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PERFORMANCE OF LIQUID-LIQUID EXTRACTION EQUIPMENT

Barney Rubin and Hugh R. Lehman

May 24, 1950

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PERFORMANCE OF LIQUID-LIQUID EXTRACTION EQUIPMENT

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May 24, 1950

ABSTRACT

A pulse column one-half inch in diameter has been built and operated utilizing the systems water, uranyl nitrate, cyclohexanone, and water, uranyl nitrate, pentaether. The reported efficiency of such columns has been verified in that minimum values of heights per equivalent theoretical stage (HETS) have been obtained in the range of seven to ten inches. The column has been found simple to operate and stable in operation. It appears to offer decided advantages for applications where columns of high extraction efficiency are desired.

A general correlation has been developed for predicting the performance of countercurrent liquid extraction towers, in which values of over-all heights per transfer unit (HTU) are expressed in terms of simple linear functions of the flow rates of both phases. The validity of the correlation is demonstrated by applying it to a large number of published extraction data.

HTU values determined experimentally for the pulse column have been found to obey the above correlation with reasonable accuracy. Values of the individual film HTU's may be predicted approximately for pentaether under the conditions of a pulse amplitude of 0.25 gallons per pulse per half-cycle and two inch plate spacing from the equations:

$$(HTU)_G = 3.8G \text{ inches}$$

$$(HTU)_L = 5.0 \text{ inches}$$

at 25 pulses per minute, and

$$(HTU)_G = 3.6G \text{ inches}$$

$$(HTU)_L = 4.0 \text{ inches}$$

at 45 pulses per minute. It is suggested that the above correlation offers

a method for interpreting column behavior in terms of pulse frequency and amplitude, plate spacing, number of perforations per plate, perforation diameter, and properties of the separate phases, as fundamental variables.

INTRODUCTION

The use of continuous liquid-liquid extraction as a method for purification and separation of chemical substances has come into widespread industrial use within recent years. The application of extraction presents advantages in certain cases as a separations method where other operations such as distillation are prohibited for such reasons as sensitivity of materials to temperature or lack of difference in volatility.[1,2]

Until recently the operation of most continuous countercurrent extraction equipment depended upon the turbulent motion of the two phases flowing past each other (because of their density difference) to provide the agitation or mixing necessary to achieve sufficient mass transfer. The degree of mixing is thus a function of the physical properties of the two liquids, and of their flow rate through the apparatus. To increase the agitation, most columns are filled with some irregular packing which forces the liquid to flow in tortuous paths, although some authors have shown that the efficiency of an unpacked (spray) column approaches that of packed columns under certain conditions.[3]

In order to improve on the efficiencies of gravity-powered equipment, recent workers have constructed various mechanically powered devices which are meeting with approval in extraction applications. In the Podbelniak Centrifugal Contactor,[4] two liquids flow through a whirling spiral of sheet metal which is perforated with many small holes. One liquid is thus squirted through the other under the influence of a high centrifugal field. Such machines have been used for solvent extraction of penicillin[4] with success.

A more conventional apparatus constructed by Scheibel[5] consists of a vertical tower fitted with a central shaft on which are mounted propellers spaced at regular intervals. Beds of conventional fixed packing alternate with the propellers and provide semi-quiescent settling zones between the agitated sections.

Innumerable other such devices probably could be constructed, but an objectionable feature which they all have in common is the presence of moving mechanical parts with their attendant servicing and repair requirements. This objection is especially important if radioactive materials are to be used in the apparatus, since the prime prerequisites of equipment for such purposes are simplicity of operation and a minimum of servicing or repair.

The Pulse Column

One powered device which has been described in a patent by Van Dijck[6] does meet the above requirements. This apparatus, to be known hereafter as a pulse column, consists of a vertical column in which are regularly spaced, tight fitting perforated plates. The holes or perforations in these plates are small enough so that countercurrent flow under quiescent conditions is prevented by resistance to flow through such holes. That is, the pressure drop due to flow through the holes is greater than the pressure drop due to density difference. Connected to the column at some point is a pulsator, i.e., a piston or bellows type pump without valves. The function of this pulsator is to impart alternating surges of liquid in both direction.

The action in such a column may be considered to be as follows: assume a section of column between two adjacent perforated plates to contain heavy phase lying in the lower half of the section, and light phase filling the remaining upper half. On the upsurge, the light phase will be forced through the holes in the top plate where it can then rise through the heavy phase

lying above that plate; simultaneously light phase enters the section through the lower plate and streams through the heavy phase in the lower part of the section. On the down-surge exactly the reverse action occurs and heavy phase moves from plate to plate. Countercurrent flow thus occurs by a combination of gravity flow in the sections between plates and mechanical action through the plates. With proper length and frequency of pulse, quite efficient agitation can be obtained in the column. The advantage of such a column, in addition to high efficiency, is the absence of moving mechanical parts within the column.

In addition to the viscosities and densities of the phases and their interfacial tension, the principal variables to be considered in the construction and operation of a pulse column are the pulse frequency and displacement, plate spacing, number of perforations per plate, and perforation diameter. Pending further theoretical correlation, optimum conditions must be determined empirically on each separate system. However, certain generalizations may be predicted from elementary considerations. First, the volumetric throughput rate for each of the two liquids cannot exceed the product of stroke frequency and displacement (i.e., the pulse volume). Second, the optimum displacement per pulse should be somewhat less than the volume between plates, in order to avoid longitudinal mixing which will tend to reduce the extraction efficiency. Third, the holdup should be diminished and the throughput increased by adjusting the pulse frequency to the largest value that is consistent with high extraction efficiency; for a column of given height that is handling a fixed throughput, this will allow the plate spacing to be decreased with a probable increase in extraction efficiency. These conclusions were incorporated in the design and operation of the column to be described in this report, although time did not permit a complete study of all operating

variables; the same conclusions have been reached by the Hanford Works staff in their experimental studies of pulse columns.[7]

Favorable results with pulse columns up to six inches in diameter have been reported by Hanford where the extraction of uranium nitrate and of nitric acid have been studied in the solvent system water-methyl isobutyl ketone.

EXPERIMENTAL STUDY

In view of these favorable reports and in connection with extraction work with uranium nitrate in other solvents, it was decided to construct and operate a pulse column with the purposes of verifying its high efficiency and of obtaining fundamental operating data with respect to operating variables such as flow rates, plate spacing, pulse amplitude, and pulse frequency.

In connection with simultaneous investigations conducted in this laboratory on the extraction characteristics of uranyl nitrate in water and various organic solvents, it was of interest to test the efficiency of the pulse column on these systems and to determine whether the physical behavior of these solvents was satisfactory for countercurrent extraction operations.

Apparatus

A three-foot pulse column was assembled by coupling a one-foot section of half-inch precision bore pyrex pipe to a similar two-foot section. One-inch industrial pyrex pipe tees were used as headers and were coupled to each end of the column with one-inch to half-inch pyrex adapters. The column volume was approximately three hundred and fifty milliliters.

All metal parts including valves were of stainless steel, and all gaskets were of teflon (polytetrafluoroethylene). The solutions contacted nothing other than glass, teflon, or stainless steel.

Sets of half-inch diameter discs cut from one-sixteenth inch teflon sheet and drilled with varying numbers and sizes of holes served as the

perforated plates. The plates were centrally supported on a one-eighth inch rod and were evenly spaced by means of lengths of three-sixteenths inch tubing slipped over the central rod. Fig. 35, Appendix C, is an assembly drawing of the column.

Eight-liter bottles were clamped near the laboratory ceiling and served as feed tanks. The bottles were connected through rotameters to the columns with three-sixteenths inch tubing. One-eighth inch needle valves were used for metering the feeds.

Overflow streams were led through burets with stop-cocks, so that known volumes could be collected over a measured time interval and the overflow rates thus measured.

The pulsator was a bellows pump one and three-eighths inches in diameter with an adjustable stroke length. Pulsator frequency could be changed in steps, by a change of the belt drive pulley, and fine control between steps was achieved by varying the voltage to the Bodine gear-reduction drive motor with a Variac transofrmer. The pulsator was connected to the column through the organic solvent feed line. Because this caused the solvent feed rotameter to pulse and give a variable reading, an additional pressure of ten pounds per square inch of nitrogen was applied to the top of the solvent feed bottle. The pressure was maintained constant by means of a mercury leg which allowed excess nitrogen pressure to escape by bubbling through the mercury.

An electronic level controller was used to maintain the interface either at the top or bottom of the column. Platinum wire probes sealed in glass with the tips exposed were extended through the top and bottom plates to the positions desired for the interfaces. Connection was made to whichever probe was to be used. Operation of the controller depended upon differences in conductivity of the organic and aqueous phases; when the probe was immersed

in the conducting (aqueous) phase, the voltage was sufficient to fire a thyratron which in turn operated a relay connected to a solenoid valve in the aqueous overflow line. (The electrical circuit for the level controller is given in Fig. 36, Appendix C.) When the solenoid valve opened, aqueous phase would drain until the interface dropped below the probe; since it was then in a non-conducting phase, the thyratron no longer actuated the relay, and the valve closed. This controller was stable and operated satisfactorily throughout the experiments.

During later runs it was noticed that there was a considerable heat effect upon extraction, and since it was feared that changes in temperature would effect the equilibrium distribution, the column was jacketed so that water could be used to maintain isothermal conditions. The water for the jacket was led through twenty feet of steel tubing immersed in a thermostat bath; at a flow rate of 0.5 liter per minute, its temperature did not increase more than 0.3°C. during any one run.

Sixty-degree thermometers calibrated in tenths of degrees were inserted at the top, bottom, and two-thirds junction point of the precision-bore section of the column. At the specified cooling-water flow rate, temperatures within the column were maintained constant to within 4°C. or better; without cooling, temperature variations as large as 15°C. were encountered.

Chemical Materials

The two solvents investigated in this work were cyclohexanone and penta-ether (dibutoxy-tetraethylene glycol). These solvents were obtained from the Carbide and Carbon Chemicals Corporation as a technical grade and used as such.

Distilled water was used for all aqueous solutions.

Crystalline uranium nitrate hexahydrate (Mallincrodt) was used for preparing uranium nitrate feed solutions.

All solutions were filtered through coarse filter paper before being used.

Physical properties of the solvents, including their mutual solutilities with water, are listed in Appendix C.

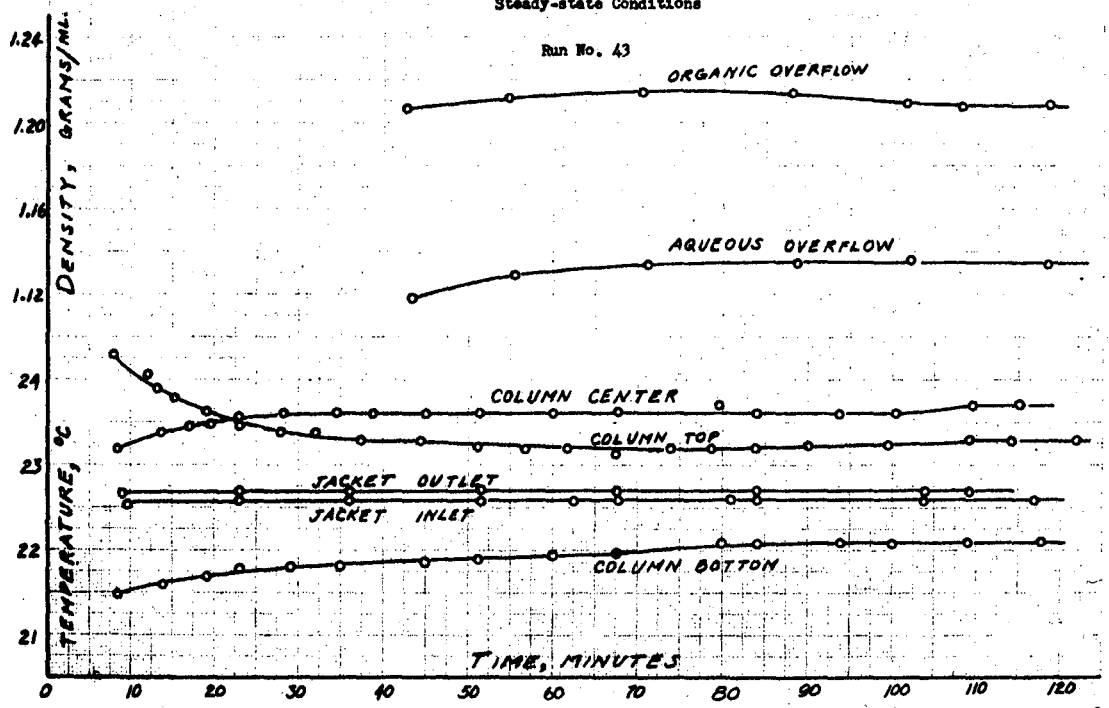
Operating Procedure

Pressure was built up to ten pounds per square inch on the solvent feed bottle before starting operations.

To start the column, it was filled with whichever phase was to be "continuous" (as determined by the subsequent location of the interface). The level controller was connected to the proper probe; the pulsator was set to the desired stroke length and frequency; and the feeds were started at a flow rate corresponding to a selected rotameter reading. The cooling water was then started through the jacket. All thermometers were observed at frequent intervals, and readings were plotted versus time from the start of the experiment. After about forty-five minutes, overflow rates were measured every ten or fifteen minutes, and the densities of the overflow streams were measured by a Westphal balance. These rates and densities were also plotted versus time; the experiment was continued until all variables appeared to have leveled out and were holding steady. This usually took about three hours, time for about ten throughputs of the slower-flowing phase. Fig. 1 shows a typical set of values for these variables as functions of time.

Feed rates were calculated from timed differences of levels of the feed solutions in calibrated sight glasses on the feed bottles; the rotameters were used only to give an approximate value of the feed rates and as a means of keeping the rates constant over the duration of the run.

Figure 1
Plot of Typical Experimental
Data Showing Approach to
Steady-state Conditions



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Analytical Methods

On the cyclohexanone solutions and initially on the aqueous solutions, uranium analyses of all samples were made by use of the Cary Recording Ultraviolet Spectrophotometer; the uranyl nitrate concentration was computed from a known molal extinction coefficient (See Appendix C). Samples whose optical density was greater than 1.5 were diluted accurately to bring them within the range of the spectrophotometer. This method gave results within a precision of about two percent.

When attempts were made to apply the method to pentaether solutions, it was found that the technical grade solvent had sufficient impurities to give considerable background absorption over the spectrum of interest and to render unreliable analyses of dilute solutions. For this reason analysis by density measurement was adopted. The densities of pentaether solutions of known uranium nitrate concentration were determined at 25°, and a graph of concentration versus density (Fig. 26, Appendix C) was drawn.

Since densities could be determined rapidly and accurately with a Westphal balance, the method was used for aqueous solutions also. Density data for aqueous uranium nitrate solutions was obtained from the Project Handbook and are shown in Fig. 25, Appendix C. Concentrations determined in this manner are accurate to within three percent.

Calculations

Cyclohexanone Experiments.-- Material balances for all runs were calculated for each of the three components cyclohexanone, water, and uranium nitrate. Unfortunately, material balances with this solvent were in error on the average by about ten percent. These errors were attributed to lack of attainment of steady state conditions in the tower; for this reason no detailed interpretation of these results will be attempted. However, concentrations were converted

to mole-ratio units (since operating lines represented on mole-ratio coordinates are essentially straight lines) and the number of theoretical stages for each run was obtained graphically by the usual horizontal and vertical steps between operating and equilibrium curves. Fig. 2 illustrates a typical calculation. HETS values (heights equivalent to a theoretical equilibrium stage) were calculated by dividing the number of theoretical stages so obtained into the tower height, and these values are reported for the cyclohexanone-uranium-nitrate-water system in Table I. Equilibrium distribution diagrams for uranyl nitrate in cyclohexanone and water will be found in Appendix C. (Definition of all symbols may be found in the Table of Nomenclature.)

Pentaether Experiments.— Material balances for pentaether experiments were in agreement to within five percent or better for most runs; this was due to more complete attainment of steady state conditions and to refinements in experimental technique. Mole-ratio units were used in all calculations, and it was assumed for calculation purposes that the species extracting into the organic phase was uranium nitrate hexahydrate.

Fig. 3 taken from the data of Stover[8] shows a plot of the total water concentration in pentaether as a function of uranyl nitrate concentration in that phase. In operating the column, the pentaether was introduced dry and withdrawn approximately 0.7M in uranium. The plot shows that the assumption of six water molecules per uranyl ion is a convenient approximation.

HETS values for each run were calculated similarly to those for cyclohexanone, and in addition, values of the over-all aqueous and solvent phase HTU's were computed from the tower height and the number of over-all transfer units for each phase. (The concept of HTU's will be discussed in a later section.) The number of over-all transfer units were calculated from the

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Figure 2

Calculation of HETS for
a Typical Experiment

Run No. 12

Continuous phase: Cyclohexanone

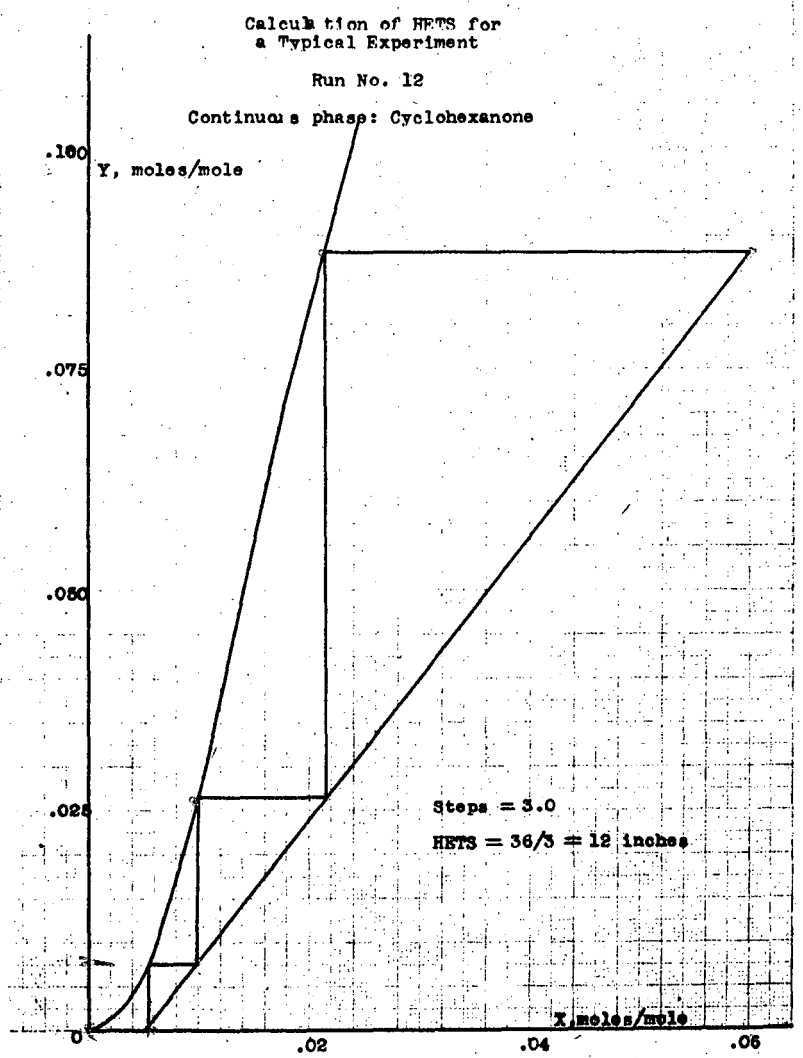

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Figure 3

Concentration of Water vs. Concentration of Uranyl Nitrate in Ferrsether

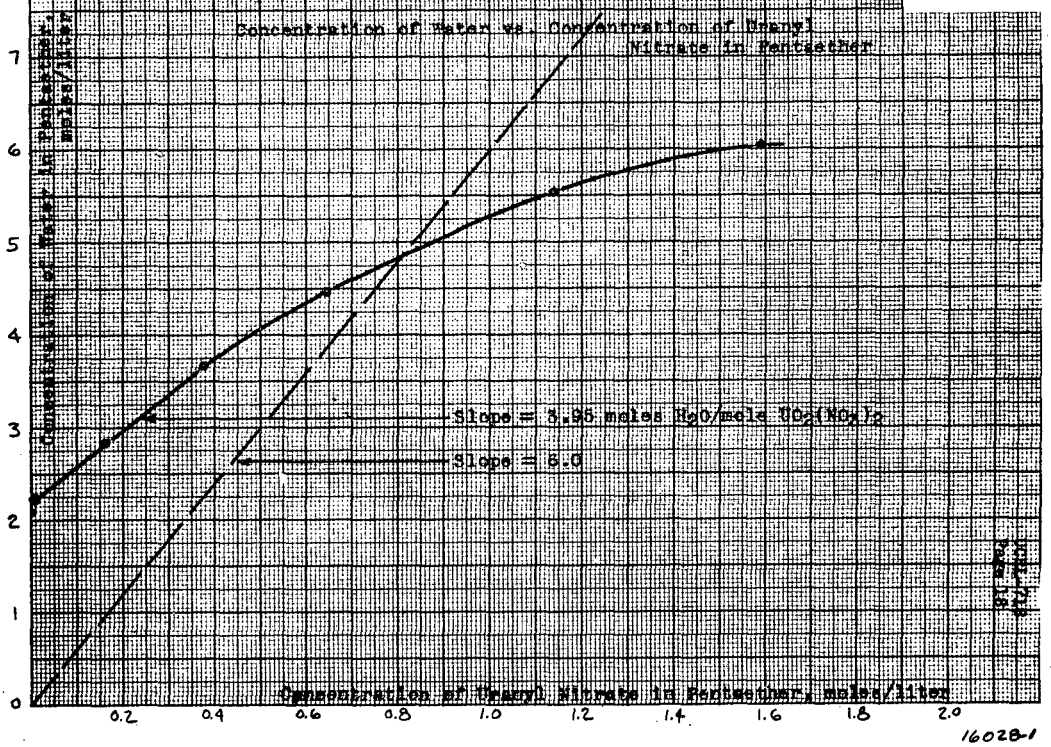

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Table I. Cyclohexanone Runs
36-inch tower height

