THE UNIVERSITY OF MICHIGAN

INDUSTRY PROGRAM OF THE COLLEGE OF ENGINEERING

ENGINEERING STUDY OF A PERFORATED PLATE PULSE COLUMN

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A dissertation submitted in partial fulfillment
of the requirements for the degree of
Doctor of Philosophy in The
University of Michigan
1960

February, 1960

IP-417

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ACKNOWLEDGMENTS

I wish to thank first of all the members of the doctoral committee in particular Dr. H. A. Ohlgren who initiated this project; Dr. J. T. Banchero for his counsel during the experimental work and Dr. J. L. York whose helpful guidance and brought this thesis into its present form.

I further wish to thank Tom Jameson and John Jackson who helped in the erection and operation of the equipment, Lovi Mathur for supervision during the typing of this thesis and Mamu Tayyabkhan for intellectual stimulation during many a disheartening hour in this period. My appreciations also extend to the Chemical and Metallurgical Office and Shop Staff, to the Staff of the Industry Program and to Carbide and Carbon Chemicals Company for providing the chemicals.

Finally I gratefully acknowledge the financial assistance by the UMRI and The University of Michigan during the course of this research.

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INTRODUCTION

Liquid-liquid extraction has long been used in industry for separation processes. However, its widespread use has been hampered by the poor extraction efficiencies associated with gravity-operated towers. This was inevitable, because in such towers the sole driving force for phase dispersion is the density difference between the two liquids, which is relatively small.

The search for improved extraction efficiencies has produced a new class of liquid-liquid contactors, all of which provide an external means of agitation to aid phase dispersion. The pulse column belongs to this category of liquid-liquid contactors. In the original design, the pulsation was obtained by reciprocating the plates in a perforated plate column in order to force the liquids through the holes. The present practice, however, is to maintain the plates stationary and to pulse the liquids.

Pulsation is characterized by the amplitude, the frequency and the pulse wave form. Of these, the most common and convenient wave form is sinusoidal, which can be generated by the use of an eccentric on a motor shaft. Other wave forms require far too complicated equipment. Hence, only amplitude and frequency are sufficient to define pulsation. Although pulsation improves extraction efficiencies, the mechanism by which it does so is not properly known. Besides, there is no clear measure of pulsation. A simple product, as well as several other combinations of amplitude and frequency, has been suggested and found unsatisfactory. Even the relationships between increase in amplitude, frequency and extraction efficiency are uncertain. In addition to pulsation, the liquid flow rates and column geometry affect the extraction efficiency. Although high flow rates are desirable, their effect on efficiencies is not clear. The conventional correlations do not apply here because of the pulsation.

The problem becomes more complicated because, along with good mass transfer rates, good throughputs are also desirable. Thus, in regard to the plate geometry, although a small hole size or a small percent free area gives smaller droplets and better mass transfer, it also gives small throughputs. Similarly, a close plate spacing prevents drop coalescence but offers a larger resistance to liquid flow rates. Some optimum hole size, free area and spacing must exist. However, it is not clear whether a small hole size with a large fraction free area of the plates is better than a large hole size with a small free area. The drop formation is also a function of the plate-wetting conditions. The same set of plates can give a spectrum of drop size depending on the condition of the plate surface, all other conditions being the same.

Analysis of the system from the first principles becomes too complicated, so all the workers in the field have resorted to empiricism. The effects of the different variables reported by various workers are not in agreement. It should be pointed out that most investigators have studied the effects of a few variables while maintaining the rest at some constant levels. It is necessary to resolve several of the conflicts by studying all the variables to find out the effect of each variable independent of the other variables.

This dissertation was undertaken to clarify these problems. The study was limited to what we consider the region of industrial interest, viz., high liquid flow rates and high mass transfer rates. In this region the effect of amplitude, frequency, plate hole size, spacing, fraction free area, and liquid flow rates has been studied by means of a factorial design of experiments.

LITERATURE SURVEY

One of the earliest types of pulsed columns is that described by Van Dijck (V1). It consisted essentially of a perforated plate tower in which the plates were reciprocated vertically. Since then, however, it has been common practice to hold the plates stationary and pulse the liquid in the column.

Belaga and Bigelow (B2) used a sieve plate column 45 inches long and 1-1/2 inches in diameter. The plates had 1/32 inch diameter holes and 23% free area; they were spaced at 1 inch intervals. Pulsing was carried out by a positive displacement pump with check valves removed. The variables studied were amplitude from 1/8 inch to 2 inches and frequency from 20 - 80 cycles/min. The product of amplitude and frequency, called "pulse volume," was considered as a measure of pulsation and an empirical correlation of H.T.U. and pulsation was obtained. The H.T.U.'s passed through a minimum as the pulse volume was increased. The system used was Methyl isobutyl ketone - acetic acid and water.

Griffith, Jasny and Tupper (G2) studied the separation of cobalt from nickel using methyl isobutyl ketone as the solvent. The continuous phase was a solution of cobalt and nickel chlorides and ammonium thiocyanate in water. They compared pulsed plate columns with spray columns and found higher efficiencies but lower capacities for the plate columns. The H.T.U. values are reported to vary inversely with amplitude and frequency at low pulsation, whereas at high pulsation H.T.U.'s vary directly with frequency, the effect of amplitude being uncertain. The column was 2 inches in diameter and 140 inches long. The plates had 1/32 inch holes and 23% free area. The solvent-water ratio was 1.57 and plate spacing was 1 and 2 inches.

Billerbeck et al (B3) used the system methyl isobutyl ketone-acetic acid-water, to study the effect of frequency on pulsed spray columns. At a constant amplitude, increase in frequency above 200 cycles/min. improved mass transfer. From visual observations they concluded that pulsation causes breakup of large drops and increases mass transfer area, thus increasing the column efficiency.

Feick and Anderson (F1) studied the extraction of benzoic acid and acetic acid from toluene by means of water. The column was 1-7/16 inches in diameter and 36 inches long. Two types of packing, 1/2 inch McMahon saddles and 3/8 inch Raschig rings, were used. Pulsing was achieved by means of a diaphragm attached to the bottom of the column and operated by an eccentric. The amplitude ranged from 1/16 inch to 1/4 inch and frequency from 200 - 1000 cycles/min. Of the two solutes they used, benzoic acid has the larger mass transfer resistance in the water phase, whereas with acetic acid the major resistance lies in the solvent phase. They observed that the increase in extraction efficiencies with agitation is of the same order of magnitude for the two systems. From this they concluded that pulsation increases the interfacial area rather than the mass transfer coefficient.

Cohen and Beyer (C4) used a 1 inch diameter, 20-inch long column with plates having 1/25 inch holes and 9% free area, spaced at 2-inch intervals. Two types of pulsing mechanisms were used -- a bellows driven by a cam and lever and a proportioning pump with check valves removed. The amplitude and frequency range was .1 inch to .6 inch and 17 - 72 cycles/min., respectively. The system was isoamyl alcohol-boric acid-water. The product of amplitude and frequency (pulse volume) was considered as a measure of pulsation. In the range studied, they found that the H.E.T.S. decreases with increase in pulse volume up to a certain point, beyond which there is no effect. The H.E.T.S. values are reported to be insensitive to flow rates and cannot be demarked as a function of the same.

Edwards and Beyer (E1) used an identical column to study the flooding characteristics using a solute-free, methyl isobutyl ketone-water system. A mathematical derivation to describe column operation at the lower limits of pulsation is presented. This operation is referred to as "flooding due to insufficient pulsation." Flooding due to excessive pulsation is reported to be extremely sensitive to plate-wetting characteristics. Straight-line plots are presented to demonstrate the effect of nitric acid washing of the column on flooding rates.

Sege and Woodfield (S1) used a 3-inch diameter, 9 feet long column to study the extraction of uranyl nitrate from nitric acid solution into tributyl phosphate diluted with carbon tetrachloride or hydrocarbon mixtures. The pulse was applied by a stainless-steel bellows or by a reciprocating pump with check valves removed. The range of amplitude was 0 - 1.5 inches and that of frequency 0 - 200 cycles/min. The product of amplitude and frequency was considered the "simplest though not complete" measure of pulsation. They found that in general H.T.U.'s pass through a minimum as pulse volume is increased, although the behavior may vary from system to system. H.T.U. was also found to pass through a minimum as flow rates were increased. H.T.U.'s are reported to be a function of concentration of the solute, being higher at the dilute end. H.T.U.'s are also sensitive to plate-wetting characteristics and dual plates with a plastic (wetting the organic) and stainless steel (wetting the water) on proper faces are reported to be better than single-material plates both for flooding and mass-transfer rates. A 2-inch plate spacing was found to be optimum, although Wiegandt and Von Berg (W1) claim that closer spacing is more advantageous. The mass-transfer rates improve with small hole size, whereas the capacities decrease. A 1/8 inch hole diameter was considered optimum. Likewise, 23% free area was considered best.

Burger and Swift (B4) studied the effect of amplitude, frequency, flow ratios and plate spacing on backmixing in a pulse plate column. The column was 2 inches in diameter and the plates had 1/8 inch holes with 24.5% free area. The technique was to introduce a tracer solution into the continuous phase at the bottom of the column and to sample the solution along the height of the column. A stainless steel capillary is inserted through the plate perforations. A fluorothene capillary is attached to the stainless steel and the sample is withdrawn by siphoning.

Backmixing was insensitive to plate spacing, flow rate of the dispersed phase and frequency. It increased with increasing amplitude and with decreasing continuous phase flow rate. Backmixing effect is supposedly high for high values of the extraction factor (L/MV or MV/L). For the minimum value of the extraction factor, backmixing accounts for 24% of the height of a transfer unit. Backmixing diffusivities and the half height (of the column in which the tracer concentration becomes half of initial concentration) are reported.

Burkhart and Fahien (B5) used a 1 inch diameter perforated plate pulse column having a contacting section of 18-3/8 inches. The plates were stainless steel, electropolished to a bright luster. The system was Methyl iso-butyl ketone, acetic acid and water, the transfer taking place from the aqueous to the organic phase.

They performed a 2^5 factorial experiment, keeping the liquid flow rates constant at 36 g/min. each and studying the following variables at two levels each: amplitude .25 and 1 inch, frequency 25 and 100 cycles per min., hole size 1/32 and 1/16 inch, percent free area 13 and 25, and plate spacing 1 and 2 inches. The results were interpreted by an analysis of variance.

The hole diameter was found to have the greatest effect on the extraction efficiency whereas the percent free area was found to have the least effect. Increasing amplitude and frequency decreased the extraction efficiency in spite of the decrease in drop size. This effect was explained by the "recycle or backmixing" present in the pulse column. The backmixing was postulated to have a profound effect on the extraction efficiency, large enough to mask the increase in interfacial area due to the smaller drop size.

The five factors studied showed three two-factor interactions between amplitude-plate spacing, amplitude-percent free area and amplitude-hole diameter. These interactions were not interpreted because of the varying backmixing present in the runs.

Chantry, Von Berg and Wiegandt (C2) studied packed, as well as plate pulse column with a 20 mm diameter, 4 foot long column. The packed column contained 1/4 inch Raschig rings, whereas the plate column had 11 plates spaced at 3-inch intervals with 24 holes in each plate. Two hole sizes, 3/64 and 5/64 inch, were studied. Two types of pulsers, a copper bellows driven by a cam and a proportioning pump, were used. Two systems -- methyl isobutyl ketone-water and ethyl acetate-water -- were used with acetic acid as the solute. Most of the runs were made with the former system. Amplitude and frequency ranges were 0 - 10 mm and 0 - 300 cycles/min., respectively. The flow rates in most cases were about 140 gallons/sq.ft.hr.

At constant flow rates for the packed column, the H.E.T.S. values go through a minimum with increased amplitude and frequency. The product of amplitude and the square of frequency gave the same H.E.T.S. and hence may be considered as a measure of pulsation. The H.E.T.S. is unaffected by acid feed rate, whereas it varied with the organic feed rate raised to .37 power. Low amplitudes and high frequencies are reported to be advantageous from the viewpoint of power consumption.

For the sieve-plate column, the maximum plate efficiencies are reported to be 70% for 3/64 inch hole size compared to 30% for 5/64 inch holes. It may be noted that this effect may be due to the 2-1/2 times increase in fraction free area in addition to that of increased hole size. The small hole size gave inconsistent results because of clogging and erratic flooding at some of the plates. The best operation for sieve plate columns is reported to be one with high holdup and high turbulence. These conditions correspond to high pulsation and high flow rates. The sieve plate columns are inferred to have higher throughputs than the packed columns.

Thornton (T2) presents a generalized correlation for the flooding rates and for mass transfer data on a 3-inch diameter pulsed plate column. The correlation is a slight modification of the one for the rotary disc contactor, presented in a previous article of the series (L2).

The variables investigated are the plate spacing, hole size, percent free area and the pulse wave form. Flooding rates were determined for six solvent-water systems and mass transfer data for both direction of transfer were obtained for two systems.

The correlation is essentially based on two dependent variables introduced by the author. One is the "characteristic velocity" -- $\overline{V}_o$ and the other, "the power function" -- ψ .

The characteristic velocity $\overline{V}_o$ is defined as the velocity of the droplets at the limiting condition when the velocity of the continuous phase is zero.

The term ψ is the maximum power dissipated by fluid friction in the plate perforations per unit mass of the fluid. It is evaluated from the orifice equation. ψ is thus a function solely of the pulsing conditions, i.e., the amplitude, the frequency and the pulse wave form and of the column geometry, i.e., the plate spacing and the percent free area. These variables do not affect the characteristic velocity $\overline{V}_o$ except through the function ψ.

The ultimate drop size is postulated to be determined by a balance of viscous and turbulent forces, the physical properties of the system, the hole size of the plates and the rate of energy dissipation. These factors determine the characteristic velocity $\overline{V}_o$. The final correlations are obtained by a dimensional analysis of the system. The final expressions are as follows:

For flooding:

$$\frac{\overline{V}_o \mu_c}{\gamma} = 0.60\left(\frac{\psi \mu_o^5 g_c}{\rho_c \gamma^4}\right)^{-.24} \left(\frac{h \rho_c \gamma}{\mu_c^2}\right)^{.9} \left(\frac{\mu_c^4 g}{\Delta\rho \gamma^3}\right)^{1.01} \left(\frac{\Delta\rho}{\rho_c}\right)^{1.8} \left(\frac{\mu_d}{\mu_c}\right)^{.30}$$

For mass transfer:

$$\frac{HTU)_{oc}}{(\mu_c^2/g\ \rho_c^2)^{1/3}} = K\left(\frac{\mu_c\ g}{\overline{V}_o^3(1-x)^3\rho_c}\right)^{\frac{2m}{3}}\left(\frac{\Delta\rho}{\rho_c}\right)^{\frac{2(m-1)}{3}}\left(\frac{V_d}{V_c}\right)^{.5}\left(\frac{V_c^3\ \rho_c}{g\ \mu_c\ x^3}\right)^{1/3}$$

where

μ is the viscosity

γ is the interfacial tension

ρ is the density

h is the plate hole size

g is the force of gravity

x is the fraction hold up

g_c is the conversion factor

and subscripts "c" and "d" refer to the continuous and the dispersed phase respectively.

For design purposes the constant "K" and the exponent "m" must be determined from model tests with the given system in the laboratory column. For the direction of transfer from water to solvent, $\overline{V}_o$ may be calculated from the preceding equation. For the reverse direction of transfer, the $\overline{V}_o$ is different and an enhancement factor must be determined before applying the correlation.

The correlation involves the holdup for which no expression is derived. Since the holdup changes with the pulsing conditions and column geometry, it seems that the correlation needs a large number of trial runs in a laboratory column for proper design applications.

In a subsequent article, Logsdail and Thornton (L3) present the effect of column diameter on the flooding rates and mass transfer rates.

They report that the flooding rates vary linearly with the cross sectional area of the column, whereas the H.T.U.'s increase with increasing diameter by a factor of exp. D/2. The application of the results to the design of pulse columns is discussed in the article.

In a more recent article, Thornton (T1) discusses the application of the preceding correlations to the design of pulse plate columns. He emphasizes the "Enhancement factor," (which is the ratio of the experimental to calculated values of $\overline{V}_o$) caused by the direction of mass transfer. This effect, which is claimed to be constant, is about 10 - 15% smaller for the water to solvent direction of mass transfer and about 200 - 300% larger for the reverse direction as compared to solute free systems.

Callihan (C1), using the system carbon tetrachloride-acetone-water, studied the flooding and mass transfer rates in terms of pulsation in a packed column.

At low pulse rates the product of amplitude and frequency has a determining effect on H.E.T.S., i.e., the amplitude and frequency affect the H.E.T.S. to the same degree. However, at high rates, frequency is more beneficial than amplitude. Under certain pulse conditions (a = 5 mm f = 216 cycles/min), the H.E.T.S. was 14 times lower than an unpulsed run. Increase in column diameter from 2 inches to 3 inches had no effect on H.E.T.S. if the packing, pulsing and superficial flow velocities were the same.*

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* Logsdail and Thornton (L3) report that H.T.U. varies with exp. D/2.

Presence or absence of the solute (1% acetone) predominantly affected flooding rates. With transfer from the organic to the aqueous phase, the flooding rates were twice as high as those in absence of the solute.* When the water to carbon tetrachloride flow ratio was increased, flooding rates were lower, presumably because of a composition pinch at the bottom of the column. Increase in column height decreased the flooding rates, the effect being attributed to decreased mass transfer per unit height. The effect of other variables on flooding was difficult to measure since their influence was much smaller than that of the solute.

From the design viewpoint, the author doubts the validity of flooding data taken in absence of mass transfer.

Li (L1) studied the effects of pulsation and flow rates on the mass-transfer coefficient in a 2 inch diameter, 20 inches long perforated plate column. The plates were made of brass with a hole diameter of 1/16 inch and a free area of 8.51%. The system was toluene-benzoic acid-water.

The amplitude and frequency were incorporated into a Reynolds number having the hole diameter as the length characteristic. The viscosity and density of the mixture were determined from an average holdup. The product of the amplitude and frequency (which has the dimensions of velocity) based on the fraction free area of the plates constituted the velocity term. The Reynold's analogy between momentum and mass transfer is confirmed by comparing the variation of the pressure drop and the "overall mass transfer coefficient K_wa" with Reynolds number.

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* This is quite in accordance with the observations of Thornton (T1).

Although the independent variable is in the form of a Reynolds number, it is in effect the product of amplitude and frequency, since the plate geometry was kept constant. In addition, the "$K_w a$" is shown to vary the same way as the "pressure drop" through the plates. We may therefore conclude that the interfacial area and the mass transfer coefficient are affected to the same degree by pulsation since the mass transfer coefficient "K" and the friction factor are related by the j factor.

Note: The use of Reynolds number as presented by Li is basically unsound. If we increase the hole size, while keeping the amplitude, frequency and free area constant, the Re No will increase and should give a high "$K_w a$". In practice, this effect is just the opposite.

Crico (C5) presents a mathematical derivation to correlate mass transfer data in a packed pulse column. The method separates the mass transfer coefficient "K" from the interfacial area "a" without the direct measurement of either variable.

The basic assumptions are that the droplet diameter is determined by the equilibrium between the interfacial energy and the mean droplet energy. In the pulse column, mean droplet energy is provided by pulsations across the continuous phase to the drops by friction. The droplet diameter therefore becomes a function only of pulsation, packing characteristic and the physical properties of the system. It is independent of liquid flow rates. The superficial liquid velocity of the drops is tied to the absolute velocity by the holdup. The absolute droplet velocity for a given drop size is determined by Stoke's law.

With these assumptions, an expression is derived to calculate the interfacial area. The mass transfer coefficient may be calculated from the value of "Ka" and "a".

The author points out that this method of separating "K" and "a" has never given good results with conventional columns. The relations are applicable only to pulse columns where pulsations are large enough to make the dispersed phase a suspension of small uniform droplets clearly separated from the packing. The relations have not been tested with experimental data.

Eguchi W. and Nagata S. (E2) studied the effects of longitudinal mixing in pulse columns, using the system methyl isobutyl ketone-acetic acid-water. They used a 58 mm diameter, 550 mm long column containing 9 plates at 52 mm spacing, 1.5 mm hole size and 8.1% free area. The variables studied were the amplitude, the frequency and the liquid flow rates and ratios.

They measured the variations of concentrations along the length of the column by sampling the two phases immediately above and beneath each plate. The backmixing diffusivity is correlated with the operating variables. They show that after eliminating the effect of backmixing, the overall mass transfer coefficient can be correlated with the function $\frac{\text{"af"}}{Vc}$ where "a" is the amplitude, f is the frequency and Vc is the flow rate of the continuous phase.

Shirotsuka et al. (S4) studied the mechanism of mass transfer in a packed pulse column, using two systems -- water-uranyl nitrate-ethyl ether and water-acetic acid-benzene. They correlated the holdup with liquid flow rates in terms of the drag coefficient of the liquid droplets. Measuring the drop size, they calculated the interfacial area.

The interfacial area was found to increase with pulsation up to a certain point, beyond which it remained constant. Pulsation is reported to affect the interfacial area far more than the mass-transfer coefficient. Increasing the height of the packed section increased the H.T.U. values, from which the authors infer that the mass-transfer coefficient is abnormally high at the bottom of the column.

Shirotsuka and Ōya (S5) compared pulsed and unpulsed behavior of packed, plate and spray columns, with benzene-acetic acid-water as the system. The amplitude range was .1 inch to .5 inch although they used .1 - .2 inch for most of the runs. Frequency ranged from 100 - 200 cycles/min. and the flow rate velocities were fixed at 8 meters/hr. for the continuous phase and 1.45 meters/hr. for the dispersed phase.

The plates used had 0.08 - .16 inch hole size and 1% to 22% free area. The hole size and percent free area were changed simultaneously, keeping the number of holes constant.

The results are correlated using a^2f^3/p^2 as the independent variable and "Ka)pulsed/Ka)unpulsed" as the dependent variable. Flooding rates are reported for some pulse conditions. Some disagreements with Pratt's (P2) correlation of characteristic velocity are noted.

DESIGN OF EXPERIMENTS

"Experiment" - Webster's dictionary defines the word experiment as "a trial or special observation made to confirm or disprove something doubtful, especially one under conditions determined by the experimenter; an act or operation undertaken to discover some unknown principle or effect or to test, establish or illustrate some suggested or known truth." To define this "something doubtful," we only need to go back to the literature survey.

Belaga and Bigelow (B2) were the first to use the product of "amplitude" and "frequency" as a single variable, calling it the "pulse volume." They found that H.T.U. values went through a minimum with increase in pulse volume. A scrutiny of their work, however, shows a significant scatter in data. Griffith et al. (G2) found that at high pulsation, the H.T.U. values increase with frequency, but that the effect of amplitude is uncertain. Cohen and Beyer (C4) found that H.E.T.S. values decrease with the increase in pulse volume (a x f) up to a certain point, beyond which they remain constant. Sege and Woodfield (S1) inferred that the pulse volume is a convenient and simple measure of pulsation, though incomplete. They felt that $(a \times f)^n$ may be a better variable. They agreed with Belaga and Bigelow (B2) that H.T.U. values pass through a minimum with increase in pulse volume, although the behavior varies from system to system. Chantry (C2) found that H.E.T.S. values go through a minimum with increase in amplitude at constant frequency. The behavior is the same when frequency is varied at constant amplitude. From a cross plot of their data, they found that H.E.T.S. is a function of the product of amplitude and the square of frequency. We may then conclude that although the different authors agree on the effect of frequency on H.T.U., they differ on the effect of amplitude and on the use of (a x f) to define the pulse characteristics. It is significant to note that most workers have maintained the other variables, such as liquid flow rates, plate hole size or packing characteristics constant in each series of runs. However, the constant variables are at different levels with different workers. The data are obtained on different systems. If the effect of one variable depends on the levels of the other variables, then their conclusions may be expected to differ.

