

TABLE OF CONTENTS

	Page
I. INTRODUCTION	2
II. LITERATURE REVIEW	4
Definitions and Concepts	4
Liquid-Liquid Extraction	4
Uses of Liquid-Liquid Extraction	5
Feed	7
Solute	7
Solvent	7
Carrier Solvent	7
Extract Phase	7
Extract	7
Raffinate Phase	8
Raffinate	8
Selectivity	8
Distribution	9
Extraction Stage	10
Ideal Stage	10
Stagewise Contact	10
Continuous Contact	10
Transfer Unit	11
Classification of Extraction Operation by Solvent Systems	12

	Page
Classification of Extraction Operations by Flow Sheets	13
Distillation Analogy	13
Equilibrium Relations	14
Distribution	14
Distribution for the System Carbon Tetra- chloride-Acetone-Water	17
Binodal Curves	20
Equilibrium in Extraction Processes	20
Rate Relations	24
Mass Transfer Concept	24
The Transfer Unit	25
Height Equivalent to a Theoretical Stage	27
Relation Between H.E.T.S. and H.T.U.	27
Methods of Calculation	29
Equipment	30
Liquid-liquid Extractors	30
Classification of Liquid Extractors	30
The Use of External Power in Extraction Equipment	32
Centrifugal Extractors	33
Film-Type Extractors	34
Spray-Tower Extractor	35
Packed Extraction Columns	36
Baffle Extractor	37
Perforated-Plate Extractor	38

	Page
Bubble-Cap Extractors	39
Mixer and Settler Extractors	40
The Van Dijk Column	41
The Feick Column	42
Pulse Column	43
Rate Relations and Flow Capacities	44
Flow Capacities	44
Rate Relations for Pulse Columns	45
Rate Relations for the Feick Column	47
Rate Relations for the Scheibel Column	47
Rate Relations for the Multistage Mixer Column.	48
III. Experimental	50
Purpose of Investigation	50
Plan of Experimentation	50
Literature Review	51
Design and Construction	51
Refractive Index Data Determination	52
Experimental Tests	52
Flooding Determinations	53
Correlation of Data	54
Materials	55
Acetone	55
Air	55
Carbon Tetrachloride	55
Water	55

	Page
Apparatus	56
Method of Procedure	69
Construction of the Column Frame	69
Construction of the Column Flanges	70
Construction of Bellows Flanges	72
Construction of Baffle Plates	72
Assembly of Column	72
Construction of Gas Assembly	76
Construction of the Push-Rod Frame	78
Construction of a Push-Rod Assembly	79
Construction of Storage Section	81
Tubing	81
Analytical Procedure	82
Determination of Refractive Index Data	83
Preparation of Column for Operation	84
Operation of Column	88
Experimental Tests	89
Completion of Tests	91
Data and Results	92
Sample Calculations	109
IV. DISCUSSION	125
Discussion of Results	125
Experimental Errors	125
Design Features and Operations of the Pulse Extractor	137

	Page
Limiting Liquid Flow Rates	142
Analysis of Stage Efficiency	151
Recommendations	169
Limitations	173
V. CONCLUSIONS	175
VI. SUMMARY	179
VII. BIBLIOGRAPHY	182
VIII. ACKNOWLEDGMENTS	192
IX. VITA	193

LIST OF TABLES

	Page
Table I. Comparison of Nomenclature for the Diffusional Operations of Extraction and Distillation	15
Table II. Distribution Data for the System Carbon Tetrachloride-Acetone-Water	18
Table III. Solubility Data for the System Carbon Tetrachloride-Acetone-Water	22
Table IV. Plate Efficiencies for a 1-1/2-inch Pulse Extractor Operating With a Pulse Frequency of 24 Per Minute and an Amplitude of 1.20 Inches	46
Table V. Bill of Materials for an Experimental Two-inch Pulse Extractor and Auxiliary Apparatus	59
Table VI. Distribution and Refractive-index Data for the System Carbon Tetrachloride-Acetone-Water at 22° C	93
Table VII. Operational Data for a Two-inch Experimental Pulse Extraction Column Extracting Acetone from Carbon Tetrachloride with Water	97
Table VIII. Summary of Acetone Material Balances for a Two-inch Experimental Pulse Extractor Extracting Acetone from Carbon Tetrachloride with Water	98
Table IX. Stage Efficiency for a Two-inch Experimental Pulse Extraction Column Extracting Acetone from Carbon Tetrachloride with Water	99
Table X. Results of the Analysis of the Two-inch Experimental Pulse-Extractor Efficiency Tests	101

	Page
Table XI. Comparison of Actual and Calculated Stage Efficiencies for a Two-inch Experimental Pulse Extractor	102
Table XII. Limiting Liquid Flow Rates in an Experimental Two-inch Pulse Extractor Using the System of Carbon Tetrachloride and Water	104

LIST OF FIGURES

	Page
Figure 1. Distribution Data for the System Carbon Tetrachloride-Acetone-Water	19
Figure 2. Solubility Data for the System Carbon Tetrachloride-Acetone-Water	21
Figure 3. Specific Gravity of Equilibrium Solutions for the System Carbon Tetrachloride-Acetone-Water	94
Figure 4. Refractive Index of Equilibrium Solutions for the System Carbon Tetrachloride-Acetone-Water	95
Figure 5. Distribution of Acetone Between Carbon Tetrachloride and Water at 22° C	96
Figure 6. Comparison of Experimentally Determined and Calculated Stage Efficiencies	103
Figure 7. Limiting Liquid Flow Rates in a Two-inch Pulse Extractor	105
Figure 8. Limiting Liquid Flow Rates in a Two-inch Pulse Extractor	107
Figure 9. Limiting Liquid Flow Rates in a Two-inch Pulse Extractor	108
Figure 10. Graphical Solution for the Number of Theoretical Stages	112
Figure 11. Effect of the Heavy-Phase Flow Rate on the Stage Efficiency of a Two-inch Experimental Pulse Extractor Extracting Acetone from Carbon Tetrachloride with Water	163
Figure 12. Effect of the Light-Phase Flow Rate on the Stage Efficiency of a Two-inch Experimental Pulse Extractor Extracting Acetone from Carbon Tetrachloride with Water	166

LIST OF DRAWINGS

		Page
Drawing 1.	Detail Drawing of Pulse Extractor	58
Drawing 2.	Detail Drawing of Pulse-Extractor Top Flange	60
Drawing 3.	Detail Drawing of Pulse-Extractor Bottom Flange	61
Drawing 4.	Detail Drawing of Pulse-Extractor Baffle Plate	62
Drawing 5.	Detail Drawing of Pulse-Extractor Top Bellows Flange	63
Drawing 6.	Detail Drawing of Pulse-Extractor and Push-Rod Frame	64
Drawing 7.	Detail Drawing of Pulse-Extractor Cam Assembly	66
Drawing 8.	Detail Drawing of Pulse-Extractor Push- Rod Assembly	67
Drawing 9.	Schematic Flow Diagram for Pulse Extractor.	86

I. INTRODUCTION

Liquid-liquid extraction is the unit operation that consists of the transfer of one or more components from one liquid phase to another liquid phase that is immiscible, or nearly immiscible, with the first phase. The many uses and the great importance of this operation are too well known to repeat in this introduction. For years the conventional apparatus used for liquid-liquid extraction has been towers employing countercurrent flow of the liquids and equipped with various means to increase the interfacial area. These columns are either filled with various types of packing; equipped with bubble-cap trays, sieve plates, or any of several arrangements of baffles; or utilize one or more spray heads. More recently mixer-settler combinations and centrifugal extractors have been developed.

The countercurrent columns suffer from the disadvantage that the only energy available for the maintenance of the large interfacial area is the density differences of the two liquid phases. Thus the available energy, no matter how efficiently it is utilized by skillful design of packing or baffles, is too small to overcome drop coalescence and the resulting decrease in the area of contact. The many mixer-settler type of extractors, while offering a partial solution, suffer from the disadvantage that they are inherently batch extractors in operation. Recently several new

Attention Patron:

Page 2 omitted from
numbering

types of extractors have been developed that combine the desirable countercurrent features of the towers and the good mixing of the mixer-settler combinations. These devices usually take the form of a number of mixing sections, mounted one above another to form a column, separated by packed or baffled settling sections. An alternative design is to use an unpacked column with the fluid being forced through perforated baffles by mixers.

Another recent design for the improvement of liquid-liquid extraction equipment is a column utilizing countercurrent flow, mechanical agitation, and baffled settling zones. The column is fitted with sieve plates that may be moved up and down relative to the liquid producing the agitation, while the liquids flow countercurrently through the plates. Settling takes place during the pauses between the strokes. The operation and construction of this column can be simplified by using stationary plates and agitating by imparting a reciprocating motion to the liquids relative to the plates.

The purpose of this investigation was the design, construction, and operation of a liquid-liquid pulse extractor. During the operation of the column the effect of the flow rates, frequency of the pulses, and endurance of the pulses on extraction efficiency was studied. In addition to this data, flooding flow rates of the extractor were determined.

II. LITERATURE REVIEW

According to Treybal(68) three major factors must be considered in the design of an extraction process. These factors are the phase equilibrium relations, the rate at which extraction proceeds, and the equipment used. These three factors will be considered in this review after some general concepts and definitions are covered.

Definitions and Concepts

The following definitions and general concepts are given with a twofold purpose in mind: First, to familiarize the reader with the broad principles and general theory underlying liquid-liquid extraction; and second, to present the meaning in which some controversial terms and ideas will be used in this review.

Liquid-liquid Extraction. One of the problems that the chemical engineer has to face frequently is the separation of the components of a liquid solution(68). The engineer has his choice of several techniques when selecting a method of separation; however, one method is usually advantageous either because of certain properties of the system and/or because of the economics involved. In general, it is desired to effect this separation by physical methods rather than by chemical methods. A few of the methods of

separation that might be mentioned are distillation, evaporation, adsorption, and liquid-liquid extraction.

All of these separation techniques depend upon the tendency of a substance when distributed between two phases to occur in different concentrations in each of the phases at equilibrium⁽⁶⁸⁾. Liquid-liquid extraction⁽²²⁾ is the separation process that consists of the transfer of one or more components from one liquid phase to another liquid phase.

Liquid-liquid extraction may be carried out in several different ways. The most important way is to add another liquid that is immiscible, or nearly immiscible, with one or more of the components of the original mixture⁽⁶⁸⁾. Alternately, the original solution may be heated or cooled to form two phases⁽⁶⁸⁾; a third component, that takes no direct part in the extraction method, may be added to cause the original mixture to form two phases⁽⁶⁸⁾; or, finally, the mixture may be treated with two immiscible liquids each of which dissolves a separate component of the original mixture⁽²²⁾.

Uses of Liquid-liquid Extraction. According to Elgin⁽²²⁾ extraction may be preferable to other separation operations when the following conditions are present: The boiling points of the components of the liquid mixture are within a very few degrees of each other, one or more of the liquids are sensitive to heat, a comparatively small amount of solute is present in the original mixture and the solute is less volatile than the other components, and/or when the solvent used has a lower heat of vaporization than

the original solution. Brown⁽⁷⁾ adds the obvious fifth condition; that is, when the two liquids are non-volatile or nearly so.

Treybal⁽⁶⁸⁾ points out that before deciding to use any extraction operation the following points must be considered. First, the addition of more components has several disadvantages; namely, a large amount of capital is tied up in inventories, the corrosion problem is increased, the handling and storage problems are increased, and the danger of contamination of the final product is increased. Secondly, the extraction process must be followed by a solvent recovery process, usually distillation. This solvent recovery step quite often represents a major portion of the final cost of the extraction process.

Benedict⁽³⁾ points out that all physical separation processes are ones in which power is expended to perform a separation. In extraction the principal use of power is in the form of heat needed to recover the solvent. Thus, if the power expended to effect a given separation is the least for extraction this is the most economical process. This should not be used as the sole criteria of selection⁽⁴⁴⁾ as other factors, such as solvent loss, may become important.

A few of the many industrial uses of liquid-liquid extraction that should be mentioned are the refining of lubricating oils⁽¹⁾, the extraction of acetic acid from pyroligneous acid liquors⁽²⁸⁾, the recovery of phenol⁽⁴⁵⁾, the purification and concentration of

vitamins and antibiotics⁽⁶⁷⁾, the refining of vegetable oils^(27, 35), and the recovery of alcohols from Fischer-Tropsch synthesis gas⁽³⁷⁾.

Feed. The original liquid mixture to be separated is referred to as the feed⁽²²⁾.

Solute. The solute⁽²²⁾ is the component or components of the feed to be extracted. The solute is not necessarily the desired final product. In the refining of lubricating oils, for example, impurities are extracted from the oil stocks. The impurities were extracted, and hence, were the solute, but were not the desired final product.

Solvent. The solvent⁽¹¹⁾ is the liquid that is added to form the second phase. If no other liquid is added the solvent is the component of the extract phase that occurs in the greatest concentration, exclusive of the solute.

Carrier Solvent. The carrier solvent⁽¹¹⁾ consists of all the components in the original feed except the solute.

Extract Phase. The extract phase⁽⁶¹⁾ consists of the solvent after it has contacted and has been separated from the other phase. This layer contains, in addition to the solvent, the solute and whatever other components it has picked up. This phase is often referred to as the solvent-rich phase.