Run No.	Phase Continuous	Pulse Freq. Cycles/min.	Pulse Amplitude gal/ft^2/1/2 cycle	L lb. moles / hr. ft^2	G lb. moles / hr. ft^2	Concentrations mol/l. Aq. Feed	Org. Feed	Aq. Raff.	Org. Raff.	Plate Spacing inches	Total Plates	No. of Holes	Diam. of Holes inches	HETS inches
1	Cyclohex	144	0.093	1.98	1.65	2.0	0	0.099	0.662	1	33	60	0.025	9.0
2	"	134	0.093	2.14	1.83	2.0	0	flood		1	33	60		--
3	"	80	0.093	2.01	1.78	1.84	0	0.220	0.438	1	33	60	0.025	10.0
4	"	148	0.063	2.04	1.78	1.84	0	0.243	0.420	1	33	60	0.025	10.4
5	"	46	0.093	4.37	1.76	1.84	0	0.195	0.669	1	33	60	0.025	4.5
6	"	80	0.093	4.37	1.76	1.84	0	flood		1	33	60	0.025	--
8	H_2O	45	0.312	1.93	1.56	1.84	0	0.214	0.276	1	33	60	0.025	17.2
9	"	45	0.312	2.04	1.76	2.02	0	0.353	0.460	1	33	60	0.025	12.6
10	Cyclohex	80	0.125	8.75	6.85	2.06	0	0.260	0.568	2	18	53	0.040	13.9
12	"	32	0.218	6.48	3.69	2.06	0	0.290	0.75	2	18	53	0.040	12.0
13	"	26	0.218	--	0.834	0	0.56	0.695	0.05	2	18	53	0.040	8.5
14	H_2O	74	0.125	3.21	2.76	2.09	0	0.230	0.56	2	18	53	0.040	12.6
15	"	74	0.125	6.45	5.50	2.08	0	0.230	0.52	2	18	53	0.040	12.6
16	"	72	0.125	6.42	1.83	2.09	0	0.76	1.09	2	18	53	0.040	19.2
17	"	46	0.093	5.02	5.50	0.235	0	0.16	0.025	2	18	53	0.040	36.0
18	"	80	0.125	10.0	2.38	0.235	0	0.18	0.025	2	18	53	0.040	52
19	Cyclohex	46	0.125	--	--	0.235	0	0.162	0.030	2	18	53	0.040	42
20	"	52	0.048	5.0	1.83	0.235	0	0.145	0.01	1	33	12	0.040	67

expressions

$$N_{OG} = \int_{Y_1}^{Y_2} \frac{dY}{Y^*-Y} + 1/2 \ln \frac{(1+Y_2)}{(1+Y_1)} \qquad (1a)$$

and

$$N_{OL} = \int_{X_2}^{X_1} \frac{dX}{X-X^*} + 1/2 \ln \frac{(1+X_1)}{(1+X_2)} \qquad (1b)$$

In these equations it is assumed that the concentration of solute in the film equals the arithmetic average of the bulk and equilibrium concentrations in the specified phase. Values of the integrals in equation (1a) and (1b) were obtained by graphical integration for each run, and values of the over-all HTU's were obtained by dividing the number of over-all transfer units into the tower height.

The average slope of the equilibrium curve had to be determined in each run, for use in correlating the data; for the relatively concentrated solutions used, the equilibrium line becomes quite curved. Some uncertainty exists in the literature as to the proper method of averaging this slope (m). Koffolt[9] and co-workers have used an integrated average with respect to concentration. This is defined by the equation

$$\bar{m} = \frac{\int m dX}{X_2-X_1}$$

for the aqueous phase and by a similar expression for the solvent phase. Because of the manner in which m is used (cf. equations (3) and (4) below), it is more correct to take an average of m with respect to the number of transfer units in the particular run under study. Accordingly, average values of m were computed from the equation and are reported in Table II.

$$\bar{m} = \frac{\int_{Y_1}^{Y_2} \frac{m dY}{Y^*-Y}}{\int_{Y_1}^{Y_2} \frac{dY}{Y^*-Y}}$$

Table II. Pentaether Runs

19-inch column height 2-inch plate spacing 12-0.040 inch holes/plate

Run No.	Phase Continuous	Pulse Frequency cycles/min.	Pulse Amplitude gal/ft²/1/2 cycle	L lb. moles/hr./ft²	G lb. moles/hr./ft²	Concentrations moles $UO_2(NO_3)_2$ Per Liter Aq. Feed	Org. Feed	Aq. Raff.	Org. Raff.	N_{OL}	N_{OG}	$\bar{m}$	$(HTU)_{OL}$ in.	$(HTU)_{OG}$ in.	HETS in.
22	water	46	0.25	4.35	0.760	2.090	0	0.365	0.775	3.88	1.412	15.1	5.03	13.45	7.72
23	penta	78	0.25	4.25	0.767	2.090	0	0.330	0.808	4.34	1.629	15.1	4.38	11.7	6.85
24	water	78	0.25	4.95	1.410	1.905	0	0.320	0.795	4.42	1.903	15.4	4.30	10.0	6.35
26	penta	46	0.218	7.80	2.02	1.905	0	0.365	0.705	3.55	1.324	16.6	5.35	14.35	8.24
27	"	46	0.218	13.20	2.15	1.905	0	0.441	0.761	3.185	1.233	17.3	5.96	15.4	9.50
28	"	46	0.197	4.22	0.821	1.905	0	0.290	0.293	3.55	0.552	16.8	5.35	34.4	10.3
30	"	46	0.218	7.98	0.877	1.905	0	0.327	0.797	4.19	1.806	17.0	4.54	10.5	6.63
31	"	46	0.25	13.75	0.745	1.960	0	0.990	1.408	1.91	2.387	13.8	9.94	7.95	9.90
33	"	46	0.25	5.07	1.90	1.960	0	0.345	0.377	3.20	0.590	14.4	5.94	32.2	10.9
34	"	46	0.25	5.50	1.90	1.960	0			Flood					
35	"	46	0.25	3.23	1.103	1.960	0	0.348	0.410	3.31	0.704	14.6	5.74	27.0	10.0
36	"	46	0.25	3.23	1.00	1.960	0	0.320	0.473	3.67	0.852	15.9	5.18	22.3	9.50
37	"	46	0.25	3.42	0.359	1.960	0	0.442	1.106	3.83	2.359	16.9	4.96	8.07	6.34
38	"	46	0.25	11.20	0.618	1.824	0	0.882	1.351	2.12	2.631	15.4	8.95	7.23	8.53
40	"	25	0.25	3.70	2.09	1.824	0	0.265	0.200	3.24	0.448	14.6	5.87	42.4	10.6
41	"	25	0.25	3.38	1.48	1.824	0	0.260	0.293	3.56	0.683	15.8	5.34	27.8	8.5
42	"	25	0.25	3.59	0.931	1.824	0	0.274	0.507	4.56	1.413	15.8	4.17	13.45	7.23
43	"	25	0.25	3.57	0.434	1.824	0	0.442	0.878	3.35	1.71	18.3	5.66	11.1	7.90
44	"	25	0.25	10.00	0.434	1.824	0	0.992	1.441	2.08	3.16	17.1	9.15	6.01	8.3
45	"	25	0.25	3.48	0.243	1.824	0	0.274	0.167	3.34	0.342	16.4	5.70	55.6	11.1
46	"	25	0.25	3.68	0.492	1.824	0	0.442	0.804	3.22	1.43	18.3	5.90	13.3	8.73
47	"	25	0.25	3.48	0.993	1.895	0	0.268	0.489	4.25	1.156	16.6	4.47	16.4	7.60
49	"	25	0.25	2.24	0.495	1.895	0	0.370	0.580	3.21	1.07	17.7	5.92	17.8	9.5
50	"	25	0.25	6.10	0.477	1.895	0	0.695	1.237	2.97	2.821	13.8	6.40	6.74	6.73
52	"	25	0.25	3.18	0.504	1.895	0	0.427	0.794	3.23	1.366	18.0	5.89	13.9	8.63

The equilibrium diagrams for pentaether and plots of m versus concentration which were utilized will be found in Appendix C.

GENERAL CORRELATION OF LIQUID-LIQUID EXTRACTION DATA

The approach to countercurrent column performance calculations proposed by Colburn[11,23] and subsequent investigators has been very fruitful in yielding correlations consistent with actual column behavior.[13] So successful has been the application of Colburn's HTU concept to gas absorption tower calculations that this idea has become thoroughly grounded in the literature.[24,25] Numerous applications of the HTU concept to liquid-liquid extraction have been made. However, attempts to interpret experimental data on spray and packed towers in terms of this concept have not been entirely successful, and at the present time no generally reliable method exists for predicting the performance of liquid extraction equipment.

In view of the fundamental character of the assumptions upon which Colburn's equations are based, it is felt that with the addition of suitable refinements to allow for the behavior of liquid-liquid systems, they still should be applicable to liquid extraction towers.

In terms of the solvent phase, the general equation for the height of a tower in which transfer of an inert solute occurs between two immiscible fluid phases passing countercurrent to each other may be written as

$$Z = \int_{Y_1}^{Y_2} \frac{(1+Y)_M \; G dY}{k_g a_v c_{av} \; (Y-Y_i)(1+Y)} \qquad (1)$$

Colburn first noticed that the group $G/k_g a_v c_{av}$ remained remarkably constant over the tower length for liquid-gas systems in any particular column run for a wide range of conditions. He proposed that this group be factored out of the integral expression for Z; since it has the dimensions

of feet, he designated it as the height of a transfer unit, $(HTU)_G$. The physical significance of this group has been explained in terms of other material-transfer concepts such as the height of an equivalent theoretical plate.[10]

The remaining dimensionless integral is denoted as the number of transfer units N_G, since the product $(HTU)_G N_G$ must equal the tower height. A corresponding equation may be written for the liquid phase, denoted by L. (For liquid-liquid extraction, the polar (aqueous) phase will be designated by L and the non-polar (organic) phase by G.)

Because equation (1) includes terms relating to interface properties which are not directly measurable, the concept of over-all HTU's has been introduced.[11] The definition of the number of over-all transfer units, N_{OG}, in this case includes equilibrium concentration instead of interface concentrations; thus

$$N_{OG} = \int_{Y_2}^{Y_1} \frac{(1+Y)_{OM}}{(1+Y)} \frac{dY}{Y-Y^*} \qquad (2)$$

The height of an over-all transfer unit, $(HTU)_{OG}$ is evaluated by dividing the number of over-all transfer units N_{OG} into the tower height. Unfortunately, the $(HTU)_{OG}$ and the corresponding term for the liquid film $(HTU)_{OL}$, vary in an unpredictable manner for various operating conditions. However, equations relating the two film HTU's to the over-all HTU's were derived by Colburn; for the case of dilute solutions, these are

$$(HTU)_{OL} = (HTU)_L + \frac{L}{mG} (HTU)_G \qquad (3)$$

$$(HTU)_{OG} = (HTU)_G + \frac{mG}{L} (HTU)_L. \qquad (4)$$

The validity of these equations for liquid-gas systems has been demonstrated by numerous investigators who have plotted $(HTU)_{OL}$ and $(HTU)_{OG}$ vs.

$\frac{L}{mG}$, $\frac{mG}{L}$, respectively. The slopes and intercepts of the resulting straight lines are easily interpreted from equations (3) and (4) to be the values of the separate film HTU's.

In liquid extraction calculations, the use of equations (3) and (4) has been generally encouraging, but sometimes has given anomalous results in the form of negative film HTU's.[12,9] For this reason numerous investigators have concluded that $(HTU)_L$ and $(HTU)_G$ are not constant from one run to another in liquid-liquid systems, but are functions of other variables such as flow rates or ratios of flow rates.

There is considerable justification for such views, based on results of dimensional analysis and the mass transfer--heat transfer analogy. The equation for heat transfer to or from liquids flowing inside round tubes:[14]

$$\frac{h\,d}{k} \qquad \frac{hk}{d} = \gamma (Re)^{0.8} (Pr)^{0.4} \qquad (5)$$

is well established. It has been found that the analogous equation for material transfer

$$\frac{RD}{d} = \gamma' (Re)^{\alpha} (Sc)^{\beta} \qquad (6)$$

where k is the coefficient of material transfer, holds quite well for wetted-wall gas absorbers where the interfacial area is known;[15] and the same form of equation has been found to apply in the case of a wetted wall liquid-liquid extraction tower.[16]

In spray or packed columns, the interfacial area is not known. In spite of this fact, most investigators have assumed a simple power dependence of HTU on Reynolds number and have obtained equations of the form

$$(HTU)_L = r \left(\frac{L}{G}\right)^{\alpha} \qquad (7)$$

$$(HTU)_G = s \left(\frac{L}{G}\right)^{\beta} \qquad (8)$$

where r, s, α, and β are constant for each system studied. However, no two

systems have given the same values for α and β.

It might be remarked that dimensional analysis does not require the coefficient of material transfer to be a power function of the Reynolds number, but only some unspecified function of it. With reference to the heat transfer analogy, it should be noted that for the thoroughly investigated case of fluids passing normal to single cylinders,[14] the heat transfer coefficient cannot be represented by a simple power function of the Reynolds number.

Practically all previous investigators have assumed HTU's to be functions of L/G in accordance with equations (3) and (4). If one examines the data of careful investigators such as Koffolt, Row, and Withrow,[9] who made a very large number of measurements on spray and packed towers, one finds that although according to equation (3) the data are claimed to fall on a single curve, they seem actually fall on several curves. It was noticed that for each curve, the flow rate of one phase or the other has been held constant so that the graph is really a superposition of several graphs of H_{OL} versus L and 1/G. A typical set of these authors' data is plotted in Fig. 4 for H_{OL} vs. L/mG. Here a grouping of the data into several separate curves at constant G is readily observed. Deviations occur at higher flowrates, due probably to the approach to flooding conditions. Another family of such curves could be drawn through points of constant L.

It would thus appear that H_{OL} and H_{OG} may be separate functions of L and 1/G, and 1/L and G, respectively. This has been verified in the present study. Plots of H_{OL} versus L and versus 1/G, and plots of H_{OG} vs. G and versus 1/L have been prepared from almost all the published data for countercurrent liquid extraction (Appendix A-II). In most cases, the data are represented fairly well by straight lines.

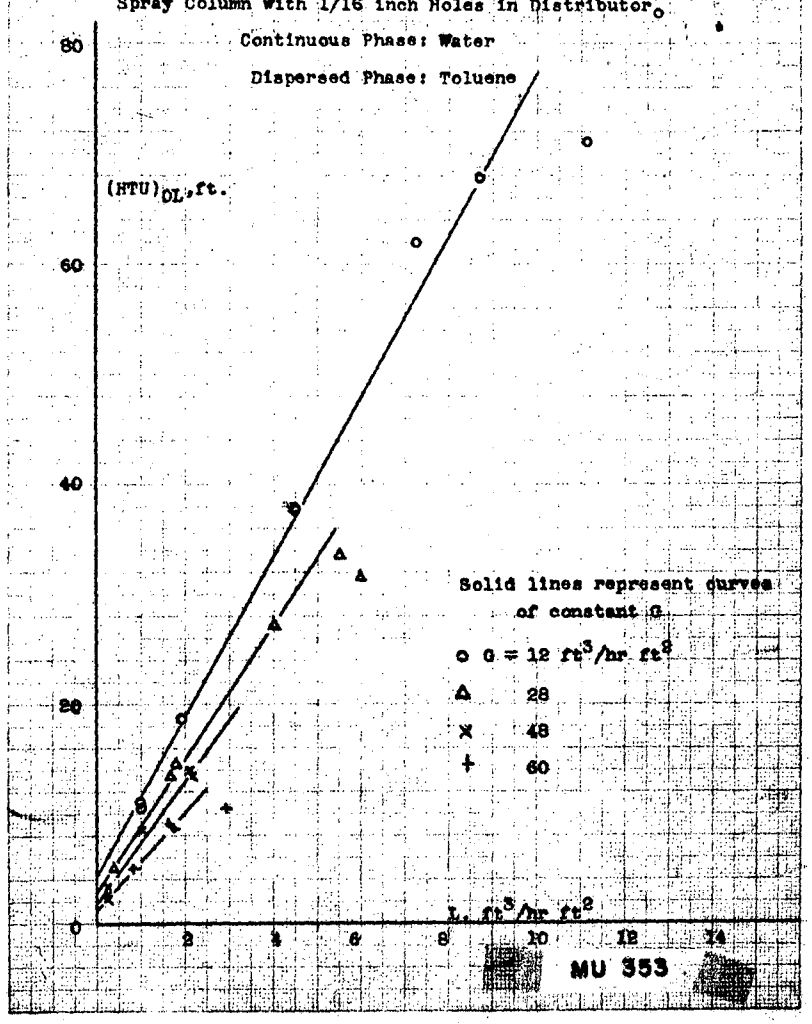
Figure 4

Data of Koffolt, Row, and Withrow

Spray Column with 1/16 inch Holes in Distributor

Continuous Phase: Water

Dispersed Phase: Toluene



A physical justification for these results may be given by the following analysis for H_{OL}; analogous expressions will hold for H_{OG}. Apparently, it is true that

$$H_{OL} = f_1 (L, 1/G). \tag{8}$$

Assume now that

$$H_L = f_2 (L, 1/G) \tag{9}$$

and

$$H_G = f_3 (L, 1/G) \tag{10}$$

Then from equation (3),

$$\left(\frac{\partial H_{OG}}{\partial L}\right)_G = \left(\frac{\partial H_L}{\partial L}\right)_G + \frac{L}{mG} \left(\frac{\partial H_G}{\partial L}\right)_G + \frac{H_G}{mG} \tag{11}$$

$$\left(\frac{\partial H_{OL}}{\partial 1/G}\right)_L = \left(\frac{\partial H_L}{\partial 1/G}\right)_L + \frac{L}{mG} \left(\frac{\partial H_G}{\partial 1/G}\right)_L + \frac{LH_G}{m} \tag{12}$$

