# Mass Transport Phenomena and Hydrodynamics in Packed Beds with Gas-Liquid Concurrent Upflow


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Several experimental comparisons on multiphase, fixed-bed catalytic reactors operated in the upflow vs. downflow mode for hydrodesulfurization of crude or heavy oil by Ohtsuka et al. (1968), Takematsu and Parsons (1972), Montagna and Shah (1975), and Ohshima et al. (1976), and for selective hydrogenation of phenylacetylene in styrene solution by Mochizuki and Matsui (1976), showed superior performance with the upflow mode (higher conversion, better selectivity, longer catalyst life, and better temperature control). The studies, however, on the liquid-solid mass transfer rates in fixed beds with gas-liquid concurrent upflow have started only very recently.

In this note, studying the recent research results on the multiphase fixed beds, an attempt was made to express the mass transfer rates in an unified form, and some interesting correlations between the transport and hydrodynamic phenomena were pointed out.

## EFFECTS OF PARTICLE DIAMETER

In the previous paper (1974), liquid-solid mass transfer coefficients measured by the diffusion current method at low liquid flow rates depend only slightly on liquid velocity. Based on hydraulic diameter, the correlation is

$$Sh = 48 Re_G^{1/4} \tag{1}$$

At high liquid flow rates, the Sherwood number depends on gas and liquid flow rate according to

$$Sh/Sh_o = 1 + 4 Re_G^{0.55}/Re_L^{0.70} \tag{2}$$

where

$$Sh_o = 0.75\, Re_L^{1/2}\, Sc_L^{1/3}$$

is the corresponding Sherwood number for single phase flow.

Those relations, however, are valid only for particular particle size (5 × 5 mm cylinder). No study has been reported on the effects of particle diameter on the mass transfer rates. Very recently, Sato et al. (1976) measured the mass transfer coefficients of benzoic acid and copper oxide-zinc oxide particles in water-air concurrent upflow systems (particle diameters were from 3.2 to 12 mm sphere), and they found the Sherwood number not only depended on gas and liquid flow velocities but also depended on particle diameters. In gas-liquid multiphase flow systems, it seems quite natural tnat particle diameter or void volume in the packed beds would play a very important role in governing the gas bubble movement and eventually would define the transfer rates. Now there are several reports which are useful for analyzing the particle size effects on the mass transfer: Nosaka et al. (1976) (mass transfer from benzoic acid particle to water) which is shown in Figure 1, and Sato et al. (1976) (from single sphere to water) shown in Figure 2. These two results are quite similar to our previous work (1974).



## HIGH REYNOLDS NUMBER

First, consider the effects of particle diameter on the mass transfer in the regime with the liquid Reynolds number greater than the critical Reynolds number $Re_{LC}$.

Assuming Equation (2) is also valid for the observed data in Figure 1, except the constant of the second term of the right side of Equation (2), the observed data were rearranged to find this constant which is denoted Y:

$$Y = (Sh/Sh_o - 1)/(Re_G^{0.55}/Re_L^{0.70}) \tag{3}$$

The calculated results are shown in Table 1. It can be concluded that Y is a constant, and the average value is 8.25. The same procedure was done on the measured value in a smaller particle bed (Goto et al., 1975). In the small particle regime, little effect of particle diameter on the Sherwood number was observed. Probably the gas bubbles move and distribute ununiformly in the particle beds. Thus, the largest particle diameter of their study (2.4 mm) was chosen for the analysis. The calculated results are shown in Table 2.

Based on the calculation, it is found that Y is proportional to the 1.2 power of particle diameter as

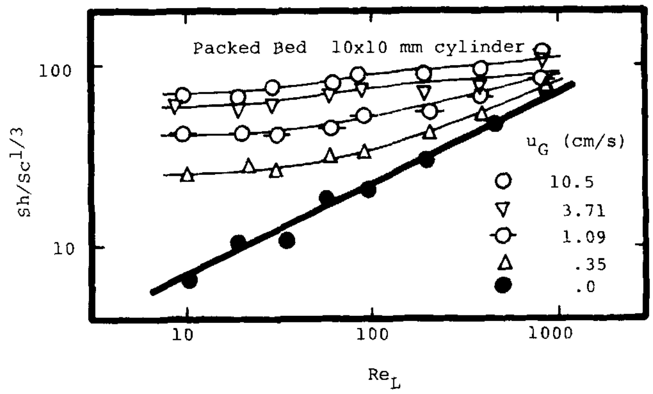


Fig. 1. Particle-liquid mass transfer in packed beds (Nosaka et al., 1976).