Extract. The extract phase after the solvent has been removed is referred to as the extract⁽⁶¹⁾. The extract composition is equal to the extract-phase composition on a solvent-free basis.

Raffinate Phase. The raffinate phase⁽⁶¹⁾ consists of the original feed after it has been contacted and separated from the solvent. This phase contains the solute that was not extracted by the solvent as well as any solvent that has been picked up. This phase is often referred to as the carrier-solvent-rich phase.

Raffinate. The raffinate phase after the solvent has been removed is called the raffinate⁽⁶¹⁾. The raffinate composition is equal to the raffinate-phase composition on a solvent-free basis, the two being equal when the extract phase and raffinate phase are immiscible.

Selectivity. The solvent is said to be selective⁽⁶¹⁾ for that component of the feed which occurs in the greater ratio in the extract than in the feed. That is, the solvent is selective for the solute by definition.

The selectivity of any solvent, S, for any solute, A, is given by the following equation⁽⁷⁰⁾:

$$\beta = \frac{X_{AS}/X_{CS}}{X_{AC}/X_{CC}}$$

where:

- β = selectivity coefficient, dimensionless
- X_{AS} = concentration of solute in extract phase, weight fraction
- X_{CS} = concentration of carrier solvent in extract phase, weight fraction
- X_{AC} = concentration of solute in raffinate phase, weight fraction
- X_{CC} = concentration of carrier solvent in raffinate phase, weight fraction.

For extraction to take place in the desired direction, the selectivity coefficient must be greater than unity⁽⁷⁾.

Distribution. Glasstone⁽²⁶⁾ states the distribution law in the following manner: "If to a system of two liquid layers, made up of two immiscible or slightly miscible components, is added a quantity of a third substance soluble in both layers, then the substance is found to distribute, or divide, itself between the two layers in a definite manner." The definite manner being that the ratio of solute concentration in the two phases is constant. It was once thought that this ratio was independent of the absolute concentration; but it is now known that this fact is true for very few systems, and in these systems, only at low concentration⁽⁶⁹⁾.

The distribution coefficient is given by the following equation⁽²⁶⁾:

$$m = \frac{X_{AS}}{X_{AC}}$$

where:

- m = distribution coefficient, dimensionless
- X_{AS} = concentration of solute in the extract phase, weight fraction
- X_{AC} = concentration of solute in the raffinate phase, weight fraction.

The distribution coefficient, m , and the selectivity coefficient, β , are often confused. The distribution coefficient expresses the equilibrium between the two phases in liquid-liquid extraction. The selectivity coefficient expresses the relative enrichment of the solute in the extract phase compared to the raffinate phase. The selectivity coefficient is equivalent to the relative volatility in distillation⁽²⁴⁾.

Extraction Stage. Brown⁽⁷⁾ states that the physical operation of extraction consists of two steps. These steps are, first, the mixing together of the two phases, with a longer or shorter time allowed for equilibrium to be reached, and, second, the separation of the two phases. Each time that the two phases are mixed and separated once, there has been one extraction stage.

Ideal Stage. If after the extraction stage the two phases leave in equilibrium, that is, if the solute is distributed as required by the distribution law, the stage is called an ideal stage⁽²²⁾. An ideal stage represents the limiting amount of extraction for any stage.

Stagewise Contact. In any extraction process, if the two liquid phases are alternately mixed and separated the process is said to be one of stagewise contact⁽⁵⁸⁾. A more rigorous definition of a stagewise extractor would be any extractor in which the interfacial area between the two phases varies with position in the equipment or with time. The classic example of stagewise contact is the bubble-cap column. If a section is taken through this column under the liquid covering the bubble caps a large interfacial area is found; however, if a section is taken through the downcomers and the disengaging section almost no interfacial area is found.

Continuous Contact. Instead of bringing the solution to be extracted and the extracting solvent into contact in separate stages with intermediate settling, it is possible to cause the liquid phases to flow continuously past each other. This type of contact is called

continuous contact⁽⁵⁸⁾. Continuous contact may be further defined as any extraction process in which the interfacial area does not vary substantially with position in the column or with time during the operation of the column.

To avoid further confusion it must be thoroughly understood that the term "continuous" is used with two different meanings in extraction. One meaning is defined above. The second meaning is in the sense of a continuation of time. This point seems to cause great confusion in the literature. Wherever the word "continuous" appears care should be taken to see in which way it is used. Is the column a continuous extractor or does the extractor merely operate without stopping? Continuous when used in this review with the latter meaning will be set off by quotation marks.

Transfer Unit. Colburn^(15, 16) points out that the concept of "ideal stages" is not applicable to continuous contact. The difference in the physical mechanism of contact invalidates the ideal stage calculation. He proposed the use of the transfer unit as a basis with which to compare the efficiency of the continuous extraction process. The Colburn transfer unit is the concentration change experienced per unit of concentration difference causing this change. The transfer unit and the ideal stage have about the same meaning; although the former applies to continuous and the latter to stagewise contact.

Classification of Extraction Operations by Solvent Systems.

A convenient method of classifying extraction operations according to Treybal⁽⁷¹⁾ is as follows:

- I. Single solvent systems, including all those which consist of or which may be reduced to the equivalent of, three components, the two to be separated and the solvent.
 - A. Stagewise contact
 - B. Continuous contact
- II. Mixed solvents
 - A. Stagewise contact
 - B. Continuous contact
- III. Double solvents (fractional extraction)
 - A. Stagewise contact
 - B. Continuous contact

Classification of Extraction Operations by Flow Sheets.

Elgin⁽²⁰⁾ has proposed a more useful classification of extraction operations depending upon the arrangement of the equipment. His classification is as follows:

- I. Single contact
- II. Multiple contact
 - A. Co-current
 - 1. Multistage
 - 2. Differential
 - B. Countercurrent
 - 1. Continuous
 - a. with reflux
 - b. without reflux
 - 2. Multistage
 - a. with reflux
 - b. without reflux

Differential co-current contact is the limiting case of multistage co-current contact in which the solvent is added in differential portions and the extract removed as fast as it is formed. Its inclusion as a separate class is due to the work of Treybal⁽⁷¹⁾ and Varteressian and Fenske⁽⁷⁶⁾. The familiar Soxhlet extractor is an example of a differential co-current extractor⁽⁷⁸⁾.

Distillation Analogy. It is sometimes helpful to consider the close relation between distillation and extraction⁽⁷¹⁾. This

is particularly true for the average chemical engineer because of his greater familiarity with the distillation process. In distillation a mixture of substances is separated by the creation of a second phase by heat. In extraction the second phase is formed by the addition of the solvent. In Table I, page 15, some further similarities of the two processes are presented.

Equilibrium Relations

In studies on chemical engineering equipment the equilibrium relations of the systems involved are important as a basis for comparisons and further calculations. By equilibrium relations is meant the studies on distribution and phase relations.

Distribution. The equilibrium that is of importance in liquid-liquid extraction is the distribution equilibrium⁽⁶⁹⁾. The ideal form of the distribution law presented on page 9, however, is inadequate in liquid-liquid extraction.

Hernst⁽¹²⁾ suggested that all deviations from the distribution law are caused by either association or disassociation of the solute in either or both phases. Hernst's explanation may well be correct but his equations become impossibly cumbersome when applied to most systems. This is because deviations, if caused by the mechanism postulated by Hernst, are also complicated by a change in the relative amount of disassociation or association when the absolute concentration of the solute changes.

TABLE I

Comparison of Nomenclature for the Diffusional Operations
of Extraction and Distillation^a

<u>Extraction Nomenclature</u>	<u>Distillation Analogy</u>
Addition of solvent	Addition of heat
Solvent mixer	Reboiler
Removal of solvent	Removal of heat
Solvent separator	Condenser
Solvent-rich solution saturated with solvent	Vapor at the boiling point
Solvent-rich solution containing more solvent than that required to saturate it	Superheated vapor
Solvent-lean solution containing less solvent than that required to saturate it	Liquid below the boiling point
Solvent-lean solution saturated with solvent	Liquid at the boiling point
Two-phase liquid mixture	Mixture of liquid and vapor
Selectivity	Relative volatility
Change of temperature	Change of pressure

a. Treybal, R. E.: "Liquid Extraction," p. 129. McGraw-Hill Book Co., Inc., New York, N. Y. 1951.

Brancker, Hunter, and Nash⁽⁶⁾ found that the mol fraction of A in the A-rich phase when plotted against the equilibrium concentration of B in the B-rich phase gave a curved line. However, if the coordinates of the concentration of A in the A-rich phase are adjusted to obtain a straight line these same coordinates will give a straight line for most other systems. This method is similar to the method of Cox for vapor pressure data. The authors state that solutropic systems cannot be correlated by this method. A solutropic system⁽⁶²⁾ is one in which the distribution ratio changes from greater than one to less than one, or vice versa, as the concentration of solute changes.

Bachman⁽²⁾ found that the equations of the straight lines on the Brancker, Hunter, and Nash coordinates are as follows:

$$X_{AA}X_{BB} = aX_{AA} + bX_{BB}$$

where:

- X_{AA} = concentration of solvent in extract phase, weight fraction
- X_{BB} = concentration of carrier solvent in raffinate phase, weight fraction
- a = constant, weight fraction
- b = constant, weight fraction.

Thus, if X_{BB} is plotted against X_{BB}/X_{AA} on rectangular coordinate paper straight lines should be obtained.

Othmer and Tobias⁽⁴⁹⁾ found that a plot of equilibrium values of $1-X_{AA}/X_{AA}$ against $1-X_{BB}/X_{BB}$ on double logarithmic coordinate paper produced straight lines.

The three previous modifications of the distribution law all suffer from the disadvantage that the concentration of the solute, C_2 , is not in the expression. Hand⁽³⁰⁾ showed that a double logarithmic plot of X_{CA}/X_{AA} against X_{CB}/X_{BB} is usually rectilinear. This method is generally considered to be the most satisfactory of the many forms of the distribution law⁽⁴⁶⁾. Hand's method amounts to writing the distribution law in the form shown below.

$$\frac{X_{CB}}{X_{BB}} = k \left(\frac{X_{CA}}{X_{AA}} \right)^n$$

where:

- X_{CB} = concentration of solute in raffinate phase, weight fraction
- X_{BB} = concentration of carrier solvent in raffinate phase, weight fraction
- X_{CA} = concentration of solute in extract phase, weight fraction
- X_{AA} = concentration of solvent in extract phase, weight fraction
- k = constant, dimensionless
- n = constant, dimensionless

Distribution for the System Carbon tetrachloride-Acetone-Water.

The distribution data⁽¹⁰⁾ for the system carbon tetrachloride-acetone-water at several temperatures are given in Table II, page 18. In Figure 1, page 19, the distribution curve is drawn on rectangular coordinates. As this system is solutropic, it was found that it could not be represented by the Hand equation.

TABLE II

Distribution Data for the System
Carbon Tetrachloride-Acetone-Water^a

Temperature	Acetone concentration	
	Carbon tetrachloride phase	Water phase
°C	wt %	wt %
30	0.80	3.35
30	3.95	10.30
30	4.60	11.50
30	13.90	22.30
30	16.50	24.40
30	24.20	30.40
30	33.30	35.80
30	39.60	40.20
30	47.20	46.00
30	51.90	49.80
30	59.04 ^b	59.04 ^b
20.8	2.10	6.90
20.8	6.20	14.30
20.8	13.80	22.50
4	2.21	9.50
4	4.90	14.90
4	9.00	20.60
4	13.80	25.30

a. Buchman, R. H.: Liquid-Liquid Equilibrium Data for the System Carbon-Tetrachloride-Acetone-Water, Ind. Eng. Chem., 44, 2449 (1952).

b. Plait point.

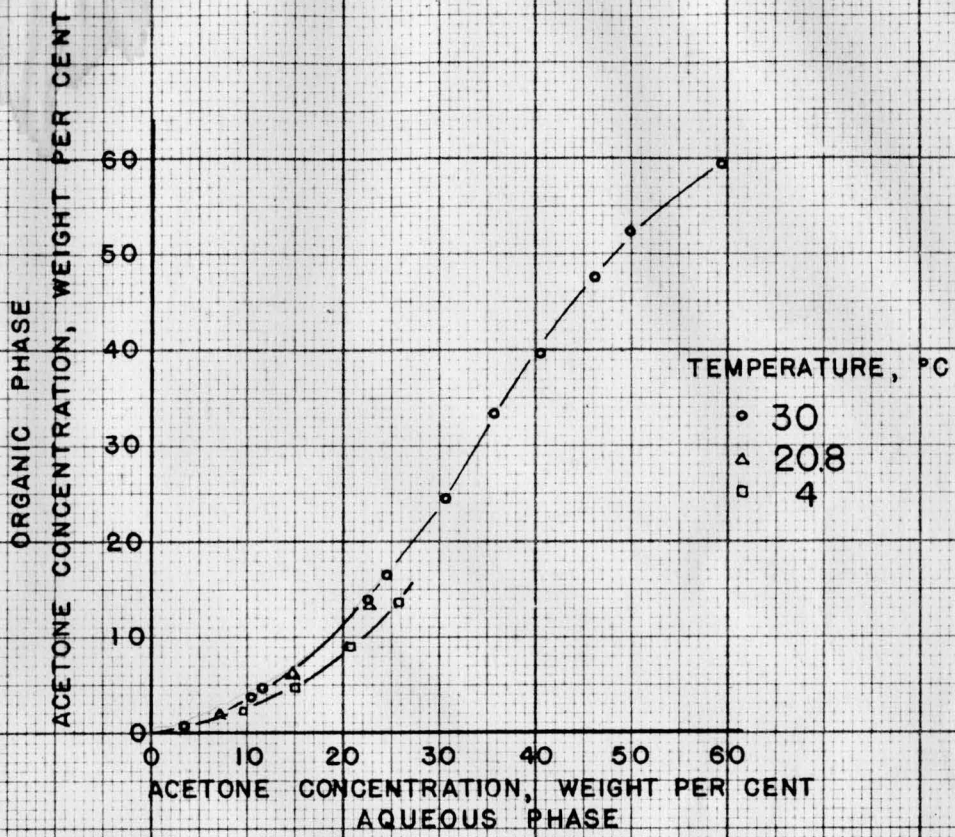


FIGURE I. DISTRIBUTION DATA FOR THE SYSTEM CARBON TETRACHLORIDE-ACETONE-WATER

BUCHMAN, R. H.: LIQUID-LIQUID EQUILIBRIUM DATA FOR THE SYSTEM CARBON TETRACHLORIDE-ACETONE-WATER, IND. ENG. CHEM., 44, 2449 (1952).

