

$$\bar{N}_i = \frac{N_i}{ak_0C}$$

Subscripts

- s = solid coal
 g = gas
 H_2 = hydrogen
 I = inert gas
 v = stable volatiles
 v^* = reactive volatiles

Superscripts

- c = value in core
 λ = value at reaction interface

LITERATURE CITED

- Anthony, D. B., "Rapid Devolatilization and Hydrogasification of Pulverized Coal," Sc.D. thesis, Dept. Chemical Engineering, Mass. Inst. Technol., Cambridge (1974).
 ———, and J. B. Howard, "Coal Devolatilization and Hydrogasification," *AIChE J.*, **22**, 625 (1976).
 ———, H. C. Hottel, and H. P. Meissner, "Rapid Devolatilization and Hydrogasification of Bituminous Coal," *Fuel*, **55**, 121 (1976).
 Aris, R., *The Mathematical Theory of Diffusion and Reaction in Permeable Catalysts*, Clarendon Press, Oxford, England (1975).
 Badzioch, S., D. R. Gregory, and M. A. Field, "Investigation of the Temperature Variation of the Thermal Conductivity and Thermal Diffusivity of Coal," *Fuel*, **43**, 267 (1964).
 Benson, S. W., *The Foundations of Chemical Kinetics*, McGraw-Hill, New York (1960).
 Dutta, S., C. Y. Wen, and R. J. Belt, "Reactivity of Coal and Char in CO₂ Atmosphere," *Am. Chem. Soc., Div. of Fuel Chem. Preprints*, **20**, No. 3, 103 (1975).
 Graff, R. A., S. Dobner, and A. M. Squires, "Flash Hydrogenation of Coal: 1. Experimental Methods and Preliminary Results; 2. Yield Structure for Illinois No. 6 Coal at 100 atm," *Fuel*, **55**, 109, 113 (1976).
 Greene, M. I., "Engineering Development of the Cities Service, Short Residence Time (CS-SRT) Process," *Am. Chem. Soc., Div. of Fuel Chem. Preprints*, **22**, No. 7, 133 (1977).
 Hirschfelder, J. O., C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids*, Wiley, New York (1954).
 Hite, R. H., and R. Jackson, "Pressure Gradients in Porous Catalyst Pellets in the Intermediate Diffusion Regime," *Chem. Eng. Sci.*, **32**, 703 (1977).
 Hutchings, J., and J. J. Carberry, "The Influence of Surface Coverage on Catalyst Effectiveness and Selectivity. The Isothermal and Nonisothermal Cases," *AIChE J.*, **12**, 20 (1966).
 Johnson, J. L., "Kinetics of Bituminous Coal Char Gasification with Gases Containing Steam and Hydrogen," in *Coal Gasification*, p. 145, Advances in Chemistry Series, No. 131, Am. Chem. Soc., Washington, D.C. (1974).
 Loison, R., A. Peytary, A. F. Bower, and R. Grillo, "The Plastic Properties of Coal," in *Chemistry of Coal Utilization*, Supplementary Vol., H. H. Lowry, ed., p. 150, New York (1963).
 Mason, E. A., A. P. Malinauskas, and R. B. Evans, III, "Flow and Diffusion of Gases in Porous Media," *J. Chem. Phys.*, **46**, 3199 (1967).
 Mazamdar, B. K., and N. N. Chatterjee, "Mechanism of Coal Pyrolysis in Relation to Industrial Practice," *Fuel*, **52**, 11 (1973).
 Moseley, F., and D. Paterson, "The Rapid High-Temperature Hydrogenation of Coal Chars, Part 2: Hydrogen Pressures up to 100 Atmospheres," *J. Inst. Fuel*, **38**, 378 (1965).
 Nusselt, W., "Die Verbrennung und die Vergassung der Kohle auf dem Rost," *Ver. Deut. Ing.*, **60**, 102 (1916).
 Reid, R. C., and T. K. Sherwood, *The Properties of Gases and Liquids*, McGraw-Hill, New York (1958).
 Scheidegger, A. E., *The Physics of Flow through Porous Media*, Univ. Toronto, Canada (1960).
 Steinberg, M., and P. Fallon, "Coal Liquefaction by Rapid Gas Phase Hydrogenation," *Am. Chem. Soc., Div. of Petroleum Chem. Preprints*, **20**, No. 2, 542 (1975).
 Suuberg, E. M., W. A. Peters, and J. B. Howard, "Product Composition and Kinetics in Lignite Pyrolysis," *Am. Chem. Soc., Div. of Fuel Chem. Preprints*, **22**, No. 1, 112 (1977).
 Van Krevelen, D. W. F., *Coal*, Elsevier, Amsterdam (1961).
 Vand, V., "A Theory of the Irreversible Electrical Resistance Changes of Metallic Films Evaporated in Vacuum," *Proc. Phys. Soc. (London)*, **A55**, 222 (1943).
 Zahradnik, R. L., and R. A. Glenn, "Direct Methanation of Coal," *Fuel*, **50**, 77 (1971).
 Manuscript received December 5, 1977; revision received August 11, and accepted August 25, 1978.

Continuous Phase Mass Transfer During Formation of Drops from Jets

This study enables the extension of a recent design procedure for perforated plate extraction columns to include operation under jetting conditions. Correlations of experimental data are given for jet length, jet contraction, drop size, and mass transfer rates in continuous phase controlled liquid-liquid systems. Systems exhibiting very low mass transfer rates were used, and this gave significantly different results from those obtained earlier with moderate rates of mass transfer.

A. H. P. SKELLAND

and

Y-F. HUANG

Chemical Engineering Department
 The University of Kentucky
 Lexington, Kentucky 40506

SCOPE

A method for the design of perforated plate extraction columns was published recently by Skelland and Conger (1973). The procedure is in terms of rate equations and

relevant hydrodynamics and, after machine computation using Fortran IV language, yields the number of real plates required for a given separation, the column diameter, the number of holes per plate, and the cross-sectional area of the downcomers. In view of Mayfield and Church's

(1952) demonstration that plate efficiencies improve substantially when operated with jets issuing from each perforation, a previous paper (Skelland and Huang, 1977) correlated the necessary aspects of operation under jetting conditions for dispersed phase controlled systems.

