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THE PERFORMANCE OF PULSED PRESSURE WETTED-WALL DISTILLATION COLUMNS

A thesis submitted for the Degree of Doctor of Philosophy in the School of Pure and Applied Science of Loughborough University

by

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August 1970.
ACKNOWLEDGEMENTS

I am sincerely grateful to Mr J Glover for his continuing guidance and suggestions so readily given during the course of this work. I wish to express my special gratitude to Prof. D. C. Freshwater for his valuable encouragement and constant support.

I am also grateful to the Albright and Wilson Group for financing this project. My thanks are also due to my colleagues at the Department of Chemical Engineering, Loughborough University, for providing interesting atmosphere of work and for several stimulating discussions.

I would like very much to thank Mrs. N. Brown for her careful typing of this thesis.

SABAH S. GABBAY
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Abstract

The main purpose of this work was to investigate the characteristics and operational performance of pulsed wetted-wall distillation columns at total reflux and using the binary system methylcyclohexane/toluene.

The results of a detailed study on three concentric-tube, wetted-wall columns are given both with the application of pulsations on the vapour phase and without. Marked improvement in the performance of the columns expressed as number of transfer units (N.T.U.) was obtained by using pulsations in the range of 0-21 cycles/sec, even to the extent of quadrupling the efficiency. It was found that an increase in efficiency was obtained for both increase in amplitude and frequency and that there exists a maximum efficiency at an approximate frequency of 17 cycles per second.

The introduction section contains an account of the various correlations and work on wetted-wall columns, and the individual influence of liquid and gas film transfer coefficients is discussed. The various theories relating to two-phase mass transfer are presented and a general account of the advantages gained by the application of pulsations to other processes is presented with particular reference to the influence of pulsations to gas/liquid systems.
The experimental section gives a description of the apparatus used and the experimental procedure followed for both the pulsed and the non-pulsed conditions. The analysis method, mainly based on refractometric determination of the samples and the method for the determination of the number of transfer units of the columns are described.

The results of the investigation are reported in section 3 and some of the results of the initial runs are included. The effect of the parameters of pulsations (frequency and amplitude) on efficiency are emphasized and the data for the non-pulsed operation are shown for comparison. Possible mechanisms of transfer under pulsations are proposed in the discussion, section 4, and an attempt was made to clarify the existence of a maximum efficiency of the columns at a certain frequency. A correlation was obtained for the two columns (1" and 1½" inner tube), characterizing the main pulsation parameters with the increase in performance obtained, for the system studied.
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SECTION ONE

1.1 Wetted-Wall Columns

Introduction

In a wetted-wall column the liquid flows as a thin film down the inside of a vertical tube with the gas flowing counter-currently. The important advantage of this apparatus is that the interfacial area can be measured accurately.

The rate of mass transfer is obtained from the analysis of the liquid and gas entering and leaving the column. The gas film coefficient $K_G$, is either measured by the rate of absorption of a highly soluble gas in a suitable liquid or by measuring the rate of evaporation of a pure liquid in a gas stream. The latter method has the advantage that the conditions could be adjusted so that there would be no resistance due to diffusion in the liquid. Liquid film coefficients, $K_L$, on the other hand, are found by determining the rate of absorption or desorption of gases of low solubility or pure gases.

The results obtained for gas-film coefficients are found to be consistent, reliable and independent of the column height. The experimental results for liquid film coefficients in this apparatus, however, vary with the length of column and depend on the amount of rippling developed in the liquid film, which has pronounced effects on the liquid film coefficients.\(^{(1,2)}\) The liquid flow
rate also affects \( K_L \), and for these reasons, it has been difficult to relate the liquid film performance of a wetted-wall column to packed towers. Liquid film coefficients are, however, usually obtained using a disc column. The theory of fractional distillation in wetted-wall columns with the vapour and liquid in streamline flow was discussed by Kuhn (3) and Westhaver (4).

Westhaver devised an equation at total reflux, assuming adiabatic operation, laminar flow of vapour and an ideal liquid film - i.e.: negligible surface resistance and a uniform radial composition, as follows:-

\[
H\text{-E.T.P.}, \text{(cm)} = \frac{17}{35} \cdot \frac{V_a W^2}{D} + \frac{D}{V_a} \quad \text{(1.1)}
\]

(for \( 1 < \alpha < 1.1 \))

where:- \( V_a \) = linear vapour velocity, cm/sec.
\( 2W \) = width of annulus, cm.
\( D \) = diffusion coefficient, sq. cm/sec.

A more exact solution of Westhaver's equation for all values of the relative volatility, has been given by Kuang-Chun and Malyusov (5) (6).

It was reported that when the vapour is in streamline flow and the column is of sufficient length to establish uniform conditions the dimensionless group, \( K_g \frac{d}{D} \) is given by:- \( K_g \frac{d}{D} = 3.66 \).

The height of a transfer unit \( H_g \), is given by:-
1.2 Gas and Liquid Film Coefficients - Empirical Correlations

Chilton and Colburn\(^{7}\), used the Prandtl-Taylor modification to Reynolds analogy for mass and momentum transfer in a pipe and derived an equation which related the gas film coefficient with the flow rate and the physical properties of the system.

The equation appears in various forms in literature, the usual versions (corrected for absorption and desorption processes by including the drift factor \(P_{BM}/P\)) are as follows:

\[
\begin{align*}
\frac{K_G RT}{v} \times \frac{P_{BM}}{P} \times (S_c)^{\frac{2}{3}} &= J_D \\
\frac{K_G P_{BM} M_M}{G} \left(\frac{S_c}{S}\right)^{\frac{2}{3}} &= J_D \\
\frac{K_G \Gamma_{BM}}{G_M} \left(\frac{S_c}{S}\right)^{\frac{2}{3}} &= J_D
\end{align*}
\]

where:

- \(M_M\) = the mean molecular weight of the gas.
- \(v\) = the gas velocity, ft/hr.
- \(G\) = the mass velocity \(= \rho v\), lb/(hr.)(ft\(^2\))
- \(G_M\) = the molar velocity \(= G/M_M\), lb. mole/(hr.)(ft\(^2\))
\[ J_D = \text{the mass transfer factor.} \]

The equation applicable for distillation in a wetted-wall column may be written in the following form:

\[ K_G P = 0.034 R_{e_v}^{-0.2} S_c^{0.67} \]

where \( P \) is the total pressure.

Further studies to find the effect of the channel diameter on the mass exchange during rectification in the turbulent flow, resulted in the following:

\[ H_{OG} = 11.1 d^{0.64} (R_{e_v})^{0.23} S_c^{0.67} \] \hspace{1cm} (1.3)

This is recommended for calculations involving the rectification process over a range of Reynolds numbers of 1,000-15,000 and pressure range of 50-760 mm Hg.

Gilliland and Sherwood (8), studied the evaporation of water and several organic liquids into a turbulent air stream, in a wetted-wall column. The mass transfer data was correlated within a spread of \( \pm 15\% \) by the equation:

\[ d = 0.023 R_{e_v}^{0.83} S_c^{0.44} \]

where:

- \( d \) = the tube diameter.
- \( x \) = the effective thickness of the laminar film.

Reynolds numbers ranged from 2,000 to 35,000 and Schmidt numbers from 0.6 to 2.5 and pressures from 0.1 to 3 atmospheres.

They also presented their results in the following form:
\[ \left( \frac{K_G RT}{u} \right) \times \left( \frac{P_{BM}}{p} \right) \times \left( \frac{\mu}{\rho D} \right)^{0.56} = 0.023 \left( \frac{p u d}{\mu} \right)^{-0.17} \] .... (1.4)

or:

\[ \frac{K_G P_{BM}}{G M} = 0.023 \, R_{e_v}^{-0.17} \, S_{c}^{-0.56} \]

where, \( u \) is the gas velocity taken relative to the tube wall.

The equation is similar to the theoretical equation of Chilton and Colburn, for heat and mass transfer, but the predicted coefficients by the theoretical equation is about 30% lower than the measured ones. This may be due partly to the effect of ripples developed in the liquid film travelling down the column, thus causing turbulence and increasing the surface area of the liquid exposed to the gas.

Morris and Jackson (9) studied absorption of hydrogen chloride in water, ammonia in water, and dilute acid and drying of air with sulphuric acid in wetted-wall columns, and gave a general correlation in the form of the following relation:

\[ \left( \frac{K_G RT}{u} \right) \left( \frac{P_{BM}}{p} \right) = 0.04 \, R_{e_v}^{-0.25} \, S_{c}^{-0.5} \]

or

\[ \frac{K_G P_{BM}}{G M} = 0.04 \, R_{e_v}^{-0.25} \, S_{c}^{-0.5} \] .... (1.5)
The best correlation was found when the gas velocity \( u \), in the above equation was taken relative to an effective liquid surface velocity, \( u_l \). For a counter-current flow of gas and liquid, the gas velocity is then given by:

\[
u = u_l + u_g
\]

where \( u_g \) is the actual gas velocity in the tube and \( u_l \) is the effective liquid surface velocity and was found to be about 70% of the calculated surface velocity assuming streamline flow in the liquid film.

i.e. \( u_l = 0.7 \times u_{l\text{ max}} \)

and from theoretical considerations;

\[
u_{l\text{ max}} = \left( \frac{2}{8} \times \frac{\rho g q^2}{\mu_l} \right)^{\frac{1}{3}} = \left( \frac{2}{8} \frac{g \sqrt{2}}{\rho L \mu_L} \right)^{\frac{1}{3}}
\]

where:

\( q \) = the volume of liquid flowing in unit time through a film of unit width.

\( \sqrt{g} = \rho_L q \), is the mass flow rate.

Johnstone and Pigford (10), studied distillation of several mixtures including acetone/water, ethanol/water, benzene/toluene and correlated their results on the basis of gas-film mass transfer coefficient, by the equation:

\[
(H.T.U.)_G = 7.63 \cdot d \left( \frac{\rho u d}{\mu} \right)^{0.23} \left( \frac{\mu}{\sqrt{g \rho D}} \right)^{0.67} \quad \cdots (1.6)
\]

Morris and Jackson claimed his results to be in good agreement with those of Johnstone and Pigford for distillation. Norman (11) found good agreement for distillation
of ethanol/water mixtures in a wetted-wall column and the Chilton-Colburn equation.

Jackson and Ceagleske\textsuperscript{(12)} studied the vapourisation of water, toluene and 2-propanol, and of water from aqueous glycerol solution and also the distillation of 2-propanol/water mixture in a wetted-wall column. They found that their results were in good agreement with the Chilton-Colburn equation, taking the gas velocity as its velocity relative to the liquid surface. They furthered the investigation by taking liquid samples from the column wall at several different heights in the column and reported that point values of the gas film transfer coefficients varied with the composition in a manner not predicted by the Chilton-Colburn equation.

Chari and Storrow\textsuperscript{(13)} studied the distillation of methanol/water, ethanol/water, and acetone/water in a wetted-wall column. By measuring the vapour temperature at several points along the column, they estimated the vapour composition and found that $H_{OG}$, varied with concentration of alcohol in a manner similar to the change in $m$, the slope of the equilibrium curve. They also attempted to separate the gas and liquid resistances and gave the following relationship for $H_{OG}$ at any particular composition:

$$H_{OG} = \alpha \left( \frac{R_{ev}}{R_{L}} \right)^{0.17} + mB \left( \frac{4}{\mu_L} \right) \frac{\mu}{\mu} \cdots (1.7)$$

where $\alpha$ and $\beta$ were constants which included the vapour and liquid Schmidt numbers. The height of a gas phase
transfer unit (\(H_G\)) over the range of alcohol concentration used (about 20-80 mole %) could be represented by the following two equations:

\[
\frac{H_G}{d} = 11.4 \left( R_{ev} \right)^{0.17} \left( S_{cv} \right)^{0.25}
\]

or:

\[
\frac{H_G}{d} = 13.9 \left( R_{ev} \right)^{0.17} \left( S_{cv} \right)^{0.56}
\]

Thus the gas phase resistance in distillation was larger than the Gilliland-Sherwood equation for evaporation of a pure component with a gas stream (unidirectional diffusion). The liquid film resistance was correlated by the equation:

\[
H_L = \beta \left( \frac{4 \sqrt{F}}{\mu_L} \right)^{2/3}
\]

where \(\beta\) was an empirical factor taking into account the variations in the physical properties of the liquid over the concentration range under consideration. The exponent of the liquid Reynolds number in the above equation was the same as the modified form of the equation for \(H_L\) given by Surowiec and Furnas (14).

Similar expressions for the liquid-film resistance have been derived for wetted-wall columns on the assumption of streamline flow in the liquid phase and turbulent motion in the vapour phase. A general expression is given as:

\[
H_L = \beta \frac{M}{M_f} \cdot \frac{R^P}{e_L} \cdot \frac{S^Q}{C_L} \cdot G^T_a
\]
where \( p, q, r = \text{constants} \)

\[
M = \text{point value of the molecular weight.}
\]

\[
M_M = \text{mean molecular weight over height } z.
\]

\[
G_n = \text{Galilee number } = \frac{D_L^2}{g\zeta^3}.
\]

Chari and Storrow, concluded that liquid film resistance was a significant part of the overall resistance to mass transfer; however, due to lack of data on the variation of the liquid phase diffusion coefficient with composition, the results could not be correlated by a single equation. Storrow (15) later determined experimentally values for the liquid phase diffusion coefficients at different compositions and analysed data on the rectification of ethanol/water mixtures in a wetted-wall column but again no satisfactory correlations were obtained. His analysis confirmed, however, the conclusions of Chari and Storrow.

Querishi and Smith (16) presented data on the rectification of six binary mixtures in a wetted-wall column. For all these systems, \( H_{OG} \) was found to vary with composition, passing through a minimum about the middle of the concentration range and increasing as the concentration of the more volatile component approached zero or unity. To counteract this variation, it was found useful to multiply \( H_{OG} \) by a function, \( y(1-y) \), so that \( H_{OG} y(1-y) \) may become independent of composition.

Boyarchuk and Planovskii (17) studied the kinetics of mass transfer in rectification apparatus of the film type, in an attempt to:
(a) establish regularities in the change of the coefficients of mass transfer, $K_G$ and $K_L$ due to hydrodynamic and physiochemical factors.

(b) to split the overall mass transfer coefficients to the individual film coefficients $K_G$ and $K_L$ and to find a more precise method of calculation of rectification columns of the film type by making use of the individual film coefficients of mass transfer. They chose three binary systems, namely, acetone/water, carbon tetrachloride/toluene, and benzene/heptane for their test mixtures and the experiments were carried out in a laboratory glass rectification column of the film type. The experiments were conducted at total reflux and the compositions of samples were determined by refractometric methods. The calculation of mass transfer coefficients were made according to:

$$M = K_{OG} \Delta y_m \cdot a \cdot$$

$$= K_{OL} \Delta x_m \cdot a \cdot$$

(1.10)

where:

- $M$ = the quantity of the M.V.C. transferred Kg. mole/hr.
- $K_{OG}$ and $K_{OL}$ = the overall mass transfer coefficient Kg. mole/$(m)^2$.(hr).
- $\Delta y_m$ and $\Delta x_m$ = the mean driving forces, kg. moles/(kg. mole).
- $a$ = the surface of the phase contact, $m^2$.

$\Delta y_m$ and $\Delta x_m$ were given by the following equations:
\[ \Delta y_m = \frac{y_t - y_b}{\int y_t \, dy} \quad \text{and} \quad \int y_b \, dy \]

\[ \Delta x_m = \frac{x_t - x_b}{\int x_t \, dx} \quad \text{and} \quad \int x_b \, dx \]

also \( M = G(y_t - y_b) = L(x_t - x_b) \)

where \( G \) = the quantity of vapour passing through the columns, kg/hr.

and \( L \) = the quantity of reflux liquid, kg/hr.

It was concluded that \( K_{OG} \) and \( K_{OL} \) depended on the composition of the liquid and vapour as well as the velocity. This conclusion has also been reached by other workers (18), for acetone/water, methanol/water and ethanol/water systems.

\( K_{OG} \) increased with an increase in the concentration of the M.V.C. and \( K_{OL} \) decreased with such increase in the concentration. These results are in agreement with those of Orlov and Planovskii (19), Kol'tsov and Planovskii (20) and Kesatkin and Chekhov (21).

The relation between the overall and the individual film coefficients was according to the following equation:

\[ \frac{1}{K_{OG}} = \frac{1}{rG} + \frac{m_a}{K_L} \quad \text{........... (1.11)} \]
where $m_a$ is the mean tangent of the angle of inclination of the equilibrium line on the operating section; $m_a$ was defined by:

$$m_a = \frac{y}{x_m}$$

The general mass transfer equations for the mixtures investigated were:

$$N_{u_v} = 0.62 \times 10^{-5} \quad Re_v \quad \frac{0.76}{\left( \frac{\mu}{\mu_v} \right)^{2.3}}$$

$$= 22.4 \quad KG \quad de/D_v$$

and

$$N_{u_L} = 0.525 \times 10^{-6} \quad Re_L \quad P_{RL}$$

$$= K_L \quad M_L \quad \text{m/}v_{LM} ^* \quad D_{LM}$$

where:

de: the equivalent diameter of the channel; (m)

$D_v$: coefficient of diffusion, $m^2$/hr.

$M_L$: mean molecular weight of the liquid phase of the mixture, $m^2$/hr.

$v_{LM}$: mean specific gravity of the liquid, Kg/m$^3$.

$M$: the thickness of the film.

$\mu_L$ and $\mu_v$ are the viscosities of the liquid and vapour phases, Kg. sec/m$^2$.

Equation for $N_{u_v}$ is valid within the limits

$$Re_v = 2000 - 12,000$$

and $\mu_L / \mu_v = 26 - 50$

Equation for $N_{u_L}$ is applicable within limits of $Re_L$

$$= 186-1900 \quad \text{and} \quad P_{RL} = 80-250.$$
1.2.1 **CONCLUSIONS**

The following points can be drawn resulting from the discussion treated so far:

1. Mass transfer coefficients depend on the physical properties of the liquid and vapour, as well as on their velocity.

2. The mass transfer coefficient relative to the vapour phase \( K_{OG} \) increases with a rise in concentration of the M.V.C. and \( K_{oL} \) decreases with such increase in composition.

3. Mass transfer in apparatus of the film type can be expressed by means of the partial coefficients of mass transfer in the vapour and the liquid phases.

4. The mass transfer coefficients \( (K_G) \) and \( (K_L) \) do not depend on changes in concentration but vary strongly as a function of the velocities of the vapour and liquid phases.

5. The dimensionless equations obtained permit making calculations of the film rectification columns with the use of the mass transfer coefficients for the vapour and liquid phases.

6. In calculating the Reynolds numbers for the vapour and liquid phases, it is necessary to use the relative velocities of the vapour streams and the liquid film.
1.3 Theories of Interphase Transfer

1.3.1 Introduction

In 1922, Peters (22) introduced the concept of the height equivalent to a theoretical plate (H.E.T.P.) as a measure of a column performance. The first complete theory of distillation was advanced by Kirschbaum (23) in 1931, and was based on considerations of the heat transfer taking place between the two phases.

Chilton and Colburn (24) in 1935, formulated a theory based on countercurrent mass transfer. It represented distillation as a diffusional process and used the Whitman model for its evaluation. At the same time Higbie put forward the idea of systematic surface renewal which formed also the basis of the penetration theory. This has subsequently been modified by Danckwerts and Kishinevskii. A different theory was proposed by Bosnjakovic (25) on kinetic considerations of disturbed equilibrium between the phases.

The various theories and models proposed for continuous transfer processes have been developed from work on packed distillation and wetted-wall columns and it is considered to be worthwhile to discuss them in some detail, with the intention of emphasising on the assumptions made for their derivations and also their limitations.
1.3.2 The Two-Film Theory of Whitman

The two-film theory assumes the existence of laminar layers on both sides of the interface with the bulk of the phases in turbulent motion. The transfer of mass through the laminar layers takes place by molecular diffusion and thus represents a steady state process.

The final rate equation through the vapour film is given by:

\[ N_i = \frac{D g P}{RT} \left( y_i - y \right) \]  \hspace{1cm} (1.12)

where:

- \( N_i \) = rate of transfer in moles/unit area and unit time;
- \( D \) = diffusion coefficient in the vapour phase;
- \( g \) = effective thickness of the vapour film;
- \( y_i \) = interface composition of the diffusing component.

The stagnant laminar layers, as visualised by the two-film theory can only exist if:

(a) The velocity gradient reaches zero at the interface.

