Liquid Distribution in Trickle-Bed Reactors

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Experimental measurements on liquid distribution in a trickle-bed reactor are presented, as well as the effect of liquid and gas flow on radial liquid distribution. The influence of liquid density and surface tension was also investigated. The model for liquid distribution proposed by Herskowitz and Smith in 1978 was modified to incorporate the effect of particle orientation. Model parameters are correlated with liquid and gas flow rates, as well as liquid surface tension.

Introduction

Trickle bed reactors (TBR) consist of a fixed bed of catalyst particles over which gas and liquid phase flow cocurrently downward. In certain cases the liquid phase and gas phase flow cocurrently upward. Downward flow is mostly used in industry, because flooding is not a problem at high flow rates. Trickle-bed reactors have wide application in the petroleum, petrochemical, and chemical industries. These reactors are also used in environmental and biochemical processes. Recently, Saroha and Nigam (1996) compiled an exhaustive review on trickle-bed reactors.

In a trickle-bed reactor, as the phases flow downward, the liquid has a tendency to move toward the reactor wall, which is undesirable since the wall is not catalytically active. This tendency is attributed to the lower resistance to flow due to increased voidage near the wall. Normally reactor interiors are necessary to take care of any maldistribution that has a bearing on a pressure drop across the reactor.

Sundaresan (1994) reported that if the reactor is to operate in the trickle-flow regime, the startup procedure is important for uniform wetting. However, this trend was not observed in pulse flow. For pulse flow Sundaresan (1994) suggested injecting the gas directly into the packed bed through uniformly spaced nozzles to promote the formation of pulses uniformly over a cross section of the bed. Niu et al. (1996) used a sheet of convex hemispheres in a staggered arrangement to line the inside of a tube to eliminate wall effect in a packed bed. It was noticed that the lining was effective in reducing the voidage adjacent to the reactor wall, which led to improved liquid distribution. Moller et al. (1996) suggested using a large-particle top layer to improve liquid distribution. Lutran et al. (1991) also studied the liquid distribution in trickle beds by using computer-assisted tomography. They observed that the flow pattern depends strongly on whether the bed had been prewetted by flooding the column with liquid or was initially dry.

Wall flow is defined as the excess liquid flow near the reactor wall. Wall flow depends on the ratio of reactor diameter to catalyst particle diameter; physicochemical properties of the liquid (density, viscosity, surface tension); liquid and gas flow rates; wettability, porosity, shape, and orientation of the catalyst particles. Compared to other hydrodynamic parameters of TBR, relatively few studies have been reported on the radial liquid distribution. Reactor-to-particle-size ratio (D/d_p) has been reported to have a significant effect on radial distribution. Different values of D/d_p above which no or reduced wall flow has been observed, have been reported in the literature. Baker et al. (1935) proposed a minimum D/d_p value of 12, while Porter et al. (1968a,b) suggested a value of 20 to 25. Prchlik et al. (1975a,b) noticed wall flow to be less than 10% for $D/d_p > 25$, while Herskowitz and Smith (1978) suggested a minimum value of 18 for D/d_p for uniform flow. Al-Dahhan and Dudukovic (1994) reported that, even at elevated pressures, a value of $D/d_p > 20$ minimizes the liquid maldistribution. Herskowitz and Smith (1978) noticed that uniform equilibrium distribution was achieved at lower ratios of D/d_p, and at lower bed depths for granular particles than for spheres or cylinders. This variation in the D/d_p ratio observed by different investigators could be an effect of particle

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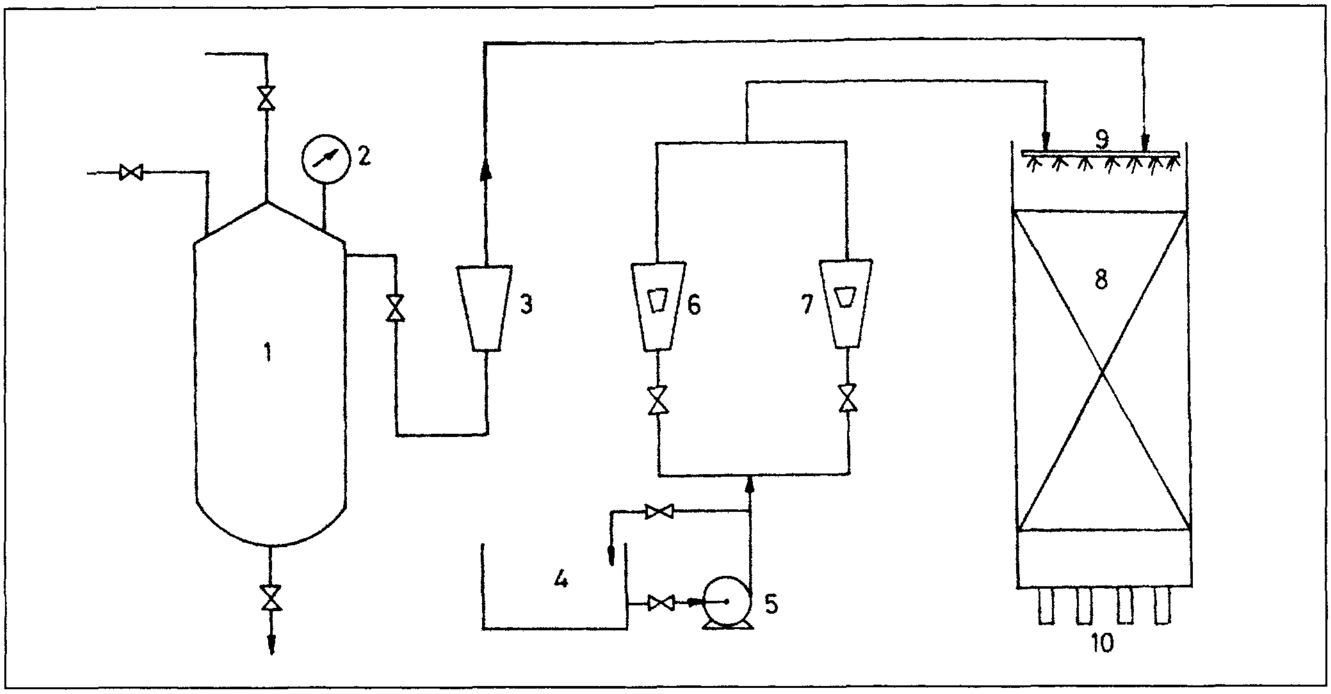

Figure 1. Experimental setup.