CONCLUSIONS AND SIGNIFICANCE

The hydrodynamic portions of this work led to correlations for jet length, jet contraction at the point of breakup, and size of drops formed at the end of the jet. Fortuitously, the systems selected exhibited much lower rates of mass transfer than in the previous dispersed phase controlled study (Skelland and Huang, 1977). It was found that this led to corresponding substantial differences in jet length and, to a lesser degree, in jet contraction and drop size, compared with the previous work.

Mass transfer rates were measured during drop formation under jetting conditions and the results compared with predicted values. The predictions were obtained using the present correlations for jet length, jet contrac-

The present paper set out to obtain corresponding correlations for continuous phase controlled systems. Results showed, however, that some hydrodynamic aspects of the operation are substantially dependent upon the rate at which mass transfer is occurring.

tion, and drop size, but continuous and dispersed phase mass transfer coefficients for drop formation, free fall, and coalescence were computed using correlations established for drops formed at a nozzle tip instead of at the end of a liquid jet. The predictions were consequently somewhat conservative (low).

The procedure for extraction column design, extended now to include jetting operation, was applied to the jetting, high interfacial tension data obtained in a six-plate column by Mayfield and Church (1952). The number of plates predicted to give several different observed separations agreed well with those actually used.

Perforated plate columns are widely used in liquid-liquid extraction. When the flow rate of disperse phase through the perforations is greater than the jetting velocity, jets are formed which break into drops at some distance from the plate. Mayfield and Church (1952) have shown that plate efficiency may increase up to two and one-half fold when columns are operated in this way, with accompanying increase in throughput rates.

Skelland and Huang (1977) have measured and correlated the hydrodynamic and mass transfer aspects of drop formation under jetting conditions in dispersed phase controlled systems. The present paper reports comparable work in continuous phase controlled systems but with much lower rates of mass transfer. The two studies in combination should enable the recently published design procedure of Skelland and Conger (1973) to be extended to columns operating with the advantages of drop formation from jet breakup.

PREVIOUS WORK AND THEORY

The relevant hydrodynamics of drop formation under jetting conditions involve the jet length, the jet contraction or diameter at the point of breakup, the jetting velocity, and the resulting drop size. Some previous work on these aspects was outlined by Skelland and Huang (1977), who noted that seemingly conflicting reports on the effects of mass transfer on jet length were qualitatively consistent with the conclusions of Burkholder and Berg (1974). The latter authors found that transfer of a surface tension lowering solute either into or out of the jet may be either stabilizing (longer jets) or destabilizing (shorter jets), depending on physical properties of the system and the mass transfer rate. Skelland and Huang (1977) then presented correlations of their own

experimental results for most of these hydrodynamic quantities and their attendant mass transfer phenomena, using dispersed phase controlled systems undergoing mass transfer at moderate rates.

Consider mass transfer between two consecutive plates in a perforated plate extraction column operating under jetting conditions. The total transfer rate is the sum of that occurring from the jet, during drop formation, during free fall (or rise), and during coalescence at the next plate. In the present work, the solute concentration was very low in the dispersed phase and effectively zero in the continuous phase. Mass transfer rates were also very low, and so

$$q = (K_{cj}A_j + K_{cf}A_f + K_{cr}A_r + K_{cc}A_c)\Delta C_A \quad \Delta C_A = C^*_{Ac} \text{ at } C_{Ad} = (C_{Adi} + C_{Adf})/2 \quad (1)$$

The overall coefficients of mass transfer are assembled in the usual manner from the individual coefficients for each phase as

$$\frac{1}{K_{cj,f,r, \text{ or } c}} = \frac{1}{k_{cj,f,r, \text{ or } c}} + \frac{1}{mk_{dj,f,r, \text{ or } c}} \quad (2)$$

The expressions used in the present study for $k_{cf,r, \text{ or } c}$ and $k_{df,r, \text{ or } c}$ are exactly the ten listed in Table II in the paper by Skelland and Conger (1973), with due regard for whether the falling drops were stagnant, circulating, or oscillating, but with the right side of each expression multiplied by M/ρ to accommodate the use of ΔC_A type driving forces in Equation (1) instead of mole fractions. This procedure involves the important assumption that these correlations for $k_{cf,r, \text{ or } c}$ and $k_{df,r, \text{ or } c}$ from data for drops formed at a nozzle are also applicable to drops formed at the end of a liquid jet. Comparison with experiment will test the validity of this assumption.

TABLE 1. PHYSICAL AND TRANSPORT PROPERTIES

System	σ , dyne/cm	ρ_d , g/ml	ρ_c , g/ml	μ_d , g/cm-s	μ_c , g/cm-s	D_d , cm ² /s	D_c , cm ² /s	Temp., °C
1	25.1	1.1025	0.9971	0.006925	0.008937	1.70×10^{-5}	1.21×10^{-5} *	25
2	31.6	1.5825	0.9971	0.006364	0.008937	9.1×10^{-6} *	1.21×10^{-5} *	25
3	26.8	1.2143	0.9971	0.03693	0.008937	5.28×10^{-6}	1.21×10^{-5} *	25

* From Perry (1963).

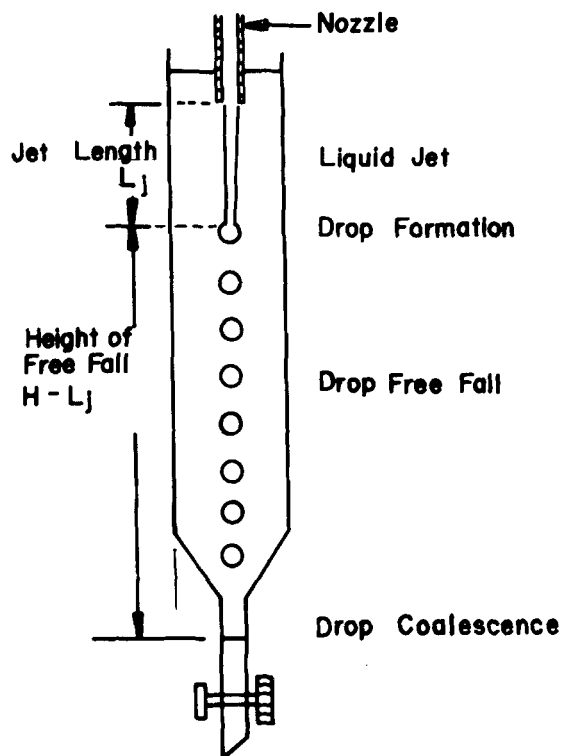


Fig. 1. The extraction column operating under jetting conditions.