Assume next that H_{OL} is a continuous function of L and 1/G as illustrated in Fig. 5. It becomes apparent from these diagrams that

$$\frac{H_{OL}-(i_L)_G}{L} = \left(\frac{\partial H_{OL}}{\partial L}\right)_G \tag{13}$$

and

$$\frac{H_{OL}-(i_{1/G})_L}{1/G} = \left(\frac{\partial H_{OL}}{\partial 1/G}\right)_L \tag{14}$$

where $(i_L)_G$ and $(i_{1/G})_L$ are the intercepts of the tangents to the curves on the H_{OL} axis at constant G and L, respectively. Equating the left member of equation (13) to the right member of equation (11), multiplying by L, and subtracting equation (3) gives the relation

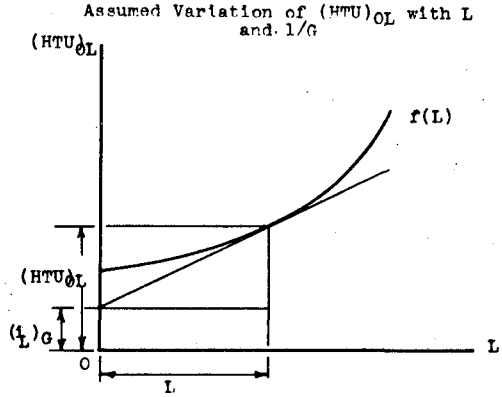
$$H_L = (i_L)_G + L \left(\frac{\partial H_L}{\partial L}\right)_G + \frac{L^2}{mG} \left(\frac{\partial H_G}{\partial L}\right)_G \tag{15}$$

Similar operations will yield

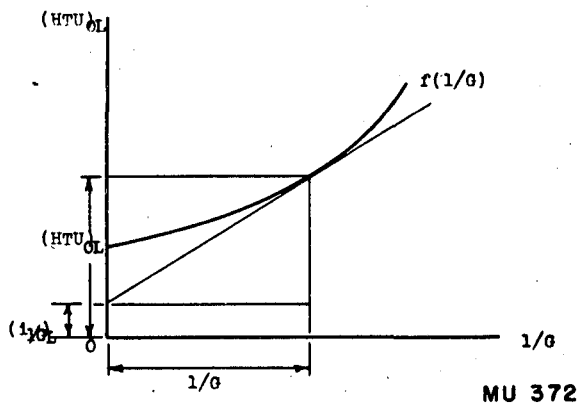
$$H_L = (i_{1/G})_L + \frac{1}{G} \left(\frac{\partial H_L}{\partial 1/G}\right)_L + \frac{L}{mG^2} \left(\frac{\partial H_G}{\partial 1/G}\right)_L \tag{16}$$

Figure 5

Assumed Variation of $(HTU)_{OL}$ with L and 1/G



$(HTU)_{OL}$ vs. L



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$(HTU)_{OL}$ vs. 1/G

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These equations are general and must be satisfied by any equations which express relations between the film HTU's and flow rates. In order to continue, therefore, information is required about the form of the dependence of H_L and H_G on L and 1/G.

Colburn and Welsh,[17] and Laddha and Smith[18] have performed experiments which give some indication of the desired relation. In both of these investigations, immiscible liquids with slight mutual solubilities were run countercurrent to each other in packed towers and HTU's were calculated for the degree of approach of each phase to saturation with the other. In this case the HTU's as measured are directly the film values, since molecules of pure solvent are diffusing and effectively they meet no resistance to transfer within their own phase.

Two conclusions may be drawn from the data of these investigators:

(1) The film HTU of the discontinuous phase is nearly independent of either flow rate.

(2) The film HTU of the continuous phase is a linear function of the continuous-phase flow rate and of the reciprocal of the discontinuous-phase flow rate. For G discontinuous, these conclusions provide that

$$H_G = c \text{ (a const)} \qquad (17)$$

$$H_L = f_1(G)(p+qL) \qquad (18a)$$

$$H_L = f_2(L)(p'+q'1/G) \qquad (18b)$$

or, combining equations (18a) and (18b),

$$H_L = a + bL + d/G + e\frac{L}{G} \qquad (18)$$

where a, b, d, and e are constants.

For L discontinuous, these equations take the form:

$$H_L = c' \text{ (a constant)} \qquad (19)$$

$$H_G = a' + b'G + \frac{d'}{L} + e'\frac{G}{L} \qquad (20)$$

In each of these investigations, the workers themselves did not reach conclusion (2) above. Instead they both claimed dependence of the continuous-phase HTU on a power of L/G. Nevertheless, linear functions definitely fit the data of Colburn and Welsh better than did their power functions; both types of function fit the data of Laddha and Smith equally well, since their exponent on L/G was found to be essentially unity, and the constants a, b, and d, in the present correlation are found to be zero for their data. Plots of their data for the continuous phase are given in Appendix A-I. It was not considered necessary to replot the data for the discontinuous phase.

The approximately constant value of the film HTU for the discontinuous phase has been explained by Colburn and Welsh on the assumption that the ratio of interfacial area to quantity of dispersed phase in the column remains constant. It may be recalled that $H_G = G/k_G a_v c_{av}$; hence constant G/a will lead to constant H_G. An increase in flow rate of the dispersed phase produces more drops and streams in the column, but the effect of increased interfacial area is just counter-balanced by the increased number of drops. This explanation is even more reasonable if it is remembered that the drops of dispersed phase travel through the column at a velocity dependent upon their densities and physical properties, and almost independent of the superficial flow rate of either phase. Increased flow rate, therefore does not greatly increase relative drop velocity, and increased material transfer due to increased interfacial velocity does not occur. Actually the film HTU for the discontinuous phase appears to decrease slightly upon increasing the flow rate of the continuous phase. This is consistent with a slight retardation of the discontinuous phase and a consequent increase in its holdup, which would be predicted in order to change $(HTU)_G$ in proportion to $(1-\epsilon\frac{L}{G})$, where ϵ depends on the linear velocities of the two phases.

The result for the continuous phase HTU may be explained also in terms of the interfacial area, since at constant flow rate of the continuous phase, the resistance to mass transfer should be inversely proportional to the interfacial area and therefore inversely proportional to the flow rate of the dispersed phase. However, the appearance of linear terms in L and 1/G (in addition to terms in L/G) indicates this interpretation has been oversimplified.

The direct proportionality of the continuous phase HTU to its own flow rate may be considered as a residence time effect; it is reasonable to assume that the amount of mass transfer occurring is proportional to the length of time the phase remains in the tower.

Combination of the empirical equations (17), (18), (19), and (20) with the theoretical equations (15) and (16) leads to the result that $(i_L)_G$ and $(i_{1/G})_L$ in the latter equations are of the simple form

$$(i_L)_G = a + d/G \qquad (21)$$

$$(i_{1/G})_L = a + bL \qquad (22)$$

The final form of the correlation now can be obtained by substituting equations (17), (18), (19), and (20) into equation (3).

For G discontinuous, the result is

$$(HTU)_{OL} = a + bL + d/G + L/G\ (e + c/m) \qquad (23)$$

and for L discontinuous

$$(HTU)_{OL} = (c' + \frac{e'}{m}) + \frac{b'L}{m} + \frac{d'}{mG} + \frac{a'L}{mG} \qquad (24)$$

Equations (23) and (24) state that H_{OL} is a linear function of L and of 1/G for L both continuous and dispersed; this is the result which was desired in order to explain the straight-line plots obtained from reported data.

A similar derivation for H_{OG} utilizing the same assumptions will yield the relations:

For G discontinuous,

$$(HTU)_G = (c + em) + mbG + \frac{md}{L} + ma\frac{G}{L} \qquad (25)$$

and for L discontinuous,

$$(HTU)_{OG} = a' + b'G + \frac{d'}{L} + \frac{G}{L}(e' + mc') \qquad (26)$$

HTU Surfaces

The assumption that $(HTU)_{OL}$ and $(HTU)_{OG}$ are functions of the separate flow rates suggests the possibility of plotting the over-all HTU's as functions of two variables in a three-dimensional plot. Fig. 6 represents a plot of equation (23); equations (24), (25), and (26) may be expressed in similar form.

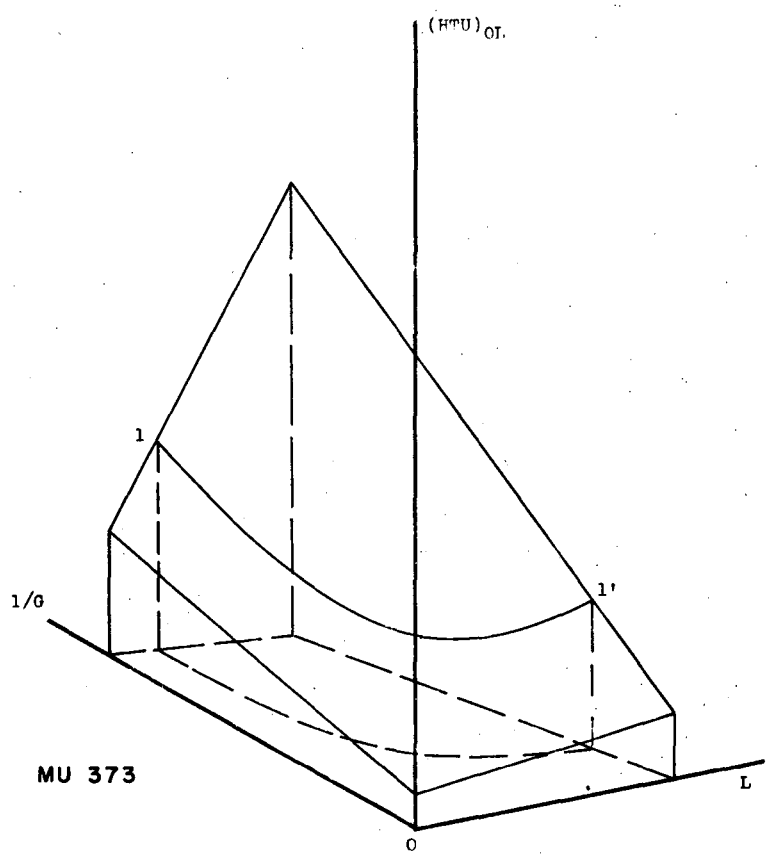
It is of interest to interpret the customary plots of $(HTU)_{OL}$ versus L/G in the light of the present correlation as represented in Fig. 6.

On such a surface, lines of constant L/G such as 1-1' are curves generated by the intersections (with the surface) of a line parallel to the $(HTU)_{OL}$ axis and moving along rectangular hyperbolas in the L-1/G plane. Such curves are not necessarily parallel to the L-1/G plane, and it is apparent therefore that values of $(HTU)_{OL}$ cannot be proportional to L/G without some correction for the separate magnitudes of L and 1/G. It is not surprising, however, that some degree of correlation has been obtained for $(HTU)_{OL}$ as a function of L/G since the variation of $(HTU)_{OL}$ from 1 to 1' probably is not large.

It should be pointed out that the constants a, b, and d, which represent the intersections of the surface with the axial planes, do not necessarily have any physical interpretation; in a real column, flooding would occur as the operating conditions approached the axial plane 1/G = 0.

Figure 6

HTU Surface



A knowledge of either over-all HTU for any given set of operating conditions would enable complete tower-height calculations to be made, since it is always possible to calculate the number of over-all transfer units required for any extraction from equilibrium data, the ratio of flow rates, and recovery specifications. Unfortunately, the above correlation does not permit calculation of the individual film HTU's. Equations (23), (24), (25), and (26) are of such a form that the primed and unprimed quantities a, b, d, and (e+mc) may be determined from graphs of experimental data. However, the values of e and of c (which equals the dispersed phase HTU) are not separable in these expressions.

In view of the previous discussions in regard to the constancy of the dispersed phase HTU, it might be expected that values of this quantity will be dependent on tower packing characteristics and on physical properties of the solute and solvents such as viscosity, density, coefficient of diffusion, and interfacial tension. Laddha and Smith tried to find a correlation for the HTU of the dispersed phase in terms of these quantities but arrived at no satisfactory conclusion. They did not include interfacial tension, however, and recent work by Farmer[19] has shown that interfacial tension is of some importance in correlating material-transfer coefficients for a spray tower. It is reasonable to assume that this property would have great influence on the formation of interfacial area in the column; however, in view of the inadequate data available on the subject, no conclusions can be drawn at present.

The form of the above equations does suggest an approximation which can be made for the value of c if m either is very large or very small. For example, considering equation (24) with L discontinuous, it may be observed that each term except c contains m in its denominator. If m is large, this

would cause $(HTU)_{OL}$ to be almost constant and equal to c. This condition should be true especially at low L and high G; while reverse conditions will hold $(HTU)_{OG}$ constant according to equation (25). It has been observed that in reported data either $(HTU)_{OL}$ or $(HTU)_{OG}$ usually does tend to be nearly constant. In the absence of a well-substantiated correlation for values of the dispersed phase HTU, however, such approximations should be made with caution.

In addition, it should be emphasized that the above correlation holds rigorously only in dilute solution where the operating and equilibrium lines can be assumed straight. In more concentrated solutions the assumptions upon which the basic equations (3) and (4) were derived do not hold, and caution should be exercised in applying the correlation to such data.

CORRELATION OF PULSE COLUMN DATA

As previously mentioned, HTU's were calculated only for pulse column runs with pentaether, since it was felt that the precision obtained in runs with cyclohexanone was not sufficient to enable good correlations to be made.

Accordingly, the over-all HTU's listed in Table II have been plotted versus the various flow rate functions required by equation (24). Results are shown in Figs. 7 through 10, for pulse frequencies of 25 and 45 cycles per minute and at a pulse volume of 0.25 gallons per square foot per half cycle.

Examination of the figures indicates that the data obey the correlation in a fairly satisfactory manner. It might have been expected that, in a powered apparatus such as the pulse column, the dispersed-phase HTU would not be constant since the distinction between continuous and discontinuous phases is decreased considerably by the mechanical dispersion. Experimentally, however, the over-all HTU for the dispersed phase was found to be essentially

Figure 9.
2-inch Pulse Column

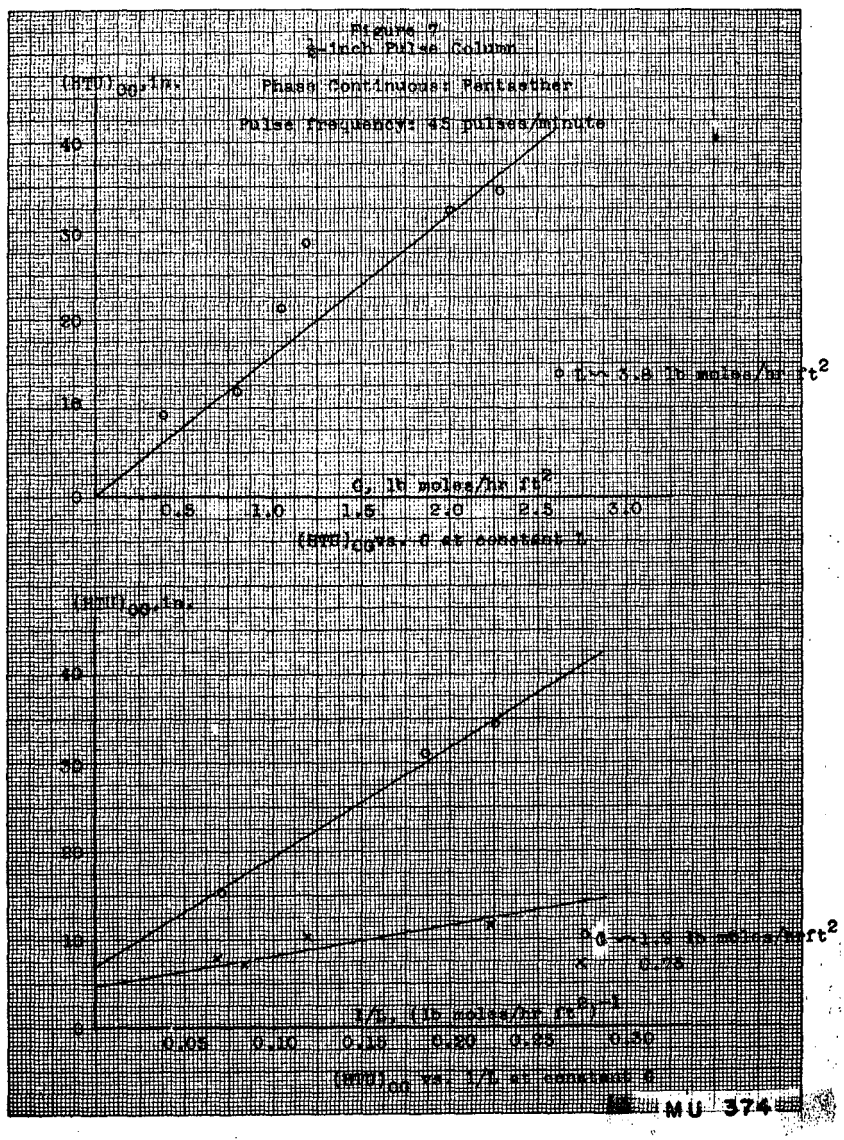


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Figure 3
½-inch Pulse Column
Phase Continuous: Pentanether
Pulse Frequency: 45 pulses/minute
Pulse volume: 0.25 gal/ft^2 ½ pulse

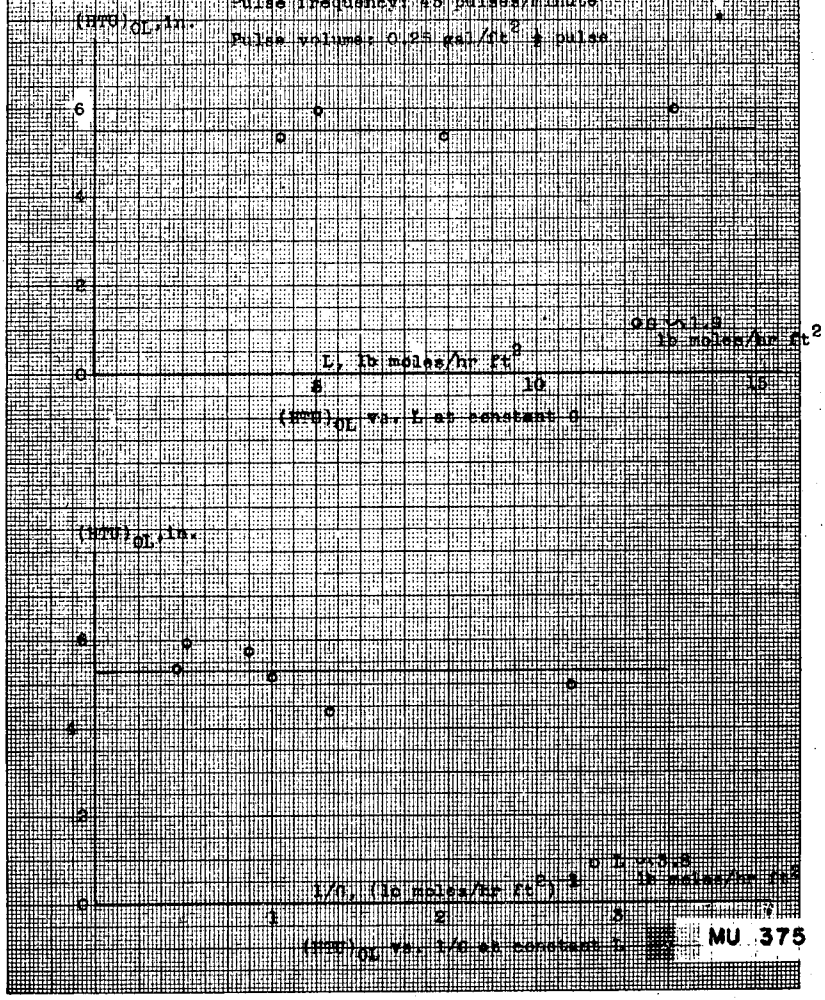

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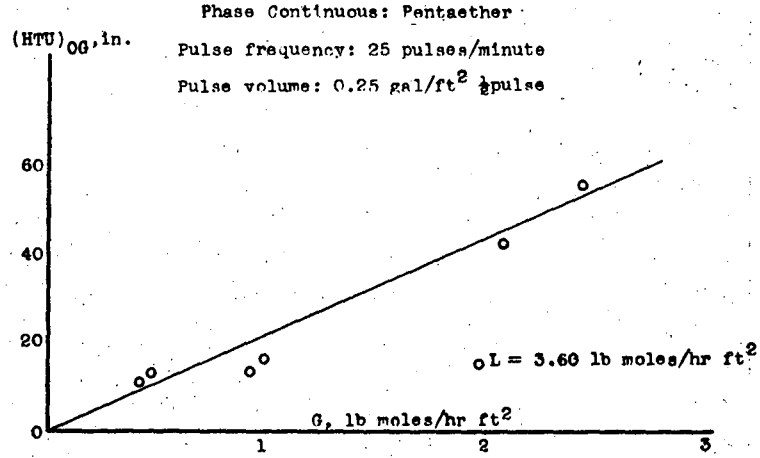
Figure 9
½-inch Pulse Column

