-Bo Qi et al [5] for the terminal zone

$$(1-\varepsilon) = 0.125\left(\frac{W}{\rho_p\left(u_o+u_t\right)}\right)\left(\frac{u_o}{\sqrt{gD_p}}\right)^{0.25} Ar^{0.15} \qquad (6)$$

and is assumed to represent the acceleration zone as well.

Zhang and Zhu [6] observed that there can be particle mixing accompanied by gas mixing in downers at low gas velocities. Radial non uniformity in particle holdup and clustering of particles can create mixing of continuous (gas) phase similar to liquid mixing in bubble columns operating in churn turbulent regime. Based on the experimental observations on axial concentration profiles reported by Fan et al (2008), there could be a lot of longitudinal backmixing. In view of this, the parameter (LHS of the equation)

$$\frac{L(1-\varepsilon)}{n\,u_o\left\{(1-X)^{-1/n}-1\right\}}$$

was evaluated for various values of n ranging between 1 to 10000. This parameter is observed to vary as a function of gas velocity and particle flux. This could be due to gas to particle mass transfer coefficient term in the RHS of equation (5).

Luo et al [7] investigated mass transfer between gas to porous particles in downers and observed that mass transfer coefficients are a function of the parameter ($u_o\rho_p$/W). Raghuramulu et al [8] investigated mass transfer between gas to non-porous coarse sand particles in downers and proposed

$$Sh_{pd} = 8\ .10^{-6}\left(\frac{u_o\rho_p}{W}\right)^{1.43} \qquad \text{for} \quad D_t=2.5\text{ cm}$$

$$Sh_{pd} = 5\ .10^{-5}\left(\frac{u_o\rho_p}{W}\right)^{0.94} \qquad \text{for} \quad D_t=5\text{ cm} \tag{7}$$

For gas to particle mass transfer in risers, assuming particles move as clusters, Subbarao [9] proposed an equation of the form

$$\frac{Sh_{pr}}{Sh_{sp}} = c_1\left(\frac{u_o\rho_p}{W}\right)^m \tag{8}$$

From eqs (5) and (8)

$$\frac{L(1-\varepsilon)}{n\ u_o\left\{(1-X)^{-1/n}-1\right\}} = \frac{1}{k_r\rho_p} + \frac{D_p^{\ 2}}{6D_m Sh_{sp}c_1}\left(\frac{W}{u_o\rho_p}\right)^m \tag{9}$$

This equation suggests a linear relation between

$$\frac{L(1-\varepsilon)}{n\ u_o\left\{(1-X)^{-1/n}-1\right\}} \quad \text{and} \quad \frac{1}{Sh_{sp}}\left(\frac{W}{u_o\rho_p}\right)^m$$

To proceed with the calculations, values for n and m have to be assigned; values 1, 2, 3, 4, 5, 10, 100, 1000 and 10000 are explored for n; m values in the range of 0.9 to 1.4 are explored. The intercept is $(1/k_r\rho_p)$ and has to be positive. Fan et al [4] reported k_r value as 0.098 ml/g cat/s and density ρ_p is 1.747 g/cm^3. With these values, the intercept is expected to be 5.84 s. However the intercept is much lower indicating the rate constant, most probably, to be much higher. Also, experimentally observed conversions in downer reactor are much higher than what can be expected in a plug flow reactor with reaction rate constant of 0.098 ml/gcat/s. In view of this, m value for possible intercept values of 0, 0.006, 0.01 and 0.02 as a function of n value are estimated and shown in fig.3. Intercept value of 0.006 corresponds to 0.098 m^3/kg cat/s and is used for further discussion. Axial concentration profiles suggest that n value is closer to 1. For a value of n equal to 1, value of m is 1.22.

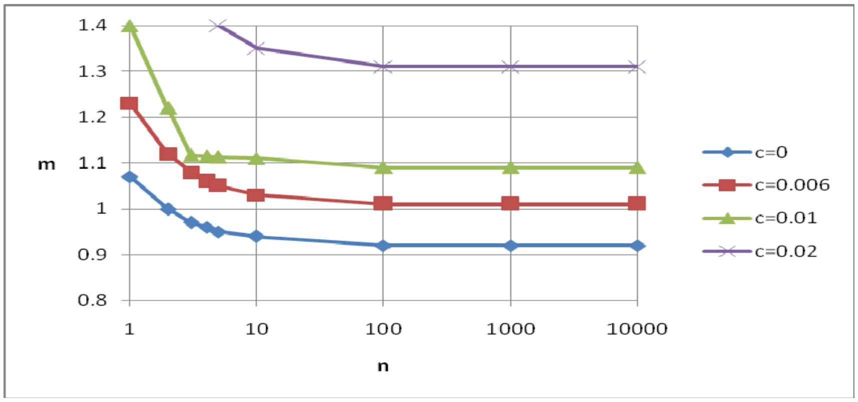

Fig3. Parameter m as a function of n for various values of the intercept.