The quantity k_{dj} was evaluated from Penetration Theory as

$$k_{dj} = 2 \left(\frac{D_d}{\pi t_e} \right)^{1/2} ; \quad t_e = L_j / U_j \quad (3)$$

in accordance with Skelland and Huang's (1977) findings. Similarly, k_{cj} was estimated by

$$k_{cj} = 2 \left(\frac{D_c}{\pi t_e} \right)^{1/2} ; \quad t_e = L_j / U_j \quad (4)$$

In some of the sections to follow, distinctions are made between phenomena occurring with mass transfer rates that are either zero, very low, or moderate.

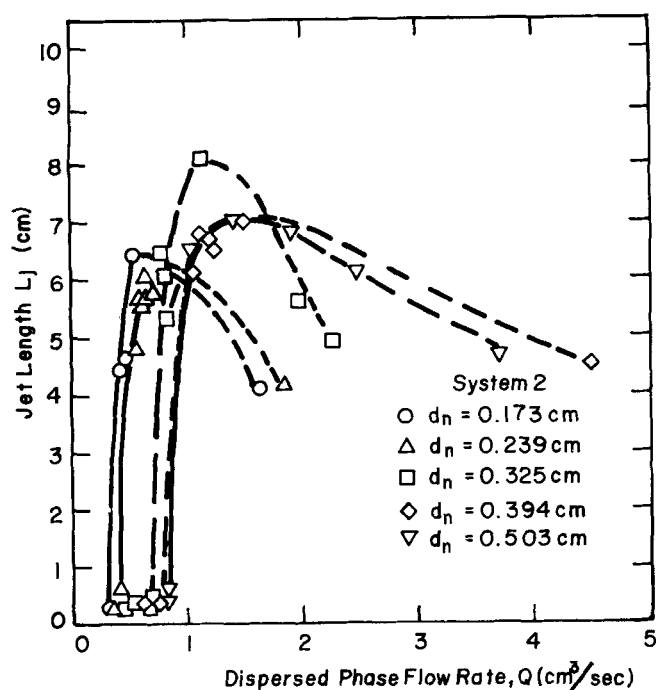


Fig. 3. Jet length vs. dispersed phase flow rate for system 2 (carbon tetrachloride-benzoic acid-water).

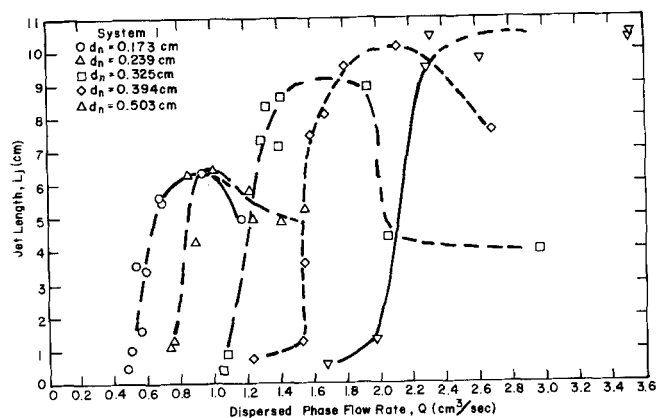


Fig. 2. Jet length vs. dispersed phase flow rate for system 1 (chlorobenzene-benzoic acid-water).

EXPERIMENTAL DETAILS

Choice of Materials

The following three continuous phase controlled systems were used, in which the dispersed phase is mentioned first, followed by the solute and continuous phase, respectively.

System 1: chlorobenzene-benzoic acid-water

System 2: carbon tetrachloride-benzoic acid-water

System 3: (Nujol-carbon tetrachloride)-benzoic acid-water

Solute transfer was from the organic to the aqueous phase throughout, with initial solute concentrations of about 1.0% by weight.

Equilibrium distribution relationships at the run temperature of 25°C and over the relevant ranges of concentration were measured as follows:

Sys-tem	Relation	C^*_{Ac} range, g mole/ml
1	$C^*_{Ad} = 4.6224 \times 10^5 (C^*_{Ac})^{1.9364}$	10^{-6} - 10^{-5}
2	$C^*_{Ad} = 1.3736 \times 10^2 (C^*_{Ac})^{1.287}$	10^{-6} - 2×10^{-5}
3	$C^*_{Ad} = 2.6343 \times 10^5 (C^*_{Ac})^{2.0241}$	3×10^{-6} - 2×10^{-5}

Distilled water was used and the chemicals were reagent grade, used as supplied. Physical properties were determined as described in the paper by Skelland and Huang (1977), and are listed in Table 1.

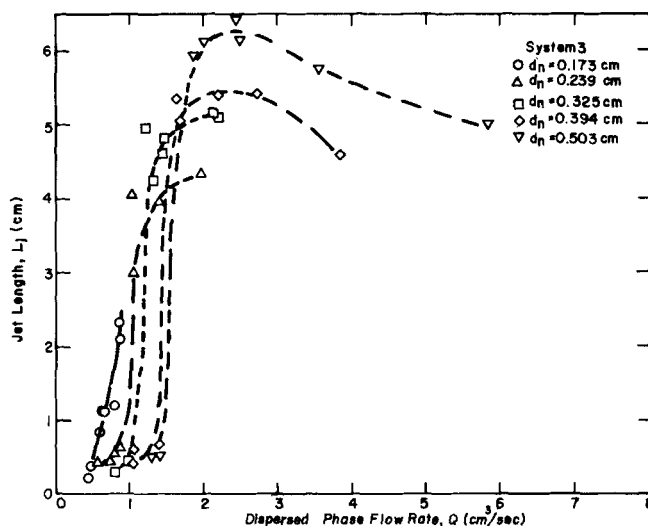


Fig. 4. Jet length vs. dispersed phase flow rate for system 3 (Nujol-carbon tetrachloride)-benzoic acid-water.

