

ψ = stream function
 ξ, χ = normalized radial coordinates (equal to $r/\sqrt{k_1}$, $r/\sqrt{k_2}$, respectively)
 ξ_1 = $R/\sqrt{k_1}$
 θ = polar direction

Superscripts

\wedge, \sim = inner permeable sphere, the outer porous medium
 The absence of a superscript denotes the fluid region in the cell.

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APPENDIX

The following formulas give G :

$$\alpha = [1/\xi_1 - \text{th}\xi_1(1 + 1/\xi_1^2)] \cdot [\text{th}\xi_1/\xi_1 - 1]^{-1}$$

$$\beta = (k_1/k)^{1/2} \cdot (\chi_2^3 + \chi_2^2 + 2\chi_2 + 2) \cdot (\chi_2^2 + \chi_2 + 1)^{-1} \chi_2^{-1}$$

where $\chi_2 = (k_1/k)^{1/2} \cdot \xi_1 \cdot \tilde{c}^{-1/3}$

$$\bar{A} = \xi_1^3 - 5\alpha \xi_1^2 + 10 \xi_1; \quad \bar{B} = -\xi_1^2 + 2\alpha \xi_1 - 4$$

$$\bar{G} = 2/\xi_1 + 2\alpha/\xi_1^2 + 5/\xi_1^3$$

$$\zeta = \xi_1 \tilde{c}^{1/3}; \quad \bar{A} = 3 \zeta^2 - 90 k/k_1 + 5\bar{B} + 2\xi_1^2 \bar{A}/\zeta^3$$

$$\bar{B} = -\bar{G} + 3/\zeta + \bar{B}/\zeta^3$$

$$\bar{C} = 12 \zeta^3 \beta + 30 \zeta^2 + 10 \zeta \beta (6 k/k_1 + \bar{B}) + 60 k/k_1 - 2\beta \xi_1^2 \bar{A}/\zeta^2$$

$$\bar{D} = 2\zeta \beta \bar{G} - 3\beta + 6/\zeta + (\bar{B} - 6 k/k_1) \beta / \zeta^2 + 12 \zeta^{-3} k/k_1$$

$$G = 9\bar{C} \cdot (\bar{A}\bar{D} + \bar{B}\bar{C})^{-1}$$

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Flow Patterns and Some Holdup Experimental Data in Trickle-Bed Reactors for Foaming, Nonfoaming, and Viscous Organic Liquids

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Determination of flow patterns and liquid holdup in trickle beds is of importance in the modeling, design, and scaling-up of these reactors extensively employed in the hydroprocessing of petroleum fractions (Henry and Gilbert, 1973; Mears, 1974; Paraskos et al., 1975; Satterfield, 1975; Charpentier, 1976). For porous catalyst particles usually employed in trickle-bed reactors, several holdup data have been proposed recently, either for air-water systems (Sato et al., 1973; Goto and Smith, 1975; Colombo et al., 1976) or for gas foaming and nonfoaming hydrocarbons (Satterfield and Way, 1972; Charpentier and Favier, 1975; Midoux et al., 1976; Schwartz et al., 1976). The purpose of this note is to present complementary results on flow patterns and liquid holdup for foaming and nonfoaming organic liquids, to quantify the effect of the liquid viscosity, and to compare these results with those proposed in the literature.

Experiments were carried out in a 5 cm ID column packed to a length of 1.20 m with spherical porous cobalt/molybdenum/aluminum oxide catalyst particle ($d = 2.4$ mm; $\epsilon = 0.385$) operating at atmospheric pressure over the range 18° to 25°C. The details of the experimental

equipment and procedure to measure the holdup by the weighting method are identical with the ones described by Charpentier and Favier (1975).

Hydrocarbon and organic liquid properties are presented in Table 1. In the presence of a sufficient air flow rate, methanol, kerosene, and desulfurized gas oils have a tendency to foam which does not happen with cyclohexane and ethyleneglycol. As explained previously by Charpentier and Favier, it was not possible to characterize this phenomenon by the physicochemical parameters. Superficial mass velocity of 0.5 to 9 and 0 to 0.9 kg/m²·s for the liquid phases and for the air, respectively, were mainly used to explore the whole field covered by the various flow patterns.