The effect of liquid flow rates on the H.T.U. is well studied for conventional liquid-liquid extraction columns. The two-film theory is generally accepted and the H.T.U.'s are correlated in terms of the extraction factor ($\frac{MG}{L}$ or $\frac{L}{MG}$). Griffith et al. (G2) report that for pulsed-plate columns, H.T.U. values increase with flow rates whereas for spray columns the trend is just the opposite. Feick and Anderson (F1) report that under certain pulsing conditions, the H.T.U. can be made independent of the dispersed flow rate. Cohen and Beyer (C4) found that H.E.T.S. values are insensitive to flow rates and cannot be clearly demarked as a function. However, the flow ratios were correlated in terms of the extraction factor. Thornton (T3) confirmed the two-film theory. For a corrugated plate column, $HTU)_{oc}$ increases with liquid flow rates up to 50% of the flooding rates and from then on remain constant. Sege and Woodfield (S1) used the sum of both phases as the variable and report that $HTU)_{oc}$ passes through a minimum as the flow rates increase. However, the effect of flow ratios on $HTU)_{oc}$ was observed only when the plates were wetted by the continuous phase. Chantry (C2) using a packed column, found that H.E.T.S. values increase with increase in organic flow rate, but were independent of the aqueous flow rate. Billerbeck et al. (B3) using a spray tower, found that $HTU)_{oc}$ increased with increasing aqueous flow rate, whereas $HTU)_{od}$ decreased, regardless of pulse frequency. Thornton (T2) has offered a correlation of $HTU)_{oc}$ as a function of "characteristic velocity," flow ratios and the flow rate of the continuous phase.

Thus in the case of flow rates, we again see uncertainties in their effect on mass-transfer rates. The data of different workers cannot be compared directly for the same reason given in the discussion of amplitude and frequency. However, "figures do not lie." Probably we see only a part of the picture. It is necessary that the picture be viewed as a whole.

The Role of Statistics

The use of statistics in experimentation is not new. The method of least squares, which is so frequently used in engineering practice, is basically a statistical tool. This simple technique is applicable to most cases where the error due to random effects is small. In other cases, where the functional relationships are not clear, one has to choose some criteria to define the system. Mathematical statistics provide just such criteria. It imposes a numerical discipline and thus eliminates the bias of the experimenter's judgment.

Apart from this, the theory of statistics can be used to estimate population characteristics from samples. To achieve this, one must formulate a hypothesis about the population and verify it from analyses of the samples. This verification requires systematic collection of data and the design of experiments is essentially the pattern of the data to be collected. Data obtained to test one hypothesis may be quite useless to test another. In other words, data, unless taken in a particular fashion, cannot be submitted to any simple statistical analysis -- as is the case in most of the data in pulse-column literature.

It may then be said that the formulation of a hypothesis, the design of experiments to test the hypothesis and the analysis of the variance of data constitute the role of statistics in experimentation.

Formulation of a Model

The flow patterns in a pulse column are so complex that an analysis of mass transfer mechanism from the first principles is a phenomenal task. Even if one were to do it, the resulting expressions would be too complex to be of any practical use. Empirical analogy with momentum transfer can give erroneous models such as the one presented by Li (L1). It is even doubtful whether the individual variables or their combinations affect the overall mass-transfer coefficient. Such doubts can be resolved by a polynomial representation of the response surface as follows:

$$y = c_o + c_1x_1 + c_2x_2 + \dots + c_{11}x_1^2 + c_{22}x_2^2 + c_{12}x_1x_2 + \dots + \epsilon$$

where

y = dependent variable, in our case the $HTU)_{od}$

c's = regression coefficients

x's = individual variables

ϵ = experimental error.

With such a model, if the effect of a factor x_1, on the response y is linear, only one term, i.e., c_1x_1 will adequately describe that effect. If the effect is nonlinear, say parabolic, then the use of two terms, i.e., $c_1x_1 + c_{11}x_1^2$ is sufficient. If the effect of one factor is dependent on the level of other factors, the term $c_{12}x_1x_2$ will describe this "interaction." Whenever an individual effect is small as determined by a statistical test, the particular term can be dropped from the polynominal expression and included in the error term. We feel that such a model is simple in itself and yet incorporates the relationships between different variables and their effects on the overall mass-transfer coefficient. Correlation by this method is called the "multiple correlation method" in the field of statistics.

Factorial Design

When a study involves a large number of variables, it is necessary to assess the effect of each variable independent of the others. The traditional method of changing one variable at a time is quite unsuitable for this purpose. The proper way to achieve this objective is to arrange the variables at different levels of interest and experiment on all of them simultaneously, i.e., to move in jumps rather than in steps. In a complete factorial design, all the combinations of the factors are tested at all the levels of interest. In other words, we form a grid of the variables and their levels and test every point on this grid. By this method, we can not only determine the effects of each variable independent of the others but also find the manner in which the effect of each variable depends on the levels of the other variables. Such effects are called "interactions." In addition, designs of this type provide their own estimate of the experimental error.

Analysis of Variance

Statistical analysis of factorial experiments is centered around one statistic: Variance. This is defined as the "sum of squares of the deviations of each observation from the sample mean, divided by the number of degrees of freedom." It is also referred to as the "Mean Square." "Standard deviation" or "Standard error" is the square root of the "Variance." Statistical analysis provides us with the variance of each "variable" and of each "interaction."

To determine whether these variances are real, we compare them with the variance of the experimental error using the "F" test or the "Variance Ratio Test." The F-distribution is used to calculate the probability that two independent estimates of variances of a single normal universe shall have any particular ratio. The distribution depends only on the number of degrees of freedom associated with each estimate of variance. The common confidence limit for such tests is 95%. If the ratio of the variance of a treatment and that of the error exceeds that predicted by the F test, then that variance is real and may not be called the experimental error.

The experimental error can be calculated from theoretical knowledge or from a previous set of experiments. When this is not available, the higher-order interactions may be used to estimate the error. This is justified on the grounds that higher-order interactions are small or negligible. On this assumption, the mean squares of the higher-order interactions are added to give an estimate of the experimental error based on a large number of degrees of freedom. The hypothesis that certain interactions may be so used can be tested by Bartlett's criterion (B1).

In our case all but one of the factorial subsets are four factors designs. We assumed that the three and four factor interactions were negligible and used them to get an estimate of experimental error. Calculations justified this assumption and satisfied the preceding criterion.

The Field of Interest

The range of variables within which a perforated plate pulse column can operate is very large. A study of the entire range would involve a sizeable number of experiments. To get a good picture of column behavior within a limited time, it is necessary to define a certain region of interest. We selected the range of industrial interest, which would involve high throughputs and high mass-transfer rates. This is supported by Chantry's (C2) conclusion: "The best operation of a pulsed sieve-plate column is obtained with a combination of high hold up and sufficient turbulence -- feed rates must be maintained at a high enough level for sufficient hold up." On our laboratory equipment, the following range of variables was found to be convenient.

Liquid flow rates:	660 and 1000 gal/hr.sq.ft.(sum of both phases)
Amplitude :	0.21, 0.423 and 0.62 inch
Frequency :	30-100 cycles/min with infinite variation
Hole size :	1/32, 1/16, 3/32 and 1/8 inch
Percent free area of the plates :	16, 24.55, 32.7 and 40

However, many of the combinations of the above variables needed for a complete factorial experiment were either not available, or not operable due to flooding, or fell outside the range of interest, i.e., gave large drop size, low holdup and low mass transfer rates.

Thus, for example, with 1/32 inch hole size, the plates could not physically be drilled to more than 24.5% free area. Moreover the frictional resistance of these plates was too high to permit operation with even the low flow rates. Similarly with 1/8 inch hole size the plates could not be drilled to 40 per cent free area. Further, operation with 1/8 inch hole sized plates gave large drops, low holdups and low mass transfer rates. These ranges of hole size were subsequently discarded and the per cent free areas for the factorial experiments narrowed to the 24.55 and 32.7 per cent range.

Probably a bigger problem was the column operation with all the combinations of amplitude and frequency. With the preceding range of hole size and per cent free area, only the following combinations of amplitude and frequency were available. With 0.21 inch amplitude, the frequency range was 60-100 cycles/min. With 0.423 inch amplitude, it was 40-80 cycles/min. and with 0.62 inch amplitude, it was 30-60 cycles/min. Below these respective ranges of frequency, pulsation was insufficient for column operation whereas above the high values, flooding occurred due to excessive pulsation.

With these problems of operation and restrictions on the range of variables, a full factorial experiment becomes impossible. Fractional factorial experiments could not be carried out since no prior information on the interactions was available. The experiments were therefore conducted with a few sets of variables at a time and completing factorial designs of these subsets. Such a procedure has an advantage that information gained at each step can be used in further work, without having to wait for the entire set of experiments. As an example, initially, amplitude was tested at three levels in a 3 x 2 x 2 - amplitude, hole size and percent free area -- factorial subset. No points of inflection were noticed and in the rest of the work the intermediate level of amplitude was dropped. These factorial subsets and the analysis of their variance are presented in Tables 6.1 to 6.9.

From the information gained from these subsets, several data were obtained in the boundary regions of those variables before correlating them by the polynominal model.

EXPERIMENTAL SETUP

Column

The column consisted of a 30-inch precision-bore glass tube with flanges at both ends. The overall length of the tube was 34 inches, which provided a contacting section of 32 inches. The inside diameter was 1.000 ± .002 inches. The calming sections were 3-inch ID glass pipe with a 1-inch side outlet.

The feed solutions were introduced from the top and from the bottom by a 3/8-inch stainless steel tubing extending to 3 inches from the contacting section. No feed distributors were used. The solvent was withdrawn from the side outlet at the top. The aqueous raffinate was withdrawn from the bottom via a jack leg used to control the interface. The pulse line was connected to the side outlet at the bottom.

A sketch of the setup is presented in Figure 1, and a photograph in Plate 1.

Plates

Stainless steel and brass plates were used. The plates were cut from a 20 gauge sheet and machined so that the clearance from the column wall was less than .005 inches. They were supported on a 1/4-inch stainless steel center rod and kept in position by spacers made from 3/8-inch stainless steel tubing. All sets of plates were drilled to furnish the specific hole size and percent free area. All holes were on a triangular pitch.

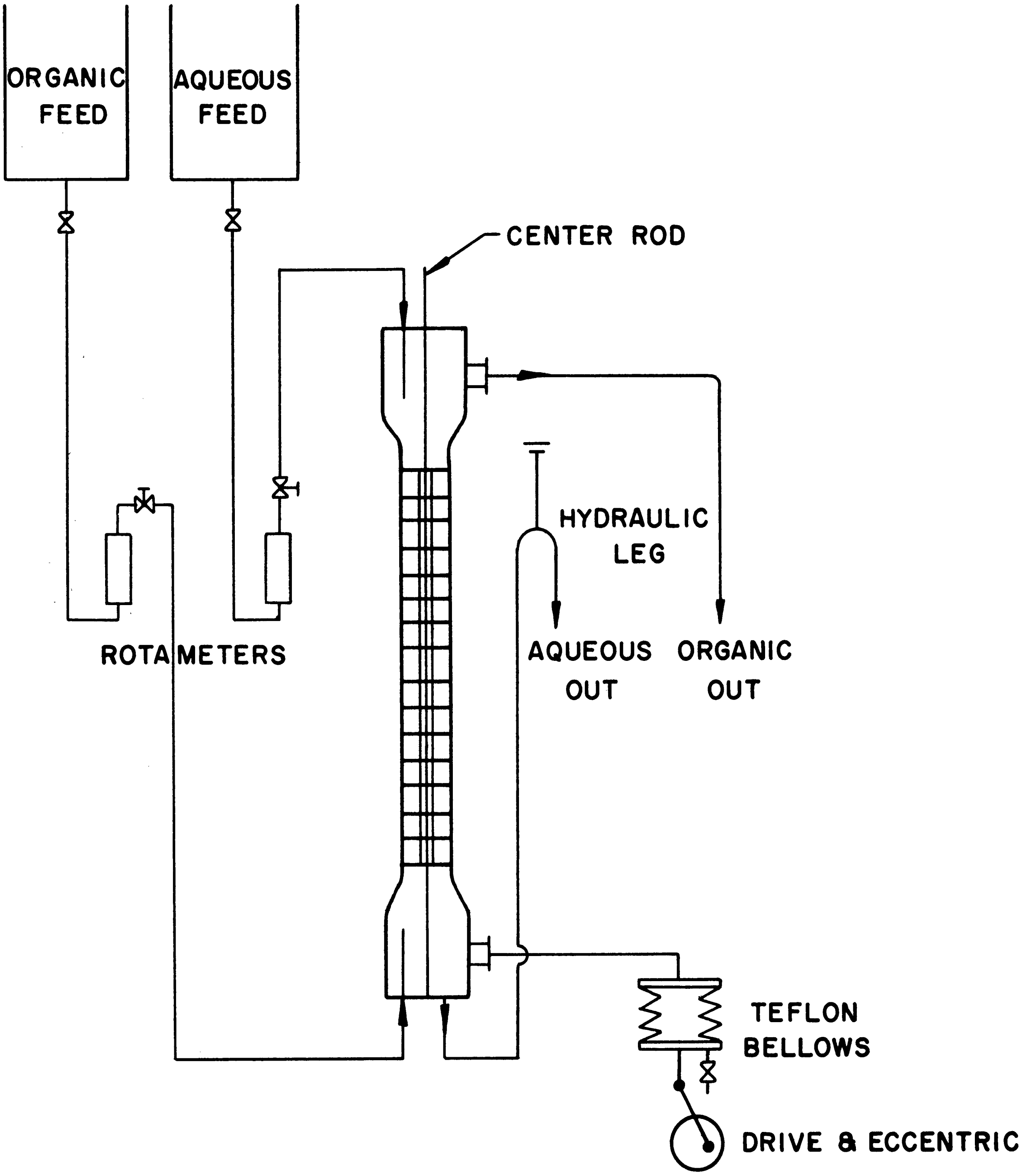


Figure 1. Schematic Diagram of the Equipment.

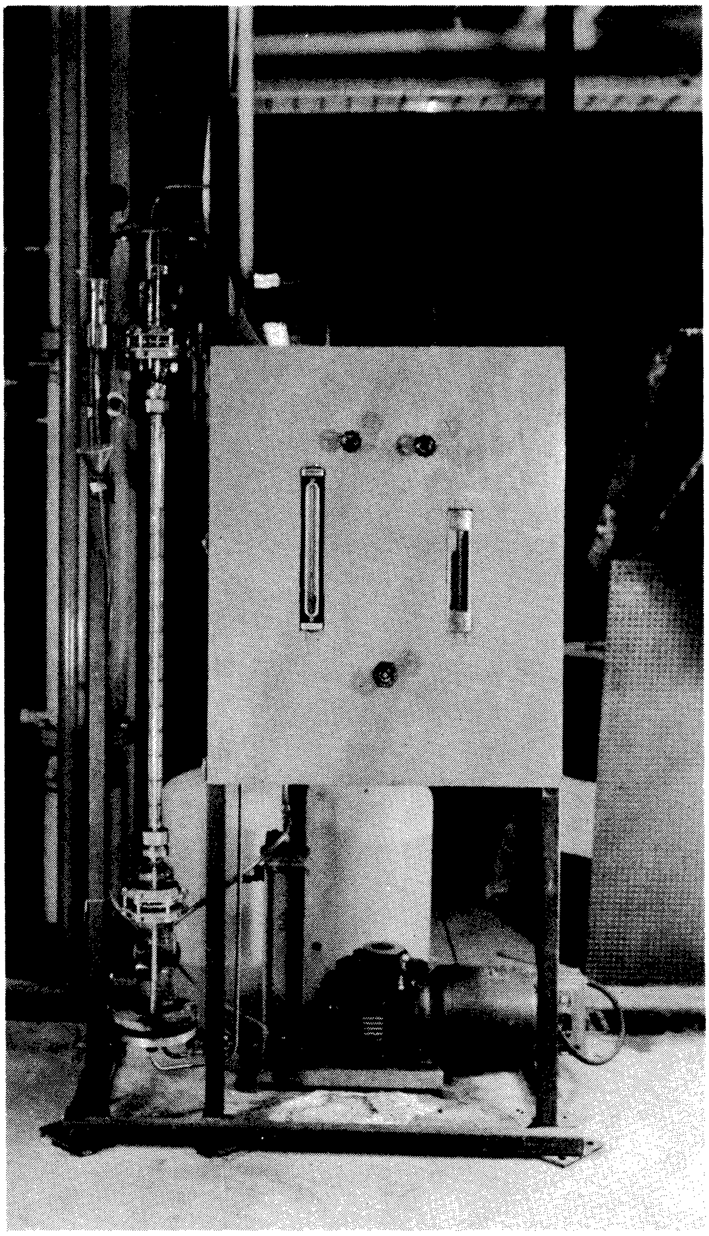


Plate 1. General View of the Equipment.

Pulser

The pulser consisted of Teflon bellows driven by a Graham motor with a built-in speed reducer. Two Teflon bellows, one 1-inch and the other 1-1/2 inches were used. The infinitely variable transmission provided a frequency range of 0 - 350 rpm. The amplitude was controlled by an eccentric attached to the shaft of the motor. A stepwise change of amplitudes from about 1/4 inch to 2.5 inches was achieved by the use of different eccentrics and proper bellows.

A photograph of the pulser is shown in Plate 2.

Feed Tanks

Two stainless steel 55 gallon drums served as the feed tanks. Advantage was taken of gravity feed by locating the feed tanks about 6 feet above the top of the column. Polyethylene and stainless steel tubings conveyed the feed solutions to the column. The flow rates were controlled by needle valves and read by rotameters. All equipment coming in contact with the solutions was made out of stainless steel or Teflon.

The System

The system used was methyl isobutyl ketone as the solvent, acetic acid as the solute and water. The direction of solute transfer was from the aqueous phase into the solvent phase. The solvent and the solute were provided by Carbide and Carbon Chemicals Company, S. Charleston, West Virginia.

This system afforded several advantages. The ketone is inexpensive, has a high density difference compared to water, and has been

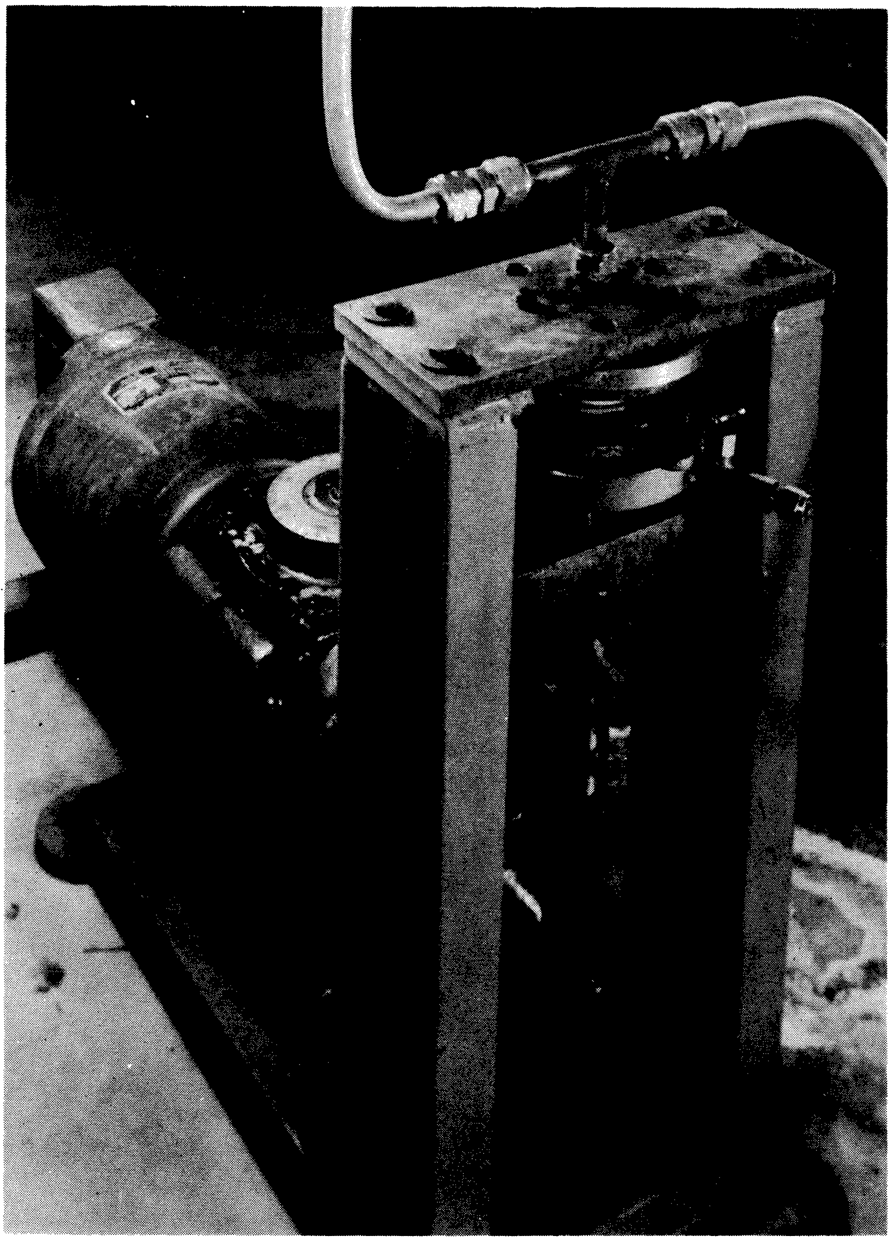


Plate 2. Pulser.

used by several investigators in pulse columns and in classical mass-transfer investigations. The analysis of the solutions is simply an acid-base titration.

The properties of the system are given in Appendix D.

Equilibrium Data

Sherwood et al. (S3) have reported the equilibrium, mutual solubility and density data on the system methyl isobutyl ketone-acetic acid-water. These data cover the concentration range of 0 - 35% acid in the water phase. Belaga and Bigelow (B2) report equilibrium data on the same system for the concentration range of 0 - 16.5% acid in the water phase. Seltzer (S2) presents similar data for the concentration range of 0 - 6% acid in the water phase.

Upon examination of the preceding data in our laboratories, we found that Seltzer's data agreed with those of Belaga and Bigelow, whereas both showed differences from Sherwood's data. Check runs in the laboratory confirmed Seltzer's data and since these data lay in the concentration range of 0 - 1 normal, they were used in the present work. The other data are presented in Appendix A.

The equilibrium data in the concentration range up to 6 weight percent acetic acid in water could be represented by a straight line, which greatly facilitated the calculation of H.T.U. values. To minimize the error, the data were fitted for the concentration range of 1N to 0.1225N, a range within which most of the experiments were conducted (see Appendix C).

The data and the linear relation are given in Table 1.

TABLE 1

EQUILIBRIUM DATA FOR THE SYSTEM METHYL ISOBUTYL KETONE-ACETIC ACID-WATER (S2)

Conc. of Acid in Water, X, Normality	Conc. of Acid in Ketone, Y, Normality
1	0.5370
0.9	0.4797
0.7450	0.3917
0.5538	0.4147
0.4975	0.2825
0.4147	0.2059
0.3092	0.1526
0.2334	0.1135
0.2027	0.0976
0.1442	0.06836
0.1225	0.05932

Equation representing the data

$$Y = - 0.01403 + 0.5456\ X \pm 0.0042$$

EXPERIMENTAL PROCEDURES

Measurement of Variables

Measurement of Amplitude

The amplitude was measured by the method described by Thornton (T3). This method makes use of the difference in electrical conductivity between the aqueous and the solvent phases. The arrangement consists of a stainless steel probe, the tip of which is moved vertically in the region of the interface by a micrometer depth gauge. The top end of the probe is insulated from the column and wired to the ground with suitable resistances across a neon indicator light and the mains. The circuit was automatically completed since the column was supported by a steel frame.

When a measurement was made, the probe was lowered until the tip just contacted the interface on the upstroke of the pulse. This extinguished the neon indicator light. The upstroke of the pulse was thus marked by intermittent flashing of the light. The probe was then lowered until it just continued to make contact with the interface on the downstroke. This was indicated by a permanent extinction of the neon light. The difference in the two positions of the probe was a direct measure of the amplitude transmitted to the column.

Errors in Amplitude Measurements.--No mention has been made in literature about the variation in amplitude with change of frequency. In the particular system used in the present work, it was noted that the amplitude varied with frequency, depending on the particular bellows used for the pulse.

With the 1-1/2 inch Teflon bellows, it was found that the amplitude measured in the column decreased with increased frequency up to a value of 50 rpm beyond which it increased for all higher values of frequency. This dip was always at about 50 rpm and remained so for all the eccentrics used. Consequently, the 1-1/2 inch Teflon bellows were discarded. These variations are shown in Figure 2. The data are given in Appendix E. With the use of the 1 inch bellows, there was no minimum (see Appendix E). However, the amplitude transmitted to the column increased slightly with frequency. The difference was not appreciable for the range of frequency used in this work and an average value of the amplitude was used throughout the runs.

The amplitudes were extremely difficult to measure during the experimental run. With pulsation, the interface oscillated in the form of a wave traveling from the center to the periphery. Probably this was caused by the impingement of the droplets on the interface. In absence of the dispersed phase, no oscillation of the interface was observed. Hence, the amplitude was measured before the start of the runs, when the column was filled only with the continuous phase. Periodic checks showed that the amplitude remained essentially constant during the entire period of experimentation.