(b) The turbulence decreases to zero at the interface.

Owing to the high mobility of the phases it is difficult to visualise these conditions in gas-liquid systems unless the phases are virtually at rest.

For a long time, it was supposed that the conditions required by the two-film theory were set up in a wetted-wall column. However, it has been found that it is almost impossible to eliminate the formation of ripples and a mathematical analysis by Kapitsa\(^{(26)}\) showed that
wave motion in falling films is more stable than laminar flow.

1.3.3. The Transfer Unit Theory

Since the transfer unit theory, as developed by Chilton and Colburn was based on the two-film theory, it will consequently be subject to the same criticism.

In recent years, experimental results were obtained which cast doubt on the validity of this theory, particularly on the applicability of the equation:

\[ H_{OG} = H_G + \frac{mG}{L} \]

(1.13)

This equation is often referred to as the Colburn equation. Its main application was considered to be in the possibility it offered for splitting the overall H.T.U. into film H.T.U's by plotting \( H_{OG} \) versus \( \frac{mG}{L} \).

Furthermore, on the basis of this equation, various claims were advanced concerning the controlling influence exercised by either of the films according to the variation of \( H_{OG} \) with \( \frac{mG}{L} \).

In 1943, Gerster, Koffolt and Withrow produced results plotted as \( H_{OG} \) versus \( \frac{mG}{L} \). The graph showed that the intercepts were negative throughout and that both the intercepts and the slopes varied with reflux ratio, Table (1.1.)
According to the conventional interpretation of the Colburn equation, a negative intercept would mean a negative value of the gas film H.T.U., but this is obviously impossible. The authors concluded that it is more satisfactory to plot $H_{OG}$ versus $m$, with $G/L$ as parameter than $H_{OG}$ versus $m \times G/L$.

Large negative intercepts have also been obtained by Smith, who attributed them to the failure of the two-film theory to represent adequately the conditions at the interface. On plotting $H_{OG}$ versus $m \times G/L$ for the distillation of isopropanol-water mixtures at total reflux.
Deed, Schurtz and Drew obtained a curve instead of a straight line. They interpreted it as an indication of a variation in the relative importance of the film resistances with concentration.

Experiments on distillation in wetted-wall columns, carried out by Johnstone and Pigford provided evidence, based on the Colburn equation, that the vapour film was controlling. However, Furnas and Taylor found a marked influence of \( m \) on \( H_{OG} \) which on the basis of the Colburn equation, they explained as being due to the controlling influence of the liquid film.

Although Pratt, in his treatment of the mass transfer coefficient assumed distillation to be a vapour film controlled process, Bowman and Briant introduced a postulate into their calculation which was equivalent to assuming that both film resistances were equal.

The main objections to the transfer unit theory can be summarized as follows:

(a) Assumption of the Whitman model for the mechanism of transfer.

(b) Assumption of constant molar overflow (this condition is only fulfilled if the molar latent heats of the components are equal).

(c) The existence of negative intercepts.

(d) The variation of slopes and intercepts with \( G/L \)
(e) The variation of slopes and intercepts with m.

where c, d, e refer to the graphical representation of equation (1.13).

1.3.4 The Penetration Theory and other Theories of Surface Renewal

The aim of the penetration theory as formulated by Higbie, (29) is to give a more realistic picture of the hydrodynamic conditions occurring at the interface. It presents a case of unsteady state diffusion and can be solved subject to some boundary conditions to give the following expression:

$$N' = (C_o - C_e) \sqrt{\frac{D}{\tau}} \quad \ldots \ldots \quad (1.14)$$

where $N'$ = local rate of mass transfer in moles per unit area and unit time.

$C_o$ = initial bulk concentration in the liquid phase.

$C_e$ = equilibrium concentration.

$\tau$ = time of exposure of a surface element.

This theory, in this respect, is considered superior to other theories based on the Whitman model.

The main objections which can be raised towards Higbie's theory are:

(a) The assumption of an infinite depth of liquid.

(b) The assumption of regular surface renewal.

(c) The assumption of molecular diffusion during the time of contact.
The first assumption can be regarded as justified if the time of contact is short e.g. in packed columns. The second assumption cannot be justified in the case of columns packed at random but this objection can be overcome by assuming Danckwert's modification by introducing the surface age distribution function in this treatment. The third assumption is, however, common to both the Higbie and Danckwert's model and it has been criticised by Kischenevskii.

Kishenevskii (30) studied the absorption of CO$_2$ into Na$_2$CO$_3$ - solution and used the results obtained to evaluate the energy of activation of the rate determining step in the reaction involved. By comparing these values with those determined by other methods, he found that the theory of Danckwerts gives wrong values for the energy of activation, and indicates that this quantity should vary with turbulence (eddy diffusion).

In the theories of surface renewal, the values for the addition of resistances are still applicable. Thus, if a gas-phase resistance is present, then according to Danckweart's theory:

\[
N' = \frac{C_o - C_e}{\sqrt{D_s}} \left( \frac{1}{K_G} + \frac{m}{K_L + \frac{K_G}{m}} \right) = \frac{C_o - C_e}{1 + \frac{m}{1 + \frac{K_G}{K_L} + \frac{K_G}{m}}} \quad (1.15)
\]

\(s\) = rate of surface renewal (fraction per unit time).
1.3.5. The Theory of Kirschbaum

The theory of Kirschbaum has not been widely accepted and experimental evidence for its support is lacking. The mechanism assumed in the original theory is that of thermal distillation with vapour being generated within the liquid phase.

This mechanism is very unlikely and it has been dropped by Kirschbaum in favour of the theory of equivalent heat and mass transfer.

The main criticism of the latter is the assumption of the two-film theory.

1.3.6. The Theory of Bosnjakovic

The limitations of the theory have been indicated by Bosnjakovic. He mentioned that in most practical cases, the removal of the molecules from the interface and not the transfer across the interface controls the process. The resistance to transfer, as visualised by Bosnjakovic is of kinetic character. However, according to Schrage, this type of resistance does not exceed 10% of the total resistance, even under most favourable conditions.

Bosnjakovic, introduced the concepts of invasion and evasion and by correlating the respective coefficients with temperature and pressure, using the kinetic theory of gases, he finally obtained the following expression:-
\[ N' = \frac{\gamma P}{M^2} \cdot \frac{Y_e}{T_L} - y \]  
\[ \text{...............(1.16)} \]

where:
\( \gamma \) = coefficient depending upon the state of motion.
\( M \) = Mol. weight of the M.V.C.
\( T_L \) = absolute temperature of the liquid surface.
\( T_G \) = absolute temperature of the vapour.

Smith, modified this by introducing a liquid phase resistance of the surface renewal type and obtained the following expression:-
\[ \frac{1}{H_{OG}} = C \cdot \left( 1 - (bm + 1) e^{-bm} \right) \]  
\[ \text{...............(1.17)} \]

where \( C \) and \( b \) are constants.

1.4. Vibrations and Pulsations

1.4.1. Introduction

The Chemical engineering literature contains a vast amount of evidence that externally applied vibrations and pulsations, of controlled intensity, have beneficial effects.

Many of the published investigations have been on a laboratory scale, although industrial applications also exist in a few cases e.g. in liquid-liquid extraction. The various applications which have been improved by pulsations are indicated in Figure(1.0)as a function of frequency and the generators available are also shown.
FREQUENCY RANGES FOR VARIOUS EFFECTS OF VIBRATION AND PULSATIONS AND TYPES OF GENERATORS
A brief discussion is presented on some of the more general chemical engineering operations in which applications of pulsations are being developed and practiced.

1.4.2. Liquid-Liquid Extraction

The performance of the perforated-plate column for liquid-liquid extraction was greatly improved by superimposing pulsations. Fluid pulsations at low frequencies (1-5 c/s) has been found to be an ideal method of applying moderate agitation to continuous extraction columns. The original patent (32) proposed vibration of the sieve plates in the column, but later development has been concentrated on columns with fixed plates or packing and pulsed flow of the continuous phase. It soon became clear that pulsations not only reduced the stage height by a factor of (3) or more, but also raised the permissable throughput by preventing flooding.

Thornton(34) and Chantry et al. (35) investigated pulsed packed columns and reported that the H.T.U. reached a minimum value at certain amplitude-frequency combinations. Increasing the pulsations above the optimum level reduced the contacting efficiency because of back-mixing.

Konovalov and Romankov (36), have studied the effect of vibrations on mass transfer in an inclined extraction column and reported yield increases of 2-3 times for identical flows and energy consumption. The effect of pulsations was studied in extraction columns with packings, sieves, etc. and yield increases of more than 200% were obtained (37).
It is believed that in extraction, pulsation improves the efficiency in two ways:—

(a) It increases the contact area between the phases.
(b) It decreases the thickness of the boundary layer which controls the interphase mass transfer because of increased turbulence.

1.4.3 Gas-Liquid Contacting

Bubble Dispersions

Several workers have found that gas absorption rates from bubble dispersions can be increased with the application of vertical vibrations. The largest effects have been observed in the frequency range of 10-50 c/s with well-defined maximum values at certain frequencies (38)

The behaviour of vibrating bubble dispersions can be largely explained by three different effects:—

(a) Forces acting on bubbles:

Bjerknes (39), showed that a pulsating body immersed in a synchronously vibrating fluid was subject to a uni-directional force and also that two adjacent pulsating bodies attracted each other if the pulsations were in phase and were mutually repelled if the pulsations were 180° out of phase.

At high vibration intensities, gas bubbles may be prevented from rising, or entrained downwards from the liquid surface. Absorbers operated under such conditions produce extremely high mass transfer rates. According to the simple theory (40) a bubble is halted by vibrations if
M is unity, where:

\[
M = \omega^4 \frac{A^2 \rho \gamma}{2g \rho_0}
\]  

(1.18)

Observations\(^{(41)}\) confirm this theory at low vibrational Reynolds numbers, but as the Reynolds number increases, the critical value of M rises to about 2.

(b) Resonance of bubbles:

A gas bubble, immersed in a liquid, has a characteristic resonant pulsation frequency given by Minnaert\(^{(41,43)}\) as:

\[
\omega = \frac{\left( \frac{3 \rho_0 \gamma}{\rho} \right)^{1/2}}{r}
\]

(1.19)

(c) Resonance of bubble dispersions:

The velocity of sound in a gas-liquid dispersion can be calculated\(^{(42)}\) to be very low, of the order of 30 m/s. It follows that a column of dispersion should resonate at correspondingly low frequencies and this was confirmed experimentally\(^{(43)}\).

Recent investigations of gas absorption in vibrated bubble columns are summarized in Table (1.1)

1.4.4 Fluidization

Bretsnajder et al\(^{(44)}\) found that fluidization of a vibrating bed could be achieved at much lower gas velocities than were required for static bed. Heat transfer is greatly enhanced by vibrating the bed and wall heat transfer coefficients of up to 40 Btu/(ft)\(^2\).(h)(°F) have been reported\(^{(45)}\). Conditions within a vibrating bed are
<table>
<thead>
<tr>
<th>INVESTIGATORS</th>
<th>Bretsnadjer &amp; Pasiuk (49)</th>
<th>Harbaum and Houghton (38)</th>
<th>Buchanan et al. (50)</th>
<th>Jameson (51)</th>
<th>Baird (52)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Column dia, cm.</td>
<td>5.0</td>
<td>5.2</td>
<td>7.0</td>
<td>7.6</td>
<td>10.3</td>
</tr>
<tr>
<td>Freq. range, c/s</td>
<td>0-80</td>
<td>200-2000</td>
<td>20-50</td>
<td>27-34</td>
<td>1.3</td>
</tr>
<tr>
<td>Amplitude range, cm.</td>
<td>0.83</td>
<td>0.1</td>
<td>0.05-1.0</td>
<td>0.614</td>
<td>5.0</td>
</tr>
<tr>
<td>System investigated</td>
<td>CO₂/water (continuous)</td>
<td>CO₂/water (continuous)</td>
<td>Air/Na₂SO₃ (continuous)</td>
<td>O₂/Na₂SO₃ (batch)</td>
<td>Air/Na₂SO₃ (batch)</td>
</tr>
<tr>
<td>Max. improvement factor</td>
<td>3.0</td>
<td>1.7</td>
<td>-</td>
<td>-</td>
<td>1.95</td>
</tr>
<tr>
<td>Most effective frequencies, c/s</td>
<td>14</td>
<td>125 - 75 - 57</td>
<td>No peaks</td>
<td>31</td>
<td>-</td>
</tr>
<tr>
<td>Corresponding column height, cm.</td>
<td>35</td>
<td>15.4 - 23.2 - 30.8 (15-45)</td>
<td>23</td>
<td>(250)</td>
<td></td>
</tr>
<tr>
<td>Power input, hp/100 gall.</td>
<td>-</td>
<td>10</td>
<td>-</td>
<td>100*</td>
<td>4</td>
</tr>
</tbody>
</table>

* Approximate estimate based on 10 per cent loading of ½ hp motor.

**TABLE 1.2. GAS ABSORPTION IN VERTICALLY VIBRATING BUBBLE COLUMN**
virtually isothermal and vibration has been recommended for exothermic gas-solid reactions.

Resonance phenomena are not usual in particle beds, but an exception was reported by Mitkevich (47) who calcined sodium bicarbonate and found pronounced resonance at frequencies between 23.3 and 50 c/s depending on the height of the bed.

Sound waves of frequency 50-500 c/s have been applied to fluidized beds by Morse (48). No effect was found in the case of free-flowing materials such as plastic beads; perhaps because of the "impedance mismatch" involved in transmitting sound to solids. However, the sound resulted in a substantial improvement in the fluidization characteristics of non-fluent solids such as plaster of Paris.

1.4.5 Chemical Reactions

The effects of sonic and sub-sonic frequencies are to accelerate those reactions which are limited by some physical process such as diffusion. Low frequency liquid-phase pulsations have been applied to a small fixed-bed reactor resulting in rate increases of up to 80%. The reaction studied was the catalytic hydrogenation of methyl styrene which is known to be diffusion-controlled and the results obtained were comparable with Krasuk and Smith's (54) work on mass transfer in pulsed flow through a fixed bed. The improvement in rate due to pulsation decreases as the steady flow rate increases.
As well as improving yields in reactors, pulsations can reduce chemical fouling i.e. the build-up of viscous reaction products on the reactor walls. A recent patent describes a resonant pulsed reactor developed for the latter purpose. Pulsations were applied to the liquid in a tubular reactor by a push-pull system comprising two gas chambers connected upstream and downstream. The resonant frequency of the arrangement was approximately 0.5 c/s, a low figure consistent with the length (197 ft) of the reactor tube.

1.5. Pulsations - Effects on Heat and Mass Transfer
1.5.1. GENERAL

The earliest work on vibration-associated heat transfer was by Martinelli and Boelter who found that natural convection from a \( \frac{3}{4} \)-inch tube to water was increased by up to 400% by vibration (amplitude up to 0.1 in., frequency up to 40 c/s).

Vibrations were also applied by Lemlich to an electrically heated nichrome wire resulting in up to fourfold increase in heat transfer. Lemlich proposed the "stretched film" model, based on the observation that cigarette smoke passed around the vibrating wire as if it were a plate. A stretched vibrational Reynolds number was defined as follows:

\[
R_{es} = (d + 2A) (4A f/\nu)
\]
It was possible to correlate data on both heat and mass transfer\((58)\) in terms of \((Reo)\).

\((59)\) Fand and Kaye observed particularly intense streaming \((\text{steady circulation wave occurs near a fixed object in a standing sound wave})\) at heated horizontal cylinders and christened the effect "thermoacoustic streaming". Later experiments\((60)\) led to correlations\((61)\) for heat transfer from:

(a) horizontally vibrating cylinders.
(b) cylinders in horizontal sound fields.

Deaver et al\((62)\) considered heat transfer from an oscillating horizontal wire to water. They were able to correlate their results, with some success, by the simple expedient of forming dimensionless groups similar to those used in correlations for combined free and force convection in steady flow past a cylinder. Detailed literature surveys on the subject have been given by Lemlich and Fand and Kaye\((63)\).

Low and Hodgins\((64)\) measured local evaporation rates of napthalene into an air stream in the presence of a 286 C/s standing wave. The transfer rate at the modes was slightly reduced by sonic radiation, but the overall effect was positive with an improvement of up to 170% at the antinodes. The enhancement increases with sound intensity and decreases with increasing frequency. The amplitude of the oscillation is a dominant factor in the enhancement.
The sound flux changes both concentration and velocity profiles in the column, probably by alteration of the boundary layer. It was reported that if oscillations are intended to produce turbulence, it is generally more effective to use a high amplitude and low frequency than to use a low amplitude and high frequency.

The relative importance of amplitude is confirmed by Lemlich who correlated his extensive results on heat and mass transfer from vibrating cylinders in terms of a "stretched film" Reynolds number, which emphasized amplitude more than frequency. Similarly, Kalashkinov and Chernikin found that heat transfer from vibrating cylinders was strongly dependent on amplitude.

Lemlich studying the effect of vibrations of frequency 20 to 118 c/s on small horizontal cylinders sublimming to room air reported increases of up to 66% in the coefficient of mass transfer. Two regions of improvement in the coefficient were distinguished; one of low improvement and the other of rapidly rising improvement with the differences between them attributed to differences in streaming pattern.

The improvement in coefficient is correlated in terms of the stretched vibrational Reynolds number by the following:

\[ \frac{K_g}{K'_g} = 0.117 \left( \frac{R}{Re} \right) 0.85 \quad \ldots \ldots \ldots (1.21) \]

\[ Re_s = (D + H) \frac{\bar{V}}{\sqrt{\mu}} \]

\[ \bar{V} = 2 HF. \]
$K_G =$ mass transfer coefficient
$K'_G =$ mass transfer coefficient for no vibration.
$\overline{V} =$ average vibrational velocity, mm/sec.
$D =$ diameter of sample, mm.
$H =$ path length; amplitude, mm.

The results are qualitatively in accord with earlier corresponding work in heat transfer. A modification of the correlation to include the heat transfer results (above $Re'_s = 20$) can be represented as follows with a standard deviation of ± 16%.

$$
\frac{K_G}{K'_G} = 0.038 \ Re'_s^{0.85} \ \phi_s^{1.13} \ \cdots \ \cdots \ (1.22)
$$

$$
\frac{h}{h'} = 0.038 \ Re'_s^{0.85} \ \phi'_s^{1.13} \ \cdots \ \cdots \ (1.23)
$$

Of course further work is required to see how well these relationships can be extended beyond the range of variables studied.

Goh (66) conducted a similar investigation, vibrating a cylindrical sample of naphthalene in a vertical sinusoidal motion, obtaining mass transfer rates of up to five times as high as those due to free convection alone. His data did not correlate well with those of Lemlich and Levy. Knight and Ratkowski (67) attempted to correlate both sets of data with the empirical equation:

$$
\frac{K_G}{K'_G} - 1 = 0.021 \ Re'_V \left(\frac{A}{A'}\right)^2 \ \cdots \ \cdots \ (1.24)
$$
Re_v : $2fA \frac{d}{\nu}$ vibrational Reynolds number.

A : effective surface area of cylinder

$(\pi d+2A)L$, cm$^2$.

A' : surface area of cylinder, $\pi dL$, cm$^2$.

L : length of cylinder, cm.

but the data of Goh, were still somewhat lower by a few per cent.

Shirotuka and Honda(68) studied mass transfer through an inside tube wall coated with p-naphthol, to a pulsation flow. They concluded that:

(a) Mass-transfer coefficient increases in direct proportion to the increase of amplitude and frequency, but in inverse proportion to that of Re number.

