EXTRACTION IN PACKED COLUMNS
PULSED AND UNPULSED

Thesis for the Degree of Ph. D.
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Vito Joseph Sarli
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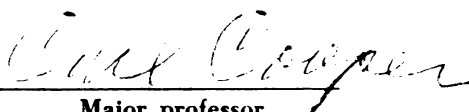
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EXTRACTION IN PACKED COLUMNS

PULSED AND UNPULSED

By

VITO JOSEPH SARLI

AN ABSTRACT

Submitted to the School for Advanced Graduate Studies of
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EXTRACTION IN PACKED COLUMNS, PULSED AND UNPULSED

ABSTRACT

This thesis presents a new method for calculating HTU (height of a transfer unit) in packed columns from the physical properties of the fluids and packing characteristics. To the author's knowledge no method has been presented previously for calculating HTU values without the use of experimental data. The calculations are based on hydrodynamic and mass transfer considerations of droplet, or disperse phase, flow in an immiscible continuous phase. Because of the fundamental nature of the analysis it should be possible to extend the method to other types of extractors such as spray, sieve plate, and baffle columns.

In the experimental work of this thesis good agreement was obtained between the calculated and experimental HTU values for the transfer of acetone between water and carbon tetrachloride phases. The experimental values of HTU in this thesis and values of HTU reported by other investigators, deviated from the calculated values by an average of 30 percent in spite of a 20 fold variation in the values of HTU.

From pulsed column operation the agreement between experimental and calculated values of HTU was of the same order of magnitude. However, the basis for calculating the effect of pulse is not as sound from a theoretical viewpoint as the rest of the calculations.

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In the experimental work on pulsed columns in this thesis the height equivalent to a theoretical stage (HETS) is improved (i.e. lowered) when the pulse rate is increased, when total flow is reduced, and when packing size is reduced. Even at high pulse rates a 50 percent reduction in total flow causes a 20-25 percent reduction in HETS. HETS values are not affected much by changing the flow ratio or by changing the diameter of the column.

At low flow rates better mass transfer was obtained when the phase with the larger volumetric rate was dispersed. At flow rates near the flood point or at high pulse rates, the choice of which phase is dispersed is immaterial.

The method developed in this thesis to calculate HTU can also be used to calculate the flooding velocities in extraction columns. The agreement between experimental and calculated flooding rates is as good as is obtained from currently accepted flood point correlations. However, none of these correlations are extremely reliable. Extraneous effects such as localized packing orientation, bridging (especially in small size packing), or the tendency of droplets to coalesce or to resist coalescence can cause variations of two fold or more in flooding rates.

It was found in this thesis that mass transfer strongly affects the tendency of droplets to coalesce. Transfer from the dispersed to the continuous phase promotes coalescence, while transfer in the reverse direction inhibits coalescence.

Approved _Carl Cooper_

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INTRODUCTION

The importance of liquid-liquid extraction in packed columns for
the separation and purification of chemicals has created an interest
in and a need to understand mass transfer and fluid flow in packed beds.

Liquid-liquid extraction in packed columns involves the counter-
current flow of two immiscible liquids through the packing. One
phase is usually denoted as the continuous phase and the other as
the dispersed phase. The continuous phase fills the interconnected
voids in the packed column and flows through the column as if it
were passing through an irregularly shaped conduit. The dispersed
phase is made up of droplets which pass through the continuous phase
in tortuous paths determined by the distribution of the packing.

The success of packed columns is dependent on the ability of
the packing to modify favorably the flow pattern of the two phases.
It increases turbulence in the continuous phase. It improves the
distribution and increases the holdup of the disperse phase. It
also distorts and sometimes breaks up the dispersed drops in their
hindered movement, thus presenting fresh surfaces for mass transfer.
If the disperse phase is the one which preferentially wets the
packing, the transfer surface may be considerably extended.

The larger the area that the two phases have in common, the
higher is the efficiency of transfer of the solute which must pass
from one phase into the other. The countercurrent liquid-liquid
extraction column could be made more efficient by subdividing the

droplets further so that the surface will be greater than that realized

when this type of apparatus is used in its simplest form. The only

energy available in a simple packed column for maintenance of disper-

sion and agitation is provided by the velocity of the entering streams

and the density difference of the two phases. This energy is small,

usually insufficient to produce optimum subdivision of the droplets,

maintain a high degree of turbulence in the continuous phase, and

counteract the tendency of droplets to coalesce. Interaction of the

two phases with insufficient energy results in a low interfacial area.

A packed column is therefore less effective as a liquid-liquid extrac-

tion column than it is as an absorber or distillation column because

the gravity difference in gas-liquid systems is much greater.

Several types of contacting devices have been developed and

studied to circumvent the disadvantages inherent in continuous liquid-

liquid contactors as a result of insufficient energy in the two streams.

These devices supplement the gravitational energy by mechanical means.

LIQUID-LIQUID CONTACTORS WITHOUT POWER AGITATORS

A significant amount of research in liquid-liquid extraction and
the various types of liquid-liquid contactors (of which the packed
column is one) has been reported. Liquid-liquid contactors have been
classified by Morello and Poffenberger[38], according to whether the
separation of the two immiscible phases is caused by gravity, i.e.
density difference of the immiscible liquids, or by centrifugal force.
Industrially the former type is of greater importance and has received
practically all of the attention of researchers in mass transfer
studies. Liquid-liquid contactors dependent on gravity for separation
of the two phases can be further classified as to whether the energy
is inherent in the streams or is supplemented by mechanical devices.

Centrifugal contactors. The most extensively used extractor
which owes its success to centrifugal forces to enhance mass transfer
and separation of phases is the Podbielniak, Inc., centrifugal con-
tactor. The two liquids are brought into forced contact in a spiral
confinement. A high speed rotor develops the centrifugal force and
causes the liquids to flow countercurrently. In a paper by Bartels
and Kleinman[3] it is reported that one centrifugal contactor is
equivalent to four theoretical stages. Podbielniak, Inc., claims that
as many as fifteen theoretical stages can be realized under certain
ideal conditions. Its main advantage is a short holdup time.

Liquid-liquid contactors which are most extensively used for
research studies are the spray column and the packed column. The

latter, already introduced, possesses many similarities with the spray column.

Spray columns. The spray column is the simplest of the continuous contacting devices. It consists of a vertical empty shell with nozzles for dispersing one of the liquids into fine droplets. The advantages of the spray column are its high capacity, ease of cleaning, and low cost. Its chief disadvantage is the inability to provide adequate transfer in a reasonable height of tower. The packed column generally gives a better efficiency than the spray column without packing[34][44][52]. In the spray column a large fraction of the mass transfer takes place at the nozzle where the drops are formed. The rate of mass transfer diminishes after the droplet leaves the nozzle, due to stagnation of the surfaces formed in the dispersing nozzle[16][31]. The absence of packing results in little opportunity for distortion of the drops, for formation of fresh droplet surfaces, for prevention of back mixing, and for creating turbulence in the continuous phase.

Although the spray column is the simplest of continuous liquid-liquid contacting devices, the number of variables which appear to affect the rate of mass transfer is large. Physical properties of the fluids that exert an influence on extraction rates are viscosity, density, interfacial tension, and diffusivity. Also of prime importance is the direction of mass transfer and the phase which occurs as the dispersed phase. For spray columns it has been reported that the phase having the larger flow rate should be made the dispersed phase; when

the ratio of the two phases is nearly unity the phase receiving the

solute should be dispersed[19][22][30][58]. As a result of increased

holdup, a larger transfer area is obtained by dispersing the phase of

greater flow rate. When mass transfer is from the continuous to the

dispersed phase, a smaller drop size is experienced and coalescence does

not take place as readily as in the reverse situation.

Minard and Johnson[38], in studying the flow capacities of a four-

inch diameter extraction column, present an empirical correlation for

maximum throughput in terms of flow ratio and physical properties of

the fluids. The reliability of the correlation is questioned by

Treybal[57] in view of the small size of the column and the criterion

of flooding used.

Packed Columns. The packed column, which has been introduced

already, has been the subject of many papers.

The performance of packed columns is noticeably affected by the

same variables that determine the efficiency of spray columns. In

addition to the physical properties of the system, the following vari-

ables have great influence on mass transfer rate in packed liquid-

liquid extraction columns:

1. Choice of dispersed phase.

2. Wetting characteristics of dispersed and continuous phase.

3. Size and type of packing.

4. Height of tower and end section design.

5. Direction of mass transfer.

6. Holdup of dispersed phase.

7. Velocity of liquid flow.

For solute transfer between two immiscible liquids in random packed towers the mass transfer area per cubic foot, a, and the mass transfer coefficient, k, cannot be measured separately. These two factors, which determine the capacity coefficient, are often combined into a mass transfer coefficient, ka, based on one cubic foot of packed volume rather than on one square foot of mass transfer area.

The mass transfer coefficient, $k_G a$ or $k_L a$, is related to two other frequently used quantities that give a measure of column performance, namely, the height of a transfer unit, HTU, and the height equivalent to a theoretical stage, HETS or HTS (see Appendix A).

Certain basic trends can be noted from a study of the experimental data published; namely, that the quantities which measure the performance of liquid-liquid contactors appear to depend more on the effect that the variables have on the mass transfer area, a, than on the mass transfer coefficient, k.

Flow rates. The flow rate of either phase affects the over-all volumetric mass transfer coefficients, $k_G a$ or $k_L a$. The principal effect of increasing the flow rate of either phase is to increase the holdup of the dispersed phase and consequently the area for mass transfer. The flow rate of the dispersed phase appears to influence the value of the interfacial area much more than the flow rate of the continuous phase[1][29][52].

Sherwood, Evans, and Longcor[52] studied the extraction of acetic

acid from water by benzene, with benzene as the dispersed phase. The

coefficient increased with increased flow rate of the continuous phase

owing to increased holdup of the dispersed phase, until at the higher

flow rates coalescence caused an actual reduction in the mass transfer

area and volumetric mass transfer coefficient.

Laddha and Smith[33] and Colburn and Welsh[13] measured individual

film coefficients and heights of transfer units. Two pure liquids of

limited solubility were contacted and the approach to saturation of each

phase was calculated in terms of "heights of individual transfer units".

Flow rate of the continuous phase showed little influence on mass trans-

fer coefficients. In every case the value of HTU_d, height of transfer

unit of the dispersed phase, was noted to be constant; whereas HTU_c was

reported to be strongly influenced by the flow rates. HTU_c decreased

as the ratio of dispersed to continuous phase flow rate increased. This

supports the findings of other investigators who have proposed that the

phase of higher flow rate should be made the dispersed phase.

In a later study reported by Leibsom and Beckman[35], in which

diethylamine was extracted from a water phase by toluene, the volumetric

mass transfer coefficient, $k_L a$, increased linearly with an increase in

flow rate of the dispersed phase (toluene). The data indicated only a

slight increase in volumetric mass transfer coefficient for an increase

in flow rate of the continuous phase (water) in contrast to the effect

of increasing the dispersed phase flow rate.

It is also suggested[26][35][36] that 3/8-inch packing is a transition size for droplet behavior. For low dispersed phase rate the performance of the column corresponded to that reported for 1/2-inch packing and at higher dispersed phase rates the values for the mass transfer coefficient were smaller than those reported for the 1/4-inch packing.

Choice of dispersed phase. In some systems with a strong preferential wetting of the packing by the dispersed phase there are no droplets. Under such conditions the dispersed phase flows through the continuous phase and along the packing in the form of a film or rivulets. The performance of liquid-liquid extraction equipment can be very different when the dispersed phase preferentially wets the packing, owing to the small increase in surface area with increased dispersed phase flow rate. For low dispersed phase flow rates the dropwise flow produces a higher interfacial area than the filmwise flow. This is due to channeling and nonuniform distribution of the disperse phase, expecially when the disperse phase flow rate is low. This phenomenon has been reported by several investigators[29][52] whose data indicate a higher volumetric mass transfer coefficient when the phase which preferentially wets the packing is made the continuous phase. It is possible, however, to observe a higher mass transfer coefficient with the preferential wetting phase dispersed when its flow rate is considerably higher than the continuous phase[1][13][30]. For some systems a reversal of the dispersed and continuous phases has been observed when the phase that preferentially wets the packing is initially the dispersed phase. The

reversal occurs with increased flow rate of the dispersed phase owing to displacement of the "nonwetting" continuous phase by the "wetting" dispersed phase[2]. This phenomenon is more likely as flow rates approach flooding conditions.

Packing size. The droplet size and the holdup are dependent on packing size in the column. The holdup and droplet size directly determine the interfacial area and consequently the volumetric mass transfer coefficient.

In general it has been noted that the smaller the packing size, the better the mass transfer[35][52]. Gayler and Pratt[24] have shown that the holdup decreased as the size of the packing was increased. A critical size of packing was reported by several investigators[35][36]; namely, the droplet size was less influenced by packing 1/2-inch or greater and the largest size drop was observed for 1/4-inch packing. For 1/4-inch Raschig rings the droplets rose through the packing as irregularly shaped drops that remained in the interstices of the packing until impacted by one from behind. For 1/2-inch Raschig rings and larger the drops appeared uniform in size and smaller than the drops observed for the 1/4-inch packing. The flow observed for 3/8-inch packing was observed as a transition between the droplet behavior through 1/4-inch rings and that through 1/2-inch rings. At low flow rates of the phases the drops behaved as observed for 1/2-inch and larger packing; however, as the dispersed or continuous phase flow rate was increased the droplet size increased and the behavior was

similar to that observed for 1/4-inch packing. In spite of the increased

drop size for 1/4-and 3/8-inch packings, the increased holdup of the

dispersed phase and the increased agitation and turbulence of the con-

tinuous phase more than offset the adverse effect of the larger drops.

In the study by Leibson and Beckman[35] it was reported that the

column-to-packing ratio should be at least eight in order to eliminate

a "wall effect" that results from a larger than normal void fraction

of the packing. For a specific packing size, all other factors remain-

ing constant, the mass transfer performance is improved as the column

diameter is increased. If the wall effect is large the holdup of the

dispersed phase is usually reduced owing to the abnormal localized

high flow along the wall of the column. A lower retention or contact

time is experienced, together with a reduction in interfacial area

and volumetric mass transfer coefficient.

The performance of a packed column for various kinds of packing

material has been reported by several investigators[39][44][52]. A

column packed with unglazed ceramic Raschig rings indicated a higher

efficiency than one packed with unglazed ceramic Berl saddles. The

reverse effect was noted between ceramic saddles and carbon rings[52].

In the latter situation, wetting characteristics of the packing by

the dispersed phase may have been the controlling factor.

Sieve plate columns. In order to take advantage of the fact

that a large fraction of the mass transfer takes place during and

immediately after droplet formation, sieve plate columns have been

developed[57]. Perforated plates spaced along the length of the

column cause the formation of fresh droplets and coalescence of the
dispersed phase prior to passing through the succeeding plate. The con-
tinuous phase is caused to flow across the plate and through a spout to
the next plate. In order to obtain reasonable success in experimental
columns of this type it is essential that proper flow rates (a very
narrow range) be maintained for the specific plate design under consid-
eration.

LIQUID-LIQUID CONTACTORS WITH POWER AGITATORS

Mixer-Settlers. The conditions for more efficient operation and performance of phase contactors can be improved by supplying additional energy for agitation and phase dispersion. When the energy is properly supplied both mass transfer area and mass transfer coefficient are substantially increased; and mass transfer capacity will in many cases be several times greater than can be realized without the application of external energy.

The mixer-settler, using alternate chambers in series for agitation and decantation of the immiscible phases, was the first attempt at increasing mass transfer capacity. With a counterflow arrangement, equilibrium in each mixer can be easily reached and the performance of such equipment can be readily calculated. With a large number of contact stages, the mixer and settler arrangement is too bulky and expensive for practical application.

Scheibel Columns. A modified mixer-settler device was developed and studied by Scheibel et al.[45][46][47]. The unit consisted of a vertical column with a central rotating shaft on which were mounted stirrers. The tower was equipped with packed sections between the stirrers. The packed sections served as calming sections where separation of the phases took place. Each combination of mixing and calming sections was equivalent to one stage. Scheibel columns often gave more than one theoretical stage for a combination of stirrer and calming section owing to the mass transfer that takes place in the packed section.

The packed section, as well as serving as a calming section, prevents backmixing and internal circulation. The height of the packed section must be adequate to disengage the two phases for the rotor speeds used. As the rotor speed is increased the dispersion becomes finer with a corresponding increase in mass transfer area. With very high rotor speed emulsification or excessive backmixing is experienced, limiting the successful performance of the column.

The flow capacity of such columns is strongly dependent on the rotor speed and is somewhat lower than the maximum reported in packed columns. The same factors that are considered for conventional packed columns in choosing the dispersed phase are important for mechanically agitated columns and have been considered by the investigators cited. Other variables that have been studied are: rotor speed, height of mixing section, height of calming section, and total throughput.

Pulsed Columns. The pulsed column is another arrangement for combining countercurrent action and mechanical agitation. The mechanical agitation is superimposed as an up-and-down motion over the entire volume of the liquids at the same time that the liquids flow countercurrently as in a conventional liquid-liquid extraction column. Mechanical agitation may be supplied by means of a piston and cylinder located outside the column. Construction within the column may take any of the conventional forms for performing the dispersing and coalescing operations. For a series of sieve plates the liquids are forced through the small perforations causing a fine dispersion. In a packed

column the droplets are dispersed by the forced contact with the packing,

and increased turbulence is experienced by the continuous phase. Since

the mechanical energy is caused by the pulse generator the intensity of

agitation can be controlled by regulating the frequency and amplitude of

reciprocating motion.

Sieve Plate Columns. In 1935, van Dijck[17] was granted a patent

for a pulsed column with a number of perforated plates so constructed

that they could be moved up and down. The plates were not provided

with downcomers or risers, so the continuous phase had to pass through

the perforated plates. No appreciable flow was experienced when the

pulsing mechanism was not in operation. Another arrangement described

by van Dijck[17] required that the plates be fixed and the liquids

pulsed. The latter design arrangement was investigated and reported in

numerous articles.

An excellent report was presented by Sege and Woodfield[50] who

investigated a large number of variables involved in the operation of

sieve plate columns. The system investigated was a 3-inch diameter

sieve plate column in which uranyl nitrate was extracted from water

with tributyl phosphate. The effects of operating conditions and sieve

plate design on extraction and flow capacity were discussed.

For the pulsed sieve plate column three distinct types of phase-

dispersion behavior were observed as a function of throughput rate and

pulsing conditions. Mixer-settler type operation occurred at low

throughputs and low pulsed frequencies. This type of operation was

characterized by a complete disengagement of the two immiscible phases between adjacent sieve plates. The less dense phase was dispersed during the upward movement of the fluids and the dense phase was dispersed during the downward movement.

The emulsion type operation was experienced at higher throughputs and frequencies. It was characterized by uniform dispersion of the dispersed phase with no separation of the phases into distinct layers between the plates. The mass transfer capacity of the column was increased several times in this region because of increased interfacial area of contact and high degree of turbulence.

At still higher throughputs and frequencies unstable operation was encountered. In this region the column flooded and the mass transfer capacity was reduced owing to coalescence of the dispersed phase in various regions of the column.

In the same paper Sege and Woodfield reported that at low frequency and amplitude the flow capacity of the column was equal to the pulsed volume. At higher frequencies and amplitudes the flow capacities were found to be less than the pulsed volume, and after a maximum flow was attained the throughput of the column was reduced with further increases in pulsed volumes. For a given flow rate two regions of flooded conditions existed, one owing to insufficient pulsing and the other to excessive pulsing.

Performance of the pulsed sieve plate column was found to be relatively insensitive to variations in flow rate provided that

"emulsion type" dispersion was maintained. An exception occurred at high and low flow rates where an increased value of HTU was experienced.

The same considerations that applied to the choice of disperse and continuous phase for packed columns appear to hold for the pulsed sieve plate column operation.

In another investigation reported by Sege and Woodfield on a 23.5-inch column[51] a louver plate redistributor was discussed to prevent channeling of the denser fluid at the top of a column.

In a paper by Cohen and Beyer[12], the performance of a pulsed perforated plate column was discussed and data presented. The column used was 1 inch in diameter equipped with ten perforated plates at 2-inch intervals. The system studied was iso-amyl alcohol-boric acid feed solution and distilled water.

The data presented showed trends similar to those reported by Sege and Woodfield[50], i.e. the values of HETS (height equivalent to a theoretical stage) was relatively insensitive to flow variations at the higher pulse frequencies (characterized by emulsion type operation). A critical frequency-pulse combination was discussed above which little improvement in extraction performance was evident. Performance of the column with pulse indicated that a reduction in height to one half or one third that required without pulse was possible. Higher rates of extraction were observed with water as the continuous phase.

In a later paper presented by Edwards and Beyer[18], flooding characteristics of a perforated pulsed column were discussed. An analysis of column operation led to derivation of an equation for

predicting conditions of inadequate pulsing in the perforated plate column.

Chantry, Von Berg, and Wiegandt[11] reported the data for some exploratory runs with a 1.57-inch diameter pulsed sieve plate column. The few data reported indicated trends similar to those reported by the investigators already mentioned. An HETS as low as 0.360 feet was obtained for the extraction of acetic acid from methyl isobutyl ketone with water under pulsed conditions.

Griffith, Jasny, and Tupper[26] used a two-inch diameter sieve plate column, 140 inches long, to carry out the separation of cobalt from nickel by extraction with methylisobutyl ketone. Over-all HTU values from 1.1 inches at low flow rates to 34 inches at high flow rates were reported, compared with 24 and 57 inches respectively for the system operated as a spray column. The data reported were somewhat erratic, probably as a result of the complex multicomponent system investigated.

Belaga and Bigelow[4] used the system water-acetic acid-methyl isobutyl ketone. The evaluation of pulsed columns was carried out in a sieve plate column 45 inches long by 1 1/2 inches in diameter. The plates spaced at 1-inch intervals were drilled with 1/32-inch holes, to give 23% free area. HTU values were found to range from 2.63 to 6.25 inches. The product of frequency and amplitude was considered to be a measure of the rate of pulsing. The amplitude and frequency were varied from 1/8 to 2 inches and 20 to 80 cycles per minute respectively.

The study was made with the aqueous phase dispersed and at a fixed flow ratio. Although the data were indicative of the same trends previously reported in this review, considerable variation existed within a family of curves.

Thornton[54] investigated the performance of a pulse sieve plate and packed column using the system toluene-acetone and water. Two types of pulsing units were discussed. Advantages were pointed out for a unit that relies on a pocket of air or inert gas to isolate the pulsing mechanism from the process liquors, over a unit that transmitted the pulse directly to the process fluids. Typical plots utilizing experimental data were presented for mass transfer performance and maximum throughputs.

Pulsed Spray Columns. The performance of a spray column under pulsed operation was reported by Billerbeck et al.[6] The system investigated was methyl isobutyl ketone-acetic acid and water in a 1.5-inch diameter column. The performance indicated as much as eight-fold reduction in HTU as a result of pulsing.

Pulsed Packed Columns. The application of pulsation to liquid-liquid extraction in packed and sieve plate columns has been carried out in a series of investigations at Cornell University. In a publication by Chantry, Von Berg, and Wiegandt[11] reporting the results of these studies the optimum operating conditions of pulse frequency and amplitude for one 3-component system at constant feed rate, the effect of varied feed rate, and the effect of pulsation on flooding capacity were cited.

The packed column contained a 27-inch section with dumped 1/4-inch porcelain Raschig rings. The system studied was methyl isobutyl ketone-acetic acid and water. In nearly all the runs, 20% by weight acetic acid solution in water was extracted by neutral solvent as the dispersed phase.

It was reported that efficiency of the column increased with increased dispersed phase flow rate, and was independent of continuous phase flow rate when the system was pulsed. For constant flow rates, performance of the column measured in HETS, height equivalent to a theoretical stage, passed through a minimum as either the pulse frequency or amplitude was increased. An HETS of 3.2 inches was reported.

Two runs were made at flooding conditions with pulse, and compared with an unpulsed flooding run. The data showed a reduction in maximum throughput when the column was pulsed.

In an earlier study reported by Feick and Anderson[21], benzoic acid was extracted from toluene with water, the latter being the continuous phase fluid. The column employed was 36 inches long and 1 7/16 inches in diameter. Two types of packing material were used, 1/2-inch stainless steel McMahon saddles and 3/8-inch ceramic Raschig rings. The pulse amplitudes ranged from 1/16 to 1/4 inch and the frequencies from 200 to 1000 cycles per minute.

The increased values of over-all mass transfer coefficients as a result of pulsing were considered to be caused either by the increase in interfacial area (owing to finer drops and increased holdup) or to the additional turbulence and its influence on the mass transfer

coefficient. The investigators assumed that the pulsing action had less influence on turbulence in the droplets than in the continuous phase. By making further runs with a solute, namely acetic acid, where the major diffusional resistance was in the dispersed toluene phase, the same order of increase in the mass transfer coefficient was observed as in the case of benzoic acid. On this basis it was concluded that the main effect of pulsation was to increase the interfacial area of contact.

Mention was also made of the fact that maximum flow capacity of the pulsed column was reduced as a result of pulsation.

Schuler[48], in a study using the system carbon tetrachloride — acetone and water, investigated the effect of pulsation in a two-inch packed column. The height of a theoretical stage was reduced from 23.6 inches to 6.3 inches as a result of pulsation. Callihan[10], working with the same system and columns of two sizes, attained 14 times as many theoretical stages with pulsation than with unpulsed operation. It was also demonstrated that HETS varied less with flow ratio then HTU. Increasing the column diameter by a factor of 2.5 did not significantly change the HETS provided that the columns were operated at the same conditions. This was due to elimination of chanelling by pulsation.

The influence of mass transfer on maximum throughput capacity was found to be very important. Higher throughputs were experienced because mass transfer was taking place, and the large effect of mass transfer made it difficult to measure the much smaller influences of other variables.

FLOODING IN PACKED COLUMNS

A number of investigators have presented data for maximum flow rates

in packed columns, and have correlated their results by dimensional

analysis[7][8][14][15][28]. Flooding conditions, according to the des-

cription presented by these investigators, occurred when both inlet

streams were simultaneously tending to overflow into the outlet tubes of

the opposite phases. Also it became difficult to distinguish a contin-

uous from a discontinuous phase in the column at flooding.

Ballard and Piret[2], on the other hand, presented "transition

point" data and reported that three types of behavior can occur, namely

flooding, phase reversal, and slugging. The so-called "transition point"

data were approximately 50 percent below the flooding rates obtained by

the other workers. According to Pratt[43], the transition point data

measured by Ballard and Piret correspond to the upper limit of the

region of rapidly increasing holdup.

Gayler, Pratt, and Roberts[23][25] described three regimes of flow

in packed liquid-liquid extraction columns for packing sizes greater

than 1/2 inch. In the region of linear holdup, the holdup increases in

proportion to the dispersed phase flow rate. In the region of rapidly

increasing holdup, the holdup increases sharply with slight increases

in the disperse phase flow rate; this is evident above holdups of about

10 percent, and is interpreted by Pratt and co-workers as due to

hindered settling of the droplets. The third region is the region of

constant holdup, where coalescence of droplets occurs and droplet

shapes are changed in such a manner that the throughput can increase

without further increase in holdup. This region is terminated by the

flood point.

Very little data exist on the affect of nonequilibrium distribution

of solute on the flood point. Good evidence is presented by Callihan[10]

and other investigators[24][30][54][55] that markedly different flood-

points are encountered for solute transfer. The direction of transfer

also affects the flood point.

Pratt and co-workers from A.E.R.E. Harwell, England, in recent

years have contributed several papers in which a fundamental approach is

presented for liquid-liquid extraction columns. Certain of their results

have been mentioned above. However, the real importance of their studies

is the presentation of hydrodynamic considerations basic to column oper-

ations. Further considerations of their works are discussed in a later

section of this thesis.

SCOPE OF THE INVESTIGATION

The investigations carried out in this research are divided into three areas of interest:

1. Investigation of certain previously overlooked phenomena that affect the maximum capacities of extraction columns. These include the effect of deposits that adhere to the packing elements, the influence of mass transfer, and the direction of mass transfer.

2. Investigation of the effect of certain variables on HETS. These variables include flow rates, flow ratios, pulsation rates, position of the interface, and packing characteristics.

3. Presentation of a theory, supporting calculations, and experimental data which demonstrate a general approach for the estimation of mass transfer in extraction.

THEORY AND CALCULATIONS

The measure of performance of liquid-liquid extraction equipment requires the determination, by measurement or calculation, of two types of information, namely flooding velocities and mass transfer characteristics. Flooding velocities are correlated in terms of hydrodynamic principles and the physical properties of the liquids under consideration. Because of the complex mechanisms of countercurrent flow in columns randomly packed with a dispersing medium, the correlations normally presented are empirical or, at best, guided by dimensional analysis. The correlations for flow in packed extraction columns, expressed as maximum allowable throughput, i.e. flooding velocities, are directly applicable in designing for the diameter of a column.

The required height of a column is dependent on the difficulty of transfer of the solute from one of the immiscible phases to the other. In extraction columns the difficulty of transfer is measured by the number of transfer stages or equilibrium stages. Because of the complex nature of mass transfer, research on towers has led to empirical correlations. The correlations are useful for limited situations in design problems.

In general the method for representing extraction performance in a packed column makes use of at least one of the related terms commonly referred to as the mass transfer coefficients, K_L or K_G; the height of a transfer unit, HTU; the height equivalent to a theoretical stage, HETS; or the height of a theoretical plate, HTP. The height of a column can

be fixed as the product of the number of theoretical plates, NTP, and the height of a theoretical plate, HTP. The latter value is determined experimentally, as it is not easy to calculate it from "a priori" arguments.

The relationships between over-all mass transfer coefficient, height of transfer unit, and height of a theoretical plate are derived in Appendix A. Some of the more useful equations are presented below:

$$\left(K_L/K_G\right) = D$$

$$\left(1/K_G a\right) = \left(D/k_L a\right) + \left(1/k_G a\right)$$

$$\left(1/K_L a\right) = \left(1/k_L a\right) + \left(1/D \; k_G a\right)$$

$$HTU_{OL} = L/K_L a \; A; \quad HTU_L = L/k_L a \; A$$

$$HTU_{OG} = G/K_G a \; A; \quad HTU_G = G/k_G a \; A$$

$$HTU_{OL} = HTU_L + \left(L/GD\right)\left(HTU_G\right)$$

$$HTU_{OG} = HTU_G + \left(HTU_L\right)\left(DG/L\right)$$

$$HTP = \ln \; \left(L/GD\right)/K_G a \; A \left[\left(1/G\right) - \left(D/L\right)\right]$$

$$NTP = \log \left[\left\{\left[X_2 - \left(x_1/D\right)\right]/\left[x_1 - \left(y_1/D\right)\right]\right\}\left[1 - \left(L/GD\right)\right] + \left(L/GD\right)\right]\Big/\log\left(DG/L\right)$$

<u>Mass Transfer Coefficients</u>. It is difficult to measure the individual mass transfer coefficients, k_L and k_G, and the mass transfer area per unit volume of tower, a. Instead, the factors $k_L a$ and $k_G a$ have been evaluated experimentally as single combined quantities. It is even difficult to measure individual combined quantities $k_L a$ and $k_G a$; therefore most experimental data has been reported in terms of over-all volumetric mass transfer coefficients, $K_L a$ and $K_G a$.

<u>Height of a Transfer Unit</u>. Although the HTU is closely related to the mass transfer coefficient, it is simpler to visualize. Its

dimension is simply length and its magnitude does not vary over wide limits. In packed towers the individual film coefficients, k_Ga and k_La, increase with G and L; however, the ratios G/k_Ga and L/k_La, and therefore the transfer units, are nearly independent of flow rates.

The HTU can be stated as being that height of column which results in a change in concentration equal to the driving force.

Height Equivalent to Theoretical Stage, HETS. The theoretical stage concept considers the tower to be subdivided into a number of equilibrium contacts or theoretical plates, NTP or NTS. When the column height is divided by NTP or NTS, HTP or HETS is obtained. The NTP in terms of end concentrations was originally derived for gas absorption by Kremser[32] and modified by Souders and Brown[53].