1. Air saturation tank; 2. pressure gauge; 3. air rotameter; 4. liquid tank; 5. liquid feed pump; 6, 7. liquid rotameter; 8. trickle-bed reactor; 9. liquid distributor; 10. liquid collector.

orientation in the bed that is of particular significance for cylindrical extrudates, as reported by Ng and Chu (1987). It implies that the method of loading the bed has an effect on the liquid distribution. It has also been reported that for low surface tension of liquids, wall flow decreases. Onda et al. (1973) observed almost negligible dependence of liquid spreading factor on the liquid viscosity and the liquid flow rate. They also noticed an increase in the spreading factor with the increase in particle diameter.

Published information on the modeling of liquid distribution is rather scanty. Herskowitz and Smith (1978) proposed a mechanistic model to predict radial liquid distribution. The model parameters were estimated from experimental results for $D/d_p < 30$, using only an air–water system. Moreover Herskowitz and Smith (1978) have reported the values of model parameters for specific packing material without correlating them with the operating flow rates and physicochemical properties of the liquid. This article presents the experimental data on liquid distribution obtained with kerosene and water as liquids and cylindrical alumina extrudates as packing to represent industrial TBR used in the petroleum industry. The mechanistic model suggested by Herskowitz and Smith (1978) was adopted with appropriate modifications that take the particle orientation into consideration. Model parameters are described as a function of liquid and gas flow rates and liquid surface tension.

Experimental Setup

The experimental setup is shown in Figure 1. Experiments were performed in a 15.2-cm-ID glass column wherein liquid and gas can be fed through a distributor at the top of the column. The column was packed with commercial 1.5-mm alumina extrudates supported on a 2-mm mesh screen. Extrudate length distribution is shown in Figure 2. The average extrudate length using this distribution is estimated to be 7.5 mm. Liquid was pumped from a tank through the rotameters (low- and high-range). Air, drawn from a compressor, passed through an air saturator, pressure regulator, control valves, and air rotameter before entering the column at the top. The liquid distributor (Figure 3) was made of 6.4-mm-ID stain-

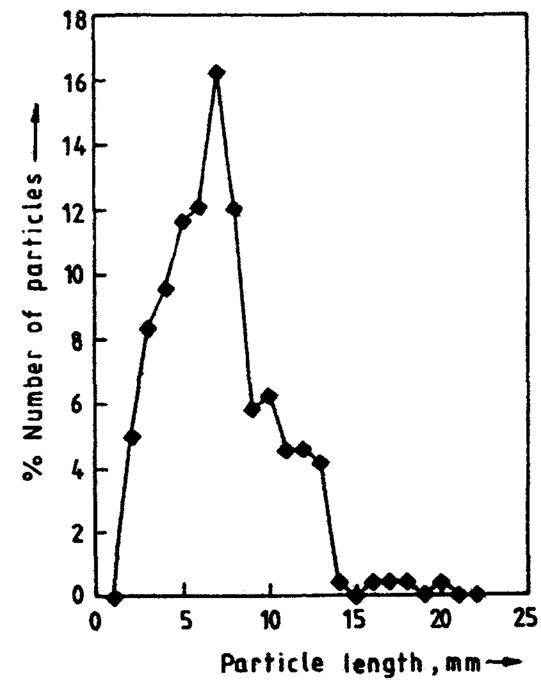

Figure 2. Particle-size distribution.

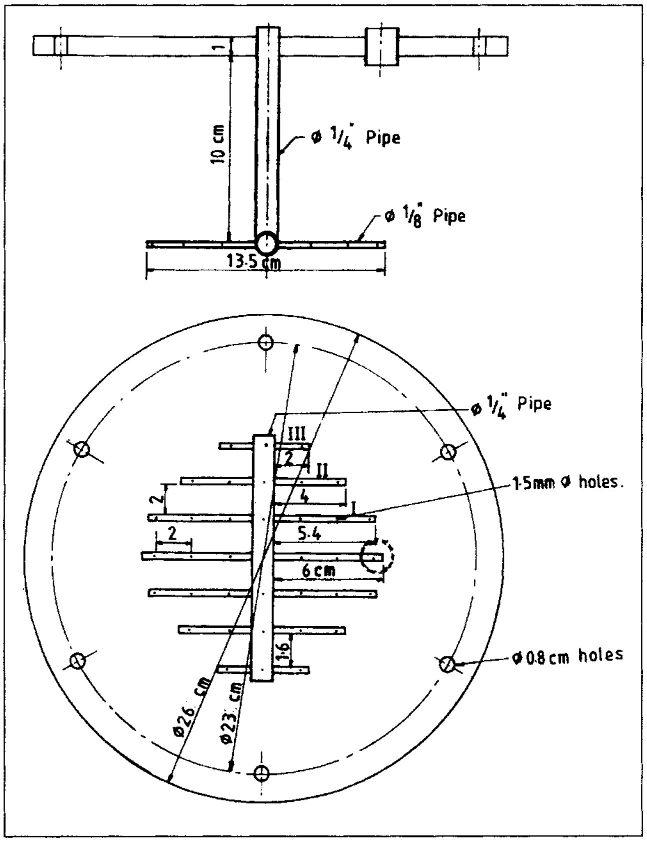

Figure 3. Liquid distributor.

