Liquid Holdup and Dispersion in Trickle-Bed Reactors

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Knowledge of liquid holdup and dispersion in trickle beds is of importance in proper modeling and design of these reactors as well as in interpreting pilot plant data and in the scaling up of such data. Satterfield (1975) summarized a number of holdup studies for liquid flows 0 to 14.7 kg/m²s. Some of the available correlations for liquid holdup for flow rates typically encountered in trickle-bed reactors ($0.1 < L < 10$ kg/m²s; $10^{-4} < G < 10^{-1}$ kg/m²s) are presented in Table 1. Most investigators have attempted to correlate holdup with liquid superficial mass velocity, particle Reynolds number, or single phase friction losses. The system used predominantly has been air-water and glass beads ($0.3 < d_p < 1$ cm). Large nonporous packing ($0.3 < d_p < 5.08$ cm) in the form of Raschig rings, Berl saddles, and Lessig rings, and some hydrocarbons as the liquid phase, have also been used. Charpentier et al. (1971) determined holdups in beds packed with porous catalyst particles ($0.3 < d_p < 0.5$ cm). Charpentier and Favier (1975) reported holdups in a variety of gas-liquid systems with beds packed with different types of spherical and cylindrical catalyst packing ($d_p > 0.3$ cm). Recently, Goto and Smith (1975) reported holdup data in beds packed with smaller packing ($0.051 < d_p < 0.41$ cm).

The typical size of porous catalyst particles in trickle-bed reactors is between 0.08 cm and 0.32 cm (Satterfield, 1975). This indicates that the available correlations, all of which have been based on data obtained with nonporous and porous particles larger than 0.3 cm, have been tested only at the upper limit of the particle size range of interest in trickle-bed reactors.

Numerous models have been proposed to account for liquid backmixing in trickle-bed reactors. A summary of these is presented by Schwartz and Roberts (1973). They conclude that the one parameter axial dispersion model is adequate to account for the effect of liquid backmixing on conversion. To employ this model, or to estimate the importance of liquid backmixing from the relationships developed by Burghardt and Zaleski (1968) and Mears (1971), it is necessary to know the value of the dispersion number, that is, the inverse of the particle Peclet number (Bodenstein number). Some correlations for the Peclet number based on experimental studies with packed beds with trickle flow have been developed (Sater and Levenspiel, 1966; Hochman and Effron, 1969; Michell and Furzer, 1972). These correlations are presented in Table 2. Particle size used was much larger, and shapes of particles were different than the ones of interest in trickle-bed reactors.

The purpose of this note is to present recently obtained results on liquid holdup and dispersion in a trickle bed (1.35 cm I.D., 30.5 cm height) packed with either nonporous or porous alumina of equal size ($d_p = 0.06$ cm) and to compare them with data of other investigators and the predictions of various correlations.

The small irregular particles used are at the lower end of the size range currently employed in the petroleum industry in trickle beds, but even smaller sizes may become important in other processes (Kittrell and Kamionski, 1976). The data presented here were obtained by means of a new, two-tracer technique for simultaneous determination of liquid holdup and contacting efficiency. The liquid was hexane and the gas was helium, both fed concurrently from the top of the column. Details of the experimental procedure are presented elsewhere (Schwartz et al., 1976).

Liquid holdup is obtained from the first moment of the exit age distribution curve determined by use of the nonadsorbable tracer (heptane):

$$\bar{t}_L = t_{\text{heptane}} = \frac{H_T V}{Q_L} \tag{1}$$

The bed axial Peclet number is calculated from the variance of the residence time distribution of the nonadsorbable tracer:

$$\frac{\Delta\sigma^2}{\bar{t}_L^2} = \frac{2}{Pe} + \frac{1}{Pe^2}\{e^{-Pe}[e^{-Pe} + 4(1 + Pe)] - 5\} \tag{2}$$

This equation is a special case of the one developed by Bischoff (1960). Only variances of the residence time distributions obtained with nonporous packing are used. For beds with porous packing, the variance depends both on bed dispersion and internal diffusion into the porous pellets. This makes it impossible to extract the information on bed dispersion from the variance alone.

Holdup and dispersion data were obtained for liquid flow rates from 0.3 to 5 kg/m²s. Gas flow rates in the range from 10^{-4} to 2×10^{-3} kg/m²s had negligible effects on holdup and dispersion.