The primary advantage of HETS over the HTU concept is that for fully developed turbulent flow the value of HETS is nearly independent of the flow ratio[10]. High throughput flow rates and pulsed column operation represent conditions at which fully turbulent flow is developed.

Calculations of Tower Performance From Theoretical Considerations

Instead of using an empirical approach to tower research, this thesis proposes to use theoretical calculations based on an understanding of the important variables on mass transfer. It is desirable whenever possible to calculate performance from theoretical considerations. Experimental performance data is then taken to verify or test the theoretical considerations.

The problem of calculating the amount of extraction in a packed

column leads to the necessity of accounting for the hydrodynamics of packed columns and the mass transfer across interfaces. Any theory that is proposed must take into account the resistance associated with the continuous phase, the resistance associated with the disperse phase, and the effective area for mass transfer.

The approach formulated here is an attempt to calculate the individual resistance of films expressed in terms of k_G and k_L. Also, an effective transfer area calculation is proposed based on average drop diameters determined from free fall considerations and the quantity of liquid holdup in the tower. The effects of velocity on each of the resistances and transfer area are included.

In general the packed column is considered to be analagous to a series of short spray columns, with lengths equal to the distance between packing contacts. The hydrodynamic principles which normally apply to spray columns are modified for the presence of packing. In the present development of the theory for calculating the performance of packed columns, it is assumed that the continuous phase is always that phase which preferentially wets the packing. The disperse phase therefore moves through the packing in the form of droplets. This leads to a limitation which will be discussed later.

Hydrodynamic Considerations

Terminal Diameter, d_o. In packed liquid-liquid extractors free fall or rise of droplets through a continuous phase is always encountered to some extent. In a spray column no packing is present and the drops rise or fall unimpeded. In packed columns or sieve plate columns the

drops are in intermittent free fall. In the case of unimpeded free fall the drop rapidly attains its terminal velocity. In the case of impeded free fall the droplets accelerate and decelerate, and the average velocity is some fraction of the terminal velocity.

For spray and sieve plate columns the drop assumes a size such that the surface tension forces counterbalance the drag or friction forces. This implies that large drops ensuing from inlet openings break up as the terminal velocity is approached. Small drops will tend to coalesce if contact occurs between them. On the average it can be expected that each drop will assume some equilibrium size that satisfies the drag force resulting from its terminal velocity. For packed columns the average velocity is some fraction of the terminal velocity.

Consequently the final drop size is somewhat larger than that experienced in spray or sieve plate columns. This is a direct result of the reduced drag experienced at reduced velocity.

In a paper by Pratt and co-workers[25], the following relationship is recommended for the characteristic diameter.

$$d_0 = .92 \sqrt{\gamma / g \Delta \rho}$$

Pratt and co-workers refer to the characteristic diameter as the actual diameter of a drop emerging from a packed section when the continuous phase flow rate is zero, and the disperse phase flow rate is so small that only a few drops are falling through the packing. The form of the relationship can be justified by assuming that the surface tension effects counterbalance the drop weight as it falls through the packed space.

This relationship was determined from extensive measurements of drop diameters of several immiscible systems with different physical properties.

An expression of the form proposed by Pratt and co-workers with an undetermined constant can be derived as follows:

a. Large drops tend to break up at the terminal velocity because forces tending to distort them are large relative to the forces holding them together.

b. The force which holds them together is proportional to the interfacial tension and the drop circumference, $\pi d_0 \gamma$.

c. The force which distorts the drop is proportional to the drag on the drop. The value at the terminal velocity is $(\pi/6) d_0^3 \Delta \rho$.

d. The critical distortion at which the drop breaks up occurs when the distortion forces are a certain proportion of the forces holding the drop together.

$$\pi d_0 \gamma = C (\pi/6) d_0^3 \Delta \rho$$

Solving for the diameter d_0, the resulting expression is

$$d_0 = C\sqrt{\gamma / \Delta \rho g}$$

Terminal Velocity, v_t. The terminal velocity is calculated from drag coefficients for the free settling of spheres with the characteristic diameters[25][42]. Stokes' Law relates the terminal velocity to the characteristic diameter of spheres. The drag force, F_d equals

$3 \pi \mu v d_0 / g_c$.

Terminal settling velocity: $v_t = \dfrac{d_0 \Delta \rho g}{18\mu}$

F_d, drag force, lbs

g, gravity constant, ft/sec^2

v_t, terminal velocity under action of gravity, ft/sec

μ, viscosity of continuous phase, lbs/ft-sec

$\Delta\rho$, difference in density between spheres and continuous phase, lb/ft^3

v, velocity of drop

Characteristic Velocity, v_0. The velocity of a drop falling unimpeded attains the terminal velocity. In packed columns the average velocity of a drop is less than the terminal velocity. At zero continuous phase flow rate and small dispersed phase flow rate, the average velocity of fall or rise of a drop through the packing is referred to as the characteristic velocity, v_0. The distance of free fall in packed columns is less than the nominal packing size. Pratt and co-workers[25] use drag coefficient data for calculating the average rate of fall or rise through a distance equal to 0.38 times the nominal packing size minus the characteristic drop diameter, d_0. This checks well with measured velocities except when the drop diameter is close to or greater than 0.38 times the packing size.

Pratt's approach has merit since the characteristic velocity cannot exceed the terminal velocity. The use of $(0.38\, d_p - d_0)$ to express the average distance traversed by drops between collisions seems quite arbitrary. Its use is justified because it fits the experimental data quite well when the characteristic velocity is more than 25 percent of

the terminal velocity.

Some unexplained inconsistencies in the theory are:

a. If the distance between collisions with the packing is 0.38 d_p, free fall is 0.38 d_p-d_o; the rest of the fall, d_o, cannot be calculated by Stokes' law.

b. The acceleration used in the theory presented by Pratt and co-workers is only that of the disperse phase. The continuous phase is also accelerated.

c. No attempt was made to include the drag between the drop and packing when free fall does not occur. An equation of the form:

$$v_0 = k_1 d_P \left/ \left[\left(d_0 / k_2 d_P^n \right) \left(\mu / g \Delta \rho \right) + \left(k_1 d_P - d_0 \right) \middle/ v_s \right] \right.$$

can be written to include the influence of wall drag. The value $k_1 d_p$ represents an average distance traversed by the drop in free fall and friction type flow between collisions with packing. Here, v_s is the average velocity of fall through the distance $k_1 d_p$-d_o. The term $(k_2 d_p^n)$ $(g \Delta \rho / \mu)$, from the modified Poiseuille equation, represents the velocity with which the drop flows by friction type flow after collision with the packing.

In spray columns the characteristic velocity of drops is essentially the terminal velocity calculated from Stokes' Law. In sieve plate columns the characteristic velocity is the average velocity of a drop falling a distance equal to the plate spacing. In many sieve plate columns the terminal velocity calculated from Stokes' Law should be applicable. An expression which relates the characteristic velocity to

the Stokes' Law terminal velocity is derived in Appendix B.

Holdup, x. The amount of the dispersed phase present in the tower
is its holdup. In packed columns as in spray columns the holdup increases
with increasing flow of the dispersed or continuous phase. For packed
columns the volume fraction of column occupied by the packing is repre-
sented by the value, $(1-\epsilon)$. ϵ represents the volume fraction of the voids.
The term x is that fraction of the voids occupied by the dispersed phase.
It follows that ϵ x represents the fraction of column occupied by the
dispersed phase and $(1-x)\epsilon$ that fraction of column occupied by the con-
tinuous phase.

Beckman and co-workers[37][59] describe several types of dispersed
phase holdup in packed columns: free, operational, and total holdup.
Free holdup is that which can be drained readily from the packing. Opera-
tional holdup is that portion of the dispersed phase holdup which is
active in mass transfer. According to the above authors no simple
"operational" dispersed-phase flow holdup exists. They encountered a
complex dispersed phase movement, which involves part of the permanent
holdup depending on the previous history of the flow rates. Total holdup
is, of course, the sum of permanent and operational holdup.

An approach analogous to spray columns and to the equations proposed
by Pratt and co-workers[23][25] can be used. They considered that the
normal holdup was involved in the mass transfer.

Two general bases for calculation are:

Case 1: This assumes that all drops are of characteristic diameter,
d_o, and fall at a relative velocity equal to the characteristic velocity.

This is expressed as:

$$V_d / x + V_c / (1-x) = \epsilon v_0$$

and

$$X = \frac{-(V_c - V_d - \epsilon v_0) + \sqrt{(V_c - V_d - \epsilon v_0)^2 - 4 \epsilon v_0 V_d}}{2 \epsilon v_0}$$

V_d, superficial velocity of dispersed phase based on empty column, ft/hr.

V , velocity of continuous phase based on empty column, ft/hr

x, fraction of dispersed phase

$1-x$, fraction of continuous phase

ϵ , void fraction of tower

v_0, characteristic velocity

Case 2: Pratt and co-workers[25] recommend:

$$V_d / x \quad V_c / (1-x) = \epsilon v_0 (1-x)$$

At very low flow rates the two cases are identical. Also, it is to be noted that at very low flow rates the conditions used to define the characteristic velocity and terminal characteristic diameter of droplets are approached. At finite flow rates the drop diameter is a function of flow ratio and holdup.

At moderate flow rate, x = 0.1 to 0.2, Pratt and co-workers claim that Case 2 fits measured holdup better. At flooding rates these cases carried to their logical conclusions give flooding rates as a function of flow ratio. For the derivations, refer to Appendix C.

At flooding:

Case 1 - $V_d/x + V_c/(1-x) = K = \epsilon v_0$

$V_c = K(x - x^2)/(R - Rx + x)$

$x = \left(R - \sqrt{R}\right)/\left(R - 1\right)$ where $R = V_d/V_c$

Case 2 - $V_d/x + V_c/(1-x) = v_0 \epsilon (1-x)$

$V_c = Kx(1-x)^2/R - Rx + x$

$x = \left(-3 + \sqrt{1 + (8/R)}\right)/\left[(4/R) - 4\right]$ where $R = V_d/V_c$

Case 1 leads to the sum of the square roots of the two flow rates being constant at flooding. This has often been reported.[10][19] Case 2 leads to a more complicated relation; Pratt recommends not using it for packed columns at flooding conditions.[43] Actual flooding rates apparently are closer to Case 1, both in absolute value and in variation with flow ratio.

For both cases the average distance of fall, $s = 0.38 \, d_p - 0.92\left(\dfrac{\gamma}{\Delta\rho g}\right)^{\frac{1}{2}}$ must be greater than zero, i.e., $d_p \geqq 2.42 \, (\gamma/\Delta\rho g)^{1/2}$

Mean Drop Diameter, d_{vs}. It was shown earlier that the equilibrium drop diameter is related to the balance of the drag forces and the surface tension forces. A terminal or characteristic drop diameter, d_0, was defined at the conditions of zero continuous phase flow rate and low dispersed phase flow rate. In order to evaluate the drop diameter at flow rates other than near zero, Gayler and Pratt[24] propose the relationship $d \propto 1/v$. There is some merit for this because it is shown by Gayler and Pratt[24] that the characteristic diameter, d_0, and characteristic

velocity, v_o, have an inverse relationship. On this basis the mean drop diameter at finite flow rates, d_{vs}, may be expressed in terms of d_o and v_o,

$$d_{vs} = d_o \, v_c \big/ \bar{v}$$

Pratt and co-workers[24] define $\bar{v}$ as the mean velocity of the droplets relative to the stationary packing. Expressed in terms of V_d, ϵ, x, the mean drop velocity as defined by Pratt and co-workers is, $\bar{v} = V_d/\epsilon x$. The consequence of the above relationship is that the drop diameter increases with increasing V_d. At low values of V_d the droplets free themselves from the packing before collision occurs with another drop. At higher dispersed phase flow rate, V_d, there is greater probability of impact before the drops free themselves. Coalescence takes place and a larger drop results.

Although Pratt and co-workers ignored the effect of the continuous phase flow rate, in this thesis an additional correction for the continuous phase is made. We consider that a droplet has equal difficulty in freeing itself from the packing when the continuous phase superficial velocity, V_c, is increased. For this thesis the mean drop velocity is not relative to the stationary packing but, rather, relative to the continuous phase flow rate. Expressed in terms of V_d, V_c, ϵ and x, the mean drop velocity relative to the continuous phase is,

$$v = V_d \big/ \epsilon x \;+\; V_c \big/ \epsilon \, (1-x)$$

The actual drop diameter, d_{vs}, is

$$d_{vs} = d_0 \; v_0/v$$

Interfacial Area of Contact, a. Since the specific surface (i.e. the surface area per unit droplet volume) of a drop of diameter, d_{vs}, is $6/d_{vs}$, the area of contact in a packed column per unit volume of column is expressed by:

$$a_d = 6 \, \epsilon x / d_{vs}$$

For spray towers the superficial area is given by:

$$a_d = 6 x / d_{vs}$$

Mass Transfer Considerations

Mass Transfer in Packed Beds. In this section we are concerned with the calculation of the mass transfer rate in a liquid-liquid extraction column. For this purpose it is proposed to use a general form of mass transfer factor introduced by Ergun[20]. In the development put forth by Ergun the existence of analogies between mass-transfer rate and pressure loss was demonstrated. Equations were developed relating mass transfer factor to the Reynolds Number of fluid flowing in a packed bed. The relationships were successfully tested on existing mass transfer data available for one-phase flow through a stationary packed bed. In this thesis application of the mass transfer factor is extended to two-phase flow in packed beds, after suitable modification.

A correlation is presented by Ergun which expresses the mass transfer factor,

$$\left(D_p/H\right)\left(\epsilon/1-\epsilon\right)\left(\mu/D_f\rho\right) \, \ln\left[\left(Y^*-Y_1\right)/\left(Y^*-Y_2\right)\right]$$

as a function of the actual Reynolds Number of the fluid flowing through the bed, $Re/(1-\epsilon)$.

Modification of Mass Transfer Factors and Reynolds Number

The definition of NTU is expressed as,

$$NTU = \int_{Y_1}^{Y_2} dY/\left(Y^*-Y\right)$$

For systems where the solute concentrations are not large

$$NTU = \ln\left[\left(Y^*-Y_1\right)/\left(Y^*-Y_2\right)\right]$$

From previous development in this thesis (Appendix A)

$$HTU_L = L/k_a A = H/NTU_L$$

For a packed column with one phase flow

$$HTU = L/k_a A = H/NTU$$

$$V = L/A$$

The effective diameter of particles can be established as follows:

Area of sphere/volume of sphere $= 6/D_p = a_p/(1-\epsilon)$, and $D_p = 6/a_p(1-\epsilon)$.

Substituting the above into the mass transfer factor proposed by

Ergun, one obtains

$$\left[6\,\epsilon(\text{Sc})k \right]\Big/ V$$

The actual velocity of the fluid in the packed section is V/ϵ and the transfer factor is written as $\left[6(\text{Sc})k \right]/V_{\text{actual}}$.

The Reynolds number to be used in the Ergun correlation for mass transfer in a packed bed is given as $\left[D_p V\rho \right] / \left[\mu(1-\epsilon) \right]$. Substituting $D_p = 6/a_p\,(1-\epsilon)$, a general form for the Reynolds number is obtained in terms of area of packing per unit of tower volume, i.e.

$$(6V\rho)\Big/(a_p\mu).$$

Application of the Ergun Correlation to Liquid-Liquid Mass Transfer

Ergun[20] developed a correlation for mass transfer in packed beds in which he relates the mass transfer factor, $\left[6\,\epsilon(\text{Sc})k \right]/V$, to a Reynolds number, $(6V\rho)/(a_p\mu)$. In order to make use of this correlation for extraction it is necessary to view the liquid-liquid contactor in terms of a packed bed with one-phase fluid flow. For a packed bed with one-phase liquid flow, the film through which the solute is transferred is stationary. The velocity of the fluid relative to the packing and the velocity of the fluid relative to the film are therefore the same. This velocity is given by V/ϵ.

In a packed column for liquid-liquid extraction, there are two moving fluid phases and the stationary packing. Each phase has a velocity relative to the packing and a velocity relative to the interface between the disperse phase and continuous phase. If the velocity relative

to the packing and the velocity relative to the interface are the same, we have a situation analogous to a packed bed in which one fluid moves through the bed at this relative velocity. In order to apply the Ergun correlation to the flow of one of the phases in a packed liquid-liquid extractor, it is necessary to assume that the velocity of the fluid relative to the packing and the velocity relative to interface are equal. The question arises whether the relative velocity of a phase should be equal to the actual phase velocity relative to packing or to the actual phase velocity relative to the liquid-liquid interface. We can reasonably expect that the velocity relative to the interface would have the most effect in determining the resistance of the liquid film through which mass transfer occurs. There is no mass transfer to or from the packing. Thus in a relatively stationary continuous phase the velocity relative to the packing is near zero but the mass transfer coefficient in this case could be greatly increased by the rapid motion of dispersed phase droplets through it.

The velocity of the phases relative to the interface is taken as the important velocity for mass transfer in a packed column. The sum of the velocities of the two phases relative to the interface must equal the velocity of either phase relative to the other. If the viscosities of the two fluids are equal, then the velocity of both phases relative to the interface are approximately equal. In the case where one fluid is very viscous, the velocity of the interface relative to the viscous phase will approach zero.

The mass transfer factor of either the disperse phase or the continuous phase can be expressed as $6(Sc)k/u_r$. If the velocities of the two phases relative to the interface are different, the value of u_r will depend upon which phase is being considered. The velocity of each phase relative to the interface, when their viscosities are equal, is

$$u_r = u_{rd} = u_{rc} = 1/2\left[V_d/\epsilon x + V_c/\epsilon(1-x)\right].$$

When the continuous phase preferentially wets the packing, its flow through the extractor is impeded and made more turbulent because it has to flow around particles of packing and also droplets of the disperse phase. In extending the Ergun correlation to this type of flow, the surface area of droplets must be taken into account as well as the surface of the packing. This may be done by adding the two surface areas together, so the Reynolds number expression for the continuous phase becomes:

$$Re_c = \left[6\, u_{rc}\, \rho_c\, \epsilon\, (1-x)\right]\bigg/\left[(a_p + a_d)\mu_c\right]$$

For disperse phase flow with continuous phase wetting the packing, the effective area is the area of the droplets only. The disperse phase does not come into direct contact with the packing. The Reynolds number for the dispersed phase is then expressed as

$$Re_d = \left[6\, u_{rd}\, \rho_d\, \epsilon\, x\right]\bigg/\left[a_d\, \mu_d\right]$$

DESCRIPTION OF APPARATUS

Main Column. The main column was constructed as shown in Figure 1.

It consisted of a Pyrex glass pipe two inches in internal diameter by

six feet in length, fitted with expanded end sections four inches in

internal diameter and thirteen inches in over-all length. The expanded

end sections and the main column were standard stock items supplied by

the Corning Glass Company. The end covers were constructed of 1/4" stain-

less steel, and had stainless steel fittings to accommodate inlet lines

and pipes, exit lines, the pulse line, manometer taps, and the vent line.

The end sections and covers were fitted with Teflon gaskets and connected

with Corning aluminum flanges and inserts. The packing was supported on

a three-inch diameter grid constructed from lengths of eighteen gauge

stainless steel one-half inch wide, arranged on edge, and spaced one-

fourth inch apart. The grids were provided with four legs (8-1/2 inches

x 3/4-inch 18-gauge stainless steel) spot-welded to the edge of the

circular grid support. The grid strips were also provided with saw-tooth

edges to act as drip points to reduce the tendency for liquid to hang on

the grid. The bottom packing support rested on the end cover and the

top packing support was held in place between the packing and the top

end cover. The upper support held the packing stationary when the column

was pulsed. The end sections provided sufficient reduction of exit stream

velocities to permit complete separation of the two phases and to prevent

the entering stream from being carried out with the exit fluid. The

packed section of the column included part of the expanded chamber. The

FIG. 1

DETAILS OF MAIN COLUMN
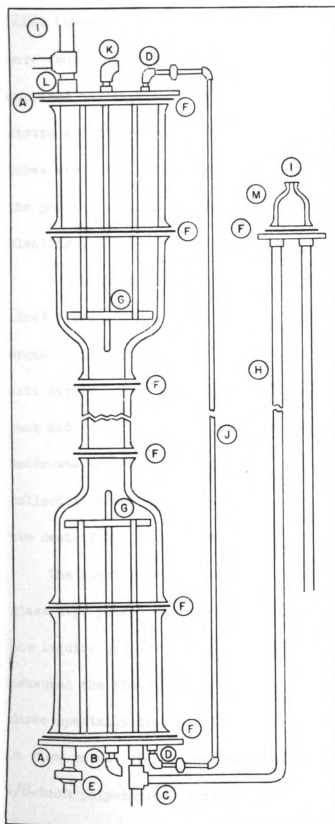
A. END FLANGES WITH COUPLINGS MACHINED INTO 1/4" STEEL PLATE

B. WATER INLET 1/4" FITTINGS

C. CCl_4 OUTLET AND DRAIN 3/4" FITTINGS

D. MANOMETER TAPS 1/4" FITTINGS

E. PULSE GENERATOR CONNECTOR 3/4" FITTINGS

F. TEFLON GASKET (FLANGE NOT SHOWN)

G. PACKING SUPPORTS

H. ADJUSTABLE OVERFLOW LEG

I. ATMOSPHERIC VENTS

J. MANOMETER TUBE

K. CCl_4 INLET 1/4" FITTINGS

L. WATER OUTLET 3/4" FITTINGS

M. OVERFLOW LEG PRESSURE BALANCE ASSEMBLY 1"x3" GLASS REDUCER AND 3/16" PLATE WITH 3/4" COUPLINGS

NOTE: ALL FITTINGS ARE 316 STAINLESS STEEL

inlet streams were introduced directly into the packing by passing the

inlet tubes between two strips of the packing support. The inlet tubes

were constructed from stainless steel tubing 3/16" in internal diameter,

machined and flattened at one end to permit the tube to pass between the

strips and to protrude one inch. Details of the packing support and inlet

tubes are shown in Figure 2. This arrangement provided one-way flow past

the packing support at reduced superficial velocities as recommended by

Blanding and Elgin[7].

A schematic diagram of the equipment is shown in Figure 3. Two glass-

lined storage tanks of 250-gallon capacity each were used to contain the

organic liquid; one contained the feed stream and the other collected the

exit stream. Liquid in the collecting tank was pumped back to the feed

tank and recirculated to the column before the start of each run. The

water was contained in a stainless steel tank, and no attempt was made to

collect it after passage through the column. The liquids were pumped by

two centrifugal pumps, Eastern Industries, Model D-11.

The liquids passed through filters constructed of three-inch diameter

glass pipe packed with cheesecloth as filtering media. From the filters

the liquids passed through 1/8-inch needle valves to rotameters which

measured the flows entering the column. The rotameters were fitted with

three specially designed floats to permit measurement of small variations

in flow rates. Lines from the storage tanks to the rotameters were

5/8-inch polyethylene tubing. Inlet lines to the column from the rota-

meters were 1/4-inch polyethylene tubing. Exit lines from the column

FIG.2

44

DETAILS OF PACKING SUPPORT GRIDS

3" DIA

1/4"
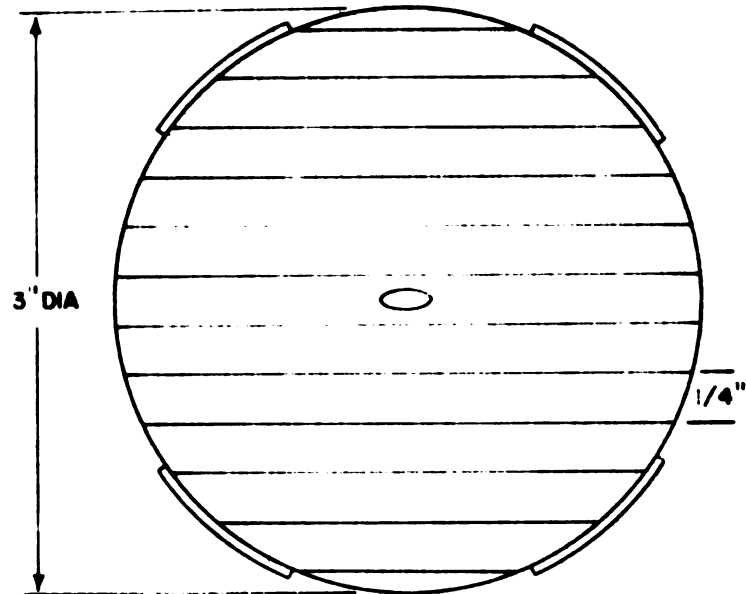
CONSTRUCTION MATERIAL:
18 GAGE STAINLESS STEEL

12 SERRATED STRIPS

1/2"

8 1/2"

3/4"
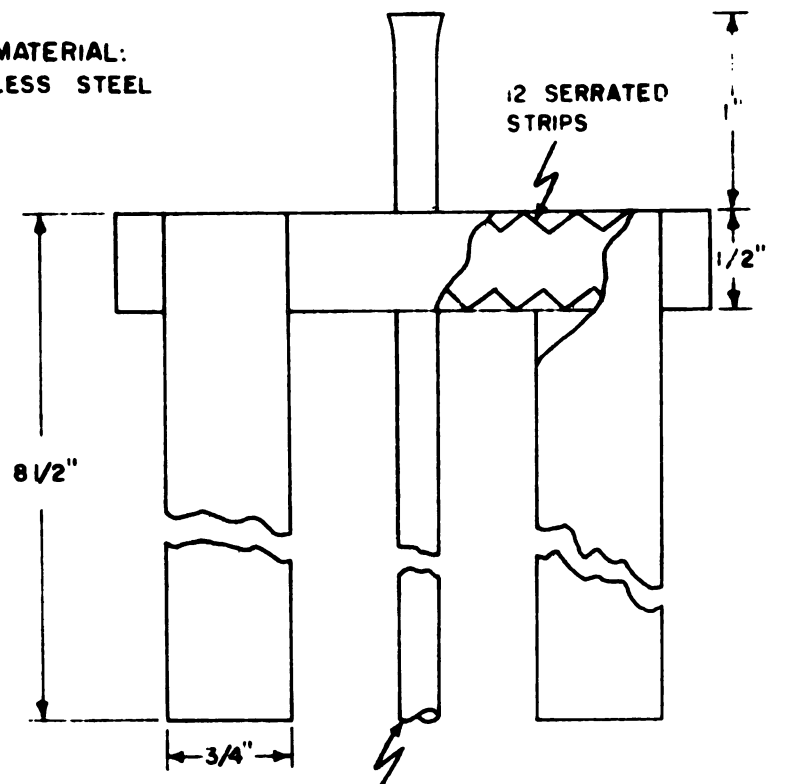
3/4" I.D. STAINLESS TUBE 9 1/2"
LONG MEASURED FROM INSIDE
SURFACE OF END FLANGE, TOP
SECTION FLATTENED

FIG. 3   45

ARRANGEMENT OF EQUIPMENT
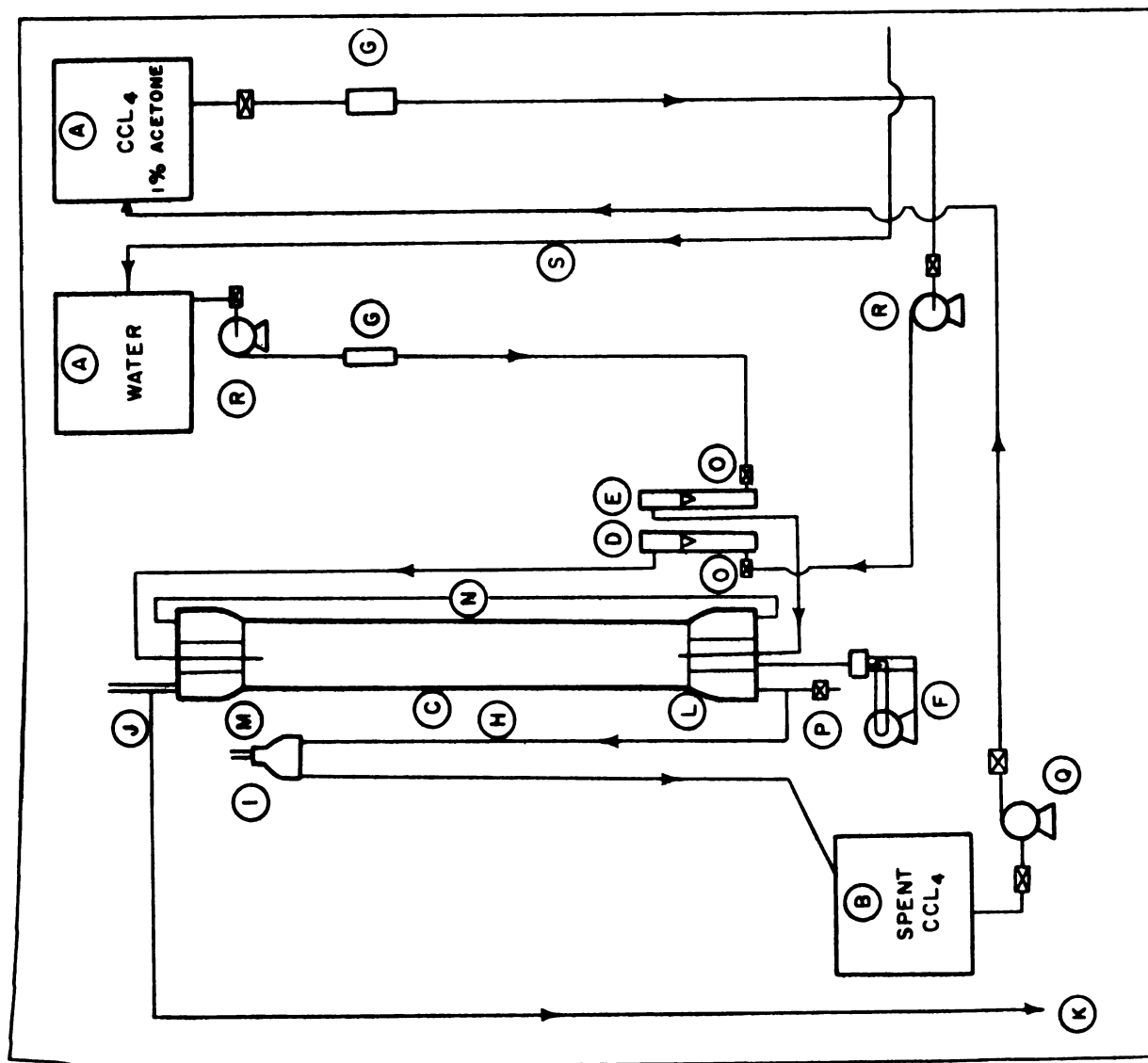
A. FEED TANKS

B. CCL$_4$ COLLECTION TANK

C. PACKED COLUMN

D. ROTAMETER--CCL$_4$ FLOW RATE

E. ROTAMETER--WATER FLOW RATE

F. PULSE GENERATOR

G. FILTERS

H. FLEXIBLE OVERFLOW LEG

I. ATMOSPHERIC VENT

J. WATER OUTLET AND VENT

K. TO WATER DRAIN

L. PACKING SUPPORT GRIDS

M. PACKING SUPPORT GRIDS

N. MANOMETER TUBE

O. FLOW RATE CONTROL VALVES

P. COLUMN DRAIN VALVES

Q. CCL$_4$ RETURN PUMP

R. FEED PUMPS

S. STEAM LINES

were 7/8-inch polyethylene tubing. This size was more than adequate to eliminate any pressure drop in the column exits. Both exit lines were fitted with vents to prevent siphoning. In addition, the exit line from the bottom of the column was constructed to permit variation of its height and control of the interphase of the immiscible liquids in the column.

Pulse Generator. For pulse operation a pulse generator was constructed and connected to the proper fitting on the bottom end cover. A union was placed in the connecting line to facilitate its removal for maintenance. The pulse generator consisted of a reciprocating piston connected to an adjustable eccentric cam. The assembly was driven by a 1/4-HP motor with a speed of 1750 rpm. The eccentric cam was connected to a speed reducer with 10.4 reduction ratio. The motor and speed reducer were each fitted with a three-speed pulley and connected with a V-belt. This arrangement permitted the speed of the eccentric to be varied between 65 and 215 rpm.