Attention Patron:

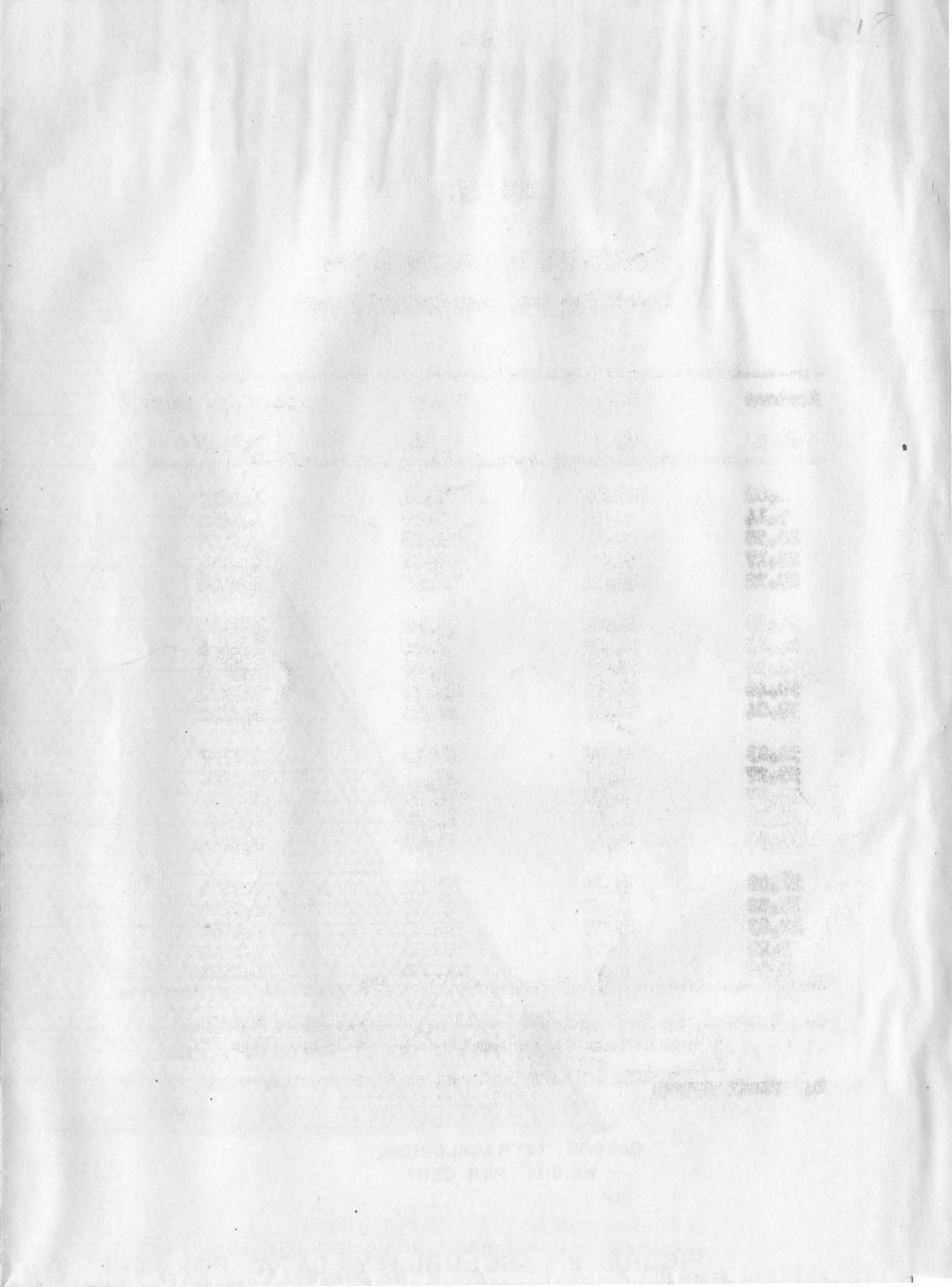
Page 18 repeated in numbering

Binodal Curves. In addition to the distribution data the composition of the saturated solutions of A and C in B and of B and C in A are needed⁽²³⁾. This type of information is usually plotted on a triangular composition diagram. The use of this type of diagram for presenting solubility and phase composition data for ternary systems has been adequately covered by Bull and Coli⁽¹¹⁾, Brown⁽⁸⁾, Elgin⁽²³⁾, and Treybal⁽⁷¹⁾. An example of this type of diagram for the system carbon tetrachloride-acetone-water is presented in Figure 2, page 21, from the data presented in Table III, page 22.

Equilibrium in Extraction Processes. Extraction may be carried out either in discreet steps or as a continuous counter-current process. The former process is called countercurrent stagewise extraction, and the latter process is called continuous countercurrent extraction. The equilibrium relations, representing optimum operation, are of course different for these two processes.

Equilibrium in Stagewise Extraction. If, in any stage-wise contactor, the two exit streams are in equilibrium the stage is called an ideal stage⁽⁶¹⁾. In actual practice the stages are always less than perfect, and the two liquids leave each stage before equilibrium has been reached. This, of course, suggests that the various types of stages or contactors can be compared by means of their relative approach to equilibrium. For this purpose the stage efficiency is defined

Figure 2. Solubility Data for the System Carbon
Tetrachloride-Acetone-Water



ACETONE,
WEIGHT PER CENT

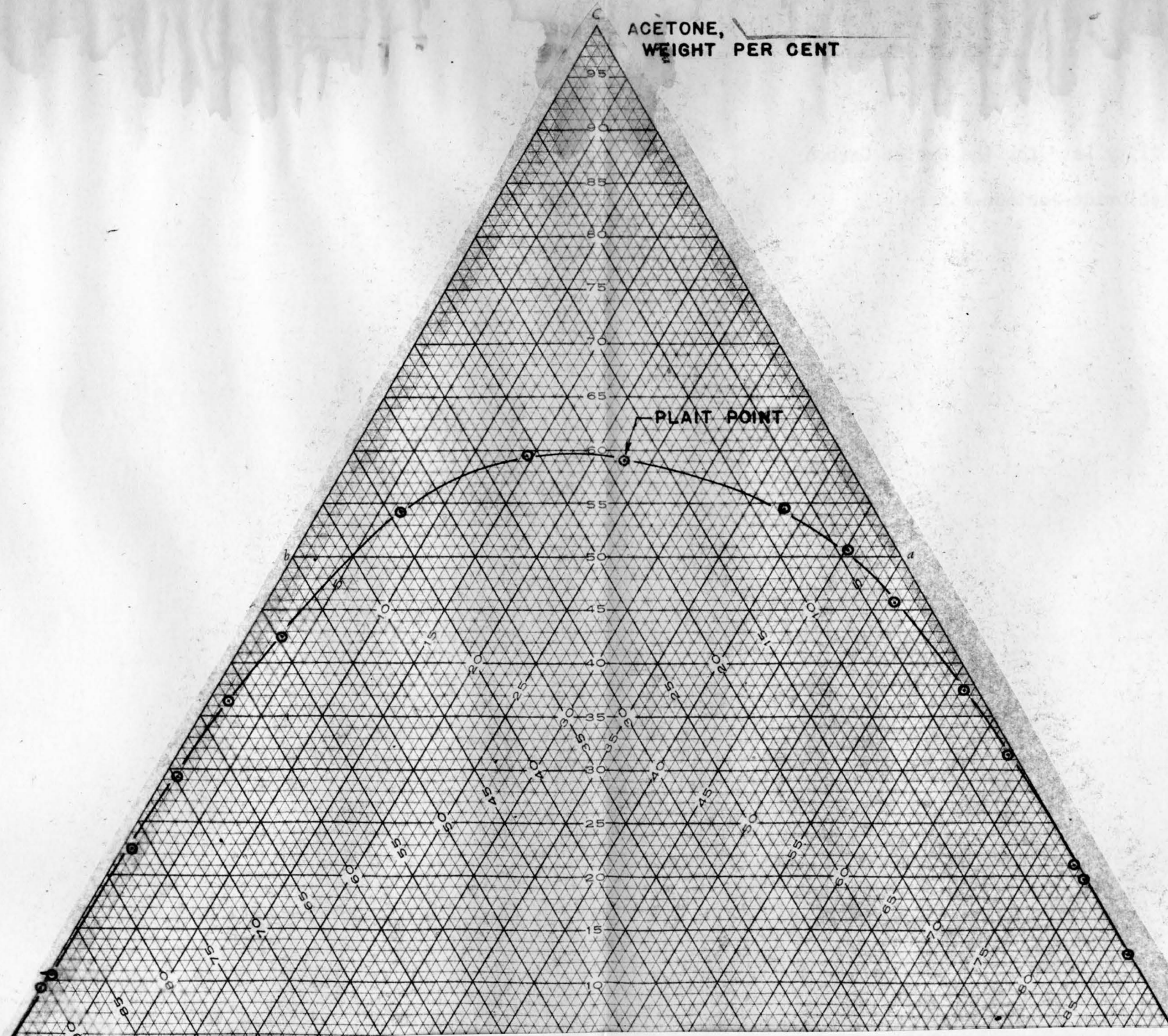


TABLE III

Solubility Data for the System
Carbon Tetrachloride-Acetone-Water^a

Acetone wt %	CCl ₄ wt %	Water wt %	Specific gravity 30°/4° C
0.00	100.00	0.00	1.5750
9.14	90.78	0.08	1.4410
10.58	89.19	0.23	1.4230
22.17	76.98	0.85	1.2852
29.32	69.61	1.07	1.2120
36.40	61.90	1.62	1.1470
24.63	54.67	2.70	1.0936
54.34	38.73	6.93	1.0000
59.46	26.15	14.39	0.9524
59.04	18.01	22.95	0.9330 ^b
54.83	7.57	37.60	0.9197
50.27	4.69	45.05	0.9325
45.77	2.73	51.50	0.9292
47.32	1.36	61.32	0.9411
31.58	0.69	67.73	0.9494
21.08	0.40	78.52	0.9657
19.82	0.39	79.78	0.9680
12.83	0.28	96.90	0.9780
3.22	0.22	96.56	0.9910
0.00	0.00	100.00	0.9956

- a. Buchman, R. H.: Liquid-liquid Equilibrium Data for the System Carbon-Tetrachloride-Acetone-Water, Ind. Eng. Chem., 44, 2449 (1952).
- b. Plait point.

as the percentage ratio of the approach to equilibrium for any stage(23).

In actual practice, where several stages are mounted together to form a tower, the total number of equilibrium stages to perform the actual separation is found. The number of real stages making up the tower divided into the number of ideal stages gives the average stage efficiency(23) for the column.

Equilibrium in Countercurrent Extraction. For continuous countercurrent extraction the ideal relation between the two countercurrent streams at any point is given by the following equation(23).

$$Y = \frac{\beta X}{1 + X(\beta - 1)}$$

where:

Y = solute in extract phase, weight fraction

X = solute in raffinate phase, weight fraction

β = selectivity coefficient.

Thus, a measure of efficiency, similar to stage efficiency, may be calculated by comparing the ideal Y from the above equation to the real Y at any given point. As will be pointed out later, this method, although quite valid and general, is seldom used.

Rate Relations

Once the equilibrium rates on performance are fixed, the rate relations make it possible to calculate the time required to approach equilibrium under actual conditions. In the more general case, if the time is fixed, the actual approach to equilibrium can, itself, be calculated.

Mass Transfer Concept. The two film theory⁽¹⁷⁾, although undoubtedly inadequate to completely describe extraction behavior, has been widely accepted as the closest available approximation. In this theory the interface between the two phases is believed to be formed of two compressed, laminar films of the type encountered in heat transfer. In this theory, it is assumed that all the resistance to transfer of the solute, from one phase to the other, takes place in the films. Experimental results show that there is a measurable resistance⁽⁶¹⁾ to the transfer of solute from phase to phase. In effect, this means that a finite time is required for a system to approach equilibrium. The reciprocal of the equilibrium time function is called the overall mass transfer coefficient. The overall mass transfer coefficient is a measure of the rate at which extraction takes place.