Data Fan et al [4] for n equal to 1 and m equal to 1.22 are presented in fig 4.
With this mass transfer correlation for this data is obtained as

$$\frac{Sh_{pd}}{Sh_{sp}} = 2.19\text{x}10^{-7}\left(\frac{u_o \rho_p}{W}\right)^{1.22} \tag{10}$$

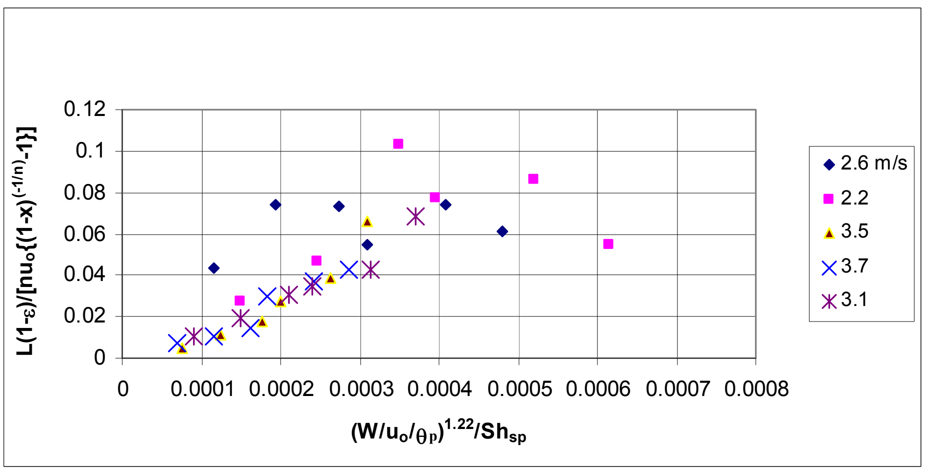

Fig.4 Data Fan et al [4] for n equal to 1 and m equal to 1.22

In fig.5, data of Fan et al [4] for n equal to 10000 and m equal to 1 are presented for comparison with fig.4.
With this mass transfer correlation for this data is obtained as

$$\frac{Sh_{pd}}{Sh_{sp}} = 4.2\ .10^{-7}\left(\frac{u_o \rho_p}{W}\right) \tag{11}$$

The difference in the estimated mass transfer correlation is not very significant. However, Axial concentration profiles will be significantly different. As the experimentally observed axial concentration profiles indicate significant axial mixing, one tank appears to be more representative.

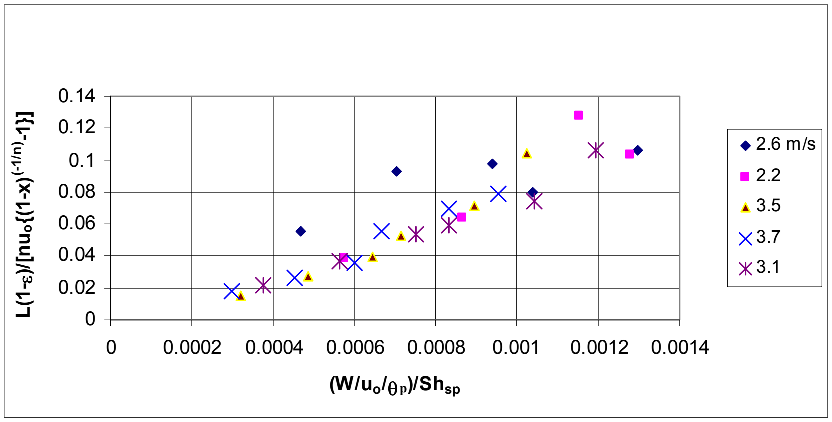

Fig.5 Data of Fan et al [4] for n equal to 10000 and m equal to 1

Concluding Remarks

This value of c_1 (2.19×10^{-7} or 4.2×10^{-7}) is much lower than the value reported by Raghuramulu et al [8] for sand particles. In risers, for fine particles, model of Subbarao [9] brought out the effect of particle size by the following equation

$$\frac{Sh_{pr}}{Sh_{sp}} \propto \left(\frac{D_p}{D_v}\right)^2 \left(\frac{\rho_P u_o}{W}\right)^m \qquad (12)$$

This equation brings out the importance of particle size. Even this equation predicts an order of magnitude higher Sherwood numbers than the ones estimated from the downer reactor data of Fan et al [4]. Thus, it appears that Sherwood numbers in downers are smaller compared to risers. Further experimental study on the effect of mass transfer on downer reactor performance is needed.

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Nomenclature

Ar	Archimedes Number
c_1	Constant
c_{i-1}	Gas phase concentration in the (i-1)th compartment
c_i	Gas phase concentration in the ith compartment
C_{in}	Reactor inlet gas phase concentration
C_{out}	Reactor outlet gas phase concentration

C_p	Particle phase concentration in the ith compartment
Dm	Diffusion Coefficient
D_p	Diameter of particles
D_t	Diameter of the Downer column
D_v	Diameter of the void
g	Acceleration due to gravity
i	Compartment number
K_m	Mass transfer coefficient
K_r	Reaction Rate Constant
L	Length of the downer
m	index, constant
n	Number of compartments in series
s_p	External surface area of catalyst particle
Sh_{pd}	Sherwood Number based on particle size in downers
Sh_{pr}	Sherwood Number based on particle size in risers
Sh_{sp}	Sherwood Number based on particle size for single particle
u_o	Gas velocity
u_t	Particle terminal velocity
v_p	Volume of particle
W	Particle mass flux
X	Conversion of Ozone

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