FLOW PATTERNS

Charpentier et al. (1975, 1976) observed various flow patterns in trickle beds. At low liquid and gas flow rates ($L < 5$ and $G < 0.01$ kg/m²·s), a trickling flow exists where the flow of the liquid is little affected by the flow of the gas (small gas-liquid interaction regime). An increase of gas and/or liquid flow rates leads to pulsing and spray flow for nonfoaming liquids, and foaming, foaming-

TABLE 1. FLUID PROPERTIES

Liquid	β_c	σ_L , dyn/cm	ρ_L , g/cm ³	μ_L , cp	A	a	λ	ψ	Foaming (*)	Key
Methanol	0.071	25.08	0.805	0.698	0.237	0.408	0.897	2.970	+	■
Cyclohexane	0.078	25.55	0.769	0.904	0.275	0.402	0.877	3.274	-	○
Kerosene	0.079	25.80	0.810	1.346	0.290	0.366	0.900	3.571	+	●
Desulfurized gas-oil	0.105	29.00	0.840	5.793	0.424	0.242	0.917	5.022	+	▼
Ethylene-glycol	0.222	48.96	1.118	17.218	0.560	0.138	1.057	3.051	-	★

(*) + foaming liquid; - nonfoaming liquid.

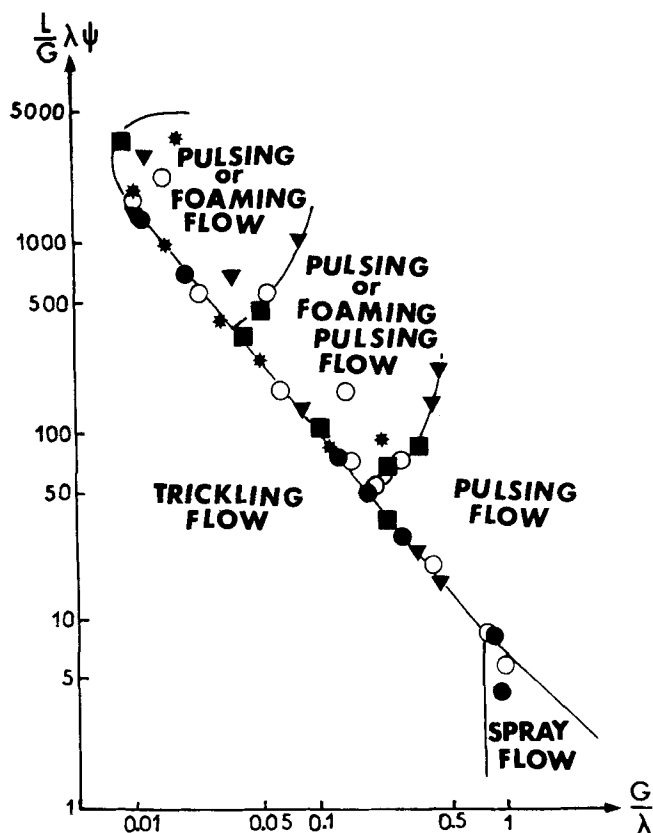


Fig. 1. Flow patterns diagram (see Table 1 for the key).

pulsing, pulsing, and spray flow for foaming liquids (high gas-liquid interaction). By taking into account the properties of the fluids, a flow pattern diagram was proposed with lines to separate the flow regimes.

In Figure 1, the experimental results obtained in this work are plotted together in one diagram. An excellent fitting is observed whatever the viscosity of the liquid (for the meaning of the key, see Table 1).

LIQUID HOLDUP

The present results concern the data of the external holdup β_{nc} and the residual or capillary holdup β_c . The capillary holdup varies typically from about 0.07 to 0.11 for the hydrocarbons and from 0.07 to 0.22 for the other organic liquids considered here. The noncapillary holdup is a function of gas and liquid flow rates and properties as well as catalyst characteristics. For the trickling flow of liquid which is obtained at a zero gas flow rate or at two-phase flow for very small gas flow rates ($G < 0.01$ kg/m²·s), β_{nc} is correlated as proportional to L^a as already observed by Charpentier et al., 1968, 1975):

$$\beta_{nc} = AL^a \quad (1)$$

where a is a function of the liquid texture, that is, the liquid and particle properties (Table 1). An attempt to

present a more general correlation has led to correlate the exponent a vs. $\log \mu_L$ and to propose the explicit equation (within $\pm 10\%$)

$$\beta_{nc} = 0.86 \left[\frac{L}{19.9} \right]^{-0.198 \log \left[\frac{\mu_L}{90.8} \right]} \quad (2)$$

which represents the experimental results of Figure 2a with a 10% accuracy. The upper limit of Equation (2) is obtained for $L = 19.9$ kg/m²·s and $\mu_L = 90.8$ cp. It has to be noted that for the higher values of the liquid viscosity the bed is flooded at any liquid flow rate in absence of a gas flow rate.