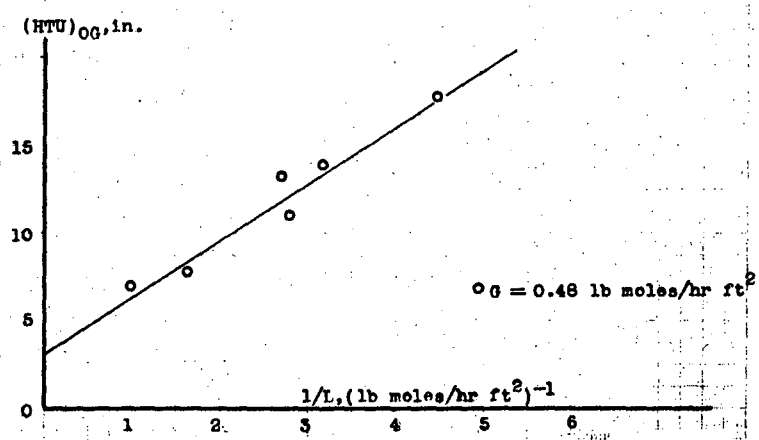
Phase Continuous: Pentaether

Pulse frequency: 25 pulses/minute

Pulse volume: 0.25 gal/ft^2 ½pulse



(HTU)$_{OG}$ vs. G at constant L



(HTU)$_{OG}$ vs. 1/L at constant G

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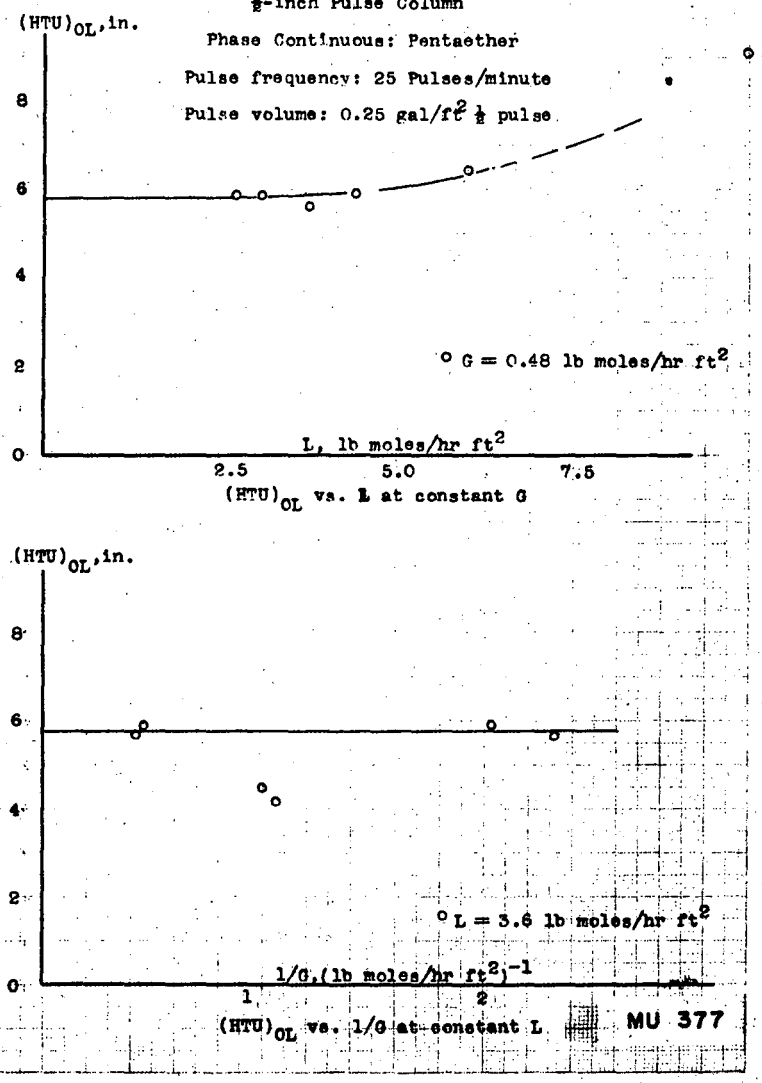
Figure 10
½-inch Pulse Column



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constant.

Examination of the data indicates that the constants a and d in equation (24) are very small, and within the accuracy of the data may be set equal to zero. Evaluation of the two remaining constants leads to the result:

At 25 cycles per minute:

$$(HTU)_{OG} = 3.8\ G + 75\ \frac{G}{L} \text{ inches} \qquad (27)$$

$$(HTU)_{OL} = 5.85 \text{ inches} \qquad (28)$$

If $(HTU)_{OL}$ is estimated from the product $(HTU)_{OG} \times \frac{1}{mG}$, the value obtained is

$$(HTU)_{OL} = 0.24L + 4.7 \text{ inches} \qquad (28a)$$

which agrees with equation (28) within the accuracy of the assumption that m is constant (m ~16).

Because m is large, equation (24) indicates that $(HTU)_L$ (or c') will be very nearly equal to $(HTU)_{OL}$. This assumption leads by equation (4) to a relation for $(HTU)_G$ which would permit it to become negative at certain values of L/G:

$$(HTU)_G = 3.8G - 18\ G/L \text{ inches} \qquad (29)$$

However, as the coefficient of G/L in equation (29) is very sensitive to small changes in $(HTU)_L$, it seems reasonable to assume that it too is equal to zero, giving

$$(HTU)_G = 3.8\ G \text{ inches} \qquad (29a)$$

and, from this expression,

$$(HTU)_L = 5.0 \text{ (average) inches} \qquad (30)$$

Similarly, the data for 45 cycles per minute yield the expressions:

$$(HTU)_{OG} = 3.6G + 60\ G/L \text{ inches} \qquad (31)$$

$$(HTU)_{OL} = 5.3 \text{ inches} \qquad (32)$$

$$(HTU)_G = 3.6G \text{ inches} \qquad (33)$$

$$(HTU)_L = 4.0 \text{ (average) inches} \qquad (34)$$

In a previous section it was mentioned that the film HTU's must be considered to depend upon the tower packing since the degree of agitation or turbulence produced as the liquids flow over the irregular surfaces will be dependent on the shapes and spacings of those surfaces. The pulse column is unique in the respect that it may be considered as a tower whose packing characteristics can be arbitrarily varied by changes in known variables. Thus one would expect an increase in pulse frequency to be reflected in increased agitation and a corresponding decrease in the values of the film HTU's. This conclusion indeed is true as may be seen from equations (28) and (32) where the HTU decreases from 5.0 to 4.0 inches with an increase in pulse frequency from 25 to 45 cycles per minute. Likewise, it may be said that the effect of increased frequency also is to decrease $(HTU)_G$.

No data were obtained with pentaether on the effect of changing plate spacing or the sizes or numbers of holes per plate.

It is of interest to note that a plot of HETS versus the numerical sum of the two flow rates (Fig. 11) exhibits a minimum, in agreement with data on pulse column behavior obtained at Hanford.[7] A plot of (HTU_{OG}) versus this total flow yields a random scatter of points; thus it appears that there is no theoretical justification for such behavior with HETS. However, since values of HETS are easily computed, such a plot is useful practically as it offers a rapid means of estimating optimum flow conditions.

ACKNOWLEDGMENTS

The senior author expresses his thanks to Hugh R. Lehman for his collaboration throughout this investigation, and to Drs. T. Vermeulen and T. E. Hicks for their general direction of the work.

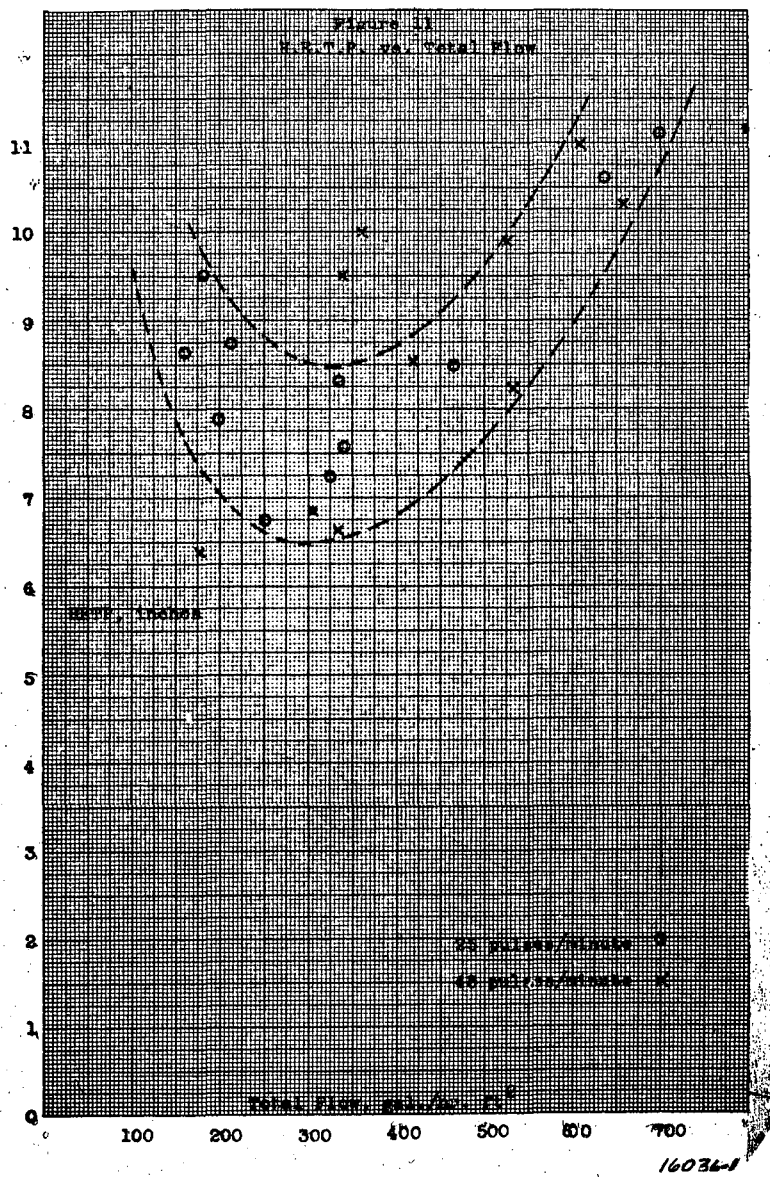


Figure 11
N.P.T.P. vs. Total Flow
11
10
9
8
7
6
5
4
3
2
1
0
100
200
300
400
500
600
700
16036-1

SYMBOLS AND NOMENCLATURE

a, a'	constants in equations (18) and (20)
a_v	interfacial area in ft^2/ft^3
b, b'	constants in equations (18) and (20)
c, c'	constants in equations (17) and (19)
c_{av}	average concentration of solvent, $mole/ft^3$
d, d'	constants in equations (18) and (20)
e, e'	constants in equations (18) and (20)
G	nonpolar phase flow rate, $moles/ft^2$ hr.
HETS	height of an equivalent theoretical stage, inches
HTU	height of a transfer unit
$(HTU)_L$	height of a transfer unit for L phase
$(HTU)_G$	height of a transfer unit for G phase
$(HTU)_{OL}$	height of an over-all transfer unit for L phase
$(HTU)_{OG}$	height of an over-all transfer unit for G phase
k_g	coefficient of material transfer, ft/hr.
L	polar phase flow rate, $moles/ft^2$ hr.
m	slope of equilibrium curve, dY/dX, mole/mole
$\bar{m}$	average slope of equilibrium curve, dY/dX, mole/mole
N_L	number of transfer units for L phase
N_G	number of transfer units for G phase
N_{OL}	number of over-all transfer units for L phase
N_{OG}	number of over-all transfer units for G phase
Pr	Prandtl number, $\frac{\text{specific heat x viscosity}}{\text{thermal conductivity}}$ (dimensionless)
p, p'	constants in equations (18a) and (18b)
q, q'	constants in equations (18a) and (18b)

r	constant in equation (7)
Re	Reynolds number, $\frac{\text{mass rate of flow x viscosity}}{\text{thermal conductivity}}$ (dimensionless)
s	constant in equation (8)
Sc	Schmidt number $\frac{\text{molecular diffusivity x density}}{\text{viscosity}}$ (dimensionless)
X	concentration in L phase, moles/mole
X_i	concentration in L phase at interface moles/mole
X^*	concentration in L phase in equilibrium with Y, moles/mole
Y	concentration in G phase, moles/mole
Y_i	concentration in G phase at interface, moles/mole
Y^*	concentration in G phase in equilibrium with X, moles/mole
$(1+Y)_M$	logarithm average of $(1+Y)$ and $(1+Y_i)$
$(1+Y)_{OM}$	logarithm average of $(1+Y)$ and $(1+Y^*)$
$\frac{hk}{d}$	$\frac{\text{coefficient of heat transfer x thermal conductivity}}{\text{characteristic dimension of system}}$ (dimensionless)
$\frac{kD}{d}$	$\frac{\text{coefficient of material transfer x molecular diffusivity}}{\text{characteristic dimension of system}}$ (dimensionless)
Z	tower height, inches
α	constants in equation (7)
β	constants in equation (8)
γ, γ'	constants in equations (5) and (6)
ϵ	constant in expression for flow dependence of dispersed phase HTU

APPENDIX A-I

CORRELATION PLOTS OF PUBLISHED DATA ON INDIVIDUAL HTU VALUE DETERMINATIONS

As previously stated, the results of experiments performed by the two sets of investigators Colburn and Welsh, and Laddha and Smith, on the determination of individual film HTU's allow important conclusions to be drawn concerning the nature of the dependence of film HTU's on flow rates. These results depend upon the empirical fit of their data to straight lines.

A. Data of Colburn and Welsh[17]

Water and isobutanol were run countercurrent to each other as continuous and dispersed phases in a 3 11/16 inch diameter column packed with 1/2 inch Raschig rings. It may be observed from the plots that the authors apparently were unable to obtain reproducible data; it is possible that this scatter was due to lack of attainment of steady state conditions in their experiments. The authors state that a period of five minutes was allowed for attainment of steady state; this length of time corresponds to slightly over one volume throughput at a typical operating condition, and not less than five throughputs usually are considered to be required for this purpose.

Figs. 12 and 13 represent these authors data in the form suggested in the previous discussions.

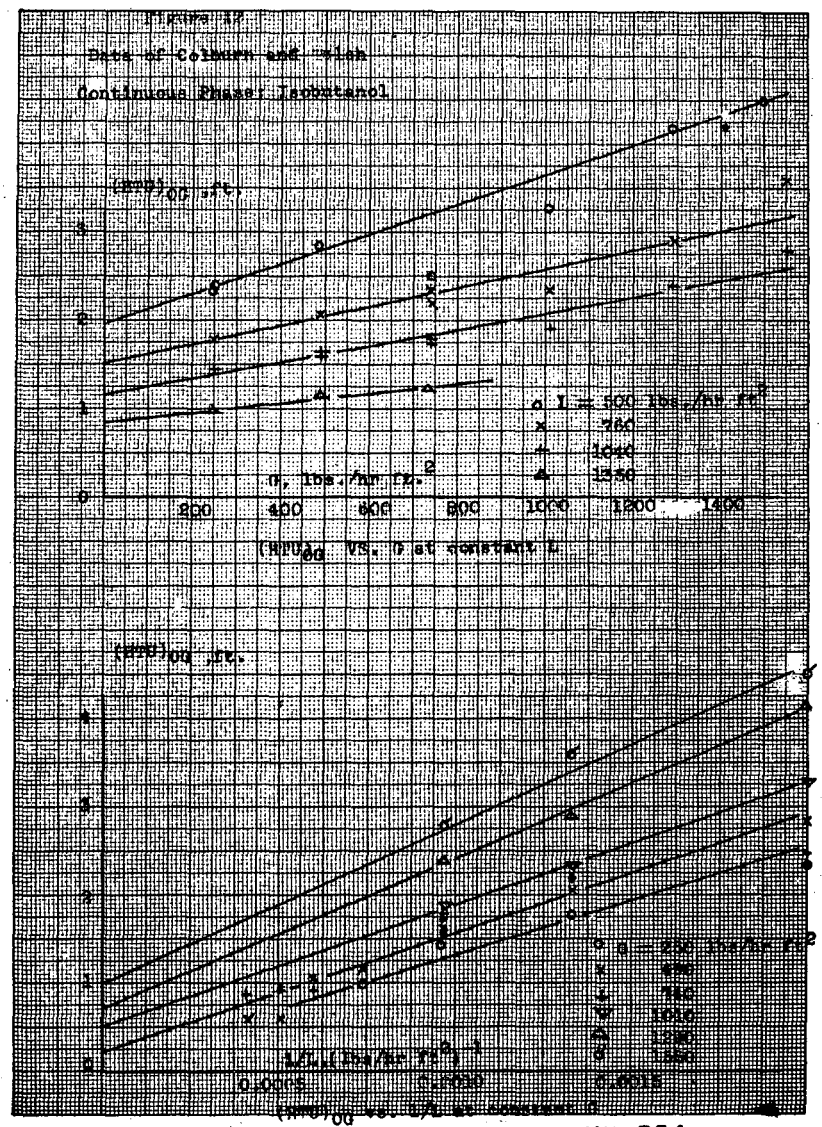
B. Data of Laddha and Smith[18]

These investigators performed essentially the same type of experiments as Colburn and Welsh; their work covers the two systems water-3 pentanol; and water-isobuteraldehyde. Their experimental results appear to be more precise than those of Colburn and Welsh, and lead to the same conclusions. The data are plotted in Figs. 14 and 15.