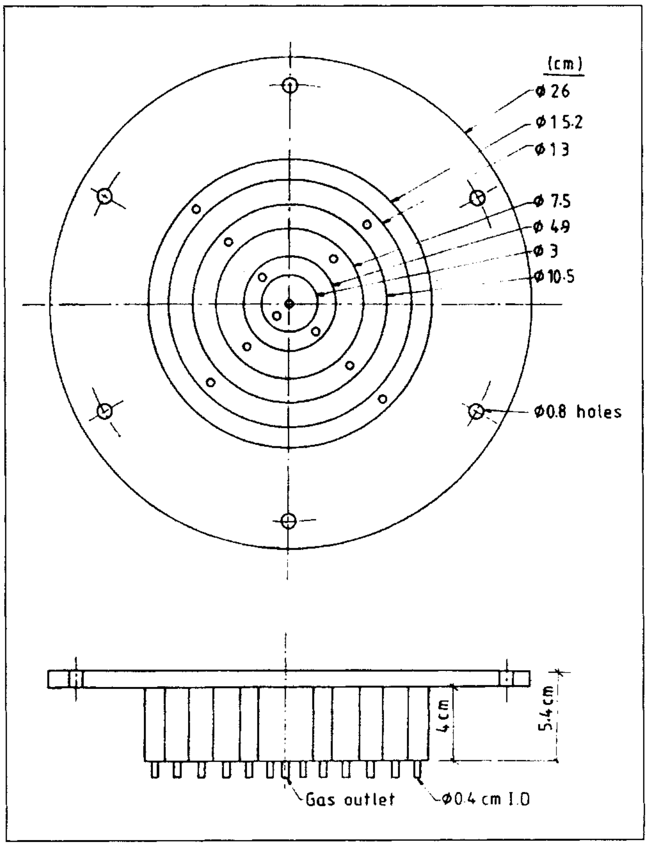

Figure 4. Liquid collector.

Table 1. Collector Dimensions

Annulus Number	Annulus Dia. (m)	Area (m^2)	% of Total Area
1 (Center)	0.03	0.000706	3.89
2	0.049	0.001178	6.56
3	0.075	0.002532	13.90
4	0.105	0.004241	23.37
5	0.13	0.004614	25.43
6 (Wall)	0.152	0.004872	26.85

less-steel tubes, to which 3.2-mm-ID tubes were attached. There were 37 1.5-mm holes arranged in a square pitch of 2 cm. The distributor was placed 10 cm over the catalyst bed. A 1-cm-ID tube, located at the center of the column and placed 1 cm above the bed, was used in the experiments where liquid was fed from a point source.

The liquid collector (Figure 4) at the bottom of the column consisted of six annular sections to facilitate the measurement of liquid distribution over the cross section of the column. The dimensions of the annular areas and annular fractions are given in Table 1. Liquid in the annulus was collected through 12 holes; two holes were located diametrically opposite in each annulus. The experiments were performed with water, ethylene glycol, and kerosene. The surface tension of water was reduced from 0.073 N/m to 0.031 N/m by adding 0.22% (by weight) surfactant Hydroxid X 200. The physical properties of the liquids and the range of liquid and gas flow rates and bed heights studied are listed in Table 2. A total of 71 experiments with a uniform feed source and 216 experiments with a point-source feed were performed at ambient conditions (Saroha, 1998).

Results and Discussions

Effect of liquid flow rate

An increase in liquid flow rate was found to reduce the percent of liquid flow in the outermost annulus. This reduction was observed both with and without gas flow rates, as presented in Figure 5. Increased uniformity of liquid distribution at higher liquid loads is obvious, a trend that was reported in earlier studies (Moller et al., 1996).

Effect of gas flow rate

At constant liquid flow, the increase in gas flow rate was found to decrease the liquid flow in the last annulus. This is shown in Figure 6. An increase in either the gas or liquid flow rate results in a higher pressure drop across the bed,

Table 2. Physical Properties of Liquids and Range of Operating Conditions

Liquid	Density (kg/m^3)	Surface Tension (N/m)	L ($kg/m^2 \cdot s$)	G ($kg/m^2 \cdot s$)
Water	1,000	0.072	1.56–5.06	0–0.027
Kerosene	796	0.031	1.29–4.34	0–0.021
Ethylene Glycol	1,100	0.049	0.69–3.59	0–0.021
Water + surfactant	1,000	0.031	1.56–5.06	0.0

For point-source feed, experiments were performed at bed heights of 0.55, 0.3, 0.15, and 0.1 m, whereas for uniform feed distribution bed height was kept constant at 0.55 m.

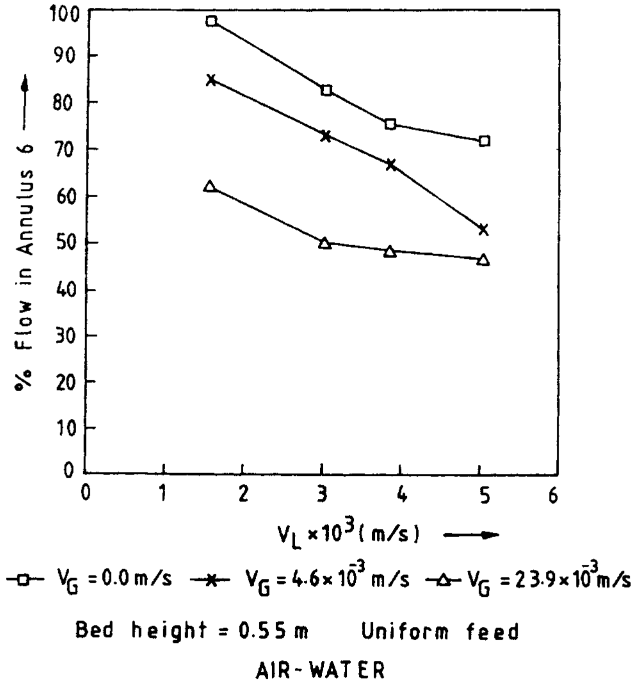

Figure 5. Effect of liquid flow rate on wall flow.