As explained by Schwartz et al. (1976), it is apparent that a simple correlation of β_{nc} vs. Re cannot fit the data of Equation (2). Moreover, the previous correlation of Satterfield et al. (1969) β_{nc} vs. $(Re/Ga)^{1/3}$ seems in better agreement (see Figure 2), but it should be noted that the agreement with exponent 1/3 is only confined to small values of liquid viscosity and flow rate. Indeed, the dynamic regime of the trickling liquid certainly changes from gravity viscosity to gravity inertia (Satterfield, 1975) then to gravity surface, and the exponents in a relation β_{nc} vs. $\mu_L^b L^a$ are not systematically $a = 0.33$ and $b = 0.25$ or 0.33 but may have different values depending on L , μ_L , and d .

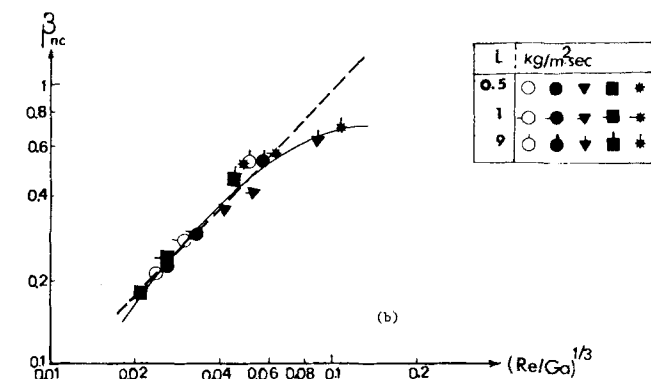
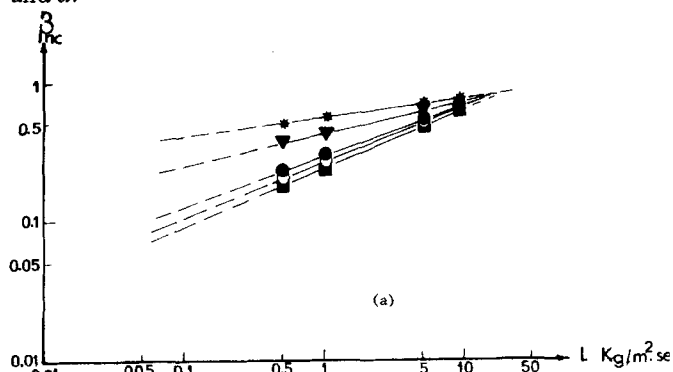


Fig. 2. Influence of the liquid flow rate and viscosity on the non-capillary liquid holdup (Figure 2a). Comparison with the correlation of Satterfield et al. (1969) (Figure 2b).

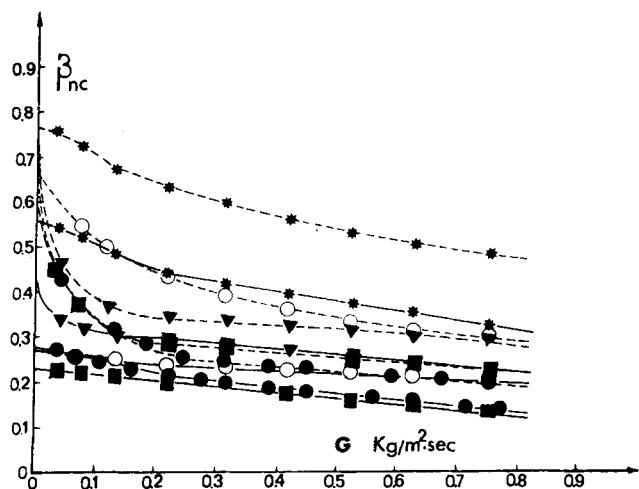


Fig. 3. Influence of the gas flow rate on the noncapillary holdup: ———— $L = 1 \text{ kg/m}^2\cdot\text{s}$; - - - - - $L = 9 \text{ kg/m}^2\cdot\text{s}$.