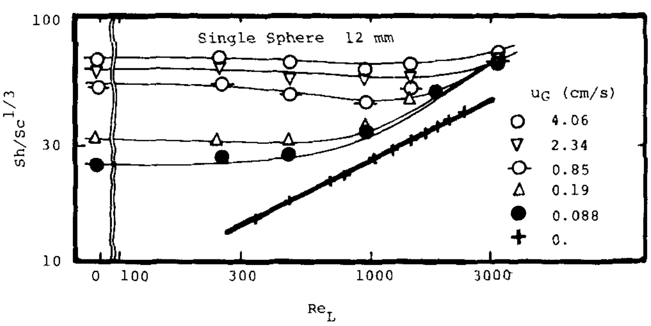


Fig. 2. Single sphere to liquid mass transfer in a bubble column (Sato et al., 1976).

TABLE 1. CALCULATION FOR Y, 10 × 10MM CYLINDER

| No. | $Re_L$ | $Re_G$ | $Sh/Sc^{1/3}$ | $Sh/Sh_o$ | Y |
|---|---|---|---|---|---|
| 1 | 90 | 2.54 | 32 | 1.524 | 7.31 |
| 2 | 90 | 7.92 | 53 | 2.52 | 11.35 |
| 3 | 90 | 27.0 | 71 | 3.38 | 8.90 |
| 4 | 90 | 76.3 | 90 | 4.29 | 7.06 |
| 5 | 200 | 2.54 | 42 | 1.31 | 7.65 |
| 6 | 200 | 7.92 | 54 | 1.69 | 9.00 |
| 7 | 200 | 27.0 | 70 | 2.19 | 7.45 |
| 8 | 200 | 76.3 | 90 | 2.81 | 6.82 |
| 9 | 400 | 2.54 | 52 | 1.16 | 6.19 |
| 10 | 400 | 7.92 | 66 | 1.47 | 9.92 |
| 11 | 400 | 27.0 | 76 | 1.69 | 7.46 |
| 12 | 400 | 76.3 | 95 | 2.11 | 6.79 |
| 13 | 800 | 2.54 | 70 | 1.08 | 4.96 |
| 14 | 800 | 7.92 | 85 | 1.31 | 10.62 |
| 15 | 800 | 27.0 | 110 | 1.69 | 12.18 |
| 16 | 800 | 76.3 | 120 | 1.85 | 8.38 |
| | | | | Average | 8.25 |

$$Y = 7.45\ d_{pe}^{1.2}$$

where $d_{pe}$ is surface equivalent particle diameter expressed in centimeters. The generalized correlation of the mass transfer coefficients between particle liquid is

$$Sh/Sh_o = 1 + 7.45\ d_{pe}^{1.2}\ Re_G^{0.55}/Re_L^{0.70} \quad (4)$$

Applying the same procedure to the observed data on single sphere liquid (Figure 2) and on wall liquid in a gas bubble column (Figure 3, Miyashita et al., 1975), a similar formula to Equation (4) was obtained for both systems:

$$Sh/Sh_o = 1 + 65 Re_G^{0.30}/Re_L^{0.70} \quad (5)$$

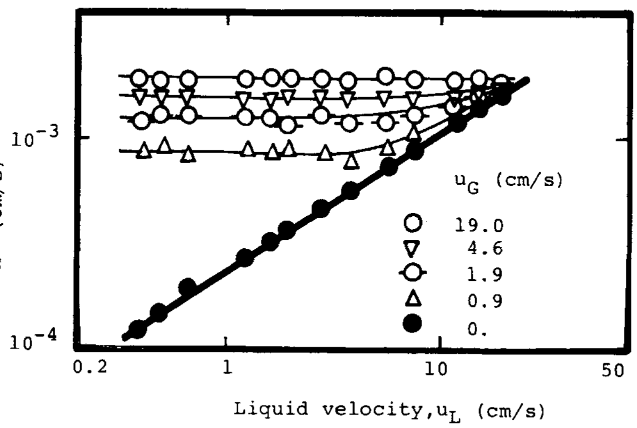


**Fig. 3. Wall-liquid mass transfer in a bubble column (Miyashita et al., 1975).**

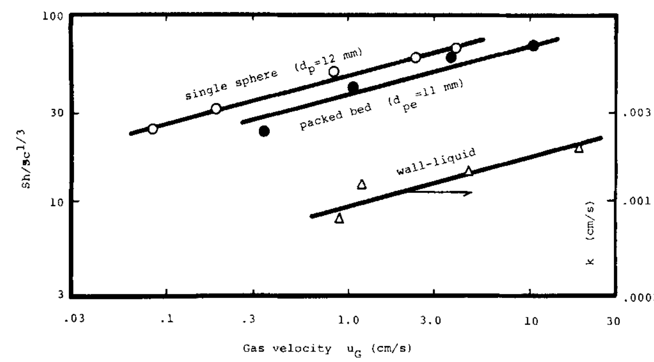


**Fig. 4. Effect of gas velocity on the mass transfer coefficient.**

TABLE 2. CALCULATION FOR Y, $d_p$ = 2.41MM SPHERE

| No. | $Re_L$ | $Re_G$ | $Sh/Sh_o$ | Y |
|---|---|---|---|---|
| 1 | 10 | 1.23 | 1.33 | 1.36 |
| 2 | 10 | .61 | 1.22 | 1.47 |
| 3 | 10 | .31 | 1.16 | 1.56 |
| | | | Average | 1.47 |

## LOW REYNOLDS NUMBER

For the regime with $Re_L$ less than $Re_{LC}$, the mass transfer rates are governed by only gas bubble movement as Equation (1). Similar correlations to Equation (1) were obtained for packed bed, single sphere, and bubble column systems as shown in Figure 4 and Table 3. In this connection, it is of particular interest to study the wall-liquid heat transfer properties in bubble columns in relation to the mass transfer phenomena in packed beds with two phase flow.

Having studied several works (Fair et al., 1962, and others) and their own experiments, Nagata et al. (1975) found that the wall-liquid heat transfer coefficients in bubble columns were proportional to one-fourth power of superficial gas velocity. In this connection, Calderbank (1967) suggested that wall-liquid heat and mass transfer coefficients in bubble columns would be correlated with one-fourth power of the energy dissipation, which had been established in aerated stirring vessels derived by Kolmogoroff's theory of isotropic turbulence (Calderbank and Moo-Young, 1961). Accordingly, for mass transfer

$$k \propto [(P/\rho V)\nu]^{1/4}/Sc^{2/3}$$

for heat transfer

$$h/\rho C_p \propto [(P/\rho V)\nu]^{1/4}/Pr^{2/3}$$

where, according to Nagata et al. (1975), aeration power $Pa$ in bubble column is given by

$$P_a = u_G \rho V$$

Thus, the aeration power dissipation by the liquid phase per unit mass of liquid is

$$P_a/\rho V = u_G$$

As a result of the above considerations and with Equation (1) and Figure 4, it may be concluded that the solid-liquid heat and mass transfer coefficients in packed beds with concurrent two phase upflow or in bubble columns

TABLE 3. UNIFIED EXPRESSIONS

| $Re_L \; Re_{LC}$ | | | $Re_{LC}$ | Note |
|---|---|---|---|---|
| Packed bed | $Sh = 48\ Re_G^{1/4}$ | $(d_{pe} = 6\ \text{mm})$ | 10 | $\left.\right\}$ $Sh = 300 \exp(-.062/V_\epsilon) Re_G^{1/4}$ |
| Packed bed | $Sh = 250\ Re_G^{1/4}$ | $(d_{pe} = 11\ \text{mm})$ | 90 | |
| Single sphere | $Sh = 300\ Re_G^{1/4}$ | $(d_p = 12\ \text{mm})$ | 500 | |
| Wall-liquid | $k = .001\ u_G^{1/4}$ | $(d_T = 62\ \text{mm})$ | 1600 | Bubble column |
| Wall-liquid | $h = 2800\ u_G^{1/4}$ | | | Heat transfer in bubble column (Nagata et al., 1975) |
| $Re_L \; Re_{LC}$ | | | | |
| Packed bed | $Sh/Sh_o = 1 + 7.45\ d_{pe}^{1.2}\ Re_G^{.55}/Re_L^{.70}$ | | | |
| Single sphere | $Sh/Sh_o = 1 + 65\ Re_G^{.3}/Re_L^{.7}$ | | | |
| Wall-liquid | $Sh/Sh_o = 1 + 65\ Re_G^{.3}/Re_L^{.7}$ | | | |

are correlated with the energy dissipation by the liquid phase per unit mass of liquid or one-fourth power of superficial gas velocity $u_G$. With low $Re_L$, $P/\rho V$ will be almost equal to $P_a/\rho V$.