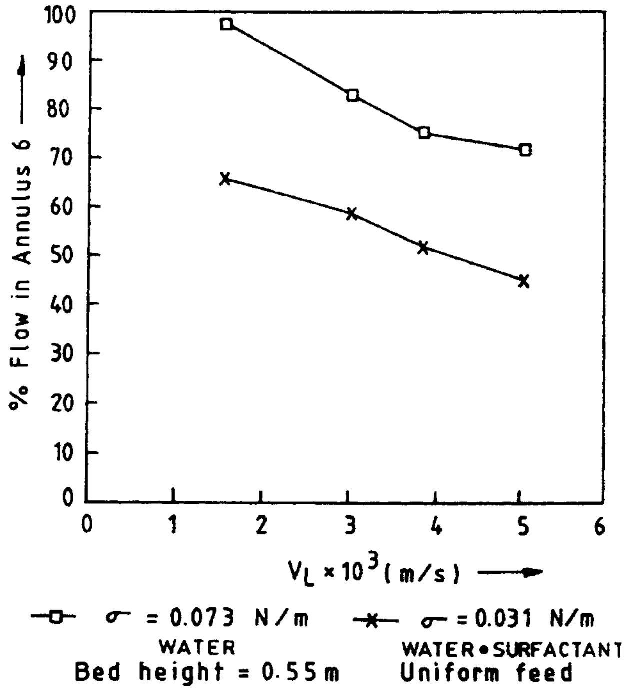

Figure 7. Effect of liquid surface tension on wall flow.

which in turn improves the liquid distribution. Sylvester and Pitayagulsarn (1975) also noticed better liquid distribution at higher gas flow rates for a certain range of liquid flow rate.

Effect of surface tension

Figure 7 shows the effect of surface tension on the flow in the last annulus for a fixed liquid viscosity and density. The reduction in surface tension was achieved by adding 2,200 ppm surfactant in water. Experiments were performed without any gas phase, as excessive foaming was observed with gas flow. Wall flow was found to decrease with a decrease in liquid surface tension. This observation is in agreement with that of Herskowitz and Smith (1978) for air–water system, while Moller et al. (1996) observed no such trend. This lack of agreement could be due to the different approaches used in these studies to deduce this behavior. Moller et al. (1996) used second-order rate expression to assess the reactor utilization parameter and, in turn, estimate liquid distribution, while both the present study and Herskowitz and Smith (1978) made actual liquid flow measurements.

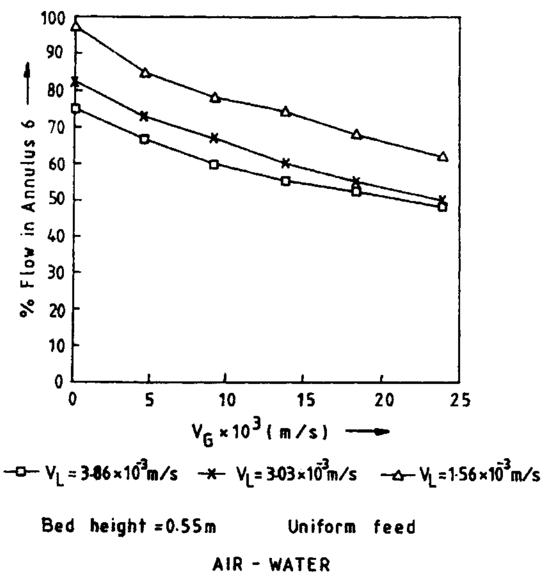

Figure 6. Effect of gas flow rate on wall flow.

Effect of density

The effect of density was deduced by performing experiments with kerosene and water doped with surfactant. Both liquid systems had the same surface tension but different densities. Figure 8 presents the liquid-density effect on the flow in the annulus near the wall (annulus 6). It is evident that decrease in density improves liquid distribution. This effect is more pronounced at higher liquid velocity.

Model for radial liquid distribution

Theory. Herskowitz and Smith (1978) have proposed a mechanistic model to predict radial liquid distribution in trickle-bed reactors. Radial distribution of liquid in a packed bed is conceived through the particle contact points, both radially outward and inward. The ratio of outward contact points to inward contact points dictates the probability of outward flow (toward the wall) of liquid, which increases with an increase in particle size. If the reactor bed is assumed to be packed with n layers of particles in the vertical direction and m concentric annulus particle rings in each layer, then the number of vertical layers, n, can be given as

$$n = H/d_p \tag{1}$$

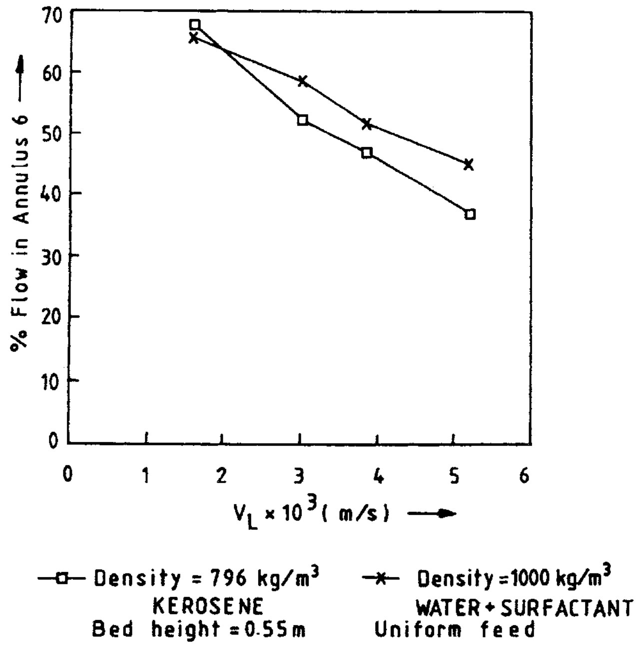

Figure 8. Effect of liquid density on wall flow.

and the number of annulus rings, m, can be given as

$$m = (D/2)/d_p. \tag{2}$$

The flow network at the center and at the wall of the column is shown in Figures 9, 10 and 11. S and f are the model parameters, which are assumed to be independent of the bed height. Here S is the fraction of liquid in an annular ring moving in the radial direction (both inward and outward), while f is the fraction of the wall flow returning to the contact points in the mth layer of the packed bed.