There have been many attempts to correlate the physical properties of the extraction system and the rate constant. Among these proposed correlations may be mentioned the work of Row,

Koffolt, and Withrow⁽⁵³⁾ and Skrzec⁽⁶¹⁾. However, as yet, no really satisfactory method of predicting the overall mass transfer coefficient for commercial equipment is available.

The Transfer Unit. The performance of stagewise equipment can be adequately described in terms of the stage efficiency. In order to obtain a measure of equal utility for use with continuous countercurrent extraction Colburn introduced the transfer unit. Colburn^(15, 16) defined the number of transfer units as equal to the integral of the differential change in concentration divided by the difference in concentration tending to cause transfer. Stated in symbols the number of transfer units may be defined as follows:

$$N = \int_{y_2}^{y_1} \frac{dy}{y^* - y}$$

where:

- N = the number of Colburn mass transfer units, dimensionless
- y = concentration of solute in one phase, mol fraction
- y* = concentration of solute in the same phase that is in equilibrium with the concentration of solute in the other phase at the same point, mol fraction
- y₁ = concentration of solute in the same phase at the feed end of the column, mol fraction
- y₂ = concentration of solute in the same phase at the solvent end of the column, mol fraction.

The transfer unit is a measure of standard difficulty of extraction. Just as one equilibrium contact is a measure of extraction independent of the system; so is one transfer unit.

In multistage columns the number of theoretical stages is divided by the actual stages to give the stage efficiency. This calculation cannot be carried out for continuous extractors as there are no actual stages present. To get around this difficulty Colburn (15, 16) stated that a unit length of the column could be defined as the continuous countercurrent equivalent of a theoretical stage. Thus, the total active height of the column divided by the number of transfer units is a measure of column efficiency. As this measure has the units of length it is commonly called the height of a transfer unit or more commonly H.T.U. The H.T.U. is different from efficiency in that a decrease in H.T.U. is equivalent to an increase in efficiency. In symbols the H.T.U. is defined as follows (9, 15, 16);

$$\text{H.T.U.} = \frac{H}{N}$$

where:

- H.T.U. = height of a transfer unit, feet
- H = height of the column, feet
- N = number of Colburn transfer units, dimensionless.

The H.T.U. is a direct measure of efficiency of extraction. If one column has a lower H.T.U. than the other it is a more efficient extractor. However, the H.T.U. can be used for comparison only for continuous countercurrent extractors; because, for a stagewise extractor the integral defining H.T.U. becomes discontinuous. The H.T.U. is related to the mass transfer coefficient and is used to calculate the value of the latter (9).

Height Equivalent to a Theoretical Stage. Instead of using the stage efficiency, stagewise columns may be compared by means of the height equivalent to a theoretical stage⁽²⁴⁾ or H.E.T.S. The H.E.T.S. is the total active height of a stagewise extractor divided by the number of theoretical stages. This number, the H.E.T.S., is a measure of extractor efficiency; but it is based on extractor height rather than on perfection of contact. The H.E.T.S. is analogous to the H.T.U. but is used for stagewise extractors where the H.T.U. is not applicable.

Relation Between H.E.T.S. and H.T.U. Many commercial extractors, for which performance data are available, are rated as having a certain H.E.T.S. Other commercial extractors are rated as having a certain H.T.U. In order to compare these extractors it is necessary that some idea be obtained of the relationship between H.E.T.S. and H.T.U.

There is, in general, no simple, direct relation between the H.T.U. and the H.E.T.S.⁽²³⁾. If equilibrium data for the system used is available, calculations can be made and the exact equivalence found. However, without making such calculation, no comparisons should be made between equipment rated by H.T.U.'s and equipment rated by H.E.T.S.'s.

Elgin⁽²³⁾ reports that for the special case where both the operating line and the equilibrium line are straight and parallel the H.E.T.S. and H.T.U. are equal. However, a calculation is still required to determine the operating line.

Along similar lines is the growing tendency of reporting performance of all types of extractors in H.T.U.'s. In the author's opinion this practice is to be discouraged. The calculation of H.T.U.'s for a stagewise extractor represents a mathematical fallacy. The calculation of H.E.T.S.'s for a continuous column is to describe the action of the column in erroneous terms. H.E.T.S. and H.T.U. when calculated as described in this paragraph have no real meaning as far as equipment performance is concerned.

The errors introduced by the indiscriminate use of H.T.U.'s and H.E.T.S.'s may be illustrated by an example from the author's own experience. A two-inch diameter laboratory spray tower, four feet long, used for student experimental studies gave a H.T.U. (32) from 12 to 30 feet. Using the same conditions and the same system, but with the data calculated as if the column were stagewise a H.E.T.S. (64) from 0.75 to 1.50 feet was obtained. The extractor was obviously the same and performing equal separation in both cases. For this particular system and extractor one transfer unit represents from 16 to 20 times as much extraction as one theoretical stage. If this fact is known, it makes little difference, from a practical standpoint, how the column is rated. However, if this fact were not known, and it seldom is, by rating the column in H.E.T.S.'s it would make the column appear to be a much more efficient extracting device than it really is.

Methods of Calculation. Algebraic methods for calculating the number of theoretical stages have been reported by Varteressian and Fenske^(76, 77) and Hunter and Nash^(33, 34). These methods are extremely difficult to apply except in rather special cases. Graphical simplifications of the above methods have been proposed by Maloney and Schubert⁽⁴⁰⁾, Hunter and Nash^(33, 34), and Varteressian and Fenske^(76, 77).

Maloney and Schubert⁽⁴⁰⁾ have modified the method of Ponchon and Saveritt to apply to extraction. This method makes use of the solvent content-concentration diagram for a ternary system. It is covered in detail by Brown⁽⁸⁾, Perry⁽²³⁾, and Bull and Coli⁽¹¹⁾.

The triangular phase diagram, an example of which is shown on page 21, supplies the necessary relation between the composition and quantity of the extract layer leaving any stage and those of the raffinate layer leaving the same stage. Simultaneous graphical solutions of the material balances on this chart furnish a graphical solution to a countercurrent stagewise extraction operation. This method was proposed by Hunter and Nash^(33, 34) and is covered in detail by Perry⁽²³⁾ and Bull and Coli⁽¹¹⁾.

The McCabe-Thiele method may be modified to present a graphical solution to extraction problems. This method was first proposed by Varteressian and Fenske^(76, 77) and covered by them and Perry⁽²³⁾.

Equipment

In this section some of the design features of liquid-liquid extractors will be discussed. After a general review of commercial equipment, a rather thorough study of some new power extractors will be made.

Liquid-liquid Extractors. A liquid-liquid extractor may be defined⁽²⁴⁾ as any type of equipment that may be used to effect liquid-liquid extraction. Liquid-liquid extractors, like most unit operations equipment, are exemplified by a great variety of commercially used types. This condition was caused by several factors⁽⁴⁴⁾: Lack of reliable performance data on available equipment, development by different industries before the emphasis on unit operations began, attempts to modify existing distillation and absorption apparatus to extraction, the need for rather specialized extractors for several purposes, research designed to make existing equipment fit rather than design new equipment, and many others.

Classifications of Liquid Extractors. Three methods of classification of extractors are those presented by Elgin⁽²⁴⁾, Treybal⁽⁷²⁾, and Morello and Poffenberger⁽⁴⁴⁾.

Elgin⁽²⁴⁾ classifies extraction equipment according to either mixer-settler (batch) or "continuous" extractors. He subclassifies further as follows:

- I. Mixer-settler extractor
 - A. Air agitators
 - B. Mechanical mixers
 - C. Flow mixers
 - D. Column mixers
 - E. Pumps

- II. Countercurrently columns operating "continuously".
 - A. Wetted wall
 - B. Spray
 - C. Packed
 - D. Sieve or perforated plate
 - E. Modified bubble plate
 - F. Baffle column
 - G. Columns with internal agitators

This classification has little to recommend it, except that it is used in Perry's Chemical Engineers' Handbook.

Treybal⁽⁷²⁾ borrowed Elgin's classification and modified it to further subdivide the "continuous" columns. However the distinction between "continuous" and continuous does not appear to have been used by him.

Morello and Poffenberger⁽⁴⁴⁾ used an entirely different scheme of classification. They divided equipment into classes depending upon the method used to separate the phases. Their further subdivisions are as follows:

I. Two phases separated by gravity

A. Contact made through the interfacial area as a film

1. Extended surfaces
2. Packed towers

B. Contact made through the interfacial area with one phase dispersed as droplets

1. No additional power required for dispersion
 - a. Spray towers
 - b. Baffle towers
 - c. Perforated plate towers
 - d. Bubble-cap towers
 - e. Packed towers
2. Additional power required to redisperse the phases
 - a. Mixer and settler combinations
 - b. Baffle tower with movable membranes

II. Two phases separated by centrifugal force.

The Use of External Power in Extraction Equipment. One of the most important questions to be solved in liquid-liquid extractor design is the utility of extractors using external power⁽⁷²⁾.

External power⁽⁴⁴⁾ may be defined as any power supplied to the extractor in addition to that power necessary to cause the flow of the liquid phases.

The additional power supplied—if efficiently utilized—will cause the formation of a very high interfacial area and, consequently, achieve better extraction⁽⁵⁶⁾. However, this gain in extraction rate is set against the additional mechanical complexity of the extractor and the additional cost of operation. Some of the mechanical features of power extractors that often cause trouble are leaks in stuffing boxes, undesirable flexing and strains in long shafts, and increased erosion and corrosion of moving parts⁽⁴⁴⁾.

Centrifugal Extractors. The only successful extractors of this type are modifications of the extremely versatile Podbialinak⁽⁵¹⁾ centrifugal contactor. This device consists of a passage, rectangular in cross section, wound upon a shaft to form the rotor. This spiral rotor is contained in a casing. When assembled the entire extractor looks somewhat like a centrifugal pump. The liquids to be contacted flow into and out of the extractor through the shaft. A successful modification consists of perforating the spiral passage⁽⁷³⁾.

These extractors must be carefully designed and constructed to withstand high mechanical stresses and to avoid injurious vibrations⁽⁵¹⁾. The first cost and the operating cost are quite high. However, these extractors have the following advantages:

A wide range of feed to solvent ratios are possible, small compact units, low liquid holdup (nine gallons in a commercial 2400 gallons-per-hour unit), and systems that tend to emulsify are easily handled.

Extractors of this type are widely used in the pharmaceutical industry (penicillin, streptomycin, etc.)⁽⁶⁶⁾ where the product can easily absorb the higher first and operating costs. In June, 1950, there were approximately 400 of these units in commercial operation⁽⁴⁴⁾. The capacity of these units varies from 500 cubic centimeters per minute to 2400 gallons per hour⁽⁷³⁾. These units contain from four to seven equilibrium stages⁽⁵⁹⁾.

Film Type Extractors. There are three types of extractors representing this type of equipment. The first two, the wetted-wall and the horizontal-tube extractor, are examples of laboratory experimental columns. The last, the film-type extractor of Gordon and Zeigler⁽²⁹⁾, is an example of a commercial type extractor.

The wetted-wall column^(18, 74) is used for fundamental studies where it is necessary to be able to measure the interfacial area accurately. These extractors provide for one phase to flow either up or down the inside wall of a circular column. The other phase flows countercurrently through the inside of the column as a central core. Only a limited range of flow rates for either phase is possible.

The column of Skrzec⁽⁶¹⁾ consists of a horizontal tube with the interface lying along the center of the column. The heavy phase flows along the bottom and the light phase flows counter-currently along the top of the tube. This column also allows an accurate interfacial area to be measured. A much larger range in flow rates is possible with this extractor.

The film extractor of Gordon and Ziegler⁽²⁹⁾ consists of passages, rectangular in cross section, inclined at an angle to induce layer formation within each duct. This design allows liquids to be treated that are prone to emulsify if dispersed. A typical example of a commercial model has the following dimensions: Ducts one-quarter-inch deep, one-inch wide, 15-feet long, and inclined at an angle of 45° . This unit was very efficient as it contained nearly 20 theoretical stages.

Spray Tower Extractor. The spray tower extractor⁽⁷³⁾ is the simplest and cheapest continuous countercurrent extractor. It consists of a container, usually cylindrical, through which the liquids to be treated pass countercurrently to each other. One of the liquids, called the continuous phase, normally fills the tower and the other liquid, called the dispersed phase, is sprayed through the first liquid.

Although widely used in laboratory studies because of its simplicity, the spray tower is little used industrially⁽⁴⁴⁾. This is, at least partly, caused by its two greatest defects.

These defects are, first, the coalescence of the dispersed phase, and, second, the recirculation of the continuous phase. Attempts have been made to eliminate or alleviate these defects by improving spray nozzle design and by changing the basic cylindrical shape to obtain better continuous-phase flow patterns. Work of this type is beyond the scope and interest of this review. The reader is referred to Blanding and Elgin⁽⁴⁾ for a particularly interesting discussion on spray tower design.