## OPTIMAL PARTICLE DIAMETER

From a practical point of view, it is very interesting to find the optimal particle diameter which will give the maximum volumetric mass transfer coefficient in the packed beds. The gas movement will be restricted by the size of void volume in the packed beds; then the mass transfer coefficients in packed beds will be correlated with the void volume of the beds. Based on the results of mass transfer correlations in Table 3, a trial plot of $Sh/Re_G^{1/4}$ vs. $1/V_\epsilon$ is shown in Figure 5, where $V_\epsilon$ is the average void volume defined as

$$V_\epsilon = (\text{average particle volume}) \times (\text{void fraction of the bed})$$

expressed in milliliters, and $V_\epsilon = \infty$ for single sphere in a bubble column system. The effect of the void volume on the mass transfer rate is expressed in the correction term of $\exp(-0.062/V_\epsilon)$. Therefore, the general expression for the packed beds with two phase flow is

$$Sh = 300 \exp(-0.062/V_\epsilon)\ Re_G^{1/4} \tag{6}$$

Using this correlation, the optimal particle diameter which gives the maximum volumetric particle-liquid mass transfer coefficient can be derived. If the overall process is a mixed process of liquid-solid mass transfer and surface reaction, the optimal diameter becomes smaller. These relations are shown in Figure 6 with various $k/k_r$.

