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Dynamic Liquid Holdup in Two-Phase Downflow in Packed Beds: Air-Silicone Oil System

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There are three purely fluid mechanical aspects to the design and operation of two-phase, cocurrent packed bed catalytic reactors. These are the overall pressure drop through the bed, the fluid flow regime, and the dynamic liquid holdup. In this note, new data are presented for the dynamic liquid holdup in systems which were chosen to be closely analogous to fairly heavy petroleum fractions found in various hydrotreating reactors.

The dynamic liquid holdup, as used here, is that fraction of the total voidage of the packed bed occupied by readily drainable liquid. This excludes liquid trapped in the catalyst pore volume and the thin liquid film, which tends to adhere to the catalyst surface after complete drainage.

The importance of the dynamic liquid holdup in effective operation of hydroprocessing reactors is well documented (Mears 1974, Ross 1965, Goto and Smith 1975). The amount of liquid held in the interstices of the catalyst bed affects the chemical reaction rate and the average liquid residence time. In planning shutdown procedures, it is important to know how much liquid is held in the bed in order to estimate drainage times.

EXPERIMENTAL PROGRAM

To replicate hydrocarbon liquid phase properties as closely as possible, without the attendant hazards of using hydrocarbons, the experimental liquids used were Dow-Corning DC 200 series silicone fluids. These fluids had measured viscosities of 3.2 and 8.2 centipoise, densities of 0.9 to 0.92 gm/cc, and a surface tension (compared to air) of 18 to 20 dynes/cm. Air was the gas phase in most experiments, but R12 (dichlorodifluoromethane) was used in a few tests to investigate gas viscosity and density effects on holdup.

All of the data were taken in a glass test column 55 mm ID and 1.5 m long. The several packing-fluid-gas systems studied are shown in Table 1. All of the packings were commercial catalysts.

The experimental procedure was relatively simple. The column would be allowed to stabilize at a desired combination of liquid and gas flow rates. The liquid and gas flow rates were chosen so that the data could be analyzed in a block, with comparisons either along lines of constant liquid or constant gas flow rate. The liquid holdup measurements were taken by simply cutting off the liquid flow and collecting the liquid as it drained out. The liquid was collected in timed fractions for a period of 30 min. When the timed data are plotted as in Figure 1, the initial volume of liquid can be obtained by extrapolation to zero time.

TABLE 1. PACKING-LIQUID-GAS SYSTEMS.

Packing Size	Void Fraction	Liquid	Gas
1.04 mm extrudate	0.361	3.2 cp silicone	Air
1.04 mm extrudate	0.361	3.2 cp silicone	R12
1.04 mm extrudate	0.361	8.2 cp silicone	Air
1.39 mm extrudate	0.330	3.2 cp silicone	Air
3.35 mm extrudate	0.320	3.2 cp silicone	Air

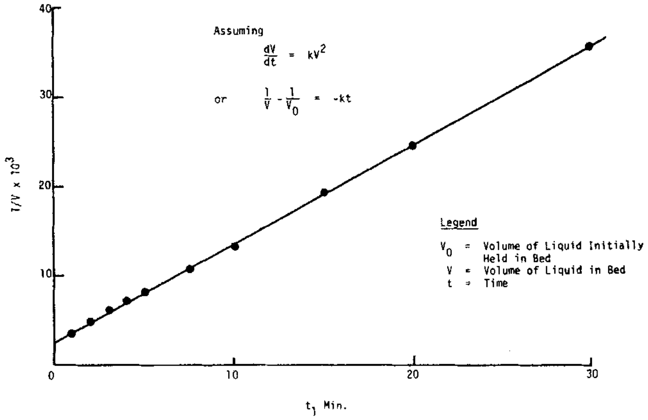

Figure 1. 1/*V* versus *t*.

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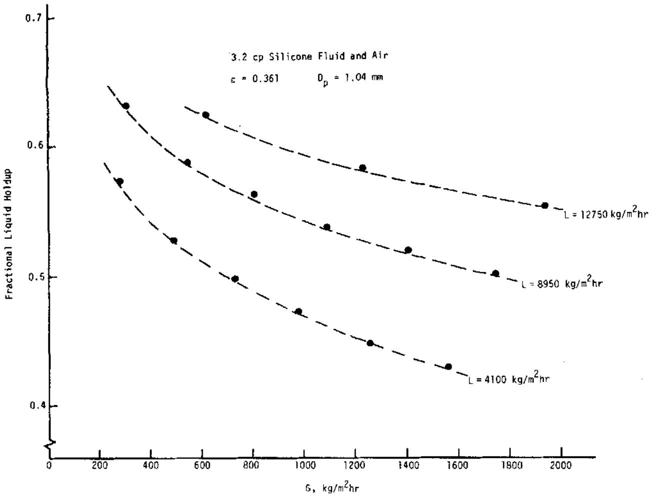

Figure 2. Effect of gas rate on dynamic liquid holdup.