Measurement of Frequency

The highest frequency used in this work was 100 rpm. The frequency was therefore measured by counting the revolutions with the aid of a stop watch.

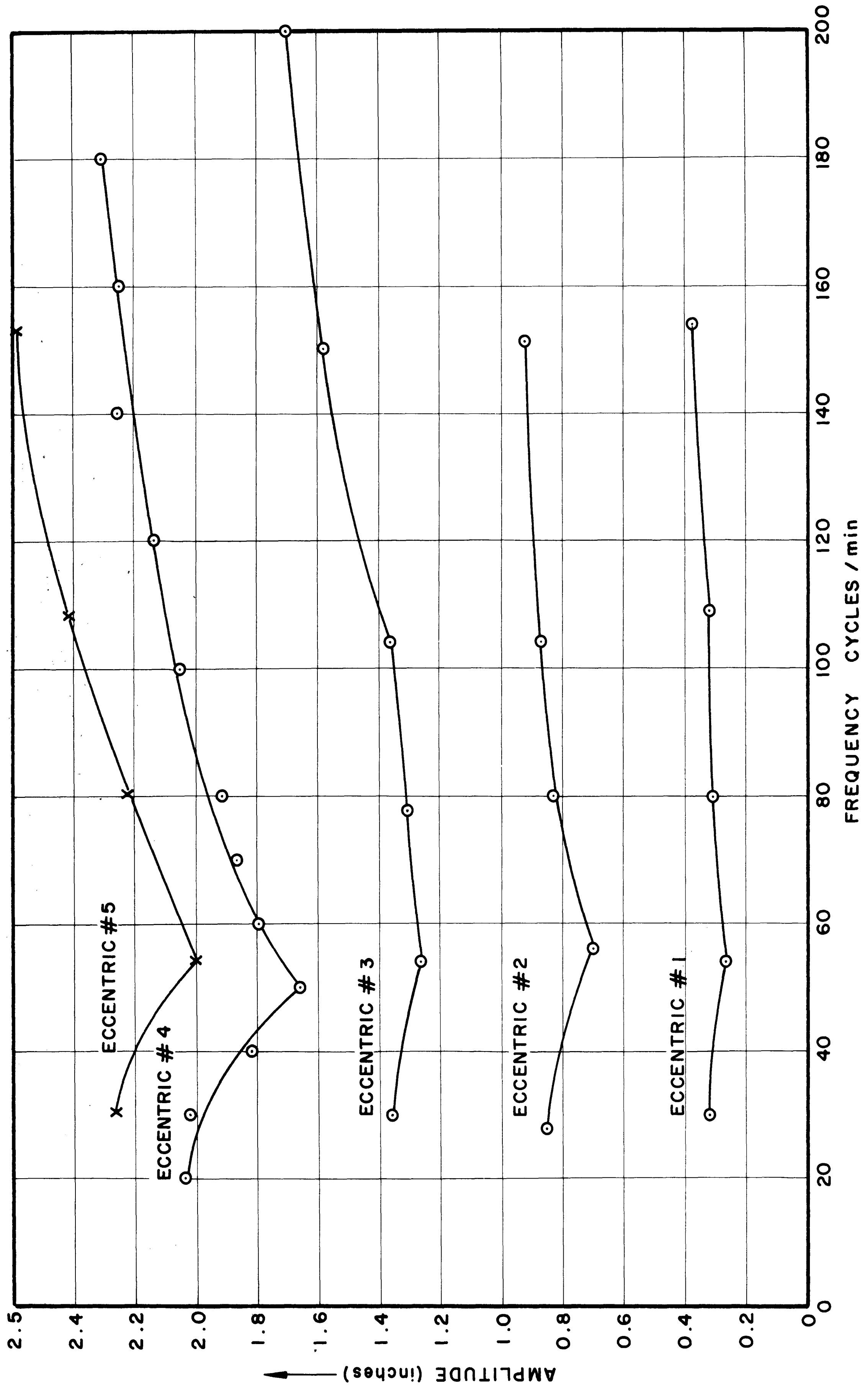


Figure 2. Calibration of 1-1/2 Bellows.

Measurement of Liquid Flow Rates

The feed solutions flowed through calibrated rotameters to the column. The rotameter reading introduced a large error in the calculation of material balances. Hence, after steady state conditions were reached, as indicated by a steady interface, the liquid flow rates were measured directly from the outlet streams with a graduating cylinder and a stop watch.

Startup and Column Operation

The experimental run was made in the same procedure described by Edwards and Beyer (E1). In the beginning the column was filled with the continuous phase to a few inches below the interface level. The desired pulsing conditions were adjusted and the pulse generator was started. The flow rates were then set at the desired levels. Subsequently, the hydraulic leg was adjusted to maintain the interface about 2 inches below the solvent outlet level.

It was found that approximately four volume changes of the column were needed before attaining steady state conditions. Usually the experiment was continued to permit about six volume changes before taking any measurements.

When steady state conditions were reached, the liquid flow rates were measured. Samples from the outlet streams were taken at 15 minute intervals and analyzed.

At the end of the run the pulser was switched off and the feed valves were closed. The hydraulic leg was then raised to stop the aqueous discharge. The hold up was determined by measuring the thickness of the solvent layer beneath each plate.

Analysis of the Liquid Streams

The liquid streams were analyzed by a simple acid-base titration against standard sodium hydroxide solution. For the solvent sample, distilled water was added and the sample was titrated in the same fashion until the aqueous layer changed color. The indicator was a 1% phenolphthalein solution in alcohol. The method was quite reliable and simple.

Feed Solution Makeup

In the entire series, the feed water solution was fixed at 1 Normal or 6% acetic acid. After adding the required amount of acid distilled water, the solution was saturated with the ketone by pumping in a closed circuit. The ketone was usually reused. The extract solvent was repeatedly washed with water until the acid concentration was less than .0005 N. Makeup ketone was added to the extract to make sure that the feed solvent was saturated with water.

THEORY

Behavior of a Pulse Column

The primary advantage of a pulse column lies in its high mass transfer rates. This is achieved mainly by dispersing the liquid into fine droplets, thereby increasing the interfacial area. The sieve plate columns usually use plates with 1/16 - 1/8 inch hole size. If such a column is not pulsed, the dispersed phase will merely accumulate at the plates. The frictional resistance of the plates and the surface tension forces of the liquids will be larger than the buoyancy force present by virtue of the difference in the densities of the two phases.

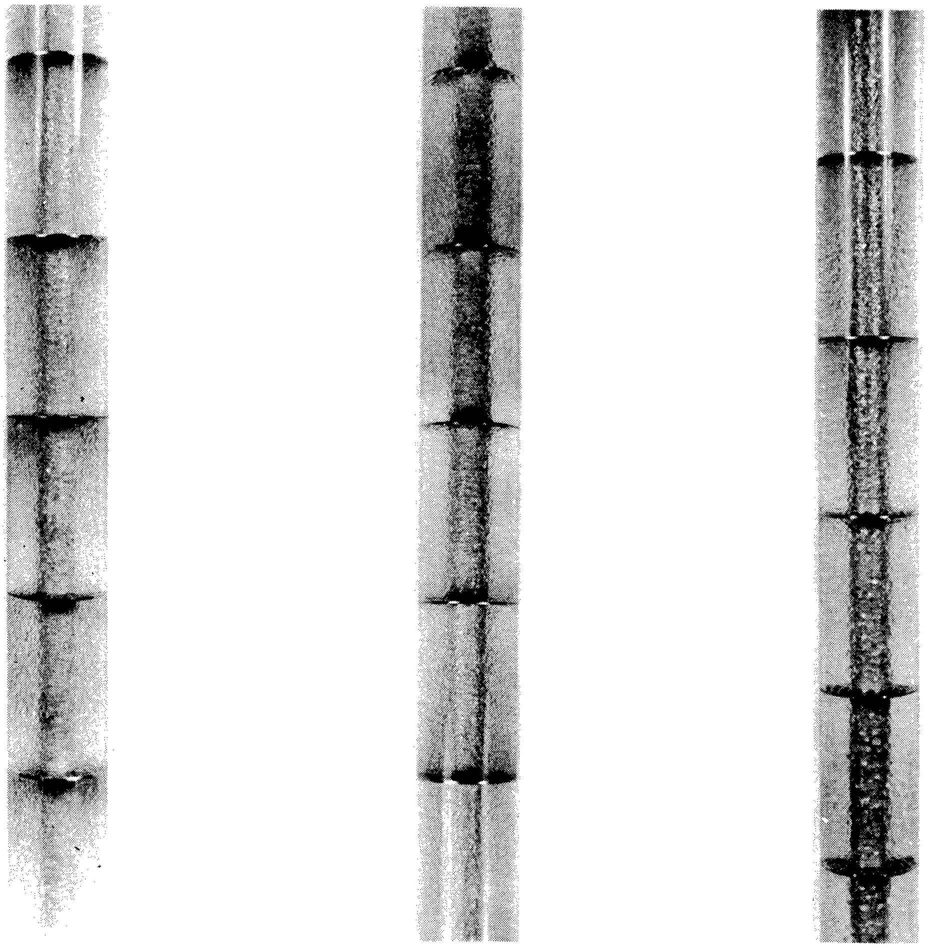
The pulser overcomes this barrier. It displaces in the column a volume exactly equal to its own (displacement x frequency). On the upward stroke a volume of the light phase equal to the pulse volume is pushed through the plates and on the downward stroke the same process is repeated for the heavy phase. If the pulse volume is less than the dispersed phase flow rate, it causes an accumulation of that phase in the column and the column ceases to operate.

By increasing the pulse rate, the pulse volume can be made just equal to the dispersed phase flow rate. This defines the lower limiting condition for the operation of a pulse column. Edwards (El) has derived mathematical relations to define this condition.

In the vicinity of this condition, the behavior of the column is unique. The contacting section is clearly divided into layers of the two phases at each plate. On the upward stroke the light phase beneath the plate rises in fine droplets through the plate and through the heavy phase to coalesce beneath the next plate. On the downward stroke the heavy phase descends through the plates in the form of drops through the light phase. This behavior is analogous to having a number of contacting and separating sections on top of each other and is often referred to as a "mixer-settler" type of operation. Eguchi and Nagata (E2) report the concentration change of the continuous change from plate to plate for such an operation. As expected, there is a distinct step-wise change in the concentration from plate to plate. The column, in other words, behaves truly like a plate column, the plates being the determining factor. However, this type of operation gives low mass-transfer rates and consequently is of little industrial importance.

As the pulse rate is increased, fewer drops of the dispersed phase coalesce. Eventually, at high pulsing rates no coalescence at the plates will occur. Under these conditions, both the continuous and the dispersed phases move back and forth through the plates, the droplet size diminishes and the plates act only as the resistance to flow. This type of operation is often referred to as "Emulsion" type operation. The mass transfer rates are high in this region partly because of the fine drop size and partly because of the high turbulence. In such an operation drops do not coalesce and reform, but are sheared and broken at each plate. Photographs of the column operating in this range are shown in Plate 3.

In the present work the light solvent phase was the dispersed phase. Visual observations demonstrated that the drop size was much finer in the top section of the column than in the bottom. The plates, therefore, behave like packing with an exceptionally high open area. Eguchi and Nagata (E2) report that the concentration change in the column is almost uniform for such an operation.



Top Section

Middle Section

Bottom Section

Plate 3. Droplet Distribution Along the Column.

Criterion for the Mass Transfer Rate

For a stage-wise contacting device, the performance of individual extraction stages can be described by the approach to equilibrium actually realized at the stage. The "Murphree Stage Efficiency" neatly defines this performance. Likewise, the performance of a continuous contacting device can be described by the number of theoretical stages to which it is equivalent. The "Height Equivalent to a Theoretical Stage" (H.E.T.S.) is a simple package by which the experimental data can be extended to design purposes. The only drawback of the H.E.T.S. concept is that it erroneously describes a continuous concentration change by a step-wise change.

An equally empirical, but theoretically sounder concept describing continuous mass-transfer operation is the "Transfer Unit" proposed by "Chilton and Colburn" (C3).

The validity for the transfer unit concept is its basis on the two-film theory for mass-transfer mechanism. An advantage of the same, is that it permits the direct calculation of the product of mass transfer coefficient "K" and the interfacial area "a". In certain continuous contacting devices such as the "Wetted Wall Column," the interfacial area "a" is easily calculated. In others, such as the spray column, it may be calculated by measuring the drop size without too much difficulty.

With this in mind, the "Height of a transfer unit" (H.T.U.) was selected as the criterion for the performance of the pulsed plate column, in preference to the "Height Equivalent to a Theoretical Stage" (H.E.T.S.).

Equivalence of H.T.U. with H.E.T.S. in the Present Study

In the present case the equilibrium line and the operating line, were assumed to be straight lines. For such a special case an analytical solution of the McCabe-Thiele method can be used to find the theoretical stages for any given conditions. If, moreover, the operating line has the same slope as the equilibrium line, as was the case in most of the experiments in this study, then the following relations should hold true.

$$HTU)_{od} = HTU)_{oc} = H.E.T.S.$$

The derivation of the preceding relationships is given below.

Relation Between H.T.U. and H.E.T.S.

For a stripping column, where solute transfer takes place from the aqueous phase to the solvent phase, the following relations hold good if the operating line and the equilibrium line are linear (P1).

$$a = \frac{L}{G} = \text{slope of operating line}$$

$$m = \text{slope of equilibrium line}$$

and subscripts 2 and 1 refer to concentrated and dilute ends respectively.

N_s = No. of theoretical stages

$$= \frac{\ell n[(1 - \frac{a}{m})(\frac{mx_2 - y_1}{mx_1 - y_1}) + \frac{a}{m}]}{\ell n \frac{a}{m}}$$

N_{oc} = No. of transfer units based on the continuous phase

$$= \frac{\ell n[(1 - \frac{a}{m})(\frac{mx_2 - y_1}{mx_1 - y_1}) + \frac{a}{m}]}{1 - \frac{a}{m}}$$

Since

$$N_s = \frac{Z}{H.E.T.S.} \quad \text{and} \quad N_{oc} = \frac{Z}{H.T.U.}$$

we get the following relationships.

$$H.E.T.S. = HTU)_{oc} \; \frac{\ell n \frac{a}{m}}{\frac{a}{m} - 1}$$

Similarly

$$H.E.T.S. = HTU)_{od} \; (\frac{m}{a}) \; \frac{\ell n \; \frac{a}{m}}{\frac{a}{m} - 1}$$

when in the limit $\frac{a}{m} \rightarrow 1$, i.e., the operating line and the equilibrium line are parallel,

$$H.E.T.S. = HTU)_{oc} = HTU)_{od}$$

Height of a Transfer Unit

Material Balance in a Stripping Column

Consider a stripping column as represented in the diagram.

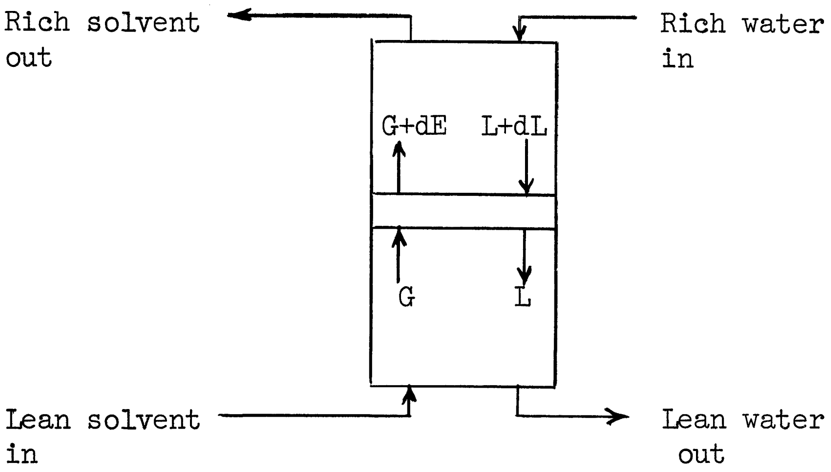


Figure 3. Stripping Column.

G = solvent phase rate, moles/hr.

L = water phase rate, moles/hr.

y,x = mole fraction of solute in the solvent and the aqueous phase resp.

Z = height of the column, ft.

s = cross sectional area of the column, sq.ft.

a = interfacial area of mass transfer, sq.ft./c.ft.

G' = solute free solvent rate, moles/hr.

L' = solute free aqueous rate, moles/hr.

Y,X = solute free mole fractions of solvent and aqueous phase resp.

For a differential column height dZ, a material balance of the solute transfer may be written as

$$d(Gy) = d(Lx) \qquad (1)$$

If the two phases are considered mutually insoluble

$$G'dY = L'dX \qquad (2)$$

The rate of mass transfer may be expressed by a rate equation

$$\begin{aligned} \text{Rate} &= k_g a \ sdz \ (f_i - f_g) \\ &= k_\ell a \ sdz \ (f_\ell - f_i) \\ &= K \ a \ sdz \ (f_\ell - f_g) \end{aligned}$$

where f_g, f_i and f_ℓ the fugacities of the solute in the phase G, at the interface and in the phase L respectively.

Equating the rates based on the material balance and the rate equation,

$$\begin{aligned} -G'dY = -L'dX &= k_g a \ sdz \ (f_i - f_g) \\ &= k_\ell a \ sdz \ (f_\ell - f_i) \\ &= K_a \ sdz \ (f_\ell - f_g) \end{aligned}$$

Grouping the above variables on a single phase,

$$-\frac{dY}{f_i - f_g} = \frac{k_g\ a\ s}{G'}\ dZ$$

and

$$-\frac{dY}{f_\ell - f_g} = \frac{K\ a\ s}{G'}\ dZ$$

Similar equations may be written for the other phase.

Driving Force

The terms f_i-f_g and f_ℓ-f_g represent the difference in fugacities of the solute between the interface and phase G, and between the phase L and phase G respectively. This difference is the driving force or the potential difference. In liquid-liquid systems it is necessary to assume that fugacities are proportional to molal concentrations. If the systems are very dilute, the fugacities may be assumed to be proportional to the concentration expressed as moles per unit volume.

If the two phases are in equilibrium with respect to the solute, then $f_g = f_\ell$. However the concentrations of the solute in the two phases may be different. In other words, the proportionality factor between fugacity and concentration may vary from system to system.

It is convenient to express the driving force in terms of a single phase. Here we introduce the term Y* to represent f_ℓ, and Y to represent f_g. Y* then would be the concentration of the G phase, which would be in equilibrium with the concentration X in the L phase. Similarly X* would be the concentration of the L phase which would be in equilibrium with the concentration Y in the G phase. The following diagram explains the above terms.

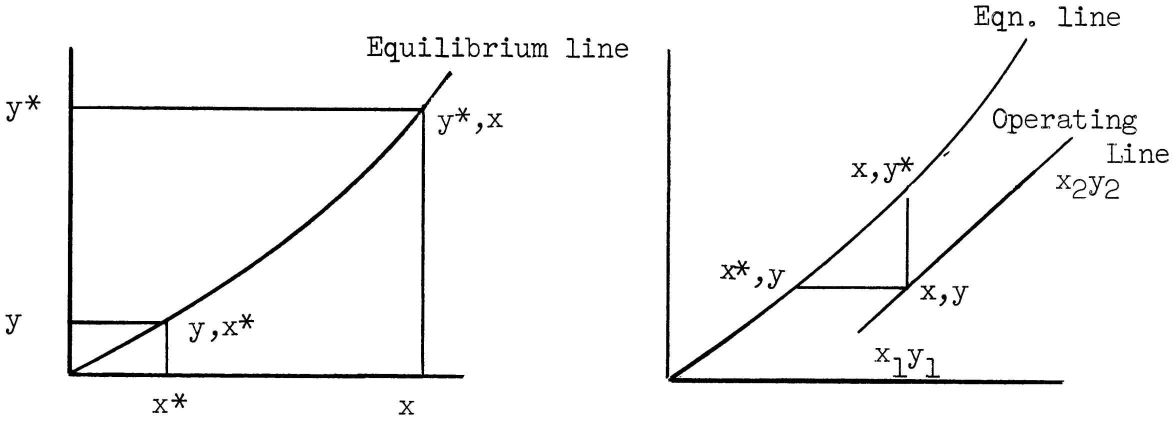


Figure 4. Equilibrium and Operating Lines.

The Transfer Unit

The material balance and the rate equations may now be written as follows:

$$-\frac{dY}{Y_i - Y} = \frac{k_g a\ s}{G'}\ dZ$$

$$-\frac{dY}{Y^* - Y} = \frac{K_{OG} a\ s}{G'}\ dZ$$

or

$$-\frac{dy}{(1-y)(y_i - y_g)} = \frac{k_g a\ s}{G'}\ dZ$$

and

$$-\frac{dy}{(1-y)(y^* - y_g)} = \frac{K_G a\ s}{G'}\ dZ$$

If the solutions are dilute, $1 - y \rightarrow 1$ and $G' \rightarrow G$

Then

$$\frac{dy}{y_i - y} = \frac{k_g a\ s}{G}\ dZ \qquad (A)$$

and

$$-\frac{dy}{y^* - y} = \frac{K_G a\ s}{G}\ dZ\ . \qquad (B)$$

Equation (B) is far more convenient to use than Equation (A) since y* is easy to calculate compared to y_i. Hence we integrate Equation (B), assuming K_Ga to be constant.

$$\int_{y_1}^{y_2} \frac{dy}{y^*-y} = -\frac{K_Ga\ s}{G} \int dZ$$

The integrated value of the left hand side gives the N_{od} or the number of transfer units, so called because the left hand side is a measure of the amount of mass transfer.

So $$N_{od} = \int \frac{dy}{y^*-y} = -\frac{K_Ga\ s}{G}\ Z$$

For the phase L, we may write

$$N_{oc} = \int \frac{dx}{x-x^*} = -\frac{K_La\ s}{L}\ Z$$

The height of a transfer unit (H.T.U.) then simply becomes the height of the column divided by the number of transfer units

$$HTU)_{od} = \frac{Z}{N_{od}} = \frac{Z}{\int \frac{dy}{y^*-y}} = -\frac{G}{K_Ga\ s}$$

Similarly

$$HTU)_{oc} = \frac{Z}{N_{oc}} = \frac{Z}{\int \frac{dx}{x-x^*}} = -\frac{L}{K_La\ s}$$

<u>Calculation of H.T.U.</u>

$$N_{od} = -\frac{K_ga\ s\ z}{G} = \int \frac{dy}{y^*-y}$$

To evaluate the integral, one must know the relationship between y* and y. If the equilibrium line is not straight, then this relationship becomes complex and the integral has to be evaluated by graphical integration by plotting y against y*-y and measuring the area under the curve.

However, if the equilibrium line is a straight line in the region of interest (not necessarily passing through the origin) then an analytical solution can be found for the integral. Thus

$$y^* = mx + c \quad \text{and} \quad \frac{y-y_1}{x-x_1} = \frac{L}{G}$$

Substituting the value for y*, we get

$$\int_{y_1}^{y_2} \frac{dy}{y^*-y} = \frac{y_2 - y_1}{\dfrac{(y_2^*-y_2)-(y_1^*-y_1)}{\ell n \dfrac{y_2^*-y_2}{y_1^*-y_1}}} = \frac{\Delta y}{\ell n_m (y^*-y)}$$

Application to the Present Study

In the present system, the equilibrium curve is fitted to a straight line. The inlet concentration of the rich aqueous phase was fixed at 1 mole/liter. The fugacities are assumed to be proportional to the concentration in moles/unit volume, and the operating line assumed to be straight. The H.T.U. values are calculated by using the above solution.

WETTING CHARACTERISTICS AND REPRODUCIBILITY PROBLEMS

It has been reported in some articles in the literature (B6,E1,S1) that plate-wetting characteristics play an important role in pulse column performance. However, only Sege and Woodfield (S1) have studied plate materials as a variable. They used stainless steel, fluoroethene and dual-surface plates with stainless steel on one side and paint on the other. Most of the other investigators have used stainless steel, brass or some polymer.

We did not plan to study the wetting characteristics or, for that matter, to study different plate materials. It was more by accident that we found variations in column performance due to the condition of plate surface. In the laboratory, any of the following conditions may affect the plate surface: dust from the atmosphere, an oil film deposited from vapors in the air*, leaving the plates in the column with a layer of organic phase beneath each plate, leaving the plates in water over the weekend, the degree of polish on the plates and of course the plate material.