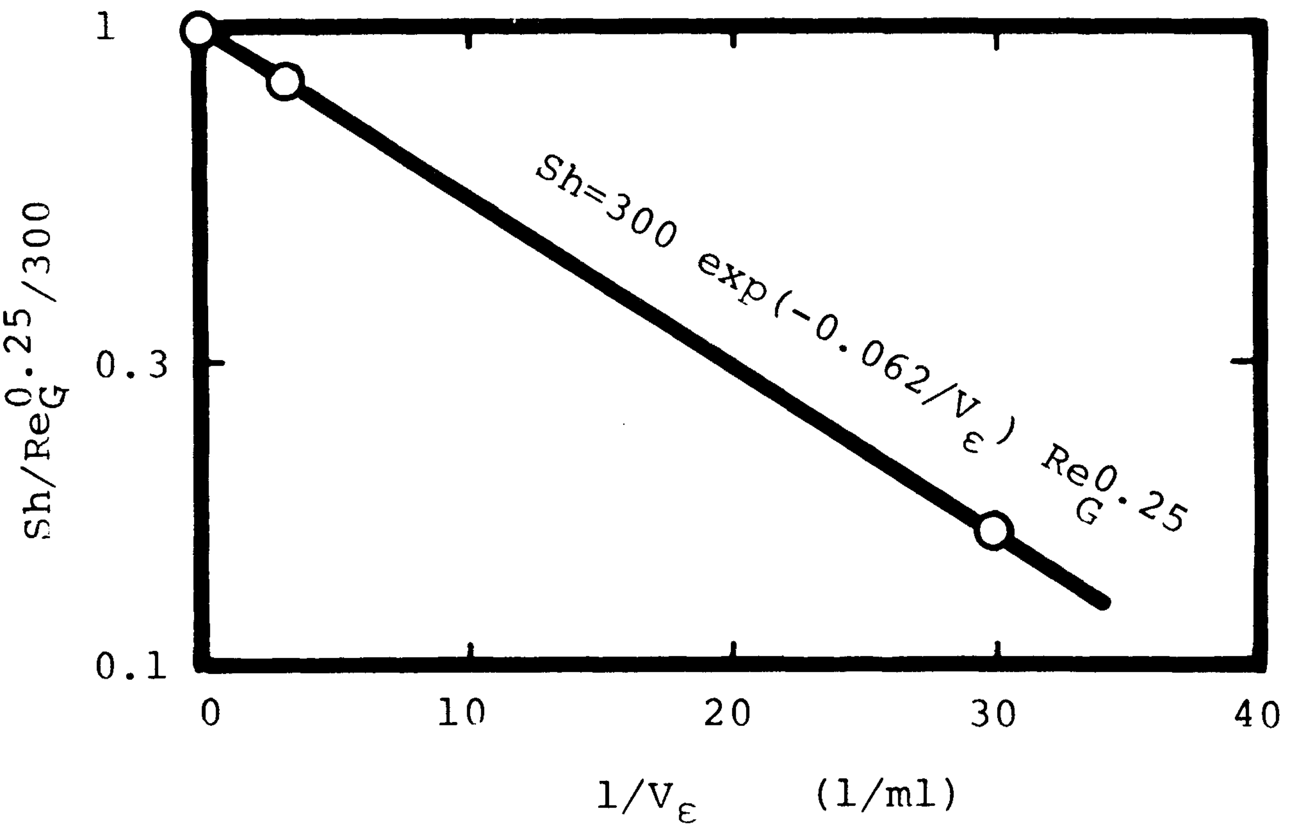


Fig. 5. Effect of void volume on the mass transfer.

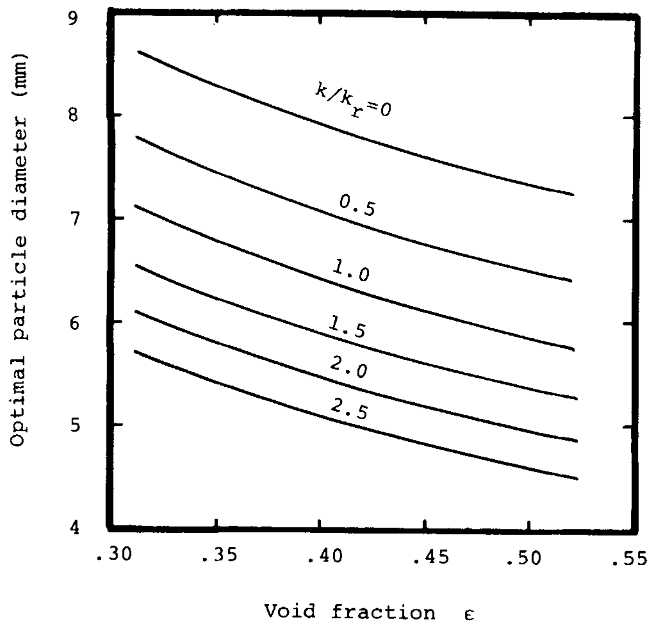


**Fig. 6. Optimal particle diameter for maximum overall volumetric solid-liquid transfer coefficient with first order surface reaction.**

## CRITICAL REYNOLDS NUMBER

Inspecting the critical Reynolds number $Re_{LC}$ in Table 3, it seems to correspond to the transient Reynolds number in a single phase flow, such that for the bubble column, $Re_{LC}$ is about 1 600, which corresponds to the transient Reynolds number from laminar flow to turbulent flow in a pipe, and for a single sphere system, this $Re_{LC}$ is about 500, which corresponds to the transient regime from Stoke's law range to Newton's law range for a sphere in moving fluid, where the first separation of the boundary layer occurs at a point just forward of the equatorial plane. Knowing these relations, it may be said that $Re_{LC}$ expresses the transient regime of boundary layer on the liquid around the packed particles. Thus, the hydrodynamic conditions in packed beds could be estimated by measuring the mass transfer coefficient between solid liquid in the fixed beds.

## DILUTED BEDS AND ALL ACTIVE BEDS

Finally, some consideration will be given to the relation between the mass transfer rates in diluted beds and in all active particle beds. Since the data were obtained by using diffusion current method in the system of an active particle in an inert bed, the concentration field around the single particles is probably different than it would be if all particles were active. On this subject, Miyauchi (1975) studied and concluded that in the regime with film thickness less than the average void thickness to adjacent active particles, the mutual interaction between them could be ignored. For a single fluid system, this regime is defined with $Pe_h = ud_h/\epsilon D$ greater than 200. For gas-liquid systems, strong turbulence caused by gas bubbles results in very thin film thicknesses; therefore, film coefficients of mass transfer between solid liquid in the diluted packed beds should be little different from those in all active particle beds. Thus, as discussed in this note, the results with diffusion current method have been well correlated with other studies in all active particle beds, and also it was verified by successful application to analyzing a chemical reaction system (Mochizuki and Matsui, 1976).