Figure 12

Data of Column and Flash

Continuous Phase: Isobutanol

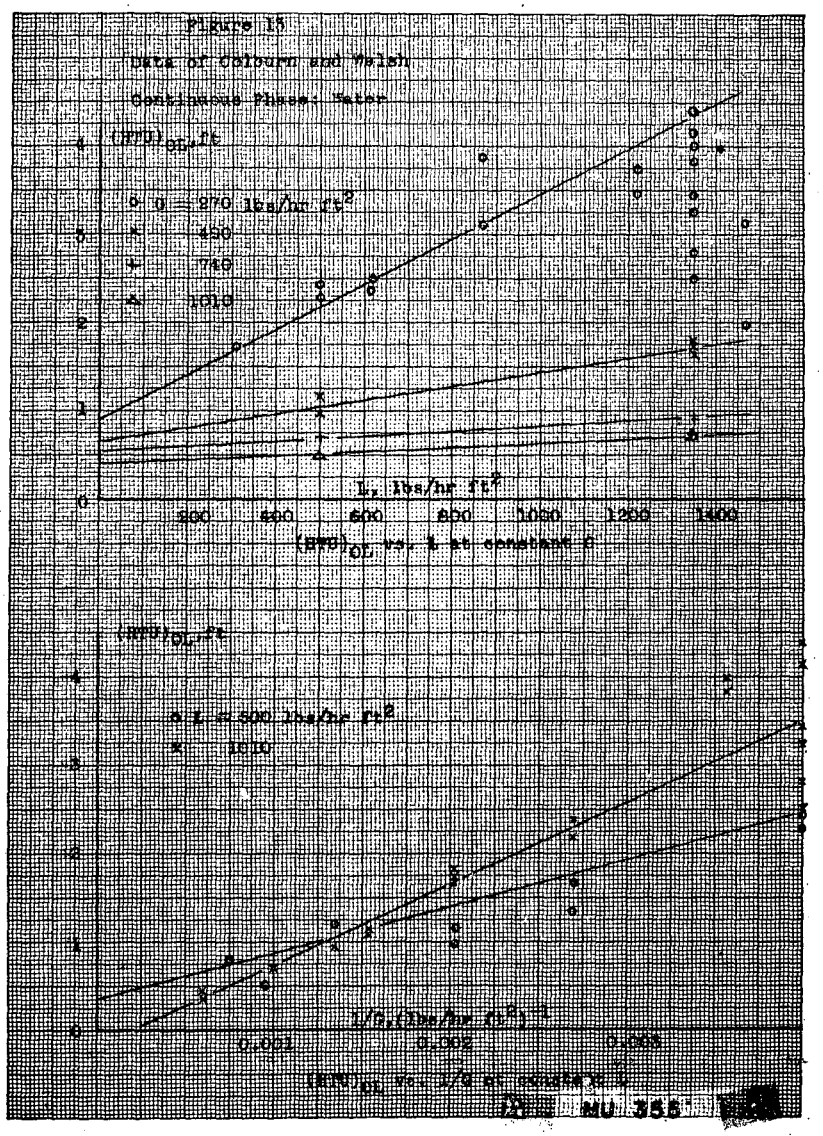


MU 354

Figure 15

Data of Colburn and Welsh

Continuous Phase: Water



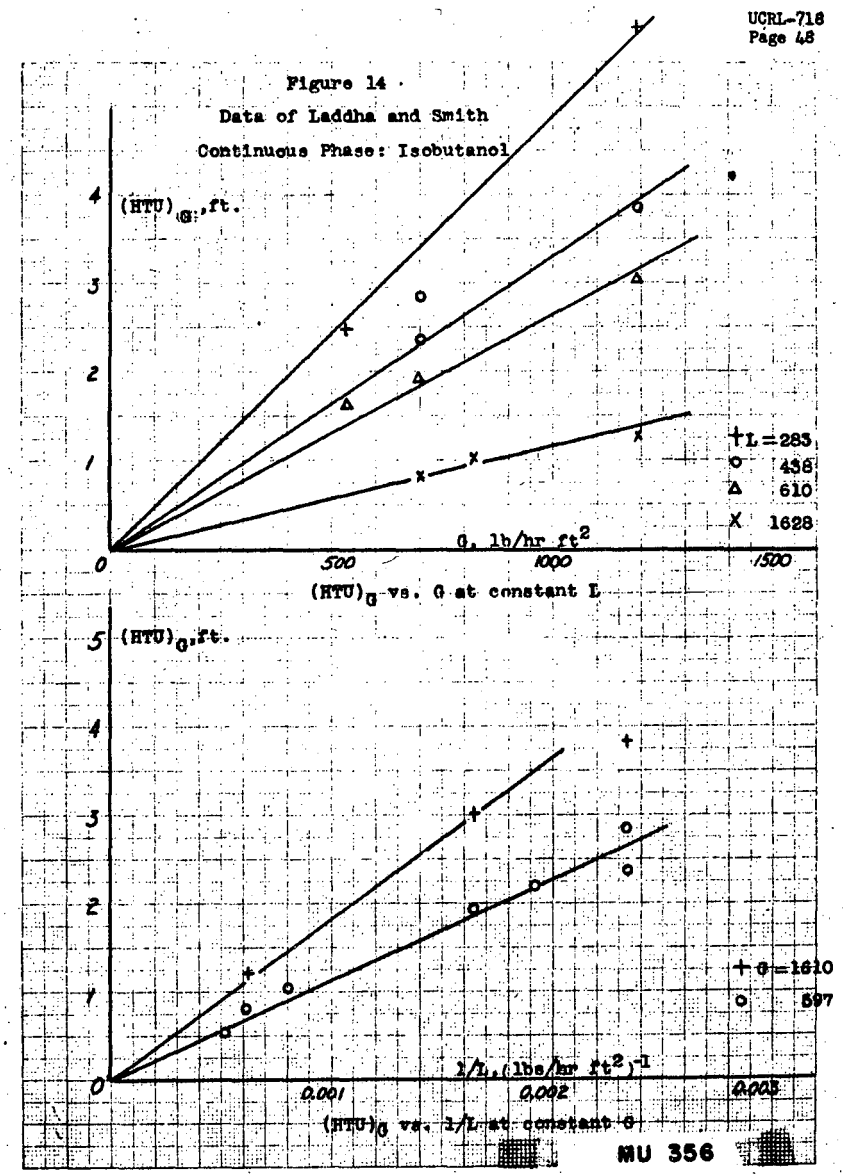
MU 355

16038-1

Figure 14

Data of Laddha and Smith

Continuous Phase: Isobutanol



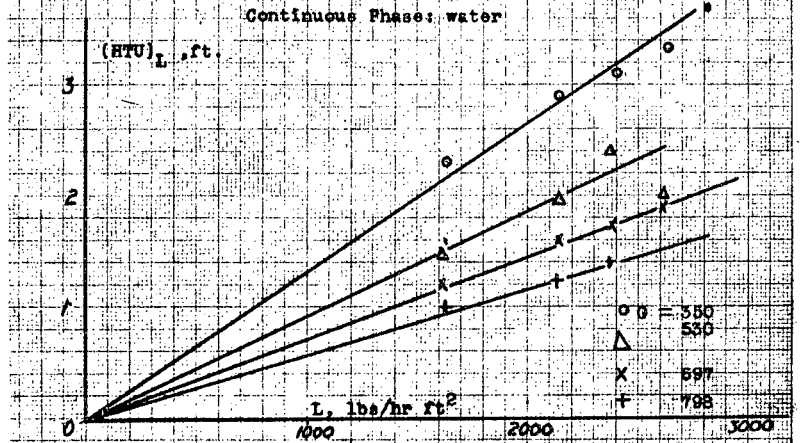
MU 356

160331

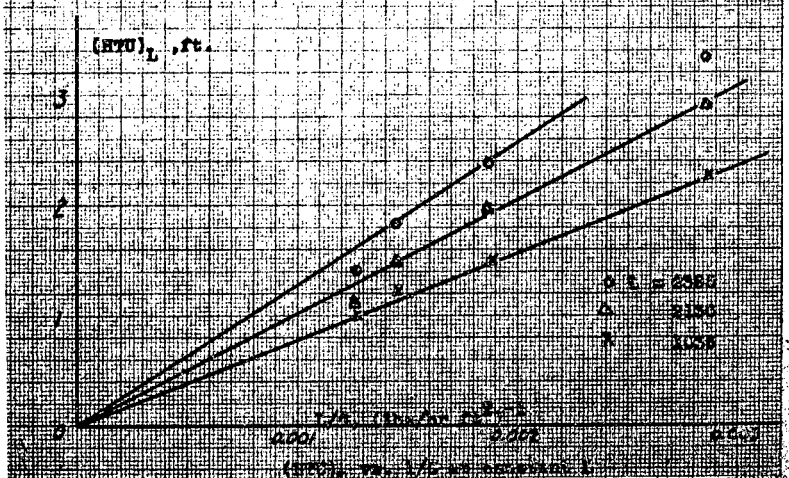
Figure 15

Data of Laddha and Smith

Continuous Phase: water



(HTU)$_L$ vs. L at constant G



(HTU)$_L$ vs. L/G at constant L

APPENDIX A-II

CORRELATION PLOTS OF PUBLISHED EXPERIMENTAL DATA ON OVER-ALL HTU DETERMINATIONS

A. Data of Koffolt, Row, and Withrow[9]

The data presented by these authors constitutes by far the most comprehensive coverage to date of different types of countercurrent extraction equipment by any single set of investigators. Their experiments utilize the transfer of benzoic acid between the solvents water and toluene, and were performed in an 8.75 in diameter column which was operated consecutively as: a spray column with various sizes of dispersing nozzles; a column packed 1/2 inch Berl saddles, with 1/2 inch Raschig rings, or with strips or rolls of copper gauze; and as a bubble cap and perforated plate tower. Figs. 16a through 19 represent the data of these authors for representative sets of their experiments.

B. Data of Allerton, Strom, and Treybal[20]

These investigators performed experiments on the transfer of benzoic acid between water and toluene or kerosene in a 3-5/8 inch diameter tower operated with perforated plates and with half-inch carbon rings. Figs. 20 and 21 illustrate their data.

C. Data of Sherwood, Evans, and Longcor[21]

These authors investigated the transfer of acetic acid between water and methyl isobutyl ketone or benzene in a tower packed with 1/2 inch carbon rings and 1/2 inch Berl saddles. Their data are shown in Fig. 22.

D. Data of Hou and Franke[22]

The experiments of these authors were performed in a laboratory glass column for the purpose of investigating the effect on HTU of changes in

packing material. Data were obtained with fine glass and stainless steel helices; the dependence of extraction behavior on packing characteristics is illustrated by this work. The data are plotted in Fig. 23.

E. Data of Knight[12]

The data reported by this author are sufficient to give only one curve; however, the experiments were performed on the transfer of furfural between water and toluene in relatively concentrated solution where the equilibrium line is quite curved. For this reason a plot (Fig. 24) is included in order to show the fit of this type of data to the correlation.

F. Data of Elgin and Browning[26] and of Johnson and Bliss[3]

In addition, the data of Elgin and Browning and of Johnson and Bliss appear to fit the correlation; plots of their data have been prepared, but for brevity these graphs have not been included.

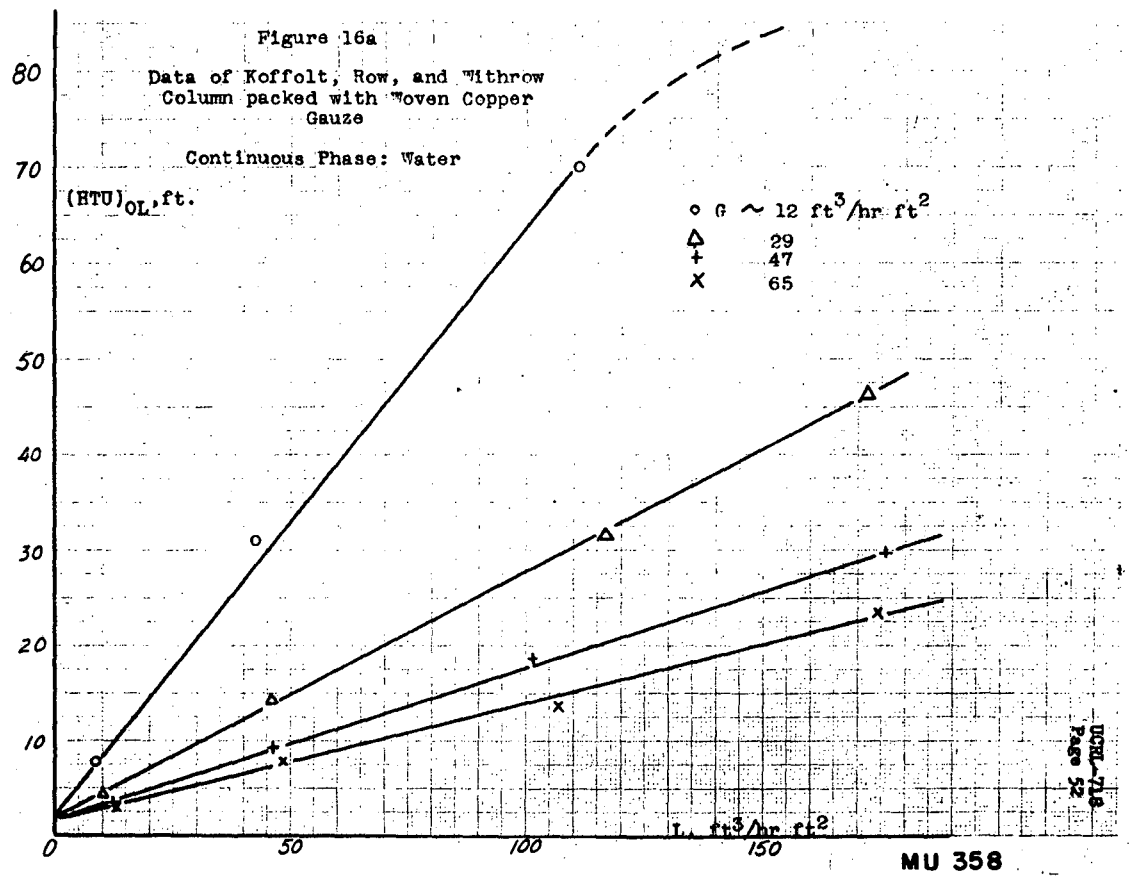


Figure 16a
Data of Koffolt, Row, and Withrow
Column packed with Woven Copper
Gauze
Continuous Phase: Water
(HTU)OL, ft.
○ G ~ 12 ft3/hr ft2
△ 29
+ 47
× 65
80
70
60
50
40
30
20
10
0
50
100
150
L, ft3/hr ft2
MU 358

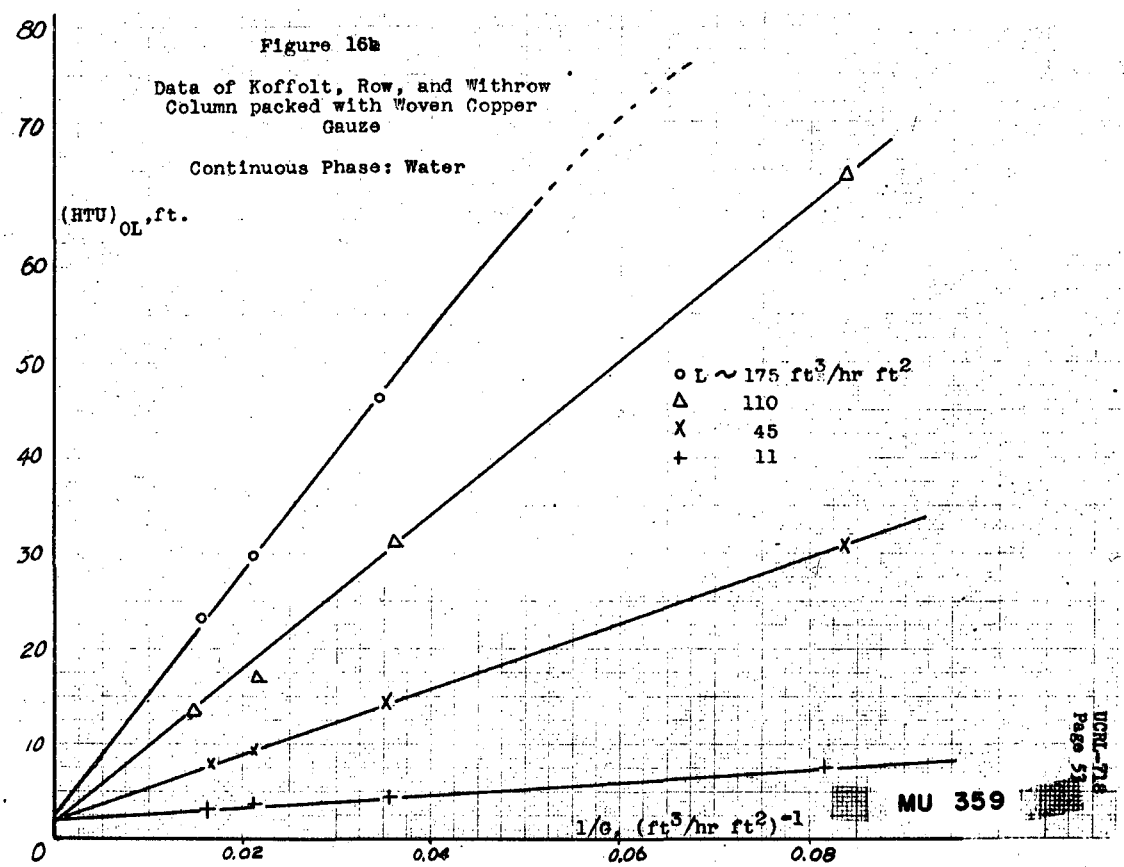


Figure 16b
Data of Koffolt, Row, and Withrow
Column packed with Woven Copper
Gauze
Continuous Phase: Water
(HTU)OL, ft.
o L ~ 175 ft3/hr ft2
Δ 110
X 45
+ 11
1/G, (ft3/hr ft2)-1
0
10
20
30
40
50
60
70
80
0.02
0.04
0.06
0.08
MU 359

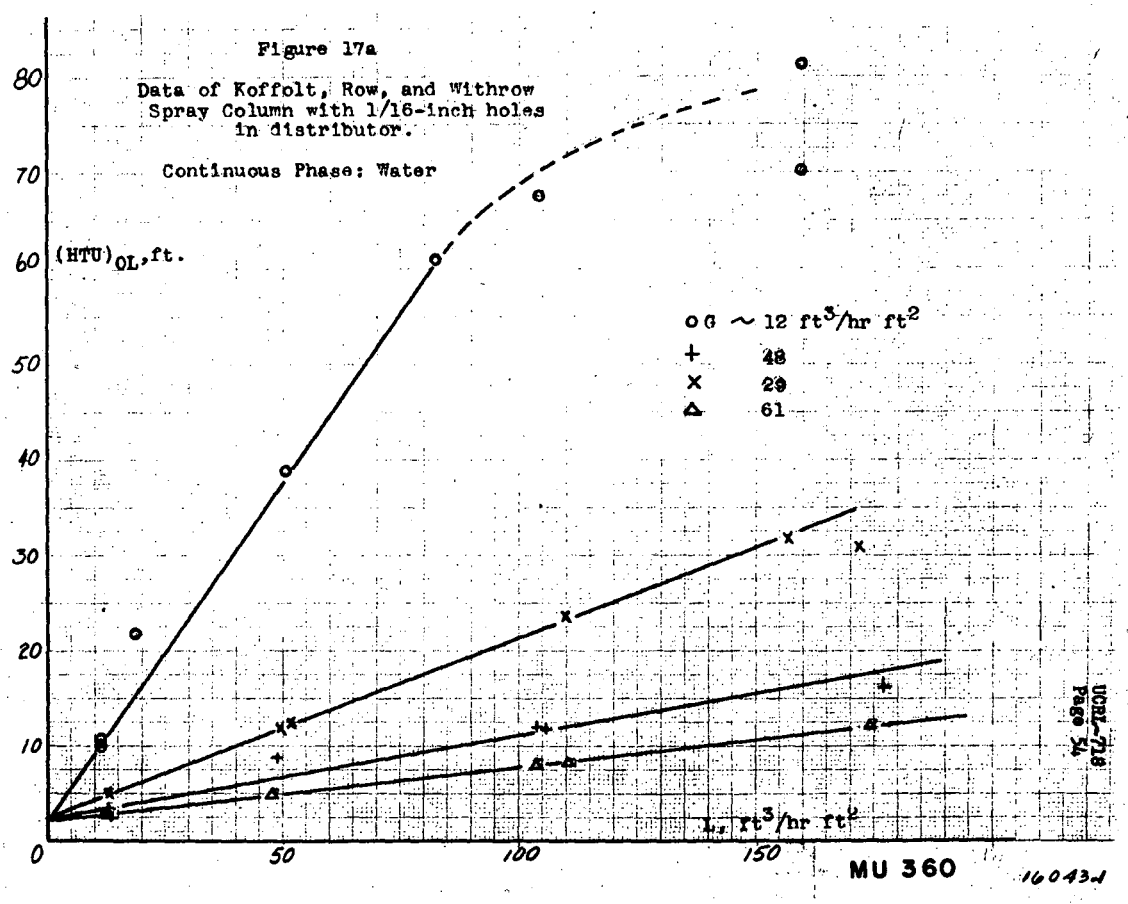


Figure 17a
Data of Koffolt, Row, and Withrow Spray Column with 1/16-inch holes in distributor.
Continuous Phase: Water
(HTU)OL, ft.
o G ~ 12 ft3/hr ft2
+ 48
x 29
Δ 61
L, ft3/hr ft2
0
10
20
30
40
50
60
70
80
50
100
150
MU 360
160434