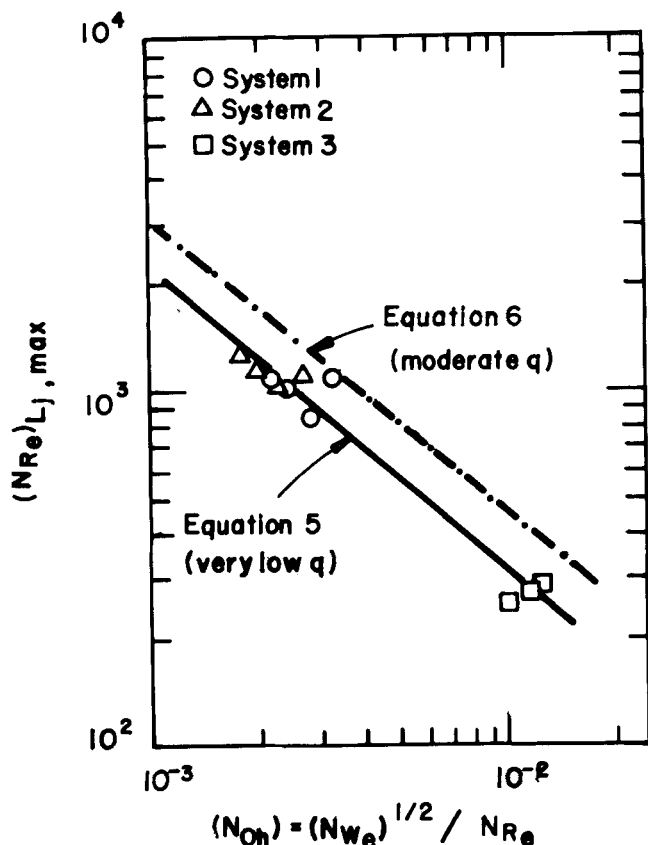


Fig. 5. Correlation of flow rate at maximum jet length. Data from all five nozzle sizes are included here.

Apparatus and Procedure

The jet-forming apparatus has been described in detail in previous papers by Skelland and Johnson (1974) and Skelland and Huang (1977), to which the reader is referred.

Briefly, the dispersed phase was supplied from a calibrated, constant-head reservoir at sufficient elevation to ensure controlled jetting conditions from a single vertical nozzle immersed in the continuous phase. Each nozzle was 60 cm long to provide fully developed velocity profiles, and the inner and (outer) diameters of the five nozzles used were as follows: 0.173 cm, (0.292); 0.239, (0.401); 0.325, (0.495); 0.394, (0.597); 0.503, (0.711).

Two glass columns, with inner diameters of 7 cm and lengths of 32 and 55 cm, were used to contain the continuous phase. Each column had a conical section at its base, tapering to a narrow exit tube with stopcock control, as shown in Figure 1. The shorter column was used to establish the control valve settings on the jetting device so as to instantly provide a jet of the desired length at start-up when the shorter column was replaced by the longer one. All extraction runs were made in the longer column, over a vertical height of about 45 cm. This, of course, involved analysis of the water phase after each run.

Photographs taken during every run enabled determination of the jet length and the number of drops in free fall at any instant. For photographic measurements of drop size and jet contraction, a 19-l aquarium with plane glass sides was used instead of the extraction column.

More details of construction, operating procedure, photography, and analysis are given by Skelland and Huang (1977).

DISCUSSION OF EXPERIMENTAL RESULTS

Jet Length

Figures 2 to 4 show plots of the measured jet lengths vs. dispersed phase flow rates (L_j vs. Q) for all the data. The flow velocity at maximum jet length, exhibited by

many of the curves, is correlated in Figure 5 as $(N_{Re})_{L_j, max}$ vs. N_{Oh} , following the suggestion of Grant and Middleman (1966). The least-squares correlation of these data is

$$(N_{Re})_{L_j, max} = 5.6994 (N_{Oh})^{-0.8663} \quad (5)$$

which contrasts with the following correlation by Skelland and Huang (1977) for data corresponding to substantially higher levels of mass flux across the jet surface:

$$(N_{Re})_{L_j, max} = 10.5514 N_{Oh}^{-0.8197} \quad (6)$$

(average absolute deviation = 13.69%). Equations (5) and (6) are both plotted on Figure 5. Skelland and Huang (1977) worked with dispersed phase controlled systems with high interfacial tension, undergoing mass transfer at moderate rates. This means that their molal fluxes from the jet surface $[k_{Aj}(\Delta C_{Aj})_{avg}]$ were one and sometimes two orders of magnitude greater than in the present work (approximately $k_{cj}\Delta C_A$). To illustrate more quantitatively, consider a particular run from each of the two studies, selected to provide closely similar operating conditions. In both cases, chlorobenzene jetted into water; in the other study, acetic acid transferred from the jet, with $d_n = 0.503$ cm, $U_n = 13.34$ cm/s, and $L_j = 5.01$ cm, resulting in $(N_{A, jet})_{avg} = 0.381 \times 10^{-5}$ g mole/cm²s. The present run substituted benzoic acid transferring from the jet, again with $d_n = 0.503$ cm and $U_n = 13.13$ cm/s, but the resulting L_j was 10.23 cm. The corresponding $(N_{A, jet})_{avg}$ was about 0.386×10^{-7} g mole/cm²s, which is only 1% of the above flux of acetic acid. Skelland and Huang's (1977) jet lengths were correlated as follows, for flow rates up to the maximum jet length,

$$\frac{L_j}{d_n} = 5.0767 (\Delta N_{We})^{0.5499} \left(\frac{\mu_c}{\mu_d} \right)^{0.5245} \quad (7)$$

with an average absolute deviation of 39.69% for 143 data points. The present data in Figures 2 to 4 are correlated, for flow rates up to the maximum jet length, by the same form of expression, namely

$$\frac{L_j}{d_n} = 10.8341 (\Delta N_{We})^{0.5860} \left(\frac{\mu_c}{\mu_d} \right)^{0.5097} \quad (8)$$

using least-squares techniques. The average absolute deviation between Equation (8) and 88 data points was 75.05%.