The cylinder sleeve and piston surface of the pulse generator were fabricated of stainless steel. A standard leather seal for a reciprocating water pump was fitted on the piston. A drain was provided in the cylinder below the piston to remove fluid that leaked past the seal.

The pulse was transmitted to the column by means of the fluid which filled the cylinder and bottom end chamber of the column. The form of the pulse wave supplied by the piston was sinusoidal, since the cam moved through the 360 degree cycle in a uniform manner. The amplitude of the pulse to the main column could be varied from zero to two inches.

However, during operations the amplitude did not exceed one inch. If a higher amplitude were used, the liquid in the column would be forced through the top vent.

Packing Material. In this investigation two sizes and one type of packing material were used; namely 8-mm and 12-mm Raschig rings.

The superfivial area, square feet of surface per cubic foot of packing material, and the percent voids were determined for each packing used. These determinations were made after the column packing had settled completely and no movement was visible when the system was pulsed at higher frequency and amplitude than used in the experimental runs.

The procedure for calculating the fraction voids required the measurement of the weight of packing and its corresponding column volume, the density of the packing material & the displacement and weight per ring. The values calculated are given in Appendix D.

Liquid-Liquid System. The system selected for the experimental runs was carbon tetrachloride (Dow technical grade) and water. The solute distributed between the two phases was acetone (Merck, C. P. grade). This system was selected because it possessed certain desirable properties; namely large density difference between the immiscible phases, high interfacial tension, and an essentially constant distribution coefficient at low concentrations of acetone. The large density difference is essential to rapid separation of the phases; high interfacial tension caused rapid coalescence of the settled drops[56]; and the constant distribution coefficient permitted the number of transfer stages to be determined analytically[32][53]. Derivation of the equation for calculating

NTP as a function of end concentration is given in Appendix A.

The physical properties of carbon tetrachloride and water used in this thesis are given in Appendix D. The equilibrium distribution of acetone between water and CCl_4 is also given in Appendix D.

OPERATING PROCEDURE

<u>Column Operation</u>. Prior to making a series of runs the column was

filled with either 8-mm or 12-mm Raschig rings. The rings were dumped

into the column and allowed to settle through water. After the column

had been filled, the top packing grid and end chamber were fixed in place.

Settling of the packing was carried out by pulsing the system initially

at 65 cycles per minute and one inch amplitude in the main section of the

column. Additional packing material was added until the volume between

the two grid elements was completely filled and remained filled after

2 hours of continuous pulsing. The packing was further tested for

settling by replacing the water in the column with carbon tetrachloride

to take advantage of the additional buoyant force of the latter and

pulsing with a larger amplitude. When the packing had been completely

settled it was found that even after several months of operation

negligible change occurred in the packing density.

The water storage tank was filled with distilled water, distilled

insofar as the water was obtained by condensing steam available in the

laboratory. The use of distilled water was necessary in order to re-

produce the flooding data, as will be seen in the results. Some of the

experimental runs for flooding were carried out with Michigan State

University tap water. Its use had to be discontinued when inconsistencies

in flooding rate were noted for the 8-mm packing. Before any runs were

begun, the water was permitted to reach room temperature.

The carbon tetrachloride was first saturated with water by contacting with water in the packed column. Acetone was added to the carbon tetrachloride prior to each extraction run to give an initial concentration of approximately one percent. The carbon tetrachloride was collected in the storage tank after each pass through the column and reused after the acetone was replenished.

Each flooding run was started by pumping carbon tetrachloride into the top and water into the bottom of the column in a one-to-one ratio until the column was filled. The overflow leg was adjusted to a position which approximated the final position. The carbon tetrachloride rate was then increased in small increments to the desired rate indicated by its rotameter and maintained for the duration of the run. With the carbon tetrachloride rate fixed, the water rate was increased in small increments until an interface appeared at the grid support in one of the column end chambers. The overflow leg was lowered or raised depending on whether the interface appeared at the top or bottom grid support. This caused the interface to move into the packing. The procedure of increasing the water rate and adjusting the overflow leg was repeated until two interfaces appeared; one at the top grid support and one at the bottom grid support. Flow rates obtained by the above procedure were greater than those which would be obtained if the operation were limited by either the top or bottom interface. The flow rate required to maintain interfaces in the top and bottom end chambers was reported as the flooding rate. Actual flow rates were determined by collecting a

measured volume of each exit stream in a measured interval of time after equilibrium had been established.

It cannot be emphasized enough that equilibrium must be established in order to obtain reliable flooding data. If the interfaces do not remain stationary the effective flow rate of one of the phases is increased and the balance of the system is disrupted. This often causes temporary or premature flooding and requires repeating of the run. Dell and Pratt[15] attempted to circumvent the problem of moving interfaces by correcting the flow rates. In order to do this the inlet and outlet flow rates in the column were tabulated from rotameters along with readings of the movement of the interfaces in a five minute interval. In this report the flow rates were reported only for those runs in which the interfaces remained stationary or nearly so and the material balance of acetone agreed within five percent.

The flow rates indicated by the rotameters, the height of the adjustable overflow leg, and the interface level in the manometer were recorded. Samples of each stream were collected and analyzed; namely the inlet and exit carbon tetrachloride streams, and the exit water stream. It was not necessary to subject the inlet water stream to analysis because it did not contain acetone. Of course, for the flooding runs in which no acetone was being transferred, the stream samples were not taken.

The extraction runs at reduced flow rates, i.e. below flooding, were made with the overflow leg fixed by the flooding run and the ratio

of carbon tetrachloride to water being considered. The column was operated at reduced flow rates with carbon tetrachloride as the continuous phase and water as the dispersed phase, and also vice versa.

For the pulsed runs the procedure for collecting data was the same as outlined above except that it was necessary to adjust the pulse amplitude and frequency and place it in operation prior to adjusting the flow rates and overflow leg. The frequency was selected by fastening the drive belt to the appropriate pulley of the speed reducer and the motor. The amplitude was adjusted through the variable eccentric cam and measured in the top and bottom end chambers. The amplitudes referred to in the data indicate the total up-and-down motion of liquids in the main section of empty column.

Analytical Procedure. The method used for quantitative determination of acetone in water and carbon tetrachloride phases consisted of titrating the hydrochloric acid released by reaction of acetone and hydroxylamine hydrochloride. The reaction

$$(CH_3)_2CO + HONH_2 \cdot HC\ell \longrightarrow CH_3CNOH + HC\ell + H_2O$$
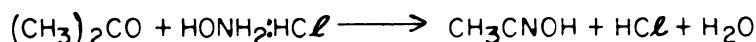
was first proposed by Hoepner[27] and investigated at later dates by other experimenters[5][9][41]. The success of the quantitative determinations is dependent on the use of an indicator and the ability to detect a definite end point. For the determinations made in this thesis, the end point was established by using a pH meter.

The procedure required the makeup of hydroxylamine hydrochloride solution by adding 14 grams of the chemical to 6 liters of distilled

water. For each determination, 600 ml of the solution was added to a
1000-ml beaker and stirred. The pH of the solution was noted and 20 ml
of a water sample or carbon tetrachloride sample was added with the aid
of a pipette. The resulting reaction released hydrochloric acid in the
solution. It was titrated with standard sodium hydroxide (0.2N) to
bring the solution to the original pH. Since the release of hydrochloric
acid was not instantaneous and the reaction proceeded only in the water
phase, the solution was stirred continuously.

The above procedure was checked against standard solutions of
acetone throughout the concentration ranges that occurred in the experi-
mental data. The results agreed within 0.5 percent.

EXPERIMENTAL RESULTS

The experimental data in this thesis are presented in three main sections. In Section I, data for flooding characteristics in packed columns are presented. In Section II, experimental mass transfer data in packed columns are tabulated. In Section III, the results calculated from hydrodynamic and mass transfer considerations of a dispersed phase flowing through a continuous phase in a packed column are presented.

The experimental data in this thesis are based, for the most part, on the operation of a 2-inch diameter column with a packed length of 81.5 inches. Packings used were 8-mm and 12-mm Raschig rings. Reference will also be made to some data collected by this writer on two columns originally constructed by Callihan[10]. This was done primarily to reproduce certain points reported by Callihan and also to obtain additional points to check anomalies.

SECTION I

FLOODING CHARACTERISTICS IN PACKED COLUMNS

Flood point data were taken under three main types of conditions.

Subsection A lists flooding data without mass transfer. Qualitative

observations, which in some instances are more important than the numer-

ical data, are presented as experimental observations. Subsection B

presents experimental flooding data with mass transfer. Subsection C

presents flooding data for which the direction of solute transfer was

considered. Observations of a qualitative nature are also listed to

demonstrate the phenomenon of coalescence in the presence of mass trans-

fer.

A. <u>Maximum Flow Rates Without Mass Transfer</u>

Numerical Data:

<u>Table I, Series I through V</u> presents flooding data obtained from a

2.127-inch diameter column packed with 8-mm Raschig rings. The packed

section was 30.75 inches long. This column was constructed for an

earlier thesis[10]. The system used for these data was carbon tetra-

chloride and tap water.

The purpose of these runs was twofold. First, it was intended to

observe the flow phenomena in packed columns. Second, it was intended

to investigate previously reported flooding curves[10] obtained in the

same column and packing, which seemed to deviate from the type reported

in the literature. The column constructed by Callihan[8] would not be

considered ideal by many because there were no expanded end sections, and

inlets were not placed directly in the packing. However, while the presence

or absence of expanded end sections and optimum inlets may change the

magnitude of the flow rates it is not likely to alter the shape of the

curves.

Series I of Table I presents the data collected by Callihan[10].

Series II of Table I presents data from the column before and after

acid cleaning. The first three points were collected after a long period

of column operation. At this time column and packing appeared discolored.

The remainder of the points in this series represents runs on the column

cleaned by acid treating.

Series III of Table I represents a second set of runs completed

after the column was acid treated.

Series IV of Table I lists data taken after four days of continuous

operation following Series III. No intermediate acid treatment was

employed.

Series V of Table I lists data obtained after the carbon tetra-

chloride inlet tube was placed directly into the packing. The packing was

not acid treated between Series IV and V. Run 34 of Series V was made

after the column was acid treated.

Table II. In Table II data are presented for pulsed and unpulsed

runs in a 2-inch diameter column with expanded end section and inlets

placed directly in the packing. The data are reported for 8-mm Raschig

rings in a packed section 81.5 inches high.

Series VI of Table II tabulates data obtained in the column operated

without pulsation. The system was carbon tetrachloride and tap water.

The data was obtained after the column was conditioned with tap water.

While the column was being conditioned, several points were checked to

observe progressively increasing flow rates with time. The last run,

47, of Series VI was completed after the column was pulsed for a short

duration of time. Run 47 indicates a reduction in flow rate. It was

also noted that much of the scum and residue between the packing materials

was lossened and washed away with pulsation.

Series VII of Table II lists data collected in the 2-inch column

with expanded end section. The packing material was the same 8-mm

Raschig rings. The liquid system in this series was distilled water and

carbon tetrachloride. Even after a long period of operation the flow rates

were observed to be lower with distilled water than with tap water.

It was observed during the operation that the flood point data for

distilled water and CCl_4 was in fair agreement with the data for tap water

and CCl_4 observed in a clean column, as after acid cleaning. The differences

may be due to different packing characteristics and end section design

between the two columns.

Series VIII of Table II represents the data collected in the 2-inch

diameter column packed with 8-mm Raschig rings under pulsed conditions.

The carbon tetrachloride and distilled water phases were pulsed at a

frequency of 125 cycles per minute and an amplitude of 5 millimeters.

Table III. In Table III data are presented for pulsed and unpulsed

runs on a 2-inch diameter column with expanded end sections and inlets

placed directly in the packing. The data are reported for 12-mm Raschig

rings in a packed section 81.5 inches high.

Series IX of Table III. In this series the system was carbon tetra-

chloride and tap water. It was interesting to observe that the flow rate

did not increase with time even after several days of operation.

Series X of Table III presents flooding data for carbon tetrachloride

and distilled water.

Series XI of Table III lists flooding data for carbon tetrachloride

and distilled water for a pulsation rate of 125 cycles per minute and 5

millimeters amplitude.

Visual Observations:

Series II, III, IV of Table I. It was observed that the maximum

throughput in the column increased progressively with time. The flow

rates decreased sharply when the column was washed with acid. However,

continued operation resulted in increased flow rates again. Also it was

observed that it was more difficult to hold the interfaces at a fixed

position immediately after the column was acid treated than after a

long period of operation.

Series VI and VII of Table II. At flooding the interfaces visible

outside of the packing supports were not easily maintained at constant

level. In many instances, after the interfaces were held in balance for

at least an hour, for no apparent reason they drifted into the packing

slowly or else moved away from each other rapidly. This behavior was

most noticeable for distilled water and carbon tetrachloride. For tap

water and carbon tetrachloride the behavior described was encountered only
when the column was clean, i.e., immediately after acid treating.

Continuous operation with tap water made it easier to hold the ex-
ternal interfaces at constant levels during flood point operation. Also
continuous operation with tap water caused the packing to appear yellow.
A scum-like film oftentimes formed at the interfaces outside of the
packing support. Foaming was also encountered.

Continuous operation with tap water resulted in large quantities of
water passing into the water storage tank. As a result the tank collected
a precipitate from the tap water which dissolved readily in hydrochloric
acid. Carbon dioxide was evolved.

Below maximum throughput a distinct interface was visible in the
packing. Above the interface carbon tetrachloride droplets moved down
through the continuous water phase, below the interface a quiescent region
was present. The water phase did not move up in droplets through the
dense carbon tetrachloride but rather the water moved up as a film and
rivulets. As the flow rates were increased the proportion of active zone
to quiescent zone in the column changed. In several instances, as the
maximum flow rates were approached, the active zone moved through the
packing in the form of slugs. At flooding the active region appeared in
the lower part of the packing. It was not clear that one liquid in the
packing formed the continuous phase and the other the dispersed phase.
The lower part of the column appeared to be predominantly water with
carbon tetrachloride falling through it. The upper section of the packing

appeared to be predominantly carbon tetrachloride with water rising, as a film, when water flows were low and as droplets when the water flow rates were high. This unstable state of balance probably had much to do with the erratic operation at flooding reported above for small packing.

Similar observations and erratic movements of the interfaces at flooding have been reported by other investigators[2] [8] [15]. Ballard and Piret[2] defined a "transition point" as the maximum throughput before sudden changes in the flow mechanism occurred. Above the transition point the column may flood or else the rates may change the flow characteristics to accommodate higher rates by a phase reversal. Ballard and Piret claimed that the "transition point" could be reproduced but above this point erratic column operation resulted.

Series IX and X of Table III. The phenomenon of progressively in-creasing flow rates was not observed with 12-mm packing for the system carbon tetrachloride-water.

The column operated more uniformly and smoothly with 12-mm rings than when 8-mm rings were used.

Series VIII of Table II and Series XI of Table III. The continuous and dispersed phase were uniformly distributed in the packing for pulsed operation at the flood point. The drops were smaller for pulsed operation than for unpulsed operation.

TABLE I

Unpulsed Runs at Flooding

(a) 2.17 inch column packed with 8 mm Raschig rings
(b) packing height 30.75 inches
(c) packing density 51.7 lbs/ft^3
(d) no acetone, tap water

Run	Flow Rates ml/min		V, ft/hr		Flow Ratio CCl_4/H_2O	Sum of sq roots of superficial velocities
	Water	CCl_4	Water	CCl_4		
Series I						
	650	343	55.84	29.46	0.528	12.90
	670	173	57.56	14.82	0.258	11.43
	685	193	58.85	16.58	0.282	11.74
	610	463	52.40	39.78	0.760	13.55
	660	150	56.70	12.89	0.227	11.12
	278	780	23.88	67.01	2.80	13.06
	640	143	54.98	12.29	0.224	10.92
	640	193	54.98	16.58	0.302	11.49
Series II						
1	720.3	407.0	61.88	34.96	0.565	13.78
2	735.0	370.0	63.14	31.78	0.503	13.58
3	425.0	535.3	36.51	45.99	1.259	12.83
4	366.9	361.7	31.52	31.10	0.986	11.19
5	358.0	395.3	30.76	34.00	1.104	11.37
6	354.5	401.8	30.50	34.50	1.133	11.10
7	227.7	1218.8	19.60	104.7	5.352	14.63
8	542.9	205.5	46.60	17.70	0.379	11.04
9	558.8	205.5	48.00	17.70	0.368	11.14
10	625.5	140.4	53.70	12.10	0.224	10.82
11	683.0	145.7	58.70	12.50	0.213	11.20

Note (left margin, vertical): Data from Callihan(10)

TABLE I
(Contd)

| Run | Flow Rates | | | | Flow Ratio | Sum of sq roots of superficial velocities |
| | ml/min | | V, ft/hr | | CCl_4/H_2O | |
	Water	CCl_4	Water	CCl_4		
Series III						
12	380.6	264.5	32.70	22.72	0.695	10.48
13	562.0	149.6	48.28	12.85	0.266	10.53
14	638.2	121.0	54.83	10.40	0.190	10.62
15	618.7	121.0	53.15	10.40	0.196	10.51
16	278.0	450.0	23.88	38.66	1.619	11.10
17	476.0	328.2	40.89	28.20	0.689	11.70
18	181.4	827.5	15.58	71.09	4.562	12.37
Series IV						
19	397.8	1269.	34.17	109.0	3.190	15.15
20	1010.	569.0	86.77	48.88	0.563	16.30
21	1361.	328.2	116.9	28.20	0.241	16.11
22	664.5	836.1	57.09	71.83	1.258	16.03
23	241.5	1591.	20.75	136.7	6.588	16.25
24	1617.	204.0	138.9	17.52	0.126	15.97
25	1500.	183.0	128.9	15.72	0.122	15.31
Series V						
26	999.7	507.9	85.88	43.63	0.508	15.86
27	1728.	257.6	148.4	22.13	0.149	16.90
28	1379.	269.0	118.5	23.11	0.195	15.71
29	1601.	150.0	137.5	12.89	0.094	14.29
30	1011.	485.0	86.86	41.67	0.478	15.77
31	689.4	753.6	59.23	64.74	1.093	15.73
32	321.0	1302.	27.58	111.8	4.056	15.81
33	338.4	1319.	29.07	113.3	3.898	16.03
34	790.2	293.0	67.88	25.17	0.371	13.26

TABLE II

Pulsed and Unpulsed Runs at Flooding

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 8 mm Raschig rings, packing density 49.12 lbs/ft^3
(e) no acetone in feed streams

| Run | | Flow Rates | | | | Flow Ratio | Sum of sq roots |
| | | ml/min | | V, ft/hr | | | of superficial |
		Water	CCl$_4$	Water	CCl$_4$	CCl$_4$/H$_2$O	velocities
Series VI	Tap water						
35 1st Day		487.0	218.0	42.28	21.16	0.448	11.10
36 2nd		527.0	218.0	51.16	21.16	0.414	11.75
37 2nd		360.0	770.0	34.94	74.75	2.139	14.56
38 3rd		582.0	216.0	56.50	20.97	0.371	12.10
39 4th		655.0	218.0	63.10	21.16	0.335	12.54
40 5th		675.0	218.0	65.52	21.16	0.323	12.69
41 5th		360.0	810.0	34.95	78.63	2.250	15.77
42		583.0	315.0	56.60	30.58	0.540	13.05
43		635.0	130.0	61.64	12.62	0.205	11.40
44		648.0	130.0	62.91	12.62	0.201	11.48
45		285.0	935.0	27.67	90.77	3.281	14.87
46		510.0	490.0	49.51	47.57	0.961	13.94
47		575.0	218.0	55.82	21.16	0.379	12.07
Series VII	Distilled water						
48		367.7	397.4	35.6	38.6	1.081	12.18
49		355.0	526.3	34.5	54.7	1.482	13.27
50		344.0	569.1	33.4	55.2	1.654	13.21
51		345.0	363.2	33.5	35.3	1.053	11.73
52		326.5	362.0	31.7	35.2	1.108	11.56
53		325.5	474.5	31.6	46.1	1.458	12.41
54		179.4	839.0	17.4	81.5	4.677	13.20
55		185.0	825.0	18.0	80.1	4.459	12.19
56		320.2	541.0	31.1	52.5	1.690	12.83
57		314.0	548.0	30.5	53.2	1.745	12.81
58		387.0	299.0	37.6	29.0	0.773	11.52
59		548.0	200.0	53.2	19.4	0.365	11.69
60		525.0	113.0	51.0	11.0	0.215	10.46

TABLE II
(Contd.)

| Run | Flow Rates | | | | Flow Ratio | Sum of sq roots of superficial velocities |
| | ml/min | | V, ft/hr | | CCl_4/H_2O | |
	Water	CCl_4	Water	CCl_4		
Series VIII	125 cycles/min, 5 mm amplitude, distilled water					
61	553.0	253.6	53.69	24.62	0.458	
62	862.0	159.0	83.68	15.44	0.184	
63	1111.0	103.0	107.8	10.30	0.093	
64	308.8	344.5	29.98	33.44	1.116	
65	93.9	415.4	9.12	40.33	4.424	

TABLE III

Pulsed and Unpulsed Runs at Flooding

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 12 mm Raschig rings, packing density 25.91 lbs/ft^3
(e) no acetone in feed streams

| | Flow Rates | | | | Flow Ratio | Sum of sq roots of superficial velocities |
| | ml/min | | V, ft/hr | | | |
Run	Water	CCl$_4$	Water	CCl$_4$	CCl$_4$/H$_2$O	
Series IX	Tap water					
66 1st day	1530.	760.0	148.5	73.78	.497	20.77
67 2nd	1530.	755.0	148.5	73.30	.493	20.74
68 3rd	1530.	755.0	148.5	73.30	.493	20.74
69 3rd	2760.	230.0	267.9	22.33	.083	21.09
70 3rd	2200.	445.0	213.6	43.20	.202	21.17
71 4th	1550.	760.0	150.5	73.78	.490	21.86
Series X	Distilled water					
72	2244.	144.0	217.8	13.98	0.064	18.49
73	2180.	242.0	211.6	23.49	0.111	19.41
74	1880.	520.0	182.5	50.48	0.276	20.62
75	1620.	750.0	157.3	72.81	0.463	21.07
76	1280.	970.0	124.3	94.17	0.758	20.85
77	1140.	1436.0	110.7	139.40	1.260	22.32
78	1040.	1405.0	101.0	136.40	1.351	21.72
79	750.	1775.0	72.81	172.30	2.367	21.65
Series XI	125 cycles/min, 5 mm amplitude, distilled water					
80	880.	1417.0	85.43	137.60	1.610	
81	1880.	730.0	182.50	70.86	0.388	
82	1504.	937.5	146.00	91.01	0.623	
83	2400.	523.0	233.00	50.77	0.218	

B. **Maximum Flow Rates With Mass Transfer**

Numerical Data:

Table IV. In Table IV flood point data with mass transfer are listed.
The solute, acetone, was present in the entering carbon tetrachloride phase
in concentrations of approximately one percent. The column in use was 2 inches
in diameter with expanded ends and with inlets located in the packing. The
81.5-inch high section was packed with 8-mm Raschig rings.

Series XII of Table IV lists data collected at flooding with no pulse
at various CCl_4/H_2O flow ratios.

Series XIII of Table IV presents data collected at flooding at a
pulsation rate of 125 cycles per minute and 5 mm amplitude.

Table V. In Table V flood point data with mass transfer are presented.
The solute was acetone entering with the carbon tetrachloride in concen-
tration of approximately one percent. The 81.5 inches of packed section
in the 2-inch diameter column consisted of 12-mm Raschig rings.

Series XIV of Table V lists data collected at flooding with no pulse
at various CCl_4/H_2O flow ratios.

Series XV of Table V presents data collected at flooding at a pulsa-
tion rate of 125 cycles per minute and 5-mm amplitude.

Visual Observations:

Series XII through XV. At high carbon tetrachloride to water flow
ratios large drops were observed in most of the packing and small drops
in the very bottom portion of the packing. The small drops appeared to
be of the same size as those observed in earlier runs with no mass transfer.

At low ratios of carbon tetrachloride to water the small drops were present

in a proportionately larger zone of the packed section.

TABLE IV

Pulsed and Unpulsed Runs at Flooding with Mass Transfer

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 8 mm Raschig rings, packing density 49.12 lbs/ft^3
(e) 1 percent acetone in CCl_4 feed

Run	Flow Rates ml/min Water	CCl$_4$	V, ft/hr Water	CCl$_4$	Flow Ratio CCl$_4$/H$_2$O	Sum of sq roots of superficial velocities
Series XII	no pulse					
84	410.8	847.6	39.88	82.28	2.063	15.39
85	606.2	538.6	58.85	52.29	0.838	14.90
86	987.5	271.0	95.87	26.31	0.274	14.92
87	927.5	249.2	90.04	24.19	0.269	14.41
88	889.2	317.1	86.32	30.78	0.357	14.83
89	904.4	258.3	87.80	25.08	0.289	14.38
90	980.0	110.0	95.14	10.68	0.112	13.02
91	888.0	158.5	86.21	15.39	0.179	13.20
92	877.0	176.0	85.14	17.09	0.201	13.35
93	868.0	213.0	84.26	20.68	0.245	13.72
94	965.0	110.0	93.68	10.68	0.114	12.95
95	900.0	225.0	87.37	21.84	0.250	14.01
96	890.0	256.4	86.40	24.89	0.289	14.28
97	870.0	304.7	84.46	29.58	0.351	14.60
Series XIII	125 cycles/min, 5 mm amplitude					
98	302.6	1102.0	29.38	107.00	3.64	15.76
99	480.0	833.3	46.60	80.90	1.74	15.81
100	520.6	756.0	50.54	73.39	1.45	15.68
101	566.2	635.4	54.97	61.63	1.12	15.27
102	558.7	523.5	54.24	50.82	0.937	14.49
103	510.7	417.8	49.58	40.56	0.818	13.40
104	397.2	363.7	38.56	35.31	0.916	12.16
105	542.8	253.3	52.70	24.59	0.466	12.22
106	1053.0	110.0	102.2	10.67	0.104	13.37
107	439.3	304.6	42.70	29.57	0.693	11.97

TABLE V

Pulsed and Unpulsed Runs at Flooding with Mass Transfer

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 12 mm Raschig rings, packing density 25.91 lbs/ft^3
(e) 1 percent acetone in CCl$_4$ feed

| Run | Flow Rates | | | | Flow Ratio | Sum of sq roots |
| | ml/min | | V, ft/hr | | CCl$_4$/H$_2$O | of superficial |
	Water	CCl$_4$	Water	CCl$_4$		velocities
Series VIV	no pulse					
108	1452.	1022.	141.0	99.2	0.704	21.83
109	2160.	440.	209.7	42.7	0.204	21.01
110	1630.	650.	158.2	63.1	0.399	20.51
111	1620.	760.	157.3	73.8	0.469	21.12
112	1226.	1230.	119.0	119.4	1.003	21.83
113	2040.	538.	198.0	52.2	0.264	21.29
114	1590.	717.	154.4	69.6	0.451	20.75
115	1000.	1390.	97.1	134.9	1.390	21.47
132	810.	1690.	78.63	164.1	2.080	21.62
Series XV	125 cycles/min, 5 mm amplitude					
116	1340.	1450.0	130.1	140.8	1.082	
117	1640.	1085.0	159.2	105.3	0.661	
118	2145.	710.0	208.2	68.9	0.331	
119	1990.	810.0	193.0	78.6	0.407	
120	1800.	910.0	174.7	88.3	0.506	
121	2810.	445.0	272.8	43.2	0.158	
122	1570.	1290.0	152.3	125.2	0.822	

C. Direction of Solute Transfer Considerations

The observations and data reported in Subsection B, above, indicated

that mass transfer under some conditions contributes to the attainment of

high flow rates and large drop formation. It was not clear whether the

phenomena encountered were peculiar to the H_2O-CCl_4-acetone system. It

was not clear that the maximum flow rates are the same with the direction

of transfer from the water phase to the carbon tetrachloride phase and

vice versa. Further the question was raised whether the flood point is

the same if it were approached with either phase the disperse phase or

the continuous phase.

It is difficult to observe these variations in the column, because

at flooding the disperse phase and continuous phase appear to be uniformly

distributed throughout the packed section. In order to clarify some of

these points bench scale experiments were performed to supplement the

column operating data.

The experiments performed in this section include: bench scale

investigations of interfacial tension and coalescence, with additional

data from column operation.

Effect of mass transfer on interfacial tension. A set of experiments

was carried out to determine if mass transfer markedly reduced the inter-

facial tension between the carbon tetrachloride and water layer. It did not.

Interfacial tension observations were made with a stalagmometer. The

observations were carried out for carbon tetrachloride and water: in the

absence of acetone, with acetone transfer from carbon tetrachloride to water,

with acetone transfer from water to carbon tetrachloride, with acetone distributed between the two phases in equilibrium concentrations. The largest interfacial tension occurred in the absence of acetone, the smallest when acetone was distributed between the phases in equilibrium concentrations. Intermediate interfacial tensions were observed when acetone transfer occurred from either the water phase or carbon tetrachloride phase. The observed differences were practically negligible for the small acetone concentrations investigated.

Effect of mass transfer on coalescence. Table VI presents the observations of qualitative bench scale experiments on the effect of mass transfer on coalescence rate. The table includes observations made in the absence of solute, with solute transfer from either phase, and with solute distributed between the phases in equilibrium concentrations.

For these experiments the disperse phase was injected with a hypodermic syringe into the continuous phase.

Effect of direction of transfer on column operation. Table VII, Series XVI presents flooding data for carbon tetrachloride and water, in the absence of acetone, with acetone distributed between the two phase in equilibrium concentrations, with acetone transfer from carbon tetrachloride to water, and with acetone transfer from water to carbon tetrachloride.

The same column and packing characteristics reported in Table V were used for these runs.

TABLE VI

Coalescence Observations

Disperse Phase	Continuous Phase	Observations
CCl$_4$	H$_2$O	Disperse phase in fine state of subdivision
H$_2$O	CCl$_4$	Disperse phase in fine state of subdivision
(CCl$_4$ + Acetone)	(H$_2$O + Acetone)	Disperse phase in fine state of subdivision
(H$_2$O + Acetone)	(CCl$_4$ + Acetone)	Disperse phase in fine state of subdivision
(CCl$_4$ + Acetone)	H$_2$O	Large drop formation and rapid coalescence
H$_2$O	(CCl$_4$ + Acetone)	Disperse phase in fine state of subdivision
(H$_2$O + Acetone)	CCl$_4$	Large drop formation and rapid coalescence
CCl$_4$	(H$_2$O + Acetone)	Disperse phase in fine state of subdivision
(CCl$_4$ + HAc)	(H$_2$O + HAc)	Disperse phase in fine state of subdivision
(H$_2$O + HAc)	(CCl$_4$ + HAc)	Disperse phase in fine state of subdivision
(CCl$_4$ + HAc)	H$_2$O	Large drop formation and rapid coalescence
H$_2$O	(CCl$_4$ + HAc)	Disperse phase in fine state of subdivision
(H$_2$O + HAc)	CCl$_4$	Large drop formation and rapid coalescence
CCl$_4$	(H$_2$O + HAc)	Disperse phase in fine state of subdivision

TABLE VI
(Contd)

Disperse Phase	Continuous Phase	Observations
Toluene	H_2O	Disperse phase in fine state of subdivision
H_2O	Toluene	Disperse phase in fine state of subdivision
(Toluene + Acetone)	(H_2O + Acetone)	Disperse phase in fine state of subdivision
(H_2O + Acetone)	(Toluene + Acetone)	Disperse phase in fine state of subdivision
(Toluene + Acetone)	H_2O	Large drop formation and rapid coalescence
H_2O	(Toluene + Acetone)	Disperse phase in fine state of subdivision
(H_2O + Acetone)	Toluene	Large drop formation and rapid coalescence
Toluene	(H_2O + Acetone)	Disperse phase in fine state of subdivision
Toluene	H_2O	Disperse phase in fine state of subdivision
H_2O	Toluene	Disperse phase in fine state of subdivision
(Toluene + HAc)	H_2O	Large drop formation and rapid coalescence
H_2O	(Toluene + HAc)	Disperse phase in fine state of subdivision
(H_2O + HAc)	Toluene	Large drop formation and rapid coalescence
Toluene	(H_2O + HAc)	Disperse phase in fine state of subdivision

TABLE VII

Unpulsed Runs Which Indicate the Effect of Direction
of Mass Transfer on Flooding

| Run | Flow Rates ml/min | | V, ft/hr | | Sum of sq roots of superficial velocities | Comments |
	Water	CCl₄	Water	CCl₄		
Series XVI						
123	1445	800	140.3	77.66	20.65	no solute, small drops
124	1425	800	138.3	77.66	20.57	equilibrium distribution of acetone between phases, small drops
125	1800	800	174.7	77.66	22.22	1 percent acetone in CCl₄ feed, large drops, rapid coalescence
126	1075	800	104.4	77.66	19.03	acetone transfer from H₂O phase to CCl₄ phase, small drops

SECTION II

MASS TRANSFER IN PACKED COLUMNS

In this research considerable data have been collected to investigate

the efficiency of mass transfer in packed columns. Data are reported in

this section based on experiments performed on two sizes of Raschig rings

in a 2-inch diameter column. The packing height in the column measured

81.5 inches, except where noted. The concentration of acetone in the

feed carbon tetrachloride was maintained at approximately one percent.