(b) The following equation represents the rate $J_D$ - factor increase, $\left[ \frac{(J_P - J_S)}{J_S} \right]_D$,

as obtained by the dimensional analysis:

$$ (J_P - J_S/J_S)_D = 8.2 \times 10^{-4} \left( \frac{Af}{w} \right)^{1.1} \left( \frac{afd}{\nu} \right)^{1.5} \left( \frac{d}{A} \right)^{1.3} \ldots (1.25) $$

(c) It was supposed that the characteristic increment of mass transfer due to pulsation can be explained in the light of increased turbulence of flow.
Jameson\(^{(69)}\) studied transfer rates of benzoic acid to glycerol-water mixtures and obtained values up to 28 times higher than those due to free convection alone. He derived an expression based upon boundary-layer theory and for mass transfer in streaming flow as follows:

\[ Sh = 0.746 \left( \frac{2\omega A\gamma/\nu}{1} \right)^{\frac{1}{3}} Sc^{\frac{1}{3}} \left( \frac{A}{r} \right)^{1/6} \] 

(valid where \( Sc \geq 1 \), \( \frac{A}{r} \leq 1 \))

Rao, Rafin and Rao\(^{(70)(71)}\) investigated mass transfer using both liquid and gas as the fluid medium. Their systems included benzoic acid into water, electrolytic redox reactions in potassium ferri and ferrocyanide and naphthalene into air. In all cases, they found that their data could be correlated by the following equation:

\[ Sh = 0.41 R_{ev}^{0.62} S_c^{\frac{1}{3}} \] 

Another investigation into mass transfer has been made by Fiklistov and Akselrud\(^{(72)(73)}\) using the system CaSO\(_4\) - H\(_2\)O. They obtained an increase in the mass transfer coefficient of up to (10) times that for free convection.

Other workers obtained an experimental equation which covers a large range of Reynolds numbers in both liquid and gas; as follows:

\[ Sh = 0.237 R_{ev}^{0.633} S_c^{\frac{1}{3}} \left( \frac{A}{d} \right)^{0.243} \]
This represents most of the work investigated so far on mass transfer with average % deviation of 26% covering a range of frequencies between 100 - 7000 c/min, double amplitudes between 0.05 - 4.0 cm, diameters between 0.07 - 1.1 cm and vibrational Reynolds numbers between 9 - 2000. In addition, the range of the ratio of double amplitude to diameter, $A/d = 0.2 - 5.7$.

Baird, (74) conducted an experimental investigation in the absence of mass transfer in order to determine some qualitative information on the flow conditions around a rigid cylinder fixed in a liquid which oscillates transversely with respect to the cylinder axis. In particular, he attempted to define a criterion which would predict the flow conditions under which the boundary layer might be expected to separate from the cylinder surface. Using a quasi-steady state approach and obtaining data by employing water as well as mixtures of toluene and liquid paraffin, he proposed that a transition would occur if a dimensionless group $\beta$, given as:

$$\beta = 2.19 \times f^{0.2} / d^{0.6} \nu^{0.2} \quad \ldots \ldots \quad (1.29)$$

exceeds a value of (2)

He also pointed out, however, that $\beta$, might not be able to be used directly when mass transfer occurs, because of the additional effect of convection.
Low frequency fluid pulsations have several practical advantages over vibrations and sound, namely:

(a) No acoustic shielding is necessary nor does provision have to be made for vibrating the equipment.

(b) Pulsations are not attenuated in long pipes, packed bed etc. as much as higher frequency oscillations.

(c) A variety of pulse generators (pistons, flow interruptors, air pulses) can be made for large scale operation at moderate cost.

The simplest quantitative approach to transfer processes in pulsed flow is to estimate the transfer rate at any instant from the fluid velocity at that instant, using a known steady-state relationship; then to integrate over one pulsation period to obtain the time average coefficient. This is the essence of the quasi-steady state theory. (75)

It can be shown that if the transfer coefficient is related to flow velocity by a simple power law:

\[ h \propto u^n \]

where: \( h \): heat or mass transfer coefficient and if the exponent \( n < 1 \), the pulsations of small amplitude should decrease the overall coefficient.
Havemann and Rao (76) worked with turbulent air streams, used pulse frequencies of 5 - 33 c/s and found a decrease in heat transfer at low frequencies but an increase at high ones. The transition frequency was in the region of 20 c/s, and depended to some extent on the pulse waveform.

It has been suggested (77) that in turbulent flow, the condition of validity of the quasi-steady state theory is given approximately by:

\[ \alpha \ll 7.4 \]

where \( \alpha \) = dimensionless frequency = \( \left( \frac{\omega^2}{u} \right)^{\frac{1}{2}} \) .....(1.30)

In laminar conditions, Poisouille flow is found at values of \( \alpha \) below 20, while if \( \alpha \) exceeds 70 the bulk of the liquid oscillates in plug flow with a boundary layer near the tube wall.

Pulsed laminar flow is more amenable to theory (54) than turbulent flow, and Krasuk and Smith have calculated the improvement factor (R) as a function of two groups, namely, the dimensionless pulsation velocity, (Y) and the dimensionless pulse frequency (\( \alpha \)).

The observations of the dissolution of \( \beta \)-naphthol from the inner wall of a \( \frac{3}{4} \)-inch tube supported the theory (68) as did the earlier data of Shirotsuka and Honda (68).

Baird (78), found the best empirical correlation for R, when investigating the performance of a heat exchanger to be:
\[ R = 0.96 + 0.0077 \frac{0.25}{R_{cm}} Y. \quad \ldots \ldots \quad (1.31) \]

\( R_{cm} \): mean \( R_e \) in pulsed flow.

\( Y \): dimensionless pulsation velocity \( = \frac{\omega A}{u} \)

It is interesting to compare equation (1.31) with the results obtained by Krasuk and Smith (54) on the investigation of the dissolution of \( \mathcal{E} \)-naphthol from the wall of a \( \frac{3}{4} \)-inch diameter tube containing water in pulsed flow. They worked chiefly in the streamline flow regime, and found the ratio \( R \) to be strongly dependent on \( Y \).

The dimensionless group \( (\alpha) \), given in equation (1.30) was also important. They also found that the effects of pulsation on mass transfer became negligible in the turbulent flow regime, disappearing entirely at a Reynolds number of 11,000. This is in contrast with equation (1.31) which shows a greater improvement as the Reynolds number is increased at constant \( Y \).

From the practical point of view, it was concluded that pulsations will improve heat transfer in the turbulent flow regime, particularly if the flow can be made to reverse in direction for part of the cycle. The maximum improvement ratio obtained was \( R = 1.41 \) on the basis of the overall heat transfer coefficient, and the theoretical improvement in the water side coefficient was estimated to give \( R = 3.25 \).
1.5.2. **Intensification of Diffusional Processes**

The speed at which diffusional processes take place depends on the rates of heat and mass transfer. Improvement in the rate of heat and mass transfer is achieved by increasing the turbulence of a stream of liquid or gas. In a turbulent flow all the constituent streams constantly change the speed and direction and, therefore, at any point within the flow of liquid or gas there exists speed pulsations.

It can be assumed that artificial excitation or pulsations would increase the rate of heat and mass transfer and this has been discussed in the previous section. It has been established that pulsations increase the heat transfer coefficients and that the increase is a function of the product of the frequency and amplitude of pulsations, \( I \). The relationship between the heat transfer coefficient and the intensity of pulsations for free-flowing solids is given by means of empirical equations such as:

\[
h = 2.26 \times 10^{-2} \quad (I)^{0.96} \quad \text{(for Al-oxide)}
\]

The sublimation of naphthalene in a current of air at room temperature was investigated \( \text{(79)} \) and pulsations proved to improve the mass transfer. Also, measurements were made of the effect of pulsating liquid on the rate of dissolution of a solid body in a form of a cylinder or a sphere suspended in the pulsating liquid. The following systems were studied: copper sulphate/water, aluminium sulphate/water, benzoic acid/water and sodium carbonate/water. The stream of water was subjected to a reciprocating motion by means of a rubber diaphragm, actuated by a pulse generator.
The frequency of pulsations could be varied from 0 - 1800 c/min. and the amplitude from 0 - 0.3 mm. The flow of water in the column was laminar or near laminar \( (Re < 2700) \).

The mass transfer coefficient was calculated from the equation:

\[
K_c = \frac{Wo - W}{\Theta} \cdot \frac{1}{A_M} \cdot \frac{1}{C_S} \quad \ldots \ldots \quad (1.32)
\]

It was observed, that the mass transfer coefficient can be increased 13 fold. A rise of frequency from 0 to 600 c/min, and of amplitude from 0 to 1.12 mm increases the mass transfer coefficient for benzoic acid by 330%. For sodium carbonate, the increase is 660%. Pulsations at a frequency of 700 c/min, and an amplitude of 1.5 mm., increases the mass transfer coefficient for aluminium sulphate by 250%. The effect of amplitude was found to be less than that of frequency; a two-fold increase in amplitude increases the mass transfer coefficient by only 20%.

1.5.3. **Gas-Phase Pulsations**

Gas or vapour-phase pulsations seem to have a promise in contacting operations, provided that the increased pressure drop and flooding tendency can be tolerated.

Ziolkowski and Filip (80) studied the effects of gas-phase pulsations on packed tower performance. Their first investigation, at a constant frequency of 6.5 c/s showed that the scrubbing of \( CO_2 \) by dilute \( NaOH \), was improved threefold by pulsations, but that the gas phase
pressure drop was raised by the same factor. This effect was attributed to wave formation on the gas-liquid surface due to the pulsations together with an increase in turbulence which acts to decrease the gas resistance.

Tudose (81) studied the effect of pulsations on absorption of the system air/ammonia/water in a 1-cm. externally wetted column, and observed rate increases exceeding 210%. He established that the total coefficient of absorption increases in direct proportion to amplitude and frequency of pulsations and that this rate increase has a pronounced peak at 9.3 c/s. The value of 9.3 c/s was independent of the liquid flow rate and was somewhat lower than the calculated resonant frequency of the column of gas.

Shridler et al. (82) studied the humidification of air in a 2.9-cm. i.d. column in presence of sound (40 - 150 c/s). Frequency had a somewhat irregular effect on performance, with the optimum values depending on the air Reynolds number. The best improvement factor of 2.9 was obtained at a Reynolds number of 3,000, and it was suggested that the pulsations acted as a "turbulence trigger".
1.6. The influence of Pulsations on Rectification

In a few instances, natural pulsations in distillation columns have been reported (83). These can only occur at high liquid and gas flow rates and result in an increase in the efficiency of the distillation.

In recent years, due to the success of application of pulsed extraction columns, attention has been directed towards finding high efficiency distillation columns and studying the effect of pulsation characteristic on the efficiency.

McGurl (84) studied the effect of pulsing the vapour phase, in a sieve plate laboratory distillation column. He found an optimum combination of pulse amplitude and frequency for any given system and that the higher the vapour rate, the lower the frequency amplitude product which causes a maximum increase in efficiency. It was envisaged that the causes of increased efficiency in plate-tray columns are the increased residence time of vapour in the froth, the continuous disruption of the boundary layers and a decrease in the tendency of the vapour to form channels in the liquid.

Olney (85) discovered that by imposing forced pressure pulsations in a counter-current vapour/liquid system, the desired mass transfer between the phases is facilitated and a closer approach to equilibrium is attained. Within the process limitations specified, it was reported, tray efficiencies increases from 4 to
over 30%.

An especially large increase in efficiency was obtained in those tests, for example, when the pulsation frequency $F \text{ c/s}$, vapour flow rate $Q = \text{cu ft/sec.}$, and liquid flow rate $N = \text{cu ft/sec}$; had values such that:

$$F \left( \frac{Q}{N} \right) > 10,000 \text{ sec}^{-1} \quad \ldots \ldots \ldots \ldots \ldots (1.33)$$

When applied to a hydrocarbon distillation process, wherein larger quantities of volatile material are repeatedly vapourised and condensed, the forced pulsation will result in even greater increases in tray efficiency. The process was also found particularly advantageous in the usual vacuum distillation process, such as employed in the separation of lubricating oil or residual fractions from crude petroleum. Such processes ordinarily involve relatively low liquid loadings with which the pulsations effect a particularly large increase in efficiency.

Baker (86), investigated the performance of a 2-inch diameter packed column, under pulsating pressure. The method of operation in which the high and low pressure were obtained was by introducing air into the column and exhausting it to the atmosphere through the condenser. They concluded that pulsating pressure at pulse frequencies of 60 c/s or less is of little value in improving fractionation efficiency although it may have limited value in simple batch distillation.
Bratu et al. (87) working on a rectifying packed, 32-mm i.d. column, drew the following observations when applying pulsations:

1. The axial vibration of the packing with a frequency of $35 \text{ Hz}$ and amplitudes of $0.5 - 4.0 \text{ mm.}$, enables the enhancement of the separation efficiency 3-5 times as against the efficiency (N.T.U.) of the non-vibrated column.

2. In the case of system n-heptane/toluene mixture, and using 4.0-mm Raschig rings packing, the maximum effect of vibrations was in the range of medium reflux rates (boil-up rate), at total reflux.

3. Within the range of high reflux rates and a vibration frequency of $35 \text{ Hz}$ the maximum efficiency is obtained at an amplitude of 3.5 mm.

4. The pressure drop across the vibrated column is practically the same as across the non-vibrated column, and the flooding limit of the axially vibrated column is near that of the flooding limit of the non-vibrated column within the range of the investigated amplitudes and frequencies.
5. The experimentally determined increase of the column's efficiency due to vibration means no upper limit. It can be presumed that at higher frequencies and amplitudes, the further investigation may lead to the achievement of still higher efficiencies of the packed columns especially at higher vibrational frequencies.

Ziolkowski and Filip (88), working on the influence of pulsations on the fractioning capacity of packed columns, using n-heptane/toluene mixture found that the efficiency, \( N_{OG} \), can be increased almost two-fold when the pulsation parameters are suitably chosen. Increase in the amplitude, in a suitable range, and of the frequency of pulsation, has a positive influence on mass transfer.

Improvement of mass transfer in the rectification process is closely linked with a corresponding increase in heat transfer between the two phases. The results did not show any particular optimum frequency as was found for pulsed wetted-wall columns (81), but an optimum amplitude of about 28 mm was observed.
1.7. **The present work**

There has been a growing amount of research work done in the field of application of vibrations and pulsations to various chemical engineering processes. Studies on laboratory-scale units, have indicated that externally applied pulsations have beneficial effects on separations and rate processes, in a variety of disciplines e.g. absorption (81), fractional crystallisation (89), chemical reactions (90), and heat transfer (63). A significant advance in the last few years, has been the application of controlled cycling to extraction processes (93,94) and to distillation (91,92).

Controlled cycling (95) basically, can be described as a mode of operating a counter-current stagewise mass transfer apparatus such that only one phase flows at a given time and the phases use the same interstage flow passages during their respective flow periods.

The successful operation of the pulsed extraction columns and the substantial advantages gained by pulsations, prompted the application of the principle to distillation. Preliminary investigations (95) carried out, have shown that the efficiency of a wetted-wall column can be improved by subjecting the liquid and the vapour phases to a rapidly oscillating pressure. This attempt has resulted in an increased efficiency of the order of 50%, expressed in terms of theoretical plates. It seemed possible that the resistance to mass transfer could be decreased without a corresponding decrease in the residence time in the column, by the application of pulsations.
The main purpose of this work was to investigate the performance of wetted-wall, pulsed distillation columns with the following goals in mind:

(a) To determine degree of enhancement and advantages attained by the application of pulsations relative to conventional operation.

(b) Determine the effect of intensity of pulsations, on the performance of three wetted-wall columns. The intensity of pulsations is the parameter relating to magnitude of the frequency and amplitude of pulsations.

(c) Attempt to understand and propose, whenever possible, the mechanism of pulsed rectification on the basis of experimental results.
SECTION II

EXPERIMENTAL
### SECTION II

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SECTION TWO

2.1. EXPERIMENTAL EQUIPMENT

2.1.1. Selection of the Experimental Column

In evaluating and selecting rectification equipment, there are two considerations of prime importance, namely: the size of the apparatus required to effect a given separation, and the pressure drop undergone by the vapour in passing through the apparatus. Pressure drop is of particular importance in vacuum distillation because the increase in the pressure down the column may cause the boiling point of the liquid in the still to be higher than desirable.

By far the most widely used rectification device is the bubble-cap plate column. The size of the apparatus required is usually reasonable. The pressure drop in such columns, however, is rather high, and the construction involved is rather expensive.

The packed column has been popular in laboratory and small scale industrial use because of its simplicity and relatively low pressure drop. However, few large diameter packed columns are in commercial use today because of inherent difficulties in maintaining uniform distribution of reflux and also because much more needs to be done in developing satisfactory relationships for use in the design of such columns. The many variables involved such as packing type and size, rates of flow
of liquid and vapour and nature and concentrations of materials fractionated made evaluation difficult and perhaps have discouraged more extensive investigations. In packed columns experience has shown that channelling of liquid often occurs and that the actual surface area available for mass transfer is variant and cannot be estimated with precision.

Investigators have used wetted-wall columns in which the surface of contact is that of a film of liquid flowing down the wall of a tube, vapour passing up through the tube with rectification taking place at the interface. In such columns the interfacial area is constant as long as the tube wall is completely wet and uniform and because of the thin film involved, this area is very nearly equal to that of the tube surface itself. The results obtained with these columns have been most useful in checking the various theoretical relationships which have been advanced for interphase transport. A typical wetted-wall column, designed to operate at total reflux is shown schematically in Figure (2.1). The transfer section consists of a vertical tube A, usually about 1-2 in. diameter and up to 6 ft. in length made either of glass or copper pipe fitted with sampling lines to allow for removal of samples from the liquid film along the length of the column, as well as the liquid reflux line (sampler ST) and the liquid removal chamber (SB).

Liquid reflux is heated to near the boiling point by contact with the rising vapour on a bubble-cap, metered
Figure 2.1.

Schematic drawing of a wetted-wall column

A - Wetted-wall section
B - Bubble cap chamber
C - Condenser
CW - Cooling Water
L1 - Liquid introduction
L2 - Liquid removal chamber
S - Sampling points
R - Reboiler
TC - Thermocouple
through a rotameter and introduced on to a wetted-wall section over a knife-edge, and removed at the bottom by means of a rounded-bell mouth. The calming sections should be at least 2 ft. long to ensure that the vapour and liquid flow has become steady and uniform before it enters the transfer section A. A reboiler, R and a condenser C, must of course be provided for the supply of vapour and reflux respectively to the column. All vapour lines must be heated electrically and lagged to prevent vapour condensation and the wetted-wall section must be sufficiently insulated to achieve close adiabatic conditions. The assumption is made that the nature of the distillation process in a wetted-wall column is the same as that of a packed column and that the transfer mechanism should not be different.

A wetted-wall column was selected in this study in favour of other distillation columns, was decided mainly on the basis of the following characteristics:

(i) Uniform liquid distribution throughout its length.
(ii) The degree of dampening the pressure-pulse signal is low as compared to other columns.
(iii) The flow of the two phases in a wetted-wall column is much less disturbed than in a packed column, apart from the formation of ripples on the surface of the liquid film.
(iv) More flexible operation and low pressure drop and hydraulic resistance.
Surface area of contact between the two phases is more easily determined.

2.1.2. General description of Apparatus

The column used in this investigation was of the wetted-wall, batch type, formerly used as a packed column and suitably modified to meet the requirements of this study. The wetted-wall column, due to its simplicity is often used in laboratory work mainly because the contact area between the interacting phases can be estimated. Another advantage is the possibility of maintaining an approximately constant temperature of the liquid reflux by heating the rectifying tube from the outside jacket and thus achieving approximate adiabatic conditions in the system.

The column assembly is shown diagrammatically in Figure (2.2) and Plates 1-3 represent the outside view of the different parts of the column with the ancillary equipment for producing and measuring pulsations of the vapour phase.

The column assembly consists of the following parts:

2.1.3. Rectification Column:

This is the body proper and can be divided into two sections:
Figure 2.2.

Pulse wetted-wall distillation column
2.1.3a. Top Section

This is about 17.625 ins. long and of inside diameter of 2\(\frac{1}{8}\)" tube made of copper. The section is enclosed with an M.S. jacket which is seal welded on a collar of 12-in. in diameter. This section is fitted with pyrotenax heating wire 14-feet long. A provision for a thermocouple well made of S.S. tube of \(\frac{1}{2}\)-ins o.d. is allowed and located flush with the inside wall of the column for temperature recording.

2.1.3b. Bottom Section

This is made of similar fabrication as the top section. It is about 42.375 ins. long and of 2\(\frac{3}{4}\)-in. i.d. copper tube. As above, it is enclosed with an M.S. jacket which is seal welded to a collar at both ends. This section is fitted with three thermocouple assemblies which are 9-ins. apart along the section and also provided with compensating heating element.

Both sections of the column are joined together by means of a 5/8" thick flanges provided at each end of both sections.