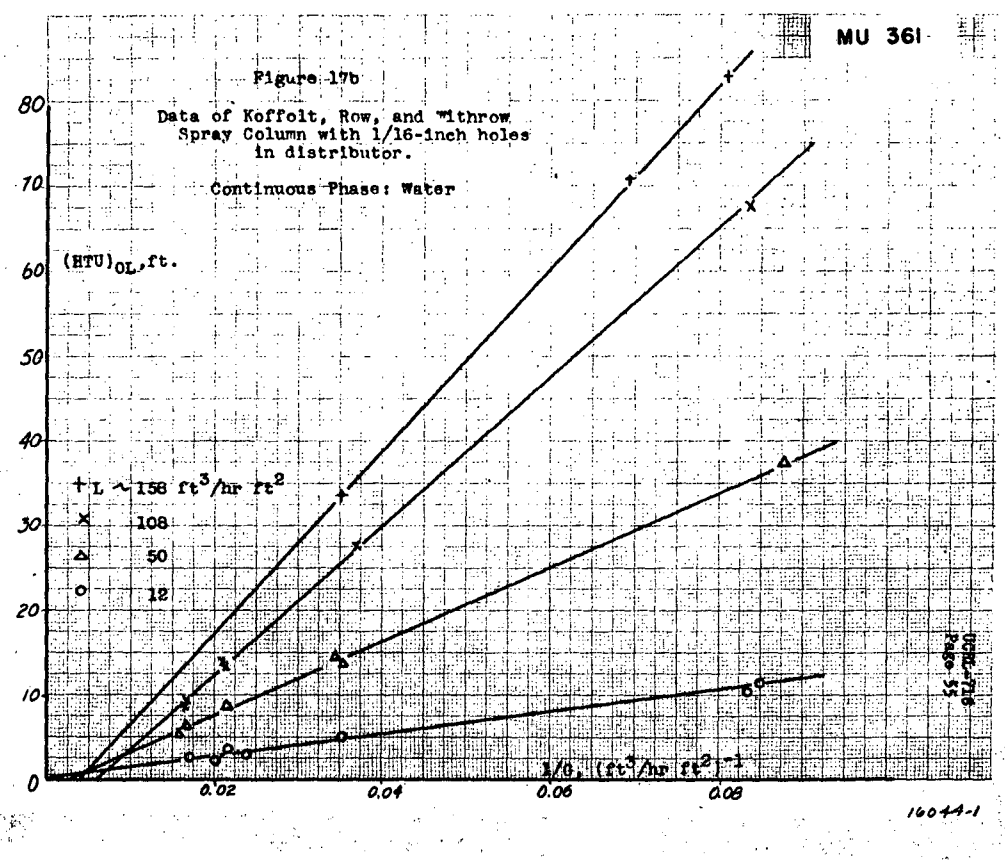

MU 361
Figure 17b
Data of Koffolt, Row, and Withrow
Spray Column with 1/16-inch holes
in distributor.
Continuous Phase: Water
(HTU)OL, ft.
+ L ~ 158 ft3/hr ft2
× 108
△ 50
○ 12
80
70
60
50
40
30
20
10
0
0.02
0.04
0.06
0.08
1/G, (ft3/hr ft2)-1
16044-1

Figure 18

Data of Koffolt, Row, and Withrow
Column packed with ½-inch Berl
Saddles

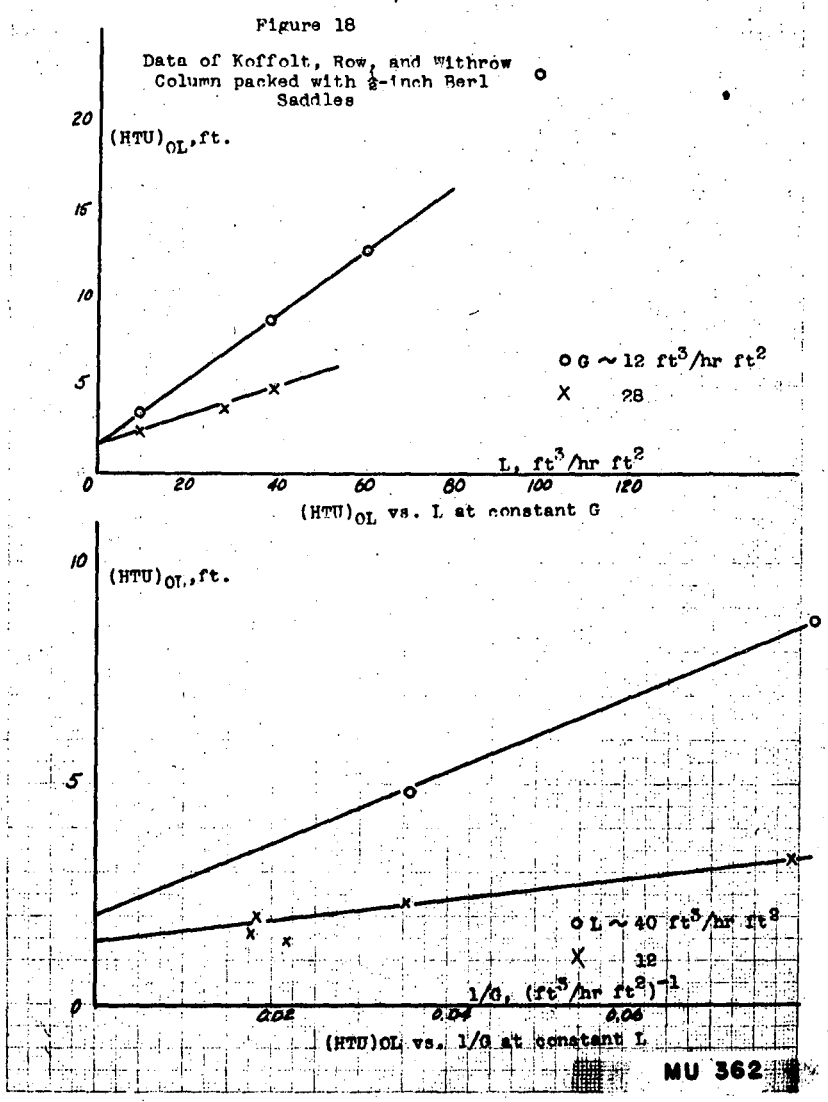
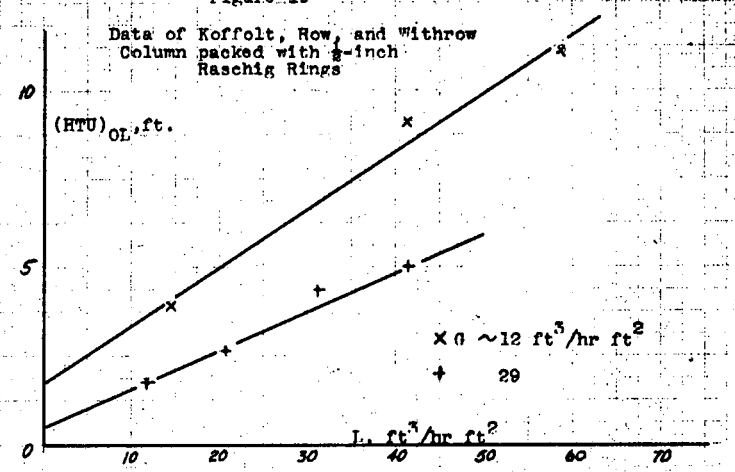
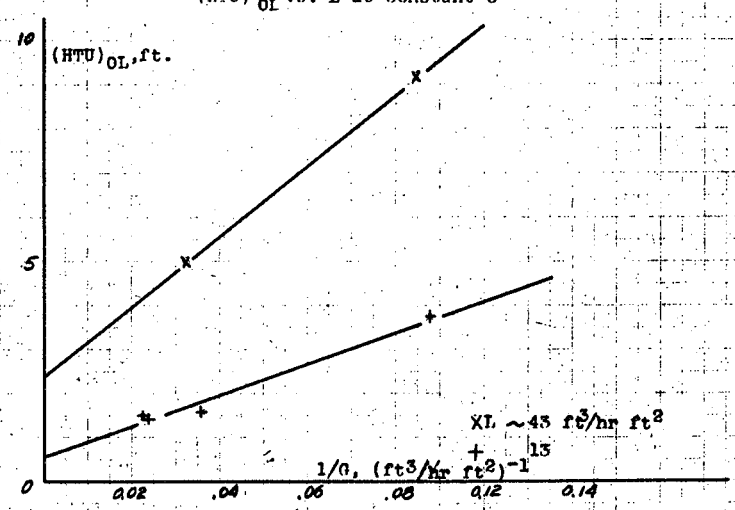


Figure 19

Data of Koffolt, Row, and Withrow
Column packed with ½-inch
Raschig Rings



(HTU) OL vs. L at constant G



(HTU)OL vs. 1/G at constant L

MU 363

MU 364

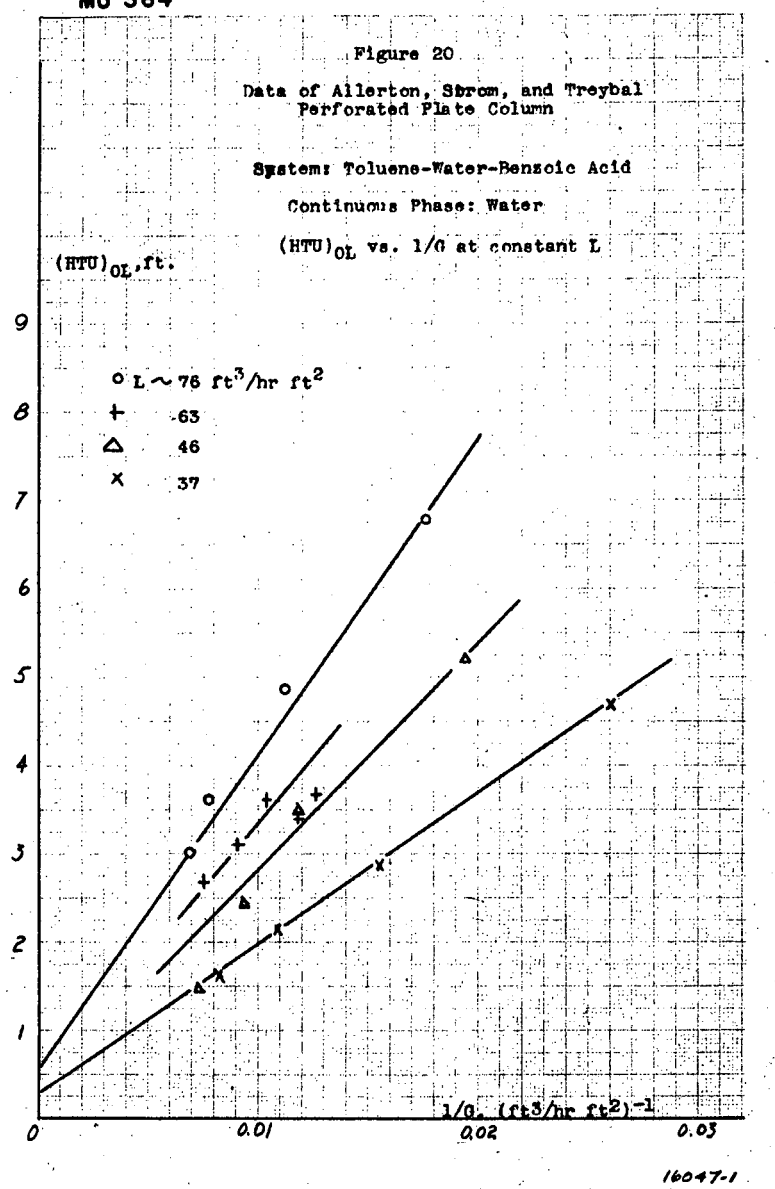


Figure 20

Data of Allerton, Strom, and Treybal
Perforated Plate Column

System: Toluene-Water-Benzoic Acid

Continuous Phase: Water

$(HTU)_{OL}$ vs. 1/G at constant L

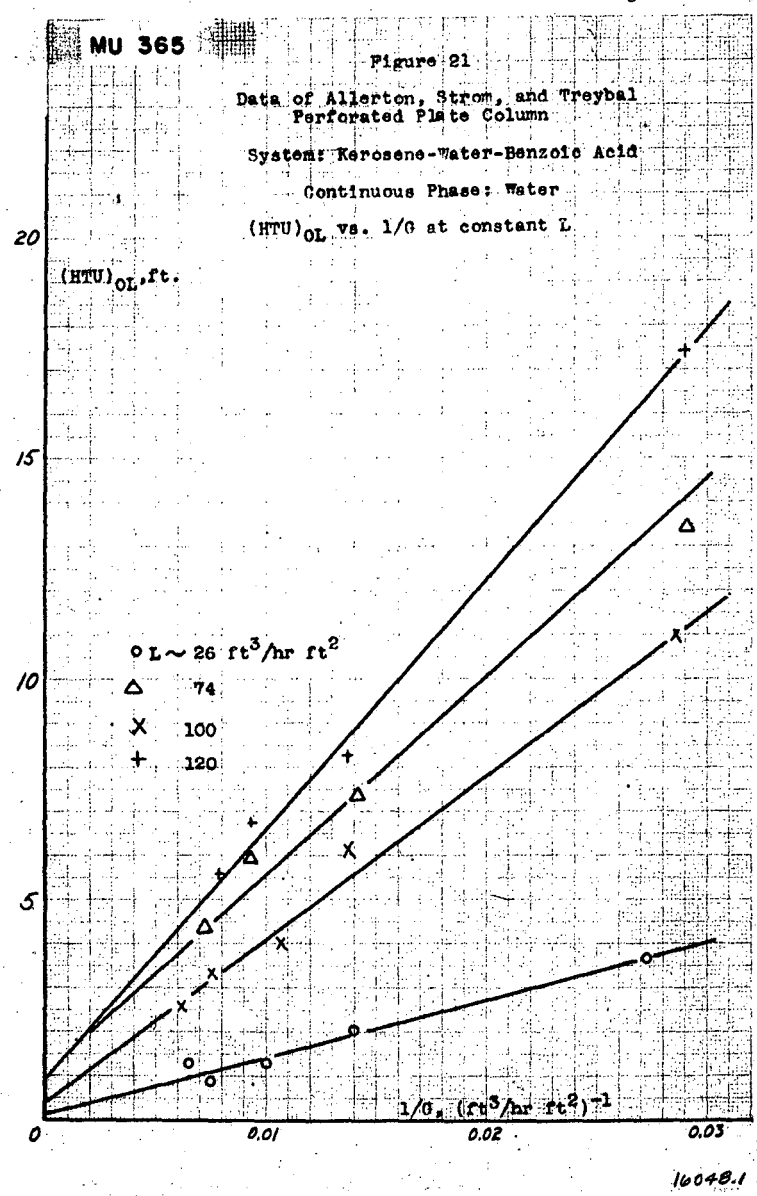


Figure 21

Data of Allerton, Strom, and Treybal
Perforated Plate Column

System: Kerosene-Water-Benzoic Acid

Continuous Phase: Water

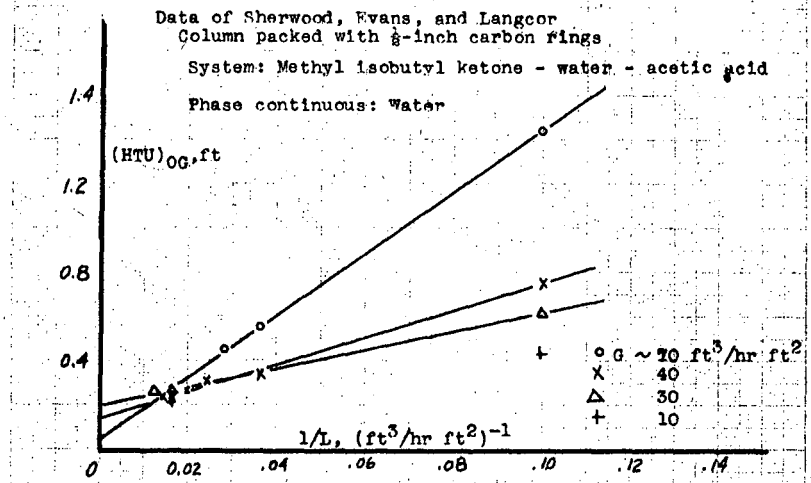
$(HTU)_{OL}$ vs. 1/G at constant L

Figure 22.

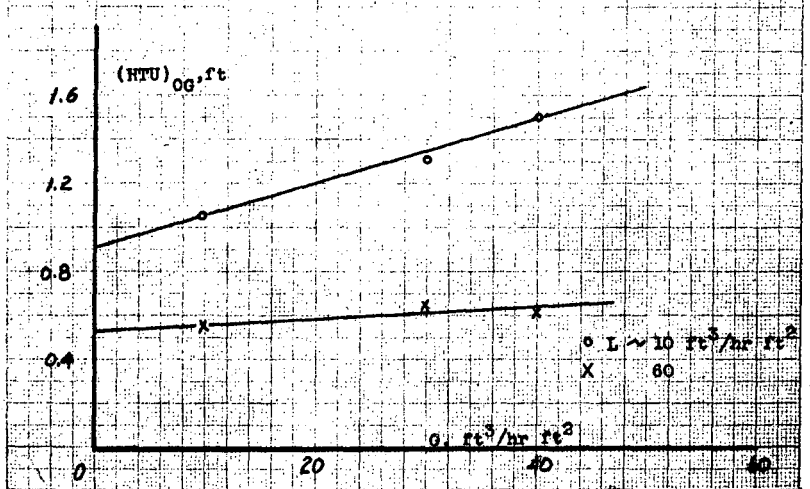
Data of Sherwood, Evans, and Langcor
Column packed with ⅜-inch carbon rings