The flow to the ith annulus and $(j+1)$ layer can be given as

$$F_{i,j+1} = \left(1-\frac{S}{2}\right)F_{i,j} + \left(\frac{S}{4}\right)\left(\frac{2r_i-1}{2r_i-2}\right)F_{i-1,j} + \left(\frac{S}{4}\right)\left(\frac{2r_i+1}{2r_i+2}\right)F_{i+1,j}. \tag{3}$$

Equation 3 is valid from $i = 2$ to $m-1$ and $j = 1$ to n.

For the annulus at the center of the packed bed (i.e., $i = 1$) and at the reactor wall (i.e., $i = m$), the flow can be written as

$$F_{1,j+1} = \left(1-\frac{S}{2}\right)F_{1,j} + \left(\frac{S}{8}\right)\left(\frac{2r_i+1}{r_i+1}\right)F_{2,j} \tag{4}$$

$$F_{m,j+1} = f^2 W_j + \left[\left(\frac{Sf}{2}\right)\left(1+\frac{1}{2r_m}\right) + \left(\frac{S}{4}\right)\left(1-\frac{1}{2r_m}\right) + 1 - S\right]F_{m,j} + \left(\frac{S}{4}\right)\left(\frac{2r_m-1}{2r_m-2}\right)F_{m-1,j}. \tag{5}$$

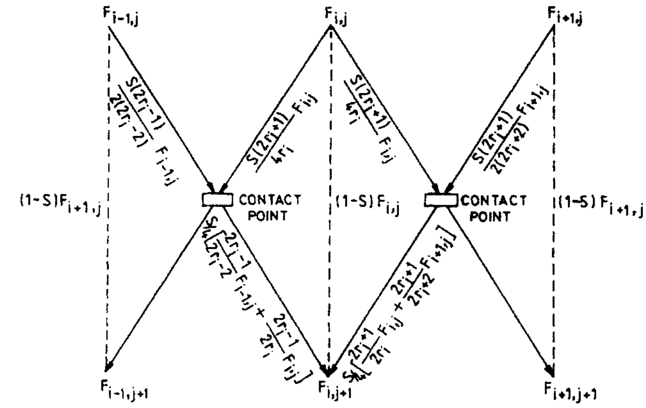

Figure 9. Flow network in the column.

The wall flow at the $(j+1)$ layer is given by

$$W_{j+1} = (1-f)\left[(1+f)W_j + \left(\frac{S}{2}\right)\left(\frac{2r_m+1}{2r_m}\right)F_{m,j}\right]. \tag{6}$$

In the preceding expressions the radial flow is governed by the ratio of outward particle contact points to inward points. For annulus i this ratio would be

$$\frac{\text{Flow}_{\text{outward}}}{\text{Flow}_{\text{inward}}} = \frac{2r_i+1}{2r_i-1}. \tag{7}$$

In Eq. 7 r_i is the dimensionless radial position. As the value of r_i increases, the ratio given in Eq. 7 approaches unity. This means that for higher values of r_i (i.e., in packed beds with a large D/d_p ratio) the liquid is distributed equally in both the outward and inward directions.

The value of r_i for a bed of fixed diameter D depends on the particle size (d_p). Equivalent particle size for cylindrical extrudates as given by Herskowitz and Smith (1978) is

$$d_p = (d_c^2/2 + d_c \cdot h_c)^{0.5}, \tag{8}$$

where d_c and h_c are the diameter and length of cylindrical extrudates.

The maximum value of r_i obtained in our experimental study using Eq. 8 is 21.6, while Herskowitz and Smith (1978) have reported that their maximum r_i value is 14.65. The comparison of estimated flow in the last annulus of the collector (this includes wall flow) using the values of S and f as reported by Herskowitz and Smith (1978) and the experimentally observed flow for the air–water system is shown in Figure 12. It can be seen that the model drastically underpredicts liquid flow near the wall. In addition, the dependence of flow in the outermost annulus on liquid and gas flow rates, as experimentally observed, is not truly represented by the constant values of the model parameters S and f. Another important observation is that the values of the model parameters estimated by Herskowitz and Smith (1978) for cylindrical extrudates are 1. These are the extreme values and represent the maximum possible radial flow for a given packed-bed

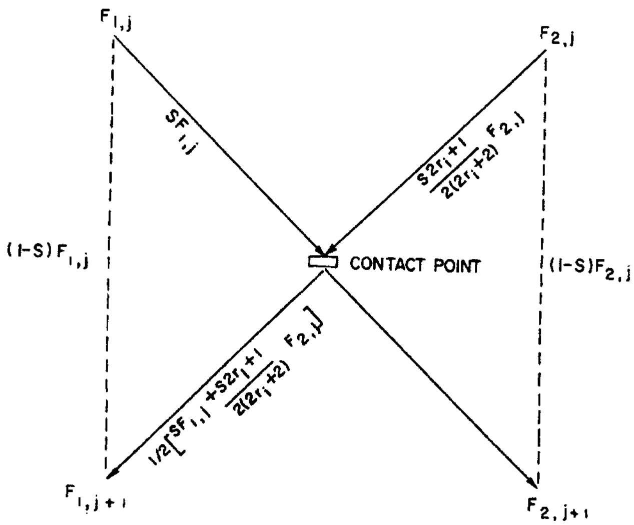

Figure 10. Flow network at the center of the column.