The entire column was insulated using 1-in. asbestos rope and fibreglass lagging. When the actual column was installed, it was lined up as vertically as possible by means of a plumb bob and levels.
PLATE 1 - PULSED DISTILLATION COLUMN SHOWING REBOILER
It was desired to study the effect of varying the surface/volume ratio of contact between liquid reflux and the vapour, using the same column, in order to establish the influence of pulsations on this parameter. Three brass tubes 62 inches long and of diameters 1, 1\(\frac{1}{2}\) and 2-in were made. Each individual tube was sealed at both ends and was inserted concentrically inside the column leaving an annulus for the vapour to pass through. A set of three fins were welded around the perimeter of each tube at both ends to ensure central alignment inside the column.

Table (2.1) shows the principal physical dimensions for the three columns used in the experimental study.

**TABLE 2.1.**

Dimensions of Columns used in this investigation

<table>
<thead>
<tr>
<th>Tube Size (ins)</th>
<th>Cross-Sectional Area of Annulus, (sq.cm), A</th>
<th>Volume of free space (cu.cm), V</th>
<th>Wetted Area/Volume Ratio, a (cm(^{-1}))</th>
<th>Equivalent Diameter, (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>20.6</td>
<td>3140</td>
<td>1.26</td>
<td>3.18</td>
</tr>
<tr>
<td>1(\frac{1}{2})</td>
<td>14.25</td>
<td>2180</td>
<td>2.1</td>
<td>1.916</td>
</tr>
<tr>
<td>2</td>
<td>5.475</td>
<td>835</td>
<td>6.3</td>
<td>0.635(\frac{1}{4})</td>
</tr>
</tbody>
</table>

Height of column 5-ft.

Diameter of Outer Tube 2.25-ins.
PLATE 2 - EXPERIMENTAL WETTED-WALL COLUMN
The data in Table (2.1) were calculated on the assumption that both the inner and outer tubes were wetted. The equivalent diameter \(d_e\) values were calculated using the following definition:

\[
d_e = \frac{4}{P} \left( r_H \right) = 4 x A = D - d.
\]

where:

\(r_H\) = hydraulic radius; cm.
\(A\) = annular cross-sectional area of column; sq. cm.
\(D\) = diameter of outer tube; cm.
\(d_0\) = diameter of inner tube; cm.
\(P\) = wetted perimeter; cm.

The Reynolds number of the vapour \(\left( Re_V \right)\) was estimated using the expression:

\[
Re_V = \frac{\rho v(D - d_0)}{\mu}
\]

where:

\(\rho\) = vapour density = 0.00302 gm/c.c.; (p.95)
\(\mu\) = vapour viscosity = 0.00845 cP.
\(v\) = velocity of vapour, cm/sec.

Table (2.1A) gives data of the columns assuming only the outer tube is wetted.

**TABLE (2.1A)**

<table>
<thead>
<tr>
<th>Tube Size (ins.)</th>
<th>Wetted Area/Volume Ratio 'a' (cm)</th>
<th>Equivalent Diameter (d_e) (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.87</td>
<td>4.6</td>
</tr>
<tr>
<td>1(\frac{1}{2})</td>
<td>1.26</td>
<td>3.18</td>
</tr>
<tr>
<td>2</td>
<td>3.28</td>
<td>1.22</td>
</tr>
</tbody>
</table>

Sample Calculation

For the 1\(\frac{1}{2}\)" inner tube column, a sample calculation is given:

i. wetted area/volume Ratio, 'a':

\[
a = \frac{\pi(D + d_0) L}{A x L} = \frac{\pi(2.25 + 1.5) x 2.54}{14.25} (L = \text{height of column}) = \frac{\pi x 3.75 x 2.54}{14.25} = 2.1 \text{ cm}^2
\]

ii. \(d_e = \frac{4 x A}{\pi(D + d_0)} = \frac{4 x 14.25}{\pi x 3.75 x 2.54} = 1.91 \text{ cm.}

iii. \(Re_V = \frac{\rho v(D - d)}{\mu}\)

Choosing Run BP-2, page (131);
\(v = 25 \text{ cm/sec},\)

and, \(Re_V = \frac{0.00302 \times 25 \times 1.91}{0.00845 \times 10^{-2}} = 1702.10\)
2.1.4. **Condenser:**

This is a water jacketed condenser made of mild steel. The diameter of the inner tube, made of copper, is 2.25-ins. and the length of the total condenser including the end flanges is approximately 3 feet. Water from the supply flows through a rotameter and into the jacket of the condenser through ½-in. i.d. P.V.C. tubing. The condenser is erected on top of the top section of the rectification column.

2.1.5. **Reboiler:**

For a column of this type and throughput, it was decided to use a Q.V.F. tube of 2-feet long and 3-in. i.d. to perform as a reboiler and capable of holding a batch of mixture of about 3 litres. This was heated electrically by means of 1200 W, 1-in. wide, 24-feet long, Electrothermal heating tape, wound externally. The power input can be varied and regulated using a variac.

The reboiler was heavily lagged with 1-in. o.d. asbestos rope and fibreglass. A Q.V.F. drain valve was fitted near the lower end of the reboiler for use when changing the batch mixture in the column. Plate 1 shows an outside view of this part of the column.

2.1.6. **Sampling Points:**

An M.S. flange (take-off flange) was designed and fabricated as shown in Figure (2.3) in order to allow the flow of liquid reflux from the inside wall of the condenser to be distributed uniformly after being condensed, and to
Figure 2.3.
Top Sample Take-off flange
Material: M.S.
N.T.S.
wet the inner wall of the outside copper tube of the column. This was located between the condenser and the top section of the column. Top product samples were taken off by means of a valve connecting a 1/8-in. o.d. S.S. tube penetrating through one point at the periphery of the flange. The length and size of this tube carrying the condensate to the valve of the sampling line was made small (about 2-cm), in order to minimise the stagnant liquid hold-up and also not to disturb equilibrium when sampling during a run.

The bottom products were sampled by using a reflux meter shown in Figure (2.4) positioned between the bottom flange of the bottom section of the column and the reboiler. This device is essentially an annulus in which vapour flows up the inner tube. This inner tube is covered with a dome which prevents the returning reflux from running down the tube. The reflux runs down the outer tube. This tube is closed at the bottom except for one line which leads to a three-way stopcock. The stopcock may be turned to allow the reflux to go directly into the reboiler, or it may be turned to allow the returning reflux to be removed for analysis. The third position of the stopcock will allow the ascending vapour to be removed. It is possible to close the stopcock completely by using an immediate position. When in this position, the returning reflux collects in the outer tube of the annulus. This outer tube has a calibration mark of 100 ml. above the stopcock. This allows the reflux rate to be measured by closing the stopcock and determining the time it takes for 50 ml. of reflux to collect. This device of
Figure 2.4.
Reflux rate measurement
Material: Glass
(N.T.S.)
measuring the reflux rate was initially calibrated and checked for accuracy of measurement before experimentation.

2.1.7. Temperature measurements

It was considered necessary to determine the temperature inside the jacket of the column in order to keep as adiabatic conditions as possible in the rectification column.

Measurement was achieved by using three Copper-Constantan thermocouples. The thermocouples were brazed at the joint individually, insulated and calibrated against mercury thermometers. They were inserted in 3 wells along the column spaced about 9-ins apart and connected to a recorder instrument (multipoint) to read directly in °F.

The control of the jacket temperature was achieved simply by means of a variac connected to the compensating heating elements located in the jacket in series.

Better control of the heating jacket temperature in accordance with that inside the column was not considered to be necessary in this study, although preferable in order to achieve adiabatic conditions.

2.2. Additional Apparatus

2.2.1. Pulse Generation

Pulse generators have generally been built to provide what is essentially a Sine wave pulse action; but some workers have found significant increase in mass transfer
or tower throughput by applying other pulse wave shapes approaching, for instance, a saw-tooth or square shape. However, it is not yet clear that such advantages will lead to any significant over-all economic savings over near sine-wave operation, and advantages, if any, need to be investigated further.

The subject of pulse-generation methods has been described in the literature. Thornton (96) gives a description of three methods of pulsing an extraction column namely, mechanical (piston), air-pulsed and the electronically-pulsed techniques. Each of the above has got a certain advantage in different applications.

Wuch and Knight (97) presented the necessary differential equations for the design of air pulsers and gave methods of their solution by numerical means. The solution requires the use of a digital computer. Baird (98) described a self-triggered resonant pulsing technique which he designated as 'water-blow pulsations'. This technique, unlike the previous methods, dispenses with electrical devices and mechanical moving parts and thus minimising on the maintenance aspect of pulsing arrangement.

In this investigation a mechanical pulsator was chosen. Pulsating air pressure is generated from the reciprocating movement of pistons in the two cylinders of a car engine, from which the valve mechanism has been isolated. The engine was driven through a pulley arrangement of a suitable diameter ratio, by a 3.5 H.P.
PLATE 3 - MECHANICAL PULSE GENERATOR
Kopp 3-phase motor. The speed of the motor can be varied by means of an integrated, variator, and hence the frequency of the generated pulsating pressure from the engine can be varied over a suitable operating range 0-25 c/s. It was decided, after initial trials, not to exceed a frequency of 22 c/s due to excessive vibration of the structure and noise.

The pulsating pressure was transmitted to the column by placing a rubber diaphragm between 2 Q.V.F. bell-shaped 9-in. diameter adaptors, and secured at the far end of the condenser. A vent valve was fitted through a T-piece on top of the condenser for use when working at atmospheric pressure and without pulsations. It was observed that the motion of the pulser (piston) was approximately sinusoidal, especially at low pulsations, as displayed on the screen of an oscilloscope.

Method of measuring the pulse amplitude and frequency is described later in (2.2.3) and plate (3) shows the general arrangement used for the pulsations and measurement with ancillary units.

2.2.2. Pressure Transducer

A S.E. 75/6 0-5 p.s.i. transducer was used to detect the pressure pulsations in the rectification column. This is of the flash-diaphragm type, having a high, long-term, temperature stability.
The instrument simply consists of a balanced bridge of inductance fed from a 3 Kc/s. oscillator. Mechanical input to the transducer, (pressure fluctuations), causes the bridge to unbalance, and the bridge output is a measure of the unbalance produced. The output from the bridge is amplified and demodulated to produce an electrical signal proportional to and in phase with the mechanical signal applied to the transducer.

The basic units of the system are:

(a) The transducer bridge.
(b) A stabilised oscillator.
(c) A balancing circuit for the transducer bridge.
(d) An A.C. amplifier to increase the A.C. output voltage from the transducer bridge.
(e) A phase sensitive demodulator to rectify the amplifier output.
(f) A low pass filter network to remove the carrier frequency components from the rectified output.
(g) A continuous recording instrument, a cathode ray oscilloscope, projecting the resultant signal on a screen.

Before using the transducer, it was necessary to perform the calibration against a range of known pressures. A clean mercury manometer was used by means which readings within 0.1 cm. H was possible. The resulting voltage (m.V.) of the signal was read on the screen of the oscilloscope and noted. The resulting chart is shown
in Figure (2.5.) and Plate (2) shows the location of the transducer in the experimental column.

2.2.3. Measurement of Pulsation Parameters

The frequency of pulsations was conveniently determined from the speed of the shaft of the engine by means of a portable tachometer. The amplitude of pulsations was simply varied over a suitable working range by throttling the pulsed air between the pistons and the diaphragm through a side valve situated at one end of a manifold connecting the engine and the top of the column. The value of the amplitude can be estimated by reading off the peak-to-peak height of the sine wave form signal in (m.V.) displayed on the oscilloscope and interpreted in terms of pressure in (cm. Hg).

The pressure variation in the column was detected by a pressure transducer, placed at a point midway along the length of the column and thus depicting an overall effect of the imposed pulsations. The signal being measured on the screen of the oscilloscope was converted to pressure by using a suitably prepared chart obtained by calibration of the instrument against a clean mercury manometer and reproduced in Figure (2.5)
Figure 2.5.
Calibration of the pressure transducer.
A double beam Oscilloscope (Solartron) was used to measure the amplitude of the pressure wave. The uncalibrated shift controls of the channel was adjusted until the bottom of the waveform coincided with a horizontal graticule line on the illuminated 8 x 8 cms. screen. The calibrated Y shift control was advanced until the top of the waveform coincides with the same graticule line. The setting on the scale of the Cal. Y shift control indicates the peak-to-peak amplitude of the signal. The sensitivity range of this instrument is 3mV/cm to 100V/cm and the accuracy is reported by the manufacturers to be ± 5% on all ranges.

Plate (3) shows the layout of the pulse generating equipment with the ancillary measuring instruments.

2.2.4. Subsequent Development of Apparatus.

When the column was first put into operation, the reboiler was heated externally by a 800 watts heating tape, but this was soon found to be inadequate and was replaced by an Electrothermal tape of 24-ft. and 1-in. wide and rated at 1200 watts. Difficulty was still encountered due to local superheating during the warming-up period and evidenced by production of occasional large bubbles of vapour, which when bursting at the surface, would produce a 'suction effect' of part of the liquid up the column. This was partly remedied by surrounding the reboiler with two layers of silver paper and later introducing porous pot in the still in order to promote regular ebullition. The whole reboiler was insulated externally with fibreglass and 1-in. o.d. asbestos cord.
Slight modification was performed at the top sampling point. It was necessary to drain almost 3 c.c. of sample to be sure that the liquid was actually the reflux which was being formed at the time. The removal of this amount of sample caused a change in the composition at the top of the column and because of this, reproducible results were difficult. The sampling arrangement was slightly modified by reducing the length of tube between the sampling point and the take-off flange from which liquid reflux was being collected and made to flow down the column. The sampling valve was also replaced by a smaller one.

Pulsations were initially produced using a 3-phase motor driving the engine via suitably sized pulleys, but this was later found unsuitable for the load in order to produce high frequency of pulsations, (above 20 c/s). This motor was replaced by a Kopp 3-phase 3.5 H.P. motor equipped with a variator by which a frequency of up to 25 c/s was produced, above which excessive vibration resulted and therefore it was decided to limit the range of working frequency below this value.

It was also observed, that after a period of running the pulse generating unit, fine drops of oil carried away from the sump of the engine were being collected via the manifold and the connecting P.V.C. 1-in. diameter line on top of the diaphragm. This was minimised by introducing some filtering medium supported by a gauze-disc in the manifold. The column was checked for leaks regularly to ensure against any extraneous vibration produced,
and all connections and flanges made air-tight by introducing P.T.F.E. gaskets between them.

2.3. THE BINARY SYSTEM

2.3.1a Selection of a Test Mixture

A binary mixture was chosen from the two components, toluene and methylcyclohexane, obtained commercially in a fairly pure condition. Data relating to the physio-chemical properties of the two components are given in Table (2.2.)

The selection of this system in this investigation, was dictated primarily by the relatively great differences in the refractive indices of the components which permitted using a simple and accurate method for their analysis. Furthermore, as is evident from the equilibrium diagram, Figure (2.6) for this system, the change in liquid composition corresponding to one theoretical plate is rather large in the range of concentration used in this study, so that it was possible to achieve good precision in the determination of N.T.P. or (N.T.U.).

The binary system of methylcyclohexane-toluene has been reported as nonideal and non-azeotropic (99) (100). It was reported also that this system was useful if the efficiency of the column to be tested is not expected to exceed 30 theoretical plates. The refractive index is a convenient and quick method of composition determination for this system and a chart of refractive index-composition has been prepared at 20°C and represented in Figure (2.8.).
Figure 2.6.
Methylcyclohexane-Toluene
Equilibrium Data at Atmospheric Pressure
Figure 2.7.

Relative volatility of M.C.H. to toluene
The relative volatility (1.3), as estimated within the working range, was large enough for relative ease of separation and the materials were readily available in high purity and low cost.

2.3.1b Method of Analysis

The two components were purified in a laboratory packed distillation column and mixtures of methylcyclohexane and toluene were prepared in the range of 5- to 95% (wt%) and were analysed by measurement of their refractive index at 20 ± 0.2°C using a Pulfrich Refractometer. This instrument is accurate up to ± 0.0002 units and the temperature was kept constant using a thermostat fitted with a contact thermometer. An accuracy of between 0.10-0.15% (mole) is expected from this procedure.

A refractive index/composition (mole fraction) chart at 20°C was prepared as shown in Figure (2.8) which was later used for analysis of samples during the experimental runs. There is a reasonably fair agreement between the results given elsewhere for the refractive index/composition diagram for this system and the values determined in this laboratory, although these are dependent on the purity of the original components obtained and used for the determination.
Figure 2.8.
Refractive Index vs. Composition
Methylcyclohexane Toluene @ 20°C.
2.3.2. Physical Properties of the Binary Mixture

(a) Vapour - Liquid Equilibrium Data

Vapour - liquid equilibrium data, at atmospheric pressure for this system was consulted from the literature. The data used in this study was taken from Krell's Handbook (101). Other reported data for this system (100), compared well with the data used in this study and also the equilibrium data have been checked for thermodynamic consistency by Thießen (102).

The equilibrium curve at 760 mm. Hg. is plotted in Figure (2.6) and Table (2.4.) represents a summary of the more important data relevant to this binary mixture. It is of interest to mention that the vapour pressure of methylcyclohexane can be expressed by:

\[ \log_e P = 7.3476 - \frac{1670.7}{T} \quad \ldots \ldots (2.1.) \]

and for toluene as:

\[ \log_e P = 7.5635 - \frac{1796.9}{T} \quad \ldots \ldots (2.2.) \]

where:

- \( T \) = temperature °K.
- \( P \) = pressure mm. Hg.

The values of the constants were found from the Circular of National Bureau of Standards.

(b) Density, \( \rho_L \)

Liquid densities of M.C.H. and toluene for a range of temperatures required for calculating boil-up rates and Schmidt numbers were taken from the data given by Timmermans (103). Mixture densities were assumed to be the molar average of the pure component values. As no volume
change occurred on mixing the two components, the densities of mixtures were calculated from the expression:

\[ \rho_L = \rho_A \frac{M_A}{M_{av}} \rho_B + \rho_B \frac{M_B}{M_{av}} \]

\[ \rho_A \rho_B = \text{densities of pure two components.} \]

\[ M_A, M_B = \text{molecular weights of the components.} \]

\[ M_{av} = \text{average molecular weight of the mixture} \]

\[ = M_A x_A + M_B x_B \]

\[ x_A, x_B = \text{mole fractions.} \]

\[ \overline{\text{Vapour Density, } \rho_v} \]

Vapour densities of the pure components were calculated by using the ideal gas laws at the established column temperature and pressure. Values for intermediate concentrations were evaluated using the molecular weight of the mixture and the corresponding condensation temperatures.

(c) Viscosity

Liquid Viscosity:

Viscosities at various temperature were obtained from the charts provided by Drickamer and Bradford \(^{(104)}\). The pure components viscosities were estimated from Bromley and Wilke's equation \(^{(105)}\).
Vapour Viscosity

Vapour mixture viscosities were calculated from the pure component values by the method of Wilke (102) given by:

\[ \eta_G = \frac{\eta_A}{1 + \frac{x_A}{x_B} \epsilon_{AB}} + \frac{\eta_B}{1 + \frac{x_A}{x_B} \epsilon_{BA}} \]  \hspace{1cm} (2.4)

where:

\[ \epsilon_{AB} = \left[ 1 + \left( \frac{\eta_A}{\eta_B} \right)^{\frac{1}{2}} \left( \frac{M_B}{M_A} \right)^{\frac{1}{2}} \right]^2 \]

\[ \frac{2 \sqrt{2} \left( 1 + \frac{M_A}{M_B} \right)^{\frac{1}{2}}}{} \]

and \( \eta_A, \eta_B = \) viscosities of pure components

\( M_A, M_B = \) molecular weights

\( x_A, x_B = \) mole fractions.

(d) Diffusion Coefficient

Liquid diffusivity, \( D_L \)

Liquid diffusivities for dilute solutions of M.C.H. and toluene were determined by the method of Wilke and Chang (106) given by:

\[ D_L = \frac{7.4 \times 10^{-8} \ (M_2)^{0.5} T}{\mu_2 V_1^{0.6}} \]  \hspace{1cm} (2.5)
where subscripts, 1 and 2, indicate solute and solvent respectively, $V_1$ is the molecular volume of the solute at its boiling point, $M_2$ is the M.W. of the solvent, and $\mu_2$ is the viscosity of the solvent.