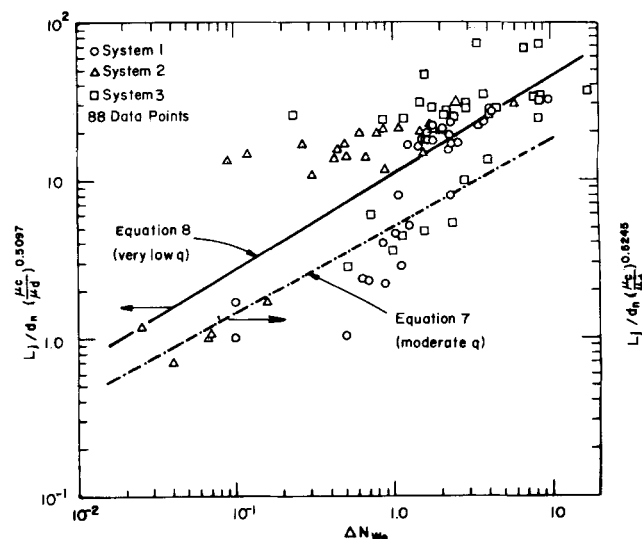


Fig. 6. Generalized jet length correlation for all three systems. Data from all five nozzle sizes are included here.

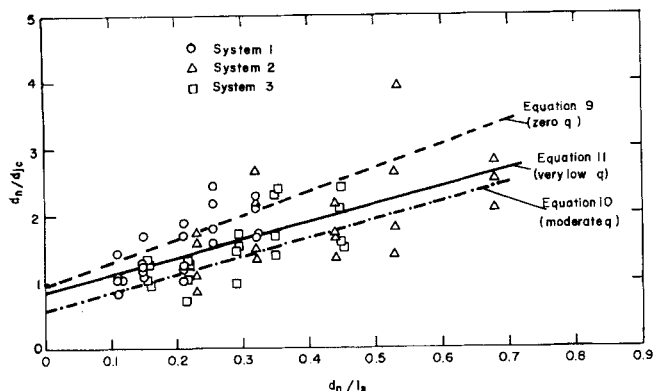


Fig. 7. Jet contraction correlation for all three systems. Data from all five nozzle sizes are included here.

Figure 6 shows a plot of Equation (8) through the present data; results from all five nozzle sizes are included. Also shown, as a dash-dotted line, is a plot of Equation (7). As an example, let $\mu_c/\mu_d = \Delta N_{we} = 1.0$; for this case the jet length under the present continuous phase controlled conditions with very low mass transfer rates [Equation (8)] is roughly double that for the dispersed phase controlled conditions with moderate mass transfer rates [Equation (7)]. This finding is consistent with Skelland and Huang's (1977) Figures 11 and 12 for one of these systems, which showed that, for $Q > Q_c$, the jet length without mass transfer was substantially longer than that with mass transfer. Apparently the present low mass transfer rates, using benzoic acid with a solubility of only 0.2% as solute, gave results approaching the zero mass transfer case where jet length is concerned. In contrast, Equation (7) was established using the infinitely soluble acetic acid as solute, resulting in molal fluxes that were greater by one and sometimes two orders of magnitude.

The use of logarithmic coordinates in Figure 6 reveals that errors in prediction increase as jet length decreases. In this regard, it is important to recall the finding of Christiansen and Hixson (1957), that oscillations in jet length increase with decrease in flow velocity below U_{nm} , defined later. Thus, still photographs of the jet will record some intermediate length between the extremes of oscillation, giving rise to scatter in the data as shown.

Jet Contraction

Figure 7 contains a plot of d_n/d_{jc} vs. d_n/l_s for the complete data from all five nozzle sizes, in the manner of Christiansen and Hixson (1957) and Skelland and Johnson (1974).

In the absence of mass transfer, Skelland and Johnson (1974) correlated their measurements as follows:

$$d_n/d_{jc} = 3.5037 (d_n/l_s) + 0.9491 \quad (9)$$

About 94% of their data were within $\pm 20\%$ of this equation.

For dispersed phase controlled systems with moderate rates of mass transfer, Skelland and Huang (1977) correlated their data as

$$d_n/d_{jc} = 2.7350 (d_n/l_s) + 0.5718 \quad (10)$$

The average absolute deviation was 20.28%.

The present continuous phase controlled data with very low rates of mass transfer are correlated by

$$d_n/d_{jc} = 2.6086 (d_n/l_s) + 0.8495 \quad (11)$$

where the average absolute deviation is 21.57%.

Consideration of the mechanism of jet breakup suggests that the jet diameter in the drop formation region will vary with time between a maximum shortly after drop detachment and a minimum just before detachment. Any still photograph will therefore record a diameter d_{jc} somewhere between these limits, accounting for the scatter in the data of Figure 7. It follows that only estimated average values of d_{jc} are obtainable from Equation (11).

Figure 7 shows the present data plotted with their correlating Equation (11). Also shown are plots of Equations (9) and (10), and there appears to be a directional trend of decreasing ordinate for a given abscissa with increasing rate of mass transfer.

Drop Size from Jet Breakup

Skelland and Johnson (1974) correlated their drop size measurements without mass transfer in a plot of d_D/d_{jm} vs. U_n/U_{nm} . The terms d_{jm} and U_{nm} were calculated from the following equations provided by Treybal (1963):

$$U_{nm} = 2.69 \left(\frac{d_{jm}}{d_n} \right)^2 \left(\frac{\sigma/d_{jm}}{0.5137\rho_d + 0.4719\rho_c} \right)^{0.5} \quad (12)$$

where the ratio of nozzle-to-jet diameter is given empirically by

$$\frac{d_n}{d_{jm}} = 0.485 \left[\left(\frac{d_n}{\left(\frac{\sigma}{g\Delta\rho} \right)^{0.5}} \right)^2 + 1 \right] \quad (13)$$

$$\frac{d_n}{d_{jm}} = \frac{1.51 d_n}{\left(\frac{\sigma}{g\Delta\rho} \right)^{0.5}} + 0.12; \quad \frac{d_n}{d_{jm}} > 0.785 \quad (14)$$

These expressions emerged from Christiansen and Hixson's (1957) results on jet breakup in liquid-liquid systems, which showed that drops assume a minimum size at the nozzle velocity U_{nm} .