Packed Extraction Columns. The successful use of packed towers in gas absorption and, to a lesser extent, in distillation suggested their use in liquid-liquid extraction. In general, the packed tower⁽⁷³⁾ may be thought of as a spray tower in which packing has been placed to modify the unhindered motion of the liquids. In a packed column additional turbulence is certainly introduced, but the overall efficiency may be poor⁽⁵⁾. This is probably caused by channeling and even greater recirculation of the continuous phase than present in a spray tower.

After the spray tower, the packed tower is probably the cheapest and easiest tower to construct. This is especially true when the needed extractor must be made of ceramic or similar material⁽⁴⁴⁾. Besides the cylindrical shell, only a simple packing support and packing of the type used in gas absorption are required. However, in spite of the wide use of this type of tower, very little work has been done on the optimum design of packing, packing supports, and nozzles for liquid-liquid extraction.

In a packed tower the relative wetting of the packing by the two liquids must be considered^(43, 70). If the dispersed phase wets the packing preferentially, the flow pattern will resemble that obtained during gas absorption with one liquid flowing over the packing in a continuous atmosphere of the other liquid. However, if the continuous phase wets the packing preferentially, the dispersed phase will flow through the tower with a flow pattern similar to the flow pattern in a spray tower. In this case, the packing serves no purpose except to redispense and agitate the dispersed phase.

A few examples of commercial packed extractors that might be mentioned are as follows. A tower used in the refining of vegetable oils is packed with raschig rings to a depth of 75 feet. This tower is 1.8 feet in diameter and 87 feet high^(35, 27, 70). A packed tower of 13 feet in diameter and 47 feet high is used in the refining of lubricating oils by furfural⁽⁴⁴⁾. A packed tower 6.7 feet in diameter and 69 feet high is used in the recovery of phenol. This tower is packed to a depth of 53 feet^(70, 44).

Baffle Extractor. The baffle extractor⁽⁷³⁾ is one of the extraction apparatus in commercial use that was developed solely for extraction. In many respects the baffle column is a satisfactory extractor. The towers are simple to construct, cheap, have low pressure drops, no small holes or orifices to plug, and operate with very little maintenance.

Elgin⁽²⁴⁾ defines the baffle extractors as "vertical towers containing horizontal baffles to direct the flow of liquids. There are three principal types: Disk and doughnut, side to side, and center to side." This is not strictly true as various modifications have been proposed and some entirely new types developed. Notable among the recent improvements in baffle tower design is the Coshran⁽¹³⁾ modified tower.

Some commercial baffle extractors that may be mentioned have the following characteristics. A disk and doughnut baffle extractor 3.5 feet in diameter and 45 feet high is used in the acetic acid recovery from wood liquors⁽²⁸⁾. This tower has 43 disk and doughnut units. There are about four ideal stages in this tower. A side to side baffle tower 6.5 feet in diameter and 48 feet high is used for the extraction of hydrocarbon oils⁽⁴⁴⁾. This tower contains 103 baffles spaced five inches apart. It takes approximately eight baffles to make one ideal stage.

Perforated-Plate Extractor. The perforated-plate extractor⁽⁷⁹⁾ operates by having the dispersed phase repeatedly coalesced and dispersed. This operation is maintained by forcing the liquids to flow through a series of trays in which a large number of small holes have been punched or drilled. This type of tower usually has means (such as downcomers, weirs, and spouts) to control the flow of the continuous phase. The dispersed phase may be, but is usually not, controlled.

The perforated-plate extractor appears to be a fruitful field for experimental work. This is witnessed by the great number of improved designs that have been patented recently. Notable among these are the vertical step plates⁽³¹⁾, the trough type plates⁽⁶³⁾, and the Koch baffle⁽³⁹⁾.

The perforated-plate extractor is used extensively in the phenol-lubricating oil refining process⁽¹⁾. Extractors used for this purpose vary in size from eight to 12 feet in diameter and from 31 to 67 feet high with from 14 to 20 plates. A plate efficiency of 3.6 to 8.6 per cent is considered average⁽⁶⁰⁾.

Bubble-Cap Extractors. Probably in no other place is the fallacy of using unmodified gas absorption or distillation apparatus more noticeable than in the bubble-cap extractor⁽⁵²⁾. The bubble-cap tray contactor in distillation and gas absorption is generally considered to be the most efficient type of contactor. However, when this contactor is used in extraction very poor results are obtained. This low efficiency is caused by the large decrease in both the density and the interfacial tension differences and by the large increase in viscosity of the dispersed phase. These changes cause a different flow pattern to exist in liquid-liquid extractions compared to distillation columns. However, both Treybal⁽⁷³⁾ and Morello and Poffenberger⁽⁴⁴⁾ believe that a bubble-cap column, designed exclusively for liquid-liquid extraction, should be quite an efficient contactor. Morello and Poffenberger⁽⁴⁴⁾ point out

that in extraction the plate spacing could be reduced to a very small fraction of that required in distillation and still allow the plate to operate.

Mixer and Settler Extractors. Mixer and settler type extractors⁽⁷²⁾ consist of a mixing chamber of suitable design followed by a settling chamber for each stage. Mixer-settler extractors were undoubtedly the earliest type of extraction equipment used. The organic chemist, with his separatory funnel, is performing extraction by mixing and settling. Exact scale up of the chemist's technique is probably the biggest reason for the continued usage of mixer and settler extractors.

The mixer-settler type of extractor, although quite dependable, is extremely bulky⁽⁷²⁾. The units that make up the extractor are commonly placed beside each other and a great amount of floor space is required.

To compromise between the high efficiency and the inherent dependability of the mixer-settler extractor and the small size of the continuous extraction towers many combinations of the two ideas have been proposed. Klarsted⁽³⁶⁾ suggested mounting a series of settling chambers one above the other. The liquids are removed, mixed, and returned to the appropriate settling chamber.

McConnell⁽⁴²⁾ and Jantzen⁽¹⁹⁾ developed independently the idea of mounting the mixing sections in the tower between the settling sections. The mixing is accomplished by a turbine mixer either

on a central shaft or inserted into each mixing section. Othmer⁽⁴⁸⁾ added additional baffling and introduced the counter-current flow of the liquids in the settling zone. Oldshue and Rushton⁽⁴⁷⁾ removed a great deal of baffling and obtained a truly countercurrent mixer-settler tower. Scheibel⁽⁵⁶⁾ added fine wire mesh packing to the settling sections to increase the rate of settling. It is reported that pilot plant units of the Scheibel column up to 12 inches in diameter are being operated successfully⁽⁵⁴⁾.

The Van Dijk Column. In 1934, Van Dijk⁽⁷⁵⁾ proposed an extractor that consisted of a vertical tower equipped with movable perforated baffles. These baffles are raised and lowered through the main body of the fluid to produce agitation.

The operation of this column may be described as follows⁽⁷⁵⁾. The two liquid phases are allowed to flow countercurrently through a tower equipped with movable perforated baffles. In a compartment bounded by two baffles both phases are present. In each of these compartments the heavier phase settles to the bottom and the light phase on top. This means that on each plate there is a layer of the heavy phase. Similarly, there is a layer of the light phase under each plate. The flow resistance of the plates is sufficient to prevent any appreciable flow through the plates because of the density difference of the liquid. When the plates begin to move upward through the stationary liquid body the

heavy phase on the top of the plates is forced through the openings in each plate in the form of jets. These jets are, of course, changed into drops or a fine spray in the lighter phase underneath each plate. The velocity of the jets is proportional to the plate movement; as the area of the holes is proportional to the total area. By adjusting, thus, the hole size, plate movement, and plate free area, any desired degree of dispersion of one phase into the other may be obtained. After the movement of the plates there is a brief pause during which settling takes place. After settling the process is reversed and the light phase is dispersed into the heavy phase by a downward movement of the plates. Instead of perforated plates wire mesh packing may be used. No idea of approximate dimensions, experimental conditions, or results are given in the patent.

The Feick Column. Feick⁽²⁵⁾ modified the idea of Van Dijk by replacing the plates with continuous packing and by moving the liquids relative to this packing. The operation of the Feick column is quite different from the operation of the Van Dijk column. The pulsations simply agitate the liquids to a greater extent than the agitation caused by their normal countercurrent flow in a packed extractor. This additional agitation results in a much greater degree of subdivision of the dispersed phase than the energy available in the density difference of the countercurrent liquids ever could produce.

This column was two inches in diameter and packed with one-half inch McMahon saddles or three-eighths inch raschig rings. The agitation was varied from a liquid travel in the unpacked column of one-sixteenth inch to one-fourth inch. The number of strokes was varied from 200 to 1,000 per minute.

Pulse Column. Van Dijck⁽⁷⁵⁾ recognized some of the practical difficulties in the design he proposed. The bulky mechanism for obtaining motion of the plates, bad enough in itself, was located inside the column. Here the mechanism would be subjected to corrosion and would be extremely inaccessible for maintenance work. Thus, Van Dijck was led to propose an alternate design. This alternate design was to make the plates stationary and move the liquids past the plates by means of a reciprocating piston. This type of extractor has been called a pulse column⁽⁴¹⁾.

Similar columns have been investigated by the Oak Ridge National Laboratories⁽⁴¹⁾, and the Departments of Chemical Engineering at North Carolina State⁽⁵⁰⁾, Oregon State⁽⁶⁵⁾, Iowa State⁽¹⁴⁾, and Georgia Institute of Technology⁽⁷⁹⁾.

The extractor at Iowa State University⁽¹⁴⁾ consists of a pyrex column one-inch in diameter and 20-inches high. This column contains ten perforated plates. The perforations are 0.040 inch in diameter and total nine per cent of the area of the plate. The pulsating motion was imparted to the liquids by a diaphragm pump with the check valves removed. Stroke frequencies

in the range of 35 to 72 cycles per minute were found to give the best operation.

The column at North Carolina State University⁽⁵⁰⁾ consists of a precision bore glass tube 1.5 inches in diameter and eight inches long. This column contains four plates with a two inch plate spacing. The plates were drilled with 0.0590 inch holes and the area of these holes totals 20 per cent of the plate area. The agitation consisted of 24.0 strokes per minute each stroke consisting of 1.20 inches of liquid travel in the empty column height. This agitation was furnished by a constant volume feed pump with the checks modified to permit pulsing.

Rate Relations and Flow Capacities

In this section the rate relations and the flow capacities important to an evaluation of the pulse column will be discussed.

Flow Capacities. The capacity of an extractor is limited by the flooding velocity. The flooding velocity is defined⁽⁷³⁾ as the velocity of flow of one phase at which the other phase is just kept from flowing through the extractor. The limit on capacity caused by this phenomena is an important design consideration.

Van Dijk⁽⁷⁵⁾ states that the flooding velocity for a pulse column is equal to the flooding velocity of a spray tower of equal column diameter. The flooding velocity, also appears to be

independent of the free area of the plates. The Iowa State⁽¹⁴⁾ pulse column flooded at a flow of about 660 gallons per hour-square foot. This is equivalent to a flooding mass velocity of 5500 pounds per hour-square foot.

Elgin and Foust⁽²¹⁾ present a chart correlating flooding data in spray towers. From this chart values in the range of 10,000 to 20,000 pounds per hour-square foot or even higher are obtained for flooding, depending upon the system and the relative velocities.

Rate Relations for Pulse Columns. The pulse column, as a countercurrent stagewise extractor, should be rated in terms of either the H.E.T.S. or the stage efficiency. However, the two columns, for which data is available, are calculated as if they were continuous countercurrent columns; that is, they are rated in H.T.U.'s. For the reasons discussed previously, these results should be viewed with caution, especially when comparing to pulse columns rated in terms of the H.E.T.S.

For the column at Iowa State⁽¹⁴⁾ values of the H.T.U. as low as 5.7 inches were reported. As no additional data were given the equivalence of the H.E.T.S. and H.T.U. could not be calculated.

For the column at North Carolina State⁽⁵⁰⁾ values of the H.T.U. as low as 0.14 foot were reported. The complete experimental results and data for this column are presented in Table IV, page 46. The results for this column were obtained using the two component system of benzene-water. As a theoretical stage for a two

TABLE IV

Plate Efficiencies for a 1-1/2-inch Pulse Extractor Operating
With a Pulse Frequency of 24 Per Minute and an Amplitude
of 1.20 Inches^a

Continuous phase	Flow rates		Plate efficiency ^b per cent
	Benzene ml/min	Water ml/min	
Benzene	107	353	17.2
Benzene	70	107	15.7
Water	73.5	235	20.2
Water	76	109	18.7
Water	77.5	106	18.7
Water	153	600	22.8
Water	43.6	598	24.2

a. Pike, F. P., D. S. Arnold, and C. A. Flank: The Performance of Contactors for Liquid-liquid Extraction. Unpublished Progress Report, Department of Engineering Research, North Carolina State College, Raleigh, N. C., July 1, 1952.

b. Calculated by the author from data obtained from the report.

component system results in the saturation of the respective phases, and as the phases did not leave saturated, it can be stated that the column did not have one complete theoretical stage. However, as the composition data of the respective phases are also available H.E.T.S.'s for this column may be calculated.

Rate Relations for the Feick Column. The Feick⁽²⁵⁾ column is a continuous countercurrent packed extraction column with additional agitation applied to the extractor by pulsing the liquids relative to the packing. Its performance should be reported in terms of the H.T.U.