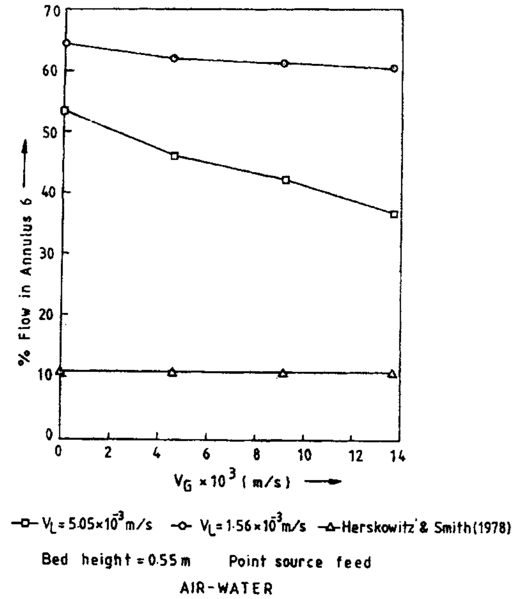

Figure 12. Comparison of flow in annulus 6 with Herskowitz and Smith (1978) model for air–water system.

configuration. Even reestimation of these model parameters with the present experimental results showed the values to be close to 1. Further attempts to use the preceding model with characteristic particle diameter as a model parameter did not lead to any acceptable parameter values. The unrealistic values of S and f clearly indicated that there is a range within which the model can be modified so it can better describe the liquid flow. This is of particular significance for the large D/d_p ratio and cylindrical extrudates used in the present study.

The procedure for packing the column is reported to affect the liquid distribution (Herskowitz and Smith, 1978; Al-Dahhan et al., 1995). The bed used in the present study was packed after filling it with water, gradually dropping the extrudates, and vibrating the column at frequent intervals. Careful observation of the bed revealed that this method of packing the bed generally orients the length of the cylindrical extrudates horizontally. This means that the same characteristic length cannot be used to estimate the number of the annulus and layers needed to represent the packed bed by the preceding mechanistic model. In view of this observation the number of vertical layers in the bed was estimated using

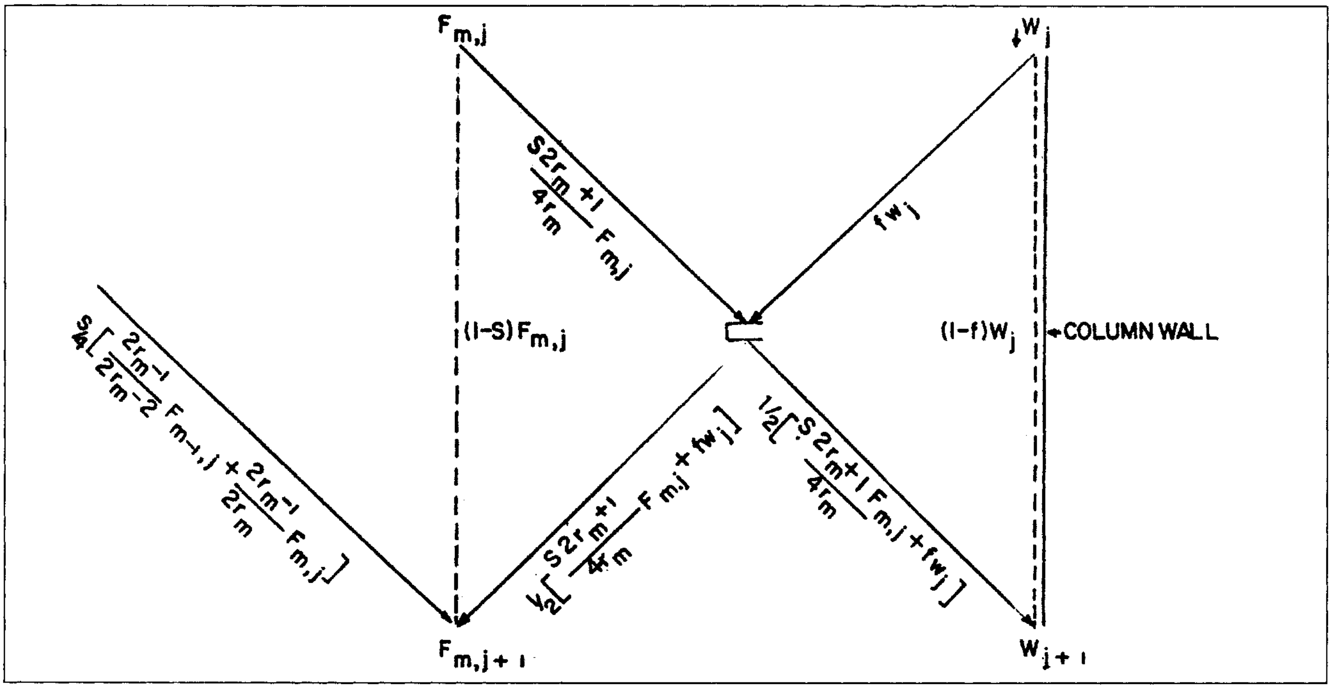

Figure 11. Flow network near column wall.

the extrudate diameter and the number of the annulus was estimated by a characteristic orientation of extrudate along the radial direction. Therefore the particle characteristic dimension in the axial direction is the extrudate diameter (d_c), while in the radial direction it can be given by the extrudate diameter multiplied by the orientation parameter, or

$$d_{p,\text{axial}} = d_c \tag{9}$$

$$d_{p,\text{radial}} = \phi \cdot d_c, \tag{10}$$

where ϕ represents the orientation of the particle with the radial direction. With these modifications in the effective particle size, Eqs. 1 and 2 can be modified as

$$m = D/2d_{p,\text{radial}} \tag{11}$$

$$n = H/d_{p,\text{axial}}. \tag{12}$$

These definitions of the characteristic particle dimensions in radial and axial direction drastically improved the liquid distribution estimates, although they required an additional model parameter ϕ, the particle orientation. Interestingly, the value of this parameter was found to be constant for all the runs conforming with the fixed-bed configuration.