Diffusivities for concentrated solutions can be calculated by:

$$
\left( \frac{D_{L1} \cdot \mu}{T} \right)_{\text{Conc.}} = \left( \frac{D_{L1} \cdot \mu_2}{T} \right)_{\text{Solvent}} - \left( \frac{D_{L2} \cdot \mu_2}{T} \right)_{\text{Solute}} x_1 + \left( \frac{D_{L1} \cdot \mu}{T} \right)_{\text{Solvent}}
$$

Diffusivities at different temperatures can be obtained by using:

$$
\frac{D_{L1} \cdot \mu_2}{T} = \text{a const.}
$$

Vapour diffusivity, $D_G$

Vapour diffusivity for M.C.H. and toluene system were predicted by the Gilliland equation as follows:

$$
D_G = \left( \frac{0.0043}{P(V_\frac{3}{1} + V_\frac{3}{2})^2} \right) \left( \frac{M_1 + M_2}{M_1 M_2} \right)^{\frac{1}{2}}
$$
where:

- $T =$ absolute temperature ($^\circ$K)
- $P =$ total pressure (atm.)
- $M =$ molecular weight.
- $V =$ molal volume.

(e) **Kinematic Viscosity, $\nu_L$, $\nu_g$**

The values of kinematic viscosity for liquid and vapour were calculated from the values of density and viscosity.

(f) **Refractive index, $\eta^20_D$**

The values of the refractive index for the binary mixture for various compositions at 20°C have been obtained experimentally and reproduced in Figure (2.7.).

(g) **Boiling point temperatures**.

The boiling point temperatures of the system methylcyclohexane-toluene were taken from Krell's Handbook (101).

The boiling point temperature and the condensation temperatures for the system were plotted in Figure (2.9.).

(h) **Relative Volatility, $\alpha$**

The volatility, $V$, of a liquid in a mixture is defined as the ratio of its partial pressure to the liquid composition i.e.:—
Figure 2.9.

Boiling Point Diagram for M.C.H./Toluene System at 760 mm Hg.
\[ \alpha_{AB} = \frac{V_A}{V_B} = \left( \frac{y_A}{x_A}, \frac{x_B}{y_B} \right) \quad \ldots \ldots \ldots \ldots (2.10) \]

The variation of the relative volatility with composition is shown in Figure (2.7).

1. **Number of Transfer Units, N\textsubscript{OG}**

The number of overall transfer units, N\textsubscript{OG}, is defined by the equation:

\[ N_{OG} = \int_{y_B}^{y_T} \frac{dY}{y_e - y} \quad \ldots \ldots \ldots \ldots (2.11) \]

This can be written as:

\[ N_{OG} = \int_{y_0}^{y_T} \frac{dY}{y_e - y} - \int_{y_0}^{y_B} \frac{dY}{y_e - y} \quad \ldots \ldots \ldots \ldots (2.12) \]

which allows for the determination of any value of N\textsubscript{OG} from a single graphical integration over practically the whole composition range. This is shown in Figure (2.10) for the system under study.

All other physical properties could be determined for the mixture from the values obtained for pure components by the equation:

\[ z_M = x_1 z_1 + x_2 z_2 \quad \ldots \ldots \ldots \ldots (2.13) \]
Figure 2.10.
N.T.U. V.S. Composition
Methylcyclohexane/Toluene
Total Reflux

Mol. % M.C.H.
where:

\[ x_1, x_2 = \text{mole fraction of components 1 and 2.} \]
\[ Z_1, Z_2 = \text{a specific physical property of pure components.} \]
\[ Z_M = \text{physical property of the mixture.} \]

Table (2.3.) represents the values of the physical properties of M.C.H./toluene binary mixture in the range of the experimental conditions.

**TABLE 2.2.**
Physical Properties of the pure Components

<table>
<thead>
<tr>
<th>Property</th>
<th>M.C.H.</th>
<th>Toluene</th>
</tr>
</thead>
<tbody>
<tr>
<td>formula</td>
<td>(C_7H_14)</td>
<td>(C_7H_8)</td>
</tr>
<tr>
<td>Molecular Weight</td>
<td>98.2</td>
<td>92.1</td>
</tr>
<tr>
<td>Density, (g/cm^3) @ 25°C</td>
<td>0.76501</td>
<td>0.8625</td>
</tr>
<tr>
<td>B. pt. @ 760 mm Hg.</td>
<td>100.8</td>
<td>110.7</td>
</tr>
<tr>
<td>Refractive Index (n_D^{20})</td>
<td>1.424(^a) 1.4206(^b)</td>
<td>1.4938(^a) 1.494(^b)</td>
</tr>
<tr>
<td>Heat vaporization (H_V, \text{Btu/lb.}) @ 25°C</td>
<td>154.83</td>
<td>177.28</td>
</tr>
<tr>
<td>@ B.pt.</td>
<td>138.9</td>
<td>156.2</td>
</tr>
<tr>
<td>Viscosity @ 20°C cp.</td>
<td>0.66</td>
<td>0.568</td>
</tr>
</tbody>
</table>

\(^a\): denotes experimental values

\(^b\): denotes literature values
### Table 2.3

Physical Properties of M.C.H./Toluene Binary Mixture at Operating Conditions.

<table>
<thead>
<tr>
<th>Property</th>
<th>M.C.H./Toluene</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>atm. pressure: 760mm Hg</td>
</tr>
<tr>
<td>Liquid density, gm/c.c.</td>
<td>0.7540</td>
</tr>
<tr>
<td>Vapour density, gm/c.c.</td>
<td>0.00302</td>
</tr>
<tr>
<td>Liquid viscosity, cp.</td>
<td>0.2676</td>
</tr>
<tr>
<td>Vapour viscosity, cp.</td>
<td>0.00845</td>
</tr>
<tr>
<td>Liquid diffusivity, $D_L$ sq. cm/sec.</td>
<td>3.69 x 10^{-5}</td>
</tr>
<tr>
<td>Vapour diffusivity, $D_G$ sq. cm/sec.</td>
<td>4.51 x 10^{-2}</td>
</tr>
<tr>
<td>Liquid Schmidt number $(S^L_c)$</td>
<td>96.2</td>
</tr>
<tr>
<td>Vapour Schmidt number $(S^G_c)$</td>
<td>0.621</td>
</tr>
</tbody>
</table>
TABLE (2.4.)
Test Mixture Methylcyclohexane/Toluene

Data

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Reported</td>
<td>Calc’d.</td>
<td>mm.Hg</td>
<td>p D</td>
</tr>
<tr>
<td>0.00</td>
<td>0.00</td>
<td>...</td>
<td>110.600</td>
<td>1.4965</td>
</tr>
<tr>
<td>0.05</td>
<td>0.075</td>
<td>0.076</td>
<td>109.55</td>
<td>1.4916</td>
</tr>
<tr>
<td>0.10</td>
<td>0.143</td>
<td>0.146</td>
<td>108.55</td>
<td>1.4871</td>
</tr>
<tr>
<td>0.15</td>
<td>0.210</td>
<td>0.213</td>
<td>107.65</td>
<td>1.4825</td>
</tr>
<tr>
<td>0.20</td>
<td>0.270</td>
<td>0.273</td>
<td>106.90</td>
<td>1.4782</td>
</tr>
<tr>
<td>0.25</td>
<td>0.326</td>
<td>0.330</td>
<td>106.20</td>
<td>1.4740</td>
</tr>
<tr>
<td>0.30</td>
<td>0.378</td>
<td>0.379</td>
<td>105.60</td>
<td>1.4699</td>
</tr>
<tr>
<td>0.35</td>
<td>0.424</td>
<td>0.426</td>
<td>105.00</td>
<td>1.4660</td>
</tr>
<tr>
<td>0.40</td>
<td>0.470</td>
<td>0.473</td>
<td>104.50</td>
<td>1.4620</td>
</tr>
<tr>
<td>0.45</td>
<td>0.515</td>
<td>0.519</td>
<td>104.00</td>
<td>1.4582</td>
</tr>
<tr>
<td>0.50</td>
<td>0.560</td>
<td>0.564</td>
<td>103.55</td>
<td>1.4544</td>
</tr>
<tr>
<td>0.55</td>
<td>0.604</td>
<td>0.608</td>
<td>103.15</td>
<td>1.4509</td>
</tr>
<tr>
<td>0.60</td>
<td>0.650</td>
<td>0.654</td>
<td>102.75</td>
<td>1.4474</td>
</tr>
<tr>
<td>0.65</td>
<td>0.694</td>
<td>0.697</td>
<td>102.45</td>
<td>1.4440</td>
</tr>
<tr>
<td>0.70</td>
<td>0.737</td>
<td>0.738</td>
<td>102.15</td>
<td>1.4408</td>
</tr>
<tr>
<td>0.75</td>
<td>0.778</td>
<td>0.778</td>
<td>101.90</td>
<td>1.4376</td>
</tr>
<tr>
<td>0.80</td>
<td>0.818</td>
<td>0.820</td>
<td>101.65</td>
<td>1.4345</td>
</tr>
<tr>
<td>0.85</td>
<td>0.860</td>
<td>0.863</td>
<td>101.40</td>
<td>1.4315</td>
</tr>
<tr>
<td>0.90</td>
<td>0.906</td>
<td>0.909</td>
<td>101.20</td>
<td>1.4286</td>
</tr>
<tr>
<td>0.95</td>
<td>0.954</td>
<td>0.955</td>
<td>101.00</td>
<td>1.4260</td>
</tr>
<tr>
<td>1.00</td>
<td>1.00</td>
<td>...</td>
<td>100.85</td>
<td>1.4235</td>
</tr>
</tbody>
</table>
2.4. Experimental Procedure

2.4.1. Method of Operation

The column was designed to operate under total reflux conditions. This was adopted because in a batch type distillation column, steady state could be attained only at total reflux and also this simplified the calculations greatly. Before beginning a series of runs, the still was charged with about 2 litres of mixture of methylcyclohexane/toluene of a known composition (about 40 mole % M.C.H.). The equipment was started by switching on the heating of the reboiler and the jacket surrounding the column. Cooling water flowing into the condenser was adjusted at a rate of 30 galls/hr. approximately and also the boil-up rate was adjusted using the variac.

Preliminary runs in this column indicated that steady state conditions were reached after about 2½ hrs. and sufficient time was allowed for each run, about 3 hrs., before product withdrawal was started. After the column had reached steady-state conditions, samples were drawn from the column from the top and bottom in pre-cooled sample bottles. The method of sampling adopted throughout was to draw a small liquid sample (about 2 c.c.) from the base first and then from the top, because while the top sample is being withdrawn, no or very little reflux flows down the column and the equilibrium would be disturbed. Also, it was thought desirable to withdraw samples at slow rate so as not to disturb the composition and to obtain reproducible results. Two samples were taken, and if their relative indices differed by more than ± 0.0002,
more time was allowed for equilibrium to be established and other samples were withdrawn until this criterion was reached on two consecutive samples.

Measurements were made of the load of the column with liquid by noting the time, by means of a stop watch, necessary to fill a volume of 30 c.c. of reflux through a stop-cock incorporated with the reflux meter as described previously in Section (2.1.6.).

The samples for analysis were always taken before the control measurement of the load in order to avoid possible error resulting from disturbance of the interphase equilibrium. The residue samples were not withdrawn from the reboiler, but from the base of the column (above the reboiler), since errors might have been introduced due to concentration gradients, and also to avoid making any assumptions about the type and operation of the reboiler.

The method of operating the column under pulsation of the gas phase was, as above, except that the pulsing unit, previously set for the desired amplitude and frequency, was switched on and after allowing sufficient period for equilibrium conditions to be attained, withdrawal of samples from the top and the base of the column was conducted as in the series of runs without pulsations.

In each series of tests, side by side with the pulsation experiments, one comparative measurement was made without pulsation, at the same or almost the same loading of the column.
2.4.2. **Efficiency Determinations**

2.4.2a **Equilibrium Time**

At the beginning of any fractional distillation, it is necessary for the column to run for some period under total reflux conditions before attempting to withdraw samples of product for analysis, in order that the essential concentration gradient may be built up and a steady-state reached.

Coulson (108), gives the following equation for estimation of the equilibrium time necessary, \( T_E \):

\[
T_E = \left( \frac{H}{V} \right) \frac{1}{x_s \left[ \frac{\alpha}{\{1 + (\alpha - 1)x_s\}} - 1 \right]^n} \]

\[
\left[ \frac{\log_{10} \left( \frac{(1 + (\alpha^n - 1)x_s/\{1 + (\alpha - 1)x_s\})}{n\log_{10}\alpha} \right)}{1 + (\alpha - 1)x_s} \right] \]

and as it can be seen \( T_E \) is proportional to the holdup \( H \), inversely proportional to the boil-up rate, \( V \) and the number of theoretical plates \( n \), and the concentration of the more volatile component in the still \( x_s \) at equilibrium and at total reflux. As a rough guide, however, it was suggested that in most cases, an equilibration time amounting to between 1 and \( 1\frac{1}{2} \) hours for every 10 theoretical plates is necessary.
The method used here is to run the column for a sufficient period as to establish steady state conditions, about 3 hours as determined from preliminary runs when it was observed that no change in the composition of the top sample product occurred after this period. A typical run showing an estimate of the time required to reach constant composition of the product at the top of the column is shown in Figure (2.11.) when pulsations are applied. As follows from the course of the curve, the operating conditions are taken to be stabilized after about 2 hours, and this method is adopted for other experimental runs under different operating conditions. The superficial column vapour velocity at the base of the column was calculated from the liquid reflux rate, column temperature near the bottom of the column, the liquid density, M.W. of the binary mixture, the cross-sectional area of the annulus and the reflux composition.

A better estimation would be if the relative gas velocity was taken relative to an effective liquid surface velocity $v_L$ as given by:

$$ v = v_g + v_L $$

when $v_g$ is the gas velocity, i.e. the volumetric flow rate divided by the cross-section and corrected for the volume occupied by the liquid film on the tube wall.
Figure 2.11.
Rate of Establishment of Equilibrium Conditions with Pulsations.

\[ f = 10 \text{ c/s} \]
\[ A = 6 \text{ cm Hg} \]
Reflux rate = 80 cc/min.

No pulsation

Start pulsation

2 hours

Refactive Index (Top Product)
2.4.2b Determination of Plate/N.T.U. Equivalence

To obtain a quantitative estimation of the performance of the wetted-wall column under study, the concepts of theoretical plates and transfer units have been used. These have been adequately described in texts and in the original article by Chilton and Colburn (109)

Two methods have been used for the determination of the efficiency:

i) Graphical

The method of McCabe-Thiele (110) was used to determine the number of theoretical plates at total reflux. A chart of refractive index versus number of theoretical plates was prepared for use with this system at 760 mm. Hg pressure. This is shown in Figure (2.12.) and was expanded to ensure greater accuracy in determining the number of stages.

Another method was to use the number of transfer units, \( N_{OG} \), as calculated by the familiar Chilton-Colburn equation (2.11.) and estimation of the above method was performed graphically with the use of a planimeter and represented in Figure (2.10.) These values were divided into the active height of the tower to give the \( H_{OG} \) values (H.T.U.). The average slope of the equilibrium curve for this system (H) was also evaluated from the relation:-
Figure 2.12.

N.T.P. VS. Refractive Index
Methylcyclohexane/Toluene
© Total reflux
and this is shown in Figure (2.13.) and found to be equal to 0.930. The value of $m$ reported in literature was 1.0495.

ii) **Calculation**

Fenske and Underwood (111) have developed an equation for determining the number of theoretical plates existing in a column under total reflux for constant relative volatility ($\alpha$) and given by:

$$n + 1 = \frac{\log \left( \frac{x_A}{x_B} \right) \left( \frac{y_A}{y_B} \right)}{\log \alpha_{AB}} \quad \cdots \cdots \cdots (2.16)$$

where:

- $n$ = number of theoretical plates in the column.
- $x_A$: mole fraction of A in liquid
- $y_A$: mole fraction of A in vapour
- $x_B$: mole fraction of B in liquid
- $y_B$: mole fraction of B in vapour

and $\alpha_{AB}$ is the geometric average of the relative volatilities relating to the extreme compositions at the top and base of the column during operation. Figure (2.7.) represents the behaviour of the relative volatility for the system used with composition.
Figure 2.13.
Estimation of \( m \): Average slope of equilibrium curve.
A value of (1.3) for the geometric average of the volatility was used for the experimental composition range.

The value of \( \lambda \) calculated by the relationship (for ideal mixtures):

\[
\log_{10} \frac{P_A}{P_B} = \left( \frac{T_{B1} - T_{A1}}{T_{A1} + T_{B1}} \right) \left\{ \frac{T_{A2}}{T_{A1} - T_{A2}} + \frac{T_{B2}}{T_{B1} - T_{B2}} \right\} \log_{10} \frac{P_1}{P_2}
\]

\[ ............ (2.17) \]

gives, \( \lambda = 1.34 \).

This differed by 4\% of the value used and it was decided to use the value = 1.3 as tolerated within the accuracy of the experiments.

where:

\[ T_{A1}, T_{B1} = B.pt. \text{ of components, } A, B \text{ at the higher pressure } P_1 \]

\[ T_{A2}, T_{B2} = B.pt. \text{ of components } A, B \text{ at the lower pressure } P_2, ^\circ \text{K} \]

Also, evaluation of the number of transfer units \((N_{OG})\) based on the gas phase was calculated from the standard equation:
\[ N_{OG} = \frac{1}{\alpha - 1} \ln \frac{Y_t(1-y_b)}{y_b(1-Y_t)} + \ln \frac{(1-y_b)}{(1-Y_t)} \] \quad \ldots \ldots (2.18)

and the height of the overall transfer unit was calculated from \( H_{OG} = Z / N_{OG} \), where \( Z \) is the total rectifying length.

2.4.3. Difficulties encountered in operation

A number of difficulties were encountered during the course of the investigation. One of the major difficulties was to achieve uniform boiling of the mixture methylcyclohexane/toluene in the reboiler. It was found necessary to introduce "boiling chips", which were small pieces of earthenware, supported on a circular brass gauze located in the lower section of the reboiler. This method produced some improvement as regards to the quantity of boiling; and the reboiler was insulated with a layer of aluminium over which the heating tape was wound. A flask with circulation heating to promote good mixing and prevent local superheating of the contents would have been favourable, but the former system was chosen because of availability.

The other difficulty was the tendency of the silicone grease to be lost to the reboiler through the 3-way valve. This was replaced with non-aq. grease which also eliminated the possibility of vapour leaks. Precautions were taken during sampling to ensure good reproducibility of the results during a run. The sampling arrangement was modified after preliminary runs and allowed a shorter length of tube between the take-off flange and the sampling point.
and it was considered good practice to discard the first 2-c.c. of sample so that the liquid was actually the reflux which was being formed and not the liquid hold-up in the tube. Removal of the sample was conducted at a slow rate so that the composition at the top of the column would not be disturbed.

2.4.4. **Accuracy of the results**

The accuracy of the Analysis which could be achieved by the Abbe refractometer was ± 0.0002, and the results of the sample composition were reproducible to the same degree of accuracy. For the non-pulsed results, the spread between the refractive indices of the top and bottom product was about 0.007. Therefore, it was possible to make an error of 2 per cent and still be within the accuracy of the refractometer. With the application of pulsations, the accuracy was increased, since the spread of the refractive indices of the two samples was increased (0.020).

The graphical method obtained by Figure (2.10.) was conveniently selected to determine the overall number of transfer units (N.T.U.). The accuracy of this method, when done on expanded graph paper was estimated to be of the order 2.3%. Tests were performed to evaluate the error involved in the determination of the reflux rate (vapour rate) and this was found to be not more than ± 3%.
SECTION III

RESULTS
### SECTION III

<table>
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<tr>
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<td>3.2.4. Results of third series of runs 2&quot; inner tube column.</td>
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3.1. **RESULTS OF THE NON-PULSED COLUMNS**

A series of runs were initially conducted at total reflux without the application of forced pulsations, at atmospheric pressure. Three columns, of different internal dimensions, as detailed in Table (2.1.), section (2), were investigated, and the performance evaluated at different vapour velocities, based on the empty Annular cross-sectional area. The majority of the runs were performed in the range of the flow of vapour (1100 < Rev < 2500).

After conducting preliminary runs, it was observed that the maximum and minimum liquid reflux rates, to give reproducible and stable operation of the columns were in the range of 50 and 120 c.c./min. respectively. Beyond this range, the operation of the column was considered to depend on factors attributed to the limitations on the operation of the still. Non-uniform boiling was encountered at the higher and lower throughputs and steps were taken to minimise this by introducing 'porous pot' in the boiling mixture. This difficulty was attributed to the design and method used for heating the still.