LIQUID HOLDUP

The agreement between volumetrically and tracer determined holdups was very good and is discussed elsewhere (Schwartz et al., 1976). Only total holdup can be determined by the tracer method, whereas the volumetric technique was used to determine dynamic and static holdups also.

Dynamic liquid holdup for nonporous packing is plotted as a function of Reynolds number in Figure 1. It is compared with some other data (Hochman and Effron, 1969; Ross, 1965; Mohunta and Laddha, 1965) on the same figure. It is apparent from this comparison that a simple correlation of holdup vs. Reynolds number alone is not obtainable. Total liquid holdups obtained on porous packing in our study and by Ross (1965) are plotted vs. liquid mass velocity in Figure 2.

The holdup level and its dependence on liquid mass velocity or Reynolds number varies from study to study. The lower dependence of holdup on liquid mass velocity in our study can be ascribed to the higher level of holdup obtained which seems to be characteristic for small particles (Goto and Smith, 1975).

Figure 3 shows the comparison of our data for nonporous packing with the data of Sater and Levenspiel (1966), Hochman and Effron (1969), and the laboratory reactor data of Ross (1965) when plotted according to the Otake and Okada correlation (1963). Our data, which were obtained at lower Reynolds numbers than those of other investigators, cannot be fitted by the correlation.

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TABLE 1. CORRELATIONS FOR LIQUID HOLDUP

Otake and Okada (1963): $H_D = 1.295\,(Re_L)^{0.676}\,(Ga_L)^{-0.44}\,(a_E e_p)$	6.4 to 22 mm spheres 12.7, 25.4 mm Raschig rings, Berl saddles
Davidson (1959): $H_E \propto (Re_f)^{0.30}\,(Ga_L)^{-1/3}\,(a_w d_p)$	Theoretical 10.3, 25.4, 50.8, mm Raschig and Lessing rings (Mitchell and Furzer 1972*a*)
Hochman and Effron (1969): $H_D = 0.00445\,(Re_L)^{0.76}$	4.8 mm glass spheres
Jesser and Elgin (1943): $H_E \propto L^{0.52 \text{ to } 0.67}$	12.7, +9.1, 25.4 mm glass spheres 25.4 mm Berl saddles 12.7 mm carbon rings
Way (1971): $H_E \propto L^{0.33}$	3 mm glass spheres, 3.2 × 3.2 mm cylinders
Larkins, White, and Jeffrey (1961): $\log_{10}\frac{H_E}{\epsilon} = -0.774 + 0.525\,(\log_{10}\chi) - 0.109\,(\log_{10}\chi)^2$ for $0.05 < \chi = \frac{\Delta P_L}{\Delta P_G} < 30$	3 mm spheres, 9.5 mm spheres, cylinders and Raschig rings
Sato, Hirose, Takahashi, and Toda (1973): $\frac{H_E}{\epsilon} = 0.40\,a_E^{1/3}\,\chi^{0.22}$ for $0.1 < \chi < 20$	2.6 to 24.3 mm glass spheres
Charpentier and Favier (1975): $\log\frac{H_E}{\epsilon} = -0.280 + 0.175\log\chi^1 - 0.047\,(\log\chi^1)^2$ for spherical packing $\log\frac{H_E}{\epsilon} = -0.363 + 0.168\log\chi^1 - 0.043\,(\log\chi^1)^2$ for cylindrical packing for $0.05 < \chi^1 = \frac{L/G}{(\Delta P_G/\rho_G g_c Z) + 1} < 100$	3 mm glass spheres, 1.8 × 6 and 1.4 × 5 mm cylinders

TABLE 2. CORRELATIONS FOR PARTICLE PECLET NUMBER IN TRICKLE FLOW

Sater and Levenspiel (1966):
$Pe_p = 7.58 \times 10^{-3}\,Re_L^{0.703}$
for: ½ in. (1.27 cm) Raschig rings and Berl saddles, countercurrent flow, $10 < \mathrm{Re_L} < 10^3$

Hochman and Effron (1969):
$Pe_p = 0.042\,Re_L^{0.5}$
for: 3/16 in. (0.48 cm) glass beads, concurrent flow, $4 < Re_L < 100$

Michell and Furzer (1972*a*, 1972*b*):
$Pe_p = 0.039\,Re_f^{0.50}$
for: 1 in. (2.54 cm) Lessing rings, countercurrent flow $50 < Re_f < 1\,000$
$Pe^1 = 1.00\,(Re^1)^{0.70}\,Ga_L^{-0.32}$
for: ¼ in. (0.64 cm), 2 in. (5.08 cm) Raschig rings, 1 in. (2.54 cm) Lessing rings, ¼ in. (0.64 cm) Berl saddles
$50 < Re^1 < 8\,000$