In this range the distribution coefficient can be considered constant.

No acetone entered with the water phase.

The data represent a wide variety of operating conditions. For

example, in Subsection A, efficiency of mass transfer at different flow

ratios for unpulsed conditions is reported for two sizes of packing. The

efficiency of mass transfer for two sizes of packing, and at flow rates

below flooding is reported. Mass transfer for different positions of the

interface at reduced flow rates have been investigated.

All of the above variations have been repeated for a variety of

pulsation rates. These data are reported in Subsection B.

Subsection C presents the contribution of the inlet tubes to mass

transfer.

A. <u>Mass Transfer in Unpulsed Packed Columns</u>

The column and packing characteristics for Tables VIII and IX are the same as for Tables IV and V.

Numerical Data:

<u>Table VIII, Series XVII and XVIII</u>. HETS data at flooding and for various ratios of CCl_4/H_2O are tabulated for 8-mm and 12-mm Raschig rings.

Table IX. In Table IX HETS data are tabulated for 8-mm and 12-mm Raschig rings at nearly constant CCl_4/H_2O flow ratios and reduced flow rates.

<u>Series XIX of Table IX</u>. HETS data are tabulated for 8-mm Raschig rings. The intermediate interface position was determined by the position of the overflow leg at flooding when the flow ratio of CCl_4/H_2O was approximately 2. The position of the overflow leg was not changed for the reduced flow rate runs. This procedure was used throughout this research where data are presented for runs below flooding with an intermediate interface.

<u>Series XX of Table IX</u>. HETS data are tabulated for 12-mm Raschig rings. The interface was located at an intermediate point.

<u>Series XXI of Table IX</u>. HETS data are tabulated for 12-mm Raschig rings with carbon tetrachloride as the continuous phase.

Visual Observations:

<u>Table IX, Series XIX, XX</u>. At low flow rates with an intermediate interface, carbon tetrachloride is the continuous phase in the lower portion of the column. Water is the continuous phase in the higher portion of the column. Carbon tetrachloride falls through the continuous water

phase as discrete drops. Water may or may not rise through the carbon tetrachloride phase as drops. When the water rate is very low it rises as a film along the packing. At higher water rates a portion of the water flow rises as a film and a portion rises as drops.

TABLE VIII

HETS for Variable Ratios of CCl₄/H₂O and Maximum Flow Rates

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 8 mm Raschig rings, packing density 49.12 lbs/ft^3
(e) 12 mm Raschig rings, packing density 25.91 lbs/ft^3
(f) 1 percent acetone in feed CCl₄, no pulse

| | Flow Rates | | | | Flow Ratio | Millimols solute/liter | | | NTP | HETS |
| | ml/min | | V, ft/hr | | | Water | CCl₄ | | | |
Run	Water	CCl₄	Water	CCl₄	CCl₄/H₂O	Exit	Feed	Exit		ft
Series XVII										
8 mm Raschig rings										
84	410.8	847.6	39.88	82.28	2.063	389.9	309.4	119.4	1.555	4.54
85	606.2	538.6	58.85	52.29	0.888	217.5	294.2	58.4	1.42	4.78
86	987.5	271.0	95.87	26.31	0.274	76.71	294.2	13.66	1.43	4.75
87	927.5	249.2	90.04	24.19	0.269	80.51	305.3	9.73	1.61	4.22
88	889.2	317.1	86.32	30.78	0.357	90.70	281.8	19.40	1.375	4.94
89	904.4	258.3	87.80	25.08	0.289	70.23	251.9	8.50	1.625	4.18
90	980.0	110.0	95.14	10.68	0.112	27.10	251.9	1.106	1.80	3.78
Series XVIII										
12 mm Raschig rings										
108	1452.	1022.	141.0	99.2	0.704	169.0	311.3	63.50	1.136	5.978
109	2160.	440.0	209.7	42.7	0.204	70.81	338.8	14.73	1.330	5.110
110	1630.	650.0	158.2	63.1	0.399	126.2	338.8	28.32	1.205	5.640
111	1620.	760.0	157.3	73.8	0.469	150.8	338.8	46.34	1.236	5.477
112	1226.	1230.0	119.0	119.4	1.003	237.9	332.2	88.71	1.167	5.81
113	2040.	538.0	--	--	0.264	--	--	--	--	--
114	1590.	717.0	154.4	69.6	0.451	136.5	351.2	45.89	1.173	5.79
115	1000.	1390.	97.1	134.9	1.390	328.6	351.2	122.9	1.200	5.66
132	810.	1690.	78.63	164.1	2.086	392.1	353.6	167.2	1.098	6.18

TABLE IX

HETS for Near Constant Ratio CCl₄ and Reduced Flow Rates

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 8 mm Raschig rings, packing density 49.12 lbs/ft³
(e) 12 mm Raschig rings, packing density 25.91 lbs/ft³
(f) 1 percent acetone in feed CCl₄, no pulse

	Flow Rates ml/min		V, ft/hr		Flow Ratio CCl₄/H₂O	Millimols solute/liter			NTP	HETS ft	Sum of superficial velocities ft/hr
	Water	CCl₄	Water	CCl₄		Water Exit	CCl₄ Feed	CCl₄ Exit			
Series XIX	8 mm Raschig rings, intermediate interface										
127*	410.8	847.6	39.89	82.30	2.063	389.9	309.4	119.4	1.515	4.48	122.2
128	322.1	637.1	31.27	65.36	2.089	420.3	348.5	141.4	1.382	4.81	96.63
129	308.7	680.2	29.97	66.05	2.207	394.8	304.8	125.3	1.490	4.56	96.02
130	190.0	376.3	18.44	36.52	1.981	444.7	350.8	115.4	1.727	3.94	54.97
131	89.87	206.2	8.72	20.01	2.292	558.5	378.8	110.1	2.371	2.86	28.73
Series XX	12 mm Raschig rings, intermediate interface										
132*	810.	1690.	78.63	164.1	2.086	392.1	353.6	167.2	1.098	6.185	242.7
133	680.	1360.	66.00	132.0	2.00	395.2	361.4	172.3	1.080	6.28	198.0
134	441.	840.	42.60	81.20	1.905	420.9	361.4	145.8	1.332	5.10	124.4
135	208.	398.	20.20	38.70	1.917	473.5	361.4	133.4	1.646	4.11	58.9
136	107.	218.1	10.38	21.18	2.047	533.4	401.8	132.9	1.822	3.73	31.6
Series XXI	12 mm Raschig rings, CCl₄ continuous phase										
132*	810.	1690.	78.63	164.1	2.08	392.1	353.6	167.2	1.098	6.185	242.6
137	670.	1384.	65.00	134.5	2.063	322.8	319.2	163.9	.924	7.35	199.5
138	438.	842.	42.60	81.70	1.92	355.9	335.8	151.3	1.10	6.18	124.3
139	214.	400.	20.70	38.91	1.87	364.0	317.9	119.4	1.386	4.796	59.61
140	118.1	215.8	11.47	20.96	1.83	407.4	323.9	97.58	1.818	3.66	32.43

*flooded

B. Mass Transfer in Pulsed Packed Column

The column and packing characteristics for Tables X, XII, and XIII are the same as reported for Tables IV and V.

Table X, Series XXII and XXIII. HETS values are tabulated for a number of CCl_4/H_2O flow ratios at flooding at a pulsation rate of 125 cycles per minute with 5-mm amplitude.

Series XXII represents the runs with 8-mm Raschig rings.

Series XXIII represents the runs with 12-mm Raschig rings.

Table II, Series XXIV represents the runs made at reduced flow rates in a 3.32-inch column with 100.75 inches of packed section, packed with 8-mm Raschig rings. The pulsation rate was 125 cycles per minute with 5-mm amplitude. This column was constructed for an earlier thesis by Callihan [10]. Four of the six points reported in Series XXIV were reported by Callihan[10].

Table XII, Series XXV presents HETS at reduced flow rates for nearly constant ratio of CCl/H_2O obtained in the 2-inch column packed with 8-mm Raschig rings. The interface was located at the intermediate location as described earlier. The pulsation rate was 125 cycles per minute with 5-mm amplitude.

Table XIII. In Table XIII HETS data for several pulsation rates and several interface locations in the packed section are reported. The runs were performed on the 2-inch column packed with 12-mm Raschig rings. The flow ratio of carbon tetrachloride to water was maintained near 2. The flow rates span the range from flooded conditions to very low values.

These data are listed as Series XXVI to XXXI.

Series XXVI of Table XIII represents HETS data for an intermediate interface and a pulsation rate of 125 cycles per minute with 5-mm amplitude.

Series XXVII of Table XIII gives HETS for an intermediate interface and a pulsation rate of 125 cycles per minute, 10-mm amplitude.

Series XXVIII of Table XIII gives HETS for water as the continuous phase and a pulsation rate of 125 cycles per minute with 5-mm amplitude.

Series XXIX of Table XIII gives HETS for carbon tetrachloride as the continuous phase and a pulsation rate of 125 cycles per minute with 10-mm amplitude.

Series XXX of Table XIII gives HETS for an intermediate interface and a pulsation rate of 125 cycles per minute with 25-mm amplitude.

Series XXXI of Table XIII gives HETS for water as the continous phase and pulsation rate of 125 cycles per minute with 25-mm amplitude.

TABLE X

HETS for Variable Ratios of CCl₄/H₂O and Maximum Flow Rates in Pulsed Column

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) packing height 81.5 inches
(d) 8 mm Raschig rings, packing density 49.12 lbs/ft³
(e) 12 mm Raschig rings, packing density 25.91 lbs/ft³
(f) pulsation rate: 125 cycles/min., 5 mm amplitude
(g) 1 percent acetone in CCl₄ feed

Run	Flow Rates ml/min Water	Flow Rates ml/min CCl₄	V, ft/hr Water	V, ft/hr CCl₄	Flow Ratio CCl₄/H₂O	Millimols solute/liter Water Exit	Millimols solute/liter CCl₄ Feed	Millimols solute/liter CCl₄ Exit	NTP	HETS ft
Series XXII	**8 mm Raschig**									
98	302.6	1102.	29.38	107.0	3.64	609.8	313.9	134.9	3.10	2.19
99	480.0	833.3	46.60	80.90	1.74	514.3	367.0	66.1	3.11	2.18
100	520.6	756.0	50.54	73.39	1.45	456.1	366.8	41.5	3.10	2.19
101	566.2	635.4	54.97	61.68	1.12	384.6	354.9	24.6	3.23	2.10
102	558.7	523.5	54.24	50.82	0.937	332.7	351.1	11.13	3.61	1.88
103	510.7	417.8	49.58	40.56	0.818	349.1	410.7	5.57	4.15	1.63
104	397.2	363.7	38.56	35.31	0.916	293.4	315.9	2.63	5.08	1.34
Series XXIII	**12 mm Raschig**									
116	1340.	1450.	130.1	140.8	1.082	282.4	332.0	83.84	1.38	4.92
117	1640.	1085.	159.2	105.3	0.661	190.3	332.0	42.26	1.48	4.58
118	2145.	710.	208.2	68.9	0.331	108.6	332.0	17.33	1.53	4.44
119	1990.	810.	193.0	78.6	0.407	--	--	--	--	--
120	1800.	910.	174.7	88.3	0.506	203.9	424.9	34.90	1.60	4.26
121	2810.	445.	272.8	43.2	0.158	67.98	424.9	3.97	1.37	4.94
122	1570.	1290.	152.3	125.2	0.822	--	--	--	--	--
147	950.	1800.	92.26	174.7	1.89	320.9	273.	104.2	1.41	4.82

TABLE XI

HETS for Near Constant Ratio CCl$_4$/H$_2$O in Two Different Columns

(a) 2.127 inches diameter, packing height 30.75 inches
(b) 3.320 inches diameter, packing height 100.75 inches
(c) 8 mm Raschig rings
(d) pulsation rate: 125 cycles/min, 5 mm amplitude

Run	Flow Rates				Flow Ratio CCl$_4$/H$_2$O	Millimols solute/liter			NTP	HETS inches	Sum of superficial velocities ft/hr
	ml/min		V, ft/hr			Water	CCl$_4$				
	Water	CCl$_4$	Water	CCl$_4$		Exit	Feed	Exit			
Series XXIV											
2.127 inch column					Approx. 2.1					3.65	24.0
										10.20	82.6
										15.70	164.0
3.32 inch* column					2.1					7.0	45.6
141	353.	646.1	12.41	22.8	1.83	686.	380.	4.0	17.01	5.93	35.2
142**	417.	950.7	14.70	33.5	2.28	645.	323.	20.4	13.57	7.42	47.2

* from Callihan(10)
** flooded

TABLE XII

HETS for Near Constant Ratio CCl_4/H_2O in Pulsed Column

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) 8 mm Raschig rings, packing density 49.12 lbs/ft^3
(d) packing height 81.5 inches
(e) 1 percent acetone in CCl_4 feed
(f) pulsation rate: 125 cycles/min., 5 mm amplitude
(g) intermediate interface

Run	Flow Rates ml/min		V, ft/hr		Flow Ratio CCl_4/H_2O	Millimols solute/liter			NTP	HETS ft	Sum of superficial velocities ft/hr
	Water	CCl_4	Water	CCl_4		Water Exit	Feed	CCl_4 Exit			
Series XXV											
143*	462.0	871.0	44.85	84.56	1.89	575.3	379.7	80.6	3.00	2.26	129.4
144	358.2	672.9	34.77	65.26	1.88	532.8	352.2	67.1	3.19	2.14	100.0
145	196.7	380.8	19.10	36.94	1.93	588.5	358.8	50.85	3.90	1.74	56.0
146	104.1	190.4	10.12	18.48	1.83	612.5	358.8	34.0	6.03	1.12	28.6

*flooded

TABLE XIII

HETS for Pulsed Column and Near Constant Ratio of CCl_4/H_2O
Below Flooding

(a) 2 inch column with expanded end sections
(b) inlets directly in packing
(c) 12 mm Raschig rings, packing density 25.91 lbs/ft³
(d) 1 percent acetone in CCl_4 feed

Run	Comments	Flow Rates ml/min Water	CCl_4	V, ft/hr Water	CCl_4	Flow Ratio CCl_4/H_2O	Millimols solute/liter Water Exit	Feed	CCl_4 Exit	Packing Height inches	HETS ft	Sum of Superficial Velocities ft/hr
Series XXVI												
147*	5 mm amp 125 cycles/min Intermediate interface	950.0	1800.0	92.26	174.7	1.89	320.9	273.0	104.2	81.5	4.82	267.0
148		680.0	1285.0	66.01	124.7	1.89	326.0	273.0	99.08	81.5	4.56	194.7
149		442.0	800.0	42.91	77.66	1.81	344.4	273.0	86.47	81.5	3.86	120.6
150		200.0	381.3	19.42	37.02	1.91	429.6	302.9	78.82	81.5	2.87	56.44
151		112.5	206.7	10.92	20.07	1.84	445.9	302.9	55.73	81.5	2.18	30.99
Series XXVII												
152*	10 mm amp 125 cycles/min Intermediate interface	1150.0	2150.0	111.6	208.7	1.86	418.3	329.8	104.6	81.5	3.84	320.4
153		840.0	1740.0	81.55	168.9	2.07	438.2	320.2	109.4	81.5	3.65	250.5
154		687.5	1390.0	66.74	134.9	2.02	442.7	320.2	100.6	81.5	3.40	201.7
155		446.0	851.1	43.30	82.62	1.908	503.8	351.9	88.68	81.5	2.78	125.9
156		212.7	389.4	20.65	37.80	1.831	534.7	351.9	63.04	81.5	2.04	58.45
157		110.1	213.8	10.69	20.76	1.942	564.9	351.9	53.79	81.5	1.725	31.45

* Flooded

TABLE XIII (continued)

Run	Comments	Flow Rates ml/min Water	CCl₄	V, ft/hr Water	CCl₄	Flow Ratio CCl₄/H₂O	Millimols solute/liter Water Exit	Feed	CCl₄ Exit	Packing Height inches	HETS ft	Sum of Superficial Velocities ft/hr
Series XXVIII												
152*	H₂O continuous phase, 125 cpm, 10 mm amp	1150.0	2150.0	111.6	208.7	1.86	418.3	329.8	104.6	81.5	3.84	320.4
158		690.0	1370.0	67.0	133.0	1.98	454.3	331.7	98.80	80.0	3.25	199.9
159		440.0	843.0	42.7	81.8	1.91	524.8	358.8	87.43	80.0	2.58	124.5
160		203.6	390.0	19.75	37.87	1.916	522.3	331.7	59.48	80.0	1.87	57.62
161		108.8	206.0	10.60	20.00	1.894	556.3	335.9	52.12	80.0	1.23	30.6
Series XXIX												
152*	CCl₄ continuous phase, Intermediate interface, 125 cpm, 10 mm amp	1150.0	2150.0	111.6	208.7	1.86	418.3	329.8	104.6	81.5	3.84	320.4
162		697.5	1395.0	67.9	135.8	2.00	429.3	319.2	111.6	80.0	3.81	203.7
163		444.0	847.0	43.10	82.23	1.91	466.8	335.8	96.72	80.0	3.10	125.3
164		187.0	386.0	18.15	37.47	2.01	496.2	317.9	76.71	80.0	2.28	55.6
165		120.9	209.3	11.74	20.32	1.73	483.9	323.9	43.46	80.0	1.77	32.04
Series XXX												
166*	Intermediate interface, 25 mm amp, 125 cpm	1200.0	2316.0	116.4	225.0	1.93	482.1	337.9	93.24	80.0	2.17	341.4
167		430.0	860.0	41.7	83.5	2.00	599.7	371.9	79.08	80.0	1.58	125.2
168		108.8	207.5	10.56	20.14	1.907	612.4	371.9	50.40	80.0	1.30	30.7
Series XXXI												
166*	H₂O continuous phase, 25 mm amp, 125 cpm	1200.0	2316.0	116.4	225.0	1.93	482.1	337.9	93.24	80.0	2.17	341.4
169		430.0	860.0	41.74	83.49	2.00	696.2	430.5	86.67	80.0	1.55	125.2
170		105.0	211.1	10.19	20.49	2.01	585.8	350.1	56.65	80.0	1.20	30.68

* Flooded

C. <u>The Effect of Inlet Tubes on Mass Transfer</u>

The data reported here were obtained by performing runs on the two

end sections attached together and packed with 8-mm Raschig rings. The

inlet tubes were placed directly in the packing to give the same configura-

tion that was used in the column. The packed section in the attached end

section measured 9.5 inches between packing supports. The bulk density

of the packing was determined to be 49.54 lb/ft^3.

<u>Table XIV</u> presents NTP in the end sections for pulsed and unpulsed

conditions for a limited ratio of CCl_4/H_2O and a large range of flow

rates. The flow rates span the range from very low rates to flooding.

<u>Series XXXII of Table XIV</u> lists NTP for unpulsed operation.

<u>Series XXXIII of Table XIV</u> lists NTP for a pulsation rate of 125

cycles per minute with 5-mm amplitude.

<u>Series XXXIV of Table XV</u> lists NTP for a pulsation rate of 125

cycles per minute with 10-mm amplitude.

<u>Table XV</u> presents the contribution of the inlet tubes for the 2-inch

diameter column end sections. Calculations for the inlet tube effect were

determined for pulsed and unpulsed column operation.

<u>Column 1 of Table XV</u> presents NTP data obtained from pulsed and

unpulsed column operation. The NTP vs superficial velocities from Series XXV

and XIX were plotted and smoothed values of NTP at various superficial

velocities were read from the plot.

<u>Column 7 of Table XV</u> lists the superficial velocities.

<u>Column 2 of Table XV</u> lists the NTP in the end sections for the same

operating conditions to which Column 1 applies. The NTP in the end sections vs superficial velocity from Series XXXIII and XXXII were plotted. The NTP at superficial velocities listed in Column 7 were read.

Column 3 of Table XV lists NTP in the length of column (72 inches) between the end sections. This is obtained by subtracting Column 2 from Column 1.

Column 4 of Table XV lists the calculated HETS for the packing between the end sections. This is obtained by dividing NTP of Column 3 into 72 inches.

Column 5 of Table XV lists the NTP in the packing of the end sections. This is obtained by dividing HETS of Column 4 into 9.5 inches.

Column 6 of Table XV lists the NTP contributed by the inlet tubes. This is given by the difference of Column 2 and Column 5.

TABLE XIV

NTP in 2 inch Diameter Column End Sections

(a) inlet tubes placed directly in packing
(b) packing height in end sections 9.5 inches
(c) packing density 49.54 lbs/ft^3
(d) 8 mm Raschig rings

Run	Comments	Flow Rates ml/min		Flow Rates V, ft/hr		Flow ratio CCl$_4$/H$_2$O	Millimols solute/liter Water Exit	CCl$_4$ Feed	CCl$_4$ Exit	NTP	Sum of Superficial Velocities ft/hr
		Water	CCl$_4$	Water	CCl$_4$						
Series XXXII	Intermediate Interface unpulsed										
171		478.0	900.0	46.40	87.37	1.88	118.7	386.7	320.5	.203	133.8
172		383.2	796.0	37.20	72.27	2.07	116.7	306.0	228.0	.273	114.5
173		285.0	581.0	27.67	56.40	2.04	73.0	355.0	303.0	.137	84.07
174		133.9	231.8	13.00	22.50	1.73	114.5	286.2	220.0	.263	35.60
Series XXXIII	Intermediate Interface 125 cpm 5 mm amp										
175		500.0	1050.0	48.54	101.9	2.1	266.6	428.2	298.6	.425	150.5
176		388.5	810.8	37.72	78.71	2.09	180.8	337.8	234.6	.384	116.4
177		265.3	539.6	25.76	52.38	2.03	193.1	309.7	204.0	.463	78.15
Series XXXIV	Intermediate Interface 125 cpm 5 mm amp										
178		600.0	1090.0	58.25	105.8	1.82	187.2	287.0	182.6	.504	164.1
179		393.5	769.7	38.20	74.72	1.96	199.0	325.0	209.1	.478	112.9
180		289.4	572.1	28.09	55.54	1.98	223.1	327.8	208.8	.513	83.64
181		163.3	245.8	15.85	23.86	1.505	229.6	303.6	229.6	.800	39.71

TABLE XV

NTP in the Inlet Tubes for 2 inch Diameter Column

NTP in column, end sections, and tubes	NTP in end sections and tubes	NTP in 72 inch length of column	HETS inches	NTP in 9.5 inches of packing in end sections	NTP in inlet tubes	Sum of superficial velocities ft/hr
Series XXV **Runs 143-146**						
3.00	0.40	2.60	27.7	0.34	0.06	129.4
3.19	0.42	2.77	25.9	0.37	0.05	100.0
3.90	0.54	3.36	21.4	0.44	0.10	56.04
6.03	0.70	5.33	13.5	0.70	0.00	28.60
Series XIX **Runs 127-131**						
1.515	0.20	1.32	54.5	0.17	0.03	122.2
1.382	0.20	1.18	61.0	0.16	0.04	96.6
1.490	0.20	1.29	55.8	0.17	0.03	96.0
1.727	0.20	1.53	47.0	0.20	0.00	54.97
2.371	0.28	2.17	33.2	0.28	0.00	28.73

SECTION III

CALCULATIONS BASED ON HYDRODYNAMIC AND MASS TRANSFER

CONSIDERATIONS

In this section particular emphasis is given to the application

of the theory presented earlier in this thesis. The results of the

calculations based on hydrodynamic and mass transfer considerations of a

dispersed phase flowing through a continuous phase in packed columns are

presented in tables. The calculations require no new experimental data.

The experimental data collected by this writer and previous investigators

serve only as a check of the theory. The calculated performance charac-

teristics are tabulated as K_Ga and the experimentally observed values

are labeled K_Ga observed.

Table XVI presents the calculations of characteristics drop diameters,

terminal velocities of characteristic drop diameters, and characteristic

velocities. The liquid systems, the pertinent physical properties, and

column characteristics are also tabulated. These data are used in the

calculations reported in subsequent tables.

In several cases the physical properties and packing characteristics

had to be estimated. Where certain values of packing characteristics

were not available from the investigator cited, the writer supplied these

data from other sources for similar packing sizes. In every case the

appropriate references are given in the tables.

Table XVII presents the results of calculations for the system

3-pentanol-water. The observed values for ka were reported by Lhadda and

Smith [33]. It is to be noted that individual volumetric film coefficients

are reported in this case.

The holdup, x, was calculated from the equation referred to as Case II

in the theory, i.e. $\left[V_d/x + V_c/(1-x)\right] = \epsilon V_0 (1-x)$.

The velocity of the dispersed phase relative to the continuous phase

is tabulated as $\left[V_d/\epsilon x + V_c/\epsilon (1-x)\right]$.

The drop diameter and the surface area of the drops are listed as

d_{vs} and a_d respectively.

The Reynolds number for the continuous and dispersed phase and

corresponding mass transfer factor are tabulated as $Re_{c,d}$ and $\left[6 Sc\ k/u_r\right]_{c,d}$

respectively. Each of these terms corresponds to the properties of the

phase being considered.

Table XVIII presents the results of calculations for the system

benzene-water with acetic acid transfer from the water phase. The

experimental over-all volumetric mass transfer coefficients based on the

benzene film measured by Sherwood and co-workers [52] are also tabulated.

In addition to the variables presented in Table XVIII, Table XIX

presents the diffusity, D_f, for each of the phases and the distribution

coefficient, D. The former is required to calculate the Schmidt number

for the mass transfer factor; the latter for the over-all mass transfer

coefficient from the expression $1/K_G = (D/k_L) + (1/k_G)$. See

Appendix A for the derivation.

Table XIX lists the calculations for the system methyl isobutyl

ketone-water with acetic acid transfer from the water phase. The observed

values of over-all volumetric mass transfer coefficients were measured by
Sherwood and co-workers [52].

Table XX lists the results of calculations for the system toluene-
water with diethylamine transfer from the continuous water phase. Leibsan
and Beckman [35] measured over-all mass transfer coefficients for this
system.

Table XXI lists the results of calculations for the system carbon
tetrachloride-water with acetone transfer from the carbon tetrachloride
phase at flooding. The conditions for which the calculations are tabulated
are consistent with the experimental runs reported as Series XVIII. The
measured values of over-all mass transfer coefficients are also listed.

The calculations for holdup, x, in Table XXI were made by applying
the equation, $\left[V_d/X + V_c/(1-X) \right] = \epsilon V_0$ at flooded conditions.
Appendix C presents the derivation for holdup at flooding.

Table XXII. The results presented in Table XXII are for the same
system and conditions as reported in Table XXI. The difference lies in
the holdup, x. In this table the holdup x was chosen as 0.5 in every
case. This will be taken up in the discussion.

Table XXIII lists the results of calculations for carbon tetrachloride-
water with acetone transfer from carbon tetrachloride below flooding. ·-
The conditions for the calculations are consistent with those reported
for Series XXI. The experimental values of over-all mass transfer coeffi-
cients are also listed.

Table XXIV presents the results of calculations for the same system

and conditions as reported in Table XXIV. The differences in tabulated values are accounted for by the mechanism of flow assumed for the dispersed water phase. In Table XXIII, discrete drops were assumed for the dispersed phase. In Table XXIV, film type flow of the dispersed phase was assumed to allow for the preferential wetting characteristics of the water phase.

Table XXV presents the calculations for the system carbon tetrachloride-water with acetone transfer from the dispersed carbon tetrachloride phase. The flow rates are below flooding.

The values for the observed values of over-all volumetric mass transfer were not measured directly. They were calculated from HETS values for carbon tetrachloride as the continuous phase (Series XXI), and HETS values for the intermediate interphase (Series XX). The interphase position was 44.5 inches from the base of the packing. The expression which relates the HETS values and interface position is:

$$(NTP)_{Intermediate} = \left[37/(HETS)_{H_2O}\right] + \left[44.5/(HETS)_{CCl_4}\right] = \left[81.5/(HETS)_{Intermediate.}\right]$$

Table XXVI presents the calculations of over-all volumetric mass transfer coefficients from HETS values. These values are reported as the measured values in Tables XXI-XXV. The equation which expresses the relationship between them is,

$$K_G a = (G/A) \left[\ln(L/GD)\right] \Big/ \left[(HETS)\ (1-GD/L)\right]$$. This expression is derived in Appendix A.

Table XXVII lists the results of calculations in pulsed columns at reduced flow rates for the system carbon tetrachloride and water with acetone transfer. The conditions for which the calculations were performed

are consistent with those reported for Series XXVII and XXIX.

With pulsation it was observed and stated previously that the holdup is higher and drop diameter smaller than without pulsation. The theory which was developed for non-pulsed operation can be applied in pulsed columns after modifications. In order to be consistent with the observed phenomena of increased holdup and reduced drop diameter, the characteristic velocity was arbitrarily reduced by one half of the pulse velocity in the packing.

Table XXVIII lists the results of calculations for the system carbon tetrachloride-water in a pulsed column at flooding. The calculated and measured values are for the operating conditions reported in Series XXIII.

Table XXIX presents the calculations of over-all volumetric mass transfer coefficients from HETS values. These values are reported as the measured values in Tables XXVII-XXVIII.