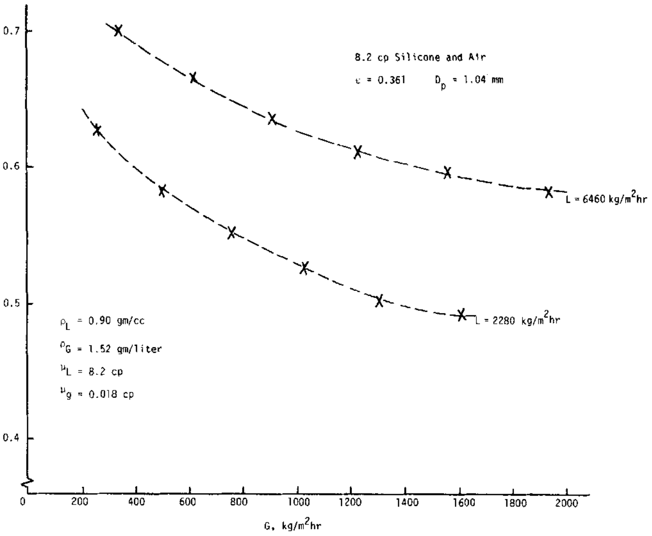

Figure 3. Effect of gas rate on dynamic liquid holdup.

PRESENTATION OF EXPERIMENTAL HOLDUP DATA

The liquid holdup data are reported as fractional liquid holdup, that is, the fraction of the available voidage filled with liquid. This removes the explicit dependence of the data upon bed void fraction and bed volume. Fractional liquid holdup data are shown in Figures 2-4.

From Figure 2, it is evident that both the liquid and gas rates play a significant role in determining the liquid holdup. Comparing the holdups shown in Figure 3 with those in Figure 2, an increase in liquid viscosity also increases the liquid holdup, everything else being equal. The results shown in curves I and II of Figure 4 indicate that changes in gas properties have some effect on liquid holdup, but not as extreme as the effect of liquid viscosity. Comparing curves II, III, and IV in Figure 4 leads to the conclusion that larger particles (larger voids) result in a lower fractional holdup.

LIQUID HOLDUP CORRELATIONS

A number of dynamic liquid holdup correlations have been presented in the literature. Most of these have been developed primarily from air-water data. The correlations considered here are those of Otake and Okada (1953) for broken solids and Rashig rings

$$\beta_{nc} = 15.1(Re)^{0.676}(Ga)^{-0.44}(a_tD_p)^{-0.6} \quad 10 < Re < 2000 \quad (1a)$$

$$\beta_{nc} = 21.1(Re)^{0.51}(Ga)^{-0.44}(a_tD_p)^{-0.6} \quad 0.01 < Re < 10 \quad (1b)$$

Larkins et al. (1961)

$$\log_{10}\beta_{nc} = 0.774 + 0.525\,(\log_{10}\chi) - 0.109\,(\log_{10}\chi)^2 \qquad (2)$$

where

$$\chi = \left[\left(\frac{\Delta P}{Z}\right)_l \Big/ \left(\frac{\Delta P}{Z_g}\right)\right]^{1/2} \qquad (3)$$

and Turpin and Huntington (1967)

$$\beta_{nc} = 0.017 + 0.132\,(L/G)^{0.24} \qquad (4)$$

In all cases, β_{nc} is the fractional, non-capillary (dynamic liquid holdup).

Charpentier and Favier (1975) suggested the expression

$$\log\beta = 0.363 + 0.168\,\log\chi' - 0.043\,(\log\chi')^2 \quad 0.05 < \chi' < 100 \qquad (5)$$

where $\beta = \beta_{nc} + \beta_c$. According to Charpentier $\beta_c \simeq 0.06$ to 0.11 and

$$\chi' = \left[\left(\frac{L}{G}\right)\Big/\left[\frac{1}{\rho_G g_c}\cdot\frac{\Delta P_G}{Z} + 1\right]\right]^{1/2} \qquad (6)$$

The principle behind this expression is the concept that the dynamic holdup is more truly dependent upon the frictional energy dissipation measured by χ' than upon the momentum dissipation measured by Larkins' χ. Equation (5) is based upon some 1,500 data points, including many hydrocarbon systems.

A recent model of dynamic liquid holdup in two-phase flow (Clements 1978) depicts flow in a packed bed as annular flow through a bundle of capillary tubes. The model requires an equilibrium between the pressure in the gas phase and the surface forces in the liquid phase at every point in each capillary. The result of the force balance suggested by this model is that

$$\beta_{nc} \propto \left[\frac{Re_gWe_g}{Re_l}\right]^n \qquad (7)$$

where $n = n(\sigma)$. Based upon only a few data, it appears that the relation between n and σ is linear. The data presented here for compounds where $\sigma = 20$ give

$$\beta_{nc} = 0.84\left[\frac{Re_gWe_g}{Re_l}\right]^{-0.034} \qquad (8)$$

Table 2 compares the new holdup data presented here with the correlation noted above. The correlation of Charpentier and Favier is seen to be quite adequate in representing these data.