In the early part of this work, 316 stainless steel plates were used. They were carefully drilled with the desired set of holes. The lubricating oil was thoroughly removed from the plates with c.p. acetone and the plates were used in the column. The plates were left in the column until a complete series of experiments was finished, i.e., the plates were not removed from the column and cleaned at the start of

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* Heat transfer experiments with oil at high temperatures were being performed next door.

each experiment. This seemed to be a satisfactory arrangement since our system was methyl isobutyl ketone - an excellent solvent - and acetic acid which is corrosive.

During these runs it was observed that all the plates did not give the same drop size. One or two plates would give smaller or larger drops than the rest. In one case, half the plates in the column were found to behave differently than the rest. These eight plates were separated and an experiment conducted to compare the two sets of plates. Under identical conditions of operation, one set gave 3.98 Transfer Units, whereas the other gave 3.22 Transfer Units -- about 20 per cent lower. The plates were removed and washed with fresh ketone, dried and the same runs were repeated after a week. The performance was now identical, i.e., 3.63 and 3.56 respectively. Subsequently, the plates were washed with fresh ketone and kept in a dust-free atmosphere.

After obtaining a few sets of data in this fashion we cleaned the equipment with soap and water. The column was then thoroughly rinsed with tap water. Subsequent operation of the column gave unusually high rates of mass transfer. These data were somewhat reproducible, i.e., while the plates were wet in the column or outside under water but not after they were removed and allowed to dry in air. To illustrate the point, using 1/16 inch hole size, 32.7 per cent free area, .21 inch amplitude, 2 inches plate spacing, 80 cycles/min. frequency and flow rates of 300 c.c./min., the untreated plates gave a $HTU)_{od}$ of 5.67 inches, the plates with soap and water treatment gave a $HTU)_{od}$ of 3.45 inches and after drying and a lapse of four days, the $HTU)_{od}$ was 4.64.

The data taken after the soap and water treatment are compared with those taken prior to it in Table 2 and are plotted in Figure 5. A scrutiny of the data brings out several interesting inferences.

First of all, the large difference in the H.T.U. is most evident from the data obtained by using a 1/8 inch amplitude (curves 1 and 1s). These values decrease from the range of 7.7 - 6.4 inches to 3.5 - 2.67 inches. With higher amplitude the difference is not as large but it is still significant. This difference can be noticed by comparing curves 2 and 2s as well as curves 4 and 4s. In this case, the effect of the plate condition is larger than the effect of either amplitude or of frequency. Moreover, it may be noticed that the range of operation is narrowed due to the soap and water treatment. Thus with 0.21 inch amplitude (curves 2 and 2s) the frequency range available is 80 to 160 cycles/min. for untreated plates, whereas that range is narrowed from 80 to 120 cycles/min. for the treated plates. Beyond 120 cycles/min., the column flooded. A similar case is observed with 0.62 inch amplitude (curves 4 and 4s). The flooding point is 80 cycles/min. as compared to 120 cycles/min. for untreated plates.

Further, one can notice that for the untreated plates, the effect of amplitude is significant (curves 1, 2, 3 and 4). But for the treated plates, the H.T.U.'s are all very close, ranging from 3.5 to 2.7 inches. It can be said that for the treated plates, the amplitude and frequency have no effect on the H.T.U. -- which brings us to an important inference that the effects of the column variables cannot be isolated from the condition of the plates.

TABLE 2

EFFECT OF SOAP AND WATER TREATMENT ON STAINLESS STEEL PLATES

UNTREATED PLATES

Amplitude inch	Frequency cycles/min.	H.T.U. inches
.21	80	5.67
	120	5.15
	160	4.85
.423	40	6.97
	80	4.95
	120	4.6
.62	40	6.0
	80	5.0
	120	4.25
.125	120	7.7
	160	7.1
	240	6.4

Constant Conditions

Hole size 1/16 inch
Free area 32.7%
Spacing 2 inches
Liquid flow
rates 300 cc/min

TREATED PLATES
(Plates washed with soap solution and rinsed with water overnight)

Amplitude inch	Frequency cycles/min.	H.T.U. inches	H.T.U., after 4 days in air
.21	80	3.45	4.64
	120	3.3	
.62	40	3.4	
	80	3.32	
.125	120	3.5	
	160	2.72	
	240	2.62	

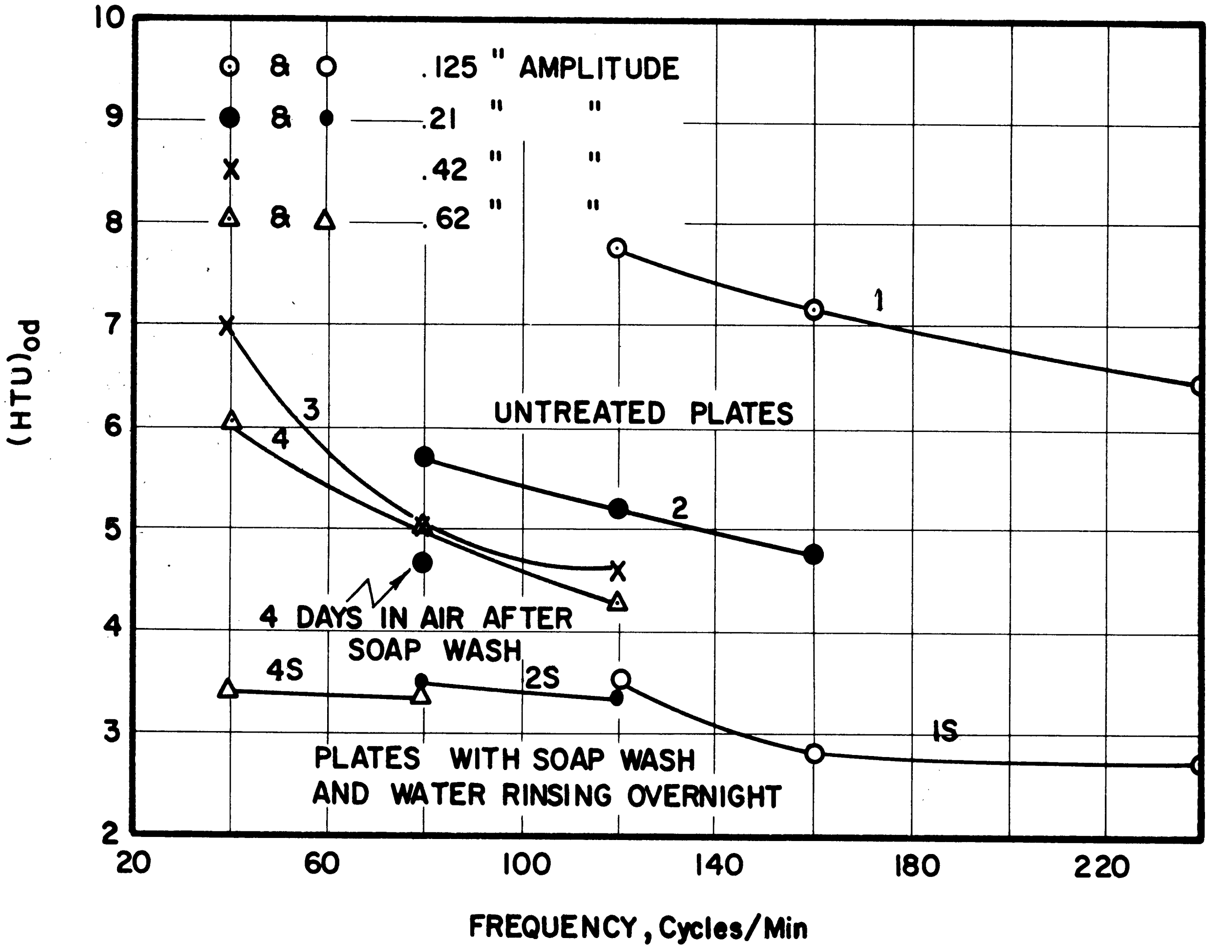


Figure 5. Effect of Soap and Water Treatment on Stainless Steel Plates. $HTU)_{od}$ in inches vs. frequency cycles/min.

We may add here that the effect of the soap treatment was not singular to the plates used in the preceding set of results. A different set of plates with the same hole size (1/16 inch) but with 40 per cent free area was subjected to a similar treatment of soapwash and overnight rinse. Using 0.21 inch amplitude and 80 cycles/min. frequency, the untreated plates gave a H.T.U. of 6.5 inches, whereas the treated plates gave 3.57 inches.

Since the plates which were maintained wet gave better mass transfer rates than those which were allowed to dry, it was felt that the surface could have adsorbed air. Soap treatment could conceivably contaminate the plates. Hence the plates were boiled with distilled water to remove any adsorbed air. So treated, the plates gave excellent mass transfer rates and the column behavior was similar to that after the soap and water treatment. The data taken with this treatment are presented in Table 3 and plotted in Figure 6. Photographs of the droplets in the column using boiled plates and plates cleaned with acetone and dried are shown in Plate 4.

A scrutiny of these data brings out the same facts as do the data from the soap and water treatment, i.e., (1) The plate conditions have a larger effect on the H.T.U. than does the amplitude or frequency, and (2) The effect of the amplitude and the frequency on the H.T.U.'s is much smaller when the plates are wetted compared to the plates which are untreated.

The data obtained from the boiled plates depended upon the length of time for which the column was run. For example, with 0.213 inch amplitude and 80 cycles/min. frequency, the $HTU)_{od}$ varies from

TABLE 3

EFFECT OF WETTING CONDITONS ON STAINLESS STEEL PLATES: BOILED WATER TREATMENT

UNTREATED PLATES				TREATED PLATES (Plates boiled in distilled water and maintained wet)		
Amplitude inch	Frequency cycles/min.	H.T.U. inches		Amplitude inch	Frequency cycles/min.	H.T.U. inches
.21	80	6.52	Constant Conditions	.21	50	4.8
	120	6.02			60	3.74
	160	5.15	Hole size 1/16 inch		80	3.13
	200	5.02	Free area 24.5%		100	2.94
			Spacing 2 inches			
.423	40	7.0	Liquid flow	.423	30	4.54
	80	6.32, 6.0	rates 300 cc/min		40	3.56
	120	5.85			60	2.97
	160	4.55				
	180	4.23				
.62	40	6.65		.62	30	4.28
	80	5.78			40	3.84
	120	4.45			50	3.42
	150	4.17			60	3.16

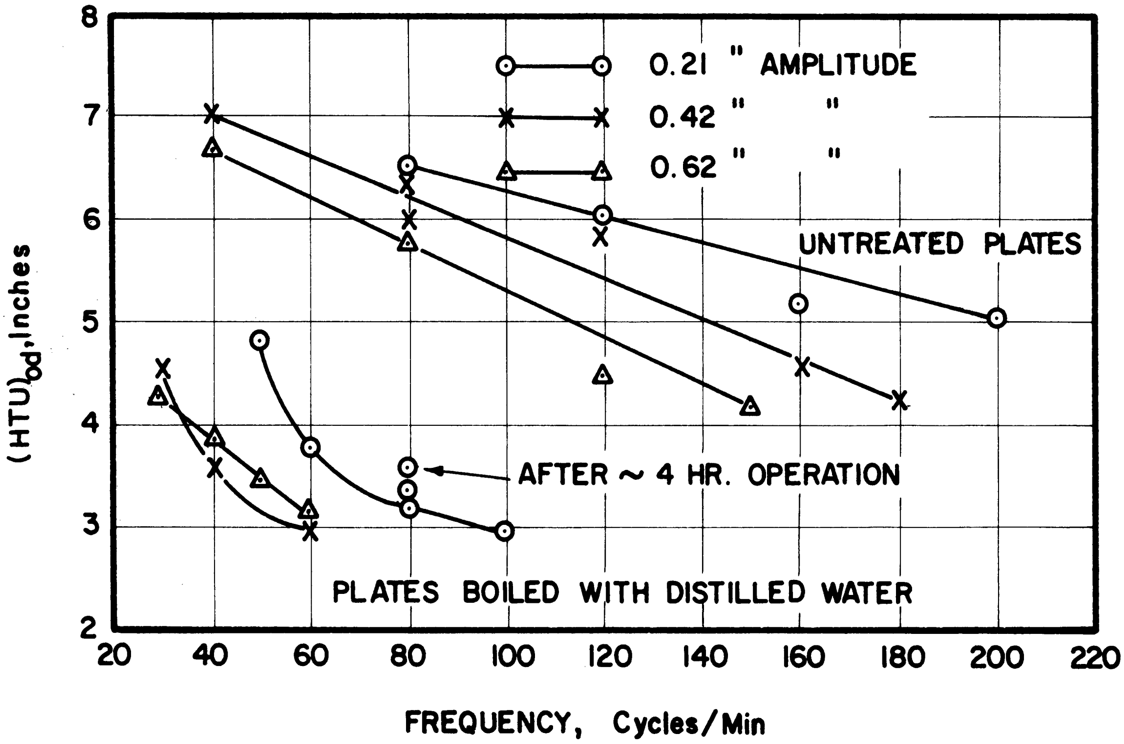
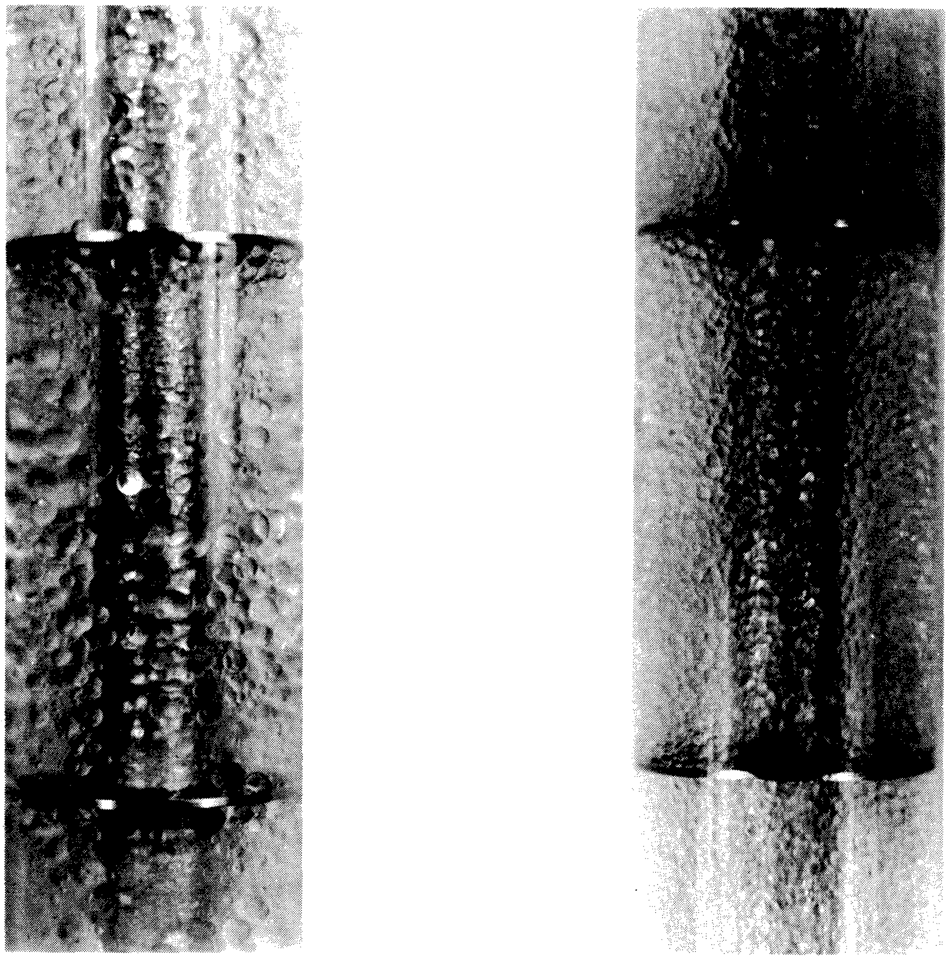


Figure 6. Effect of Wetting Conditions on Stainless Steel Plates: Boiling Water Treatment.



Droplets obtained from plates washed with ketone and dried in air.

"center plate section"

Droplets obtained from the same plates, boiled with water distilled water and maintained wet.

"center plate section"

Condition of Operation:
Amplitude: 0.21 inch
Frequency: 80 cycles/min.
Spacing: 2 inches
Free Area: 32.7%
Hole Size: 1/16 inch
Liquid flow rate: 300 cc/min.

Plate 4. Effect of Boiling Water on Stainless Steel Plates - Drop Size.

3.13 inches to 3.56 inches after four hour operation. This difference is significant even though it is not very large. Moreover, when the plates were kept immersed in water for a period of two weeks, the $HTU)_{od}$ with the same conditions of operation was 3.0 inches.

Boiling of the plates was then abandoned and recourse had to brass plates.

Self Sacrificial Behavior of Brass Plates

Originally we selected stainless steel plates in preference to brass plates because our system was mildly corrosive (1 molar acetic acid). Acetic acid attacks brass, but in doing so provides a clean, fresh surface. Before use in the column, the brass plates were washed with a 10% nitric acid. This gave to the plates a clean crystalline surface, which was maintained by the corrosive medium during a run. The transfer rates obtained from these plates were in fair agreement with those obtained from the stainless steel plates, boiled with water or washed with soap and water. Besides, the results were reproducible within experimental error. The brass plates were not immune to wetting effects. Actually if the plates were left in the column after shut down for about 2 hours with a layer of the organic phase under each plate, the mass transfer rates were about 20 per cent lower than those obtained with clean plates. Photographs of this operation are shown in Plate 5. Illustrative data are shown in Table 4 and are plotted in Figure 7. However, the data with the freshly cleaned plates were reproducible. The brass plates, in effect, sacrificed themselves to provide a reproducible set of results. In time, the hole size increased steadily but slowly. This did not introduce

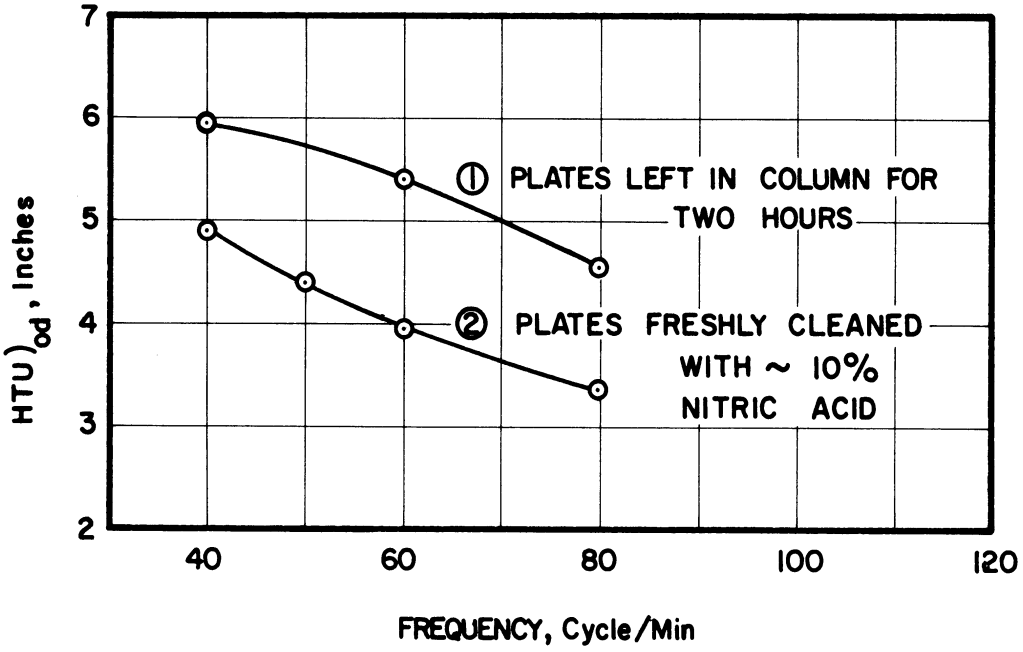
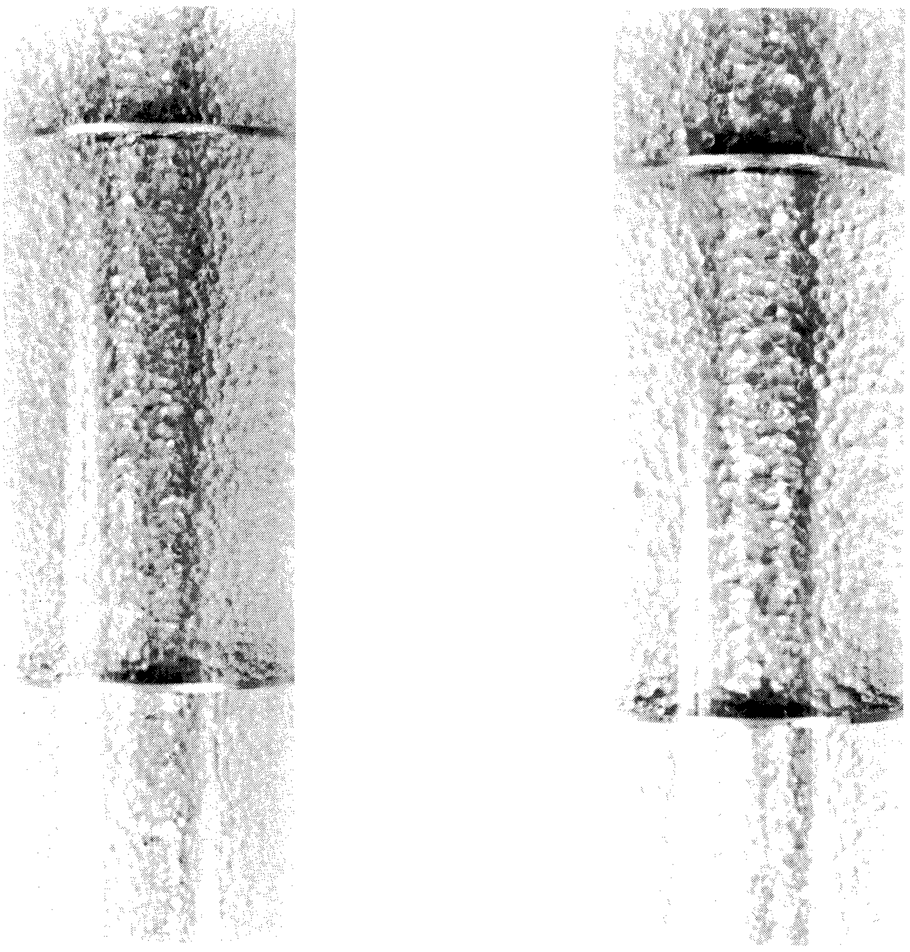


Figure 7. Effect of Wetting Conditions on Brass Plates.



Droplets obtained from Brass plates, freshly cleaned with 10% HNO_3.

"center plate section"

Droplets obtained from Brass plates left in the column with a layer of ketone under each plate for 2 hours.

"center plate section"

Condition of Operation:
Amplitude: .21 inch
Frequency: 80 cycles/min.
Plate Spacing: 2 inches
Free Area: 24.55%
Hole Size: 1/16 inch
Liquid flow rate: 300 cc/min.

Plate 5. Effect of Wetting Conditions on Brass Plates - Drop Size.

TABLE 4

EFFECT OF WETTING CONDITIONS ON BRASS PLATES
$HTU)_{od}$, INCHES VS. FREQUENCY, CYCLES/MIN.

		Frequency Cycles/Min.			
		40	50	60	80
$HTU)_{od}$	Plates freshly cleaned with nitric acid	4.9	4.41	3.95	3.35
	Plates left in the column for 2 hrs. with layers of ketone under the plates	5.91	-	5.4	4.54

any significant error since the surface effects cause a larger variation in mass transfer rates than does the doubling of hole size from 1/16 inch to 3/32 inch. In subsequent runs work on stainless steel was abandoned. The data used in the present correlation were obtained entirely from brass plates and are reported in the next chapter.

The importance of wetting characteristics cannot be overemphasized. Garner, Ellis and Hill (G1) have suggested an empirical method to select the plate material for any given system. This method is based on the measurement of contact angles made by one phase droplet in the other. The difference in the cosines of the two angles is taken as a measure of the adhesive work between the fluids and the surface in question.

However, as we have seen in the previous discussion, the plate material itself is not the determining factor. The previous history of the plates plays an important role. The plate surface after different treatments should, therefore, be studied before the final selection is made.

It is also conceivable that disagreements among the results of workers using the same system and same variables may be caused by the surface conditions of the plates.

RESULTS FROM BRASS PLATES

Nine factorial subsets comprising of 88 experimental runs are presented in Tables 6.1 - 6.9. The hole size at 1/16 inch and 3/32 inch as well as the free area at 24.55 and 32.7 per cent was tested in all the subsets.