System: Methyl isobutyl ketone - water - acetic acid

Phase continuous: Water



$(HTU)_{OG}$ vs. 1/L at constant G



$(HTU)_{OG}$ vs. G at constant L

MU 366

16049-1

MU 367

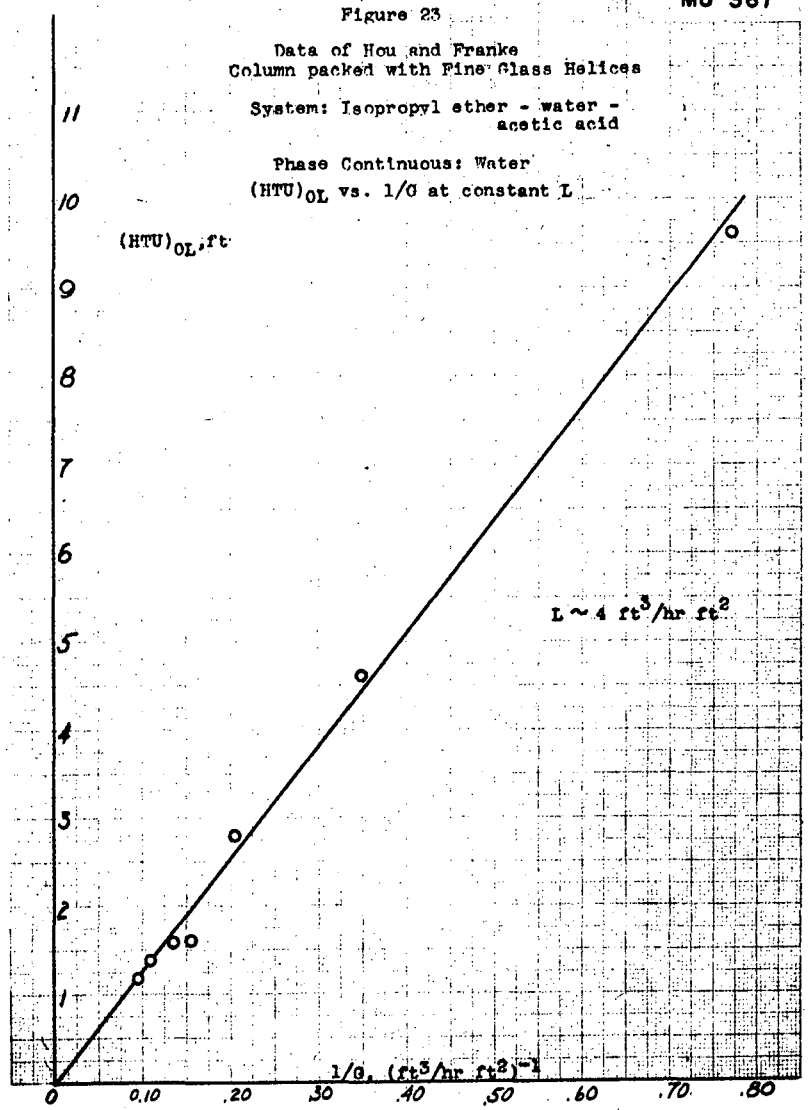
Figure 23

Data of Hou and Franke
Column packed with Fine Glass Helices

System: Isopropyl ether - water - acetic acid

Phase Continuous: Water

$(HTU)_{OL}$ vs. 1/G at constant L



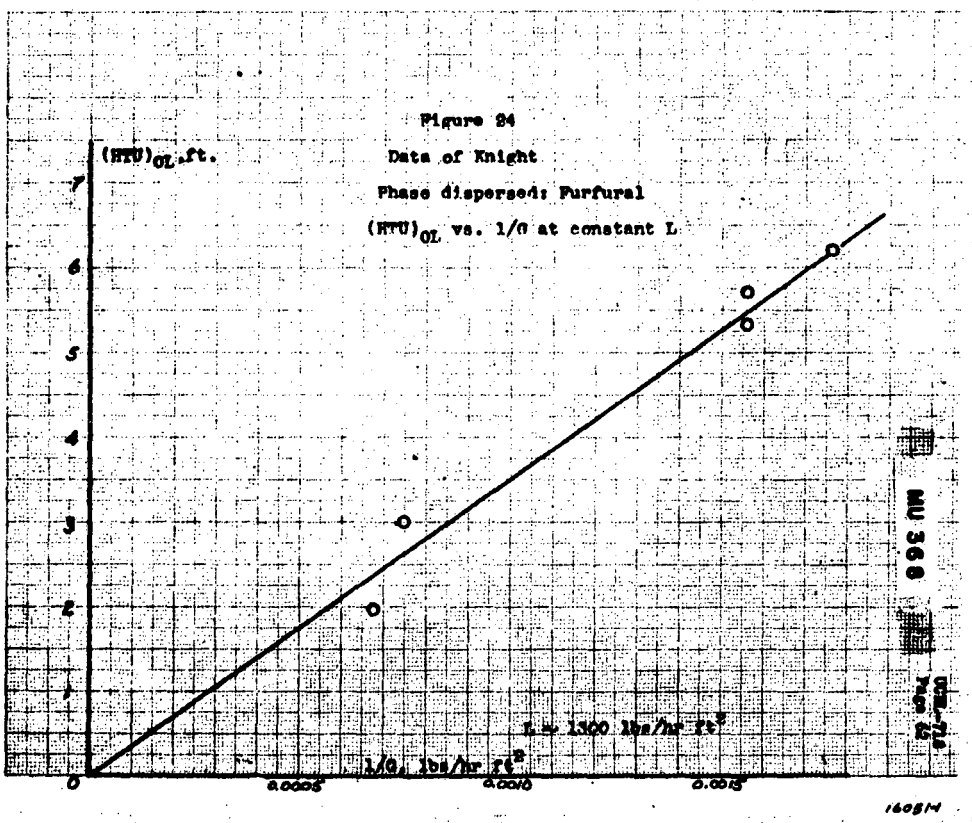


Figure 24

Data of Knight

Phase dispersed: Furfural

$(HTU)_{OL}$ vs. $1/G$ at constant L

MU 368

APPENDIX B

PHYSICAL PROPERTIES OF SOLVENTS

Table III
Physical Properties of Cyclohexanone[27] and Pentaether[28]

	Density	Viscosity	Boiling Point	Freezing Point	Flash Point	Solubility by Wt. In H_2O	Solubility by Wt. of H_2O in
Cyclo-hexanone	0.9748 20°C.		156.7°C.	-45°C.		1.02% 30°C.	
Penta-ether	0.9436 20°C.	5.7 cp. 20°C.	237°C. @ 50 mm.	-20°C.	355°F.	1.3% 20°C.	4.8% 20°C.
	0.938 25°C.						

Plots (a) Density vs. conc. for $UO_2(NO_3)_2$ in:

1. Water[29]
2. Pentaether
3. Cyclohexanone

(b) Optical density vs. conc. for $UO_2(NO_3)_2$ in:

1. H_2O
2. Pentaether
3. Cyclohexanone

(c) Equilibrium concentrations

1. $UO_2(NO_3)_2$ in penta vs. $UO_2(NO_3)_2$ in H_2O
2. $UO_2(NO_3)_2$ in cyclo vs. $UO_2(NO_3)_2$ in H_2O

(d) 1. Slope of pentaether equilibrium curve in mole ratio units vs. X

MU 379

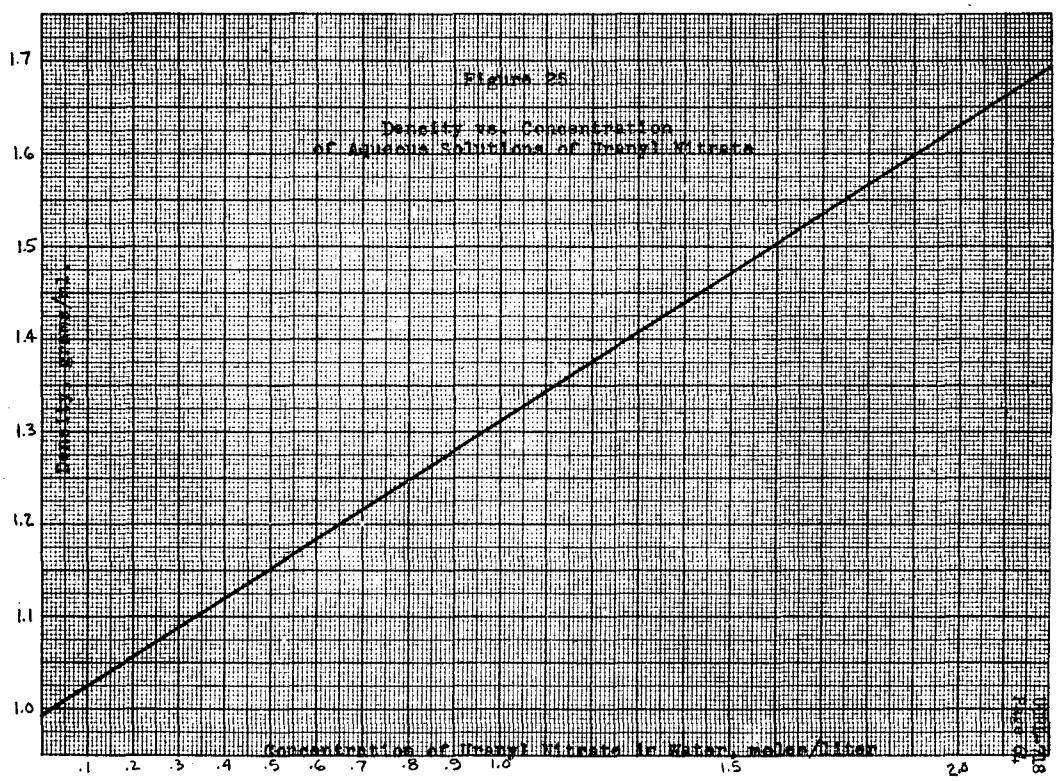
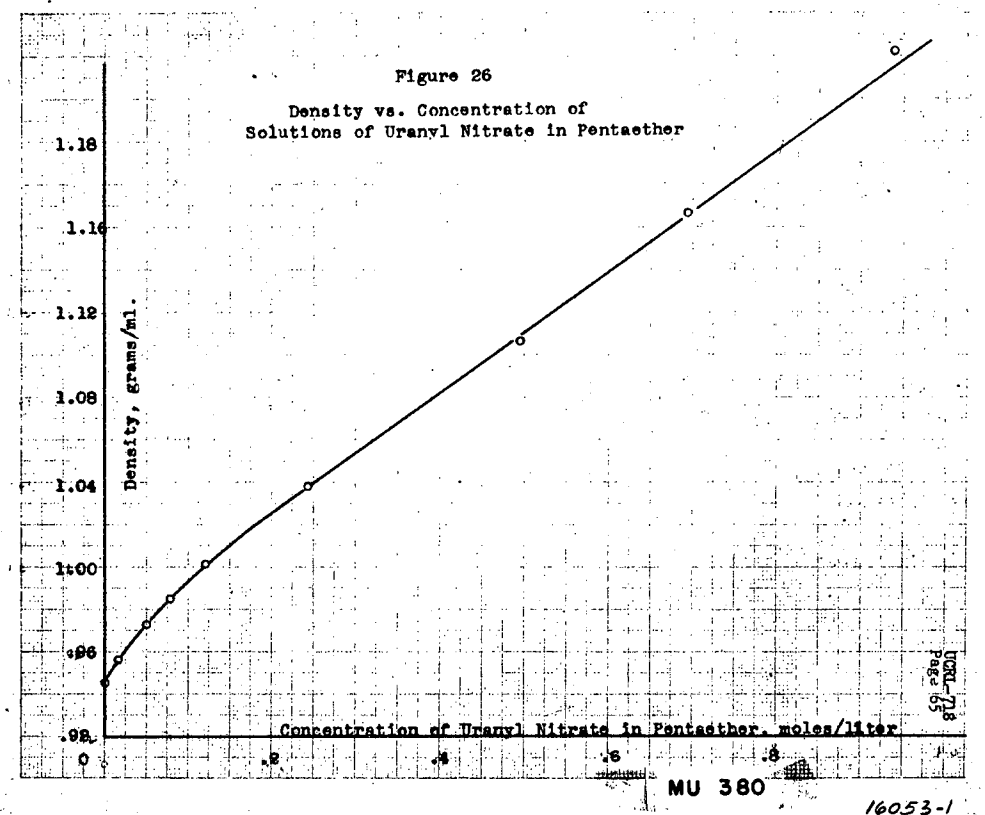


Figure 26
Density vs. Concentration
of Aqueous Solutions of Thorium Nitrate
Density, grams/cc.
1.7
1.6
1.5
1.4
1.3
1.2
1.1
1.0
Concentration of Thorium Nitrate in Water, moles/liter
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
1.5
2.0

Figure 26

Density vs. Concentration of
Solutions of Uranyl Nitrate in Pentaether



MU 380

16053-1

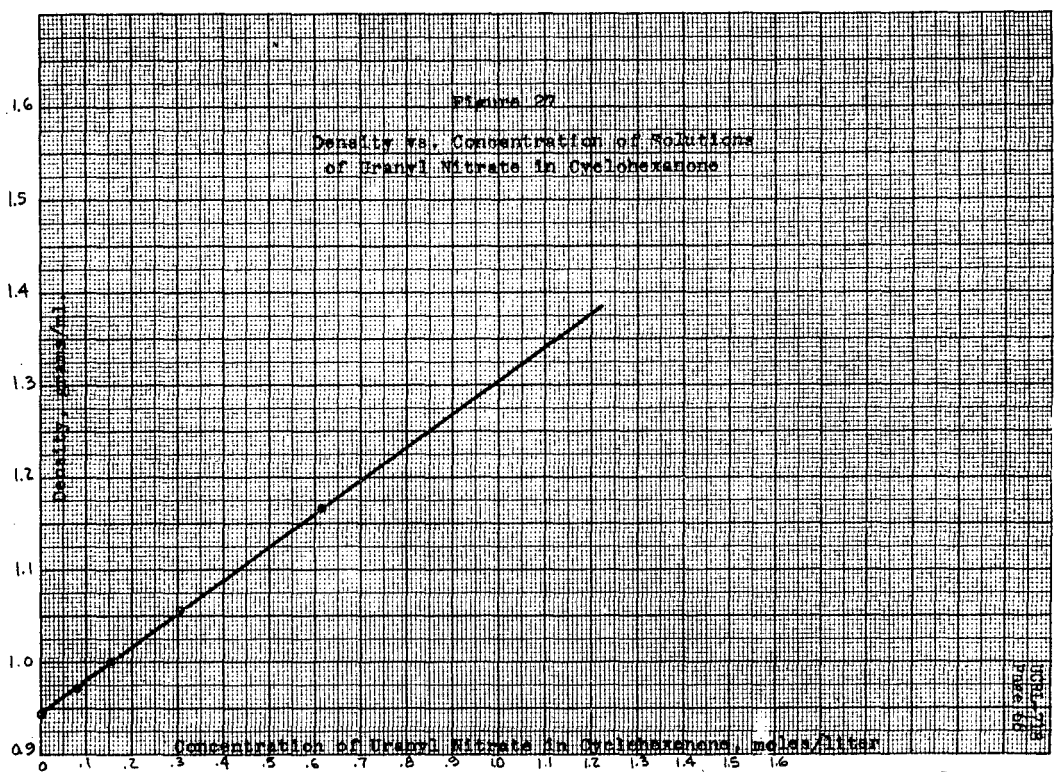


Figure 27
Density vs. Concentration of Solutions
of Uranyl Nitrate in Cyclohexanone
Density, grams/ml
1.6
1.5
1.4
1.3
1.2
1.1
1.0
0.9
Concentration of Uranyl Nitrate in Cyclohexanone, moles/liter
0
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
1.1
1.2
1.3
1.4
1.5
1.6
MU 381

16054-1

MU 382

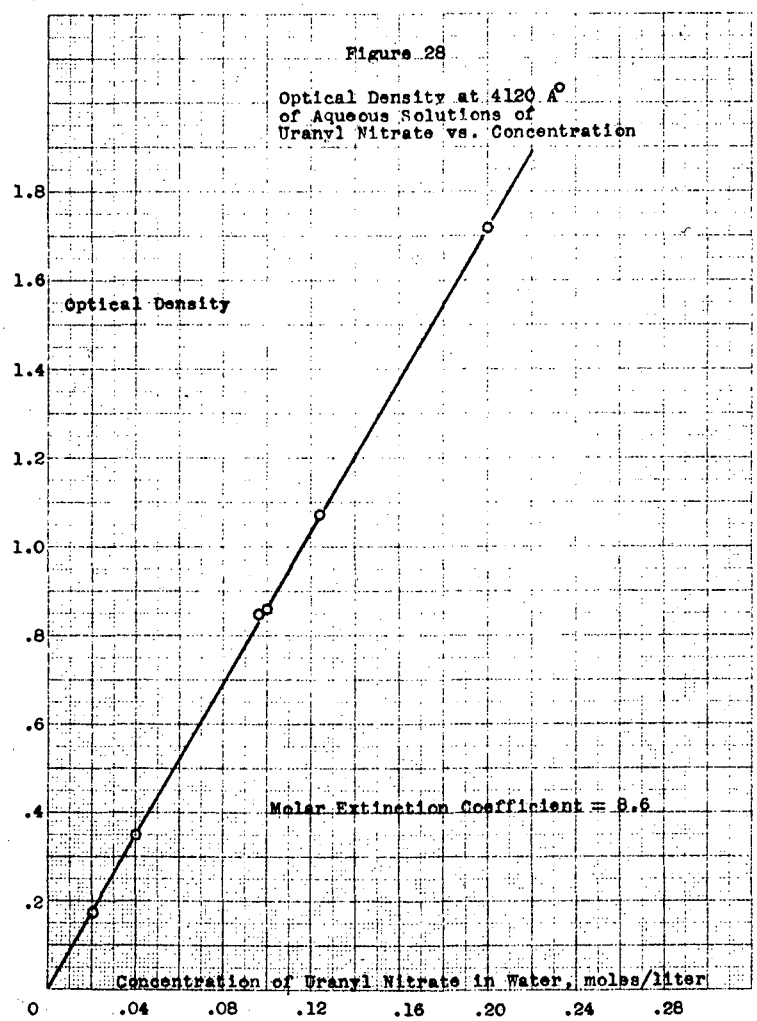


Figure 28
Optical Density at 4120 A°
of Aqueous Solutions of
Uranyl Nitrate vs. Concentration
Optical Density
Molar Extinction Coefficient = 8.6
Concentration of Uranyl Nitrate in Water, moles/liter
1.8
1.6
1.4
1.2
1.0
.8
.6
.4
.2
0
.04
.08
.12
.16
.20
.24
.28

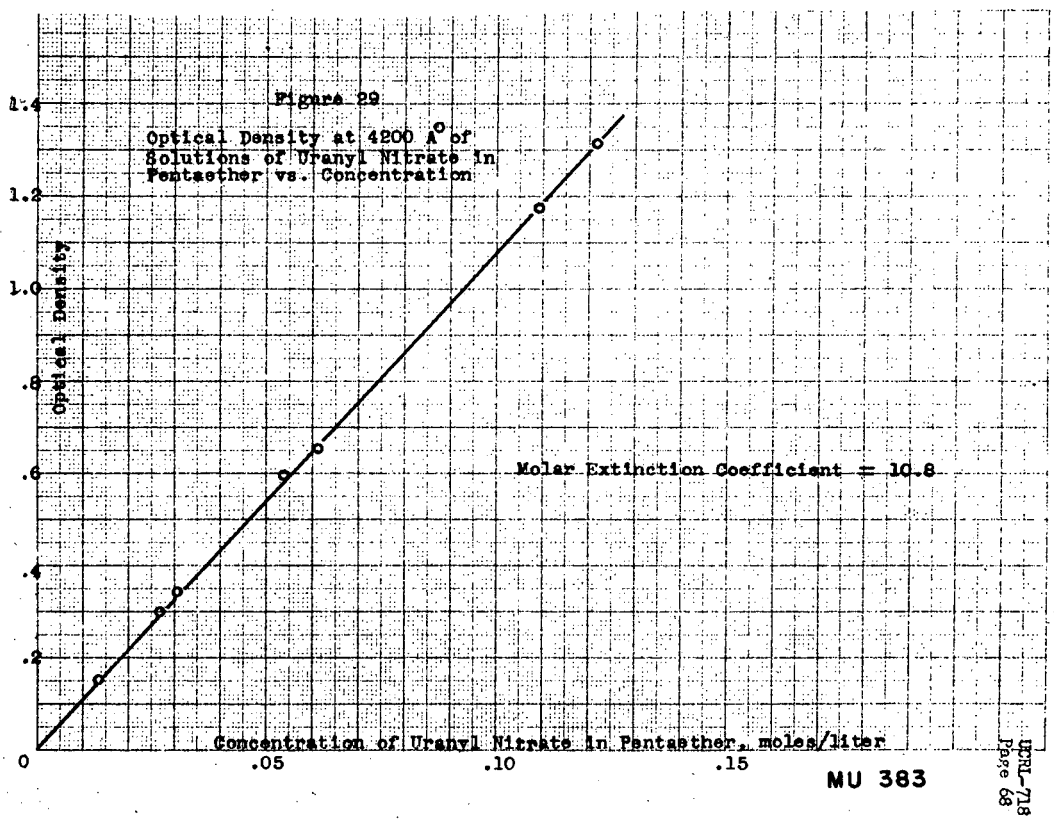


Figure 29

Optical Density at 4200 A° of Solutions of Uranyl Nitrate in Pentaether vs. Concentration