TABLE XVI

Physical Properties, Characteristic Drop Diameters and Characteristic Velocities

Liquid	Density, ρ, lbs/ft³	Viscosity, μ, lbs/ft-hr	Interfacial tension, γ, lbs/hr²
Water	62.4	2.42*	
CCl₄	99.84	2.42	1.036 x 10⁶
3 Pentanol	51.1	9.44	0.114 x 10⁶
Benzene	54.1	1.62	0.854 x 10⁶
MIK	50.0	1.40	0.3247 x 10⁶
Toluene	54.0	1.43	1.026 x 10⁶

*2.25 is used sometimes

System	Disperse phase	Ring type	d_p, ft	Column diam ft	a_p ft²/ft³	ε fraction voids	d_o ft	s ft	v_t ft/hr	Ref	v_o/v_t	v_o ft/hr
CCl₄-H₂O	CCl₄	Raschig 12 mm	.0402	.167	115.2	.821	.0075	.0078	2050	15	.487	996
	H₂O (33)	Raschig 1/4 in.	.0402	.167	115.2	.821 (8)	.0075	.0078	1785	15	.645	1150
3 pentanol-H₂O	3 pentanol (33)	Raschig 1/4 in.	.0204	.167	208.0	.534 (8)	.00452	.0034	590	33, I.C.T.	.76	448
	H₂O (52)	Raschig 1 in.	.0204	.167	208.0	.534 (14)	.00452	.0034	348	15, 52	1.00	348
MIK-H₂O	MIK (35)	carbon 1/2 in.	.0825	.292	60.7	.728	.0073	.02435	996	14, 52	.773	770
Toluene-H₂O	Toluene (52)	Raschig 1 in.	.0417	.500	103.0 (14)	.500 (14)	.0141	.00174	1390	8, 35	.155	216
Benzene-H₂O	Benzene	carbon	.0825	.292	60.7	60.7	.0152	.01645	1325		.445	590

TABLE XVII

Calculations for 3-Pentanol-Water

Ref	Continuous / Disperse	d_o, ft	v_o, ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft²/ft³
(33) R 248	3-pentanol / water	.00452	348	12.65 / 31.8	.26	261	.00602	138.5
(33) R 221	3-pentanol / water	.00452	348	23.1 / 7.02	.046	332	.00474	31.2
(33) R 176	water / 3-pentanol	.00452	448	34.10 / 10.37	.055	424	.00478	36.8
(33) R 183	water / 3-pentanol	.00452	448	26.2 / 15.6	.080	418	.00485	52.8

Ref	Continuous / Disperse	μ lbs/ft hr	$(Sc)_{c,d}$	$u_{rc,\,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	ka	ka observed
(33) R 248	3-pentanol / Water	9.44 / 2.42	3720 / 960	67 / 199	2.48 / 31.0	60. / 6.8	.186 / .235	24.9 / 32.6	23.6 / 30.9
(33) R 221	3-pentanol / Water	9.44 / 2.42	3720 / 960	85 / 247	5.9 / 30.1	28. / 6.8	.107 / .292	3.32 / 9.15	6.05 / 8.65
(33) R 176	Water / 3-pentanol	2.42 / 9.44	960 / 3720	315 / 109	100.5 / 2.48	4.0 / 55.0	.220 / .269	8.10 / 9.90	17.3 / 12.1
(33) R 183	Water / 3-pentanol	4.42 / 9.44	960 / 3720	311 / 107	89. / 2.81	4.5 / 55.	.243 / .264	12.9 / 14.0	25.9 / 18.75

TABLE XVIII

Calculations for Benzene-Water
(acetic acid solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{VB}, ft	a_d, ft²/ft³
(52) Run 33	Water Benzene	.0152	590	10 30	.078	543	.0165	20.6
(52) Run 36	Water Benzene	.0152	590	60 30	.092	539	.01665	24.1

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft²/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	K_Ga	K_Ga observed
(52) Run 33	Water Benzene	2.25 1.62	4.85 x 10⁻⁵ 7.95 x 10⁻⁵	744 371	152 391	209 219	3.0 2.9	.102 .510	.0303	.442	9.12	10.5
(52) Run 36	Water Benzene	2.25 1.62	4.85 x 10⁻⁵ 7.95 x 10⁻⁵	744 371	151 388	191 219	3.1 2.9	.105 .506	.0303	.440	10.6	10.5

TABLE XIX

Calculations for Methyl Isobutyl Ketone-Water
(acetic acid solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft^2/ft^3
(52) R 21	Water MIK	.0073	770.	10.0 40.0	.080	702	.0080	43.7
(52) R 22	Water MIK	.0073	770.	40.0 40.0	.0865	695	.00808	46.8

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft^2/hr	$(Sc)_{c,d}$	u_{rc},rd ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	$\dfrac{K_G a}{}$	$K_G a$ observed
(52) R21	water MIK	2.25 1.40	4.85 x 10^{-5} 8.70 x 10^{-5}	744 322	265 437	282 125	2.5 3.5	.1485 .792	.50	.216	9.45	21.6
(52) R21	water MIK	2.25 1.40	4.85 x 10^{-5} 8.70 x 10^{-5}	744 322	263 432	270 124.5	2.5 3.5	.1472 .783	.50	.214	10.1	44.5

TABLE XX

Calculations for Toluene-Water
(diethylamine solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft²/ft³
(35) 6-3-1	Water / Toluene	.0141	216.	7.86 / 5.18	.043	205.	.0148	10.9
(35) 6-3-14	Water / Toluene	.0141	216.	15.12 / 18.84	.21	173.3	.01755	45.1
(35) 6-3-15	Water / Toluene	.0141	216.	23.70 / 8.94	.092	197.	.01545	22.4

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft²/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\frac{6(Sc)k}{u_T}\right]_{c,d}$	$k_{c,d}$	D	K_G	K_Ga	K_Ga observed
(35) 6-3-1	Water / Toluene	2.42 / 1.43	3.39×10^{-5} / 6.11×10^{-5}	1140 / 432	84 / 121	68.4 / 67.6	5.0 / 5.0	.0614 / .234	.810	.0572	.625	1.08
(35) 6-3-1	Water / Toluene	2.42 / 1.43	3.39×10^{-5} / 6.11×10^{-5}	1140 / 432	70.8 / 102.5	36.7 / 67.8	7.0 / 5.0	.0725 / .198	.74	.0655	2.96	4.68
(35) 6-3-1	Water / Toluene	2.42 / 1.43	3.39×10^{-5} / 6.11×10^{-5}	1140 / 432	80.5 / 116.5	56.6 / 68.0	5.5 / 5.0	.0647 / .224	.752	.0622	1.44	2.34

TABLE XXI

Calculations for Carbon Tetrachloride-Water at Flooding
(acetone solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft²/ft³
Series XVIII 132	Water CCl₄	.0075	996	78.63 164.1	.590	573	.01302	220.
112	Water CCl₄	.0075	996	119.0 119.4	.500	583	.01280	192.5
114	Water CCl₄	.0075	996	154.4 69.61	.402	525	.01422	139.0
109	Water CCl₄	.0075	996	209.7 42.72	.311	535	.01398	109.3

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft²/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\frac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	$K_G a$	$K_G a$ observed
132	Water CCl₄	2.42 2.42	4.24 x 10⁻⁵ 4.67 x 10⁻⁵	915 519	287 287	44.7 156.5	6.0 3.2	.312 .295	2.135	.0957	21.0	12.7
112	Water CCl₄	2.42 2.42	4.24 x 10⁻⁵ 4.67 x 10⁻⁵	915 519	292 292	60.3 154.0	5.2 3.2	.277 .300	2.135	.0930	17.9	13.5
114	Water CCl₄	2.42 2.42	4.24 x 10⁻⁵ 4.67 x 10⁻⁵	915 519	263 263	78.8 154.5	4.5 3.2	.216 .270	2.135	.0798	11.0	11.0
109	Water CCl₄	2.42 2.42	4.24 x 10⁻⁵ 4.67 x 10⁻⁵	915 519	268 268	106.2 155.0	4.0 3.2	.195 .276	2.135	.0777	8.5	10.63

TABLE XXII

Calculations for Carbon Tetrachloride-Water at Flooding (acetone solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft^2/ft^3
Series XVIII								
132	Water	.0075	996	78.63	.50	592	.01270	194.
	CCl$_4$			164.1				
112	Water	.0075	996	119.0	.50	583	.01280	192.5
	CCl$_4$			119.4				
114	Water	.0075	996	154.4	.50	546	.01370	180.
	CCl$_4$			69.61				
109	Water	.0075	996	209.7	.50	614	.01215	203.
	CCl$_4$			42.72				

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft^2/hr	(Sc)$_{c,d}$	$u_{rc,rd}$ ft/hr	(Re)$_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	K_Ga	K_Ga observed
132	Water	2.42	4.24 x 10^{-5}	915	296	60.	5.2	.280	2.135	.0944	18.3	12.70
	CCl$_4$	2.42	4.67 x 10^{-5}	519	296	151.	3.2	.304				
112	Water	2.42	4.24 x 10^{-5}	915	292	60.3	5.2	.277	2.135	.0930	17.9	13.50
	CCl$_4$	2.42	4.67 x 10^{-5}	519	292	154.	3.2	.300				
114	Water	2.42	4.24 x 10^{-5}	915	273	59.0	5.2	.259	2.135	.0852	15.3	11.00
	CCl$_4$	2.42	4.67 x 10^{-5}	519	273	152.5	3.2	.271				
109	Water	2.42	4.24 x 10^{-5}	915	307	66.0	5.15	.288	2.135	.0975	19.6	10.63
	CCl$_4$	2.42	4.67 x 10^{-5}	519	307	153.5	3.2	.316				

TABLE XXIII

Calculations for Carbon Tetrachloride-Water Below Flooding
(acetone solute)

Ref Series XXI	Continuous Disperse	d_o, ft	v_o, ft	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft^2/ft^3
140	CCl$_4$	.0075	1150	20.96	.0126	1146	.00753	8.25
	water			11.47				
139	CCl$_4$	.0075	1150	38.91	.0234	1124	.00766	15.10
	water			20.70				
138	CCl$_4$	.0075	1150	81.7	.0526	1071	.00805	32.20
	water			42.6				
137	CCl$_4$	.0075	1150	134.5	.0915	980	.0088	51.30
	water			65.0				

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft^2/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	$K_{G}a$	K_Ga observed
140	CCl$_4$	2.42	4.67x10^{-5}	519	573	934	1.9	.350	2.135	.1180	.975	2.96
	water	2.42	4.24x10^{-5}	915	573	111	4.0	.418				
139	CCl$_4$	2.42	4.67x10^{-5}	519	562	855	1.9	.344	2.135	.1155	1.745	3.99
	water	2.42	4.24x10^{-5}	915	562	111	4.0	.410				
138	CCl$_4$	2.42	4.67x10^{-5}	519	535.5	700	1.95	.336	2.135	.1120	3.61	6.60
	water	2.42	4.24x10^{-5}	915	535.5	111	4.0	.390				
137	CCl$_4$	2.42	4.67x10^{-5}	519	490	543	2.2	.346	2.135	.1115	5.72	8.17
	water	2.42	4.24x10^{-5}	915	490	111	4.0	.357				

TABLE XXIV

Calculations for Carbon Tetrachloride-Water Below Flooding
(acetone solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft^2/ft^3
Series XXI 140	CCl$_4$ water	film	1150	20.96 11.47	.0126	1146		115.2
139	CCl$_4$ water	film	1150	38.91 20.70	.0234	1124		115.2
138	CCl$_4$ water	film	1150	81.70 42.60	.0526	1071		115.2
137	CCl$_4$ water	film	1150	134.5 65.0	.0915	980		115.2

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft^2/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	$K_G a$	$K_G a$ observed
140	CCl$_4$ water	2.42 2.42	4.67x10^{-5} 4.24x10^{-5}	519 915	573. 573.	1000.0 3.98	1.9 40.0	.350 4.17	2.135	.158	18.2	2.96
139	CCl$_4$ water	2.42 2.42	4.67x10^{-5} 4.24x10^{-5}	519 915	562. 562.	970.0 7.26	1.9 23.0	.343 2.36	2.135	.1505	17.35	3.99
138	CCl$_4$ water	2.42 2.42	4.67x10^{-5} 4.24x10^{-5}	519 915	535.5 535.5	897.0 15.5	1.9 15.0	.327 1.465	2.135	.1385	15.97	6.60
137	CCl$_4$ water	2.42 2.42	4.67x10^{-5} 4.24x10^{-5}	519 915	490. 490.	786.0 24.7	1.95 10.0	.307 .892	2.135	.124	14.30	8.17

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TABLE XXV

Calculations for Carbon Tetrachloride-Water Below Flooding
(acetone solute)

Ref	Continuous Disperse	d_o, ft	v_o, ft	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft²/ft³
	water CCl₄	.0075	996	10.38 21.18	.027	966	.00774	17.2
	water CCl₄	.0075	996	20.2 38.7	.0514	796	.0091	27.8
	water CCl₄	.0075	996	42.6 81.2	.1210	722	.01035	57.6
	water CCl₄	.0075	996	66.0 132.0	.250	616	.0121	102.0

Flow rates are the same as reported for Runs 133-136

Ref	Continuous Disperse	μ lbs/ft-hr	D_f(58) ft²/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\frac{6(Sc)k}{u_r}\right]_{c,d}$	$K_{c,d}$	D	K_G	K_Ga	K_Ga observed
136	water CCl₄	2.42 2.42	4.24x10⁻⁵ 4.67x10⁻⁵	915 519	484 484	452.0 154.3	2.4 3.2	0.216 0.497	2.135	.112	1.92	2.75
135	water CCl₄	2.42 2.42	4.24x10⁻⁵ 4.67x10⁻⁵	915 519	398 398	355.0 149.5	7.0 3.2	0.507 0.409	2.135	.139	3.86	5.55
134	water CCl₄	2.42 2.42	4.24x10⁻⁵ 4.67x10⁻⁵	915 519	361 361	233.0 154.0	10.0 3.2	0.657 0.371	2.135	.1375	7.92	10.05
133	water CCl₄	2.42 2.42	4.24x10⁻⁵ 4.67x10⁻⁵	915 519	308 308	136.0 153.3	18.0 3.2	1.01 0.316	2.135	.129	13.15	12.0

TABLE XXVI

Calculations of K_Ga From HETS

$$K_Ga = (G/A)\left[\ln(L/GD)\right]/(HETS)\ 1-GD/L$$

Ref	HETS ft	G/A ft/hr	Superficial velocity V_d+V_c, ft/hr	$L/GD = (G/A)[\ln(L/GD)]/(Y_2-Y_1)/(X_2-X_1)D$	$\ln(L/GD)$	1-GD/L	$(G/A)\ln L/GD$	HETS(1-GD/L)	K_Ga
Series XVIII									
132	6.185	78.63	242.7	.9850	-0.01518	-0.01522	- 1.192	- 0.09406	12.70
112	5.81	119.0	238.4	.4577	-0.7807	-1.1848	-93.00	- 6.8837	13.50
114	5.79	154.4	224.0	.2094	-1.5617	-3.7755	-241.3	-21.8601	11.00
109	5.11	209.7	252.4	.1023	-2.277	-8.7752	-478.0	-44.841	10.63
Series XX									
136	3.73	10.38	31.6	.9293	-0.07323	-0.0760	- 0.76013	- 0.2835	2.68
135	4.11	20.20	58.9	.9728	-0.02755	-0.0279	- 0.5565	- 0.1145	4.85
134	5.10	42.60	124.4	.9143	-0.08949	-0.09373	- 3.8123	- 0.4780	7.96
133	6.28	66.00	198.0	.9789	-0.02130	-0.02155	- 1.4058	- 0.1353	10.4
Series XXI									
140	3.663	11.47	32.43	.8431	-0.17048	-0.18609	- 1.9554	- 0.6610	2.96
139	4.796	20.70	59.61	.8590	-0.15182	-0.16414	- 3.1427	- 0.7875	3.99
138	6.18	42.60	124.3	.9035	-0.10136	-0.10680	- 4.3179	- 0.6550	6.60
137	7.35	65.0	199.5	.9733	-0.02703	-0.02743	- 1.7570	- 0.2150	8.17
	3.58	10.38	31.46	.9051	-0.09959	-0.10485	- 1.03374	- 0.3760	2.75
	3.58	20.20	58.90	.8973	-0.10824	-0.11445	- 2.1864	- 0.3940	5.55
	4.00	42.6	124.0	.8949	-0.11093	-0.11744	- 4.7265	- 0.4700	10.05
	5.30	66.0	198.0	.9367	-0.06532	-0.06757	- 4.3111	- 0.3580	12.0

H_2O continuous phase, HETS calculated from Series XX and XXI

TABLE XXVII

Calculations for Carbon Tetrachloride-Water in Pulsed Column

(a) below flooding; (b) 125 cycles/min, 10 mm amp; (c) pulse velocity, V_p, 493 ft/hr

Ref	Continuous Disperse	d_o, ft	$\dfrac{v_o-V_p/2\epsilon}{\epsilon}$ ft/hr	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/(\epsilon(1-x)))$	d_{vs}, ft	a_d, ft²/ft³
Series XXVIII								
158	water	.0075	696	67.0	.50	487	.0107	230.0
	CCl4			133.0				
159	water	.0075	696	42.7	.203	553	.0093	107.5
	CCl4			81.4				
161	water	.0075	696	10.6	.037	671	.00778	23.4
	CCl4			20.0				
Series XXIX								
162	CCl4	.0075	830	135.8	.170	687	.00906	92.5
	water			67.9				
165	CCl4	.0075	830	20.32	.018	820	.00760	11.68
	water			11.74				

Ref	Continuous Disperse	μ lbs/ft-hr	D_f(58) ft²/hr	$(Sc)_{c,d}$	$u_{rc,rd}$ ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	K_G^a	K_G^a observed
158	water	2.42	4.24x10⁻⁵	915	244	45.0	6.0	.266	2.135	.0948	21.8	19.68
	CCl4	2.42	4.67x10⁻⁵	519	244	108.0	4.0	.314				
159	water	2.42	4.24x10⁻⁵	915	277	125.5	3.8	.192	2.135	.0893	9.60	15.73
	CCl4	2.42	4.67x10⁻⁵	519	277	106.3	4.0	.356				
161	water	2.42	4.24x10⁻⁵	915	336	279.0	2.8	.1715	2.135	.0930	2.18	8.06
	CCl4	2.42	4.67x10⁻⁵	519	336	108.0	4.0	.432				
162	water	2.42	4.24x10⁻⁵	915	344	280.0	2.8	.310	2.135	.0989	9.14	17.53
	CCl4	2.42	4.67x10⁻⁵	519	344	80.5	5.0	.312				
165	water	2.42	4.24x10⁻⁵	915	410	644.0	2.1	.276	2.135	.0960	1.12	5.94
	CCl4	2.42	4.67x10⁻⁵	519	410	80.3	5.0	.374				

TABLE XXVIII

Calculations for Carbon Tetrachloride-Water in Pulsed Columns

(a) flooding; (b) 125 cycles/min, 5 mm amp; (c) pulse velocity, V_p, 246 ft/hr

Ref	Continuous Disperse	d_o, ft	$\dfrac{v_o-V_p/2\epsilon}{ft/hr}$	V, ft/hr	x, holdup	$(V_d/\epsilon x)+(V_c/\epsilon(1-x))$	d_{vs}, ft	a_d, ft²/ft³
Series XXIII								
120	water CCl4	.0075	846	174.7 88.3	.50	642	.010	246.0
118	water CCl4	.0075	846	208.2 68.9	.50	676	.00939	262.5

Ref	Continuous Disperse	μ lbs/ft-hr	D_f (58) ft²/hr	$(Sc)_{c,d}$	u_{rc}, r_d ft/hr	$(Re)_{c,d}$	$\left[\dfrac{6(Sc)k}{u_r}\right]_{c,d}$	$k_{c,d}$	D	K_G	$K_G a$	$K_G a$ observed
120	water CCl4	2.42 2.42	4.24x10⁻⁵ 4.67x10⁻⁵	915 519	321 321	56.3 322.0	5.3 2.5	.310 .258	2.135	.0868	21.4	18.70
118	water CCl4	2.42 2.42	4.24x10⁻⁵ 4.67x10⁻⁵	915 519	338 338	56.8 130.5	5.3 18.0	.326 .196	2.135	.0715	18.7	16.46

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TABLE XXIX

Calculation of K_Ga From HETS

$$K_Ga = (G/A)\left[\ln(L/GD)\right]/HETS\left[1-GD/L\right]$$

Ref	HETS ft	G/A ft/hr	Superficial velocity V_d+V_c, ft/hr	L/GD = $(Y_2-Y_1)/(X_2-X_1)D$	ln L/GD	1-GD/L	(G/A)ln L/GD	HETS(1-GD/L)	K_Ga
158	3.25	67.0	200.0	.9133	-0.09060	-0.09493	- 6.0702	- 0.3085	19.68
159	2.58	42.7	124.0	.9058	-0.09881	-0.10399	- 4.2192	- 0.26829	15.73
161	1.23	10.6	30.6	.8770	-0.13110	-0.14025	- 1.3897	- 0.17251	8.06
162	3.18	67.9	203.7	.9686	-0.03188	-0.03241	- 2.1646	- 0.12348	17.53
165	1.77	11.74	32.06	.8075	-0.21358	-0.23839	- 2.5074	- 0.42195	5.94
120	4.26	174.7	263.0	.2450	-1.4049	-3.0816	-245.436	-13.1276	18.70
118	4.44	208.2	277.1	.1617	-1.8200	-5.1843	-378.924	-23.0183	16.46

DISCUSSION

SECTION I

FLOODING CHARACTERISTICS IN PACKED COLUMNS

Various studies in the literature report the maximum flows attainable in packed columns. In all cases much data are presented and attempts are made at correlating the results in terms of packing characteristics such as void fraction and effective area of packing, and physical properties of the liquids.

At best the correlations are to be used with caution since the maximum flows in packed columns are frequently influenced greatly by various additional characteristics which are normally overlooked. Again, in small size packing much higher flow rates can be attained in dirty packing than in clean packing. For example in both small and large packing, mass transfer under certain conditions increases the maximum flows in the column and under other conditions reduces them. The flow capacity in the packing is very much influenced by the non-uniformity inherent in random packing of the column. The subsequent discussion explains these phenomena in terms of observations and measurements which have been presented in Section I of the Experimental Results.

Maximum Flow Rates Without Mass Transfer

Effect of Dirty Packing on Flooding. The maximum flow rates in packed columns can sometimes be much greater for a dirty or conditioned packing than for a clean packing. Two distinct flooding curves are shown in Figure 4. The lower curve represents flooding in the clean packing and the upper curve represents flooding in the dirty, or conditioned, packing. The dashed line in Figure 6 represents the flooding curve based on flood point data reported by Callihan for the system tap water-carbon tetrachloride.

The unusual curve resulting from the data reported by Callihan is apparently due to the different degrees of conditioning of the packing for each of the flood points. Continuous operation results in higher maximum flow rates. This effect is observed in Figure 5 by the points marked 0, 1, 2, 3, 4, and 5 days. It is apparent that a higher flooding curve could be measured for each of the 5 days.

A sharp reduction in column capacity occurred when the dirty column was cleaned by acid treating. Reference is made to Runs 1 to 3 in Table I. These points indicate higher flooding rates than the subsequent values. The runs which follow Run 3 are for the column when cleaned by acid treating. Run 34 also indicates a sharp reduction when the column is cleaned by acid treating.

The increased capacity with continuous operation is apparently due to a bridge formation at the points of contact between the packing elements. The bridge is formed from solids, dissolved in the water,

that gradually precipitate and build up on the packing. Bridge formation

reduces the randomness of flow by forming channels that aid rivulet-type

flow. The solids that form the bridge are dissolved by acid treating

and are broken down by pulsing the liquids. Evidence of the latter is

given by Run 47 of Table II in the Experimental Results.

It is difficult to bridge large packing sizes, such as 12-mm Raschig

rings, since the individual packing elements are not nearly close enough

in randomly packed situations. Therefore, it is expected that for clean

water, such as distilled water, continuous operation will not result

in increased column capacities. Also the capacities in large packing

sizes are expected to be independent of time. These phenomena are

observed in Figure 5. The lower curve represents flooding for the clean

8-mm Raschig rings with distilled water. The intermediate curve represents

flooding for dirty 8-mm Raschig rings. The upper curve represents flooding

for both clean and dirty conditions for 12-mm Raschig rings.

<u>Visual Observations at Flooding</u>. Flooding in packed columns is

characterized by increased holdup of the dispersed phase. There is

evidence that this holdup is neither uniform nor constant. The continually

changing holdup in some sections of the column results in nonuniform

flow of the dispersed and continuous phases in the section. This could

lead to velocities at the section which surpass the maximum allowable flow

rates. Flooding could start at the section and grow until the entire

column is flooded. However, it is also likely that the holdup will change

at the section before the entire column is flooded and a redistribution

of the phases will occur. This phenomenon was observed and reported as erratic column behavior in the Experimental Results. This also explains the up and down movements of the interfaces at flooding when no external changes were made in the volume flow rates to the column.

Regardless of which phase was maintained as the continuous phase below flooding, at flooding the carbon tetrachloride and the water phases were redistributed naturally until the continuous phase was predominantly water and the dispersed phase was carbon tetrachloride. Apparently this phenomenon is related to the continually changing holdup discussed above.

Two explanations are offered for the apparent phase reversal phenomenon. At flooding the dispersed and continuous phases are present in the column in approximately equal proportions. It would seem that either phase could be dispersed and for the proper conditions an interchange between the dispersed and continuous phase could occur. It is apparent that in order to establish balanced conditions at flooding, the phase which preferentially wets the packing forces the nonwetting phase from the packing. The situation for maximum stability, then, is one in which the dispersed phase is the nonwetting phase.

Another explanation of these phenomena is possible. One can logically assume that in randomly packed columns it is improbable that uniform packing densities occur throughout. Apparently, there is a cross-section for which the bulk density of the packing material exceeds the bulk density in all other sections of the column. Maximum flow rates or flooding velocities are attained in this section before any other section.

The liquids, then, pile up on either side of the maximum packing density

(minimum voids) cross-section. Above the minimum voids cross-section the

heavy phase accumulates, and below the light phase builds up.

The probability of having a very small voids cross-section increases

with the column length. In the case of Raschig rings an orientation is

possible which would completely block the flows. Yet one can calculate

only an average voids fraction for the column. This situation casts

some doubt on the general reliability of empirical correlations based

on average packing characteristics for the calculation of flooding rates.

Equations of this type have been reported by many [7], [8], [14], [15], [28].

End Section Design. Dell and Pratt [15] reported that inlet tube

placements were important for attaining high flooding rates. Flooding

data have been reported in this research with inlet tubes placed directly

in the packing and outside the packing. No apparent changes in flooding

rates were measured. This is observed in the upper curve of Figure 1. The

data for Series V and IV represents runs for the water inlet tube placed

directly in the packing and outside of the packing respectively.

Blanding and Elgin [7], and Dell and Pratt [15], reported that

expanded end sections were important for attaining high flow capacities.

Before and including Series IV expanded end sections were not used.

They were employed in all experimental runs after Series V. It was not

apparent that the expanded end sections made any difference. Other effects

such as the effect of dirty or conditioned packing, non-uniform packing

densities, non-uniform holdups, phase reversal, and mass transfer were of a
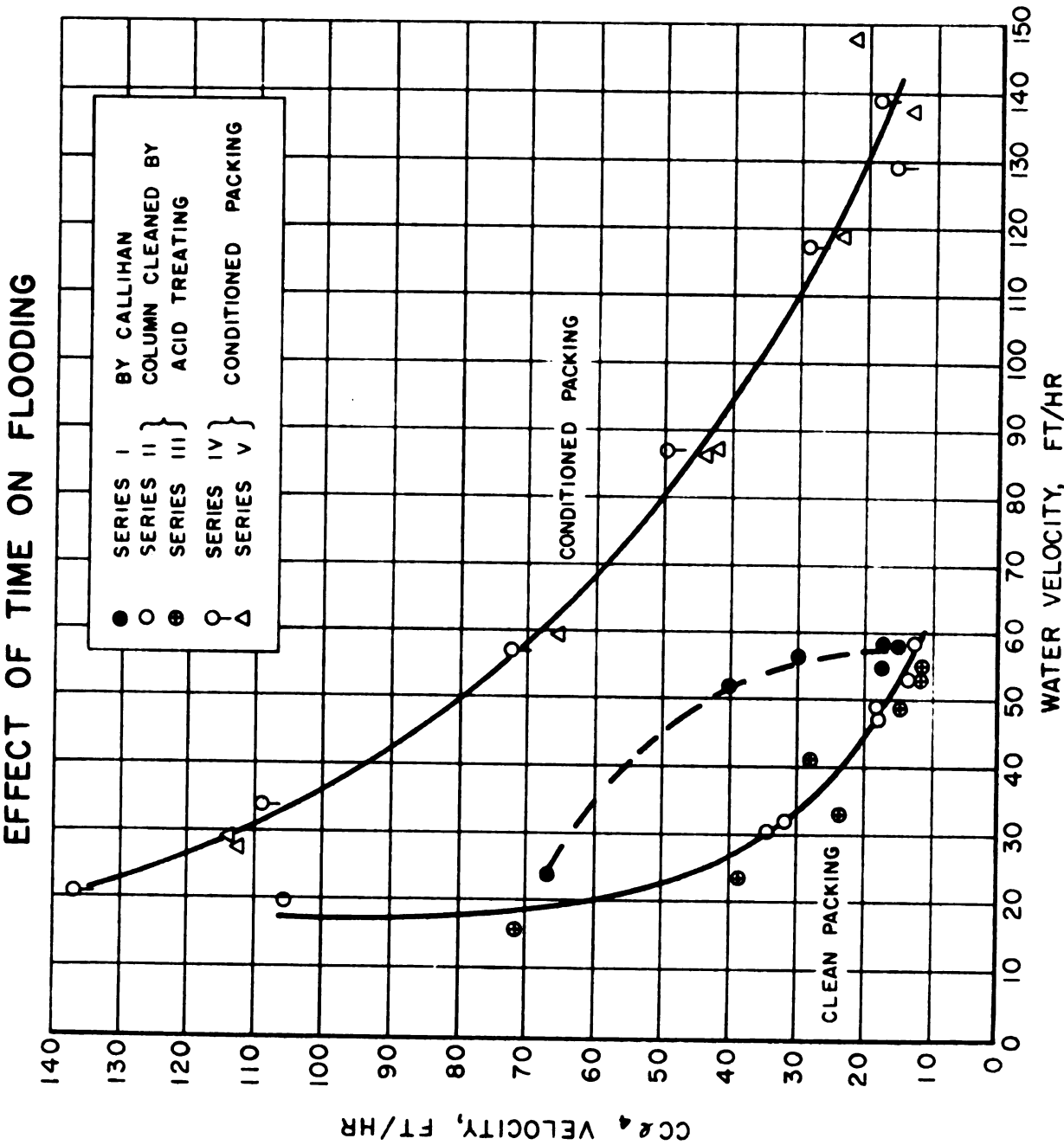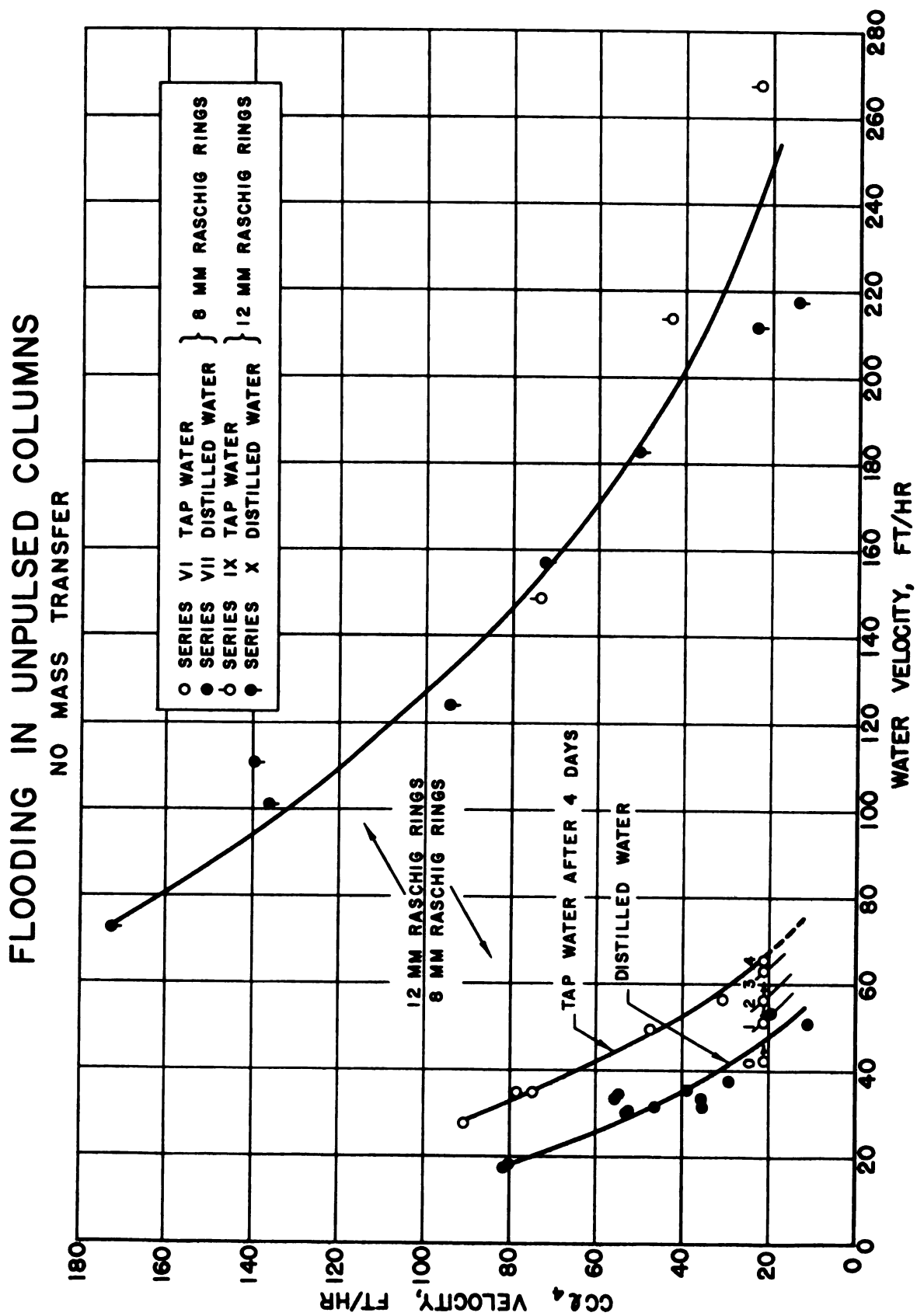
much greater significance.