Table 6.1 shows the comparison of three levels of amplitude. These could be tested at a frequency level of 60 cycles/min. The lowest and the intermediate amplitudes could be tested at two levels of frequency whereas the intermediate and the highest levels of amplitudes could be tested at three levels of frequency. These are presented in Tables 6.2 and 6.3. Tables 6.4 and 6.5 show the results of two 2^4 factorial subsets involving a l h p and a s h p. Tables 6.6 and 6.7 represent the comparisons of liquid flow rates and frequencies at the lowest and highest levels of amplitude. Tables 6.8 and 6.9 are subsets similar to Tables 6.6 and 6.7 except that they compare plate spacing instead of liquid flow rates. Liquid flow rates and plate spacing were not studied independent of each other. It may also be noted that the levels of frequency and the differences between these levels in Tables 6.2 and 6.3, Tables 6.6 and 6.7, and Tables 6.8 and 6.9 are different due to the corresponding levels of amplitude.

The analysis of variance of these factorial subsets was carried out according to the methods outlined by Davies (D1).

In addition to these subsets, 47 experimental runs were conducted in the boundary regions of the preceding range of variables. The entire set of experimental runs is presented in Tables 5.1 - 5.3.

TABLE 5.1

TABLE OF RESULTS

HTU$)_{od}$, inches as a function of operating variables

SET I, CONSTANT L_2 (300 c.c./min.) S_1 (2 inches)
Combined Flow Rate

		a_1 (.21 inch)				a_2 Levels of Amplitude (.423 inch)				a_3 (.62 inch)			
		f_3 50 rpm	f_4 60 rpm	f_5 80 rpm	f_6 100 rpm	f_2 40 rpm	f_3 50 rpm	f_4 60 rpm	f_5 80 rpm	f_1 30 rpm	f_2 40 rpm	f_3 50 rpm	f_4 60 rpm
h_1 1/16 inch	P_1 (16.35%)									99* 3.18 .324	100 3.14 .372	101 3.08 .451	Flood
	P_2 (24.55%)	89 3.84 -	1 3.67 .234	2 3.08 -	3 2.83 .406	73 3.68 .236	74 3.36 .265	75 3.24 .318	76 3.00 .465	3.5 .269	7 3.37 .291	8 3.37 .33	9 3.25 -
	P_3 (32.7%)		4 3.92	5 3.26 .261	6 3.19 .26	77 3.86 .232	78 3.61 -	79 3.28 .267	80 3.02 .332		10 3.52 .256	11 3.58 .277	12 3.27 .328
	P_4 (40.9%)		90 4.34 .187	91 3.64 .236	92 3.3 .308	93 4.27 .203	94 3.99 -	95 3.78 .215	96 3.38 .287		97 3.81 .24		98 3.52 .30
h_2 3/32 inch	P_1		107 3.42 .267	Flood						102 4.01 .24	103 3.36 .291		104 3.38 .578
	P_2	108 5.48 .209	13 5.0 .164	14 3.81 .187	15 3.1 .347	81 4.80 .207	82 3.87 .26	83 3.82 .215	84 2.88 .414	105 4.05 .223	19 3.74 .261	20 3.57 .273	21 3.29 -
	P_3		16 5.5 .148	17 4.5 .158	18 3.83 .262	85 4.95 .193	86 4.44 -	87 4.04 .193	88 3.38 .30	106 4.43 .203	22 4.25 .183	23 4.18 -	24 3.68 .236

TABLE 5.2

SET II, CONSTANT L_2 (300 c.c./min.) S_2 (3 inches)

		a_1 (.21 inch)				a_3 (.62 inches)			
		f_3 50 rpm	f_4 60 rpm	f_5 80 rpm	f_6 100 rpm	f_1 30 rpm	f_2 40 rpm	f_3 50 rpm	f_4 60 rpm
h_1 1/16 inch	P_1 (16.30%)	109 3.02 0.285	110 2.96 0.304	Flood		115 3.78 0.238	116 3.25 0.283	117 3.18 0.338	Flood
	P_2 (24.55%)		25 3.72 0.224	26 3.74	27 3.40 0.271	118 4.10 0.199	31 3.78 0.242	32 3.54 0.24	33 3.28 0.302
	P_3 (32.70%)	111 4.89 0.183	28 4.4 0.1775	29 4.05	30 3.68 0.264	119 4.60 0.195	34 4.20 0.23	35 3.94	36 3.83 0.3
	P_4 (40.9%)		112 5.22 0.168	113 4.67 0.179	114 4.08 0.197		120 4.38 0.185	121 4.03 0.213	122 3.88 0.224
h_2 3/32 inch	P_1 (16.30%)	128 3.76 0.246	129 3.87 0.232	130 3.29 0.289	Flood	123 4.27 0.199	124 3.38 0.277	125 3.16 0.396	Flood
	P_2 (24.55%)		37 5.04 0.16	38 3.96 0.232	39 3.61 0.185	126 5.38 0.18	43 5.02 0.189	44 3.96 -	45 3.52 0.294
	P_3 (32.70%)	131 5.48 0.152	40 4.92 0.16	41 4.05 0.211	42 3.7 -	127 5.12 0.17	46 4.95 0.1735	47 4.66 0.185	48 4.1 -

TABLE 5.3

SET III, CONSTANT L_1 (200 c.c./min.) S_1 (2 inches)

		a_1 (.21 inch)			a_3 (.62 inch)			
		f_4 60 rpm	f_5 80 rpm	f_6 100 rpm	f_1 30 rpm	f_2 40 rpm	f_3 50 rpm	f_4 60 rpm
h_1 1/16 inch	P_2 (24.55%)	49 4.82 0.1	50 4.15 0.117	51 3.71 0.1505	134 4.15 0.109	55 4.15 0.135	56 3.90 --	57 3.42 0.193
	P_3 (32.70%)	52 5.0 0.118	53 4.47 0.109	54 4.07 0.1425	135 4.81 0.127	58 4.65 0.119	59 4.3 -	60 3.58 0.1875
h_2 3/32 inch	P_2 (24.55%)	61 5.52 0.09	62 4.88 0.1155	63 4.23 0.12	133 4.33 0.118	67 4.30 0.119	68 4.14 0.123	69 3.54 0.166
	P_3 (32.70%)	64 5.82 -	65 5.11 -	66 4.69 -	132 4.86 -	70 4.65 -	71 4.65 -	72 4.38 -

Legend	99*	Expt. No.
	3.18	H.T.U.
	.324	Hold Up (Fraction Dispersed Phase)

FACTORIAL SUBSETS TABLES 6.1 - 6.9

Height of a Transfer Unit in Inches
as a Function of the Operating Variables.

TABLE 6.1

3 x 2 x 2 ahp Factorial Subsets

Constant levels, Liquid flow rates: 300 c.c./min.
Plate spacing : 2 inches
Frequency : 60 cycles/min.

		a_1 (.21 inch)	a_2 (.423 inch)	a_3 (.62 inch)
h_1 1/16 inch	p_1 (24.55%)	3.67	3.24	3.25
	p_2 (32.7%)	3.92	3.28	3.27
h_2 3/32 inch	p_1 (24.55%)	5.00	3.82	3.29
	p_2 (32.7%)	5.5	4.04	3.68

Analysis of Variance

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Square	Variance Ratio	Significance
a	2	2.9744	1.4872	572	*
h	1	1.8388	1.8388	706	*
p	1	0.1660	0.1660	63.8	-
ah	2	0.7777	0.3888	150	-
ap	2	.0335	0.0167	6.43	-
hp	1	.0554	0.0554	21.3	-
Error	1	.0026	0.0026		

TABLE 6.2

2^4 afhp Factorial Subset

Constant levels, Liquid flow rates: 300 c.c./min.
Plate spacing : 2 inches

		a_1 (.21 inch)		a_2 (.423 inch)	
		f_4 60/min	f_5 80/min	f_4	f_5
h_1 1/16 inch	p_2 (24.55%)	3.67	3.08	3.24	3.00
	p_3 (32.7%)	3.92	3.26	3.28	3.02
h_2 3/32 inch	p_2 (24.55%)	5.0	3.81	3.82	2.88
	p_3 (32.7%)	5.5	4.5	4.04	3.38

Analysis of Variance

Source of Variation	Degrees of Freedom	Mean Squares	Variance Ratio	Significanc
a	1	1.6384	32.7	x
f	1	2.6732	53.4	x
h	1	3.4782	69.4	x
p	1	0.7225	14.4	*
af	1	.0072	.14	-
ah	1	0.3306	6.6	*
ap	1	0.0025	.05	-
fh	1	0.5776	11.53	*
fp	1	0.0240	.5	-
hp	1	0.3660	7.3	*
Error	5	0.0501		

TABLE 6.3

2 x 2 x 2 x 3 afhp Factorial Subset

Constant levels, Liquid flow rates: 300 c.c./min.
Plate spacing : 2 inches

	a_2 (.423 inch)			a_3 (.62 inch)		
	f_2 40 cycles/ min.	f_3 50 cycles/ min.	f_4 60 cycles/ min.	f_2	f_3	f_4
p_1 (24.55%)	3.68	3.36	3.24	3.37	3.37	3.25
p_2 (32.7%)	3.86	3.61	3.28	3.52	3.58	3.27
p_1 (24.55%)	4.80	4.87	3.82	3.74	3.57	3.29
p_2 (32.7%)	4.95	4.44	4.04	4.25	4.18	3.68

Analysis of Variance

Treatment	Degrees of Freedom	Sum of Squares	Mean Squares	Variance Ratio	Significance
a	1	0.6273	0.6273	89.7	x
h	1	2.1841	2.1841	312.3	x
p	1	0.4538	0.4538	64.9	x
f	2	1.1558	0.5779	82.6	x
ah	1	0.2688	0.2688	38.4	x
ap	1	0.0096	0.0096	1.37	-
af	2	0.2398	0.1199	17.1	x
hp	1	0.1067	0.1067	15.2	x
hf	2	0.1584	0.0792	11.3	x
p f	2	0.0610	0.0305	4.4	-
Error	9	0.0613	0.007		

TABLE 6.4

2^4 alhp Factorial Subset

Constant levels, Frequency : 60 cycles/min.
Plate spacing : 2 inches

		a_1 (.21 inch)		a_3 (.62 inch)	
		l_1 200cc/min	l_2 300cc/min	l_1	l_2
h_1 1/16 inch	p_1 (24.55%)	4.82	3.67	3.42	3.25
	p_2 (32.7%)	5.00	3.92	3.58	3.27
h_2 3/32 inch	p_1 (24.55%)	5.52	5.0	3.54	3.29
	p_2 (32.7%)	5.82	5.5	4.38	3.68

Analysis of Variance

Source of Variation	Degrees of Freedom	Mean Squares	Variance Ratio	Significance
Amplitude a	1	7.344	120	x
Flow rates l	1	1.266	20.8	x
Hole size h	1	2.103	34.5	x
Free area p	1	0.436	7.15	*
al	1	0.168	2.76	-
ah	1	0.585	9.6	*
lh	1	0.053	.87	-
ap	1	0.002	.03	-
lp	1	0.006	.01	-
hp	1	0.126	2.07	*
Error	5	0.061		

* Significant at 5%, but not at 1%
\- Not Significant at 5%
x Significant at 1%

TABLE 6.5

2^4 ashp Factorial Subset

Constant levels, Frequency : 60 cycles/min.
Liquid flow rate: 300 c.c./min.

		a_1 (.21 inch)		a_3 (.62 inch)	
		s_1 (2 inches)	s_2 (3 inches)	s_1	s_2
h_1 1/16 inch	p_1 (24.55%)	3.67	3.72	3.25	3.28
	p_2 (32.7%)	3.92	4.05	3.27	3.83
h_2 3/32 inch	p_1	5.0	5.04	3.29	3.52
	p_2	5.5	4.92	3.68	4.1

Analysis of Variance

Source of Variation		Degrees of Freedom	Mean Square	Variance Ratio	Significance
Amp.	a	1	3.610	91.9	x
Spacing	s	1	0.048	0.99	-
Hole size	h	1	2.295	25.1	x
Free area	p	1	0.391	8.69	*
	as	1	0.16	3.56	-
	ah	1	1.071	23.8	x
	sh	1	0.028	0.62	-
	ap	1	0.021	.47	-
	sp	1	0.002	.04	-
	hp	1	0.002	.04	-
Error		5	0.045		

TABLE 6.6

2 x 3 x 2 x_2 lfhp Factorial Subset

Constant levels, Amplitude : .21 inch
Plate spacing: 2 inches

		l_1 (200 c.c./min.)			l_2 (300 c.c./min)		
		f_4 60 cycles/min.	f_5 80 cycles/min.	f_6 100 cycles/min.	f_4	f_5	f_6
h_1 1/16 inch	p_1 (24.55%)	4.82	4.15	3.71	3.67	3.08	2.83
	p_2 (32.7%)	5.0	4.47	4.07	3.92	3.26	3.19
h_2 3/32 inch	p_1	5.52	4.88	4.23	5.0	3.81	3.1
	p_2	5.82	5.11	4.69	5.5	4.5	3.83

Analysis of Variance

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Square	Variance Ratio	Significance
l	1	4.8420	4.8420	159.8	x
h	1	4.0180	4.0180	132.5	x
p	1	0.8660	0.8660	28.6	x
f	2	5.878	2.939	97.0	x
lh	1	0.1291	0.1291	4.26	*
pl	1	0.0308	0.0308	1.01	-
hp	1	0.0662	0.0662	2.18	-
lf	1	0.0054	0.0027	..09	-
pf	1	0.0306	0.0153	.51	-
hf	1	0.3547	0.1773	5.85	*
Error	9	0.2704	.0300		

TABLE 6.7

2 x 3 x 2 x 2 lfhp Factorial Subset

Constant levels, Amplitude : .62 inch
Plate spacing: 2 inches

		l_1 (200 c.c./min.			l_2 (300 c.c./min.		
		f_2 40 cycles/ min.	f_3 50 cycles/ min.	f_4 60 cycles/ min.	f_2	f_3	f_4
h_1 1/16 inch	p_1 (24.55%)	4.15	3.90	3.42	3.37	3.37	3.25
	p_2 (32.7%)	4.65	4.30	3.58	3.52	3.58	3.27
h_2 3/32 inch	p_1 (24.55%)	4.30	4.14	3.54	3.74	3.57	3.29
	p_2 (32.7%)	4.65	4.65	4.38	4.25	4.18	3.68

Analysis of Variance

Treatment Comb.	Degrees of Freedom	Mean Square	Variance Ratio	Significance
l	1	1.8095	66.8	x
h	1	0.6700	24.7	x
p	1	0.9009	33.2	x
f	2	0.6135	22.6	x
hl	1	0.0198	.73	-
pl	1	0.0315	1.16	-
hp	1	0.1305	4.8	-
lf	2	0.0656	2.41	-
hf	2	0.0007	- -	-
kf	2	0.0034	--	
Error	9	0.0271		

TABLE 6.8

2 x 3 x 2 x 2 sfhp Factorial Subset

Constant levels, Amplitude : .21 inch
Liquid flow rate: 300 c.c./min.

		s_1 (2 inches)			s_2 (3 inches)		
		f_4 60 cycles/ min.	f_5 80 cycles/ min.	f_6 100 cycles/ min.	f_4	f_5	f_6
h_1 1/16 inch	p_1 (24.55%)	3.67	3.08	2.83	3.72	3.74	3.40
	p_2 (32.7%)	3.92	3.26	3.19	4.40	4.05	3.68
h_2 3/32 inch	p_1 (24.55%)	5.00	3.81	3.10	5.04	3.96	3.61
	p_2 (32.7%)	5.50	4.5	3.83	4.92	4.05	3.7

Analysis of Variance

Treatment	Degrees of Freedom	Sum of Squares	Mean Squares	Variance Ratio	Significance
s	1	0.2774	0.2774	6.66	*
h	1	2.7203	2.7203	65.06	x
p	1	0.6800	0.6800	16.27	x
f	2	5.0150	2.5075	59.97	x
hs	1	0.5104	0.5104	12.21	x
ps	1	0.0794	0.0794	1.90	-
hp	1	0.0003	0.0003	-	-
sf	2	0.1471	0.0736	1.76	-
hf	2	0.8619	0.4310	10.3	x
kf	2	0.0026	0.0013	-	-
Error	9	0.3766	0.0418	-	

* Significant at 5%, but not at 1%
\- Not Significant at 5%
x Significant at 1%

TABLE 6.9

2 x 3 x 2 x 2 sfhp Factorial Subset

Constant levels, Amplitude : .62 inch
Liquid flow rate: 300 c.c./min.

		s_1 (2 inches)			s_2 (3 inches)		
		f_2 40 cycles/min.	f_3 50 cycles/min.	f_4 60 cycles/min.	f_2	f_3	f_4
h_1 1/16 inch	p_1 (24.55%)	3.37	3.37	3.25	3.78	3.54	3.28
	p_2 (32.7%)	3.52	3.58	3.27	4.2	3.94	3.83
h_2 3/32 inch	p_1 (24.55%)	3.74	3.57	3.29	5.02	3.96	3.52
	p_2 (32.7%)	4.25	4.18	3.68	4.95	4.66	4.1

Analysis of Variance

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Square	Variation Ratio	Significance
s	1	1.3585	1.3584	46.7	x
h	1	1.4950	1.4950	51.4	x
k	1	0.8325	0.8325	28.6	x
f	2	1.3336	0.6673	22.9	x
hs	1	0.0693	0.0693	2.38	-
ps	1	0.0198	0.0198	.68	-
hp	1	0.0392	0.0392	1.35	-
sf	2	0.2568	0.1284	4.41	-
hf	2	0.2841	0.1421	4.88	*
pf	2	0.0522	0.0261	.89	-
Error	9	0.2617	0.0291		

* Significant at 5%, but not at 1%
\- Not Significant at 5%
x Significant at 1%

DISCUSSION OF RESULTS

The following discussion is based on four facets of this work: (1) The results obtained from stainless steel plates. These results illustrate the importance and the effect of the condition of the plate surface. (2) The overall results obtained from brass plates. These results cover the "range of industrial interest." (3) The analysis of variance of the factorial subsets. This is used to determine the relative importance of the variables and their interactions. (4) Comparison of our results with those of other investigators in the field. From the viewpoint of column design, none of these facets can be isolated from the rest. Hence this discussion is meant to give a perspective of the column behavior from the above angles.

In discussing the wetting effects we have pointed out several interesting and pertinent facts. Looking back at those results (Figures 5 and 6, Tables 2 and 3), it can be seen that the H.T.U.'s from the untreated plates range from 4.25 inches to 7.7 inches. Upon treatment with soap and water or with boiled distilled water, the H.T.U.'s for the same range of variables lie between 2.67 to 4.8 inches. For identical operating conditions, the H.T.U.'s for the treated plates are only about half as much as those for untreated ones. The treated plates have a narrower range of operating variables, i.e., for any amplitude, the column can be operated only in a narrow band of frequency. Whereas for the untreated plates the operating variables have a significant effect on the H.T.U.'s (Lines of constant amplitude are widely spaced), this effect is very small for the treated plates. We can then say that "given a favorable plate condition, the amplitude and frequency are really not very important." Probably the more important variables are the plate hole size, percent free area, liquid flow rates, etc., since these variables can be arbitrarily fixed and the operating range of the amplitude and frequency is in a sense dependent on them. Moreover, since the plate surface cannot be described by an standard, it becomes difficult if not pointless to compare our data with those of others directly. In the brass plates, we have found a surface which can be reproduced and which gives favorable dispersion, i.e., fine drops. Our results, i.e., H.T.U.'s may then be expected to be lower than those of most of the workers in the field. This we do notice.

Burkhart and Fahien (B5) who used the same system report H.T.U.'s from 3.65 to 8.28 inches. The majority of the results lie between 4 to 7 inches. For an amplitude of 0.25 inch, hole diameter of 1/16 inch, plate space of 2 inches, percent free area of 25% and a frequency of 50 cycles/min., they report an H.T.U. of 7.68 inches. For identical conditions except for the liquid flow rates, our value of H.T.U. is about half of their value, i.e., 3.84 inches. Since lower liquid flow rates give higher H.T.U.'s, we can probably attribute our low H.T.U.'s to the high flow rates we have used.

Chantry (C2) who also used low flow rates and a plate hole size of 3/64 inch and 5/64 inch report plate efficiencies of 30 - 70% or H.E.T.S. ranging from 4.7 inches to 11 inches. The amplitude and frequency for this performance was between 1.5 - 2.5 m.m. and 50-80 cycles/min. respectively. The range of his H.E.T.S. is also much higher than that of our results.

Eguchi and Nagata (E2) used a 1/16 inch hole size, an 8.1% free area and 2 inch plate spacing with flow rates in the same but slightly lower range than ours. They report their results in terms of the mass transfer coefficient K_Wa Their highest value of $K_W a$ is 0.008 1/sec. In the same units our lowest value is 0.017 1/sec., which is about twice their value. We are surprised at their throughputs with such a small free area. Under similar conditions in our equipment, flooding would probably occur.

Belaga and Bigelow (B2) used a 1/32 inch hole size, 23% free area and 1 inch plate spacing to study the effects of amplitude and frequency on H.T.U.'s. For the range of 1/8 inch to 1/2 inch amplitude and 20 - 80 cycles/min. frequency, their H.T.U.'s vary from 2.63 to 5.3 inches. These values are in the same range as our values, even though the hole size of their plates is 1/32 inch as compared to our values of 1/16 - 3/32 inch.

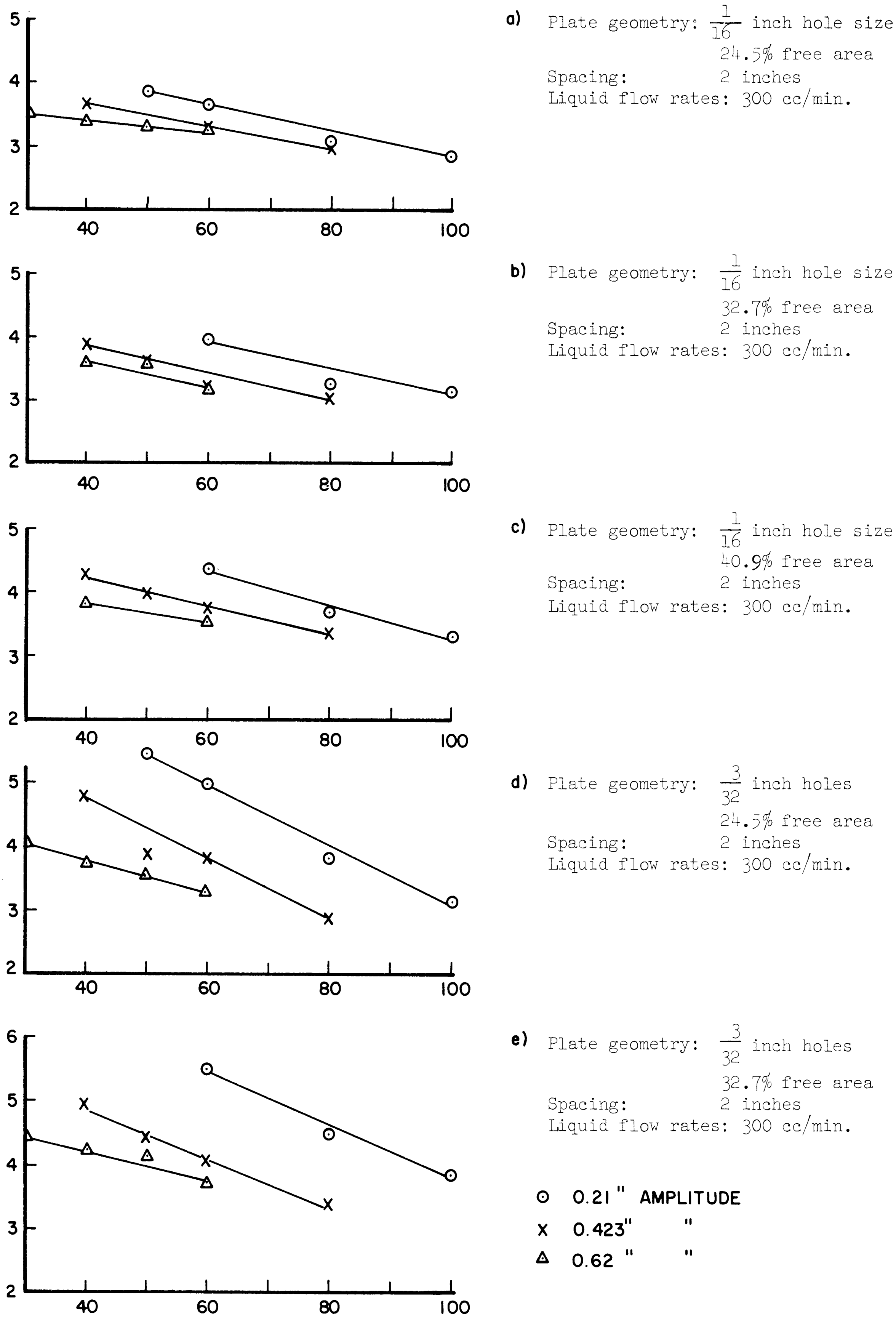
Thus we see that except for Belaga and Bigelow (B2), the rest of the workers who have used the same system as ours report H.T.U.'s much larger than our values.