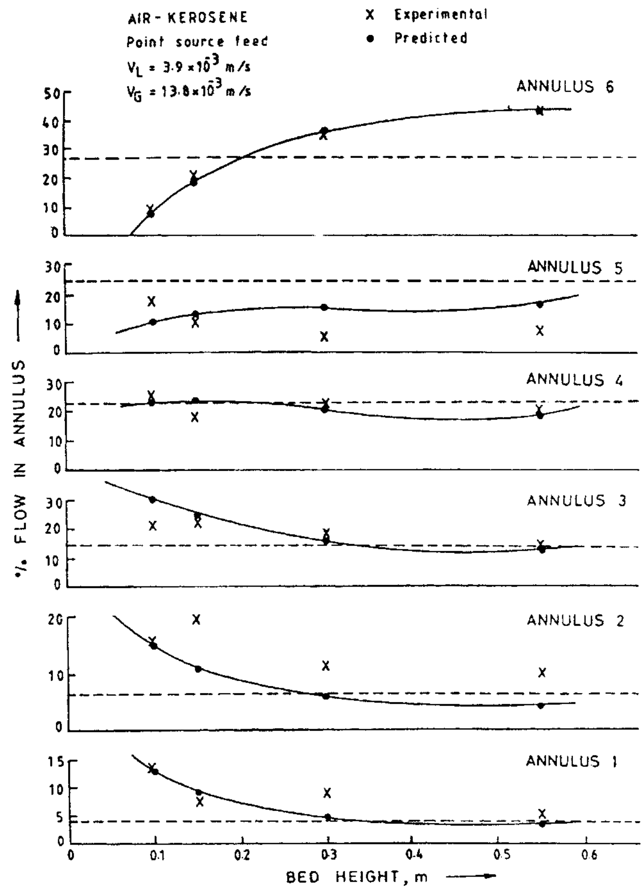

Figure 13. Comparison of experimental and predicted liquid distribution for air–kerosene system with point source feed.
$V_L = 3.9\times10^{-3}$ m/s; $V_G = 13.8\times10^{-3}$ m/s.

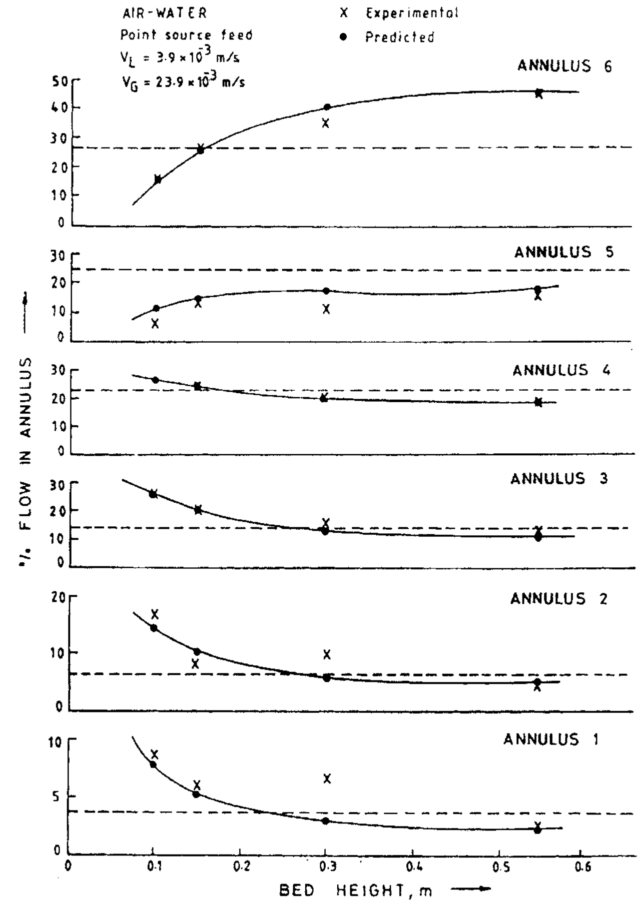

Figure 14. Comparison of experimental and predicted liquid distribution for air–water system with point source feed.
$V_L = 3.9\times10^{-3}$ m/s; $V_G = 23.9\times10^{-3}$ m/s.

To estimate model parameters, experiments were performed with point-source feed at four different bed heights. Figures 13 and 14 present the distribution rate for air–kerosene and air–water systems, respectively, in the annulus for fixed gas and liquid velocities. Model predictions using estimated parameter values are also included in these figures. A significant improvement in the liquid distribution estimates is evident once the particle orientation is considered.