FIG. 4

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EFFECT OF TIME ON FLOODING

FIG.5

FLOODING IN UNPULSED COLUMNS
NO MASS TRANSFER

Maximum Flow Rates With Mass Transfer

Mass transfer of solute from one phase to the other can cause large increases or decreases in flooding rates. This effect can be so large that it dominates all of the other variables at flooding.

Effect of Mass Transfer on Flooding. Many of the anomalous features of the experimental results can be explained by the effect of mass transfer on drop coalescence. Large drops which have been produced by coalescence give high flooding rates. Small drops result in low flooding rates. That is to say, large drops have a high settling or terminal velocity and consequently are not easily carried over by the continuous phase. The terminal velocities of small drops are low. The continuous phase easily carries small drops along with it.

Specific effects of coalescence were investigated in bench scale experiments. Mass transfer from the dispersed phase aids coalescence. However, mass transfer from the continuous phase inhibits coalescence. When no mass transfer occurs, as in the absence of a solute, or when the solute is distributed between the phases in equilibrium concentrations, coalescence is inhibited. Data supporting the above phenomena are reported in Table VI of the Experimental Results.

The flooding rates in columns can be greatly different depending on whether mass transfer aids or inhibits coalescence. Effects of direction of mass transfer on flooding were also investigated in the packed column. The data from the experimental runs corroborated the results of the coalescence study. They are presented in Table VII.

When mass transfer occurred from the dispersed to the continuous phase high flow rates were observed. For transfer from the continuous to the dispersed phase very low flooding rates were measured. When no solute was present or when the solute was distributed in equilibrium concentrations, moderately low flooding rates were measured.

In packed column operation sharp reductions in maximum flow rates are observed when the CCl_4/H_2O ratios are low, even though acetone is transferred from the dispersed CCl_4 to the water phase. This is not inconsistent with the phenomena of mass transfer and coalescence reported above. At high water flow rates and low CCl_4 flow rates the solute is rapidly washed out of the dispersed phase. Equilibrium may be established or low mass transfer rates may occur in some part of the column. With low mass transfer or equilibrium, coalescence is inhibited and the flow rates must be reduced to prevent flooding. Reduction in maximum flow rates in this research were always accompanied by very low acetone concentration in the exit CCl_4 stream (Table VIII, X).

Tables VIII and X also indicate that high maximum flows are accompanied by high acetone concentrations in the exit CCl_4 phase. That is, when the ratio of CCl_4/H_2O at maximum flows permitted mass transfer to take place throughout the packed section, high flooding rates were attained.

These phenomena of high flooding rates with high ratios of CCl_4/H_2O and reduced flooding rates for low ratios of CCl_4/H_2O are observed in Figures 6, 7, and 8. In these runs, acetone was transferred from the dispersed CCl_4 phase to the continuous water phase.

Comparison of Flooding Rates in Pulsed Columns: With and Without

Mass Transfer. The flooding rates in pulsed columns demonstrate the

phenomena of high throughput rates with mass transfer and sharp reduction

as the transfer rate is reduced. Figure VI shows that at low ratios of

CCl_4/H_2O the flooding rates approach those observed in pulsed columns

with no mass transfer.

It is interesting to observe from Figure VI that the reduction in

maximum flow is sharper for the 8-mm Raschig rings than for the 12-mm

Raschig rings. The ratio of CCl_4 to H_2O at which the reduction in

flooding begins is considerably higher for the 8-mm Raschig rings, i.e.

little water is required to wash out the acetone from the carbon tetra-

chloride. This is explained by the higher efficiency of extraction in

small packing than in the large packing under pulsed conditions. This

is taken up in detail in Section II of this discussion. For now it will

suffice to say that, with pulse, the drops are broken down further in

small size packing than in large size packing.

Comparison of Flooding Rates in Unpulsed Columns: With and Without

Mass Transfer. For 8-mm packing, high flow rates are observed for mass

transfer relative to flow rates without mass transfer. However, differences

in maximum flow rates, with mass transfer and without mass transfer for

12-mm Raschig rings are small. This is observed in Figure 7 by the four

distinct curves. The abnormally high flows for 12-mm Raschig rings when

no solute is present are attributed to the extremely large voids in the

packing. The column-to-packing diameter ratio is only four, leaving large

open spaces at the wall.

Theoretically the number of 8-mm rings and 12-mm rings to produce

the same packing configuration are in the ratio $\left[(12)^3/(8)^3\right] = 3.37$.

However, for this research the ratio is $\left[49.12/.4225\right] / \left[25.91/.9855\right] = 4.42$

(Appendix D). This indicates that the 12-mm Raschig rings are more loosely

packed than the 8-mm rings.

<u>Comparison of Flooding Rates With Mass Transfer: Pulsed and Unpulsed.</u>

For 8-mm and 12-mm Raschig rings the ratio of water to carbon tetrachloride

at which the reduction in flow rates are observed is less for pulsed

operation than for the corresponding unpulsed situation. This is due to

the increased efficiency of pulsed operation.

It is seen by comparing the two curves for the 8-mm Raschig rings

(Figure 8) that the pulsed curve crosses the unpulsed curve at two points.

The upper crossing is due to the higher efficiency of pulsed operation.

At high efficiency the acetone is washed out sooner than in unpulsed

columns. Therefore the reduction in flow rates is experienced sooner in

pulsed columns. The lower intersection is brought about by the ability

of the pulse to act as a pump. The pumping action forces the liquids

through the packing, to give a slightly higher flow rate than in the

unpulsed situation.

The flooding curves for the 12-mm Raschig rings do not intersect

since a smaller efficiency is expected for the large packing than for the

small packing. Thus, the reductions in maximum flows are not as pronounced

for 12-mm Raschig rings as for 8-mm Raschig rings. In the large packing

the drop sizes are not significantly reduced by pulsation since the voids

are large relative to the drops. Also, the pulsation effect analogous

to pumping is a smaller factor in the large voids than in the small voids.

This is observed in Figure 8.

Maximum Flows Calculated from Empirical Correlations

The Hoffing and Lockhart equation is representative of the several

empirical equations for calculating maximum flow rates in packed columns.

Table XXX gives the flooding rates calculated from the Hoffing and Lockhart

equation for the 8-mm and 12-mm Raschig rings.

The flooding rates observed in this thesis fit the Hoffing and Lockhart

correlation at least as well as most of the data from which the correla-

tion was established. The calculated and observed values are tabulated

in Table XXXI and plotted in Figure 7.

Both Tables XXX and XXXI present the flood points as the sum of the

square roots of the superficial velocities, $V_c^{\frac{1}{2}} + V_d^{\frac{1}{2}}$. This expression has

been used frequently to express flooding rates and for many systems is

very nearly constant.

The writer believes that the good agreement between observed

and calculated results in Table XXXI is fortuitous. The flooding rates

can vary over a range of 100% and more as a result of orientation in

randomly packed columns, the formation of bridges between the packing

elements, and the effect of mass transfer on coalescence and drop sizes.

Since the data employed to establish the empirical correlations do not

include mass transfer, additional variations may result between calculated

and observed maximum flows as a result of mass transfer effects.

The positions that the calculated flooding curves take relative to

the observed values for the 8-mm and 12-mm rings, are apparently due to

the large voids in the 12-mm packing and to the large differences in packing

configurations.

TABLE XXX

Flood Point Calculations Based on the Equation Proposed by Hoffing and Lockhart (28)

$$V_{H_2O} = \left[3.33 \times 10^{-5}\, \rho_{CCl_4}^{.22}\, \rho_{H_2O}^{.10}\, \mu_{H_2O}^{.08}\, \mu_{CCl_4}^{.10} \left(\rho_{CCl_4} - \rho_{H_2O} \right)^{.5} \left(\frac{\gamma_{H_2O-CCl_4}}{\gamma_{H_2O-AIR}} \right)^{.5} \left(\frac{a}{\epsilon^{1.2}} \right)^{.67} f(R) \left(\frac{V_{CCl_4}}{V_{H_2O}} \right)^{.25} \right]^{1.25}$$

Physical Properties

Liquid	Density ρ, gr/c.c.	Viscosity μ, cp.
CCl4	1.60	0.975
H2O	1.00	1.00

Interfacial Tension, γ, dynes/cm

γ H2O-CCl4 = 36.3 (13)

γ H2O-Air = 71.0

f(R), ordinate of graph which is related to flow ratio and represents a summary of all pertinent data obtained by various investigators.

Flow Ratio CCl4/H2O	f(R)	8 mm Raschig Rings a, 225.1 ft²/ft³ ϵ, .689, fraction voids			12 mm Raschig Rings a, 115 ft²/ft³ ϵ, .821, fraction voids		
		Flow Rate V, ft/hr CCl4	H2O	$V_c^{\frac{1}{2}} + V_d^{-\frac{1}{2}}$	Flow Rate V, ft/hr CCl4	H2O	$V_c^{\frac{1}{2}} + V_d^{-\frac{1}{2}}$
.05	.1650	7.03	140.5	14.50	14.7	294.0	20.98
.10	.1250	11.80	118.0	14.30	24.7	247.0	20.69
.20	.0900	18.70	93.5	13.97	38.8	194.0	20.16
.50	.0600	35.30	70.6	14.34	73.5	147.0	20.69
1.00	.0400	51.00	51.0	14.28	106.0	106.0	20.58
2.00	.0260	70.20	35.1	14.33	147.6	73.8	20.73
5.00	.0150	111.50	22.3	15.27	232.0	46.4	22.04
10.00	.0092	144.00	14.4	15.79	299.0	29.9	22.61

TABLE XXXI

Calculated and Experimental Flooding Rates in Unpulsed Columns

Sum of the Square Roots of Superficial Velocities

| 8 mm Raschig Rings | | | 12 mm Raschig Rings | | |
Calculated Table XXX	No mass transfer Series VII	Mass transfer Series XII	Calculated Table XXX	No mass transfer Series X	Mass transfer Series XIV
14.5	12.2	15.4	20.98	19.4	21.83
14.3	13.3	14.9	20.7	20.6	21.0
13.97	13.2	14.9	20.2	21.07	20.5
14.3	12.4	14.4	20.7	20.85	21.8
14.3	13.2	14.8	20.6	22.32	21.3
15.3	12.2	14.0	20.7	21.72	20.8
15.8	12.8	14.3	22.0	21.65	21.5
		14.6	22.6		21.6

FIG. 6
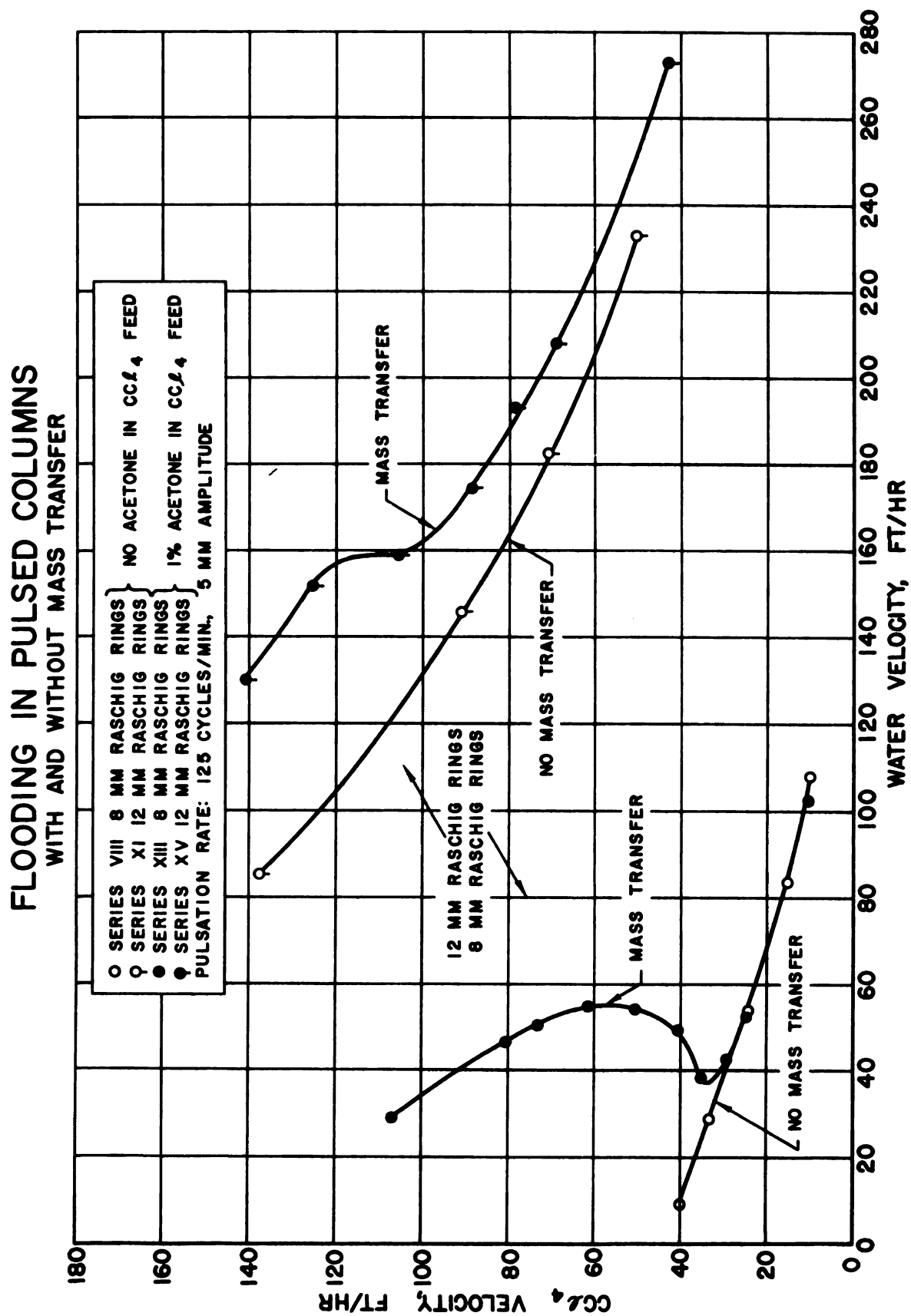
FLOODING IN PULSED COLUMNS
WITH AND WITHOUT MASS TRANSFER

FIG.7

FLOODING IN UNPULSED COLUMNS
WITH AND WITHOUT MASS TRANSFER

FIG. 8

FLOODING IN PULSED AND UNPULSED COLUMNS
WITH MASS TRANSFER

SERIES XII 8 MM RASCHIG RINGS } NO PULSE
SERIES XIV 12 MM RASCHIG RINGS }
SERIES XIII 8 MM RASCHIG RINGS } PULSATION RATE: 125 CYCLES/MIN, 5 MM AMPLITUDE
SERIES XV 12 MM RASCHIG RINGS }
1% ACETONE IN CCℓ₄ FEED

PULSED

NO PULSE

12 MM RASCHIG RINGS
8 MM RASCHIG RINGS

NO PULSE

PULSED

WATER VELOCITY, FT/HR

CCℓ₄ VELOCITY, FT/HR

SECTION II

.

MASS TRANSFER IN PACKED COLUMNS

In this section HETS values for various flows, pulsation rates, packing characteristics and other conditions such as interface positions are discussed. The data have been presented in Tables VIII through XV of the Experimental Results.

Effect of Total Flows on HETS. In pulsed and unpulsed columns HETS values are strongly influenced by the total flows of the two phases. This is observed in Figures 10 through 19.

In pulsed columns HETS values are poorest (i.e., largest) at flooding and they improve (become smaller) with decreasing total flows. Even at high pulsation rates HETS values are not insensitive to total flows. The changes in HETS are at least as large at high pulsation rates as the changes at low pulsation rates for the same percentage changes in total flows. For example, from Figure 13, with water as the continuous phase, HETS is 2.2 feet at flooding. At 50 percent of maximum total flow HETS is 1.67 feet. The change in HETS is 24.1 percent at the high pulsation rate of 125 cycles/min, 25 mm amperes. From Figure 12, with water as the continuous phase, HETS is 3.85 feet at flooding. At 50 percent of the maximum total flow, HETS is 2.92 feet. The change in HETS is 23.2 percent at lower pulsation rate of 125 cycles/min, 10 mm amperes.

Also the largest relative reduction in HETS occurs when water is the continuous phase, and the least when CCl_4 is continuous. These results are in contrast to observations reported for sieve plate columns in which

investigators$^{(11),(12),(54)}$ claim that at high pulsation HETS is insensitive to total flows. Callihan$^{(10)}$ also claims insensitivity of HETS at high pulsation rates to total flows.

In unpulsed columns HETS is not always a maximum at flooding. In Figure 11, with a continuous carbon tetrachloride phase, HETS is a maximum at 80 percent of the flooding rates. This is also observed for the intermediate interface situation. However, for the continuous water phase, the maximum occurs at flooding. This phenomenon, observed in unpulsed columns, is the result of phase reversal at flooding.

Effect of Interface Position on HETS.

Unpulsed Columns. Although the efficiency of mass transfer at reduced flows is less when carbon tetrachloride is the continuous phase than when water is continuous, the HETS and efficiencies are the same at flooding (see Figure 11). This is explained by the fact that CCl_4 is not the continuous phase at flooding. As the total flows approach the maximum flows CCl_4 is displaced by water which preferentially wets the packing. Therefore at the maximum flows the column behaves as if water is the continuous phase.

For the intermediate interface situation water is already the continuous phase in part of the column. Therefore the phase reversal at flooding and consequent reduction in HETS is expected to be small. This is observed in Figure 11.

Earlier in this thesis two explanations were offered for phase reversal. The first stated that at flooding the dispersed and continuous

phase are present in approximately equal proportions in the column. Either

phase could well be continuous. However, the situation that leads to

maximum stability occurs when the dispersed phase is the nonwetting phase.

The second explanation stated that a cross-section of minimum voids

occurs in the column. Flooding always begins from this cross-section.

The heavy CCl_4 phase accumulates above the minimum voids cross-section.

The light water phase builds up below the voids cross-section minimum. In

order to observe the same HETS at flooding, as in Figure 11, the minimum

cross-section would have to be located near the top of the packing. If

the minimum voids cross-section could be removed and the voids cross-

section be made the same throughout the column, the curves in Figure 11

would still intersect in a common flood point. The location of the new

flood point would very likely be such that HETS values would always increase

as the flows are increased to the flooding rates.

Pulsed Columns. Pulsation in packed columns is characterized by

uniform distribution of the dispersed and continuous phases. Regardless

of which phase is continuous, HETS in pulsed columns are expected to in-

crease smoothly with total flows up to flooding. Also HETS values are

expected to be less affected by the choice of the dispersed phase in pulsed

columns. At high pulsation rates HETS values are expected to be insen-

sitive to which phase is continuous. These phenomena are observed in

Figures 12 and 13.

Choice of Dispersed Phase. The dispersed phase should be selected to

give the maximum area of drops for mass transfer. If one phase exceeds

the other in flow rate by an excessive amount, then the phase with the
larger flow rate apparently should be dispersed. This has been observed
in the present study for all runs below flooding, both pulsed and unpulsed.
The ratio of CCl_4/H_2O was maintained at approximately 2. The lowest values
of HETS were observed when CCl_4 was dispersed.

At high pulsation rates, dispersing either phase will give the same
efficiency (Figure 13).

At flow ratios of approximately unity and flows well below maximum,
i.e. when flooding is not a factor, the phase receiving the solute should
be dispersed. This has been recommended by several investigators[19],[22],
[30],[58]. In Table VI of the Experimental Section it is observed that
small drops are formed when mass transfer occurs from the continuous to
the dispersed phase. Small drops give high transfer areas.

However, when the maximum flow rates are a factor in the column design
it may be advantageous to disperse the phase which transfers the solute.
This will result in increased maximum flow since large drops are formed.
The large drops are formed from coalescence of small drops in the presence
of mass transfer.

An additional advantage may be gained by this. The large drops flow-
ing in the packing tend to break down into smaller drops and to coalesce
rapidly. The repeated coalescence exposes fresh surfaces for mass transfer
and increases the turbulence within the drop. These factors directly
improve the mass transfer rate coefficient of the drop phase.

Effect of Flow Ratio on HETS. Flow ratio does not influence HETS

values at flooding. This was reported by Callihan[10] and observed in the present research also. Figure 9 presents HETS as a function of flow ratio for pulsed and unpulsed operation. The curves in Figure 9 indicate a constant HETS for flow ratios above 0.5. The low values of HETS are apparently due to a reduction in maximum total flow. The reduction in flow rates is the result of small drops that form when the mass transfer rate is reduced in some section of the column.

Small errors in the analysis of acetone concentrations in low concentration exit streams introduce large errors in HETS. It is believed that the spread of values at the low ratios of CCl_4/H_2O is due to the very low concentrations of acetone, which had to be analyzed. This is observed in Tables VIII and X of the Experimental Results.

Effect of Column Diameter on HETS. In pulsed columns HETS is not influenced by column diameter, i.e. the "wall effect" appears to have a negligible influence on HETS. Pulsing of the liquids distributes the continuous and dispersed phase uniformly in the column, thus reducing considerably the tendency towards channeling. This is observed in Figure 10. HETS is plotted as a function of total flow rates for two different diameter columns. The packing in each case is 8 mm Raschig rings.

Effect of Packing Characteristics on HETS.

Unpulsed Columns. In unpulsed columns the efficiency of transfer below flooding is not greatly different for the two types of packing employed in this research. The total flow rates have a greater influence on HETS than the packing characteristics. This is observed in Figure 19.

At flooding, the high values of HETS for the 12-mm packing and the low HETS values in 8-mm packing are directly due to the total flows. The flow capacity of the 12-mm packed column is considerably higher than that of the 8-mm column. It has been observed throughout this discussion that high values of total flow are accompanied by high values of HETS and vice versa.

The values of HETS at flooding for the 8-mm and 12-mm packing are observed in Figure 9. The flow capacities with mass transfer are observed in Figure 8 of Section I.

Pulsed Columns. At and below flooding, efficiency of mass transfer is increased sharply in 8-mm packings with pulsation (Figures 9, 18, 19). The increase in efficiency for 12-mm packing, however, is not as great for the same pulsation rates (Figures 9, 18, 19). For the 8-mm Raschig rings, this is due to the sharp decrease in drop diameters and increased holdup of the dispersed phase. Also, some improvement can be expected as a result of increased turbulence during pulsation. For the 12-mm packing, changes in drop diameter and holdup are not as pronounced because of the large voids encountered in 12-mm packing relative to 8-mm packing.

The large discrepancies are apparently due to the effect of packing characteristics on the mechanism of flow, and the mechanism by which small drops are formed in pulsed operations. In small size packing, the drop size is influenced not only by the physical properties of the liquids but also by the size of openings or voids in the packed column. In contrast to this, the drop size is not influenced greatly by large size packings. The voids are already larger than the average equilibrium drop size. In

small size packing the drops apparently are slowed up considerably by the packing and must distort to pass through the voids. During pulsation, the drops in the small packing are broken down by collisions. In the large packing the drops simply pass through the large voids as they normally would when no pulsation is imposed on the system. Therefore, the increase in transfer area is considerably greater in small size packing than in large size packing.

HETS for Inlet Tubes. The simple inlet tubes employed in all the runs of this research did not contribute to HETS. This is apparently due to the single opening in the tubes and the placement of the tubes directly into the packing. This is observed in Table XV. The number of transfer stages, NTP, in the inlet tubes are very small for both pulsed and unpulsed operation.

This is not in agreement with those investigators who claim that significant transfer occurs during drop formation at the inlets to the extraction zones of spray columns[16],[31]. Their results are apparently for shower type inlets and special weirs. Also, inlet tube effects on mass transfer are expected to be more significant in spray columns than in packed columns. Spray columns are not as efficient as packed columns. Therefore, it can be said for packed columns that unless elaborate distributing weirs and special inlets are employed, the transfer at the inlets is very small.

FIG.9   135

HETS VS FLOW RATIO AT FLOODING
IN PULSED AND UNPULSED COLUMNS

FIG. 10

136
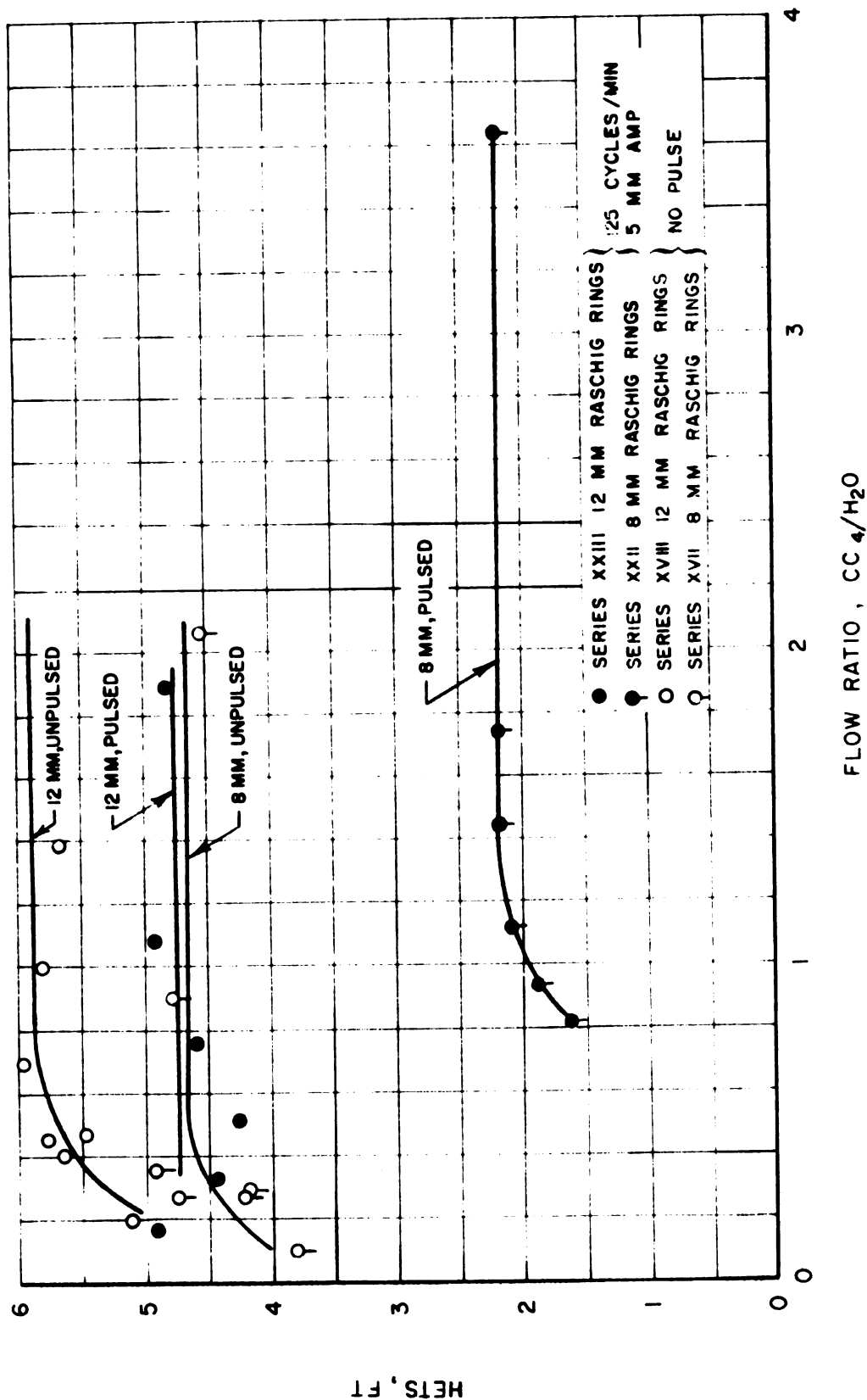
HETS VS SUPERFICIAL VELOCITY IN TWO COLUMNS
OF DIFFERENT DIAMETERS

1% ACETONE IN CCl4 FEED

FLOW RATIO: CCl4/H2O ≈ 2
PULSATION RATE: 125 CYCLES/MIN,
5 MM AMPLITUDE

SERIES XXIV
○ 2.127 INCHES DIAMETER } CALLIHAN (8)
◓ 3.32 INCHES DIAMETER
● 3.32 INCHES DIAMETER

TOTAL SUPERFICIAL VELOCITY, FT/HR

HETS, INCHES

FIG. II

137
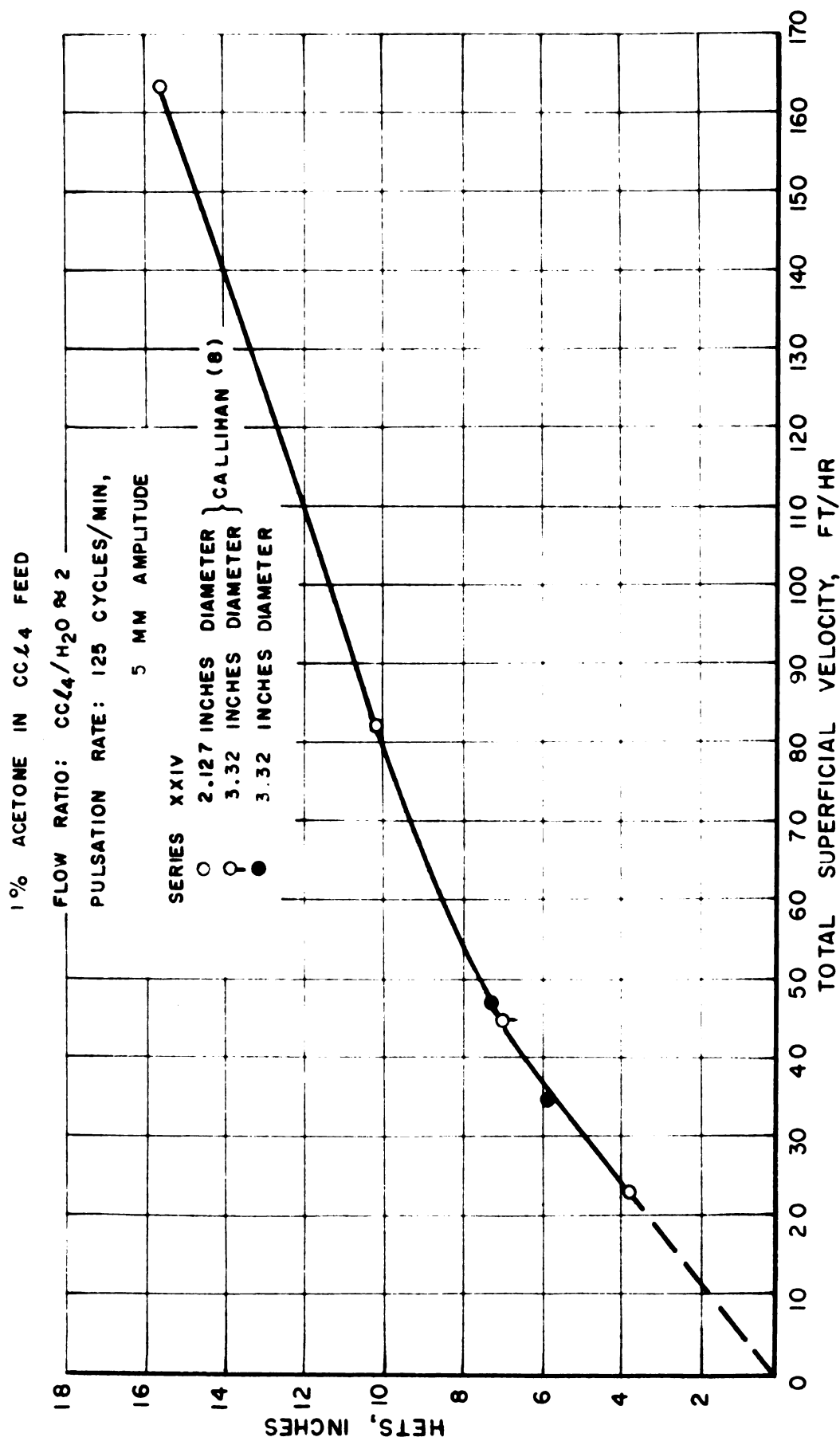
HETS VS SUPERFICIAL VELOCITY FOR DIFFERENT
INTERFACE POSITIONS IN UNPULSED COLUMN