Although the results from the Feick column will have little bearing on the problem at hand, they will be used to show the large possible increase in extraction efficiency obtained by applying extra agitation. The Feick column can be operated as a regular packed extractor for comparison purposes.

The H.T.U. was decreased from the range of 7.5 to 13 feet to the range of 1.0 to 0.5 feet when agitation was applied. These results were obtained with the system toluene-benzoic acid-water. For the system toluene-acetic acid-water similar results were obtained. H.T.U. values of 1.09 to 1.14 feet, without agitation, were reduced to values of about 0.484 to 0.185 feet with agitation.

Rate Relations for the Scheibel Column. Performance data for three different installations of the Scheibel column are available in the literature.

The first column⁽⁵⁵⁾ is 1.0 inch in diameter and consists of 13 stages, each stage being made up of a one-half inch mixing section and a two inch packed settling section. The minimum H.E.T.S. for this column was 2.2 inches which gives a plate efficiency of greater than 100 per cent. This was for a flow rate of 1340 pounds per hour per square foot of combined phases. The system water-methyl-ethyl-ketone-acetic acid gave a minimum of H.E.T.S. of 4.2 inches for the same range of variables.

In his patent application Scheibel⁽⁵⁶⁾ claims to have operated a column three inches in diameter that had a stage efficiency greater than 100 per cent. This column consisted of 11 stages; each stage being composed of a four-inch mixing section and a four-inch packed settling zone. No further details are available.

A Scheibel-type column⁽⁵⁷⁾ 12 inches in diameter has also been reported. This column has three stages, each stage consisting of a mixing section three inches high and a settling zone nine inches high. The minimum H.E.T.S. reported for this column was 9.0 inches at a combined phase flow rate of 4100 pounds per hour per square foot. Flooding occurred at a combined rate of 600 gallons per hour per square foot maximum and 350 gallons per hour per square foot minimum.

Rate Relations for the Multistage Mixer Column. The multistage-mixer column is a countercurrent, stagewise, vertical extraction column compartmented by horizontal baffles, agitated by mixing

impellers on a vertical shaft, and baffled by vertical members. Unlike the Scheibel column there is no attempt to induce coalescence between stages. For a column⁽⁴⁷⁾ six inches in diameter consisting of eight stages each stage three inches high a minimum H.E.T.S. of 3.7 inches was found. This was for a combined flow rate of 2140 pounds per hour per square foot.

18

III. EXPERIMENTAL

The experimental portion of this investigation consists of the purpose of investigation, plan of experimentation, materials, apparatus, method of procedure, data and results, and sample calculations.

Purpose of Investigation

The purpose of this investigation was to design, construct and operate a liquid-liquid pulse extractor, and to determine the effect of the flow rates and frequency and amplitude of the pulses on extraction efficiency. The limiting liquid flow rates for the extractor were also determined.

Plan of Experimentation

The plan of experimentation followed in the investigation consisted of a review of the literature on liquid-liquid extraction, design and construction of the pulse extractor, determination of refractive index data for the system used, operation of the column, determination of limiting liquid flow rates, and correlation of data.

Literature Review. The literature review was designed to acquaint the author with the principles of extraction and, in particular, extraction as applied to the pulse column. To accomplish this purpose a study of the general principles of extraction and some important definitions was made. Next, a study of the equilibrium relations important in extraction was performed. The equilibrium data for the system carbon tetrachloride-acetone-water which was used in this investigation were, of course, covered in detail. The rate relations by which equipment can be compared were then studied. Next, a brief review of equipment types, with several industrial examples given, was made. Several improved extractor designs, among them the pulse extractor, were covered in greater detail. Last, a study of the rate relations and flow capacities for the pulse extractor and some other recent designs of extractors was made.

Design and Construction. For purposes of design and construction the equipment used in this study was divided into five major groupings; namely, the pulse extractor, the pulse-extractor and push-rod frame, the push-rod assembly, the can assembly, and storage and tubing.

It was desired to construct a pulse-type extractor from a section of pyrex pipe two inches in diameter. The extractor was to be equipped with eight baffle plates; each plate spaced at four-inch intervals. The plates were drilled with 116 holes $3/32$

inches in diameter. The pulsating motion of the liquids was caused by a movable bellows driven by a push rod, operated by an eccentric cam. For the satisfactory operation of the extractor various auxiliary apparatus were required. Important among these were the column frame, the push-rod frame, the push-rod bearing, the reduction train for the cam, the storage carboys, and the tubing and valves. The column and all auxiliary apparatus were assembled at the extractor site from parts either made or purchased for this investigation.

Refractive Index Data Determination. The demonstration system of carbon tetrachloride-acetone-water was selected for this investigation. The only available data on this system made use of the specific gravity of a sample as the method of analysis. For convenience in analyzing for acetone the refractive index data for this system were desired. These data were obtained by preparing samples of carbon tetrachloride, acetone, and water, analyzing by measuring the specific gravity, and determining the refractive index at 20° C. In addition, distribution data were obtained at the same time.

Experimental Tests. To gather data on the efficiency of the pulse extractor, fifteen tests were made, seven at varying pulses and eight to study the effect of flow rates. For the first series of tests, a heavy-phase flow rate of approximately 1500 pounds per hour-square foot and a light-phase flow rate of

approximately 500 pounds per hour-square foot were used. The pulse amplitude was varied from one to three inches, and the pulse frequency was varied from 10.5 to 21.0 strokes per minute. In the second series of tests the effect on the stage efficiency was determined when the heavy-phase flow rate varied from 400 to 5000 pounds per hour-square foot and the light-phase flow rate varied from 1000 to 8000 pounds per hour-square foot. For these tests a feed mixture of ten weight per cent acetone in carbon tetrachloride was prepared. The column was started, and the frequency and amplitude of the pulse was adjusted. The desired flow rates were then set. When "steady-state conditions" had been reached the following data were taken: Amplitude of pulse, frequency of pulse, extract flow rate, raffinate flow rate, raffinate refractive index, extract refractive index, and feed refractive index.

Flooding Determinations. Three conditions were recognized as indicating unsatisfactory column operation. These conditions were defined as follows: Flooding, or the failure of one phase to flow through the column as fast as it was being introduced; dumping, or failure of the heavy phase to cease flowing during the pause between pulses; and blowing, or failure of the heavy phase to form a layer on the plates during the upward portion of the pulse. Visual observations were made on these conditions, and on

the flow rates in which satisfactory plate operation took place. Tests were made at two different frequencies and at two different amplitudes.

Correlation of Data. The correlation of stage efficiency with the variables of heavy-phase flow rate, light-phase flow rate, concentration of acetone in the feed, and amount of pulsation was accomplished by means of a statistical analysis of the data. Specifically, the variables were related by means of a multiple regression equation. The statistical significance of the regression coefficients was also determined.

Materials

The specifications of the materials used in this investigation are presented in the following sections.

Acetone. H. F., Baker and Adamson lot No H 297J. Obtained from the General Chemical Division, Allied Chemical and Dye Corporation, New York, New York. Used as the solute in the demonstration system.

Air. Compressed air at the extractor site furnished from lines in the Unit Operations Laboratory, Department of Chemical Engineering, Virginia Polytechnic Institute, Blacksburg, Virginia. Pressure varied from 50 to 75 pounds per square inch gauge, at compressor site. Used as the motive force for the heavy phase.

Carbon Tetrachloride. Technical grade, Baker and Adamson lot No B 052. Obtained from the General Chemical Division, Allied Chemical and Dye Corporation, New York, New York. Used as the carrier solvent in the demonstration system.

Water. Tap water at extractor site obtained from lines located in the Unit Operations Laboratory, Department of Chemical Engineering, Virginia Polytechnic Institute, Blacksburg, Virginia. Used as the solvent in the demonstration system.

Apparatus

The apparatus used in this investigation are described in the sections that follow.

Balance. Solution type beam balance, capacity 20 kilograms, sensitivity 1 gram. Obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used to weigh the extract phase and the raffinate phase from the pulse extractor.

Constant-temperature Bath. Fisher Unitized Constant-temperature bath, guaranteed to maintain set temperature at $\pm 0.025^{\circ}$ C. Obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used for a constant-temperature bath during specific gravity determinations.

Constant-temperature Bath. Constant-temperature bath constructed by the Department of Chemical Engineering, Virginia Polytechnic Institute, Blacksburg, Virginia. Controlled by an Aminco Thermal Switch, manufactured by the American Instrument Company, Silver Spring, Maryland. Used as a source of constant-temperature water for the refractometer.

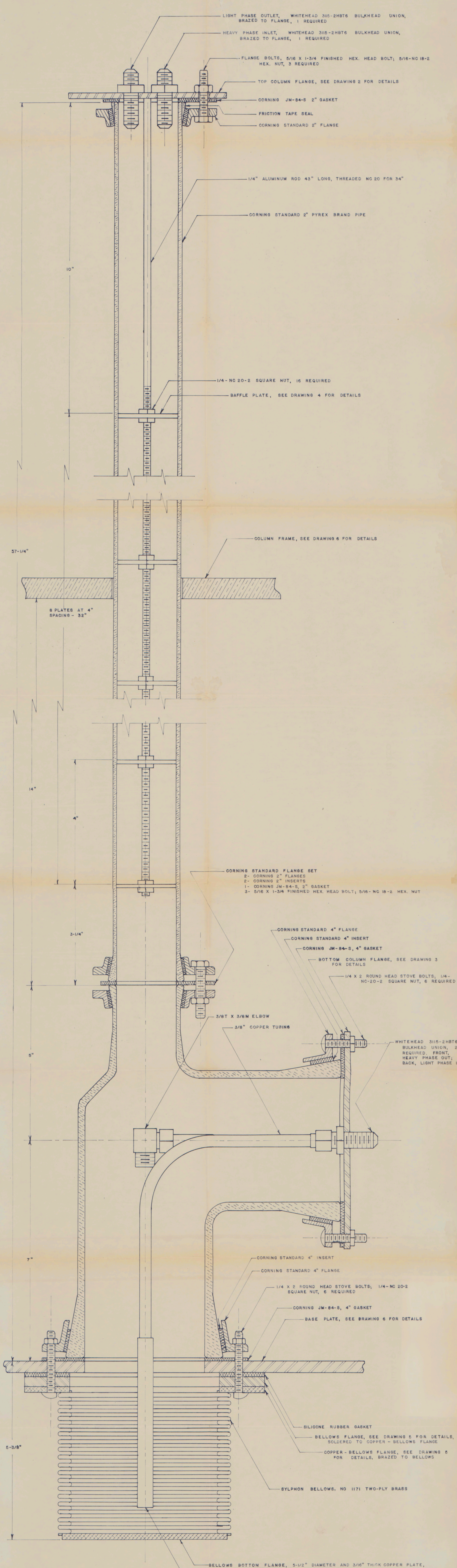
Miscellaneous Glassware. Laboratory glassware obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used for standard analytical and laboratory purposes.

Mixer. "Lightin" model L mixer, serial No 502260, 110 volts, 50-60 cycles, single phase. Manufactured by the Mixing Equipment Company. Obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used to mix the feed for the extractor.

Pails. Ten-quart galvanized pails. Obtained from the Nelson Hardware Company, Roanoke, Virginia. Used to collect the extract phase and the raffinate phase from the extractor, except during the experimental tests.

Pulse Extractor. The pulse extractor was constructed in the Department of Chemical Engineering from purchased and manufactured parts. A detailed drawing of this column is found on page 58. The bill of materials for this column is contained in Table V, page 59. To aid in understanding Drawing 1, details of the column top plate are found in Drawing 2, page 60, details of the column bottom plate are found in Drawing 3, page 61, details of the perforated baffle plates are found in Drawing 4, page 62, and details of the top bellows flanges are found in Drawing 5, page 63. The parts shown in these last four drawings are also contained in Table V, page 59.

Pulse-Extractor and Push-Rod Frame. The pulse-extractor and push-rod frame was constructed in the Department of Chemical Engineering for this investigation. Details of this frame may be seen in Drawing 6, page 64. The bill of materials for this frame is contained in Table V, page 59.



NOTE: PUSH-ROD RAM (SEE FIGURE 8 FOR DETAILS) CENTERED AXIALLY ON COLUMN. DIMENSION OF BELLOWS SHOWN FOR RAM IN FULLY RETRACTED POSITION.

REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA		
	PULSE EXTRACTOR		
FULL SCALE	DATE 4/14	CASE 83	
DRAWN BY: DAW	4/14	FILE 899	
CHECKED BY: PHW	4/15	DRAWING 1	
APPROVED BY: PHW			

Drawing 1. Detail Drawing of Pulse Extractor



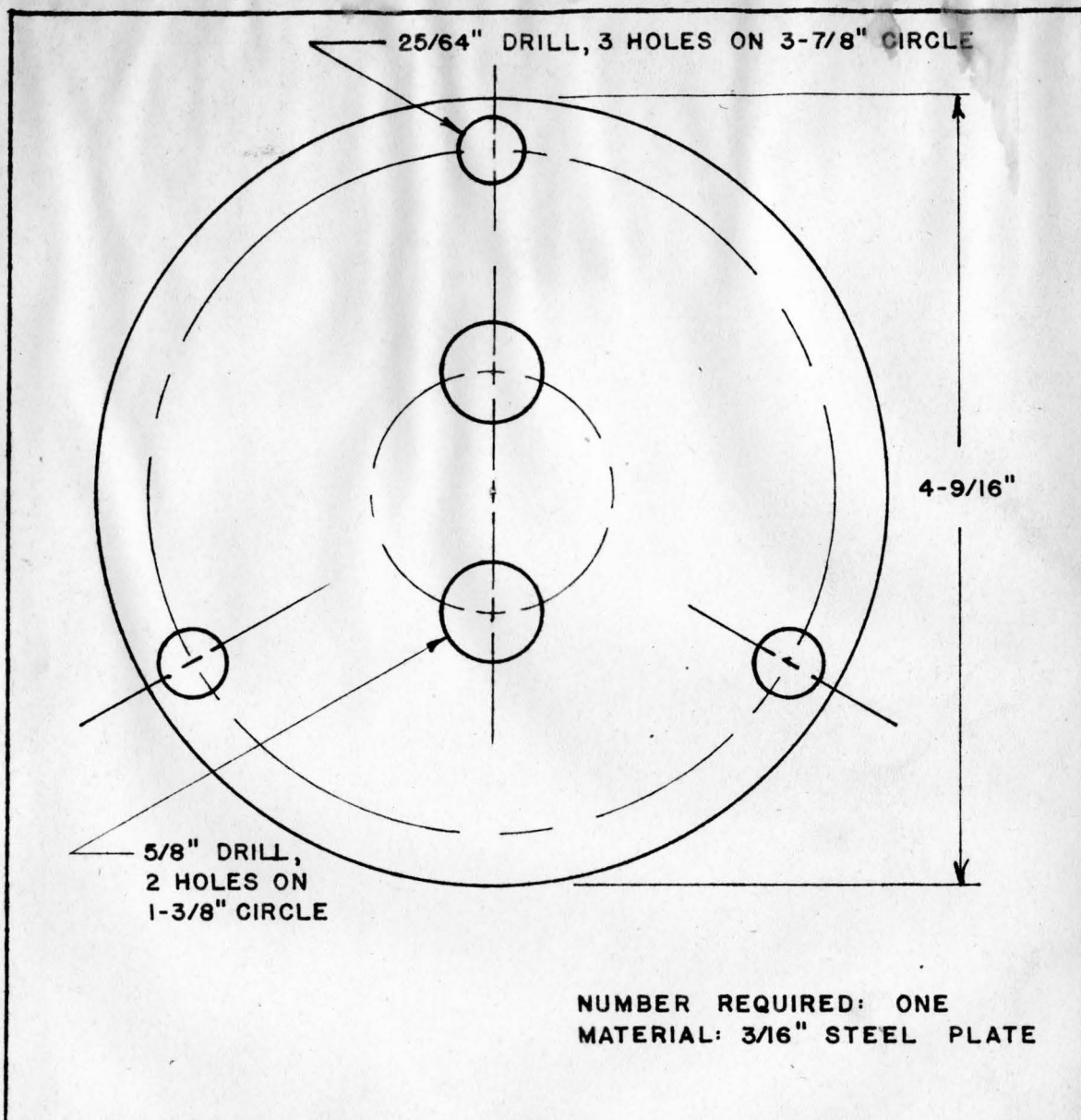
TABLE V

Bill of Materials for an Experimental Two-inch
Pulse Extractor and Auxiliary Apparatus

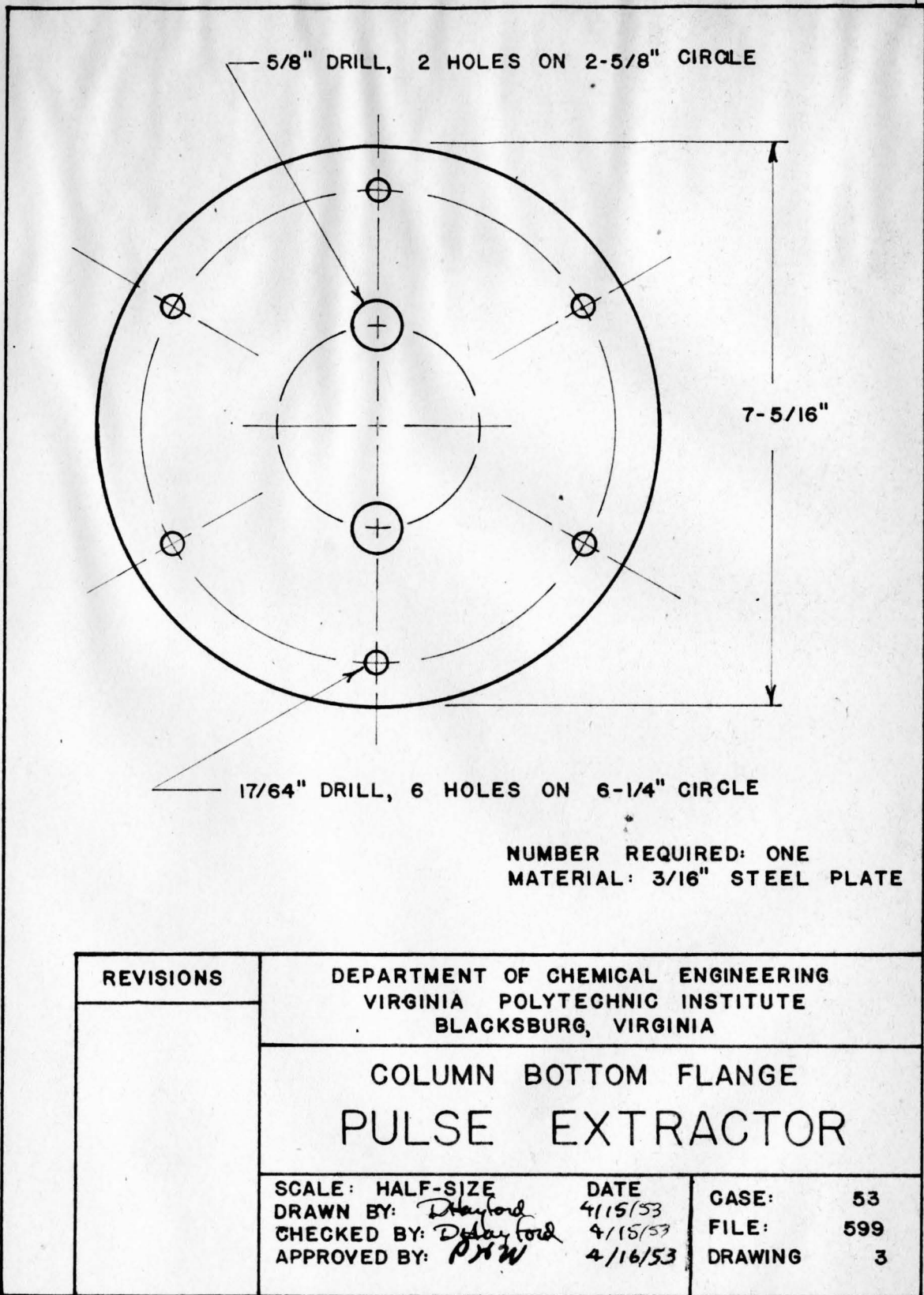
TABLE V

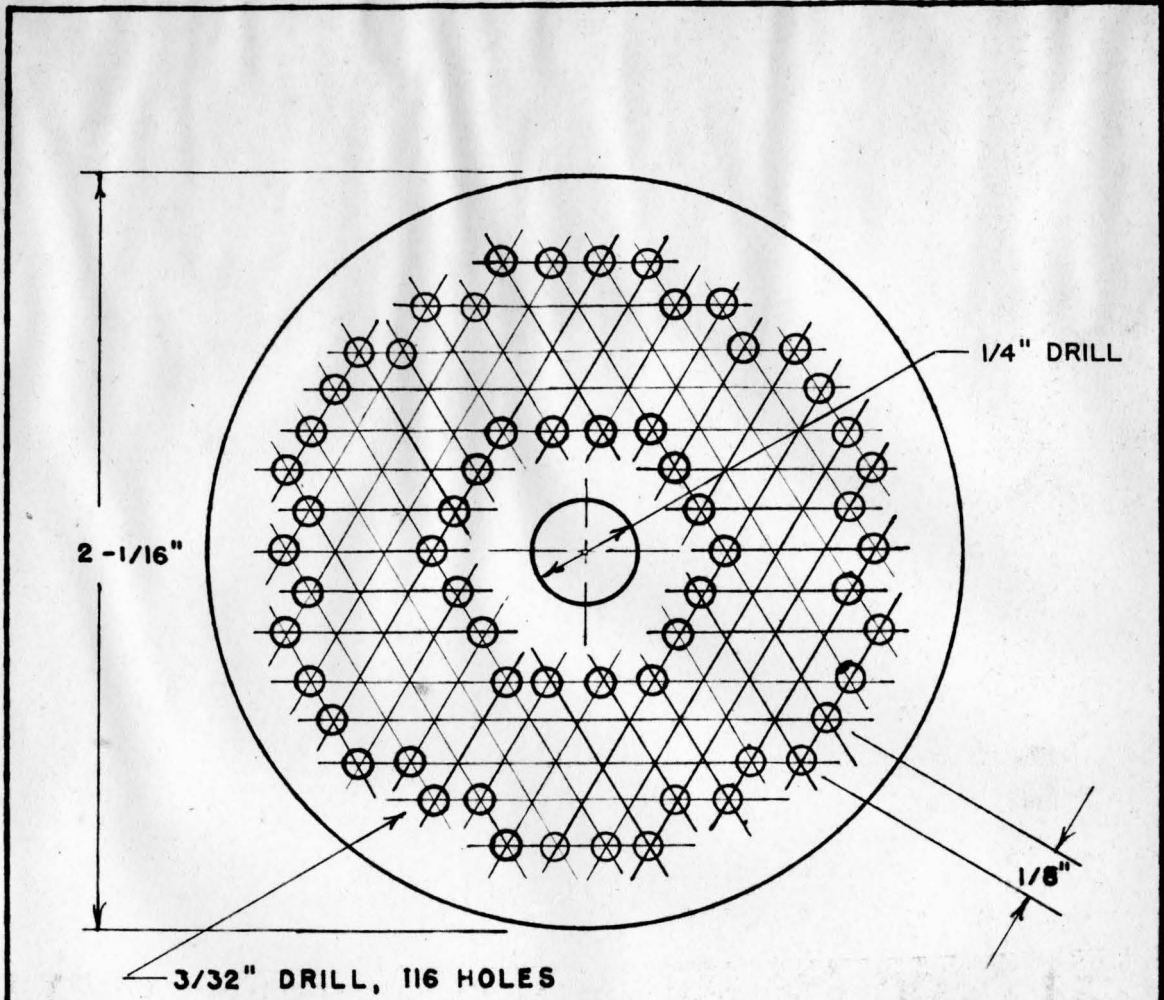
Bill of Materials for an Experimental Two-inch Pulse Extractor and Auxiliary Apparatus