The equation correlating Skelland and Johnson's (1974) results on six liquid-liquid systems with five nozzle sizes and without mass transfer is

$$\frac{d_D}{d_{jm}} = 2.6051 - 0.7747 \left(\frac{U_n}{U_{nm}} \right) + 0.3994 \left(\frac{U_n}{U_{nm}} \right)^2 \quad (15)$$

For dispersed phase controlled systems with moderate rates of mass transfer, Skelland and Huang (1977) cor-

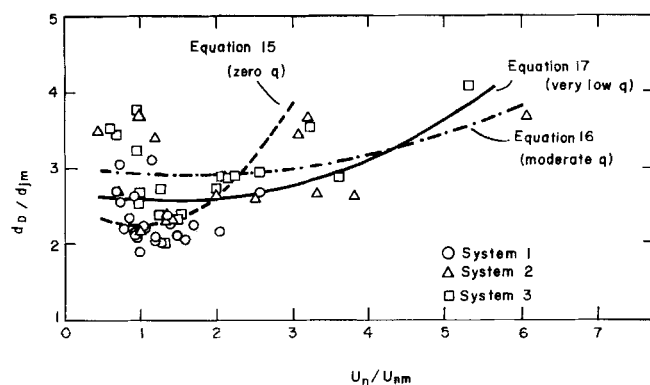


Fig. 8. Generalized drop size correlation for all three systems. Data from all five nozzle sizes are included here.

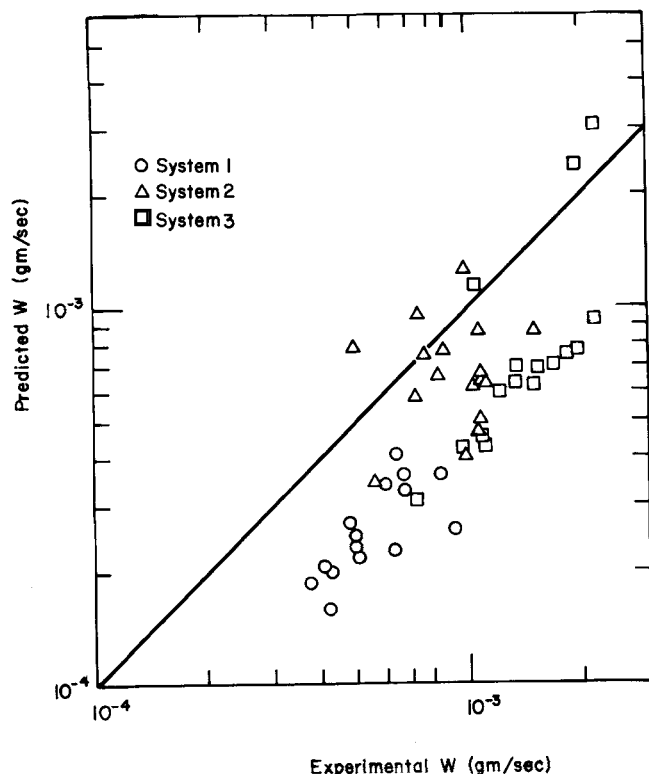


Fig. 9. Predicted vs. experimental mass transfer rates from generalized correlations for systems 1, 2, and 3. Data from all five nozzle sizes are included here.

related their data on four systems with five nozzle sizes as follows:

$$\frac{d_D}{d_{jm}} = 3.0704 - 0.1701 \left(\frac{U_n}{U_{nm}} \right) + 0.0487 \left(\frac{U_n}{U_{nm}} \right)^2 \quad (16)$$

The average absolute deviation was 13.1%.

The present results for three continuous phase controlled systems, five nozzle sizes, and very low rates of mass transfer are correlated by least squares in the following way,

$$\frac{d_D}{d_{jm}} = 2.7270 - 0.2028 \left(\frac{U_n}{U_{nm}} \right) + 0.0731 \left(\frac{U_n}{U_{nm}} \right)^2 \quad (17)$$

with an average absolute deviation of 14.6%.

Figure 8 shows a plot of the present data from all five nozzle sizes and their correlating Equation (17). Plots of Equations (15) and (16) are also shown. In each case, the curves extend only over the ranges of the original sets of data. The two curves with finite mass transfer

are fairly close throughout, whereas that corresponding to zero mass transfer shows a more rapid rate of rise to the right of the minimum. The minima in the plots of Equations (15), (16), and (17) occur at abscissa values of 0.97, 1.75, and 1.39, respectively. Whether the deviation of the last two values from unity is a real consequence of the mass transfer or merely an inaccuracy associated with the scatter in the data is unclear.

Order of magnitude predictions of drop size from all three equations seem comparable over the ranges of variables examined. The complexity of the situation, however, is again indicated by Burkholder and Berg (1974), who conclude that "the wavelength of the most unstable disturbance, (and) hence the ultimate drop size, may either increase or decrease for transfer either into or out of the jet."

Comparison between Predicted and Experimental Mass Transfer Rates

The total rate of mass transfer in the column was predicted for each run using Equation (1), omitting the final term, $K_{cc}A_c\Delta C_A$. This omission was made because the coalescence contribution was rendered negligible by maintaining the coalescence interface in the narrow exit tube of the column (see Lindland and Terjesen, 1956; Skelland and Minhas, 1971).

The mass transfer coefficients were evaluated from Equations (2), (3), and (4), plus appropriate selections from Table II in Skelland and Conger's (1973) paper. Separate experiments with aluminum particles in the disperse phase showed visually that drops of system 1 were internally stagnant, while those of system 2 were oscillating. Some runs of system 3 showed stagnant drops, while in other runs they were oscillating. To distinguish between them, the criterion that drops are stagnant for $N_{we,c} < 3.58$ and oscillating for $N_{we,c} \geq 3.58$ was adopted, following Hu and Kintner (1955).

The interfacial areas were computed as

$$A_j = \pi \left(\frac{d_n + d_{jc}}{2} \right) L_j \quad (18)$$

with L_j and d_{jc} estimated from Equations (8) and (11), respectively. The correlations for k_{cf} and k_{df} are based on the drop surface at detachment, so that

$$A_f = A_D = \pi d_D^2 \quad (19)$$

using Equation (17) to estimate d_D . Also

$$A_r = (N_D)_{avg} A_D \quad (20)$$

where $(N_D)_{avg}$ is the average number of freely falling drops in the extraction column at any instant. This value was known from the photographs taken during a run, but could alternatively be estimated as $(N_D)_{avg} = (H - L_j)/u_{tj}$. Then $W = qM_A$.