FLOW RATIO: $CCl_4/H_2O \approx 2$

12 MM RASCHIG RINGS

SERIES XXI CCl_4 CONTINUOUS PHASE

SERIES XX   INTERMEDIATE INTERFACE

— — — H_2O CONTINUOUS PHASE,

CALCULATED FROM XXI, XX
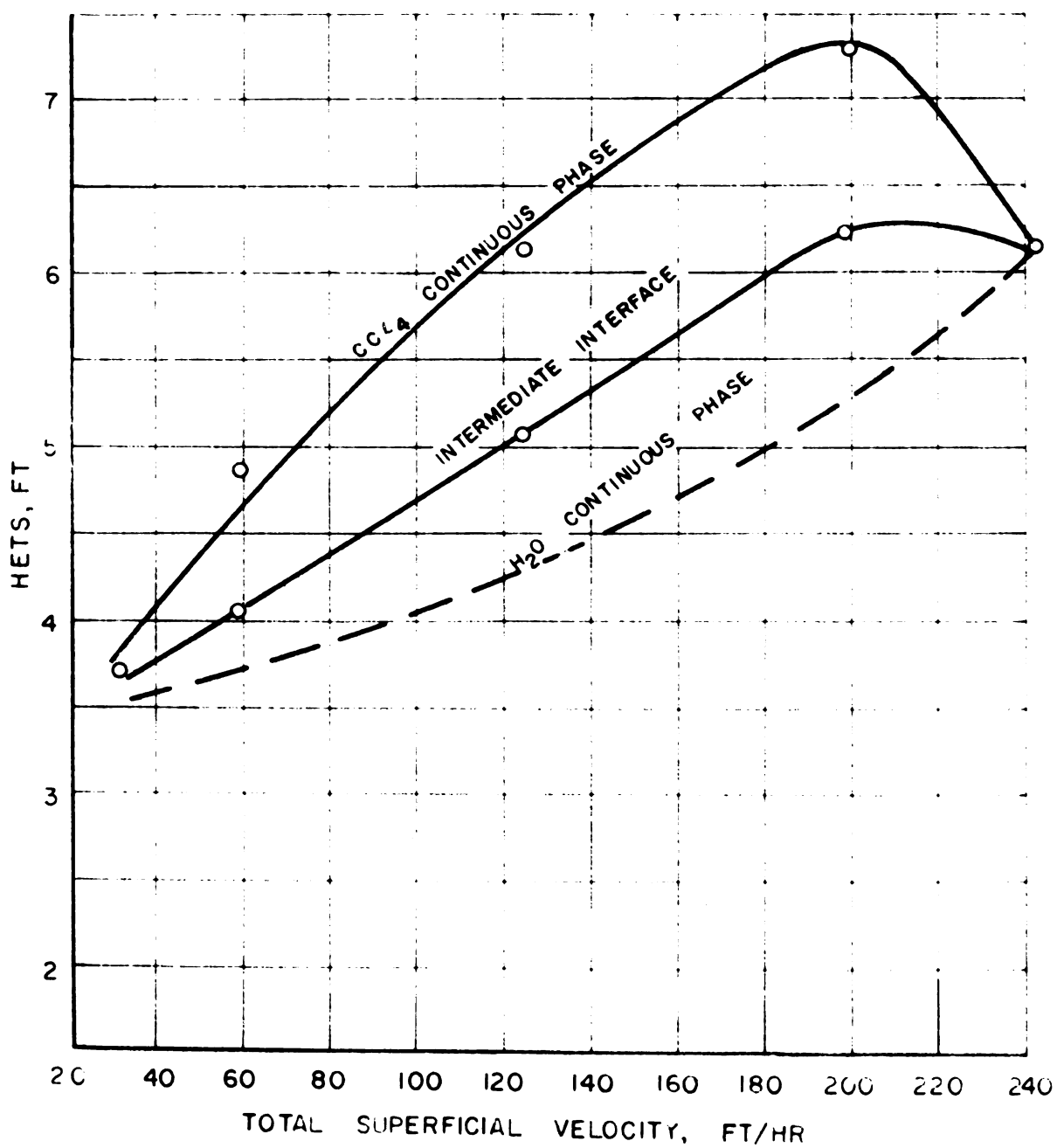
FIG.12   138

HETS VS SUPERFICIAL VELOCITY WITH DIFFERENT INTERFACE
POSITIONS AND CONSTANT PULSATION RATE

PULSATION: 125 CYCLES / MIN, 10 MM AMPLITUDE
FLOW RATIO: $CCl_4 / H_2O \approx 2$
12 MM RASCHIG RINGS

SERIES XXVII INTERMEDIATE INTERFACE
SERIES XXVIII H_2O CONTINUOUS PHASE
SERIES XXIX CCl_4 CONTINUOUS PHASE
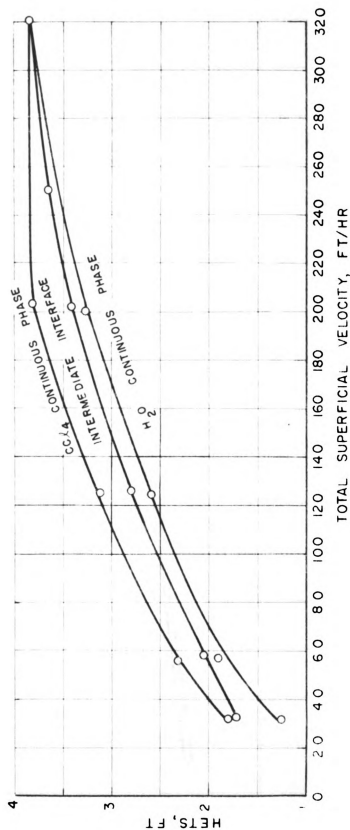
FIG.13

HETS VS SUPERFICIAL VELOCITY WITH DIFFERENT INTERFACE POSITIONS AND CONSTANT PULSATION RATE

PULSATION: 125 CYCLES/MIN, 25 MM AMPLITUDE

FLOW RATIO: $CCl_4/H_2O \approx 2$

12 MM RASCHIG RINGS

SERIES XXX INTERMEDIATE INTERFACE

SERIES XXXI H_2O CONTINUOUS PHASE
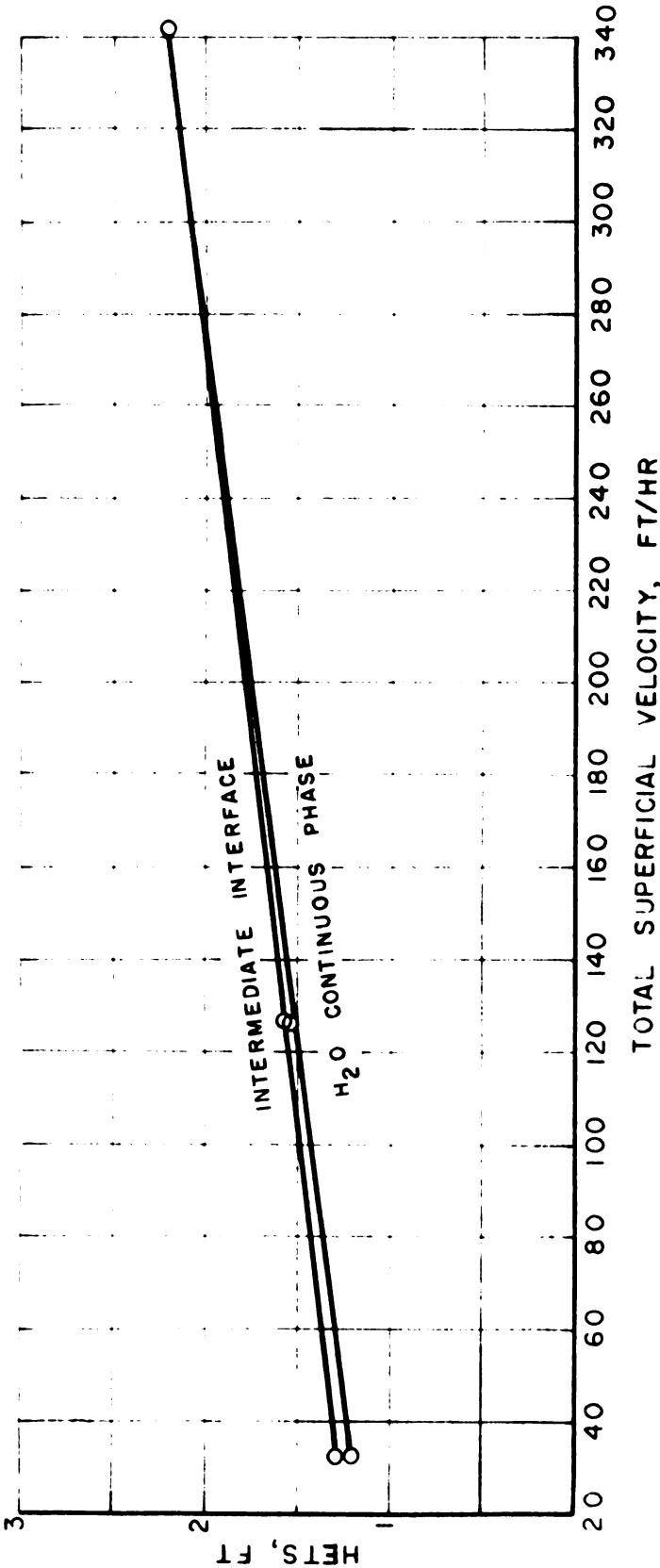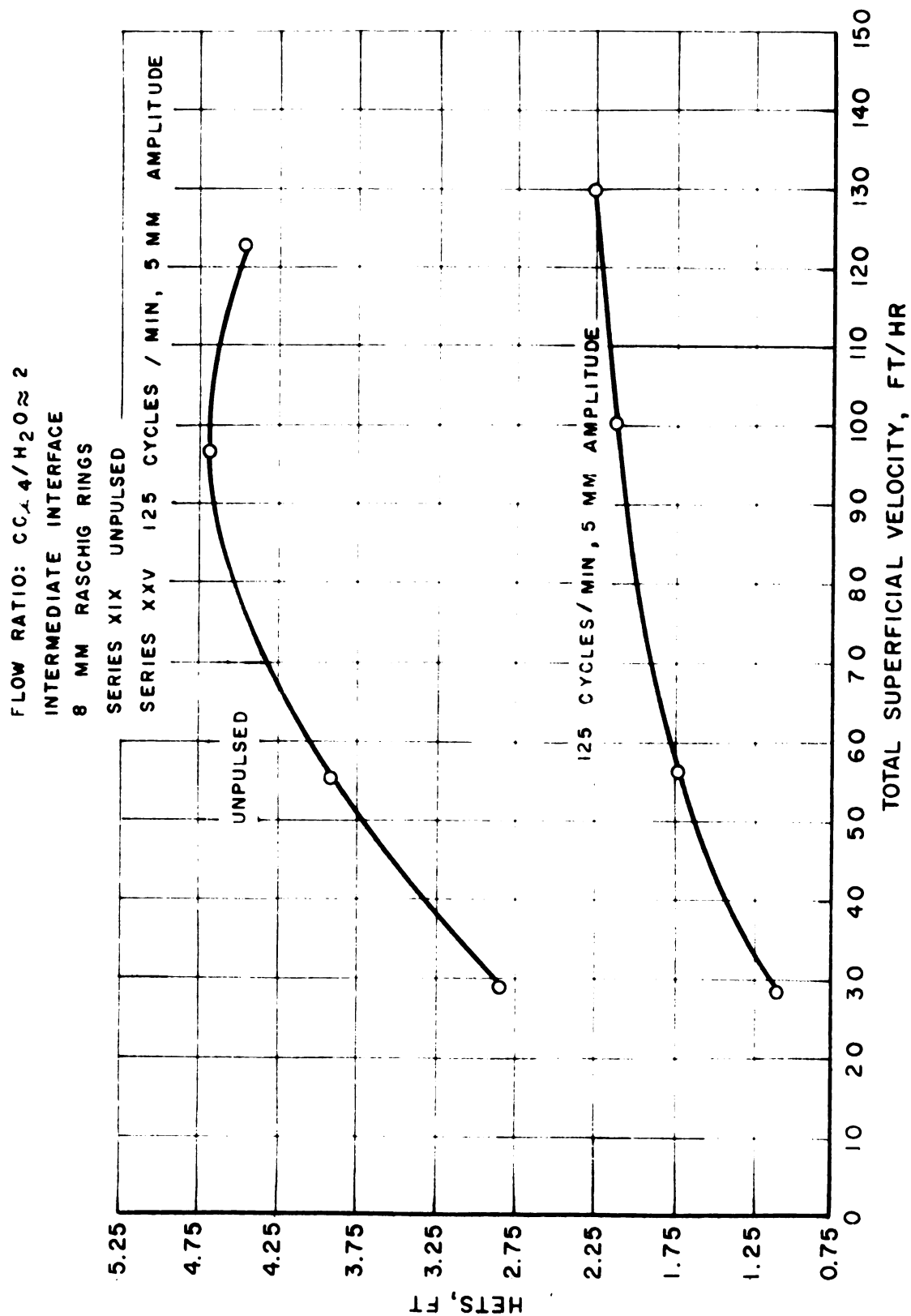
FIG.14

140

HETS VS SUPERFICIAL VELOCITY AT DIFFERENT PULSATION RATES

FIG.15

141

HETS VS SUPERFICIAL VELOCITY AT DIFFERENT PULSATION RATES

FLOW RATIO: $CCl_4/H_2O \approx 2$
CONTINUOUS PHASE: WATER
12 MM RASCHIG RINGS

SERIES XXVII 125 CYCLES/MIN, 10 MM AMPLITUDE
SERIES XXXI 125 CYCLES/MIN, 25 MM AMPLITUDE

FIG.16   142

HETS VS SUPERFICIAL VELOCITY AT DIFFERENT PULSATION RATES

FLOW RATIO: CCl₄/H₂O ≈ 2
INTERMEDIATE INTERFACE
12 MM RASCHIG RINGS

SERIES XX	UNPULSED	
SERIES XXVI	125 CPM,	5 MM
SERIES XXVII	125 CPM,	10 MM
SERIES XXX	125 CPM,	25 MM

UNPULSED

125 CYCLES / MIN, 5 MM AMPLITUDE

125 CYCLES / MIN, 10 MM AMPLITUDE

125 CYCLES / MIN, 25 MM AMPLITUDE

TOTAL SUPERFICIAL VELOCITY, FT/HR

HETS, FT

FIG.17

HETS VS SUPERFICIAL VELOCITY AT DIFFERENT PULSATION RATES

FLOW RATIO: $CCl_4/H_2O \approx 2$
CONTINUOUS PHASE: CCl_4
12 MM RASCHIG RINGS

SERIES XXI   UNPULSED
SERIES XXIX PULSATION RATE: 125 CPM, 10 MM

125 CYCLES/MIN, 10 MM AMPLITUDE

UNPULSED

TOTAL SUPERFICIAL VELOCITY, FT/HR

HETS, FT

FIG.18

144

HETS VS SUPERFICIAL VELOCITY IN PULSED COLUMNS FOR TWO
PACKING CHARACTERISTICS

PULSATION: 125 CYCLES/MIN, 5 MM AMPLITUDE

INTERMEDIATE INTERFACE

SERIES XXV   8 MM RASCHIG RINGS

SERIES XXVI 12 MM RASCHIG RINGS

FIG.19   145

HETS VS SUPERFICIAL VELOCITY IN UNPULSED COLUMNS FOR TWO
PACKING CHARACTERISTICS

INTERMEDIATE INTERFACE
SERIES XIX 8 MM RASCHIG RINGS
SERIES XX 12 MM RASCHIG RINGS

12 MM

8 MM

HETS, FT

TOTAL SUPERFICIAL VELOCITY, FT/HR
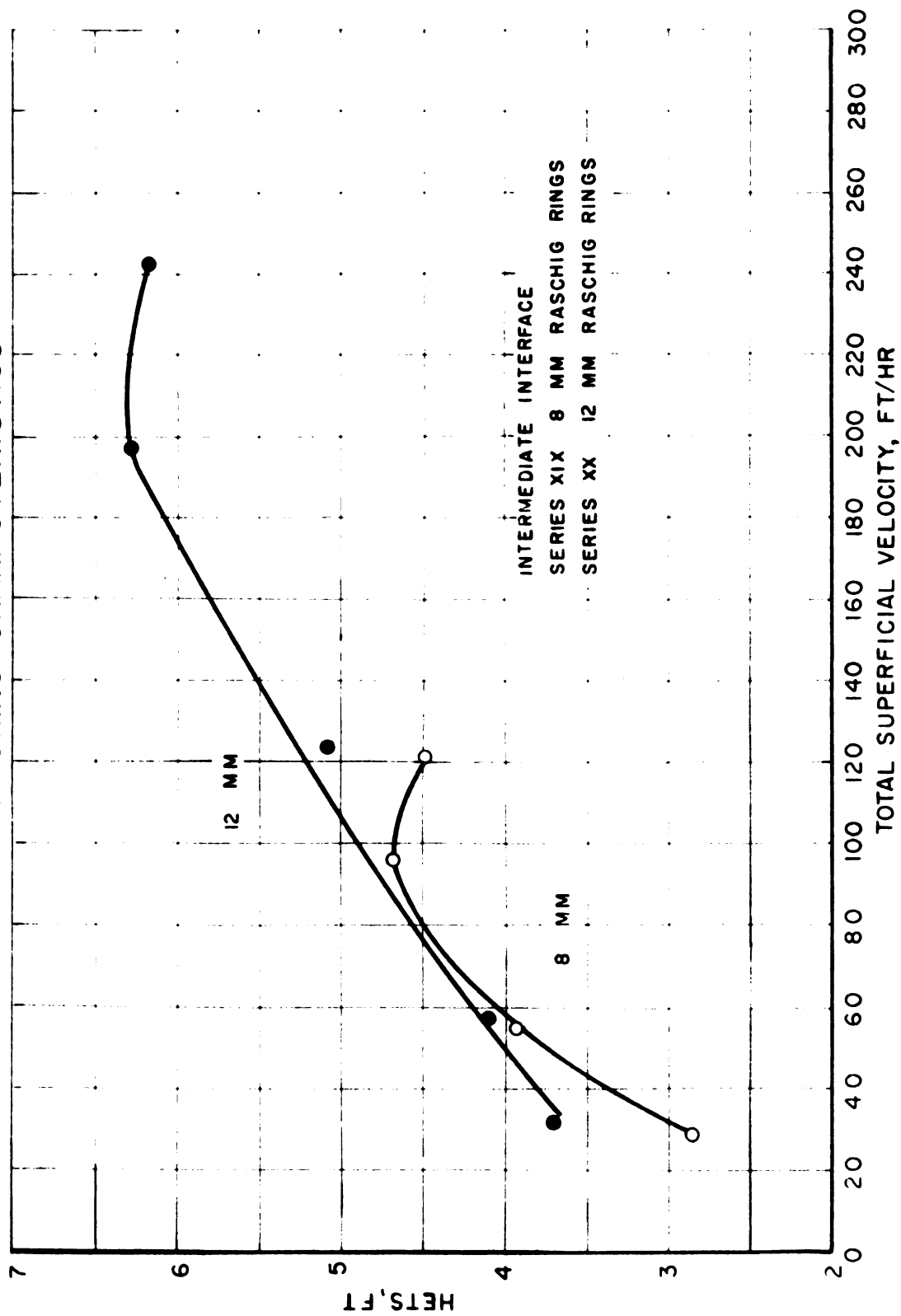
SECTION III

CALCULATIONS OF MASS TRANSFER IN PACKED COLUMS

In this section a brief discussion of the calculations of mass

transfer coefficients from fundamental and semitheoretical considerations

is presented.

The over-all mass transfer coefficient, $K_G a$, calculated by the

method outlined in Theory and Calculations of this dissertation, is in

good agreement with the observed or experimental values. The agreement

is good not only for the data collected by this investigator but also for

previously reported extraction data in packed columns. The calculations

and experimental values of $K_G a$ have been tabulated in Section III of

Experimental Results.

It is difficult to pass judgement on the general reliability of

each of the intermediate results in the analysis since many are not

subject to measurement without elaborate and extended research, For

example, drop diameters, drop velocities, holdup, effective mass

transfer areas, average distance of drop fall between collisions, and

velocities of each phase relative to the interface are some of the

intermediate results which were not measured directly. However, the

analysis which makes use of these intermediate results does indicate

the merits of the over-all approach. The agreement between observed

and calculated results is considerably better than was originally

expected.

Calculation of $K_G a$. It is seen, from Tables XVI to XXV, XXVII

and XXVIII, that the analysis proposed in the Theory and Calculations

section has considerable value in predicting $K_G a$. The method for calculating values of $K_G a$ can be considered as two separate calculations. First, the mass transfer area, a, can be determined if the droplet diameters, d_{vs}, and the disperse phase holdup, x, are known. Second, the mass transfer coefficient, K_G can be determined from k_L, k_G and D, i.e. $1/K_G = D/k_L + 1/k_G$. The resumé which follows is not to be construed as a recipe for arriving at final answers but rather a recapitulation of the general approach. This approach could well be extended to other types of extractors.

Evaluation of Droplet Velocities, v_o. The velocity of droplets relative to the continuous phase depends on droplet diameters, difference in density of the two phases, viscosities, and distance of fall. Theoretical calculations can be made provided that the drop diameters are known. In packed beds the droplet velocity is some fraction of the Stokes' Law terminal velocity. The exact relation used in the calculations is given in Appendix B.

Evaluation of Distance of Free Fall. The drops fall an average distance which is a fraction of the height of the voids in the packed bed. In this work the distance recommended by Pratt and co-workers is used; namely $s = 0.38 d_p - 0.92(\gamma/\Delta\rho g)^{\frac{1}{2}}$. This imposes the limitation that the packing size must be at least equal to or greater than $2.42 \ (\gamma/\Delta\rho g)^{\frac{1}{2}}$.

Evaluation of Drop Diameters, d_{vs}. The drop diameters are determined by the drag forces that break down the drops and surface forces that hold the drops together. A drop diameter is calculated from the

balance of the forces acting on the drop. The drop diameter resulting from the balance of drag and surface tension forces is corrected for the effect of the liquid velocities to give the diameter of the drop in the packed column, $d_{vs}=0.92(\gamma/\Delta\rho g)^{\frac{1}{2}} (v_o/v)$.

Evaluation of Holdup, x. Below flooding, the holdup x is a function of the flow ratios and fluid velocities. It is determined from the expression, $V_d/\epsilon x + V_c/\epsilon (1-x)=v_o(1-x)$.

At flooding the holdup is taken as 0.5 when the flow ratios of the phases are within normal operating limits, i.e. $0.5 \leq V_d/V_c \leq 1.5$. Also at flooding the holdup can be estimated from the relationship $x=(R-\sqrt{R})/(R-1)$.

Evaluation of k_G and k_L. These mass transfer coefficients are evaluated from mass transfer factors, J, explained in terms of the Schmidt number, $Sc=D_f/\mu\rho$, and Reynolds number, $Re=D_p u_r \rho/\mu$. For the Ergun correlation used in this thesis the mass transfer factor, J, is $\left[6(Sc)k/u_r\right]_{c,d}$ and the Reynolds number, $Re_{c,d}$ is $6u_{rc}\rho_c\epsilon(1-x)/(a_p+a_d)\mu_c$ or $6u_{rd}\rho_d\epsilon x/a_d\mu_d$, depending on the phase considered.

Evaluation of $u_{rc,rd}$. The velocities, u_{rc} and u_{rd} are the effective velocities of the continuous and dispersed phases in the packing relative to the interface. When the viscosity of the two phases are equal $u_{rc}=u_{rd}=u_r$. When the viscosities of the phases are not equal the velocities u_{rc} and u_{rd} assume different values. The more viscous phase attains a velocity relative to the interface which is less than the velocity attained by the less viscous phase. For this thesis an inverse relationship is assumed between the velocities and viscosities.

Comparison of Characteristic Drop Diameters and Drop Diameters in
the Packing. According to the theory the smaller the velocity in the
packing between the dispersed and continuous phase the smaller the drag
forces. The drop sizes that accompany small drag forces are large.
This is observed in all of the tables by comparing drop sizes d_o and
$d_{v\bullet}$. The former value, d_o, is the characteristic or terminal drop size
that corresponds to the maximum or characteristic velocity, v_o, between
the drop and continuous phase in the packing. The latter, $d_{v\bullet}$, corre-
sponds to drop diameter at the fluid velocity, v, in the packing.

Increased flows are accompanied by increased holdup of the dispersed
phase and consequently smaller values of fluid velocities in the packing.
This is observed in all of the tables in Section III of the Experimental
Results. These results are fortunate because as the holdup is increased
the drop size is increased. This compensating factor apparently cor-
rects somewhat for the normal crowding of droplets at high holdup.
However it may not account for the abnormal variations in coalescence
that result from the direction of mass transfer.

Film Type Flow vs Droplets. For the situation that the dispersed
phase wets the packing, the mechanism of flow could be different than
when the dispersed phase is the nonwetting phase. Film type flow and
droplets can occur simultaneously when the dispersed phase preferentially
wets the packing. Film type flow means that the wetting phase spreads
over the surface of the packing and flow is along the surface of the
packing.

The fraction of the dispersed phase that flows as film and the fraction that flows as droplets are difficult to determine. However it is interesting to compare the limiting cases: i.e., 100 percent droplet flow and 100 percent film type flow. For film type flow the area for transfer is approximately the area of the packing regardless of the flow ratio and holdup. However the effective area for transfer is apparently less because of stagnation zones in the packing. This is observed by comparing the results in Tables XXIII and XXIX.

For drop flow the calculated values of $K_G a$ are smaller than the observed values. For film flow the calculated values are larger than the observed values.

Holdup of Dispersed Phase; A Comparison of Equations at Flooding.

The experimental flooding rates appear to support the fact that the sum of the square roots of the superficial velocities is nearly a constant. This is observed in Table XXXI which presents data from this research and Table XXXII which presents flooding data from Dell and Pratt for the system methyl isobutyl ketone and water.

In Appendix D it is seen that the equation referred to as Case I, i.e. $V_d/\epsilon x + V_c/\epsilon(1-x)=v_o$, leads to a constant $(V_c^{\frac{1}{2}} + V_d^{\frac{1}{2}})$ at flooding. On the other hand Case II, i.e. $V_d/\epsilon x + V_c/\epsilon(1-x)=v_o(1-x)$ does not work. On this basis it is apparent that Case I is closer to the actual results in variation with flow ratios. In packed unpulsed columns the holdup is better approximated by using the results of Case I, namely that, $x=(R-\sqrt{R})/(R-1)$.

TABLE XXXII

Flood Point Calculations Based on the Equation Proposed by Dell and Pratt (13)

$$\left[1 + .835\left(\frac{\rho_d}{\rho_c}\right)^{\frac{1}{4}}\left(\frac{V_d}{V_c}\right)^{\frac{1}{4}}\right] = C\left[\frac{V_c^2\, a_p}{g\epsilon^3}\left(\frac{\rho_c}{\Delta\rho}\right)\gamma^{\frac{1}{4}}\right]^{-\frac{1}{4}}$$

Column: 3 inch diam.
Raschig rings, 10 mm
a_p: 130.5 ft²/ft³
ε : .802
C : .70

Column Characteristics and Liquids (13)

Liquids	Density ρ, lbs/ft³	Interfacial Tension, γ, dynes	Characteristic drop diameter $d_o = .92\left(\frac{\gamma}{\Delta\rho g}\right)^{\frac{1}{2}}$, ft	Characteristic velocity, v_o, ft/hr
Water	62.4			
MIK	50.0	11.4	MIK dispersed phase 7.3 x 10⁻³	501

Calculated

V_d/V_c	$1/(1+V_d/V_c)$	V_c, ft/hr	V_d, ft/hr	$V_c^{\frac{1}{2}}+V_d^{\frac{1}{2}}$
.2	.833	112.5	22.5	15.35
1.0	.500	64.0	64.0	16.00
4.0	.200	30.8	123.2	16.65
9.0	.100	18.05	162.5	17.00
16.0	.0588	11.83	189.5	17.21

Experimental (13)

V_d/V_c	$1/(1+V_d/V_c)$	V_c, ft/hr	V_d, ft/hr	$V_c^{\frac{1}{2}}+V_d^{\frac{1}{2}}$
.17	.855	116.5	19.8	15.25
.37	.730	95.2	35.2	15.70
.915	.522	68.9	63.1	16.25
1.95	.339	48.4	94.5	16.68
4.22	.1915	30.4	128.1	16.83
9.00	.100	18.0	162.0	16.96

On the other hand at flooding for normal flow ratios, i.e. not far from unity, a value of 0.5 for holdup may serve just as well. This is observed in Figure 20 which indicates that the holdup is 0.5 at flow ratios of unity for either phase dispersed. $K_G a$ calculations based on $x=0.5$ and $x=(R-\sqrt{R})/(R-1)$ are presented in Tables XXI and XXII.

Figure 20 also includes flooding curves which indicate the variations in the sum of the square roots of the superficial velocities and variations in holdup for either phase dispersed. The system employed for the calculations is MIK and water. In addition to the equations of Cases I and II, the empirical equation of Dell and Pratt[13] was employed to calculate a flooding curve for comparison with his experimental data for the system MIK-H_2O. As expected, good agreement is observed since the data was used to establish the equation.

In this research it was discussed earlier that regardless of which phase is dispersed below flooding, at flooding the water phase, which preferentially wets the packing, is continuous. In certain situations this involves a phase reversal. If however the nonwetting phase flow rate exceeds the wetting phase flow rate by a large amount, as by a factor of 10 or greater, it appears logical that the former would be the continuous phase. According to Figure 20 for MIK/H_2O equal to 10, if MIK is dispersed the holdup would be near 0.8, an improbable situation. With phase reversal the holdup would be closer to 0.2 a more likely situation. Therefore, it is possible for phase reversal to occur, when the flooding rates are approached, so that the nonwetting phase becomes

TABLE XXXIII

Flood Point Calculations Based on the Equations of Case I and Case II for the System MIK-Water

Case I: $V_d/x + V_c/(1-x) = \epsilon v_o$
Case II: $V_d/x + V_c/(1-x) = \epsilon v_o(1-x)$
Dispersed Phase: MIK; $v_o = 501$ ft/hr, $\epsilon = 0.802$

Case I

V_d/V_c	$1/(1+v_{MIK}/v_{H_2O})$	x	V_c,ft/hr	V_d,ft/hr	$V_c^{\frac{1}{2}}+V_d^{\frac{1}{2}}$
0	1.00	0	402.0	0	20.05
0.16	0.862	.31	204.0	32.5	20.05
0.36	0.735	.375	157.0	56.6	20.05
0.50	0.667	.415	137.0	68.5	20.05
0.90	0.527	.500	106.0	95.4	20.05
1.10	0.476	.500	95.8	105.5	20.05
2.0	0.333	.58	69.6	139.2	20.05
4.0	0.200	.67	44.7	178.8	20.05
9.0	0.100	.75	25.2	226.8	20.05

Case II

V_d/V_c	$1/(1+v_{MIK}/v_{H_2O})$	x	V_c,ft/hr	V_d,ft/hr	$V_c^{\frac{1}{2}}+V_d^{\frac{1}{2}}$
0	1.00	0	402.0	0	20.05
0.1	0.91	.167	186.0	18.45	18.00
0.5	0.667	.282	91.0	44.5	16.22
1.0	0.50	.333	59.5	59.5	15.40
4.0	0.20	.423	20.55	82.4	13.60
10.0	0.091	.462	9.20	92.0	12.63
100.0	0.0099	.500	0.995	99.5	10.07

TABLE XXXIII (contd.)