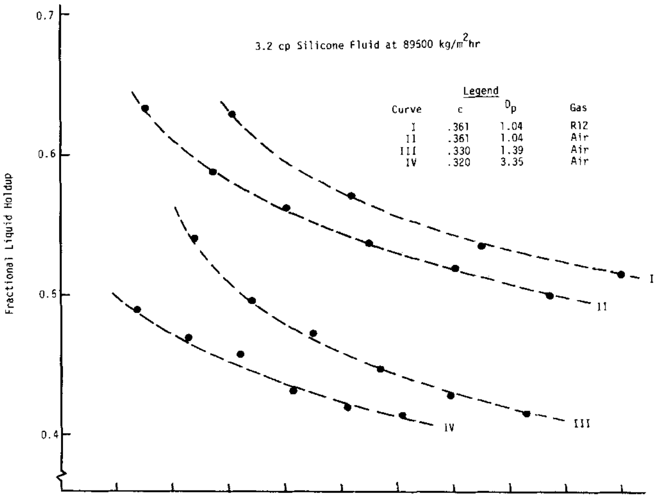

Figure 4. Effect of gas composition and bed properties on dynamic liquid holdup.

TABLE 2. RESULTS OF CORRELATION TESTING
(45 data points from this study)

Correlation	Average Absolute Deviation, %	Average Error %
Larkins et al. (1961)	57.9	−57.9
Turpin and Huntington (1967)	62.2	−62.2
Otake and Okada (1953)	30.3	−30.2
Charpentier and Favier (1975)	14.3	− 9.4
Clements (1978)	10.4	0.6

SUMMARY

1. Both the gas and liquid flow rates and liquid viscosity, as well as the packing size, are seen to have a significant (± 50 percent) effect on the dynamic liquid holdup in a packed column.

2. The gas density and viscosity appear to have a less strong (± 20 percent) influence on dynamic liquid holdup.

3. The holdup correlation of Charpentier and Favier (1975) and the capillary flow model of Clements (1978) appear to represent the data for viscous liquids in beds of small catalyst packings well.

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NOTATION

a_t = total external surface area of particles, cm^2/(cm^3 of empty tube)
D_p = effective particle diameter, cm
g = absolute acceleration of gravity, cm/sec^2
G = superficial gas flow rate, gm/cm^{2}sec
Ga = Galileo number $D_p{}^3 g\rho^2/\mu^2$
L = superficial liquid flow rate, gm/cm^{2}sec
$\Delta P/Z$ = pressure drop per unit length, atm/m
Re = Reynolds number, D_pG/μ_G, D_pL/μ_L
U_L, U_G = velocity of liquid and gas, respectively, cm/sec
We = Weber number, $U_G{}^2D_p\rho_G$

Greek Letters

β_{nc} = dynamic holdup, (cm^3 of liquid)/(cm^3 of bed)
ϵ = bed void fraction
δ_L, δ_G = predicted single-phase pressure drops for gas and liquid, respectively, atm/cm
μ_L, μ_G = liquid and gas phase viscosity, cp
ρ_L, ρ_G = liquid and gas phase density, gm/cc
χ = $[(\Delta P/Z)_1/(\Delta P/Z)_g]^{\frac{1}{2}}$
σ = surface tension, dyne/cm

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Transient Behavior of Fresh and Fouled Nickel Hydrogenation Catalyst

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The unsteady-state characteristics of catalytic reactors must be understood in order to design control systems, to investigate stability, and to determine whether to operate the reactor in a steady-state or cyclic mode. To this end, experimental studies have been conducted of the transient behavior of acetylene hydrogenation reactions, the overall stoichiometry of which may be written

$$C_2H_2 + H_2 \rightarrow C_2H_4$$
$$C_2H_4 + H_2 \rightarrow C_2H_6.$$

EXPERIMENTAL MATERIALS AND METHODS

A commercial nickel catalyst, Girdler G87RS, was employed in these investigations. It contains 43% nickel by weight on a refractory support, with a surface area of 3.4×10^4 m^{2}kg^{-1}. Prior to use, the catalyst was crushed and screened to a size range 0.84-1.2 mm, after which the pellets were pretreated for 24 h in a 5% (wt) solution of $Na_2S_2O_3$ (Komiyama and Inoue 1968). Before each experiment, the catalyst was activated in the reactor in a hydrogen stream at 477 K for 6 h.

All experiments were conducted in a spinning-basket catalytic CSTR. The feed flow rates of hydrogen and nitrogen were switched with a timer and solenoid valves, and effluent hydro-

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