The mass transfer rates predicted in this way are com-

TABLE 2. COMPARISON BETWEEN ACTUAL AND PREDICTED NUMBERS OF REAL PLATES REQUIRED FOR SEPARATIONS ACHIEVED UNDER JETTING CONDITIONS IN A PERFORATED PLATE EXTRACTION COLUMN

Run	System	Direction of transfer	No. of plates used	Predicted No. of plates	
				Oscillating drops	Circulating drops
9*	Water-†	To disp. phase	6	4	6
10	Benzoic	To disp. phase	6	4	6
11	Acid-	From disp. phase	6	4	8
12	Toluene	From disp. phase	6	3	7

* Run numbers are those in Table IV of Mayfield and Church (1952).

† In the order disperse phase-solute-continuous phase.

pared with the experimental values in Figure 9. Data from all five nozzle sizes are represented in this plot. The average absolute deviation between the forty-eight predicted and measured values, calculated as $100(W_{\text{exp}} - W_{\text{pred}})/W_{\text{exp}}$, is 46.13%. Figure 9 shows that the predictions tend to underestimate the measurements; this is readily understood in terms of the violent nature of jet breakup. Thus, even the stagnant drops of systems 1 and 3 probably began their free fall existence with some degree of oscillation induced by the breakup process, so that mass transfer estimates based on completely stagnant drops would be on the low side. The oscillating drops of systems 2 and 3 probably experienced enhanced oscillation due to the violence of jet breakup, compared to the nonjetting experiments which led to the oscillating drop correlations used for k_{cr} and k_{dr} .

The drop formation process, too, may be more violent during jet breakup than during formation at nozzles, as in the experiments leading to the k_{cf} and k_{df} correlations used here.

The overall result is that predicted mass transfer rates are on the conservative side, which is a desirable feature for design purposes.

Comparison with the Extraction Column Data of Mayfield and Church

One purpose of the present study was to extend the scope of the design procedure for perforated plate extraction columns recently presented by Skelland and Conger (1973). The Fortran IV program prepared by W. L. Conger to implement the procedure appeared in Skelland (1974); this computer program has now been revised and extended to include operation under jetting conditions by Y-F. Huang, and is available from the AIChE.* It has been applied to the jetting runs made on a high interfacial tension system in a perforated plate extraction column by Mayfield and Church (1952).

Their column was 5 cm in diameter, with a plate spacing of 61 cm, and with fifteen 0.32 cm diameter jets per plate on 0.95 cm triangular pitch. The downcomers were 1.57 cm in diameter. Benzoic acid was transferred between toluene and water in the relevant experiments. Since this system exhibits very low rates of mass transfer, Equations (8), (11), and (17) from the present study were used for prediction of L_j , d_{jc} , and d_p , respectively.

The authors identified their successful runs under jetting conditions with this system as those numbered 9 to 12 in their Table IV. The number of real plates necessary to achieve the separation actually obtained in these four runs was predicted first by assuming that the drops were oscillating after detachment and then by assuming that they were circulating. Table 2 shows that the number of plates actually used is closely bracketed by the predictions for these two forms of droplet condition after detachment.

ACKNOWLEDGMENT

This work was partially supported by National Science Foundation Grant No. ENG74-17286.

NOTATION

A_c = interfacial area during drop coalescence, cm^2
 A_D = surface area of a single detached drop, cm^2
 A_f = interfacial area during drop formation, cm^2
 A_j = interfacial area between the jet and the continuous phase, cm^2

A_r = surface area of all free fall drops at any instant, cm^2
 C_A = solute concentration, g-mole/ml
 C_{Ac}^* , C_{Ad}^* = equilibrium solute concentrations in continuous and dispersed phases, respectively, g-mole/ml
 C_{Adi} , C_{Adj} , C_{Adf} = solute concentration in disperse phase, initial, final, g-mole/ml
 ΔC_A = solute concentration driving force defined by Equation (1), g-mole/ml
 $(\Delta C_A)_{\text{avg}}$ = arithmetic mean solute concentration driving force during jetting, g-mole/ml
 D_c , D_d = molecular diffusivity of solute in continuous and disperse phases, cm^2/s
 d_D = diameter of a single free fall drop, cm
 d_{jc} = jet diameter at breakup, cm
 d_{jm} = jet diameter at breakup at maximum area point as estimated from Equations (11) and (12), cm
 d_n = nozzle diameter, cm
 g = acceleration due to gravity, cm/s^2
 H = vertical distance between nozzle tip and coalescence plane at foot of column, cm
 K_{cj} , K_{cf} = overall continuous phase coefficients of mass transfer during jetting, drop formation, free fall, and coalescence, cm/s
 K_{cr} , K_{cc} = overall dispersed phase coefficients of mass transfer during jetting, drop formation, free fall, and coalescence, cm/s
 k_{cj} , k_{cf} = individual dispersed phase coefficients of mass transfer during jetting, drop formation, free fall, and coalescence, cm/s
 k_{cr} , k_{cc} = individual dispersed-phase coefficients of mass transfer during jetting, drop formation, free fall, and coalescence, cm/s
 k_{dj} , k_{df} = individual dispersed-phase coefficients of mass transfer during jetting, drop formation, free fall, and coalescence, cm/s
 k_{dr} , k_{dc} = individual dispersed-phase coefficients of mass transfer during jetting, drop formation, free fall, and coalescence, cm/s
 L_j = length of liquid jet, cm
 l_s = system length parameter, $\pi\sqrt{\sigma/\Delta\rho g}$
 M = mean molecular weight of phase under consideration
 M_A = molecular weight of solute
 m = slope of the equilibrium curve, dC_{Ad}^*/dC_{Ac}
 $(N_{A,\text{jet}})_{\text{avg}}$ = average molal flux of component A (the solute) across the interface between the jet and the continuous phase, g-mole/ $\text{cm}^2 \text{ s}$
 $(N_D)_{\text{avg}}$ = average number of free fall drops in the extraction column at any instant
 N_{Oh} = Ohnesorge number, $\mu_d/(\rho_c d_n \sigma)^{1/2}$
 $(N_{Re})_{Lj,\text{max}}$ = Reynolds number at maximum jet length, $(U_n)_{Lj,\text{max}} d_n \rho_d / \mu_d$
 $N_{We,c}$ = continuous phase Weber number, $d_D u_t^2 \rho_c / \sigma g_c$
 ΔN_{We} = modified Weber number, $(U_n^2 - U_{nj}^2) d_n \rho_d / \sigma$
 Q = dispersed phase flow rate, cm^3/s
 Q_c = critical value of Q
 q = rate of extraction, g-mole/s
 t_e = time of exposure to mass transfer, s
 t_f = time of drop formation, s
 U_j = jet velocity, cm/s
 U_n = velocity in nozzle, cm/s
 U_{nj} = jetting velocity in nozzle, predictable from Scheele and Meister's expression, cited as Equation (1) by Skelland and Huang (1977), cm/s
 $(U_n)_{Lj,\text{max}}$ = velocity in nozzle at maximum jet length, cm/s
 U_{nm} = velocity in nozzle at maximum area point as estimated from Equation (10), cm/s
 u_t = terminal velocity of a single drop, cm/s
 W = rate of extraction, g/s