Dispersed Phase: H_2O; v_o = 486 ft/hr, ϵ = 0.802

Case I

v_d/v_c	$1/(1+v_{MIK}/v_{H_2O})$	x	v_c,ft/hr	v_d,ft/hr	$v_c^{\frac{1}{2}}+v_d^{\frac{1}{2}}$
0	0	0	382.0	0	19.55
0.16	0.138	.31	194.0	30.9	19.55
0.36	0.265	.375	149.2	53.8	19.55
0.50	0.333	.414	130.2	65.1	19.55
0.90	0.474	.50	100.8	90.6	19.55
1.10	0.524	.50	91.1	100.2	19.55
2.0	0.667	.58	66.2	132.3	19.55
4.0	0.80	.67	42.5	170.0	19.55
9.0	0.90	.75	23.95	216.0	19.55
∞	1.00	1.00	0	382.0	19.55

Case II

v_d/v_c	$1/(1+v_{MIK}/v_{H_2O})$	x	v_c,ft/hr	v_d,ft/hr	$v_c^{\frac{1}{2}}+v_d^{\frac{1}{2}}$
0	0	0	382.0	0	19.55
0.1	0.091	.167	176.8	17.53	17.55
0.5	0.333	.282	86.5	42.3	15.82
1.0	0.50	.333	56.6	56.6	15.00
4.0	0.80	.423	19.52	78.3	13.25
10.0	0.91	.462	8.74	87.5	12.32
100.0	0.99	.500	9.46	94.6	9.84

FIG.20   155

FLOODING CURVES FOR MIK-H$_2$O

CASE I: $(V_d/\epsilon x)+V_c/\epsilon(1-x)=v_o$

CASE II: $(V_d/\epsilon x)+V_c/\epsilon(1-x)=v_o(1-x)$

EXPERIMENTAL POINTS
MIK DISPERSED

CALCULATED FROM EQUATION
BY DELL AND PRATT

HOLDUP AT FLOODING
FOR MIK-H$_2$O

CASE I: $x=(R-\sqrt{R})/(R-1)$

CASE II: $x=-3+\sqrt{1+8/R}/(4/R)-4$

$R=V_d/V_c$

$1/(1+v_{MIK}/v_{H_2O})$

$v_c^{1/2}+v_d^{1/2}$

MIK DISPERSED I

H$_2$O DISPERSED I

MIK DISPERSED II

H$_2$O DISPERSED II

MIK DISPERSED I

H$_2$O DISPERSED I

II

II

continuous. Apparently in this research the flow ratios were not far enough away from unity to observe the nonwetting phase as the continuous phase at flooding.

Effect of Interface Position on Calculated K_Ga. It is interesting to observe that the semitheoretical calculations predict the trends of K_Ga values which are consistent with the choice of the dispersed phase. Table XXXIV summarizes some of the calculated and observed K_Ga values for various situations of interface position. It is seen that calculated K_Ga values are highest (HETS smallest), when CCl_4 is dispersed and smallest (HETS highest) when water flows as drops. This is also observed experimentally.

For the intermediate interphase situation it will be recalled from Section II of this discussion that HETS values are intermediate between the values observed for a continuous water phase and continuous CCl_4 phase (see Figure 11). This is also observed in Table XXXIV, when it is allowed that the calculated K_Ga results for the CCl_4 dispersed phase lie somewhere between the values for the droplet flow and film type flow situation.

Calculations for Pulsed Columns. With pulsing, the agreement between calculated and observed values are fair to good. It is difficult to correct for the pulsing action on the hydrodynamics of flow and the resulting drop sizes. The results are tabulated in Tables XXVII and XXVIII of the Experimental Results.

The modification of the calculations (see Section III of Experimental Results) for the pulsing action was made consistent with the observed

TABLE XXXIV

Summary of Calculated K_Ga and Experimental K_Ga Values for CCl_4 and H_2O

(a) Flow ratio: $CCl_4/H_2O \approx 2$
(b) Below flooding, no pulsation
(c) Acetone transferred from CCl_4 to H_2O

K_Ga CCl_4 continuous observed Table XXVI Series XXI	K_Ga CCl_4 continuous calculated film flow Table XXIV	K_Ga CCl_4 continuous calculated discrete drops Table XXIII	K_Ga Intermediate Interface observed Table XXVI Series XX	K_Ga H_2O, continuous calculated Table XXV	K_Ga H_2O, continuous observed Table XXVI	Superficial velocity ft/hr
2.96	18.2	0.975	2.68	1.92	2.75	31.46- 32.43
3.99	17.35	1.745	4.85	3.86	5.55	58.90- 59.61
6.60	15.97	3.61	7.96	7.92	10.05	124.00-124.4
8.17	14.30	5.72	10.40	13.15	12.0	198.00-199.5

phenomena, that the holdup is higher and the drop size is smaller. This can be accomplished in the equations by reducing the characteristic velocity of the drop(by an amount equivalent to $\frac{1}{2}$ the pulse velocity). There is justification for this because it is logical that the maximum fluid velocity of the fluids relative to each other are reduced with increased pulsing. In fact, for extremely high pulsing rates the drops will remain suspended in the continuous phase and the velocity between the phases becomes zero.

Another approach is offered for future consideration. The drop sizes in unpulsed columns are dependent on the acceleration of gravity. In pulsed situations the effective acceleration is increased. If this is considered in the theory smaller drops would be calculated with increased acceleration resulting from increased pulsing. This would lead to smaller maximum fluid velocities such as the characteristic velocity, v_o, and higher holdups.

CONCLUSIONS

The following conclusions have been made as a result of this research:

1. A theory is presented which leads to calculations of over-all mass transfer coefficients, $K_G a$, and HTU that are in good agreement with experimental results. The agreement is good for the results of this research as well as for results reported by other investigators with various extraction systems.

2. Mass transfer from the dispersed to the continuous phase aids coalescence.

3. High throughput rates are obtainable in packed columns when coalescence of the small drops occurs as a result of mass transfer from the dispersed to the continuous phase.

4. In the absence of mass transfer, or when mass transfer occurs from the continuous to the dispersed phase coalescence of small drops is inhibited.

5. A reduction in maximum flow rates in packed columns results when the small drops do not tend toward coalescence, as a result of mass transfer from the continuous to the dispersed phase.

6. Unusual variations in flooding rates occur when the flow ratio of CCl_4/H_2O is low and mass transfer rates are reduced in some section of the column. The flooding curves are

S-shaped. These variations in flooding rates are explained in terms of mass transfer and coalescence phenomena.

7. In pulsed columns with mass transfer from the dispersed to the continuous phase the S-shaped flooding curves are more pronounced than in unpulsed columns. This is due to the increased efficiency of mass transfer with pulsing.

8. In small size packing continuous operation with liquids that contain impurities which precipitate on the packing may lead to increased flooding rates with time. This is explained by bridging of the packing, reducing the randomness of flow.

9. In large size packing the individual packing elements are separated enough so that bridging of the packing elements does not occur.

10. Pulsing of the liquids inhibits bridge formation.

11. Phase reversal sometimes occurs in packed columns. Two explanations have been offered: One is based on preferential wetting of the packing by one of the liquids, the other on a minimum voids cross-section that results from random packing. Phase reversal does not occur with pulsing.

12. The equation of Hoffing and Lockhart, representative of empirical equations that are frequently used to predict flooding rates in packed columns, must be used with caution. These correlations do not consider the effects of mass transfer, dirty packing, or localized minimum voids that occur in random packing.

13. The efficiency of mass transfer (i.e. low HETS) in both pulsed and unpulsed situations is increased with reduced total flows. Even at high pulsing rates, the increase in efficiency as a result of reduced throughputs can be as high as at low pulsing rates. A 50 percent reduction in total flow is accompanied by as much as a 25 percent reduction in HETS for low and high pulsing rates.

14. HETS values are not dependent on flow ratios.

15. In this research the efficiency of transfer is highest when the water phase is the dispersed phase and lowest when the CCl_4 phase is dispersed. Calculations from the theory agree with this also.

16. HETS at high pulsing rates is the same regardless of which phase is dispersed.

17. HETS in pulsed columns is independent of the column diameter, i.e. the wall effect is negligible with pulsing.

18. In unpulsed columns the efficiency of transfer below flooding is not strongly dependent on the packing size. The total flow rates have a greater influence on HETS than the packing characteristics.

19. With pulsing the efficiency of mass transfer increases sharply in small size packing. The increase is not nearly as great in large size packing. This is due to the drops breaking down by collision in the small size packing. In the large packing

the voids are considerably larger and the drops simply pass

through as they normally would in unpulsed situations.

20. Expanded end sections and single inlet tubes had little

influence on flooding rates and the mass transfer efficiencies

in this research.

NOMENCLATURE

A	column cross section ft^2
a_d	surface area of drops, disperse phase, per unit column volume ft^{-1}
a_p	surface area of packing per unit volume of column ft^{-1}
D	distribution coefficient $D = Y/X^* = Y/X = Y/X$
D_f	coefficient of diffusion ft^2/hr
D_p or d_p	nominal packing size ft
d_o	characteristic diameter of drop ft
d_{vs}	mean diameter of drop ft
F_d	drag force lb
G	light phase which flows up the column ft^3/hr
g	gravity ft/hr^2
H	column height above the base ft
HTU_{OL}	over-all height of transfer unit--liquid or heavy phase film ft
HTU_{OG}	over-all height of transfer unit--gas or light phase film ft
HETS	or HTP, height of theoretical plate ft
k_L, k_G	mass transfer coefficient for liquid or heavy phase film, gas or light phase film defined by equation (1), (2) in Appendix A. ft/hr
K_L, K_G	over-all mass transfer coefficients based on liquid or heavy phase film, gas or light phase film defined by equation (3), (4) in Appendix A. ft/hr
L	heavy phase which flows down the column ft^3/hr
NTU	number of transfer units
NTU_{OL}	over-all number of transfer units based on liquid or heavy phase film $\int_{X_1}^{X_2} (X-X^*)^{-1} dX$
NTU_{OG}	over-all number of transfer units based on gas or light phase film $\int_{Y_1}^{Y_2} (Y^*-Y)^{-1} dY$

NTP	number of theoretical plates
R	ratio of superficial velocities of disperse and continuous phase V_d/V_c
$\dfrac{Re}{1-\epsilon}$	abscissa for Ergun Correlation Re_c, Re_d
Re	$(D_p V\rho)/\mu$ Reynolds number
Re_c, Re_d	$\left[6u_{rc}\rho_c\,\epsilon\,(1-x)\right]\Big/\left[(a_p+a_d)\mu_c\right]$, $\left[6u_{rd}\rho_d\,\epsilon x\right]\Big/\left[a_d\mu_d\right]$
s	average distance that a drop falls in a column $s = .38D_p - .92\left(\dfrac{\gamma}{\Delta\rho g}\right)^{1/2}$ for packed column ft
t	time hr
$Sc = \mu/D_f\rho$	Schmidt number
u_{rd}	velocity of continuous phase relative to interface ft/hr
u_{rc}	velocity of disperse phase relative to interface ft/hr
$u_r=u_{rc}=u_{rd}$	when viscosity of both phases are equal
V	phase velocity relative to empty column ft/hr
V_c	continuous phase velocity relative to empty column ft/hr
V_d	disperse phase velocity relative to empty column ft/hr
v_o	characteristic velocity of drops ft/hr
$\bar{v}$	mean droplet velocity relative to packing ft/hr
v	mean droplet velocity relative to continuous phase ft/hr
v_t	terminal settling velocity ft/hr
$V_d/\epsilon x$, $V_c/\epsilon(1-x)$	actual velocity of dispersed, continuous phase relative to packing ft/hr
X	concentration in heavy phase lb-mol/ft^3
Y	concentration in light phase lb-mol/ft^3
X_i, Y_i	concentration at the interface lb-mol/ft^3

X^*, Y^*	concentration in equilibrium with Y, X lb-mol/ft^3
x	volume fraction of voids occupied by disperse phase
$1-x$	volume fraction of voids occupied by continuous phase
ϵ	volume fraction of column occupied by voids
$1-\epsilon$	volume fraction of column occupied by packing
ϵx	volume fraction of column occupied by disperse phase
$\epsilon(1-x)$	volume fraction of column occupied by continuous phase
γ	interfacial tension between phases lb/hr^2
μ	viscosity lb/ft-hr
ρ	density lb/ft^3
$\Delta\rho$	density difference of phases lb/ft^3

Subscripts

c	refers to continuous phase
d	refers to disperse phase
i	refers to interphase
p	refers to packing
$(\ )_1$	subscript 1 refers to bottom of column
$(\ )_2$	subscript 2 refers to top of column

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APPENDIX A

EXPRESSIONS FOR MASS TRANSFER

In this section some of the relationships commonly used to express

tower performance are developed. Consider a tower in which two immiscible

fluids are continuously contacted. The composition at any cross-section

of the tower will be Y and X for the light phase, G, and heavy phase, L,
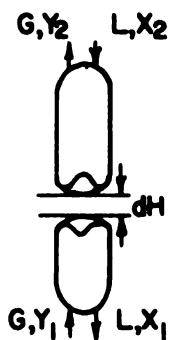
respectively.

Assumptions:

a. The immiscible liquids are separated by an interface bounded

by the heavy phase liquid film and light phase liquid film.

b. The flow rate of each stream, L and G, remains constant

throughout the tower.

c. The distribution coefficient, D, is constant.

d. The driving force is the concentration X and Y in lb-mols/ft^3.

Within a section of tower dH the rate of transfer of solute, lb-mols/hr.

can be expressed by the relationships:

$$L\,dX = G\,dY = k_L a A\,dH(X - X_i) \qquad (1)$$

$$= k_G a A\,dH(Y_i - Y) \qquad (2)$$

$$= K_L a A\,dH(X - X^*) \qquad (3)$$

$$= K_G a A\,dH(Y^* - Y) \qquad (4)$$

The mass transfer coefficients k_L, k_G are defined by equations

(1) and (2); the over-all mass transfer coefficients K_L, K_G are defined

by equations (3) and (4). The combined expressions $k_L a$, $k_G a$, $K_L a$, $K_G a$ are often referred to as mass transfer or over-all mass transfer co-efficients based on unit volume of tower.

Integrating equations (3) and (4):

$$NTU_{OL} = \int_1^2 \left[dX / (X - Y/D) \right] = (K_L a A H)/L \tag{5}$$

$$NTU_{OG} = \int_1^2 \left[dY / (DX - Y) \right] = (K_G a A H)/G \tag{6}$$

Applying $GdY = LdX$ from (1):

$$d(DX - Y) = (DG/L)dY - dY = \left[(DG - L)/L \right] dY$$

$$d(X - Y/D) = dX - (L/DG)dX = \left[(DG - L)/DG \right] dX$$

Substituting for dX and dY in (5) and (6):

$$NTU_{OL} = \left[DG/(DG - L) \right] \int_1^2 \left[d(X - Y/D) \right] / (X - Y/D)$$

$$= \left[DG/(DG - L) \right] \ln \left[(X_2 - X_2^*)/(X_1 - X_1^*) \right] \tag{7}$$

$$NTU_{OG} = \left[L/(DG - L) \right] \int_1^2 \left[d(DX - Y) \right] / (DX - Y)$$

$$= \left[L/(DG - L) \right] \ln \left[(Y_2^* - Y_2)/(Y_1^* - Y_1) \right] \tag{8}$$

Using $D = Y_2/X_2^* = Y_2^*/X_2$:

$$(X_2 - X_2^*)/(X_1 - X_1^*) = (Y_2^* - Y_2)/(Y_1^* - Y_1) \tag{8a}$$

From (5), (6), (7), (8), and definition of HTU=H/NTU:

$$NTU_{OL}/NTU_{OG} = DG/L = HTU_{OG}/HTU_{OL} = (K_L/L)/(K_G/G) \tag{9}$$

$$K_L = DK_G \tag{10}$$

Relation between $\underline{K_G}$, $\underline{k_G}$ and $\underline{k_L}$; $\underline{K_L}$, $\underline{k_G}$ and $\underline{k_L}$:

$$(DG/k_L)dY = aAdH(Y^* - Y_i) \tag{1}$$

$$(G/k_G)dY = aAdH(Y_i - Y) \tag{2}$$

$$(G/K_G)dY = aAdH(Y^* - Y) \tag{4}$$

$$1/K_G = D/k_L + 1/k_G \tag{11}$$

Combining with (10):

$$1/K_L = 1/(Dk_G) + 1/k_L \tag{12}$$

Relation between $\underline{HTU_{OL}}$, $\underline{HTU_L}$ and $\underline{HTU_G}$; $\underline{HTU_{OG}}$, $\underline{HTU_L}$ and $\underline{HTU_G}$:

From (5) and (6):

$$HTU_{OL} = H/NTU_{OL} = L/(K_L aA) \tag{13}$$

$$HTU_{OG} = H/NTU_{OG} = G/(K_G aA) \tag{14}$$

Similarly:

$$HTU_L = H/NTU_L = L/(k_L aA) \tag{15}$$

$$HTU_G = H/NTU_G = G/(k_G aA) \tag{16}$$

Substituting (14), (15) and (16) into (11):

$$HTU_{OG} = HTU_G + HTU_L(DG/L) \tag{17}$$

Substituting (13), (15) and (16) into (12):

$$HTU_{OL} = HTU_L + HTU_G (L/DG) \tag{18}$$

Derivation of Expression for NTP. The theoretical stage concept considers the tower to be subdivided into a number of equilibrium contacts or theoretical plates. Consider the previous column subdivided into N stages numbered from 1 to N. Temporarily the subscripts refer to the plate number.

Material balance around plate 1:

$$G \, Y_0 - L \, X_1 = G \, Y_1 - L \, X_2$$

$$Y_1 = D \, X_1$$

$$Y_2 = D \, X_2 = (DG/L)(Y_1 - Y_0) + D \, X_1$$

$$(Y_2 - Y_1) = (DG/L)(Y_1 - Y_0)$$

Material balance around plate 2:

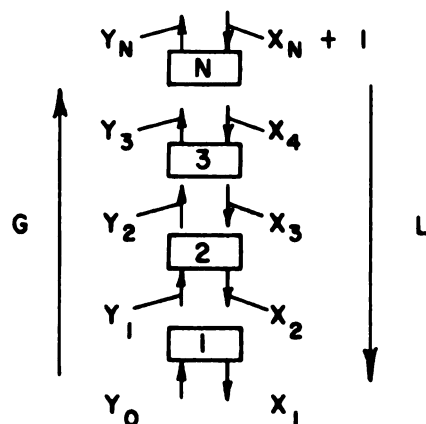$$G \, Y_1 - L \, X_2 = G \, Y_2 - L \, X_3$$

$$Y_2 = D \, X_2$$

$$Y_3 = D \, X_3 = (DG/L)(Y_2 - Y_1) + D \, X_2$$

$$(Y_3 - Y_2) = (DG/L)^2 (Y_1 - Y_0)$$

Material balance around plate N:

$$(Y_{N+1} - Y_N) = (DG/L)^N (Y_1 - Y_0)$$

Returning to original nomenclature where subscripts 1 and 2 refer to bottom and top of the tower, it follows that:

$$Y_{N+1} = D \, X_2 = Y_2^*$$

$$Y_N = Y_2$$

$$Y_1 = D \, X_1 = Y_1^*$$

$$Y_0 = Y_1$$

$$(Y_2^* - Y_2) = (DG/L)^{NTP} (Y_1^* - Y_1)$$

$$NTP \ln(DG/L) = \ln\left[(Y_2^* - Y_2)/(Y_1^* - Y_1)\right] \qquad (19)$$

$$NTP = \left[1/\ln(DG/L)\right] \ln\left[(Y_2^* - Y_2)/(Y_1^* - Y_1)\right] \qquad (20)$$

From (7), (8), and (8a):

$$\left[NTU_{OL} - NTU_{OG}\right] = \left[DG/(DG-L) - L/(DG-L)\right] \ln\left[(Y_2^* - Y_2)/(Y_1^* - Y_1)\right] \qquad (21)$$

Substituting (21) into (20):

$$NTP = \left[NTU_{OL} - NTU_{OG}\right] / \ln(DG/L) \qquad (22)$$

$$(NTU_{OL}/NTU_{OG}) = (DG/L) \qquad (9)$$

$$NTP = \left[NTU_{OL} - NTU_{OG}\right] / \ln(NTU_{OL}/NTU_{OG}) \qquad (23)$$

Substituting (5) and (6) into (23):

$$NTP = \left\{\left[(K_G/G) - (K_L/L)\right] / \left[\ln(K_G L/K_L G)\right]\right\} aAH \qquad (24)$$

In terms of K_L:

$$K_L = D K_G \qquad (10)$$

$$NTP = \left\{\left[(L/GD) - 1\right] / \left[\ln(L/GD)\right](K_L aAH)/L\right. \qquad (25)$$

$$NTU_{OL} = (K_L a A H)/L \qquad (5)$$

$$(NTP/NTU_{OL}) = \left[(L/GD) - 1\right] / \left[\ln(L/GD)\right] = (HTU_{OL}/HTP) \qquad (26)$$

In terms of K_G:

$$K_L = DK_G \qquad (10)$$

Substituting (10) in (24):

$$NTP = \left\{ \left[(1/G) - (D/L) \right] / \left[\ln (L/DG) \right] \right\} K_G a A H \tag{27}$$

$$HTP = H/NTP = \ln(L/GD)/K_G a A \left[(1/G) - (D/L) \right] \tag{28}$$

$$K_G a A = G \ln (L/GD)/(HTP) \left[1 - (GD/L) \right] \tag{29}$$

In terms of end concentrations, X_1, Y_1, X_2, Y_2:

$$NTP = \log \left[(Y_2^* - Y_2) / (Y_1^* - Y_1) \right] \Big/ \left[\log (DG/L) \right] \tag{20}$$

$$Y_2^* = D X_2, \qquad Y_1^* = D X_1$$

Material balance over tower:

$$G Y_1 + L X_2 = G Y_2 + L X_1$$

$$Y_2 = (L/G)(X_2 - X_1) + Y_1$$

$$\therefore (Y_2^* - Y_2)/(Y_1^* - Y_1) = \left[D X_2 - (L/G)(X_2 - X_1) - Y_1 \right] \Big/ (D X_1 - Y_1)$$

$$= \left[X_2 - (Y_1/D) - (L/GD)(X_2 - X_1) \right] \Big/ \left[X_1 - (Y_1/D) \right]$$

$$= \left\{ 1 \Big/ \left[X_1 - (Y_1/D) \right] \right\} \left\{ X_2 - (Y_1/D) - (L/GD) \left[X_2 - (Y_1/D) \right] + (L/GD) \left[X_1 - (Y_1/D) \right] \right\}$$

$$= \left\{ \left[X_2 - (Y_1/D) \right] \left[1 - (L/GD) \right] + (L/GD) \left[X_1 - Y_1/D \right] \right\} \Big/ \left[X_1 - (Y_1/D) \right]$$

$$= \left\{ \left[X_2 - (Y_1/D) \right] \Big/ \left[X_1 - (Y_1/D) \right] \right\} \left\{ \left[1 - (L/GD) \right] + (L/GD) \right\}$$

$$\therefore NTP = \left[1/\log (DG/L) \right] \log \left[\left\{ \left[X_2 - (Y_1/D) \right] \Big/ \left[X_1 - (Y_1/D) \right] \right\} \left[1 - (L/GD) \right] + (L/GD) \right]$$

For the special case where $Y_1 = 0$ (no solute in entering light phase solvent): $NTP = \log \left\{ \left[(X_2/X_1)(1 - L/GD) \right] + (L/GD) \right\} \Big/ \log (DG/L)$

APPENDIX B

CHARACTERISTIC VELOCITY IN TERMS OF TERMINAL VELOCITY

In packed columns the drop falls an average distance s between collisions in the time interval t.

Assumptions:

    a. Voids in the packing are larger than the drops.

    b. Viscous drag force predominates and can be represented by Stokes' law.

At any instant during accelerated motion the drop behavior may be described as follows:

$$\left[(1/6)(\pi d_0^3 \rho_d / g_c) \right] dv/dt = F_G - F_B - F_D \tag{1}$$

$$F_G = \rho_d (1/6)(\pi d_0^3), \text{ gravity force, lbs}$$

$$F_B = \rho_c (1/6)(\pi d_0^3), \text{ buoyant force, lbs}$$

$$F_D = C (v^2/2g_c) \rho (\pi d_0^2/4), \text{ drag force, lbs}$$

For cases where the drag force is entirely frictional and the particle spherical $C = 24 / \left[(d_0 v \rho)/\mu \right]$. (Ref. 42)

$$F_D = (3\pi\mu d_0 v)/g_c = \left[(3\pi\mu d_0)/g_c \right] ds/dt$$

Substituting into equation (1) and expressing the acceleration dv/dt as d^2s/dt^2 one obtains:

$$d^2s/dt^2 = \Delta\rho g/\rho_d - \left[18\mu_c/(d_0^2 \rho_d) \right] ds/dt \tag{2}$$

At terminal velocity $d^2s/dt^2 = 0$:

$$\therefore \ ds/dt = v_t = (\Delta\rho \ g_c d_0^2)/(18\mu_c)$$

Rewriting equation (2) in terms of v_t:

$$d^2s/dt^2 = \left[(\Delta\rho g/\rho_d)\right] - \left[(\Delta\rho g 18\mu_c)/(\Delta\rho g \ d_0^2 \ \rho_d)\right] ds/dt$$

$$= \Delta\rho g/\rho_d - (\Delta\rho g_c/v_t)v$$

$$= (\Delta\rho g/\rho_d)\left[1 - v/v_t\right] = dv/dt$$

$$dv\Big/\left[1-(v/v_t)\right] = \left[(\Delta\rho g)/\rho_d\right] dt$$

$$dv/(v-v_t) = \left[(-\Delta\rho g)/(v_t \rho_d)\right] dt$$

Integrating between limits $v = 0$ when $t = 0$, $v = v$ when $t = t$:

$$\ln\left[(v-v_t)/-v_t\right] = -\left[(\Delta\rho g)/(v_t\rho_d)\right] t$$

$$\ln\left[1-(v/v_t)\right] = \left[-(\Delta\rho g)/(v_t\rho_d)\right] t$$

$$e^{-(\Delta\rho gt)/(v_t\rho_d)} = 1-(v/v_t)$$

$$(v/v_t) = 1- e^{-\Delta\rho gt/(v_t\rho_d)} = (1/v_t) \ ds/dt$$

$$(1/v_t) ds = \left\{ 1-e^{-\left[\Delta\rho g/(v_t\rho_d)\right] t} \right\} dt$$

Integrating between limits $s = 0$ when $t = 0$, $s = s$ when $t = t$:

$$(s/v_t) = t + \left\{ \left[e^{-(\Delta\rho gt)/(v_t\rho_d)} \right] \Big/ \left[(\Delta\rho gt)/(v_t\rho_d)\right] \right\} - 1/\left[(\Delta\rho gt)/(v_t\rho_d)\right]$$

but

$$v_0 = s/t$$

$$(v_0 t/v_t) = t + \left[e^{-(\Delta\rho gt)/(\rho_d v_t)} -1 \right] \Big/ \left[(\Delta\rho_d g)/(v_t\rho_d)\right]$$

$$(v_o/v_t) = 1 - \left[(v_t \rho_d v_o)/(\Delta\rho gs)\right]\left[1 - e^{-(\Delta\rho gs)/(\rho_d v_t v_o)}\right]$$

Pratt and co-workers[25] suggest that for small diameter columns the wall effects are not negligible. They arbitrarily multiply the ratio (v_o/v_t) by $1 - e^{(-7.2\ d_c)}$ where d_c is the column diameter in feet.

APPENDIX C

FLOODING VELOCITY AND HOLDUP IN COLUMNS

CASE I $\quad V_d/x + V_c/(1-x) = K = \epsilon v_0$

CASE II $\quad V_d/x + V_c/(1-x) = K(1-x) = \epsilon v_0(1-x)$

CASE I $\quad$ IF $R = V_d/V_c$

$$R/x + 1/(1-x) = K/V_c = (R - Rx + x)/(x - x^2)$$

$$V_c = K(x - x^2)/(R - Rx + x)$$

$$dV_c/dx = K(R - Rx + x - 2Rx + 2Rx^2 - 2x^2 - x + Rx + x^2 - Rx^2)/(R - Rx + x)^2$$

$$(dV/dx) \text{ at } V_{maximum} = 0 = K\left[R - 2Rx + (R-1)x^2\right]/(R - Rx + x)^2$$

$$x = \left(2R \pm \sqrt{4R^2 - 4R^2 + 4R}\right)/(2R - 2) = (R - \sqrt{R})/(R-1)$$

R	x	V_c/K	V_d/K	$(V_c/K)^{\frac{1}{2}} + (V_d/K)^{\frac{1}{2}}$
0	0	1.0	0	1
0.5	0.414	0.344	0.172	1
1	0.50	0.25	0.25	1
2	0.586	0.171	0.342	1
100	0.91	0.00827	0.827	1
∞	1.0	0	1.00	1

CASE II $\quad R/\left[(1-x)x\right] + 1/(1-x)^2 = K/V_c = (R-Rx+x)/\left[x(1-x)^2\right]$

$$V_c = Kx(1-x)^2/(R-Rx+x)$$

$$dV/dx = K\left[(R-Rx+x)(1-4x+3x^2) - (x)(1-x)^2(1-R)\right]/(R-Rx+x)^2$$

$$0 = \left(R-Rx+x-4Rx+4Rx^2-4x^2+3Rx^2-3Rx^3\right.$$

$$\left. +3x^3-x+2x^2-x^3+Rx-2Rx^2+Rx^3\right)$$

$$= R-4Rx-2x^2+5Rx^2+2x^3-2Rx^3$$

$$= R-4Rx+(5R-2)x^2+(2-2R)x^3$$

$$= (2-2R)x^2+3Rx-R$$

$$x = \left(-3R \pm \sqrt{9R^2-8R^2+8R}\right)/(4-4R)$$

$$= \left(-3R \pm \sqrt{R^2+8R}\right)/(4-4R)$$

$$= \left[-3 + \sqrt{1+(8/R)}\right]/\left[(4/R)-4\right]$$

R	x	V_c/K	V_d/K	$(V_c/K)^{\frac{1}{2}} + (V_d/K)^{\frac{1}{2}}$
0	0	1.0	0	1.00
0.01	0.064	0.762	0.0076	0.96
0.10	0.167	0.464	0.046	0.90
1.0	0.333	0.148	0.148	0.768
4.0	0.423	0.0512	0.2048	0.679
∞	0.50	0	0	0

APPENDIX D

TABLE XXXV

PROPERTIES OF CCl$_4$ AND WATER

(20°C-25°C)

	Density lbs/ft^3	Viscosity, lbs/ft-hr	Interfacial Tension lbs/hr^2
Water	62.40	2.42	
Carbon Tetrachloride	99.84	2.42	1.036 x 10^6

TABLE XXXVI

PACKING CHARACTERISTICS

Raschig Rings	Material	Density lbs/ft^3	Wt/Ring gm	Volume/Ring ml	Area/Ring cm^2	Bulk Density lb/ft^3	Void Fraction	Area ft^2/ft^3
8 mm	Lime Glass	157.87	.4225	.1670	3.964	49.12	.6889	225.1
12 mm	Pyrex	144.77	.9855	.4250	8.975	25.91	.8210	115.2

TABLE XXXVII

DISTRIBUTION OF ACETONE IN WATER AND CARBON TETRACHLORIDE

ACETONE IN WATER	ACETONE IN CCl$_4$	DISTRIBUTION COEFFICIENT
millimols/liter	millimols/liter	
0.0	0.0	0.0
100.0	45.0	2.222
200.0	92.0	2.174
300.0	141.0	2.128
400.0	192.0	2.083
500.0	245.0	2.041
600.0	297.5	2.017
650.0	326.0	1.994

FIG.21

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DISTRIBUTION OF ACETONE IN WATER AND CARBON TETRACHLORIDE

JAN 31 1962

☐ In person