The experimental data were analysed in the form of (N.T.U.), calculated using equation (2.18) and the overall height of the transfer unit was obtained by simply dividing the height of the effective rectification unit (length between top and bottom sampling points) by the number of transfer units developed.
The results obtained from these runs are represented in Table (3.1.) for the 1 - in., inner tube column. The performance of the column, expressed as N.T.U. is plotted as a function of the Reynolds Number of the vapour phase ($Re_v$), is illustrated in Figure (3.1.). Similarly, Table (3.2.) and Table (3.3.) are given for the results obtained for the 1½ - in. and 2 - in inner tube columns respectively under similar operating conditions and Figure (3.2.) and Figure (3.3.) illustrate the variation of the performance of these two columns with ($Re_v$) over the permissable operating range.

The efficiency data obtained for the three columns would be considered in more detail in Section (4.1.) and fitted whenever possible with previously established relationships described by other workers for wetted-wall or similar rectification columns.
### TABLE (3.1.)

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Figure 3.1: Variation of N.T.U. VS. Rev.

1 in. inner column.
NO PULSATIONS.
Figure 3.2.
Variation of N.T.U. VS. Rev.

1 1/2 in. inner column.
NO PULSATIONS.

Reynolds Number x 10^2
(Rev.)
Figure 3.3.
Variation of N.T.U. vs. Rev.

2 in. inner column.
NO PULSATIONS.

Reynolds Number $x10^2$
(Rev.)
3.2.1. RESULTS OF INITIAL PULSED RUNS

During the initial stage of this study, it was the main objective to find the qualitative effect of pulsations on the efficiency of the column as measured by the number of theoretical plates (N.T.P.).

The N.T.P. was determined using the graphical method of McCabe and Thiele(110) and it was found that the (N.T.P.) obtained with pulsations was about 300% more than without pulsations. The selected experimental data and principal results are summarized in Table (3.4.). It was also observed that the (N.T.P.) is increased when running the column at a lower boil-up rate, but the per cent increase in (N.T.P.) for pulsating at this lower boil-up rate is practically the same.

Since it was unlikely that the conditions for maximum increase in efficiency were being used in this preliminary work, the next step was to investigate quantitatively the effect of the variables. It should then be possible to predict the maximum possible increase in efficiency and the operating conditions required to establish the optimum.
### TABLE (3.4.)

**INITIAL PULSATION RESULTS**

1" ANNULAR COLUMN

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<th>CONDITION</th>
<th>Refractive Index at 20°C (Equilibrium)</th>
<th>Boil-up Rate</th>
<th>Velocity cm/sec.</th>
<th>No. of Theoretical Stages</th>
<th>H.E.T.P. (in)</th>
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<td>Top</td>
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<td>c.c./min. lb moles/sec. per sq. ft. of Annular area</td>
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</table>
3.2.2. RESULTS OF FIRST SERIES OF RUNS (AP)

1st. INNER TUBE COLUMN

After the performance of the three columns had been investigated without the effect of pulsations for a range of boil-up rates, an experimental programme was devised in order to evaluate the effect of pulsing the vapour phase on the efficiency of the columns. It was generally established, from previous preliminary runs, Section (3.2.1.), that the efficiency of the column was considerably higher with pulsations than without.

The results of the first series of runs (AP), pertaining to the 1-in. inner tube column, are represented in Table (3.5.), summarizing the principal calculated values of (N.T.U.) characterizing the fractionating capacity of the column. Among the factors that were taken into consideration was the effect of the vapour velocity (reflux rate), and for every run, this was varied between the range of 10 -22 cm/sec. for a fixed amplitude and frequency of pulsations. Figures (3.(4-14)) show the effect of variation of reflux rate on the rectification efficiency at various frequencies and amplitudes of pulsations. This was plotted as N.T.U. v.s. Reynolds number of the vapour phase ($R_{ev}$). The effects of frequency and amplitude of pulsations were studied in more detail. Frequencies of 2, 4, 7, 10, 17 and 21 c/s were chosen to cover so wide range as was possible and for each run the efficiency of the column was determined at four vapour velocities for a fixed amplitude. This is shown
in Figures (3.4-8).

Inspection of these figures reveal an increase in efficiency with an increase in frequency and that an apparent optimal range of increased efficiency exists at a frequency of about 17 c/s.

An investigation of the effect of the pulsation amplitude on the resultant performance was also undertaken and (Figures 3.9-14) show that an increase in Amplitude results in a definite increase in efficiency of the column. At the higher frequencies and amplitudes, the influence in the variation of the reflux rate is less marked than at the lower values.

3.2.3. RESULTS OF SECOND SERIES OF RUNS - (BP)

1\frac{1}{2}" - INNER TUBE COLUMN

After the completion of the first series of runs (AP) with the 1-in, annular tube column, the column was dismantled, cleaned and polished thoroughly and modified to be replaced by the 1\frac{1}{2}-in. o.d. inner brass tube, as described in the experimental section (II).

The performance of this column without the application of pulsations was investigated for a range of throughputs and these were followed by 36 runs with pulsations to study the effects of pulsation parameters on the efficiency. It was found useful to repeat a run deliberately since it was found difficult to reproduce the parameter of vapour velocity from run to run.
The results of these runs are presented in Table (3.6.) and these were obtained in the range of vapour velocity varying between 17 - 36 cm/sec. The results are plotted in Figures (3. (15-25)) as (N.T.U.) vs. ($R_{ev}$). The results without pulsations were included for comparison and it was observed that in most cases the efficiency of this column is considerably higher with pulsations than without.

Figures (3. (15-19)) show the variation of the overall rectification efficiency (N.T.U.) with frequency of pulsations at various amplitudes for the range of reflux rates investigated. By inspection, the performance of the column increases rapidly with the frequency of pulsations and then drops beyond frequency value of the order 17 c/s. A reversal of the increased trend in efficiency is possible for the higher frequencies. This, as before, indicates an existence of an optimum region of operation.

Figures (3. (20-25)) show that an increase in performance can be obtained with increasing the amplitude of pulsations and that this increase is directly proportional to the amplitude of the pulsations.

3.2.4. RESULTS OF THIRD SERIES OF RUNS

2" - INNER TUBE COLUMN

A series of five group of runs were made using the 2-in. annular tube column and were performed for identical conditions as for (J.P) and (BP) series of runs.
These are represented in Table (3.7.). The runs were performed before any attempt was made at interpreting the results at this stage. The range of vapour velocity was varied between 50 - 110 cm/sec. When varying the pulsation parameters, (frequency and amplitude), it was observed, unlike with the two columns studied previously, that the amplitude parameter decreased with increasing frequency of pulsations. A detailed examination of the column revealed a 'bulge' on the inside wall of the outer tube, thus creating an obstruction for the pulsations effect to propagate along the full length of the column. This observation pointed a finger of suspicion as to the reliability of these data so far obtained, and it was later decided to discard them and to discontinue work on this column. This discovery could also provide an explanation for the reason of disagreement of the 2-in. column with the expression correlating the data obtained for the 1" - and 1 1/2" inner tube columns.
## RESULTS WITH PULSATIONS

### TABLE (3.5.)

**AP - SERIES EXPERIMENTAL DATA**

**1-in. INNER TUBE COLUMN**

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<th>Run No</th>
<th>Velocity cm/sec.</th>
<th>Composition mole %</th>
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<th>Still</th>
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<th>(N.T.U) P</th>
<th>(N.T.U) o = R</th>
<th>(Rev. Reynolds No.)</th>
<th>Frequency C/S</th>
<th>Amplitude (cm · Hg)</th>
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**BP - SERIES EXPERIMENTAL DATA**

**13-in. INNER TUBE COLUMN**

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### TABLE (3.7.)

**CP - EXPERIMENTAL D.T. SERIES**

**2-in INNER TUBE COLUMN**

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**Note:** The table contains data on velocity, composition, number of transfer units (N.T.U), Reynolds number, and frequency. The data is organized in a column format with specific values for each run number.
Figure 3.4.
Effect of pulse frequency on Efficiency.

1 in. inner column.

\[ A = 2 \text{ cm. Hg.} \]

- \( 2 \text{ c/s} \)
- \( 4 \text{ c/s} \)
- NO PULSATIONS.

Reynolds Number (Rev.) \( \times 10^2 \)
Figure 3.5.

Effect of Pulse Frequency on Efficiency.

1 in. inner column.

A = 4 cm. Hg.

Frequency

- 4 c/s
- 7 c/s
- 10 c/s
- 17 c/s
- 21 c/s
- NO PULSATIONS.

Reynolds Number (Rev.) x 10^2
Figure 3.6.
Effect of Pulse Frequency on Efficiency.
Figure 3.7. Effect of Pulse Frequency on Efficiency.

- 1 in. inner column.
- $A = 8 \text{ cm. Hz.}$
- $f =$ frequency
- $\bigcirc -$ 17 c/s
- $\Box -$ No pulsations.

Number of Transfer Units (N.T.U.)

Reynolds Number (Rev.) $\times 10^2$
Figure 3.8. 
Effect of Pulse Frequency on Efficiency.

1 in. inner column. 
A = 9 cm. Hg. frequency 
- 21 c/s
- No pulsations.

Reynolds Number (Rev.) \( \times 10^2 \)
Figure 3.9.
Effect of Pulse Amplitude on Efficiency.

1 in. inner column.

\[ f = 2 \text{ cycles/sec.} \]

Amplitude

\[ \circ \quad 2 \text{ cm. Hg.} \]

\[ \square \quad \text{NO PULSATIONS.} \]
Figure 3.10. Effect of Pulse Amplitude on Efficiency.

1 in. inner column.

\( f = 4 \text{ c/s.} \)

Amplitude

- \( \triangle \) 4 cm Hg.
- \( \bigcirc \) 2 cm Hg.
- \( \square \) No pulsation.
Figure 3.11.
Effect of Pulse Amplitude on Efficiency.

1 in. inner column.

$f = 7$ c/s.

Amplitude

- 4 cm Hg.
- 6 cm Hg.
- No pulsation.

Reynold Number (Rev.) $\times 10^2$
Figure 3.12.

Effect of Pulse Amplitude on Efficiency.

- 1 in. inner column.
- $f = 10 \text{ c/s.}$
- $f = 10 \text{ c/s.}$
- Amplitude
- $\bigcirc$ - 4 cm Hg.
- $\triangle$ - 6 cm Hg.
- $\square$ - No pulsations.
Figure 3.13.
Effect of Pulse Amplitude on Efficiency.

1 in. inner column

$f = 17$ c/s

Amplitude

- $4$ cm Hg.
- $6$ cm Hg.
- $8$ cm Hg.
- No pulsations.

Reynolds Number (Rev.)
$x10^2$
1 in. inner column.

\[ f = 21 \text{ c/s.} \]

Amplitude

- 4 cm Hg.
- 6 cm Hg.
- 9 cm Hg.
- No pulsations.

Figure 3.14.

Effect of pulse Amplitude on Efficiency.
Figure 3.15.
Effect of Pulse Frequency on Efficiency.

1½ in. inner column
A = 2 cm Hg.
Frequency

- 2 c/s
- 4 c/s
- No pulsations.

Reynolds Number (Rev.) x 10²
1½ in inner column.

A = 4 cm Hg.

Frequency

• 4 c/s
△ 7 c/s
● 10 c/s
○ 17 c/s
■ 21 c/s
□ No pulsations.

Figure 3.16.

Effect of Pulse Frequency on Efficiency.
Figure 3.17.
Effect of Frequency on Efficiency.
Figure 3.18.
Effect of pulse frequency on Efficiency.

$1\frac{1}{2}$" inner column

$A = \frac{9 \text{ cm Hg}}{\text{Frequency}}$

$\circ - 17 \text{ c/s}$

$\Box - \text{No pulsations}$
Figure 3.19. Effect of frequency on N.T.U.
Figure 3.20.
Effect of pulse Amplitude on Efficiency.

1\(\frac{1}{2}\) in. inner column

\(f = 2\) c/s.

Amplitude

○ 2 cm Hg.

□ No pulsations

Number of Transfer Units (N.T.U.)

Reynolds Number (Rev.)

x10^2
Figure 3.21.
Effect of pulse amplitude on efficiency.
Figure 3.22.
Effect of pulse Amplitude on Efficiency.
Figure 3.23. Effect of pulse amplitude on efficiency.

1\frac{1}{2} \text{ in. inner column}

\frac{f}{10 \text{ c/s.}}

\text{Amplitude}

- 4 \text{ cm Hg.}
- 6 \text{ cm Hg.}
- No pulsations

Number of Transfer Units (M.I.U.)

Reynolds Number (Rev.) \times 10^2

12 14 16 18 20 22 24 26 28
Figure 3.24.
Effect of Pulse Amplitude on Efficiency.
Figure 3.25.
Effect of Pulse Amplitude on Efficiency.

1\frac{1}{2} in. inner column

f = 21 c/s.

Amplitude

○ 4 cm Hg.
△ 6 cm Hg.
● 10 cm Hg.
□ No pulsations.
SECTION IV

DISCUSSION
SECTION IV

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4.1. **CORRELATION OF NON-PULSED DATA**

The variation of (N.T.U.) with the reflux rate in the case of the non-pulsed operation was investigated for the three columns: 1", 1½" and 2" inner tube columns and designated by series (A), (B) and (C) runs respectively. The overall height of the transfer unit was obtained by simply dividing the height of the effective rectification unit (length between top and base sampling points) by the number of transfer units developed.

As was expected, the efficiency of the column, in all cases, decreased with increasing vapour flow rates, for \( R_{ev} > 1100 \), below which value, the operation of the column was considered to be unstable. As indicated in Figure (3.1) the efficiency dropped by a value of 18%, in the range 1200 > \( R_{ev} \leq 2600 \), for series (A) results. The corresponding value for the (B) column was 14% and similarly for (C) column a decrease in efficiency of 23% was obtained. The results for the latter column were somewhat scattered and were treated with some reservation at this point. Other research workers, studying concentric tube rectification columns reported similar trend of the performance, and for the system used assumed that the overall phase resistance lies mainly in the vapour phase. In order to determine the validity of this assumption and to determine whether there is any resistance to mass transfer in the liquid side, the values of the overall resistance, \( \frac{1}{KOG} \), and given by the expression:
\[ \frac{1}{K_{OG}} = \frac{4 HOG/v (d-d_o)}{\ldots \ldots \ldots (4.1.)} \]

where:

- \( d \) : diameter of outer tube; cm.
- \( d_o \) : diameter of inner tube; cm.
- \( v \) : velocity of vapour, based on empty column, cm/sec.

1 values were calculated and plotted against \( \frac{1}{v^{0.8}} \), assuming \( K_G \propto v^{0.8} \) and \( K_L \) is independent of the gas velocity. The data, when plotted, were represented by a straight line passing nearly through the origin as shown in Figure (4.1.), indicating according to equation (1.11), due to Colburn, that most of the resistance lies in the vapour film under normal conditions of operation and that the resistance to mass transfer in the liquid film was assumed to be negligible so that,

\[ (H.T.U.)_{OG} = (H.T.U.)_G \quad \ldots \ldots (4.2.) \]

The measured performance of the three columns, expressed in terms of the overall height of a transfer unit (HOG), is shown in Figure (4.2.). The effect of the effective diameter possible width = \( d - d_o \), on the efficiency of the three columns is seen to be marked. The efficiency of open-tube columns, assuming negligible liquid-phase resistance has been shown in equation (1.8) to be inversely proportional to the tube diameter, i.e.:

\[ N_{OG} \propto d^{-1} \text{ or } H_{OG} \propto d \quad \ldots \ldots (4.3) \]

The experimental data were further plotted in terms of \( (HOG/d - d_o) \) versus vapour Reynolds number (Rev).
Figure 4.1. Variation of $\frac{1}{Kog}$ vs. $\frac{1}{V^{0.5}}$

A - 1" inner tube column
B - 1.5" inner tube column
C - 2" inner tube column

NO PULSATIONS

$\frac{1}{Kog}$ vs. $\frac{1}{V^{0.5}}$
Figure 4.2.
Effect of inner tube size of column on efficiency.

A - 1" inner tube column
B - 1\(\frac{1}{2}"\) " " "
C - 2\(\frac{1}{2}"\) " " "

NO PULSATIONS

Height of Transfer Unit, Hug (cms)

Reynolds Number (Rev.) \(x10^2\)
as shown in Figure (4.3.), and it was observed that although there was some scatter, especially for the data pertaining to (Group C) runs, the slopes of the three lines plotted on logarithmic scale was approximately the same and corresponds to an exponent on the Reynolds number of (0.24) especially for groups (A) and (B) runs. This would probably suggest, that in this work, turbulent conditions developed and prevail in the gas phase since vapour in the system considered, was abruptly introduced to the column, which provided no lower calming section and furthermore, the inner tubes, blocked from the lower end and perforated with two rows of \( \frac{3}{4} \) -in. diameter holes probably acted as a turbulence promoter. The fact that the data in Figure (4.3.) did not fall on a single straight line further suggests the function of the width of the annular space to be of importance on the efficiency.

On the basis of the information so far obtained, it was decided to find an expression correlating the data for the three columns and using assumptions introduced by other workers describing the operation of open-tube distillation columns, at total reflux. The power of the Schmidt number (Sc) was taken as (0.67), due to that of Gilliland equation (1.3.), and the physical characteristics of the liquid and vapour mixture during rectification were assumed to be constant and liquid film resistance was assumed negligible. It was found that the data could be best represented by the following equation:

\[
HOG = 10.26 \cdot (d-d_0)^{0.64} \cdot R_{ev}^{0.24} \cdot Sc^{0.67}
\]  

(4.4.)

where the constants were obtained using the Rosenbrock optimisation technique to minimise the sum of the squares of the difference between HOG (calculated) and HOG (observed).
Figure 4.3.
Plot of Hog/d-do vs. Rev.
No Pulsations
Tables (4.1-3.) show the results as calculated by expression (4.4) and the corresponding experimentally obtained data and Figure (4.4.) is a plot of $(\frac{HOG}{(d-d_0)0.64})$ versus $(R_{ev})$ for the three columns. It would appear that the correlation (4.4.) relates the results (A) and (B) groups satisfactorily, with little scatter. However the results pertaining to the 2-in., inner tube Column (C group), deviated grossly from the correlation. This is believed to be due, as was discovered later, to the existence of a 'bulge' in the inner tube, when pulsations were applied creating eccentricity and misalignment of the two surfaces. Contact of the inner and outer tubes due to 'bowing' resulted in a partial interruption of the flow of the reflux over the inside wall of the outer tube, thus giving rise to the formation (at points of contact), of 'rivulets' of liquid reflux. The reflux is made to course down in the form of thickened streams, instead of a thin, uniform film; a condition which is unfavourable for bringing about the desired intimacy of contact between liquid and vapour.
### TABLE (4.1.)

**SUMMARY OF RESULTS - NON-PULSED COLUMNS**

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## TABLE (4.2.)

### SUMMARY OF RESULTS - NON-PULSED COLUMN

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### TABLE (4.3.)

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Equation (4.8)

Figure 4.4.
Correlation of Efficiency Results
4.2. EFFECTS OF PULSATION PARAMETERS ON PERFORMANCE

4.2.1. INTRODUCTION - Preliminary results

Preliminary runs were initiated on the 1"-inner tube column to determine the effects of pulsations on the performance and also to gain some idea on the rate of establishment of the operating conditions in a column working with pulsating flow of the gaseous phase. Table (3.4.), Section (3.2.1.) shows the beneficial enhancement obtained by the use of pulsations. An increase by a factor of (3) was obtained using pulsations of fixed frequency of 15 c/s. It was then decided to investigate the performance for the lower range of frequencies and the results obtained are shown in Figure (4.5.). A definite increase was observed and this increased with increasing frequency. The motor driving the pulse generating unit was then replaced with a variable stroke motor to obtain a wider range of frequencies and the performance of the column could be investigated under frequency of pulsations range varying between 0-21 c/s and this was considered sufficient for the purpose of this experiment. The rate of establishment of operating conditions was discussed in Section (2), and it was concluded that the time needed would be between 2-3 hrs. for equilibrium to be established.

4.2.2. THE EFFECT OF FREQUENCY ON COLUMN PERFORMANCE

Pulsation frequencies of 2, 4, 7, 10, 17 and 21 cycles per second were used. For each of the above frequency and at a fixed amplitude, the efficiency of the Column was determined for four vapour flow rates between
Figure 4.5.
Effect of pulsation on Efficiency

1" tube
\(A = 2 \text{ cm. Hg}\)
frequency

- 2 c/s
- 4 c/s
- 6 c/s
- No pulsations
the range of 10-22 cm/sec., (11,000 < Rev < 25,000), as reported in Table (3.5.) section (III).