Our experiments with the brass plates have been restricted to what we consider to be the range of industrial interest -- involving large throughputs and high mass transfer rates. In this region, we find that the H.T.U.'s vary very little, i.e., the operating variables have a relatively small effect. However, the trends of these effects are quite clear. These trends are discussed below.

Effect of Pulsation

Both amplitude and frequency affect the mass transfer rates independently of each other. Increase in either one decreases the H.T.U. up to the point of flooding without any points of inflection. Illustrative data are plotted in Figure 8. Belaga and Bigelow (B2) and Chantry (C2), who used the same system for plate column and packed column respectively, report some optimum amplitude and frequency. However, both of them have used low flow rates. Expressed in terms of combined flow rates, the former used approximately 300 gal./hr. sq. ft. and the latter used approximately 140 gal./hr. sq. ft. Burkhart and Fahien (B5), who also used the same system, report that increasing the amplitude from 0.25 inch to 1 inch and increasing the frequency from 25 to 100 cycles/min., both increased the H.T.U. -- a conclusion contradictory to our observations. However, they used low flow rates, i.e., approximately 250 gal./hr. sq. ft. Since no intermediate amplitude and frequency were studied by these investigators, it is probable that they have missed a point of inflection in the middle. With other systems, Cohen and Beyer (C4) report a point of inflection using approximately 175 gal./hr. sq. ft. Our studies were made with 670 - 100 gal./hr. sq. ft. As mentioned before, no optima were observed. Analysis of Li's (L1) data with the flow rates of about 600 gal./hr. sq. ft. show no minimum of H.T.U. with pulsation. Eguchi and Nagata (E2), who used the same system as ours and flow rates of about 500 gal./hr. sq. ft., also report no inflections.

We infer that the effect of pulsation on mass transfer rates depends upon the level of liquid throughputs, i.e., at low liquid flow



a) Plate geometry: $\frac{1}{16}$ inch hole size
24.5% free area
Spacing: 2 inches
Liquid flow rates: 300 cc/min.

b) Plate geometry: $\frac{1}{16}$ inch hole size
32.7% free area
Spacing: 2 inches
Liquid flow rates: 300 cc/min.

c) Plate geometry: $\frac{1}{16}$ inch hole size
40.9% free area
Spacing: 2 inches
Liquid flow rates: 300 cc/min.

d) Plate geometry: $\frac{3}{32}$ inch holes
24.5% free area
Spacing: 2 inches
Liquid flow rates: 300 cc/min.

e) Plate geometry: $\frac{3}{32}$ inch holes
32.7% free area
Spacing: 2 inches
Liquid flow rates: 300 cc/min.

Figure 8. Effect of Pulsation on H.T.U.
$(HTU)_{od}$ inches vs. Frequency, cycles/min.)

rates, there probably exists some optimum amplitude and frequency, whereas at high flow rates increase in amplitude and frequency concomitantly improve mass transfer rates up to the point of flooding. The levels of amplitude and frequency which would give the points of inflection are never reached since flooding interrupts the column operation before that point.

Effect of Plate Geometry

Smaller hole sizes and smaller per cent free areas increase mass transfer rates but simultaneously decrease the throughputs. In practice, therefore, it is necessary to determine as small a hole size and per cent free area as possible consistent with reasonable throughputs. Although the preceding statements are generally accepted, very little work has been done to determine the relative importance of the two variables. Sege and Woodfield (S1) have proposed a 1/8" hole size and 23 per cent free area as the optimum, based on mass transfer rates and the liquid throughputs. These values are also generally accepted and used in most of the A.E.C. work with pulse columns.

However, from preliminary experiments, we found that 1/8 inch hole size gave large drops and low mass transfer rates. From the analysis of variance of the factorial subsets presented in Tables 6.1 to 6.9 it can be seen that the effect of the hole size is much larger than that of the percent free area. The relative effects of the change in hole size from 1/16 - 3/32 inch and the change in free area from 24.5 to 32.7 percent are given below. (Reproduced from the Tables of Analysis of Variance.)

TABLE 7

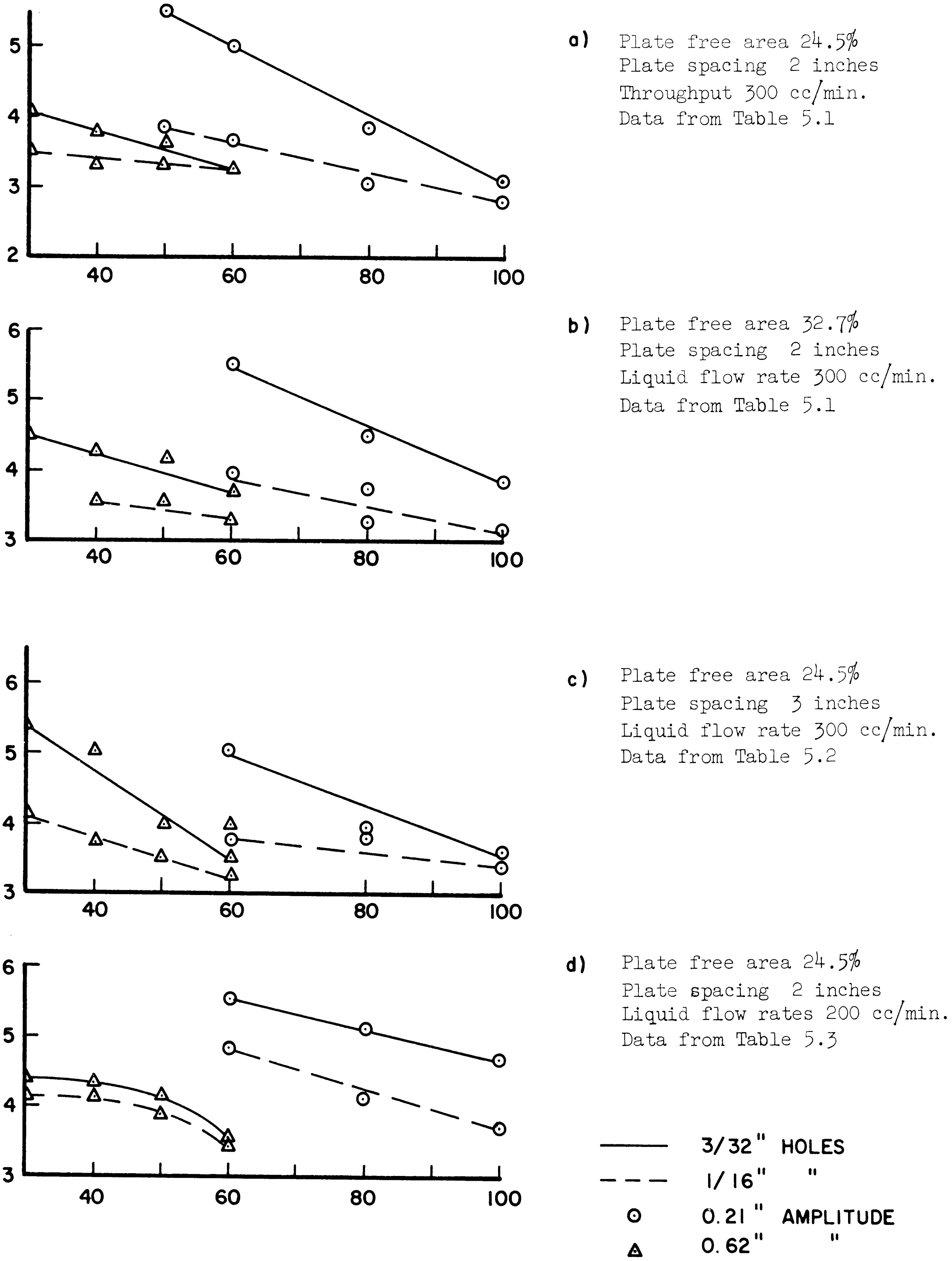
COMPARISON OF THE VARIANCE RATIOS OF THE PLATE HOLE SIZE AND PLATE FREE AREA

Table No.	Variance Ratios Hole Size	Variance Ratios Per Cent Free Area
6.1	70.6	63.8
6.2	69.4	14.4
6.3	31.2	64.9
6.4	34.5	7.15
6.5	25.1	8.7
6.6	132.5	28.6
6.7	24.7	33.2
6.8	65.1	16.3
6.9	51.4	28.6

Illustrative data are plotted in Figures 9 and 10. We would therefore recommend the use of plates with small hole size and large free area. Probably, a good plate geometry would be 1/16" holes with 33% free area.

Liquid Flow Rates and Plate Spacing

Due to the limited amount of time, the combination of low flow rates and high plate spacing was not studied. However, from the general pattern in the analysis of variance, we infer that their interaction is nonexistent or at least negligible. Increase in flow rates from 660 to 1000 gal./hr. sq. ft. improved mass transfer rates. Thornton (T2) reports that the $HTU)_{oc}$ showed little variation with flow rates in the



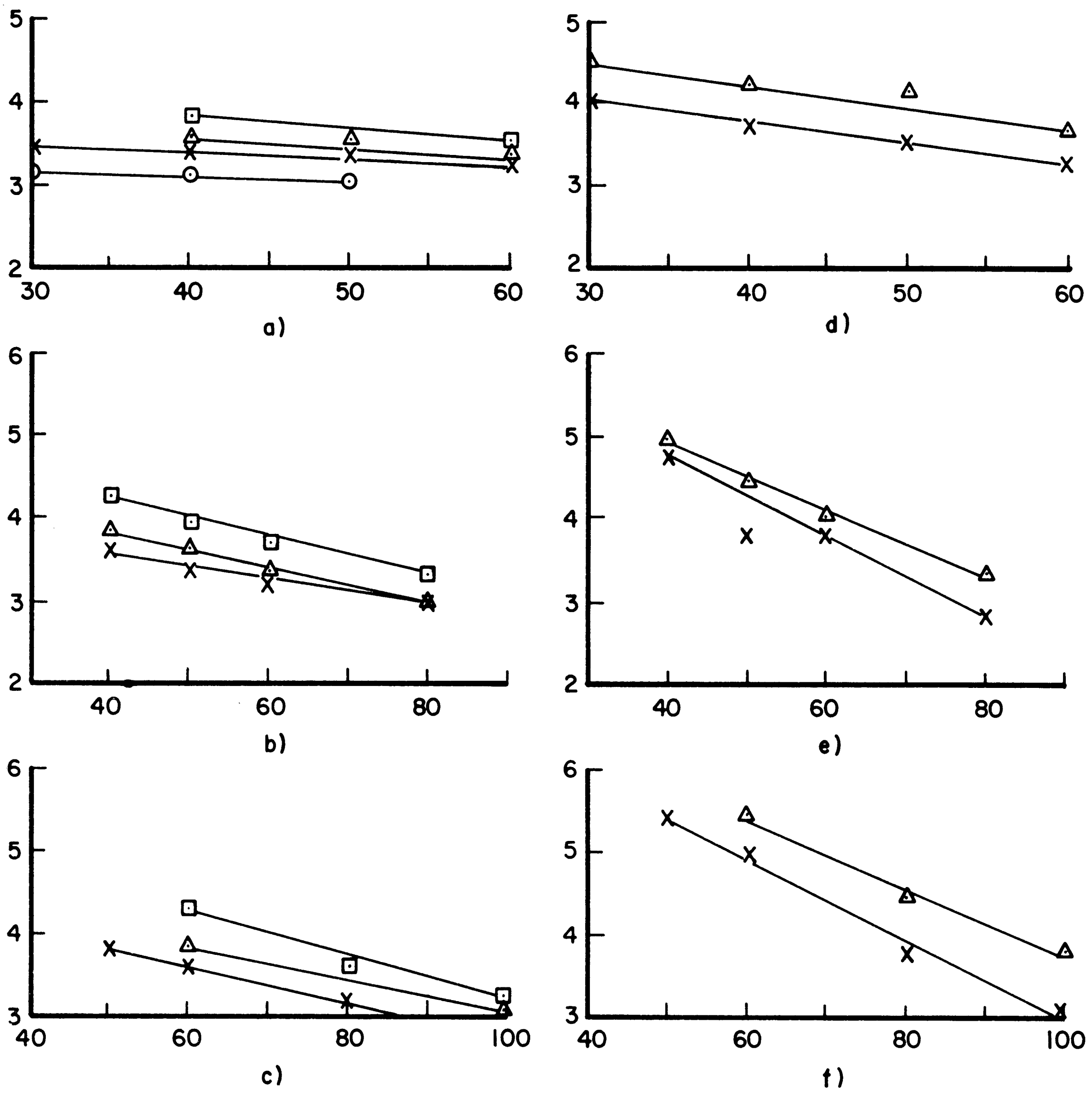
a) Plate free area 24.5%
Plate spacing 2 inches
Throughput 300 cc/min.
Data from Table 5.1

b) Plate free area 32.7%
Plate spacing 2 inches
Liquid flow rate 300 cc/min.
Data from Table 5.1

c) Plate free area 24.5%
Plate spacing 3 inches
Liquid flow rate 300 cc/min.
Data from Table 5.2

d) Plate free area 24.5%
Plate spacing 2 inches
Liquid flow rates 200 cc/min.
Data from Table 5.3

Figure 9. Effect of Plate Hole Size
$HTU)_{od}$ inches vs. Frequency cycles/min.



a) 1/16 in. hole size
.62 in. amplitude

b) 1/16 in. hole size
.423 in. amplitude

c) 1/16 in. hole size
.21 in. amplitude

d) 3/32 in. hole size
.62 in. amplitude

e) 3/32 in. hole size
.423 in. amplitude

f) 3/32 in. hole size
.21 in. amplitude

⊙ 16.3 %
X 24.5 %
△ 32.7 %
⊡ 40.9 %

Data from Table 5.1 at 2-inch spacing and 300 cc/min. throughputs

Figure 10. Effect of Plate Percent Free Area
$HTU)_{od}$ inches vs. frequency cycles/min.

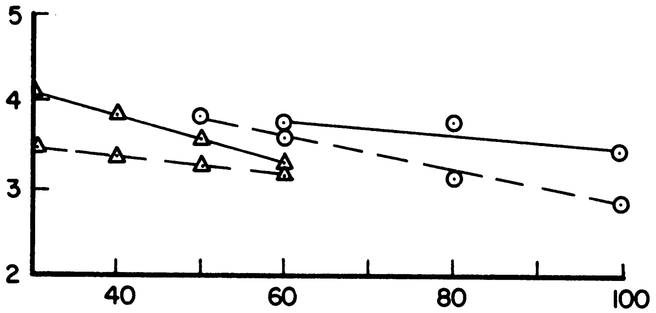
in the range 30 - 90% of the flooding rates. Cohen and Beyer (C4) report that the H.E.T.S. could not be demarked as a function of flow rates. We find that increasing flow rates increase the holdup, decrease the drop size and improve the mass transfer rates. (See Figure 12.)

A 2-inch plate spacing was found to be better than a 3-inch spacing. (See Figure 11.) Closer spacing was not studied for the reason given in the following section.

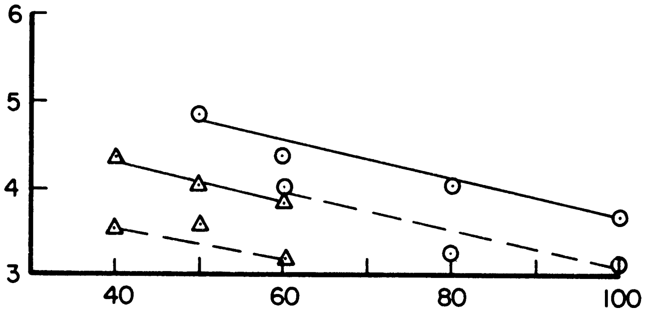
Drop Size and Holdup

From visual observations, we believe that almost all the variables affect the mass transfer rates through the drop size and the holdup. Large holdups (fraction dispersed phase) are associated with low H.T.U.'s and vice versa, although no quantitative relations could be established. Similarly, small drop sizes are associated with large holdups and small H.T.U.'s. Just before flooding points, the drop size is the smallest and holdup the largest and the mass transfer rates were found to be the highest. This held true, no matter how the flooding was approached, by increasing the flow rates, by increasing pulsation or by decreasing the hole size and fraction free area of the plates. In this region, the H.T.U.'s assume a limiting value with a variation of about ±25% (see Table 5.1; the H.T.U.'s range from 3-5 inches). It may be expected that, depending upon the system, this limiting value of the H.T.U. will change in magnitude, but the range within which it varies will be small.

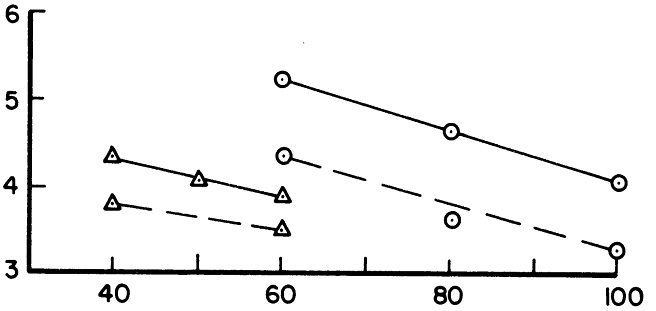
The drop size decreases and the holdup increases if the amplitude or frequency is increased. The same phenomenon is observed if the hole size or the per cent free area is decreased. However, although the



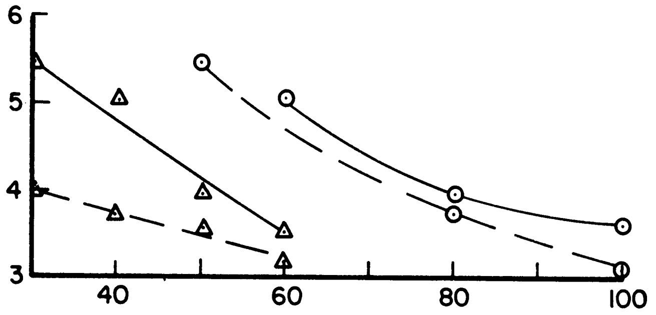
a) Hole size 1/16 inch
Free area 24.5%



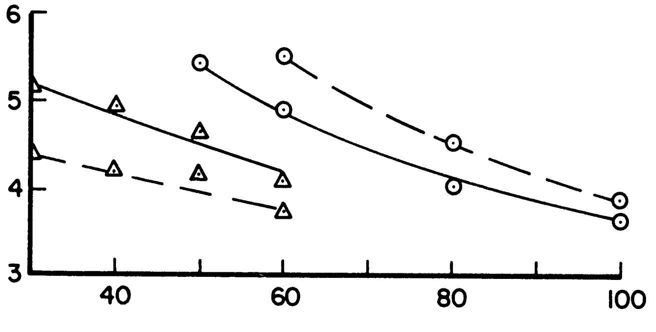
b) Plate geometry: 1/16 inch holes
32.7% free area



c) Plate geometry: 1/16 inch holes
40.9% free area

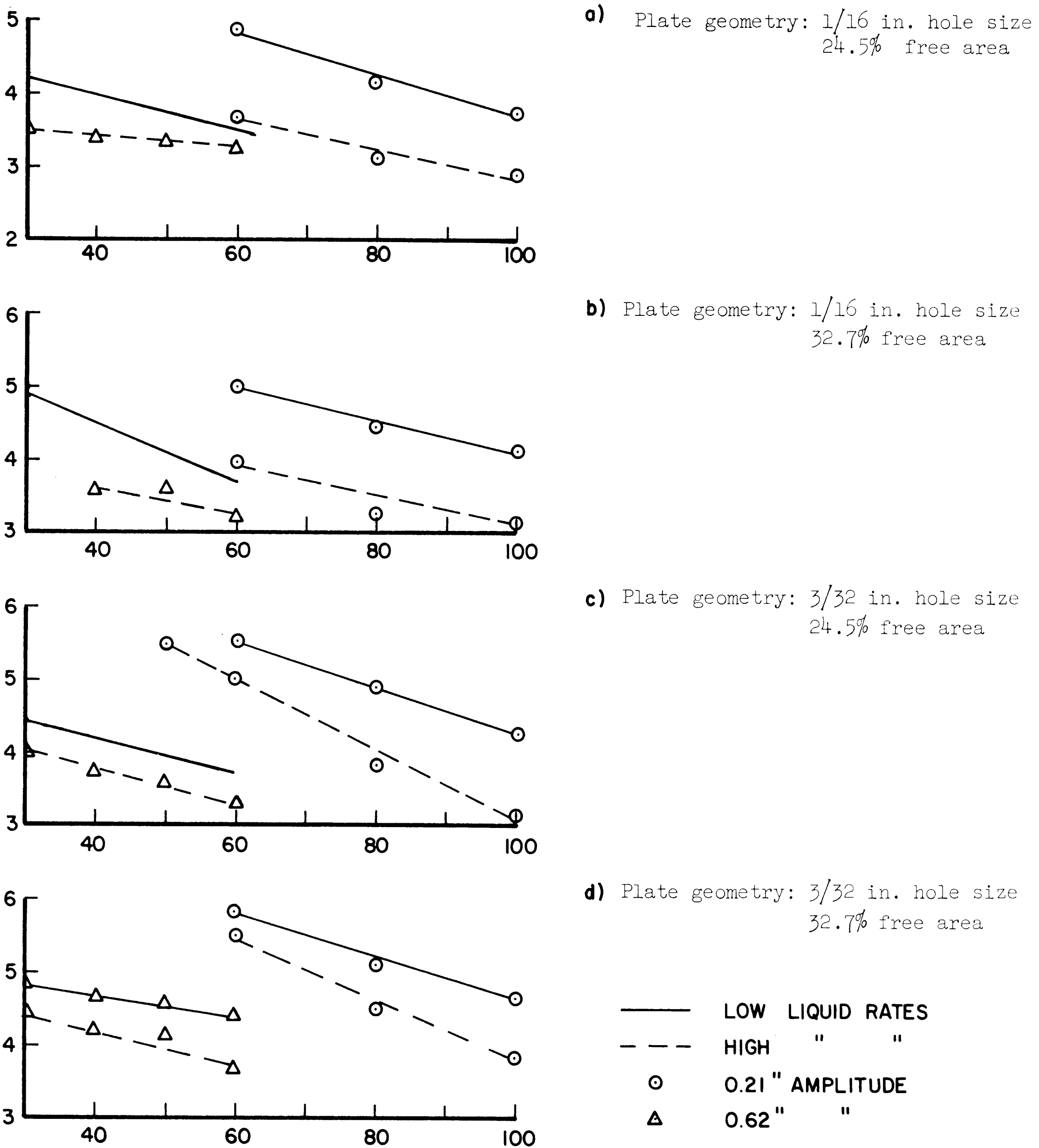


d) Plate geometry: 3/32 inch holes
24.5% free area



e) Plate geometry: 3/32 inch holes
32.7% free area

Figure 11. Effect of Plate Spacing
$HTU)_{od}$ inches vs. Frequency, cycles/min.



a) Plate geometry: 1/16 in. hole size
24.5% free area

b) Plate geometry: 1/16 in. hole size
32.7% free area

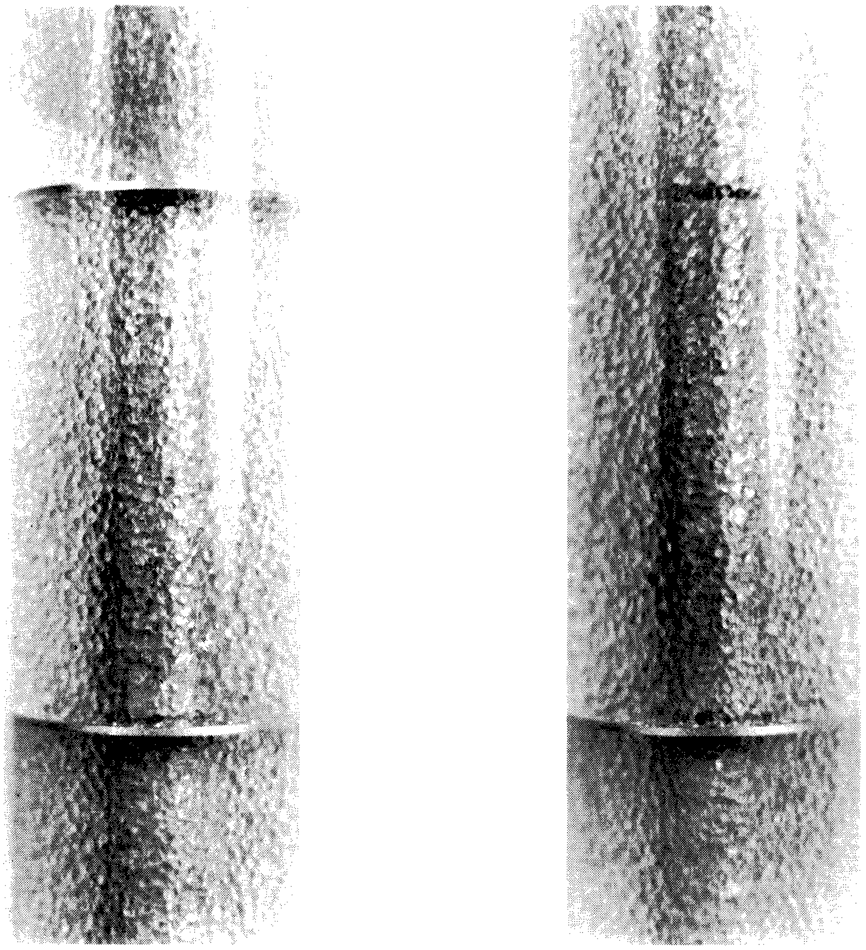
c) Plate geometry: 3/32 in. hole size
24.5% free area

d) Plate geometry: 3/32 in. hole size
32.7% free area

Data from Table 5.3 and the corresponding data from Table 5.1

Figure 12. Effect of Liquid Flow Rates
$HTU)_{od}$ inches vs. frequency, cycles/min.

hole size affects the drop size, the two are not identical. In the lower part of the column, the drops are larger than the holes, whereas in the top section, they form a fine conglomeration, much smaller than the hole diameter. One can notice the gradual decrease in drop diameter in the direction of the flow of the dispersed phase. The effect of the plates is to shear and break the drops as they pass through them. There is a lower limit to the drop size, which is reached within about two thirds of the column length. The plates in the top section thus are ineffective and function to prevent the coalescence of the drops. This behavior is illustrated in the photographs of the drops in top place section. Plate 6a shows the drops with the top plate in position. Plate 6b shows the drops with the top plate removed. Plate 7 shows the breakup of the dispersed phase by the bottom two plates. Plate 3 shows the drop sizes along the entire column length. Thus equal spacing of the plates is inefficient and although no reference is found in literature, we feel that the plates should be closely spaced at the bottom and loosely spaced at the top, or vice versa depending on the direction of the dispersed phase flow.