A modified form of Davidson's theoretical form (1959), as used by Michell and Furzer (1972*a*), which includes the effects of viscous and gravitational forces and of particle size and is primarily based on a laminar flow model over a string of spheres, was applied to our data as well as to data of other investigators for nonporous packing and is plotted in Figure 4. The data obtained in our study appeared to extend the trend of the data collected by others to the lower Reynolds number region. The somewhat lower slope of the correlation line obtained for the data in this study suggests an asymptotic approach to zero holdup at zero Reynolds number (subtracting static holdup).

The same form of the correlation was applied to the data for porous packing from our study and Ross' work. The data plotted in this manner in Figure 5 show the same

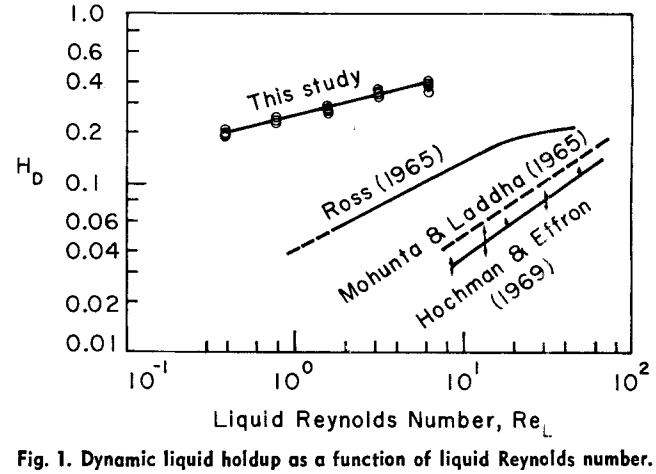

Fig. 1. Dynamic liquid holdup as a function of liquid Reynolds number.

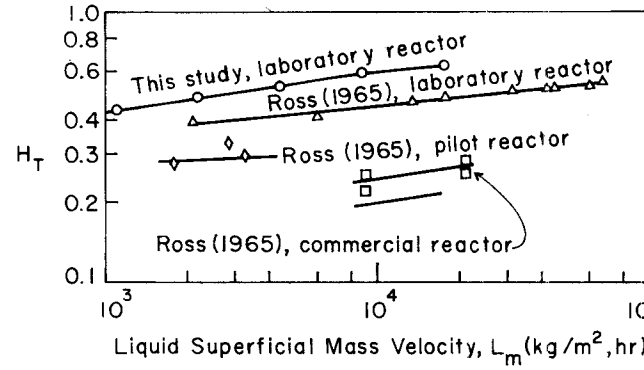

Fig. 2. Total liquid holdup on porous packing as a function of liquid superficial mass velocity.

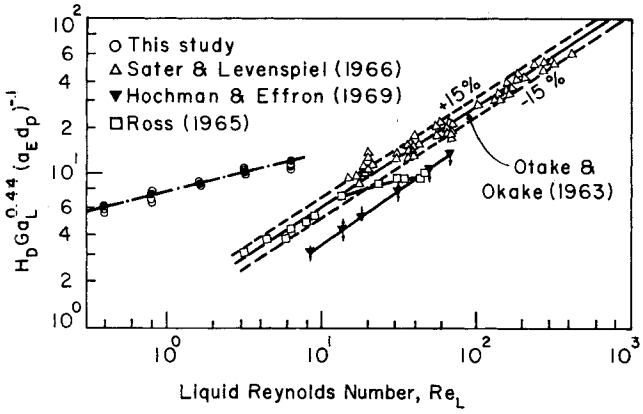

Fig. 3. Correlation of Otake and Okada (1963) applied to dynamic liquid holdup data on nonporous packing.