## NOTATION

$a$ = specific surface area of a packed bed
$D$ = molecular diffusivity
$d_h$ = $\epsilon d_p/[1.5(1-\epsilon)]$, effective particle diameter
$d_p$ = particle diameter
$d_{pe}$ = surface equivalent particle diameter, cm
$k$ = solid-liquid mass transfer coefficient
$k_r$ = first-order surface chemical reaction rate constant
$P$ = agitation power
$P_a$ = power by aeration
$P_r$ = Prandtl number
$Re$ = $ud_h/\epsilon\nu$, Reynolds number, $L$ for liquid, $G$ for gas
$Re_{LC}$ = critical liquid Reynolds number
$Sc$ = $\nu/D$, Schmidt number
$Sh$ = $kd_h/D$, Sherwood number
$Sh_o$ = for liquid-full condition
$u$ = mean fluid velocity
$V$ = liquid volume
$V_\epsilon$ = void volume, ml
$Y$ = defined in Equation (3)
$\epsilon$ = void fraction of a packed bed
$\mu$ = viscosity
$\rho$ = density
$\nu$ = kinematic viscosity

## LITERATURE CITED


Calderbank, P. H., and M. B. Moo-Young, "The Continuous Phase Heat and Mass Transfer Properties of Dispersion," *Chem. Eng. Sci.*, **16**, 39 (1961).
Calderbank, P. H., in *Mixing*, V. W. Uhl and J. B. Gray, ed., 1 ed., Vol. 2, pp. 76-81, Academic Press, N.Y. (1967).
Fair, J. R., A. J. Lamdright, and J. W. Andersen, "Heat Transfer and Gas Holdup in a Sparged Contactor," *Ind. Eng. Chem. Process Design Develop.*, 1, 33 (1962).
Goto, S., J. Levec, and J. M. Smith, "Mass Transfer in Packed Beds with Two-Phase Flow," *ibid.*, **14**, 473 (1975).
Miyashita, H., K. Kita, M. Sugata, and H. Masuzaki, "Bubbling Effect on Transfer Coefficients," Proceedings of 9th Fall Meeting of Soc. Chem. Engrs., Japan, A-307 (1975).
Miyauchi, T., "Film Coefficient of Mass Transfer in Packed Beds," *J. Fac. Eng. Univ. Tokyo*, **A-13**, 72 (1975).
Mochizuki, S., and T. Matsui, "Liquid-Solid Mass Transfer Rate in Liquid-Gas Upward Cocurrent Flow in Packed Beds," *Chem. Eng. Sci.*, **29**, 1328 (1974).
——————, "Selective Hydrogenation and Mass Transfer in a Fixed-Bed Catalytic Reactor with Gas-Liquid Concurrent Upflow," *AIChE J.*, **22**, 904 (1976).
Montagna, A., and Y. T. Shah, "Backmixing Effect in an Upflow Co-current Hydrodesulfurization Reactor," *Chem. Eng. J.*, **10**, 99 (1975).
Nagata, S., M. Nishikawa, M. Itaya, and K. Ashiwake, "Study of Heat Transfer for Aerated Mixing Vessel and Aerated Tower," *Kagaku Kogaku Ronbunshu*, 1, 460 (1975).
Nosaka, M., H. Kawasaki, and H. Tanaka, "Solid-Liquid Mass Transfer in Gas-Liquid Concurrent Flow," Proceeding of Hokuriku Meeting of Soc. Chem. Engrs., Japan, A-10 (1976).
Ohshima, T., T. Takematsu, et al., "Liquid-Phase Mass Transfer Coefficient and Gas Holdup in a Packed-Bed Cocurrent Upflow Column," *J. Chem. Eng., Japan*, 9, 29 (1976).
Ohtsuka, T., M. Koizumi, et al., "Hydrodesulfurization of Khafji Crude Oil, Part V, On the Effect of Flow Directions in a Fixed Catalyst Bed," *Rept. 6205*, Annual Meeting of Chem. Soc., Japan (1968).
Sato, Y., T. Hirose, et al., "Liquid-Particle Mass Transfer in Fixed Beds with Gas-Liquid Cocurrent Upflow," Proceeding of 10th Fall Meeting of Soc. Chem. Engrs., Japan, A-109 (1976).
Takematsu, T., and B. I. Parsons, "A Comparison of Bottom-Feed and Top-Feed Reaction Systems for Hydrodesulfurization," *Fuels Res. Centre Tech. Bull., Ottawa, TB 161* (1972).



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