Figure (4.6.) shows the variation of performance of the Column plotted as (N.T.U.) vs frequency, with amplitude being the parameter, at a fixed vapour flow rate \( (R_{ev} = 1700) \). A family of curves was obtained, and the general pattern was that, at all frequencies investigated, an increase in efficiency over the non-pulsed values, rises with increasing frequency to a region of optimal value and thereafter decreasing with further increase in frequency. The maximum increase obtained for amplitude of 6 cm. Hg. was 2.10 and for \( A = 4 \), the corresponding increase was 1.63. There exists, therefore, an optimum frequency for a fixed amplitude and vapour flowrate of a known system.

Similarly, the efficiency of the \( \frac{3}{4} '' \)-inner tube column was also investigated in the range of vapour velocity of 18-35 cm/sec. The data are tabulated in Table (3.6.) Section (III), and figure (4.7.) is a plot of (N.T.U.) versus frequency of pulsations at a fixed \( (R_{ev} = 1700) \) and the amplitude being the parameter. It was observed that the performance of the column was significantly improved and that the important feature about the variation is a region of optimal value at a frequency of about 17 c/s. as was found for the 1-in. Column. However, the maximum increase obtained for this column for amplitude of 6 cm. Hg. was 2.67 and for \( A = 4 \), the increase was 2.0.
Figure 4.6.
Effect of frequency on Performance
Figure 4.7.
Effect of frequency on Performance
4.2.3. **THE EFFECT OF AMPLITUDE ON COLUMN PERFORMANCE**

Figure (4.8.) shows the variation of (N.T.U.) versus amplitude of pulsations, at a fixed vapour flow rate with the frequency being the parameter. The amplitude used ranged from 0 - 10 cm Hg. and the data used were taken from Table (3.5.), for the 1"-column.

The plot indicates that in the region of amplitudes investigated, the increase in efficiency is directly proportional to the amplitude of pulsations.

Figure (4.9.) shows a similar plot, but for the 1½" column, and the data were taken from Table (3.6.). The increase in the efficiency is observed with increasing amplitude for a fixed frequency and from the plot the increase in the steepness of the slopes of the lines in the range of frequencies $0 < f < 17$ c/s. indicates an increase in the same direction of the efficiency as was also inferred from Figures (4.6-7).

4.2.4. **THE EFFECT OF VAPOUR VELOCITY ON PERFORMANCE**

Figures [4.(10-12)] represent the data taken from Table (3.5.) relating to the 1"-inner column. These were plotted as (R) versus Reynolds Number of the vapour phase ($Re_v$) with the frequency of pulsations as parameter. It is shown that (R) values for the pulsed Column do not change significantly over a range of vapour flow rate, especially for the higher intensity of pulsations. For the lower frequencies $0 \leq f \leq 6$, the efficiency of the pulsed column is not entirely uninfluenced by the vapour flow rate, but the effect is smaller than in the non-pulsed
Column where efficiency decreases with increase in flow rate.

Data from Table (3.6.) for the 1\(\frac{1}{2}\)" inner column were plotted in Figures [4.(13-15)] to show the variation of (R) versus (Rev). As above, this column displayed a similar behaviour, and it was thought valid to conclude that the effect of the vapour flow rate on the per cent increase in efficiency of the pulsed columns is small and would not be taken into consideration in the overall correlation of the results.

4.3. CORRELATION OF PULSED COLUMNS DATA

The amplitude-frequency (Af) product represents a useful, simple means of correlating the pulsing conditions with column performance. A more exact analysis of the data, however, indicates the need for additional considerations. One of these is that the variation of frequency has effects not fully accounted for by the corresponding (Af) variation. This is apparent in Figures (4.7.) and Figure (4.8.) which show that there exists a maximum value of efficiency at approximately 17 c/s, after which frequency, the efficiency begins to decrease with increasing pulsations. A plot of (N.T.U.) versus (Af), from the data presented in Table (3.5.) revealed a poor correlation and therefore the need for another refinement of the Amplitude - frequency product was attempted.
Figure 4.8.
Effect of Amplitude on Performance

1 in. tube column
Rev. = 1700
frequency
•— 4 c/s
△— 7 c/s
•— 10 c/s
○— 17 c/s
□— 21 c/s
Figure 4.9.
Effect of Amplitude on Performance
Figure 4.10.
Variation of R vs. Rev.

1 in. tube column
Amplitude = 2 cm Hg.
Frequency
● 2 c/s
△ 4 c/s
Figure 4.11.
Variation of R vs. Rev.
Figure 4.12.
Variation of $R$ vs. $\text{Rev.}$

1 in. tube column

$A = 6 \text{ cm Hg.}$

- $7 \text{ c/s}$
- $10 \text{ c/s}$
- $17 \text{ c/s}$
- $21 \text{ c/s}$

Reynolds Number (Rev.) $\times 10^2$
Figure 4.13.
Variation of $R$ vs. $\text{Rev.}$

$1\frac{1}{2}$ in. tube column

$A = 2 \text{ cm Hg.}$

frequency

$\circ - 2 \text{ c/s}$

$\triangle - 4 \text{ c/s}$

Reynolds Number (Rev.)
$x10^2$
Figure 4.14.
Variation of R vs. Rev.

1\frac{1}{2} in. tube column

\[ A = 4 \text{ cm Hg} \]

\[ \text{frequency} \]

\[ \bigcirc \quad 4 \text{ c/s} \]
\[ \triangle \quad 7 \text{ c/s} \]
\[ \square \quad 10 \text{ c/s} \]
\[ \bigcirc \quad 17 \text{ c/s} \]
\[ \bigcirc \quad 21 \text{ c/s} \]

Reynolds Number (Rev.) x10^2
$R = \frac{(N.T.U.)_y}{(N.T.U.)_x}$

**Figure 4.15.**
Variation of $R$ vs. Rev.

- $1\frac{2}{3}$ in tube column
- Amplitude = 6 cm Hg.
- Frequency
  - $7$ c/s
  - $10$ c/s
  - $17$ c/s
  - $21$ c/s

Reynolds Number (Rev.)

$\times 10^2$
The experimental data have indicated for both Columns, that the variations in the vapour flow rate have no significant effect on the ratio of the performance (R). Accordingly data given in Table (4.4.) were used taking the average of (R) for every run and correlated using the expression:

\[
(N_{\text{T.U.}})_p = (N_{\text{T.U.}})_o \left[ 1 + K \Lambda \sin^2 \left( \frac{\pi}{2} \frac{f}{f_0} \right) \right] \quad \ldots \ldots (4.5.)
\]

where \( K \) is constant for the 1-in inner tube column, the following expression was obtained:

\[
R = 1 + 0.189 \Lambda \sin^2 \left( \frac{\pi}{2} \frac{f}{10.59} \right) \quad \ldots \ldots (4.6.)
\]

and for 1\( \frac{3}{4} \)-in. column:

\[
R = 1 + 0.286 \Lambda \sin^2 \left( \frac{\pi}{2} \frac{f}{11.36} \right) \quad \ldots \ldots (4.7.)
\]

The constants were obtained using the Rosenbrock optimisation technique with the aid of a Computer program (see Appendix  ).

It was observed that the ratio of the two Constants 0.189/0.286, is nearly equivalent to the Area/Volume ratio, \((a)\) of the two columns i.e. \(1.26/2.1\).

This would probably suggest that the overall correlation would be represented by the expression:

\[
R = 1 + 0.15 \Lambda \sin^2 \left( \frac{\pi}{2} \frac{f}{f_0} \right) \quad \ldots \ldots (4.8.)
\]

where the value of

\( a = \text{Area/volume ratio of the Column, cm}^{-1} \)

\( f_0 = \text{frequency for optimum efficiency, c/s} \)
This correlation indicates that the increase in efficiency obtained by the use of pulsations is cyclic and has an optimum at frequency in the region of 17 c/s. It would be useful if the validity of this expression could have been investigated for higher pulsations, and systems with different physical properties over a wider range of reflux rates.
4.4. Interpretation of pulsed-columns results

4.4.1. General

The increase in efficiency that has been noted as a result of pulsations is difficult to explain on theoretical basis. In general, there are two ways by which it is possible to increase the efficiency of a distillation process. One of these would be to increase the mass transfer rate between the phases by means which would reduce the resistance to mass transfer at the interface. The other way is to increase the interfacial contact area between the phases.

When the vapour phase in a distillation column is pulsed, the turbulent nature of the flow produced appears to produce an effective vapour diffusion coefficient or "eddy diffusion", reaching as high as ten times that for laminar vapour stream. This turbulence within the phases result in an intensification of mass transfer and of the relative speed at the interface without increasing the mean vapour flow. The overall effect is that the application of pulsations to a distillation column is a practical method for efficiency improvement.

4.4.2. THEORETICAL CONSIDERATIONS IN RELATION TO MECHANISM OF PULSATIONS

In distillation process, pulsations applied to the gas phase will set in motion a flow in the vapour which is continuously accelerated and retarded. Consequently, the pulsations can create turbulence, in the gas, or, through the vapour phase, they may act upon the liquid phase.
Mass and simultaneous heat transfer can take place in the accelerating or retarding period. These periods can be considered as an incipient process in which the interface is not yet completely developed. Under these conditions, the interfacial layer is reduced and the rate of the process is increased.

An important consideration concerns the profile of the velocities and the velocity gradient during one oscillation. Franke (113), studied the profile of air velocities pulsating at frequencies of 5 - 25 Hz in a square-shaped tube. His results are shown in Figure (4.6c), and the graphs represent the variations of mean values of absolute velocities with distance from the walls at different frequencies. In a pulsating flow, contrary to the known profiles for steady-state flows, a ring effect is observed whereby the flow velocity reaches a maximum in the vicinity of the wall and then drops suddenly towards the wall. This phenomenon is due to the variability of the ratio between the forces of inertia and those of friction at a certain distance from the wall. If this ring effect be coupled with the development of the interface layer, it seems that it could be an important factor in contributing for enhancing mass transfer. The interactions between the flow of the vapour phase and the liquid phase must also be considered. A liquid film flowing over a vertical surface and having Reynolds number of Re_L > 16, will form waves on its surface. The presence of waves on the film surface has a mixing effect in this zone even for Reynolds Number
Figure (4.16 A)
Variation of gas velocity in pulsating flow as a function of distance from wall.

Figure (4.16 B)
Possible methods of transferring turbulence across gas - liquid interface.
(i) by formation of ripples.
(ii) by 'friction wheel' mechanism.
values of $Re_L < 16$, and that the more the amplitude and frequency of the waves increase, the more substantial is their effect upon the gas phase.

In the case of pulsating flow, it is possible for the vapour, being homogenous and compressible, to vibrate as a solid elastic system or to get into resonance acting upon the liquid in various ways. From this point of view, the amplitude and frequency of pulsations are of significant importance because they determine the extent of the effect of the gas phase upon the liquid phase.

Creating turbulence in the gas phase with pulsations can explain to a certain extent the increase in the efficiency obtained, but does not seem to justify the existence of a peak. It seemed also possible that there may exist a link or interdependence between the pulsation frequencies and the frequency of the liquid waves, and that at equal frequencies the pulsating vapour phase has a stronger effect upon the liquid reflux film producing a high mixing effect. However Brauer (115) has shown that with the system for an unpulsed falling liquid film the wave frequency increases with the reflux rate, whereas Figure (4.6-7) show a maximum increase at the frequency 17 c/s, for different reflux rates investigated.

It can be concluded, on the basis of visual observations of the liquid reflux having maximum rhythmic vibrations, that the pulsating vapour has the strongest effect on the liquid at approximately a frequency of 17 c/s.
Under these conditions, while the process is taking place, there is a reciprocal effect, which helps to emphasize mass transfer: the gas, by acting upon the liquid film creates turbulence and waves, and these in their turn act upon the vapour phase by amplifying its turbulence. The overall effect of this phenomenon is that surface renewal of both liquid and vapour phases coming in contact can be achieved at higher rate with the application of pulsations.

4.4.3. Mechanism of Diffusional Transport

A hypothesis of the "free-surface diffusion"\cite{116} theory could be put forward to account for the intensification of separation process under pulsations. With the liquid - fluid boundaries, the transfer necessarily takes place into a free liquid surface, as postulated by Kishinevskii \cite{117} and in addition it is envisaged that turbulence is transferred through the interface from one phase to the other. The rate of transfer through a free liquid surface would be expected to be governed by both the molecular and the eddy diffusivities.

The presence of pulsation introduces interfacial rippling as a manifestation of the transfer of turbulence as suggested by Figure \cite{4.16b}. 

### TABLE 4.4
CORRELATION OF PULSED DATA

<table>
<thead>
<tr>
<th>Tube Size (in.)</th>
<th>$(N.T.U.)_p/(N.T.U.)_0$</th>
<th>$f$ (c/s)</th>
<th>$A$ (cm. Hg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.05</td>
<td>2</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>1.17</td>
<td>4</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>1.10</td>
<td>4</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>1.52</td>
<td>7</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.30</td>
<td>7</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>1.70</td>
<td>10</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.41</td>
<td>10</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>2.62</td>
<td>17</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>2.07</td>
<td>17</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.67</td>
<td>17</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>2.38</td>
<td>21</td>
<td>9</td>
<td></td>
</tr>
<tr>
<td>2.05</td>
<td>21</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.58</td>
<td>21</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>1/2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.03</td>
<td>2</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>1.26</td>
<td>4</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>1.11</td>
<td>4</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>1.45</td>
<td>7</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.44</td>
<td>7</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>1.95</td>
<td>10</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.59</td>
<td>10</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>1.79</td>
<td>17</td>
<td>9</td>
<td></td>
</tr>
<tr>
<td>2.67</td>
<td>17</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.96</td>
<td>17</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>3.67</td>
<td>21</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>2.56</td>
<td>21</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>1.90</td>
<td>21</td>
<td>4</td>
<td></td>
</tr>
</tbody>
</table>
Before this theory can be tested quantitatively, it will be necessary to obtain data for the variation of the eddy viscosity and diffusivity across the interface e.g. by means of velocity and concentration traverses.

A related phenomenon is that of the effect of pulsations result in local variations of velocity gradient at the interface so that mixing is promoted and thus increasing mass transport across the phases.

4.4.4. Theory of partial condensation and evaporation

The subject of this thesis is a proposal for a method of column operation under the application of pulsating pressure, the column being operated first at one pressure maintained at a specified level, then at some increased pressure level, then reverting to the original pressure. The cycle being repeated continuously.

A column operating in this manner between a lower pressure \( P_1 \) and a higher pressure \( P_2 \) will come to thermal equilibrium with the boiling point corresponding to a pressure intermediate between pressures \( P_1 \) and \( P_2 \). With the application of the higher pressure \( P_2 \), superheating of the vapour phase will occur due to the work of compression applied to the system from an external source (pulsations); this compression being neither isothermal nor adiabatic but of a polytropic form. The vapour now being in a superheated condition, will be at a temperature higher than its surroundings.
and as a consequence will lose heat to them, the surroundings being defined as the liquid phase and column walls.

Since the temperature of the liquid is now below its boiling point at the pressure now applied, the superheated vapour, will upon losing its superheat and heat of vaporisation condense, the amount condensing depending upon the heat capacity of the surroundings, the rate of heat transfer and the length which the column is maintained at this pressure. The result will be a surge of liquid down the column as reflux. If, at this point, the pressure is reduced to \( P_1 \), the liquid phase will be at a temperature above its boiling and a "flash vaporisation" will occur with a surge of vapour up the column. Again, the conditions which dictated the amount of superheat vapour condensing will dictate the amount of vapour formed by flash vaporisation.

Equilibrium will be attained only when the temperature of the liquid phase coincides with its boiling point at pressures \( P_2 \) and \( P_1 \) respectively.

The ultimate result of the pulsations would be an alternate series of surges of liquid and vapour streams within the column. It seems quite logical that such surges will tend to promote turbulence in the gas and liquid streams, thereby increasing the rate of diffusion and providing more intermediate intimate contacting of the liquid and vapour phases.
Thus the hoped for effect in distillation under pulsating pressure was an increase in the efficiency of column performance.

However, although the partial evaporation and condensation theory might be applicable to pulsed distillation, it would not apply to pulsed absorption process where the absorbed component reacted in the liquid phase and so became non-volatile. Tudose (81) has shown that the absorption of Ammonia in water under pulsations in a laboratory scale apparatus has similar characteristics to the present pulsed distillation process so it is concluded that the partial evaporation and condensation effect is a negligible factor in the intensification of mass transfer by pulsations.

4.5.1. **SUITABILITY OF METHOD FOR PRACTICAL APPLICATIONS**

Liquid - liquid extraction, by far, is the only known operation in which pulsations are exploited and used. Commercially, the first large scale application of this principle was made during the second World War, when pulsed extraction was used to recover fissionable materials. It has been stated, that the reasons why pulsations of the continuous phase increases the efficiency of extraction are that the total contact area is enlarged and that the turbulence in the two phases is increased. This increase in turbulence causes a decrease in the thickness of the boundary layer which controls the resistance to mass transfer.
The problem of increasing the efficiency of distillation by 200-300% by the use of pulsations is important from a practical point of view. However, the adaptation of this procedure for practical applications must be preceded by an exact analysis of technical and economic factors involved. Work on a range of other operations, discussed in section (I), emphasize the benefits obtained by pulsations of small-scale units and little consideration has been given to "scale-up" and economic factors of the process. Results of the work obtained on plant-scale study of controlled cyclic distillation (118), have shown that the promises of earlier small-scale efforts were not completely realized in the large column and that modifications of tray design would be necessary. It is felt that unless further research can throw more light on these problems, the range of industrial applications of pulsations will remain limited.

From the information at present available it might be possible to construct a "savings curve"; savings could appear as a reduction in equipment size and cost or as an improvement in product quality. Data for the "cost curve" are lacking and it was estimated that as the pulsation intensity is raised, the operating cost will rise steeply, for example in pulsed plate columns the power consumption varies as the cube of the r.m.s. pulsation velocity (119).
It is hoped that further information would be emphasized on the following points in the future before feasibility of the process is justified:

(a) Development of simple and reliable means of pulse generation for large scale units of equipment with emphasis on power consumption and operating costs.

(b) Accurate determination of net savings, on capital cost basis due to the application of pulsations, so that an acceptable return on the investment in pulsation equipment could be assessed.

4.5.2. SUGGESTIONS FOR FURTHER WORK

1. The procedure followed in this study and based on the determination of the overall values of (N.T.U.) is not very satisfactory as it does not give a true picture of what actually happens inside the column during the period of a cycle pulsations. It would be favourable, if the column were provided with intermediate sampling points along its length in order to visualize the effects of liquid distribution and composition and thus point values of (H.T.U.) could be determined.
Another method, of course, is to devise a system for the purpose of measuring the temperatures of both phases accurately along the length of the column.

2. It is not clear whether any advantages would be obtained by applying other pulse wave shapes approaching, for example, a sawtooth or square shape so compared to a near sine-wave operation in this study.

3. Obviously, systems other than M.C.H./toluene should be studied to cover Schmidt number values (Sc) over a wide range to determine the effect of variation of physical properties on performance. Positive and negative systems would conveniently be chosen.

4. The column performance could be investigated and operated at various pressures covering a practical range of applications 0.1 - 3 atms.

5. An extension of this work should be devoted to finding the effect of pulsations on throughput of a continuous column operating at different reflux rates. It will be interesting to operate a semi-plant scale under pulsations to reveal any limitations of this method and to assess its practical validity.

6. Complete and exact data should be made available on the performance of pulsed columns at higher vapour velocities and under range of pulsation intensities than those reported here, also the problem of hydrodynamics of liquid and vapour phase during pulsations.
could be clarified by incorporating a refined system of instrumentation and control along the body of the column, to determine the temperature and velocity profiles.

7. A pulsed multitube column, similar in construction to a heat exchanger would produce low H.E.T.P., with high throughput and a relatively low pressure drop, which would be industrially interesting, particularly for vacuum distillations. More data should be obtained in this area which is, at this stage only a corollary of the present work.

4.5.3. CONCLUSIONS

The main conclusions which can be drawn from this work can be summarized by the following points:

1. Experimental studies on the performance of wetted-well columns using a mixture of methylclohexane and toluene were conducted on three annular tube columns operating under normal conditions i.e. total reflux, atmospheric pressure and non pulsations.