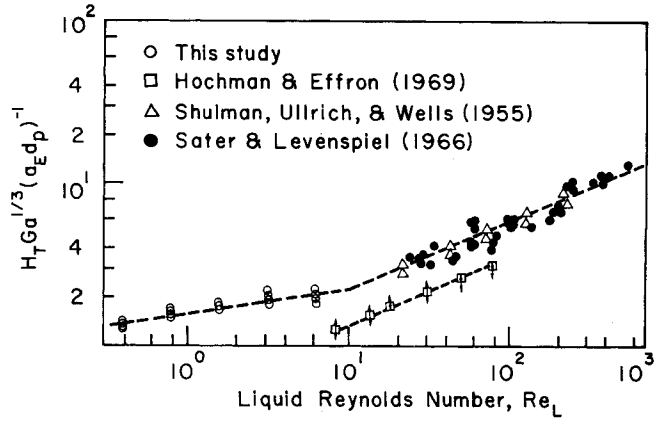

Fig. 4. Correlation of Davidson (1959) applied to total liquid holdup data on nonporous packing.

dependence on Reynolds number. The agreement of the data of our study with hexane and the data of Ross in pilot and commercial reactors with light gas oil was quite good, considering the large differences in particle size and shape, reactor size, and liquid properties.

The dynamic and total holdups from systems with varying dimensions, which were operated under a wide range of conditions, seem to follow the trend predicted by Davidson (1959). The range of variables and physical properties examined in the above cited studies are $0.4 < Re_L < 600$, 0.4 mm $< d_p <$ 17.7 mm, $2.6 < a_E d_p < 6.02$, $1.4 \times 10^1 < Ga_L < 3.2 \times 10^2$ for particle shapes consisting of Raschig rings, Berl saddles, spheres, irregular granules, and for concurrent and countercurrent flow.

The comparison of our holdup data for nonporous packing with the correlations of Larkins et al. (1961) and Sato et al. (1973) indicates that these correlations underestimate the holdup by more than 40%. The holdup values calculated from the recent correlation of Charpentier and Favier (1975) vary from 0.186 at the lowest liquid flow rate of 0.3 kg/m²s to 0.240 at the highest liquid flow rate of 5 kg/m²s. These values are from 25 to 40% lower than the experimentally determined ones. Similar discrepancies are found for holdups with porous packing.

It is apparent that, on the basis of studies to date, it is still not possible to predict holdup accurately in different trickle beds. The correlations developed for absorption towers with larger, differently shaped, predominantly nonporous packing cannot be extrapolated with certainty to trickle-bed conditions. Neither the correlations based on liquid superficial velocity nor those founded on friction losses can predict holdup accurately in trickle beds packed with small particles. Holdup levels in these beds seem to be considerably higher than holdups in absorption towers and are less dependent on liquid flow rate.

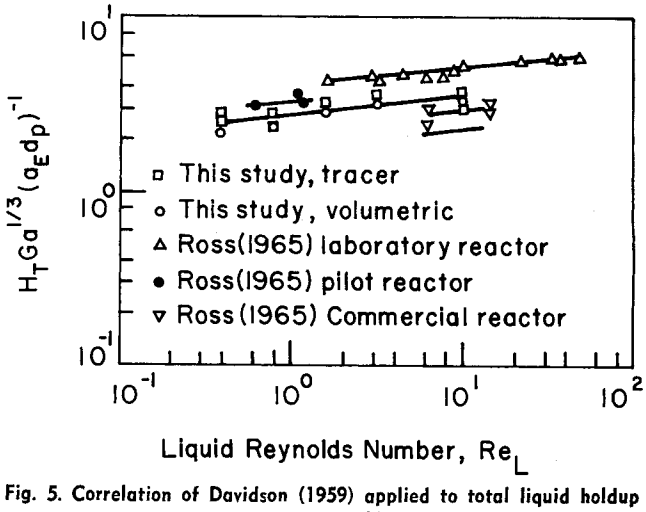

Fig. 5. Correlation of Davidson (1959) applied to total liquid holdup on porous packing.

LIQUID DISPERSION

Axial dispersion numbers, $D/U_L Z$, that is, the reciprocal of the bed axial Peclet numbers, are obtained from Equation (2). The dispersion numbers ranged from 0.006 up to 0.038 but appear to be scattered between these values. The results are plotted as particle Peclet number (Bodenstein number) versus particle Reynolds number in Figure 6. The values of particle Peclet numbers found are one fourth to one tenth of those for single phase liquid flow in packed beds at comparable Reynolds numbers (Levenspiel and Bischoff 1963). This compares well with other literature values for two phase flows. The correlation of Hochman and Effron (1969), extended to lower Reynolds numbers, fits our data quite well. The data of Sater and Levenspiel (1966) for countercurrent flow have even