Droplets with the top plate in position.

6a

Droplets with the top plate removed.

6b

Plate 6. Drop Dispersion in the Top Section of the Column.

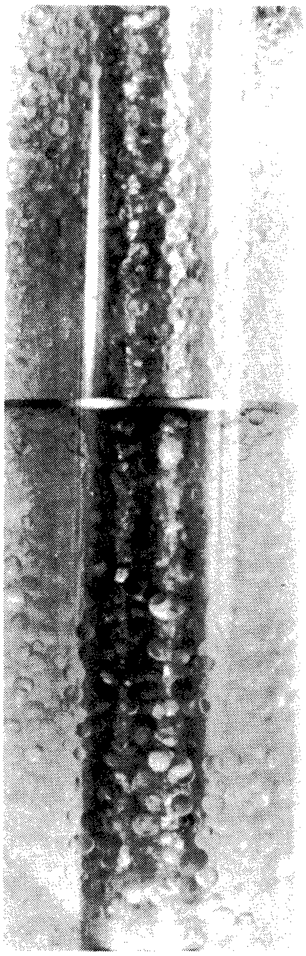


Plate 7. Drop Dispersion by the Bottom Plates.

CORRELATION OF RESULTS

At this stage we feel it pertinent to repeat some of the statements we have made previously. We have pointed out that the condition of the plate surface is of major importance. We do not have a standard method to define this plate surface. The results vary up to 100% with the plate surface. A quantitative correlation therefore cannot be of much use for industrial design of pulse columns. We have explored the region of column operation which gives high mass transfer rates. Outside this region in one direction, the mass transfer rates are lower, whereas in the other, column operation becomes difficult due to flooding. In this optimum region, we have found that the H.T.U. values vary very little. For our system, they vary from 2.8 to 5.5 inches.

For industrial design, one can say that the H.T.U.'s have one average value in this region with a variation of ±35 per cent.

Our correlation is restricted to this range. We have tried to correlate our data by several methods presented in literature. Two of these methods, i.e., use of dimensionless groups and Thornton's (T2) method are discussed later in this chapter. On comparison of these methods, we feel that the most suitable and simple representation of our data is in the form of a polynomial expressing H.T.U. as a function of the operating variables. In a general case, the polynomial takes the following form.

$$\text{H.T.U.} = c_o + c_1x_1 + c_2x_2 + c_{12}x_1x_2 + \ldots + c_nx_n + \epsilon$$

Where x's are the independent variables, c's are the coefficients and ϵ is the experimental error.

It is possible to fit any data to a polynomial by the use of a "sufficiently large" number of terms. However, to keep the expression from being clumsy, it is necessary to find the least number of terms which will represent the data within the experimental error. The analysis of variance provides a neat guide to select these terms. We start out with only those variables and interactions which are significant. Those variables and interactions which are not significant are assumed to have no effect on the dependent variable and hence are neglected. If this polynomial does not adequately represent the data, more terms have to be added. These new terms, however, must be powers of those terms which already have beenselected from the analysis. The powers of these terms represent the nonlinear effects of the variables. The variables which have the nonlinear effects can be determined by statistical analysis although the process is quite laborious.

In the present case, the analysis of variance for the various factorial subsets is presented in Tables 6.1 - 6.9. The three factor and higher order interactions are used to estimate the experimental error. Most of the two factor interactions are seen to be insignificant at the 5 per cent level, as determined by the "F" test. Some of these interaction are significant in some sets while being insignificant in others. No single two factor interaction is significant in all sets or even in the majority of the sets. The logical inference is that the two factor interactions are negligible. The variables can then be assumed to be independent of one another. In such a case, a revised estimate of the experimental error can be obtained by combining all the interactions.

The terms for the polynomial are now narrowed only to the individual variables and their powers. Using only the six variables and the constant, the equation becomes

$$\text{H.T.U.} = c_o + c_o a + c_2 f + c_3 h + c_4 p + c_5 s + c_6 \ell$$

This equation gave a large error as compared to the estimated error. Addition of two more terms viz. a^2 and f^2, changes the equation to

$$\text{H.T.U.} = c_o + c_1 a + c_2 a^2 + c_3 f + c_4 f^2 + c_5 h + c_6 p$$

$$+ c_7 s + c_8 \ell$$

Based on only the data included in Tables 6.1 - 6.9, the values predicted from the above equation came within ±0.27 inch of the observed values. The estimate of the error determined from the analysis of variance was ±0.285 inch. We consider this a satisfactory correlation. When additional data, taken in the laboratory but not included in the analysis of variance were used, the total error was ±0.31 inch. Addition of more terms decreased the error slightly but not sufficiently to justify their inclusion. The coefficients were determined by the method of least squares.

We feel that the eight terms and a constant adequately describe the polynomial correlating the H.T.U. values as a function of the operating variables.

The final correlation has the following form:

$$\text{HTU})_{od} = 5.9 - (5.85a - 4.17a^2) - (.0375f - .00009f^2) + 18.1h$$

$$+ .0506p + .314s - 4.75\ell \pm 0.31$$

where

$HTU)_{od}$: Overall height of a Transfer Unit, based on the dispersed phase, in inches.

a : Amplitude, in inches.

f : Frequency, in cycles per minute.

h : Plate hole size, in inches.

p : Area of the plate holes, per cent of cross sectional area of the column.

s : Plate spacing, in inches.

l : Combined liquid flow rates, in inches/sec.

For other systems, it may be expected that the absolute values of the H.T.U.'s will be different, but the coefficients of the expression will remain the same. In other words, the same expression will be applicable with the use of an overall factor characteristic of any particular system.

The range of variables for which the preceding equation predicts the H.T.U. values within the experimental error is as follows:

amplitude	0.21 inch - 0.62 inch
frequency	30 - 100 cycles/min.
hole size	1/16 inch - 3/32 inch
per cent free area	16 - 40
plate spacing	2 inch - 3 inch
combined liquid flow rates	600 - 1000 gal./hr. sq. ft.

The observed values, the values calculated from the preceding expression and the deviations are presented in Table 8.

TABLE 8

COMPARISON OF OBSERVED AND CALCULATED DATA

Expt No.	Observed Value HTU)$_{od}$ in.	Calculated Value	Deviation	Expt No.	Observed Value HTU)$_{od}$ in.	Calculated Value	Deviation	Expt No.	Observed Value HTU)$_{od}$ in.	Calculated Value	Deviation	Expt No.	Observed Value HTU)$_{od}$ in.	Calculated Value	Deviation
1	3.67	3.75	0.08	41	4.05	4.53	0.48	81	4.80	4.23	-0.57	121	4.03	4.22	.19
2	3.08	3.24	0.16	42	3.70	4.10	0.40	82	3.87	3.94	0.07	122	3.88	3.94	0.06
3	2.83	2.80	0.03	43	5.02	4.26	-0.76	83	3.82	3.66	-0.16	123	4.27	4.16	-0.11
4	3.92	4.15	0.23	44	3.960	3.962	0.002	84	2.88	3.15	0.27	124	3.38	3.84	.46
5	3.26	3.65	0.39	45	3.52	3.68	0.16	85	4.95	4.64	-0.31	125	3.16	3.55	.39
6	3.19	3.22	0.03	46	4.95	4.66	-0.29	86	4.44	4.34	-0.10	126	5.38	4.57	-0.81
7	3.37	3.37	0.00	47	4.66	4.37	-0.29	87	4.04	4.06	0.02	127	5.12	4.98	-0.14
8	3.37	3.08	-0.29	48	4.11	4.09	-0.02	88	3.38	3.56	0.18	128	3.76	4.50	.74
9	3.25	2.79	-0.46	49	4.82	4.46	-0.36	89	3.84	4.02	0.18	129	3.87	4.22	.35
10	3.52	3.78	0.26	50	4.15	3.96	-0.19	90	4.34	4.57	0.23	130	3.29	3.71	.42
11	3.58	3.48	-0.10	51	3.71	3.52	-0.19	91	3.64	4.07	0.43	131	5.48	5.32	-0.16
12	3.27	3.21	-0.06	52	5.0	4.87	-0.13	92	3.30	3.64	0.34	132	4.88	5.38	.50
13	5.00	4.32	-0.68	53	4.47	4.37	-0.10	93	4.27	4.49	0.22	133	4.33	4.97	.64
14	3.810	3.814	0.004	54	4.07	3.93	-0.14	94	3.99	4.19	0.20	134	4.15	4.40	.25
15	3.10	3.38	0.28	55	4.15	4.09	-0.060	95	3.78	3.91	0.13	135	4.81	4.40	-0.41
16	5.50	4.72	-0.78	56	3.90	3.80	-0.10	96	3.38	3.42	0.04				
17	4.50	4.22	-0.28	57	3.42	3.52	0.01	97	3.81	4.20	0.39				
18	3.83	3.79	-0.04	58	4.65	4.50	-0.15	98	3.52	3.63	0.11				
19	3.74	3.94	0.20	59	4.30	4.20	-0.10	99	3.18	3.27	0.09				
20	3.57	3.65	0.08	60	3.58	3.92	0.34	100	3.14	2.96	-0.18				
21	3.28	3.37	0.09	61	5.52	5.03	-0.49	101	3.08	2.66	-0.42				
22	4.25	4.35	0.10	62	4.88	4.53	-0.35	102	4.01	3.84	-0.17				
23	4.18	4.06	-0.12	63	4.23	4.09	-0.14	103	3.36	3.53	0.17				
24	3.68	3.78	0.10	64	5.82	5.44	-0.38	104	3.38	2.95	-0.43				
25	3.72	4.06	0.34	65	5.11	4.94	-0.17	105	4.05	4.26	0.21				
26	3.74	3.56	-0.18	66	4.69	4.50	-0.19	106	4.43	4.66	0.23				
27	3.40	3.12	-0.28	67	4.30	4.66	0.36	107	3.42	3.90	0.48				
28	4.40	4.47	0.07	68	4.14	4.36	0.22	108	5.48	4.60	-0.88				
29	4.05	3.97	-0.08	69	3.54	4.08	0.54	109	3.02	3.93	0.91				
30	3.68	3.53	-0.15	70	4.65	5.07	0.42	110	2.96	3.65	0.69				
31	3.78	3.69	-0.09	71	4.65	4.77	0.12	111	4.89	4.75	-0.14				
32	3.54	3.40	-0.14	72	4.38	4.50	0.12	112	5.22	4.89	-0.33				
33	3.28	3.11	-0.17	73	3.68	3.66	-0.02	113	4.67	4.38	-0.29				
34	4.20	4.01	-0.10	74	3.360	3.367	.007	114	4.08	3.95	-0.13				
35	3.94	3.80	-0.14	75	3.24	3.09	-0.15	115	3.78	3.59	-0.19				
36	3.83	3.52	-0.31	76	3.00	2.58	-0.42	116	3.25	3.27	0.02				
37	5.04	4.63	-0.41	77	3.86	4.07	0.21	117	3.18	2.98	-0.20				
38	3.96	4.13	0.17	78	3.61	3.77	0.16	118	4.00	4.10	-0.10				
39	3.61	3.70	0.09	79	3.28	3.49	0.21	119	4.41	4.60	-0.19				
40	4.92	5.04	0.12	80	3.02	2.99	-0.03	120	4.38	4.51	0.13				

Standard Deviation	0.961
Standard Error	±0.31

The approach to correlating our data by dimensionless groups and the method presented by Thornton (T2) are discussed below.

Dimensional Analysis

Dimensional analysis is a mathematical tool by which we deduce certain relationships between the physical variables in a particular system. In other words, we define the system in terms of certain combinations of variables called "dimensional groups," and in termsof "dimensional constants" used to normalize the units in the equations. Dimensional analysis imposes one basic restriction that "the dimensional formulas of the variables and of the constants must have the form of products of powers of the fundamental units."

The simple application of dimensional analysis is then to write the dimensional formulas of all the variables and find the dimensionless combinations of these variables. The number of dimensional products is the difference between the number of arguments (physical variables plus dimensional constants) and the number of fundamental units. In the present case we have studied six independent variables viz. amplitude, frequency, hole size, per cent free area, plate spacing and liquid flow rates. The dependent variable is the height of a transfer unit. Since the units are absolute, we have no dimensional constants. The number of fundamental units is two -- the length and time. Consequently, we have five dimensionless groups involving seven quantities. However, since one of our quantities viz. per cent free area is already dimensionless, we have four dimensionless groups involving six quantities. When the dimensional analysis is actually carried out, we have fourteen different combinations available for trials. None of these combinations can theoretically be shown to be superior to any other. Moreover, in all these combinations, one variable assumes a much larger importance than all others. An example of this case is shown below.

$$\frac{\text{H.T.U.}}{s} = \left(\frac{a}{s}\right)^{x}\left(\frac{fs}{\ell}\right)^{y}\left(\frac{h}{s}\right)^{z}(p)^{q}$$

Here, "s", the plate spacing, assumes predominance over others. Any of the other variables could be used in a similar fashion. The correlation by dimensional analysis then becomes a matter of trial and error to find a good fit.

Thornton's Method

Thornton (T2) has presented a generalized correlation between the HTU$)_{oc}$ and the operating variables, the physical properties of the system and the dispersed phase holdup.

Thornton assumes that the drop size of the dispersed phase is affected only by the power dissipated by friction per unit mass of fluid in the column. The power dissipated is a function of the pulse wave form and the operating variables viz. amplitude, frequency, plate hole size, spacing and per cent free area. It is assumed that the liquid flow rates have no effect on the drop size. The power dissipated is correlated with "characteristic velocity" and the physical properties of the system. The "characteristic velocity" is defined as the droplet velocity under the limiting condition when the flow rate of the continuous phase is zero. The characteristic velocity is in turn correlated with HTU$)_{oc}$ by the dispersed phase holdup and the physical properties of the system. The final correlations have the following form:

For Flooding:

$$\frac{\overline{V}_o \mu_c}{\gamma} = 0.60\left(\frac{\psi \mu_o^5 g_c}{\rho_c \gamma^4}\right)^{-.24} \left(\frac{h \rho_c \gamma}{\mu_c^2}\right)^{.9} \left(\frac{\mu_c^4 g}{\Delta\rho \gamma^3}\right)^{1.01} \left(\frac{\Delta\rho}{\rho_c}\right)^{1.8} \left(\frac{\mu_d}{\mu_c}\right)^{.30} \quad A$$

For Mass Transfer:

$$\frac{HTU)_{oc}}{(\mu_c^2/g \rho_c^2)^{1/3}} = K\left(\frac{\mu_c g}{\overline{V}_o (1-x)^3 \rho_c}\right)^{\frac{2m}{3}} \left(\frac{\Delta\rho}{\rho_c}\right)^{\frac{2(m-1)}{3}} \left(\frac{V_d}{V_c}\right)^{.5} \left(\frac{V_c^3 \rho_c}{g \mu_c x^3}\right)^{1/3} \quad B$$

For design purposes, the values of $\overline{V}_o$ can be obtained from Equation A when solute transfer takes place from the aqueous to solvent phase. The constants "K" and "m" are determined from model tests with the given system using a laboratory column. It is also necessary to carry out holdup determinations before Equation B can be used.

Using the systems toluene-acetone-water and butyl acetate-acetone-water, Thornton shows that the value of "m" is 0.5 for both systems. The scatter of the data is within ± 20 per cent of the predicted values.

With our data on the methyl isobutyl ketone-acetic acid-water, we find the value of "m" is about 0.08 with the scatter of data of approximately ± 20 per cent of the predicted values. Although this falls within the accuracy claimed by Thornton, we feel that the correlation offers no distinct advantage. For extrapolation of the data, the holdup determinations are necessary even for the same system. Since there is no correlation between the holdup and the operating variables, we feel that with a little extra effort, actual mass transfer determinations may be carried out with far greater accuracy than can be obtained by using the generalized correlation.

CONCLUSIONS

1. Increase in amplitude or frequency improves mass transfer rates in pulse columns. With high liquid throughputs, no optima in amplitude or frequency are involved. The best operation, i.e., highest mass transfer rates, is obtained by using high amplitudes and high frequencies.

2. Hole size has a much larger effect on the mass transfer rate than does the per cent free area of the plates. In order to permit large liquid throughputs and to obtain high mass transfer rates at the same time, the plates should be drilled with a small hole size and a large fraction free area.

3. Higher throughputs give larger holdup and give better mass transfer rates. Close plate spacing is more desirable but uniform plate spacing is ineffecient. The plates at the inlet section of the dispersed phase serve to break up the dispersed phase into fine droplets, whereas those at the exit section serve merely to maintain the dispersion. To minimize the frictional resistance, the plates should be closely spaced at the inlet end of the dispersed phase and loosely spaced at the exit.

4. The best operation is associated with high holdups accompanied by fine drop dispersion. The highest mass transfer rates are obtained in the vicinity of flooding. This holds true no matter how flooding is approached -- by increasing the amplitude, frequency or by using low hole size and low per cent free area. In this region the H.T.U.'s take up almost a constant value with a variation of ± 35 per cent.

5. Plate wetting conditions have a predominant influence on the mass transfer rates. The same plates can give up to 100 per cent variation

in mass transfer rates depending upon the condition of the plate surface. The effect of the plate wetting is probably larger than that of any single variable.

6. The mass transfer rates are correlated by a polynomial having the following form:

$$HTU)_{od} = 5.9 - (5.85a - 4.17a^2) - (.0375f - .00009f^2) + 18.1h$$
$$+ .0506p + .314s - 4.75\ell \pm 0.31$$

where "$HTU)_{od}$" is in inches

"a" is the amplitude in inches

"f" is the frequency in cycles/min.

"h" is the hole size of the plates in inches

"p" is the per cent free area of the plates

"s" is the plate spacing inches

"ℓ" is the combined liquid flow rates in cubic inches/sq. inch sec.

The expression may be used to predict the H.T.U. values within less than ± 10 per cent for the following range of variables:

amplitude	0.21 - 0.62 inch
frequency	30 - 100 cycles/min.
hole size	1/16 - 3/32 inch
per cent free area	16 - 40
plate spacing	2 - 3 inches
liquid flow rates	670 - 1000 gal./hr.sq.ft.

AREAS FOR FUTURE RESEARCH

We have observed that high mass transfer rates are associated with fine drop dispersion and high holdup, i.e., high interfacial area and long residence time. The operating variables such as the amplitude, frequency, plate geometry, etc., affect mass transfer rates mainly through the drop size. Future research should consequently be directed towards studying the drop size.

We have shown in this work that plate wetting or the condition of the plate surface is more important than any other variable. Fundamental work needs to be done to study the plate surface, such as: defining the plate surface by a standard and simple technique, obtaining a plate surface which will give small drop size and understanding the mechanism of drop formation with respect to the plate wetting condition.

APPENDIX A

EQUILIBRIUM DATA ON THE SYSTEM

MIBK - ACETIC ACID - WATER

Ketone	Water
% Acid	% Acid
1.87	2.85
8.90	11.70
17.30	20.50
24.60	26.20
30.80	32.80
33.60	34.60

Temp. = 25°C
Data of Sherwood(S3)

% Weight Acid	
Aqueous	Org.
2.95	1.79
6.58	4.38
8.98	6.14
12.11	8.77

Temp. = 32.5°C

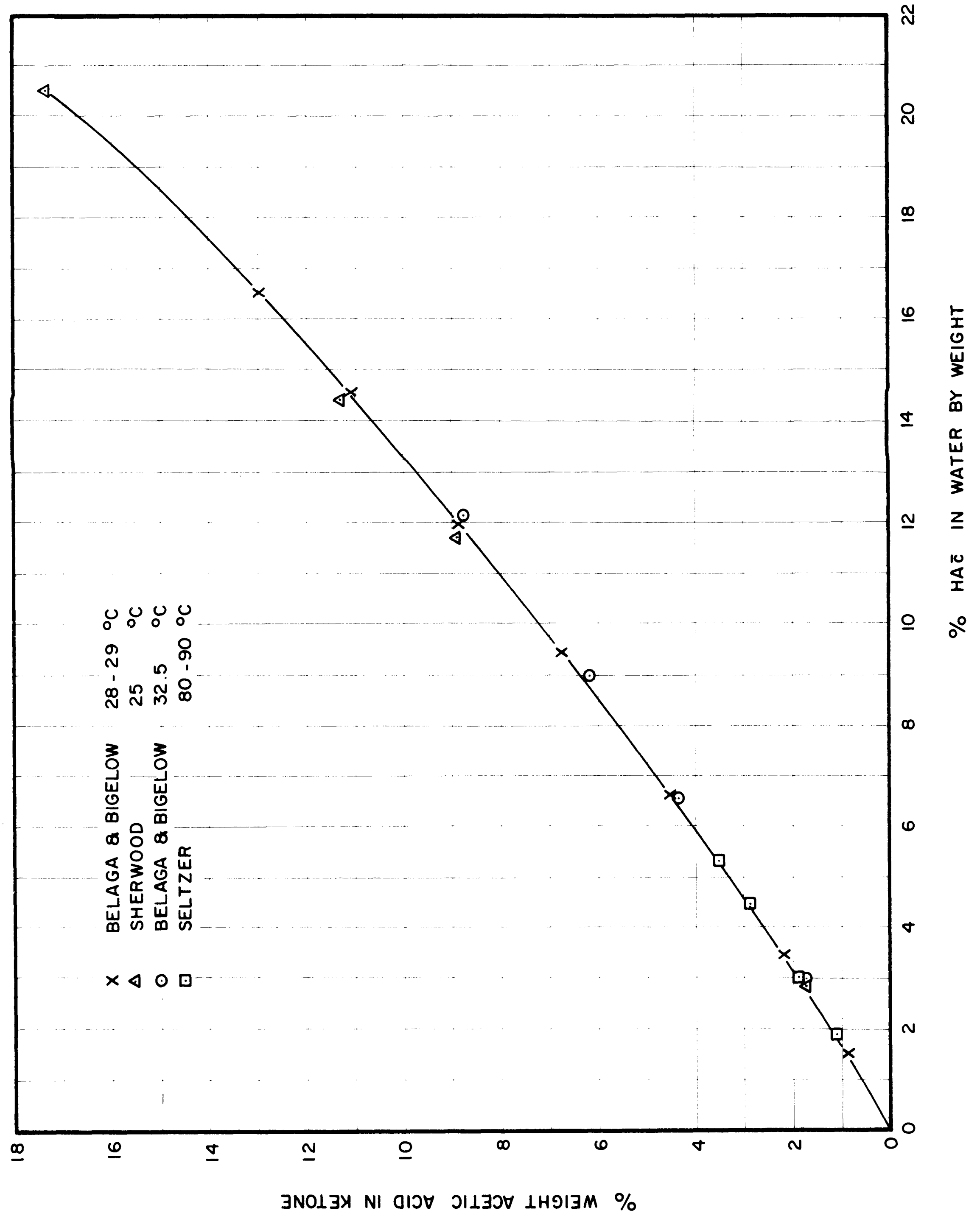
% Weight Acid	
Aq.	Org.
1.51	0.90
3.47	2.22
6.61	4.47
9.40	6.73
11.96	8.82
14.55	11.09
16.52	12.92

Temp. = 28° - 29°C

Data of Belaga and Bigelow(B2)

Normality of Acid in Water	Normality of Acid in Ketone
0.00374	0.00161
0.01061	0.00471
0.02668	0.01205
0.04307	0.02012
0.06066	0.02817
0.10659	0.05021
0.12552	0.05932
0.14421	0.06836
0.20270	0.09762
0.23340	0.11345
0.30920	0.15257
0.41470	0.20590
0.49750	0.25230
0.55380	0.28250
0.74500	0.39170
0.89880	0.47780
1.00000	0.53700
0.90000	0.47970

Temp. = 80° - 90°F
Data of Seltzer(S2)



Comparison of the Equilibrium Data Methyl Isobutyl Ketone - Acetic Acid - Water.