16056-1

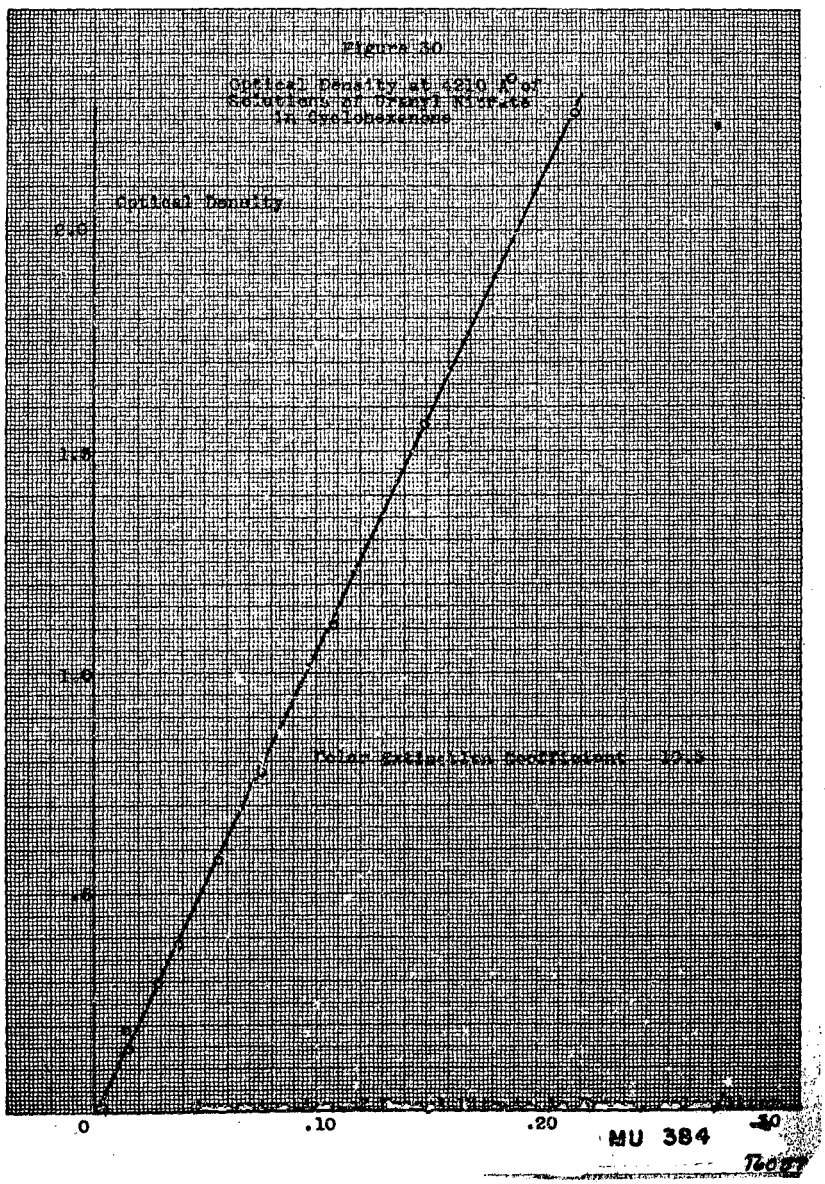


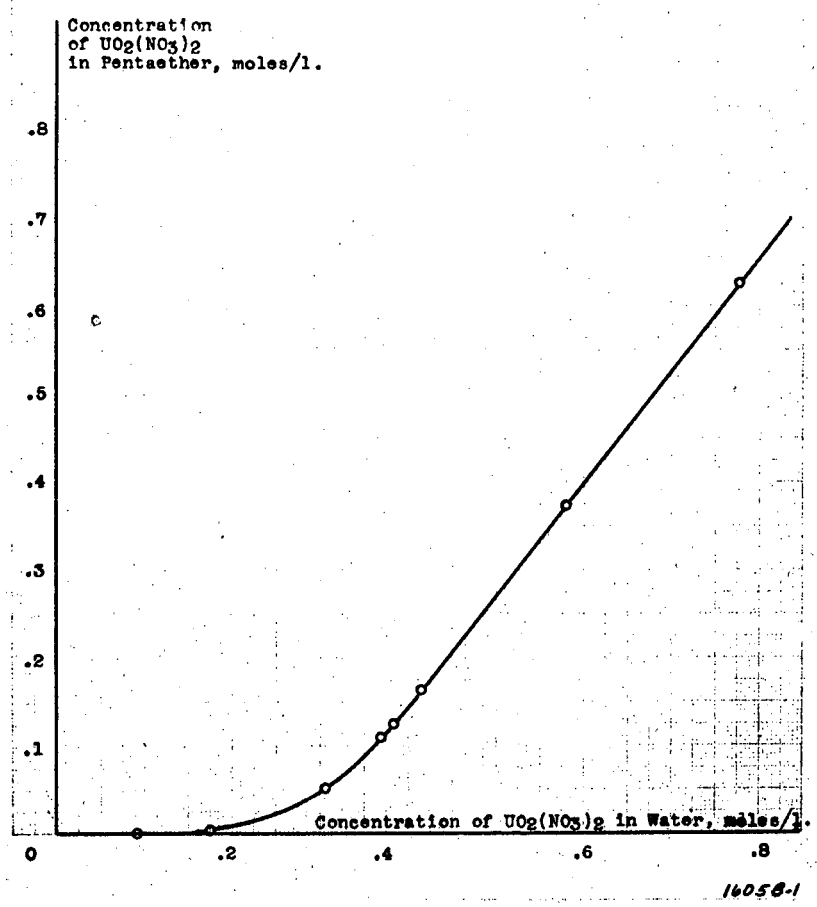
Figure 30
Optical Density at 4210 Å of
Solutions of Ethyl Nitrate
in Cyclohexane
Optical Density
2.0
1.0
.6
0
.10
.20
.30

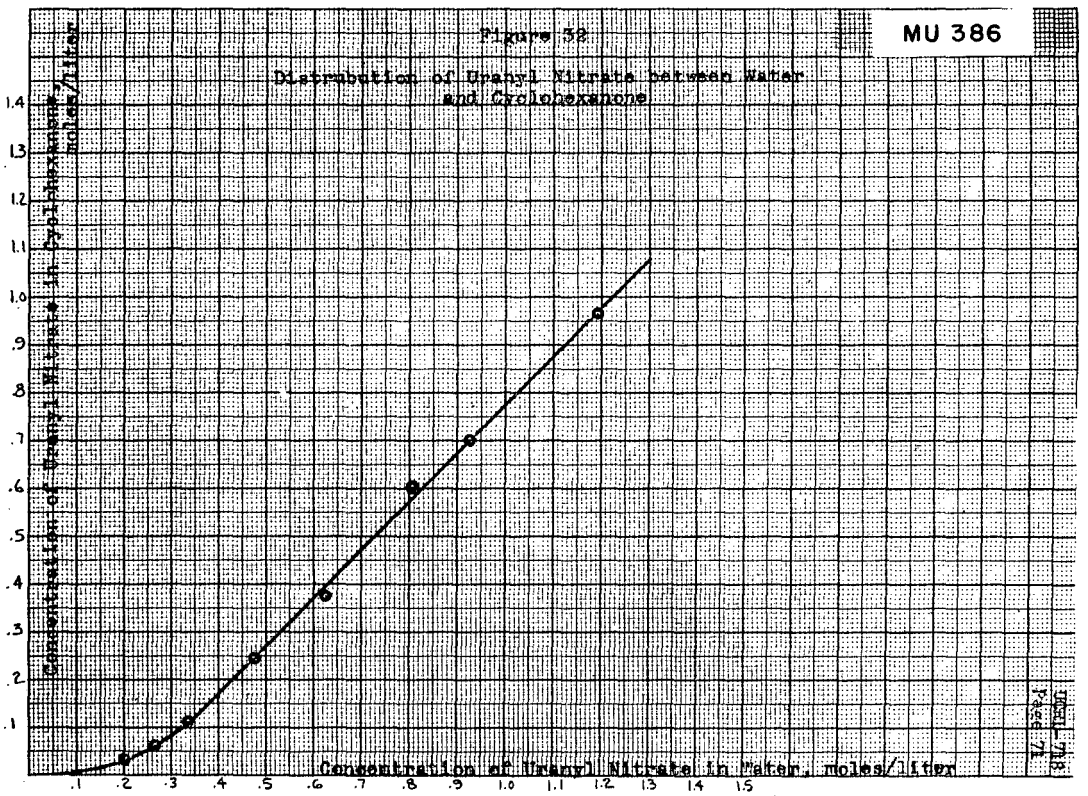
MU 384

MU 385

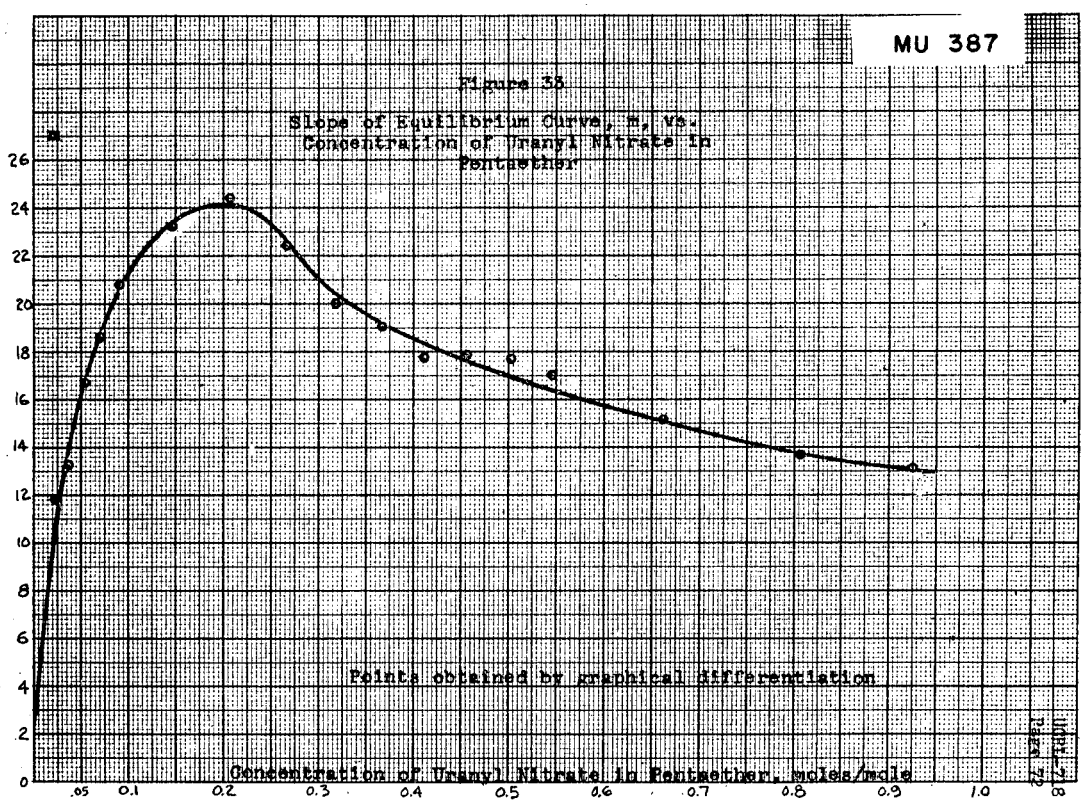
Figure 31

Equilibrium Distribution of Uranyl Nitrate between Water and Pentaether




MU 386
Figure 32
Distribution of Uranyl Nitrate between Water and Cyclohexanone
Concentration of Uranyl Nitrate in Cyclohexanone, moles/liter
Concentration of Uranyl Nitrate in Water, moles/liter
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
1.1
1.2
1.3
1.4
1.5

16059-1


MU 387
Figure 33
Slope of Equilibrium Curve, m, vs.
Concentration of Uranyl Nitrate in
Pentaether
m
26
24
22
20
18
16
14
12
10
8
6
4
2
0
Points obtained by graphical differentiation
Concentration of Uranyl Nitrate in Pentaether, moles/mole
.05
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
∞
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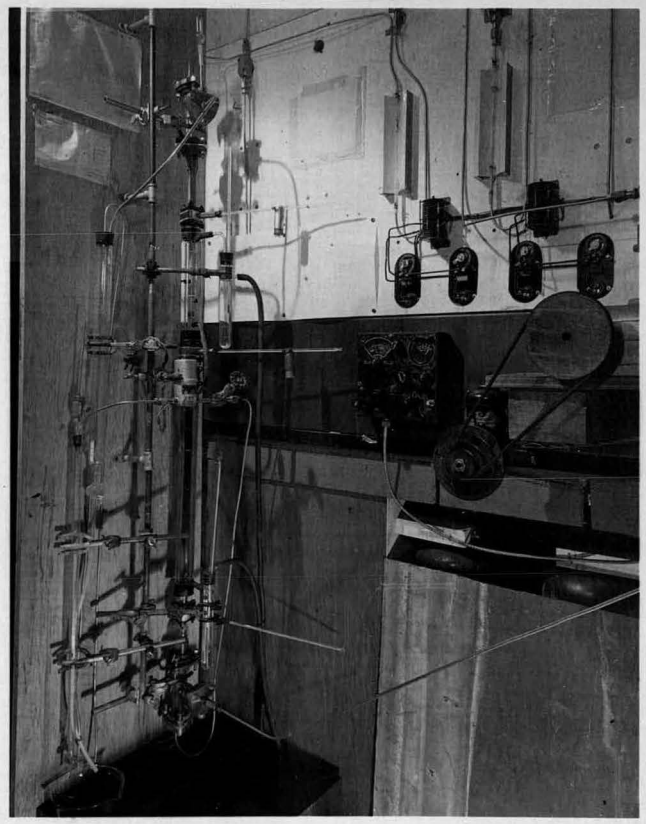
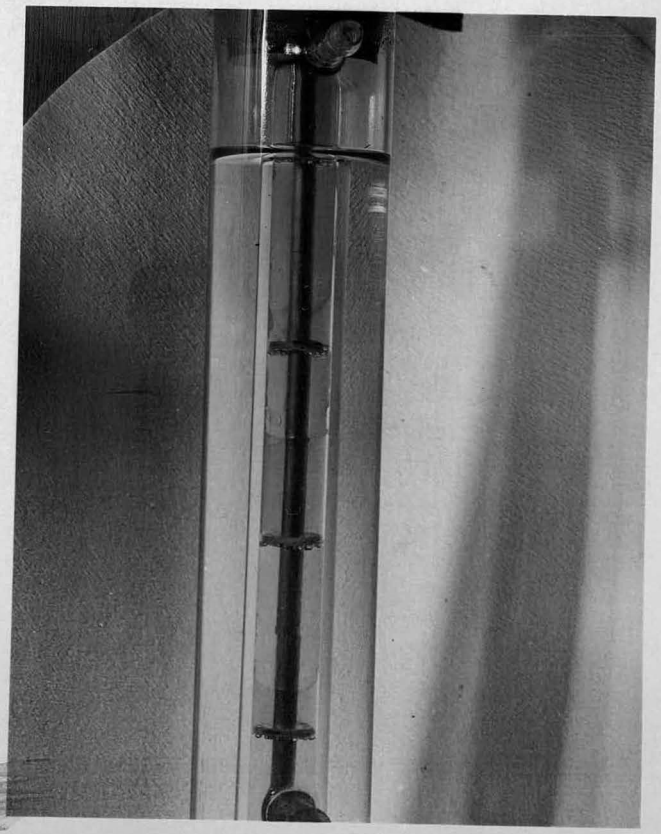
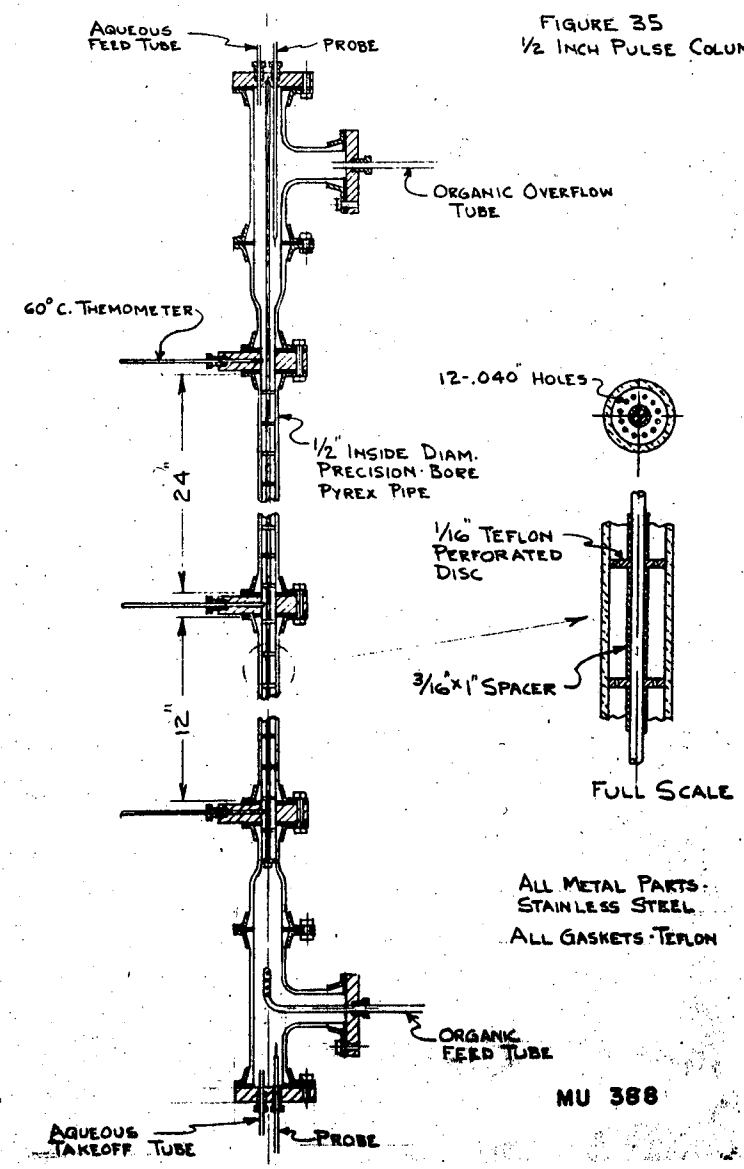




SECRET

FIG. 34

FIGURE 35
½ INCH PULSE COLUMN



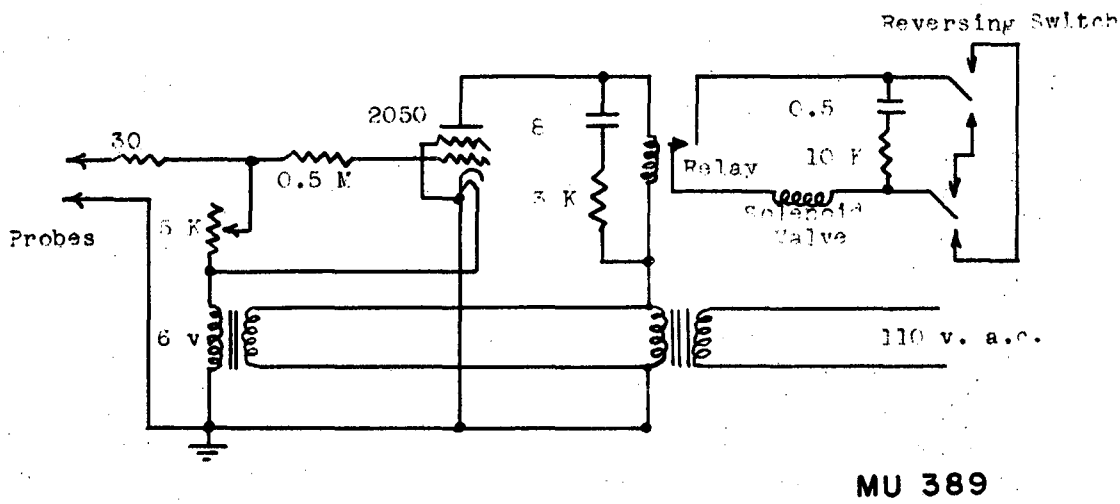


Figure 36

Electronic Interface
Controller

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