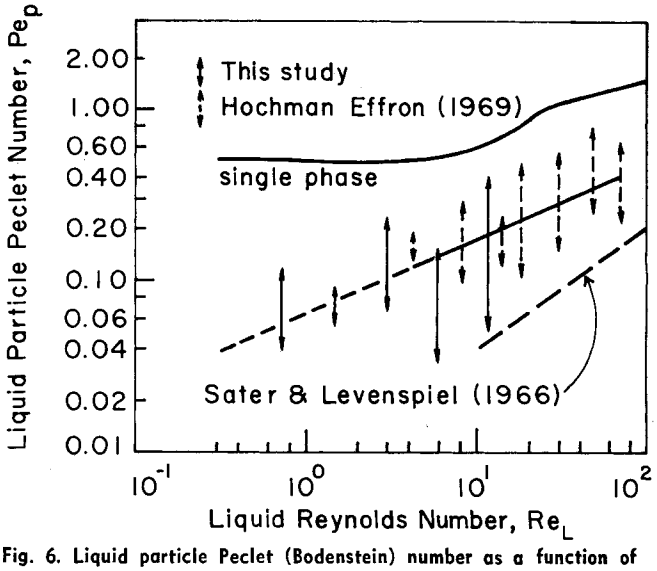

Fig. 6. Liquid particle Peclet (Bodenstein) number as a function of liquid Reynolds number.

lower particle Peclet numbers, which is to be expected, since greater backmixing occurs for the countercurrent flow pattern. The agreement of Peclet numbers with the correlation of Michell and Furzer (1972b) is as good as with the correlation of Hochman and Effron (1968).

CONCLUSIONS

The data of this study for cocurrent downward flow confirm the previous observations that liquid holdup increases with liquid flow rate and is almost independent of gas flow rate over a moderate range. However, most available correlations underestimate the holdup in trickle-beds packed with small particles (d_p = 0.06 cm), which are at the lower end of the size range of interest, by more than 40%. Holdup dependence on liquid flow rate is less pronounced than in beds packed with larger particles.

Liquid phase particle Peclet numbers are four to ten times smaller than in single phase flow indicating larger dispersion in trickle flows. The available correlations for particle Peclet numbers, obtained for beds packed with larger particles, fit the dispersion data for beds packed with small particles well.

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NOTATION

a_E = total external geometric pellet surface area per unit reactor volume, cm^{-1}
a_w = external geometric pellet surface area contacted by liquid per unit reactor volume, cm^{-1}
D = dispersion coefficient, cm^2s^{-1}
d_p = mean pellet diameter, cm
G = superficial gas mass velocity, kg $m^{-2}s^{-1}$
Ga_L = Gallileo number for the liquid phase, $(= d_p^3 g\rho_L^2 \mu_L^{-2})$, dimensionless
H_D = dynamic liquid holdup, (cm^3 moving liquid/cm^3 reactor volume), dimensionless
H_E = external liquid holdup, (cm^3 liquid external to packing/cm^3 reactor volume), dimensionless
H_T = total liquid holdup, (cm^3 liquid/cm^3 reactor volume), dimensionless
L = superficial liquid mass velocity, kg $m^{-2}s^{-1}$
Pe = axial Peclet number for the bed $(= U_LZ/D)$, dimensionless
Pe_p = particle Peclet number, Bodenstein number $(= U_Ld_p/D)$, dimensionless
Pe^1 = particle Peclet number $(= U_id_p/D)$, dimensionless
Q_L = liquid flow rates, cm^3s^{-1}
Re_L = Reynolds number $(= d_p\rho_LU_L/\mu_L)$, dimensionless
Re^1 = Reynolds number $(= d_p\rho_LU_i/\mu_L)$, dimensionless
Re_f = film Reynolds number $(= 4\rho\delta_LU_i/\mu)$, dimensionless
$\bar{t}_L$ = mean residence time of the liquid phase, s
U_i = interstitial liquid velocity, cm s^{-1}
U_L = superficial liquid velocity, cm s^{-1}
V = reactor volume, cm^3
Z = reactor length, cm

Greek Letters

ΔP_G = pressure drop for the gas phase in single phase flow, kg m^{-2}
ΔP_L = pressure drop for the liquid phase in single phase flow, kg m^{-2}
$\Delta\sigma^2$ = difference in the variance of the input and output signal, s^2
δ = laminar liquid film thickness, cm
ϵ = bed porosity, dimensionless
ρ_G = gas density, g cm^{-3}
ρ_L = liquid density, g cm^{-3}
μ_L = liquid viscosity, poise
χ = pressure drop ratio defined in Table 1, dimensionless
χ^1 = parameter defined in Table 1, dimensionless

LITERATURE CITED

Bischoff, K. B., "Notes on the Diffusion-type Model for Longitudinal Mixing in Flow," *Chem. Eng. Sci.*, **12**, 69 (1960).