It is interesting to note that one of the models for design and analysis of trickle-bed reactors which is based on the assumption that the rate of reaction is proportional to the liquid holdup leads to

$$\ln \frac{C_{in}}{C_{out}} \propto \frac{k_v \beta_{nc}}{\text{LHSV}} \quad (3)$$

C_{in} and C_{out} are the concentrations at the entrance and the exit, k_v is the intrinsic first-order reaction constant per unit volume of catalyst pellet, and LHSV is the liquid hour space velocity [$L \propto [\text{LHSV}]Z$]. If now β_{nc} is replaced by the relation (2), Equation (3) is replaced by

$$\ln \frac{C_{in}}{C_{out}} \propto k_v \frac{Z^a}{(\text{LHSV})^{1-a}} \quad \text{with } a = -0.198 \log \frac{\mu_L}{90.8}$$

The slope $(1-a)$ of $\ln[C_{in}/C_{out}]$ vs. $1/\text{LHSV}$ (at otherwise constant reaction conditions) was assumed to be $2/3$ by Henry and Gilbert (1973) which supposes $a = 0.33$. But in an experimental study on desulfurization, demetallization, and denitrogenation of various gas oils by Paraskos et al. (1975) the slope was found to range from 0.532 to 0.922 , which is comparable with our range of $0.58 < 1-a < 0.86$, this extending the model for scale-up as explained by Mears (1974).

In two-phase flow, the representative liquid holdup data are presented in Figure 3 for two constant liquid flow rates, $L = 1$ and $9 \text{ kg/m}^2\cdot\text{s}$. As already obtained by Charpentier et al. (1975, 1976), the liquid holdup decreases when liquid and gas flow rates increase. For $L = 1 \text{ kg/m}^2\cdot\text{s}$, data are representative of those of pilot plant reactors, the flow pattern stays a trickling flow in the presence of a continuous gas phase, and the gas-liquid interaction is small leading to a not too important decrease of β_{nc} . Except for the viscous fluids, Equations (1) and (2) are still applicable with a good accuracy.

For $L = 9 \text{ kg/m}^2\cdot\text{s}$, data are representative of those of commercial reactors; the flow patterns which correspond to higher gas and liquid flow rates are foaming, foaming pulsing, or pulsing flow; and the gas-liquid interaction is high, thus leading to important decrease of β_{nc} . In such cases, as already observed by Midoux et al. (1976) for a given value of L and G , the liquid holdup is smaller for the foaming systems.

Midoux et al. (1976) have recently summarized the gas-liquid flow parameters χ or χ' and corresponding semi-empirical correlations based either on momentum or on energy balances that have been used by several authors to calculate the liquid holdup for packings used in trickle-

bed reactors (Larkins et al., 1961; Sato et al., 1973; Charpentier and Favier, 1975). Values of h_K and h_B which are necessary to calculate the single-phase pressure loss and then the parameters χ or χ' used to compare the present data with these correlations are given in the notations. This comparison shows that this correlation does not fit our data, which is not surprising because it does not concern results corresponding to experimental measurements with hydrocarbons.

Except for the case when the liquid is the viscous ethyleneglycol and the holdup is thus much bigger, the comparison of the data with Sato et al.'s correlation for air-water systems indicates that this correlation always underestimates the holdup (20% when $L \leq 5 \text{ kg/m}^2\cdot\text{s}$ and 20 to 40% when $L > 5 \text{ kg/m}^2\cdot\text{s}$). The fit is also much better with the correlations of Charpentier and Favier or Midoux et al. for hydrocarbons ($\pm 20\%$). However, for the viscous liquids, a complementary correction factor was necessary to take the viscosity effect into consideration in these two correlations. Actually, a good fit of the experimental data with these correlations is obtained when β is replaced by $[\beta(\mu_{wat}/\mu_L)^{0.20}]$. For example, such a correction with the correlation of Charpentier and Favier (1975) is presented in Figure 4:

$$\log \left[\beta \left(\frac{\mu_{wat}}{\mu_L} \right)^{0.20} \right] = -0.363 + 0.168 \log \chi' - 0.043 (\log \chi')^2 \quad (4)$$

Moreover, a comparison with the empirical correlations of Specchia and Baldi (1977) for the high interaction regime has shown that these correlations fit the data with a $\pm 30\%$ accuracy only for the nonviscous liquids. For viscous liquids, the measured holdups are always between 50 and 75% higher.