Greek Letters

μ_c , μ_d = continuous and dispersed phase viscosities, poise
 ρ = mean density of phase under consideration, g/ml
 ρ_c , ρ_d = continuous and dispersed phase densities, g/ml
 $\Delta\rho$ = density difference, $\rho_d - \rho_c$, g/ml
 σ = interfacial tension, dyne/cm

Supplementary material has been deposited as Document No. 03322 with the National Auxiliary Publications Service (NAPS), c/o Microfiche Publications, 4 North Pearl Street, Portchester, N. Y. 10573 and may be obtained for \$3.00 for microfiche or \$5.00 for photocopies.

LITERATURE CITED

- Burkholder, H. C., and J. E. Berg, "Effect of Mass Transfer on Laminar Jet Breakup. Part II. Liquid Jets in Liquids," *AIChE J.*, **20**, 872 (1974).
- Christiansen, R. M., and A. N. Hixson, "Breakup of a Liquid Jet in a Denser Liquid," *Ind. Eng. Chem.*, **49**, 1017 (1957).
- Grant, R. P., and S. Middleman, "Newtonian Jet Stability," *AIChE J.*, **12**, 669 (1966).
- Hu, S., and R. C. Kintner, "The Fall of Single Liquid Drops Through Water," *ibid.*, **1**, 42 (1955).
- Lindland, K. P., and S. G. Terjesen, "The Effect of a Surface-Active Agent on Mass Transfer in Falling Drop Extraction," *Chem. Eng. Sci.*, **5**, 1 (1956).
- Mayfield, F. W., and W. L. Church, Jr., "Liquid-Liquid Extractor Design," *Ind. Eng. Chem.*, **44**, 2253 (1952).
- Perry, R. H., C. H. Chilton, and S. D. Kirkpatrick, *Chemical Engineer's Handbook*, 4th ed., McGraw-Hill, New York (1963).
- Skelland, A. H. P., *Diffusional Mass Transfer*, Chapt. 8, Wiley-Interscience, New York (1974).
- , and W. L. Conger, "A Rate Approach to Design of Perforated-Plate Extraction Columns," *Ind. Eng. Chem. Process Design and Develop.*, **12**, 448 (1973).
- Skelland, A. H. P., and Y-F. Huang, "Dispersed Phase Mass Transfer during Drop Formation under Jetting Conditions," *AIChE J.*, **23**, 701 (1977).
- Skelland, A. H. P., and K. R. Johnson, "Jet Break-up in Liquid-Liquid Systems," *Can. J. Chem. Eng.*, **52**, 732 (1974).
- Skelland, A. H. P., and S. S. Minhas, "Dispersed Phase Mass Transfer During Drop Formation and Coalescence in Liquid-Liquid Extraction," *AIChE J.*, **17**, 1316 (1971).
- Treybal, R. E., *Liquid Extraction*, pp. 467-8, McGraw-Hill, New York (1963).

Manuscript received December 8, 1977; revision received June 5, and accepted June 22, 1978.

Optimal Design of Multistage Adsorption-Bed Systems

The operation of two arrangements of multistage adsorption beds, parallel and series, was analyzed, and the optimal design problem of minimizing the capital and operating costs of the multistage adsorption bed systems was formulated. Solution of the optimization problem was obtained by using the Fibonacci search scheme. For the purpose of the study, data on the adsorption of liquid on activated carbon were taken from the literature for both single solute and multisolute adsorption systems and was used to derive the expressions for adsorption rate and the equilibrium relationship. Details of the performance of the optimally designed multistage adsorption bed systems are presented in terms of the physical and design parameters.

EDMOND SUNG

and

CHANG DAE HAN

Department of Chemical Engineering
Polytechnic Institute of New York
Brooklyn, New York 11201

and

HYUN-KU RHEE

Department of Chemical Engineering
College of Engineering
Seoul National University
Seoul, Korea

SCOPE

In the present study, we undertook an analysis and an optimal design of multistage adsorption bed systems where the problem involves nonequilibrium adsorption. More specifically, the present study deals with formulating the optimal design problem of single and multisolute adsorption systems, developing a method for solving the system equations for an optimal design of multistage adsorption bed systems, investigating the effect of physical parameters on the optimally designed systems, determining which of the two bed arrangements (parallel or series) yields lower capital and operating costs, and

investigating the effects of tight purification specifications and the size of the adsorption plant on the cost of plant operation.

Examples of wastewater treatment for single solute and multisolute adsorptions were considered, using data available in the literature. Although the specific applications of the present study have dealt with the problem of wastewater treatment, the concepts of the process design and optimization methods demonstrated here are also applicable to other liquid phase adsorption processes as well.

CONCLUSIONS AND SIGNIFICANCE

The optimal design of multistage adsorption bed systems is presented based on a theoretical consideration of transient cyclic operation. The Fibonacci search scheme

has been used to solve an integer programming problem involving a nonlinear objective function. The principal conclusion from this study is that, under optimal design conditions, the serial-bed arrangement is favored over the parallel-bed arrangement, because the leading bed in the serial-bed system can achieve a higher solute saturation than the corresponding one in the parallel-bed sys-

Correspondence concerning this paper should be addressed to Chang Dae Han. Edmond Sung is with Merck & Co., Inc., Rahway, New Jersey 07065.

0001-1541-79-2160-0087-\$01.55. © The American Institute of Chemical Engineers, 1979.