Burghardt, A., and T. Zaleski, "Longitudinal Dispersion at Small and Large Peclet Numbers in Chemical Flow Reactors," *ibid.*, **23**, 575 (1968).

Charpentier, J. C., M. Bakos, and P. Legoff, "Hydrodynamics of Two-Phase Concurrent Downflow in Packed Bed Reactors," *2nd Congr. on Quelques Applications de la Chimie Physique*, Veszpren, Hungary (1971).

Charpentier, J. C., and M. Favier, "Some Liquid Holdup Experimental Data in Trickle-Bed Reactors for Foaming and Nonfoaming Hydrocarbons," *AIChE J.*, **21**, 1213 (1975).

Davidson, J. F., E. J. Cullen, D. Harison, and D. Roberts, "The Holdup and Liquid Film Coefficient of Packed Towers, Part I: Behavior of a String of Spheres," *Trans. Inst. Chem. Engrs.*, **37**, 122 (1959).

Goto, S., and J. M. Smith, "Trickle-Bed Reactor Performance, Part I: Holdup and Mass Transfer Effects," *AIChE J.*, **21**, 706 (1975).

Hochman, J. M., and E. Effron, "Two-Phase Cocurrent Downflow in Packed Beds," *Ind. Eng. Chem. Fundamentals*, **8**, 63 (1969).

Jesser, B. W., and J. C. Elgin, "Studies of Liquid Holdup in Packed Towers," *Trans. Am. Inst. Chem. Engrs*, **39**, 277 (1943).

Kittrell, J. R., and M. Kamionski, "Trickle-Bed Studies for Gluconic Acid Production Using Immobilized Enzymes," presented at AIChE 81st National Meeting, Kansas City, Mo. (Apr., 1976).

Larkins, R. P., R. R. White, and D. W. Jeffrey, "Two-Phase Concurrent Flow in Packed Beds," *AIChE J.*, **7**, 231 (1961).

Levenspiel, O., and K. B. Bischoff, "Patterns of Flow in Chemical Process Vessels," *Advan. Chem. Eng.*, **4**, 95 (1975).

Mears, D. E., "The Role of Axial Dispersion in Trickle Flow Laboratory Reactors," *Chem. Eng. Sci.*, **26**, 1361 (1971).

Michell, R. W., and I. A. Furzer, "Trickle Flow in Packed Beds," *Trans. Inst. Chem. Engrs.*, **50**, 334 (1972*a*).

———, "Mixing in Trickle Flow through Packed Beds," *Chem. Eng. J.*, **4**, 53 (1972*b*).

Mohunta, D. M., and G. S. Laddha, "Prediction of Liquid Phase Holdup in Random Packed Beds," *Chem. Eng. Sci.*, **20**, 1069 (1965).

Otake, T., and K. Okada, "Liquid Holdup in Packed Towers," *Kagaku Kogaku*, **17**, 176 (1963).

Ross, L. D., "Performance of Trickle Bed Reactors," *Chem. Eng. Progr.*, **61**, 77 (1965).

Sato, Y., T. Hirose, F. Takahashi, and M. Toda, "Pressure Loss and Liquid Holdup in Packed Bed Reactor with Cocurrent Gas-Liquid Down Flow," *J. Chem. Eng. Japan*, **6**, 147 (1973).

Sater, V. E., and O. Levenspiel, "Two-Phase Flow in Packed Beds," *Ind. Eng. Chem. Fundamentals.*, **5**, 86 (1966).

Satterfield, C. N., "Trickle-Bed Reactors," *AIChE J.*, **21**, No. 2, (1975).

Schwartz, J. G., and G. W. Roberts, "An Evaluation of Models for Liquid Backmixing in Trickle-Bed Reactors," *Ind. Eng. Chem. Process Design Develop.*, **12**, 262 (1973).

———, E. Weger, and M. P. Dudukovic, "A New Tracer Method for Determination of Liquid-Solid Contacting in Trickle-Bed Reactors," submitted to *AIChE J.* (1976).

Way, P., "The Role of the Liquid Phase in the Performance of a Trickle Bed Reactor," Ph.D. thesis, Mass. Inst Technol. Cambridge (1971).

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