APPENDIX B

MUTUAL SOLUBILITY DATA OF S. C. DOSHI(S2)

METHYL ISOBUTYL KETONE - ACETIC ACID - WATER (80 - 84°F)

Solubility of Water in Ketone	
% Weight Acid in Ketone	% Weight Water in Ketone
0.000	1.830
1.299	2.183
2.258	2.434
3.211	2.514
3.723	3.114
5.526	3.807
7.757	4.704
11.520	6.053
17.330	8.905

Solubility of Ketone in water	
% Weight Acid in Water	% Weight Ketone in Water
0.000	1.696
1.991	1.776
2.481	1.857
2.975	1.913
3.718	1.931
9.230	2.409
18.200	3.669

APPENDIX C

STRAIGHT LINE REPRESENTATION OF THE EQUILIBRIUM DATA

MIBK - ACETIC ACID - WATER

Equation representing the data:

$$y = -0.014 + 0.5456\,x \pm 0.0042$$

where

y = Concentration of acetic acid in the ketone (normality)

x = Concentration of acetic acid in the water (normality)

Normality of water x	Normality of ketone y	y Calculated	Deviation
1.0	0.537	0.5316	-0.0054
0.9	0.4797	0.4770	-0.0027
0.7450	0.3917	0.3924	+0.0007
0.5538	0.2825	0.2881	+0.0056
0.4975	0.2523	0.2574	+0.0051
0.4147	0.2059	0.2122	+0.0063
0.3092	0.1526	0.1546	+0.0020
0.2334	0.1135	0.1133	-0.0002
0.2027	0.0976	0.0966	-0.0010
0.1442	0.06836	0.06465	-0.0037
0.1225	0.05932	0.05280	-0.0065
		Standard Deviation	±0.0042

Data of Seltzer(S2)

APPENDIX D

PROPERTIES OF MIBK - WATER SYSTEM

Density water 20/20	.996 g/cc
Density ketone 20/20	.805 g/cc
Difference in Densities	.191 g/cc
Viscosity ketone 20	.614 cps
Viscosity water 20	1.060 cps
Interfacial tension 20	10.100 dynes/cm.

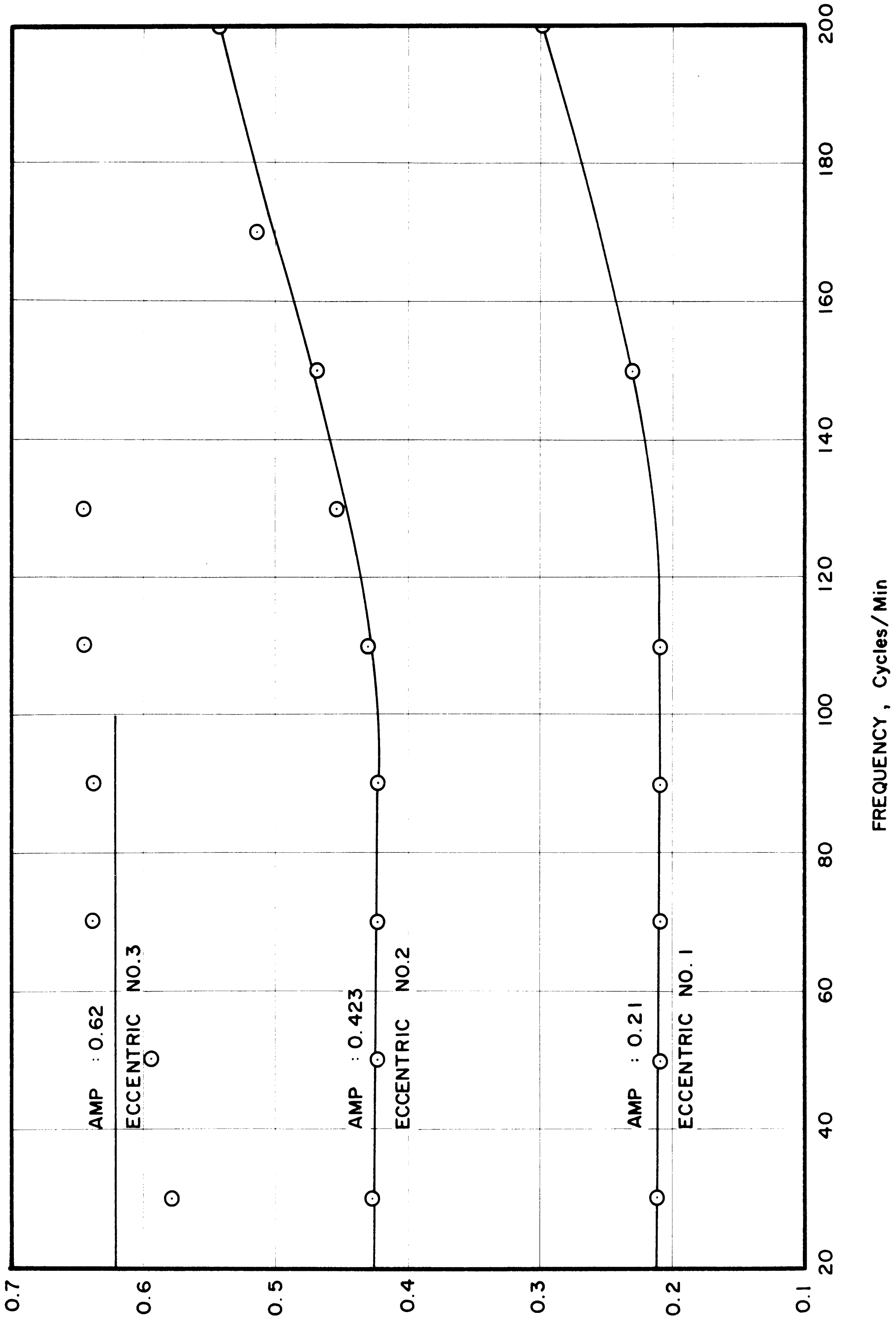
APPENDIX E

CALIBRATION OF 1 INCH BELLOWS

Frequency cycles/min	Column Amplitude, inches		
	Eccentric #1	Eccentric #2	Eccentric #3
30	.21	.428	.585
50	.21	.423	.594
70	.21	.423	.639
90	.21	.423	.639
110	.21	.432	-
130	-	.455	.664
150	.230	.468	-
160	-	-	.664
170	-	.513	-
200	.297	.548	.810

CALIBRATION OF 1-1/2-INCH BELLOWS

Frequency	Column Amplitude, inches				
cycles/min	Eccentric #1	Eccentric #2	Eccentric #3	Eccentric #4	Eccentric #5
20				2.034	
28		.851			
30	.315		1.359	2.02	2.27
40				1.818	
50				1.66	
54	.270		1.273		2.00
56		.698			
60				1.804	
70				1.867	
78			1.31		
80	.307	.837		1.917	2.23
100				2.052	
104	.315	.869	1.368		
120				2.142	
140				2.259	
150			1.58		
152		.918			
154	.387				
160				2.25	
180				2.313	
200		1.107			
202	.522				



Calibration of the 1 inch Bellows

APPENDIX F

TABLE OF MASS TRANSFER DATA

						Flow Rates cc/min.				Concentration Normality (Inlet Ketone was Acid Free)			
	Pulsation			Plate Geometry		Inlet (Nominal)		Outlet		Inlet	Outlet		
Expt. No.	Amp. Inches	Frequency rpm	Plate Spacing Inches	Hole Size Inches	Free Area %	Aqueous	Organic	Aqueous	Organic	Aqueous	Aqueous	Organic	HTU)$_{od}$ Inches
89	0.21	50	2	1/16	24.55	100	180	-	-	1.035	.161	.5025	3.84
1		60						-	-	1.04	.14	.5	3.67
2		80						-	-	1.04	.15	.518	3.08
3		100						-	-	1.04	.141	.5175	2.83
73	.423	40	2	1/16	24.55	100	180	-	-	1.04	.168	.51	3.68
74		50						-	-	1.04	.141	.508	3.36
75		60						-	-	1.04	.14	.5075	3.24
76		80						-	-	1.04	.12	.51	3.00
	.62	30	2	1/16	24.55	100	180	100	174	1.04	.15	.508	3.5
7		40						-	-	1.04	.145	.509	3.37
8		50						-	-	1.04	.145	.509	3.37
9		60						98	190	1.02	.125	.494	3.25
4	.21	60	2	1/16	32.7	100	180	-	-	.9925	.152	.479	3.92
5		80						94	176	.9925	.131	.486	3.26
6		100						92	182	1.00	.116	.484	3.19
77	.423	40	2	1/16	32.7	100	180	98	190	.9925	.139	.476	3.86
78		50						94	188	1.00	.131	.48	3.61
79		60						97	190	.9975	.118	.481	3.28
80		80						95	183	.9975	.122	.489	3.02
10	.62	40	2	1/16	32.7	100	180	95	180	.9975	.132	.481	3.52
11		50						90	196	.9975	.13	.479	3.58
12		60						94	188	.9975	.124	.484	3.27
99	.62	30	2	1/16	16.35	100	180	98	187	.9975	.118	.486	3.19
100		40						97	182	.9975	.141	.494	3.14
101		50						-	-	.9975	.14	.495	3.08
90	.21	60	2	1/16	40.9	100	180	90	184	.9925	.154	.471	4.34
91		80						94	186	.9925	.139	.48	3.64
92		100						96	188	.9925	.13	.485	3.3
93	0.42	40	2	1/16	40.9	100	180	95	181	.9925	.154	.472	4.27
94		50						96	182	.9925	.15	.479	3.99
95		60						-	-	.9925	.146	.48	3.78
96		80						100	192	.9925	.1325	.484	3.38
97	.62	40	2	1/16	40.9	100	180	91	-	.9925	.145	.479	3.81
98		60						97.5	190	.9925	.126	.48	3.52
107	.21	60	2	3/32	16.35	100	180	96	188	.9975	.125	.482	3.42
102	.62	30	2	3/32	16.35	100	180	97	190	.9975	.148	.477	4.01
103		40						97	190	.9975	.145	.49	3.36
104		60						98	183	.9975	.14	.4875	3.38
108	.21	50	2	3/32	24.55	100	180	99	188	.995	.184	.458	5.48
13		60						98	188	.995	.18	.466	5.0
14		80						97.5	184.5	.995	.164	.486	3.81
15		100						97	188	.995	.116	.486	3.1
81	.423	40	2	3/32	24.55	100	180	98	190	.995	.165	.466	4.8
82		50						98	187	.995	.144	.4775	3.87
83		60						99	188	.995	.145	.479	3.82
84		80						100	190	.995	.116	.491	2.88
105	.62	30	2	3/32	24.55	100	180	100	191	.995	.149	.475	4.05
19		40						97	190	.995	.1425	.479	3.74
20		50						97	188.5	.995	.1325	.48	3.57
21		60						96	188	.995	.12	.481	3.29
16	.21	60	2	3/32	32.7	100	180	98.5	190	.995	.19	.461	5.5
17		80						-	-	.995	.161	.468	4.5
18		100						-	-	.995	.139	.476	3.83
85	.423	40	2	3/32	32.7	100	180	96	188	.9975	.16	.461	4.95
86		50						92	187	1.00	.135	.462	4.44
87		60						99	190	.9975	.159	.481	4.04
88		80						96	189	.9975	.125	.482	3.38
106	.62	30	2	3/32	32.7	100	180	96	186	.9975	.158	.471	4.43
22		40						96	192	.9975	.14	.468	4.25
23		50						92	190	.9975	.14	.465	4.18
24		60						92	190	.9975	.1225	.474	3.68
109	0.21	50	3	1/16	16.3	100	180	96	183	1.00	.121	.491	3.02
110		60						95	188	1.00	.1085	.487	2.96
115	.62	30	3	1/16	16.3	100	180	-	-	1.00	.142	.481	3.78
116		40						98	181	1.00	.134	.49	3.25
117		50						95	192	1.00	.116	.485	3.18
25	.21	60	3	1/16	24.55	100	180	96	190	1.02	.128	.485	3.72
26		80						96	190	1.00	.129	.476	3.74
27		100						97	186	1.00	.1325	.485	3.4
118	.62	30	3	1/16	24.55	100	180	-	-	.9975	.136	.47	4.1
31		40						95	190	.9975	.126	.472	3.78
32		50						-	-	.9975	.1325	.4825	3.54
33		60						95	188	.9975	.126	.481	3.28

TABLE OF MASS TRANSFER DATA (CONT'D)

						Flow Rates cc/min.				Concentration Normality (Inlet Ketone was Acid Free)			
	Pulsation			Plate Geometry		Inlet (Nominal)		Outlet		Inlet	Outlet		
Expt. No.	Amp. Inches	Frequency rpm	Plate Spacing Inches	Hole Size Inches	Free Area %	Aqueous	Organic	Aqueous	Organic	Aqueous	Aqueous	Organic	$HTU)_{od}$ Inches
111	.21	50	3	1/16	32.7	100	180	94	188	1.02	.1505	.4575	4.89
28		60						96	184	1.02	.154	.48	4.4
29		80						-	-	1.00	.1325	.47	4.05
30		100						-	-	1.00	.135	.48	3.68
119	.62	30	3	1/16	32.7	100	180	99	190	1.02	.167	.48	4.6
34		40						93	195	1.02	.129	.474	4.2
35		50						-	-	1.00	.125	.47	3.94
36		60						92	190	1.02	.125	.481	3.83
112	.21	60	3	1/16	40.9	100	180	99	185	1.00	.189	.467	5.22
113		80						100	192	1.00	.164	.469	4.67
114		100						100	185	1.00	.16	.48	4.08
120	.62	40	3	1/16	40.9	100	180	98	186	1.00	.174	.479	4.38
121		50						97	188	1.00	.154	.479	4.03
122		60						97	190	1.00	.1475	.48	3.88
128	.21	50	3	3/32	16.3	100	180	98	187	1.00	.135	.4775	3.76
129		60						95	190	.9975	.1305	.4725	3.87
130		80						98	186	.9975	.129	.486	3.29
123	.62	30	3	3/32	16.3	100	180	95	187	.999	.144	.47	4.27
124		40						-	-	.999	.134	.4875	3.38
125		50						96	189	.999	.125	.489	3.16
37	.21	60	3	3/32	24.55	100	180	96	186	1.02	.1675	.471	5.06
38		80						-	-	1.02	.13	.48	3.96
39		100						96	186	1.00	.155	.48	3.61
126	.62	30	3	3/32	24.55	100	180	-	-	1.02	.168	.47	5.38
43		40						95	187	1.02	.165	.472	5.02
44		50						-	-	1.00	.1425	.475	3.96
45		60						96	188	1.02	.1275	.49	3.52
131	.21	50	3	3/32	32.7	100	180	94	189	.9975	.17	.4525	5.48
40		60						-	-	.9975	.159	.46	4.92
41		80						96	190	.9975	.148	.477	4.05
42		100						-	-	.9975	.137	.471	3.7
127	.62	30	3	3/32	32.7	100	180	94	190	.9975	.15	.451	5.12
46		40						96	186	.9975	.154	.4575	4.95
47		50						-	-	1.00	.1725	.48	4.66
48		60						95	190	.9975	.126	.465	4.1
49	.21	60	2	1/16	24.55	66	120	66	127	1.00	.165	.465	4.82
50		80						66	127	1.00	.1575	.479	4.15
51		100						66	126	1.00	.1475	.485	3.71
134	.62	30	2	1/16	24.55	66	120	61.4	128	1.00	.126	.465	4.15
55		40						65.3	127	1.00	.155	.477	4.15
56		50						-	-	1.00	.146	.480	3.9
57		60						67	130	1.00	.135	.486	3.42
52	.21	60	2	1/16	32.7	66	120	66	127	1.00	.165	.463	5.0
53		80						65	121	1.00	.164	.474	4.47
54		100						66	125	1.00	.15	.4775	4.07
135	.62	30	2	1/16	32.7	66	120	64	123	1.00	.1675	.47	4.81
58		40						64	121	1.00	.16	.476	4.65
59		50						-	-	1.00	.145	.47	4.3
60		60						64	123	1.00	.135	.482	3.58
61	.21	60	2	3/32	24.55	66	120	-	-	1.00	.1675	.4525	5.52
62		80						64	124	1.00	.169	.467	4.88
63		100						64	123	1.00	.1475	.4725	4.23
133	.62	30	2	3/32	24.55	66	120	64	129	1.00	.141	.466	4.33
67		40						68	124	1.00	.1675	.479	4.3
68		50						65	129	1.00	.145	.473	4.14
69		60						66	127	1.00	.159	.492	3.54
64	.21	60	2	3/32	32.7	66	120	62	128	1.00	.15	.44	5.82
65		80						-	-	1.00	.1225	.44	5.11
66		100						-	-	1.00	.112	.445	4.69
132	.62	30	2	3/32	32.7	66	120	66	126	1.00	.171	.486	4.86
70		40						62	134	1.00	.129	.455	4.65
71		50						64	122	1.00	.17	.473	4.65
72		60						64	123	1.00	.165	.476	4.38

APPENDIX G

SAMPLE CALCULATIONS AND PROBABLE ERROR

1. Sample Calculations.

Expt No. 5.

Inlet Concentration: Aqueous phase $x_2 = 0.9925$ N
Organic phase $y_1 = 0$

Outlet Concentration: Aqueous phase $x_1 = 0.131$ N
Organic phase $y_2 = 0.486$ N

Equation representing the equilibrium data: $y = -0.014 + .5456\ x$

N_{og} = Number of Transfer Units:

$$\int \frac{dy}{y-y^*} \quad \frac{y_2-y_1}{\dfrac{(y_2^*-y_2)-(y_1^*-y_1)}{\ln \dfrac{y_2^*-y_2}{y_1^*-y_1}}}$$

Calculation of y_2^* and y_1^*

$$y_2^* = 0.5456\ x_2 - 0.014$$
$$= (.5456)(.9925) - 0.014 = 0.529$$

$$y_1^* = 0.5456\ x_1 - 0.014$$
$$= (.5456)(.131) - 0.014 = 0.0575$$

$$N_{og} = \frac{0.486 - 0}{\dfrac{(0.529-0.486)-(0.0575-0)}{\ln \dfrac{0.529-0.486}{0.0575}}} = \frac{.486}{\dfrac{0.043-0.0575}{\ln \dfrac{.043}{.0575}}}$$

$$= \frac{.486}{\dfrac{.0145}{\ln 1.338}}$$

$$= \frac{.486}{.0145/.292} = 9.8$$

$$\text{HTU})_{od} = \frac{\text{Height of column}}{N_{og}} = \frac{32 \text{ inches}}{9.8} = 3.26 \text{ inches}$$

2. Probable Error in the Calculation of H.T.U.

Expt No. 5.

Error in titration:

Titrating solution: 0.25N NaOH

Error in Burette reading: .05 cc

Titrating error: $\frac{(.05)(.025)}{\text{sample size}}$

Probable error in inlet and outlet concentrations:

Inlet aqueous x_2: 0.9925 ± .00125 (10 cc sample)

Outlet aqueous x_1: 0.131 ± .00125 (10 cc sample)

Outlet organic y_2: 0.486 ± .00125 (10 cc sample)

Inlet organic y_1: 0 ± .0005 (25 cc sample)

Probable error in the calculation of y*:

$y^* = .5456\, x \pm \epsilon \quad (\epsilon = \pm\, 0.0042)$

Error in $y^* = (\epsilon_x^2 + \epsilon^2)^{1/2} = \pm\, .0044$

Probable error in H.T.U.:

Numerator: $y_2 - y_1$

Error in numerator: $(\epsilon_{y_2}{}^2 + \epsilon_{y_1}{}^2)^{1/2} = \pm\, .0013$

Denominator: log mean $(y_2^*-y_2)$, $(y_1^*-y_1)$

Error in $y_2^*-y_2$: $= (\epsilon_{y_2^*}{}^2 + \epsilon_{y_2}{}^2)^{1/2} = \pm\, 0.0046$

Error in $y_1^*-y_1$: $(\epsilon_{y_1^*} + \epsilon_{y_1})^{1/2} = \pm .0045$

Error in denominator: $(\frac{.0046^2 + .0045^2}{2})^{1/2} = \pm .0046$

Value of numerator: 0.486

Value of denominator: 0.0496

Error in N_{od}:

$$\epsilon_{N_{od}} = [(\frac{1}{\text{Den.}})^2(\epsilon_{\text{Num.}})^2 + (\frac{\text{Num.}}{\text{Den.}^2})^2(\epsilon_{\text{Den.}})^2]^{1/2}$$

$$= [(\frac{1}{.0496})^2(.0013)^2 + \{\frac{.486}{(.0496)^2}\}^2(.0046)^2]^{1/2}$$

$$= [6.42 \times 10^{-4} + .1950]^{1/2}$$

$$= \pm 0.44$$

$$\text{Probable Error in H.T.U.} = \frac{Z}{N_{od}} \times \epsilon_{N_{od}}$$

$$= \frac{32}{9.8} \times 0.44 = \pm .145$$

$$\sim \pm 0.15 \text{ or approx. } \pm 5\%$$

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NOMENCLATURE

a	Amplitude of the pulse in the column
a	Interfacial area of mass transfer
a	Slope of the operating line
c	Regression coefficient
D	Diameter of the column
f	Frequency of the pulse
f	Fugacity
G	Flow rate of the solvent phase
G'	Flow rate of the solute free solvent
g	Force of gravity
g_c	Mass-Force conversion factor
h	Diameter of the plate perforations
H.T.U.	Height of a transfer unit
H.E.T.S.	Height equivalent to a theoretical stage
K	Overall coefficient of mass transfer
K	Constant
k	Film coefficient of mass transfer
L	Flow rate of the liquid phase
L'	Flow rate of the solute free liquid
ℓ	Combined liquid flow rates
m	Slope of the operating line
m	Constant
p	Area of the plate holes, as percent of the cross sectional area of the column
q	Constant

NOMENCLATURE (CONT'D)

s	Cross sectional area of the column
$\overline{V}_o$	Characteristic velocity
x	Concentration of the solute in the aqueous phase
x	Hold up of the dispersed phase
x	Independent variable
x	Constant
y	Concentration of the solute solvent phase
y	Dependent variable
y	Constant
z	Height of the column
z	Constant

Subscripts

c	Continuous phase
d	Dispersed phase
i	Interface
g,G	Solvent phase
ℓ,L	Liquid phase
oc	Overall continuous
od	Overall dispersed

Greek Letters

ϵ	Error
γ	Interfactial tension
μ	Viscosity
ρ	Density
ψ	Power function