The model parameters S and f were correlated in terms of liquid velocity, gas velocity, and liquid properties. Surface tension was found to give better correlation for these model parameters as compared to density and viscosity. Inclusion of these properties along with surface tension in the correlations hardly improved the fit. Correlations of S and f are as follows:

$$S = 0.17 + 9.7\sigma\ (\text{N/m}) + 36.1V_L\ (\text{m/s}) - 12.4V_G\ (\text{m/s})$$
$$(\text{Average Error} < 10\%) \tag{13}$$

$$f = 0.07 + 1.03\sigma\ (\text{N/m}) + 59.71V_L\ (\text{m/s}) + 1.17V_G\ (\text{m/s})$$
$$(\text{Average Error} < 10\%). \tag{14}$$

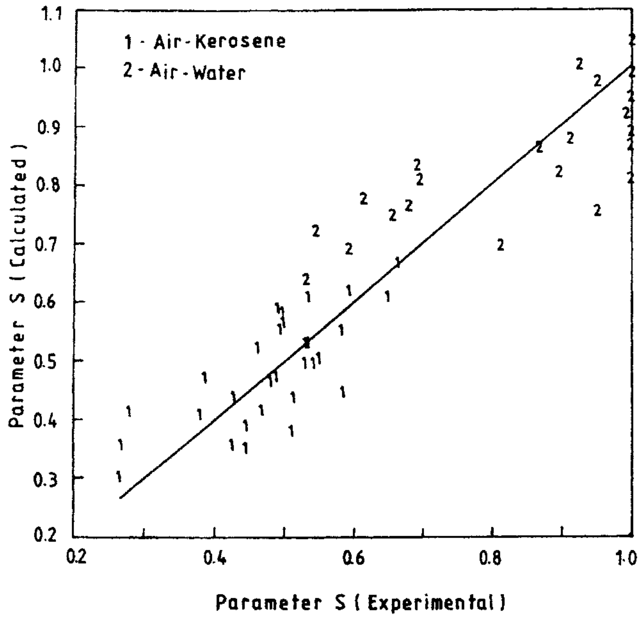

Figure 15. Parity plot for parameter *S*.

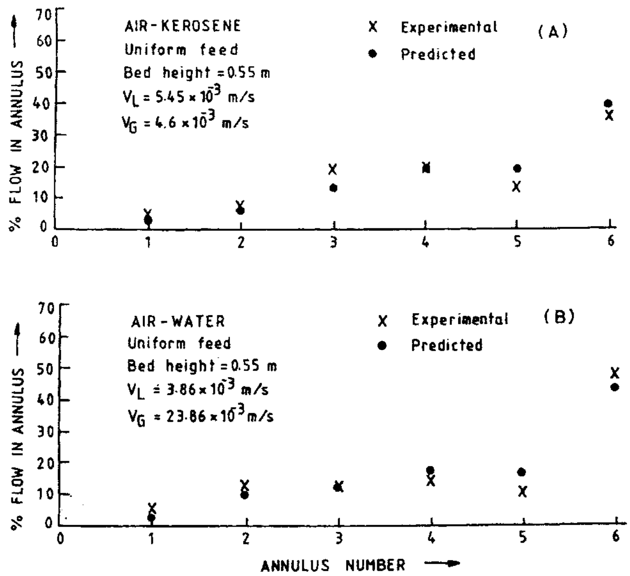

Figure 17. Comparison of experimental and predicted liquid distribution for uniform distribution.

(a) Air–kerosene system; (b) air–water system.

Parity plots between the experimental and calculated values of S and f are plotted in Figures 15 and 16, respectively. The third model parameter used in the model—orientation parameter, ϕ—was always found to have a value very close to five. Interestingly, this observation conforms with a fixed-bed configuration that suggests a characteristic particle length in the axial direction of 7.5 mm. As can be seen in Figure 2, this value represents the average extrudate length. Thus the values of parameters obtained after the model modifications are realistic. Prediction of radial flow distribution for uniform distribution at the top of the bed using parameter values calculated by Eqs. 13 and 14 are compared with experimental results in Figure 17. Good agreement is evident from the figure.

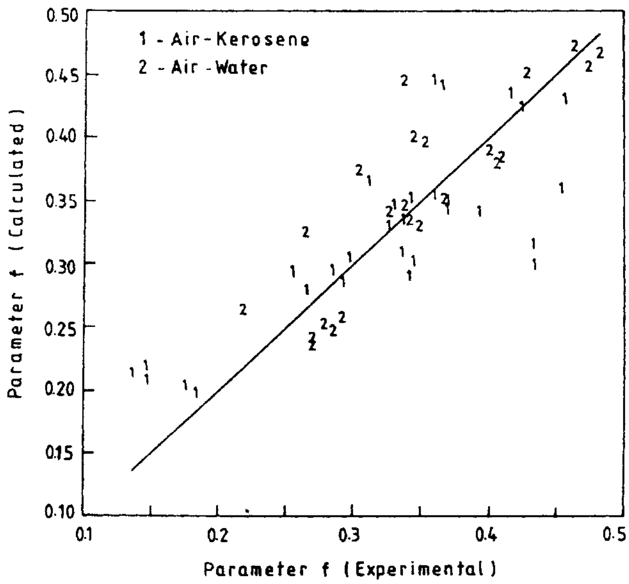

Figure 16. Parity plot for parameter *f*.

Conclusion

Experimental results on radial liquid distribution in a trickle-bed reactor are presented. Increase in the liquid and gas flow rates was found to reduce wall flow and improve liquid distribution in the column. An improved liquid distribution was observed for lower liquid surface tension and density.

The mechanistic model proposed by Herskowitz and Smith (1978) for liquid distribution was found not to be applicable for a column-to-particle-diameter ratio (D/d_p) of more than 30. The model was modified to incorporate the effect of particle orientation, and the model parameters were correlated to liquid and gas flow rates as well as liquid surface tension. With the proposed modifications the model described the liquid distribution fairly well, even for a high D/d_p ratio with realistic parameter values.

Notation

d_p = equivalent particle diameter, m
D = diameter of column, m
G = gas superficial mass velocity, kg/m^2·s
i = number of particles measured radially from the center of the bed
H = height of catalyst bed, m
L = liquid superficial mass velocity, kg/m^2·s
R = radial distance from the center of the column, m
R_0 = radius of column, m
V_G = gas velocity, m/s
V_L = liquid velocity, m/s
W = flow rate at the wall, m^3/s
σ = surface tension, N/m

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