2. A final correlation was obtained which took the form:

\[ HOG = 10.26 \left( \overline{d} - d_0 \right)^{0.64} \overline{Re}^{0.24} \overline{Sc}^{0.67} \]

This expression correlated satisfactorily the data obtained from the 1-in and the 1\(\frac{1}{2}\)-in inner tube columns.
3. The data obtained with the 2-in inner tube column was not used in the above correlation as these data were inconsistent due to mechanical problems of construction as mentioned in section (4.1.)

4. The above equation was derived on the basis that the main resistance to mass transfer is in the vapour phase and that the liquid phase resistance is negligible. The excellent correlation of the data by the equation supports these assumptions and is in agreement with the findings of other workers (112).

5. Expansion of these experiments to include work with other systems and higher vapour flow rates would be necessary to discover the validity of the correlation.

6. Application of pulsations in the range of frequencies 0 - 20 c/s and amplitudes 2 - 10 cm Hg produced an appreciable increase in efficiency relative to the efficiency (N.T.U.) of the non-pulsed columns. An increase by a factor 2-3 having been obtained.

7. Within the range of reflux rates operated; increase in efficiency of the columns were obtained with increase in frequency and amplitude of pulsations. A maximum efficiency was obtained at a frequency of about 17 cycles per second.
It was found that although the overall efficiency decreases with increase in reflux rates, the ratio \(R\), was assumed to be not dependent on the change of the vapour flow rate, in the operating range possible. This simplification was justified by the good correlation obtained of the results for the 1 and 1\(\frac{3}{4}\)-in tube columns.

The increase in the efficiency obtained could be correlated in terms of the pulsation parameters (frequency and amplitude), by the correlation which took the form:

\[
R = 1 + 0.15aA...\sin^2 \left( \frac{x}{2} \frac{f}{f_0} \right)
\]

The validity of this correlation, should be explored, however for higher amplitudes and frequencies and for different systems.

Qualitative explanations were proposed which attempted to explain the characteristics of the results obtained by pulsations. It is concluded that surface renewal of both liquid and vapour phases and the creation of free-surface diffusion by the influence of pulsations, present a favourable explanation to the mechanism of transfer.

It is proposed that expansion of this experiment should be made to large scale continuous distillation columns. This would reveal if the process has any potential commercial viability.
NOMENCLATURE

Symbols are defined in the text after the expression (s) in which they appear. Only symbols which occur repeatedly are given. Although any dimensionally consistent units can be used, the cm. gm. sec. (c.g.s.) system is adopted.

A amplitude of pulsations; cm Hg.

\( a \) interfacial transfer area; sq. cm./cu. cm.

C heat capacity; col/gm. mole (°C)

c Concentration; gm. moles/l.

d diameter of outer tube of wetted-wall column, cm.

do diameter of inner tube of wetted-wall column, cm.

D diffusion coefficient; sq. cm./sec.

G molar rate of flow of vapour; gm moles/sec.

HG height of a transfer unit, gas phase; cm.

HL height of a transfer unit, liquid phase; cm.

\( H_{OG} \) height of overall transfer unit, gas phase; cm.

\( H_{OL} \) height of overall transfer unit, liquid phase, cm.

\( K_G \) mass transfer coefficient, gas phase; gm. mole/sec.cm²( atm. )

\( K_L \) mass transfer coefficient, liquid phase gm. mole/sec. cm (mole fraction).

\( K_{OG} \) mass transfer coefficient, overall gas, gm. mole/sec. cm² (atm.)

\( K_{OL} \) mass transfer coefficient, overall liquid; gm. mole/sec. cm²(mole fraction)

\( f \) frequency of pulsations, sec⁻¹
L  molar rate of flow of liquid; gm. moles/sec.
M  molecular weight.
N  rate of mass transfer; gm. moles/sec.
N' local rate of mass transfer; gm. moles/sec.
N_i  interfacial rate of mass transfer; gm. moles/sec. sq. cm.
N_{OG}  number of transfer units, overall gas.
N_{OL}  number of transfer units, overall liquid.
P  total pressure; (atmospheres)
r  radius of tube; cm.
R  ratio of \(\frac{N.T.U. \text{ - pulsed}}{N.T.U. \text{ - non-pulsed}}\)
S  cross-sectional area of column; sq. cm.
T  absolute temperature; °K.
Z  effective length of wetted-wall column; m
S  acceleration due to gravity; cm/sq. cm.
h  heat transfer coefficient; cal/sec. (sq. cm)°C
h_o  depth below liquid surface; cm.
D  diffusional mass transfer coefficient.
m  slope of equilibrium diagram.
D  refractive index at 20°C.
K  thermal conductivity; (cal/s cm²·deg C cm)
p  partial pressure; (atm)
S  rate of surface renewal; sec⁻¹
t  temperature; °C
u  average vapour velocity; cm/sec.
V  volatility
x  mole fraction of the m.v.c. in the liquid phase, mole fraction.
\( y \) mole fraction of the m.v.c. in the vapour phase; mole fraction.

\( \lambda \) relative volatility as defined in equation (2.10); dimensionless.

\( \mu \) viscosity; gm/cm. sec.

\( \rho \) density; gm/cu.cm.

\( \lambda \) molar latent heat

\( \nu \) kinematic viscosity; sq. cm./sec.

\( \gamma \) peripheral mass liquid flow rate; gm/cm. sec.

\( \omega \) angular velocity; rad/sec

**Subscripts**

A, B refers to components A or B.

G, v vapour phase

L liquid phase

e or * equilibrium value

i interface value

av. average value

p pulsed condition

o non-pulsed condition.

m molar value
ABBREVIATIONS

H.E.T.P.  height equivalent to theoretical plate
H.T.U.  height of transfer unit.
N.T.P.  number of theoretical plates.
M.V.C.  more volatile component.
L.V.C.  less volatile component.
i.d.  inside diameter
o.d.  outside diameter

Nu  Nusselt Number, dimensionless; \( \frac{h d}{K} \)
Pr  Prandtl Number, dimensionless; \( \frac{C \mu}{K} \)
Re  Reynolds Number, dimensionless; \( \frac{d u f'}{\mu} \)
Sc  Schmidt number, dimensionless; \( \frac{\mu}{\rho \nu} \)
Sh  Sherwood Number, dimensionless; \( \frac{k d}{D} \)
Re_v  Reynolds number of the vapour phase.
\( \pi \)  constant = 3.14159
(N.T.U.)_p  number of transfer units - pulsed.
(N.T.U.)_o  number of transfer units - non-pulsed.
R.M.S.  root - mean - square.
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**FORTRAN G382**

**GLOVER**

**CHEMICAL ENGINEERING**

**MASTER PULSED COLUMN**

DIMENSION X(2), E(2), AMP(50), FREQ(50), R(50), W(1000), F(50)

COMMON X, FREQ, AMP, R, W, F, N, M, MAXFUN, ESCALE, IPRINT, INDIC

N=2

M=13

MAXFUN=1000

ESCALE=100

IPRINT=1

DO 1 I=1, M

1 READ(1,100) AMP(I), FREQ(I), R(I)

X(1)=1.

X(2)=15.

E(1)=.001

E(2)=.01

INDIC=0

CALL VA02A

CALL CALFUN

100 FORMAT(3F5.2)

STOP

END

**COMPUTER PROGRAMME**

**CORRELATION OF PULSED DATA USING POWELL OPTIMISATION ROUTINE**

**END OF SEGMENT, LENGTH 108, NAME PULSEDCOLUMN**
SUBROUTINE CALFUN

DIMENSION X(2), E(2), AMP(50), FREQ(50), R(50), W(1000), F(50)

COMMON X, F, AMP, FREQ, R, W, P, N, M, MAXFUN, ESCALE, IPRINT, INDIC

IF (INDIC.EQ.1) WRITE (2, 101)

DO 1 I = 1, M

T = SIN (FREQ(I) / X(2))

RCALC = X(1) * AMP(I) * T

F(I) = R(I) - RCALC

IF (INDIC.EQ.1) WRITE (2, 100) AMP(I), FREQ(I), R(I), RCALC, F(I)

CONTINUE

00 FORMAT (2F9.3, 1E11.2)

101 FORMAT (//, 5X, 5HAMP, 6X, 4HFREQ, 5X, 5HR-1, 4X, 9H(R-1)CALC, 5X, 5HERROR, 1//)

RETURN

END

ND OF SEGMENT, LENGTH 120, NAME CALFUN
SUBROUTINE VA02A
DIMENSION X(2),E(2),AMP(50),REQ(50),R(50),W(1000),F(50)
COMMON X,F,AMP,REQ,R,W,N,M,MAXFU,N,ESCALE,IPRINT,INDIC
MPLUS=N+1
KST=N+MPLUS
NPLUS=N+1
KINV=MPLUS*(MPLUS+1)
KSTORE=KINV+MPLUS+1
CALL CALFUN
NN=N+N
DO1 I=1,M
K=K+1
W(K)=F(I)
1 CONTINUE
IINV=2
K=KST
I=1
2 X(I)=X(I)+E(I)
CALL CALFUN
X(I)=X(I)-E(I)
DO 3 J=1,N
K=K+1
W(K)=0
W(J)=0
3 CONTINUE
SUM=0
KK=NN
DO 4 J=1,M
KK=KK+1
F(J)=F(J)-W(KK)
SUM=SUM+F(J)*F(J)
4 CONTINUE
IF (SUM) .LT. 5.5
5 WRITE(2,7) I
7 FORMAT(5X,2HVA02A E(13,2OH) UNREASONABLY SMALL)
DO 8 J=1,M
NN=NN+1
F(J)=W(UN)
8 CONTINUE
GO TO 10
6 SUM=1./SQRT(SUM)
J=K-N+1
W(J)=E(I)*SUM
DO 9 J=1,N
K=K+1
W(K)=F(J)*SUM
KK=NN+1
DO 11 I=1,I
KK=KK+MPLUS
W(I)=W(I)+W(KK)*W(K)
11 CONTINUE
9 CONTINUE
ILESS=I-1
IAMAX=N+I-1
INCINV=N-ILESS
INCNP=INCINV+1
IF (ILESS) 13,13,14
13 W(KINV)=1
GO TO 15
14 B=1.
   DO 16 J=NIPLUS,IGAMAX
       W(J)=O.
16 CONTINUE
KK=KINV
   DO 17 II=1,IILESS
       IIP=II+N
       W(IIP)=W(IIP)+W(KK)*W(II)
       JL=II+1
   IF (JL=IILESS) 18,18,19
18 DO 20 JJ=J,IILESS
       KK=KK+1
       JJP=JJ+N
       W(IIP)=W(IIP)+W(KK)*W(JJ)
       W(JJP)=W(JJP)+W(KK)*W(II)
20 CONTINUE
   B=B+W(I)+W(IIP)
   KK=KK+1INCINV
17 CONTINUE
   B=1./B
   KK=KINV
   DO 21 II=NIPLUS,IGAMAX
       BB=B+W(I),
       DO 22 JJ=II,IGAMAX
       W(KK)=W(KK)-BB*W(JJ)
       KK=KK+1
22 CONTINUE
   W(KK)=BB
   KK=KK+1INCINV
21 CONTINUE
   W(KK)=B
15 GO TO (27,24),IINV
24 I=I+1
IF (I=N) 2,2,25
25 IINV=1
   FF=O.
   KL=NN
   DO 26 I=I+1,M
       KL=KL+1
       F(I)=W(KL)
       FF=FF+F(I)*F(I)
26 CONTINUE
   ICONT=1
   ISS=1
   MC=N+1
   IIP=IABS(IPRINT)+(IABS(IPRINT)-1)
   IPC=0
27 IPC=IPC-IABS(IPRINT)
   IF (IPC) 28,29,29
28 WRITE(2,30) ITC,MC,FF
30 FORMAT (/5X,9HITERATION,14,19,16H CALLS OF CALCUN,5X,2HF=,E24.14)  
WRITE(2,31)(X(I),I=1,N)
31 FORMAT (5X,9HVARIABLES,(5F24.14))
   IF(IPRINT)1000,1001,1001
   1001 WRITE(2,32)(F(I),I=1,N)
   32 FORMAT (5X,9HFUNCTIONS,(5F24.14))
1000 IPC=IPC
GO TO (29,33) IIPS
29 GO TO (34,35) ICONT
35 IF (CHANGE=1.) 10,10,36
10 IF (IPRINT) 1003,33,37
1003 WRITE(2,1002)
1002 FORMAT(//5X,31HVA02A FINAL VALUES OF VARIABLES)
GO TO 1004
37 WRITE(2,38)
38 FORMAT(///5X,45HVA02A FINAL VALUES OF FUNCTIONS AND VARIABLES)
1004 IPS=2
GO TO 28
33 RETURN
36 ICONT=1.
34 ITC=ITC+1.
K=0
KK=KST
DO 39 I=1,N
K=K+1.
W(K)=0.
KK=KK+N
W(I)=0.
DO 40 J=1,M
KK=KK+1.
W(I)=W(I)+W(KK)*F(J)
40 CONTINUE
39 CONTINUE
DM=0.
K=KINV
DO 41 I=1,N
IIP=II+I.
W(IIP)=W(IIP)+W(K)*W(I)
JL=II+1.
IF (JL=H) 42,42,43
42 DO 44 J=JL,N
JJP=JJP+1.
W(IIP)=W(IIP)+W(K)*W(J)
WCJP=W(JJP)+W(K)*W(I)
44 CONTINUE
43 IF (DM=ABS(W(II)+W(IIP))) 45,41,41
45 DM=ABS(W(II)+W(IIP))
KL=II+
41 CONTINUE
II=II+DMPLUS*KL
CHANGE=0.
DO 46 I=1,N
JL=II+I.
W(I)=0.
DO 47 J=JPLUS,NN
JL=JL+JPLUS.
47 CONTINUE
II=II+1.
W(I)=W(JL).
WC(JL)=X(J)
IF (ABS(E(I)+CHANGE)=ABS(W(I))) 48,48,46
48 CHANGE=ABS(W(I)/E(I))
46 CONTINUE
DO 49 I=1,M
II=II+1
JL=JL+1
W(I) = U(JL)
W(JL) = F(I)

49 CONTINUE
FC = FF
ACC = 0.1 CHANGE
IT = 3
XC = 0.
XL = 0.
IS = 3
XSTEP = AMIN(0.5, ESCELE/CHANGE)
IF (CHANGE = 1.) 50, 50, 51

50 ICONT = 2.
51 CALL VD01A (IT, XC, FC, 6, ACC, 0.1, XSTEP)
GO TO (52.53, 53, 53), IT
52 MC = MC+1
IF (MC = MAXFUN) 54, 54, 55
53 WRITE (2, 54) MAXFUN
54 FORMAT (5X, 5h/maxfun, 16, 17h calls of CALFUN)
ISS = 2
GO TO 51
55 WRITE (2, 54) MAXFUN
56 FORMAT (5X, 5h/maxfun)
ISS = 2
GO TO 51
57 CONTINUE
XL = XC
CALL CALFUN
FC = 0.
DO 58 J = 2, N
X(J) = X(J) + XL*W(J)
58 CONTINUE
XL = XC
CALL CALFUN
FC = 0.
DO 58 J = 2, N
FC = FC + F(J) * F(J)
59 CONTINUE
GO TO (59, 59, 60), 18
60 KN = N
IF (FC = FF) 61, 51, 62
61 IS = 2
FMIN = FC
FSEC = FF
GO TO 63
62 IS = 1
FMIN = FF
FSEC = FC
GO TO 63
63 IF (FC = FSEC) 64, 51, 64
64 K = KSTOR
GO TO (75, 74), IS
65 K = N
66 IF (FC = FSEC) 65, 51, 66
67 CONTINUE
DO 68 J = 1, N
K = K+1
W(K) = X(J)
68 CONTINUE
DO 69 J = 1, N
K = K+1
W(K) = X(J)
68 CONTINUE
GO TO 51
53 K=KSTORE
KK=N
GO TO (69,70,69),1S
70 K=N
KK=KSTORE
69 SUM=0.
DM=0.
JJ=KSTORE
DO 71 J=1,N
K=K+1
KK=KK+1
JJ=JJ+1
X(J)=W(K)
W(JJ)=U(K)-U(KK)
71 CONTINUE
DO 72 J=1,N
K=K+1
KK=KK+1
JJ=JJ+1
F(J)=W(K)
W(JJ)=U(K)-U(KK)
SUM=SUM+W(JJ)
DM=DM+F(J)*U(JJ)
72 CONTINUE
GO TO (73,10)1SS
73 J=KINV
KK=KPLUS-KL
DO 76 I=1,KL
J=J+K+1
K=KK
W(I)=W(K)
W(K)=W(J+1)
76 CONTINUE
IF (KL=N) 77,78,78
77 KL=K+1
JJ=K
DO 79 I=KL,N
K=K+1
J=J+KPLUS-1
W(I)=W(K)
W(K)=W(J+1)
79 CONTINUE
W(JJ)=U(K)
B=1./W(KL-1)
W(KL-1)=W(N)
GO TO 88
78 B=1./W(N)
88 K=KINV
DO 80 I=1,1LESS
BD=B*W(I)
DO 81 J=1,1LESS
W(K)=W(K)-BD*W(J)
K=K+1
81 CONTINUE
K=K+1
80 CONTINUE
IF (FMIN-FF) 82,83,83
83 CHANGE=0.
GO TO 84
82 FF=FMIN
CHANGE=ABS(XC)*CHANGE
84 XL=DM/FMIN
SUM=1./SORT(SUM+DM*X)
K=KSTORE
DO 85 I=1,N
K=K+1
W(K)=SUM*U(K)
W(I)=0.
85 CONTINUE
DO 86 I=1,N
K=K+1
W(K)=SUM*(W(K)+XL*F(I))
KK=NN+1
DO 87 J=1,N
KK=KK+PLUSN
W(J)=W(J)+W(KK)*W(K)
87 CONTINUE
86 CONTINUE
85 CONTINUE
GO TO 14
END
ND OF SEGMENT, LENGTH 2397; NAME VA02A
SUBROUTINE VDO1A(I, X, F, MAXFUN, ABSACC, RELACC, XSTEP)
GO TO (1, 2, 3, 4, 5, 6).

1 I = I + 1
XINC = X + XSTEP
MC = IS - 3
IF(MC) 4, 15, 19
2 MC = MC + 1
IF(MAXFUN - MC) 12, 15, 19
3 ITEST = 4
4 X = DB
F = FB
IF(P - F) 15, 15, 44
5 X = X + XSTEP
GO TO 3
6 IF(F - F) 9, 10, 11
7 X = X + 2
XINC = XINC + XINC
GO TO 3
8 DB = X
FB = F
XINC = XINC
GO TO 13
9 DC = X
FB = F
DC = X
FC = F
X = DC + D + DA
IS = 2
GO TO 3
10 DA = DB
DB = DC
FA = FB
FB = FC
DC = X
FC = F
GO TO 14
11 IF(P - F) 16, 17, 17
12 IF(P - F) 18, 32, 32
13 FA = FB
DA = DB
14 F = F
DB = X
GO TO 14
15 IF(P - F) 21, 21, 20
16 XINC = FA
FA = FC
FC = XINC
XINC = DA
DA = DC
DC = XINC
21 XINC=DC
   IF((D-DB)*(D-DC))32,22,22
22 IF(F-FA)23,24,24
23 FC=FB
24 DC=DB
   GO TO 19
25 FA=F
26 DA=X
27 IINC=2
28 XINC=DC
29 IF((FB-FC)25,25,29
30 IINC=2
31 XINC=DC
32 IF((FB-FC)29,45,29
33 D=(FA-FB)/(DA-DB)-(FA-FC)/(DA-DC)
34 D=(DB-DC)*33,33,37
35 D=0.5*(DB+DC-(FB-FC)/D)
36 IF(ABS(D-X)-ABS(ABSACC))34,34,35
37 IF(ABS(D-X)-ABS(D*RBLACC))34,34,36
38 ITEST=2
   GO TO 43
39 IS=1
40 X=D
41 IF((DA-DC)+(DC-D))3,26;38
42 IS=2
43 GO TO (39,40),IINC
44 IINC=41,3,3
45 IS=2
46 GO TO (41,42),IINC
47 X=DC
48 GO TO 10
49 IF(ABS(XINC-X)-ABS(X-DC))42,42,3
50 X=0.5*(XINC+DC)
51 IF((XINC-X)*(X-DC))26,26,3
52 X=0.5*(DB+DC)
53 IF((DB-X)*(X-DC))26,26,3
54 ITEST=3
55 GO TO 43
56 END

ND OF SEGMENT, LENGTH 982; NAME VDO1A