In conclusion, for design purpose, flow pattern diagrams and correlations proposed by Charpentier et al. (1975, 1976) for foaming and nonfoaming hydrocarbons are still applicable with a reasonable order of magnitude for foaming and nonfoaming organic liquids. Furthermore, for gas and liquid flow rates representative of those of commercial reactors, a correction factor has been proposed to take into consideration the liquid viscosity effect when $\mu_L < 20$ cp. It would be interesting to extend these results to higher values of μ_L . Finally, for generalization, it is also noteworthy to remark that the correlation (4) is still valuable for spherical porous particles within the accuracy of the equation, though it was initially proposed for cylindrical pellets, and that this correlation concerns packing diameters bigger than 2 mm.

Indeed, a comparison with the data of Goto and

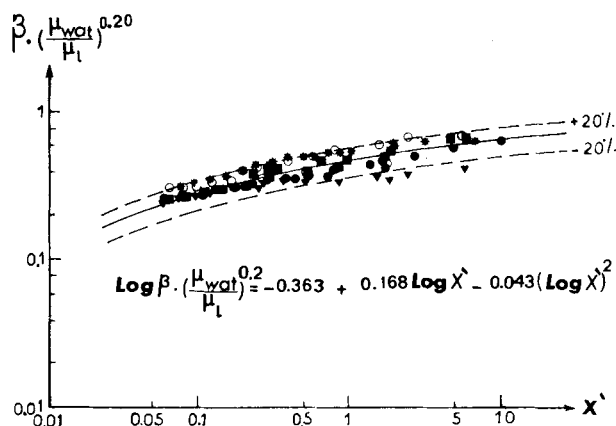


Fig. 4. Correlation of total liquid holdup.

Smith (1975) concerning 0.54 mm β -naphthol granular particles and with the data of Schwartz et al. (1976) concerning 0.6 mm alumina particle has shown that Equation (4) give values of holdup which are at least 30% lower.

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NOTATION

- A = coefficient in Equation (1)
 a = exponent in Equation (1)
 d = particle diameter (m)
 G = superficial gas flow rate ($\text{kg}/\text{m}^2 \cdot \text{s}$)
 Ga = Galileo number $= (d^3 g \rho_L^2 / \mu_L^2)$
 h_K, h_B = constant in an Ergun type of equation ($h_K = 4.796; h_B = 0.245$)
 L = superficial liquid flow rate ($\text{kg}/\text{m}^2 \cdot \text{s}$)
 Re = Reynolds number $= (L d / \mu_L)$
 Z = reactor length (m)

Greek Letters

- $\beta, \beta_{nc}, \beta_c$ = total, noncapillary and capillary holdup expressed in percentage of interparticle void $\beta = \beta_{nc} + \beta_c$
 ϵ = porosity (interparticle)
 λ = flow parameter $= \left[\frac{\rho_G}{\rho_{air}} \cdot \frac{\rho_L}{\rho_{wat}} \right]^{1/2}$
 μ_L, μ_{wat} = liquid and water viscosity (cp)
 $\rho_G, \rho_L, \rho_{wat}$ = gas, liquid and water density (g/cm^3)
 σ_L, σ_{wat} = liquid and water surface tension (dyne/cm)
 ψ = flow parameter $= \frac{\sigma_{wat}}{\sigma_L} \cdot \left[\frac{\mu_L}{\mu_{wat}} \left(\frac{\rho_{wat}}{\rho_L} \right)^2 \right]^{1/3}$
 χ, χ' = gas-liquid flow parameters defined by Charpentier and Favier (1975)

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A General Direct Digital Control Algorithm for a Class of Linear Systems

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Owing to a large increase in the use of digital computers for process control purposes, it has become in-

creasingly important to develop practical direct digital control algorithms. Several special purpose algorithms, both continuous and discrete, have been reported in the literature for lumped parameter systems. In an early paper,

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