Item	Quantity	Description	Supplier
Bellows	1 ea	Sylphon No 1171 2-ply brass	Fulton Sylphon Div. Knoxville, Tenn.
Glass pipe	4 ft	2-inch flanged pyrex	Sentinel Glass Co. Hatboro, Pa.
Glass tee	1 ea	4 x 2 x 4-inch pyrex	Sentinel Glass Co. Hatboro, Pa.
Motor	1 ea	1/4 hp, 1720 rpm, 110 volts, 60 cycle induction	Montgomery Ward Baltimore, Md.
Motor	2 ea	1/4 hp, 1750 rpm, 110 volts, 60 cycle induction	Sears, Roebuck and Co. Greensboro, N. C.
Pressure gages	4 ea	Ashcroft -30 in Hg to 100 lb per sq in	Noland Co., Roanoke, Va.
Pumps	2 ea	Roper model K5	Noland Co., Roanoke, Va.
Reducer	1 ea	Boston gear 80 to 1 ratio, input 0.15 hp at 1500 rpm	Noland Co., Roanoke, Va.
<u>Hardware</u>			
Bearings	2 ea	Tinkin series 68, 1/4 x 1-inch ball-type	Auto Spring and Bearing Co. Roanoke, Va.
	2 ea	3/4-inch, Dodge split journal, babbitted-type	Noland Co., Roanoke, Va.
Belt	1 ea	Type A-31 Veelos link V-belt	Noland Co., Roanoke, Va.
Carboys	4 ea	5-gallon glass	Department of Chemistry Blacksburg, Va.
Clamps	4 ea	1/2-inch tubing	Noland Co., Roanoke, Va.
Collars	2 ea	3/4-inch shaft	Noland Co., Roanoke, Va.
Coupling	1 ea	3/4 x 1-inch ball-type	Noland Co., Roanoke, Va.
Flange sets	2 ea	2-inch Corning standard	Sentinel Glass Co. Hatboro, Pa.
	1 ea	4-inch Corning standard	Sentinel Glass Co. Hatboro, Pa.
Funnel	1 ea	9-inch polyethylene	Fisher Scientific Co. Silver Spring, Md.
Gaskets	1 ea	3/16-inch x 9-inch diameter silicone rubber	Dow-Corning Co. Midland, Mich.
	2 ea	2-inch Corning JM-84-S	Sentinel Glass Co. Hatboro, Pa.
	2 ea	4-inch Corning JM-84-S	Sentinel Glass Co. Hatboro, Pa.
Pulleys	3 ea	1/2 x 2-inch single V-belt	Noland Co., Roanoke, Va.
	2 ea	1/2 x 4-inch single V-belt	Noland Co., Roanoke, Va.
	1 ea	3/4 x 4-inch single V-belt	Noland Co., Roanoke, Va.
Turnbuckles	9 ea	3/16 x 3-inch eye-type	Blacksburg Hardware Co. Blacksburg, Va.
Wire	100 ft	No 8 piano	Blacksburg Hardware Co. Blacksburg, Va.
<u>Structural Shapes</u>			
Angle	9 ft	3/4 x 3/4 x 1/16-inch steel	Noland Co., Roanoke, Va.
	6 ft	2 x 2 x 1/4-inch steel	Noland Co., Roanoke, Va.
Channel	1 1/2 in	4 x 2 x 1/4-inch steel	Noland Co., Roanoke, Va.
Lumber	5 ft	1 x 4-inch finished	New River Lumber Co. Blacksburg, Va.
	5 ft	2 x 4-inch semi-finished	New River Lumber Co. Blacksburg, Va.
	6 ft	2 x 4-inch finished	New River Lumber Co. Blacksburg, Va.
Plate	2 ea	3/16-inch x 8-inch diameter copper	Industrial Engineering Dept. Blacksburg, Va.
	16 ea	1/4-inch steel, 2 x 2-inch	Noland Co., Roanoke, Va.
	1 ea	1/4-inch steel, 2 1/2 x 28-inch	Noland Co., Roanoke, Va.
	2 ea	3/16-inch steel, 8-inch diameter	Noland Co., Roanoke, Va.
	1 ea	3/8-inch steel, 13-1/2 x 12-inch	Noland Co., Roanoke, Va.
	3 ea	3/8-inch steel, 13-inch diameter	Noland Co., Roanoke, Va.
	1 ea	3/8-inch steel, 8-inch diameter	Noland Co., Roanoke, Va.
Round stock	4 ft	1/4-inch aluminum	Whitehead Metal Products Co. Baltimore, Md.
	11 in	3/4-inch steel	Noland Co., Roanoke, Va.
	14 in	3/4-inch steel	Noland Co., Roanoke, Va.
<u>Fasteners</u>			
Anchors	3 ea	1/4-inch U.S.E. Korker	Noland Co., Roanoke, Va.
	6 ea	5/16-inch U.S.E. Korker	Noland Co., Roanoke, Va.
Bolts	100 ea	1/4 x 1-1/2-inch square head	Noland Co., Roanoke, Va.
	50 ea	1/4 x 2-inch square head	Noland Co., Roanoke, Va.
	20 ea	1/4 x 4-inch square head	Noland Co., Roanoke, Va.
	3 ea	3/8 x 2-inch square head	Noland Co., Roanoke, Va.
	8 ea	1/2 x 1-1/2-inch square head	Noland Co., Roanoke, Va.
	3 ea	1/2 x 2-inch square head	Noland Co., Roanoke, Va.
Lag screws	8 ea	1/4 x 2-inch square head	Noland Co., Roanoke, Va.
	3 ea	3/8 x 2-inch square head	Noland Co., Roanoke, Va.
Nuts	16 ea	1/4-NC 20-2 square	Noland Co., Roanoke, Va.
	2 ea	3/4-NC 10-2 hex	Noland Co., Roanoke, Va.
Screws	6 ea	1-inch No 9	Noland Co., Roanoke, Va.
	36 ea	1-1/2-inch No 11	Noland Co., Roanoke, Va.
Screw eyes	6 ea	1/2-inch eye No 6	Blacksburg Hardware Co. Blacksburg, Va.
Stove bolts	12 ea	1/4 x 2-inch round head	Noland Co., Roanoke, Va.
	6 ea	5/16 x 2-inch round head	Noland Co., Roanoke, Va.
Washers	100 ea	1/4-inch	Noland Co., Roanoke, Va.
	20 ea	3/8-inch	Noland Co., Roanoke, Va.
	3 ea	1/2-inch	Noland Co., Roanoke, Va.
<u>Pipe and Pipe Fittings</u>			
Bushings	1 ea	1/4 x 3/8-inch black iron	Noland Co., Roanoke, Va.
	2 ea	3/8 x 1/2-inch galvanized	Noland Co., Roanoke, Va.
Couplings	4 ea	1/4-inch black iron	Noland Co., Roanoke, Va.
Elbows	7 ea	1/4-inch black iron	Noland Co., Roanoke, Va.
	2 ea	3/8-inch galvanized	Noland Co., Roanoke, Va.
Flanges	8 ea	1/2-inch No 7 black iron	Noland Co., Roanoke, Va.
	4 ea	1/2-inch No 9 black iron	Noland Co., Roanoke, Va.
	4 ea	1-inch A.S.M.E. 125-pound	Noland Co., Roanoke, Va.
Nipples	9 ea	1/4-inch close black iron	Noland Co., Roanoke, Va.
	3 ea	1/4 x 1-inch black iron	Noland Co., Roanoke, Va.
	12 ea	1/4 x 2-inch black iron	Noland Co., Roanoke, Va.
	3 ea	1/4 x 3-inch black iron	Noland Co., Roanoke, Va.
	2 ea	1/4 x 6-inch black iron	Noland Co., Roanoke, Va.
	3 ea	3/8-inch close galvanized	Noland Co., Roanoke, Va.
	5 ea	3/8 x 1-inch galvanized	Noland Co., Roanoke, Va.
	2 ea	3/8 x 4-inch galvanized	Noland Co., Roanoke, Va.
	2 ea	3/8 x 6-inch galvanized	Noland Co., Roanoke, Va.
	1 ea	1 x 4-inch black iron	Noland Co., Roanoke, Va.
Pipe	6 ft	1/4-inch black iron	Noland Co., Roanoke, Va.
	6 ft	3/8-inch galvanized	Noland Co., Roanoke, Va.
	13 ft	1/2-inch black iron	Noland Co., Roanoke, Va.
Reducers	1 ea	1/4 x 3/8-inch galvanized	Noland Co., Roanoke, Va.
	1 ea	3/4 x 1-inch black iron	Noland Co., Roanoke, Va.
Street elbows	2 ea	1/4-inch black iron	Noland Co., Roanoke, Va.
	4 ea	3/8-inch galvanized	Noland Co., Roanoke, Va.
Tees	13 ea	1/4-inch black iron	Noland Co., Roanoke, Va.
	4 ea	3/8-inch galvanized	Noland Co., Roanoke, Va.
Unions	2 ea	1/4-inch black iron ground joint	Noland Co., Roanoke, Va.
	2 ea	3/8-inch galvanized ground joint	Noland Co., Roanoke, Va.
Valves	4 ea	1/4-inch gate, brass	Noland Co., Roanoke, Va.
	5 ea	1/4-inch globe, brass	Noland Co., Roanoke, Va.
	1 ea	1/4-inch diaphragm air reducing	Noland Co., Roanoke, Va.
	1 ea	1/4-inch swing check, brass	Noland Co., Roanoke, Va.
	3 ea	3/8-inch gate, brass	Noland Co., Roanoke, Va.
	2 ea	3/8-inch needle, brass	Noland Co., Roanoke, Va.
	2 ea	3/8-inch swing check, brass	Noland Co., Roanoke, Va.
<u>Tubing and Tubing Fittings</u>			
Connectors	7 ea	1/4T x 1/4M-inch, brass	Whitehead Metal Products Co. Baltimore, Md.
	7 ea	3/8T x 3/8M-inch compression, brass	Auto Spring and Bearing Co. Roanoke, Va.
	3 ea	3/8T x 3/8M-inch compression elbow, brass	Auto Spring and Bearing Co. Roanoke, Va.
Tees	4 ea	3/8T-inch compression, brass	Auto Spring and Bearing Co. Roanoke, Va.
	4 ea	3/8T-inch Parker 6 JBT, brass	Whitehead Metal Products Co. Baltimore, Md.
	4 ea	3/8T x 3/8M x 3/8T-inch Parker 6 MBT, brass	Whitehead Metal Products Co. Baltimore, Md.
	4 ea	3/8T x 3/8F x 3/8T-inch Parker 6 RBT, brass	Whitehead Metal Products Co. Baltimore, Md.
Tubing	115 ft	3/8-inch soft seamless	Noland Co., Roanoke, Va.
	6 ft	3/8-inch Tygon	Fisher Scientific Co. Silver Spring, Md.
Unions	4 ea	3/8T x 3/8T-inch bulkhead, Parker 3115-2 HBT6, brass	Whitehead Metal Products Co. Baltimore, Md.
	4 ea	3/8T x 3/8T-inch, sarun	Whitehead Metal Products Co. Baltimore, Md.
Valves	4 ea	3/8T x 3/8T-inch compression, brass	Auto Spring and Bearing Co. Roanoke, Va.
	16 ea	3/8T x 3/8T-inch compression plug cock, brass	Auto Spring and Bearing Co. Roanoke, Va.



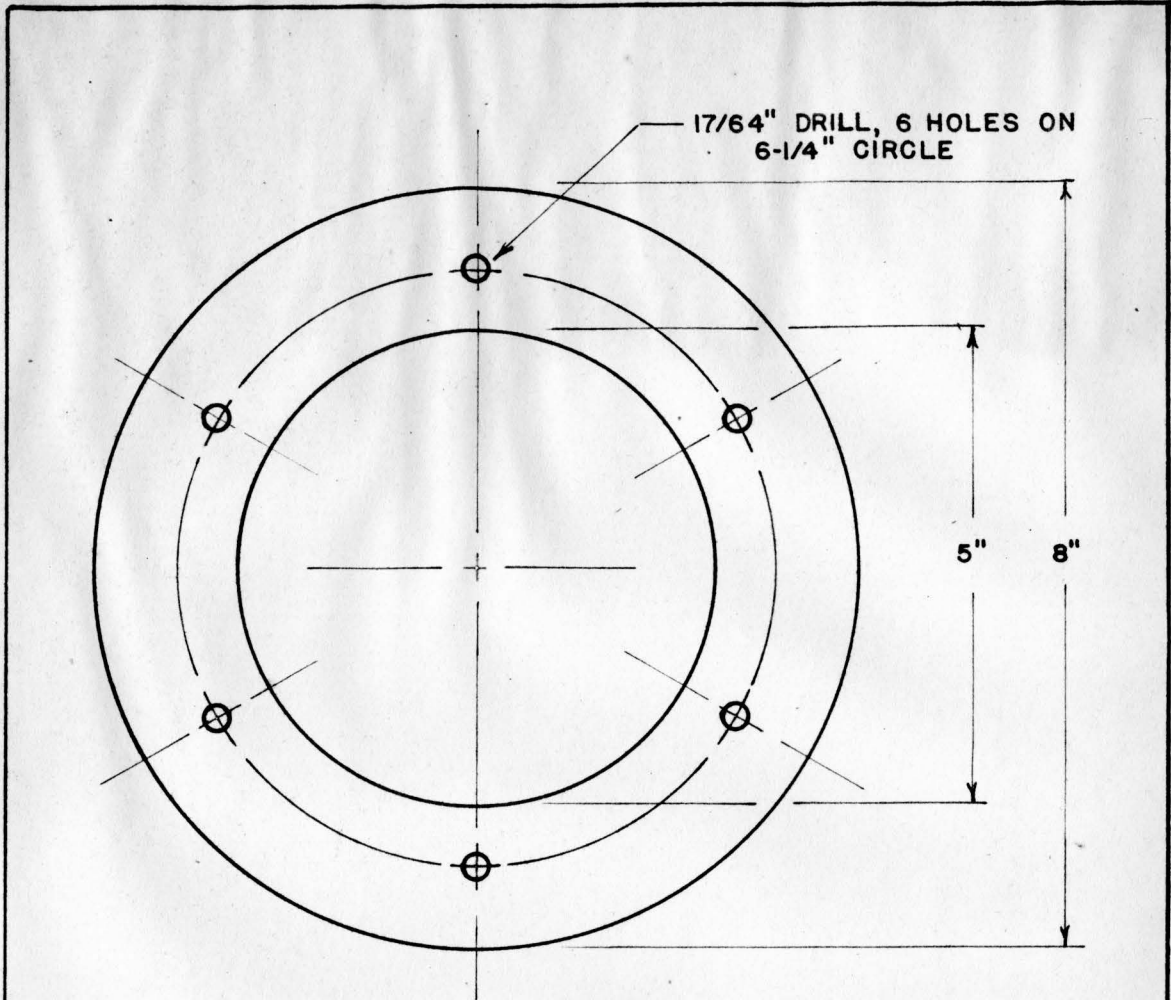
REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA	
	COLUMN TOP FLANGE PULSE EXTRACTOR	
	SCALE: FULL DRAWN BY: DAH-SHR CHECKED BY: D. Dayford APPROVED BY: PHW	DATE 4/12/53 4/15/53 4/16/53





NUMBER REQUIRED: 8
MATERIAL: 1/8" STEEL PLATE
FREE AREA: 2.59 %

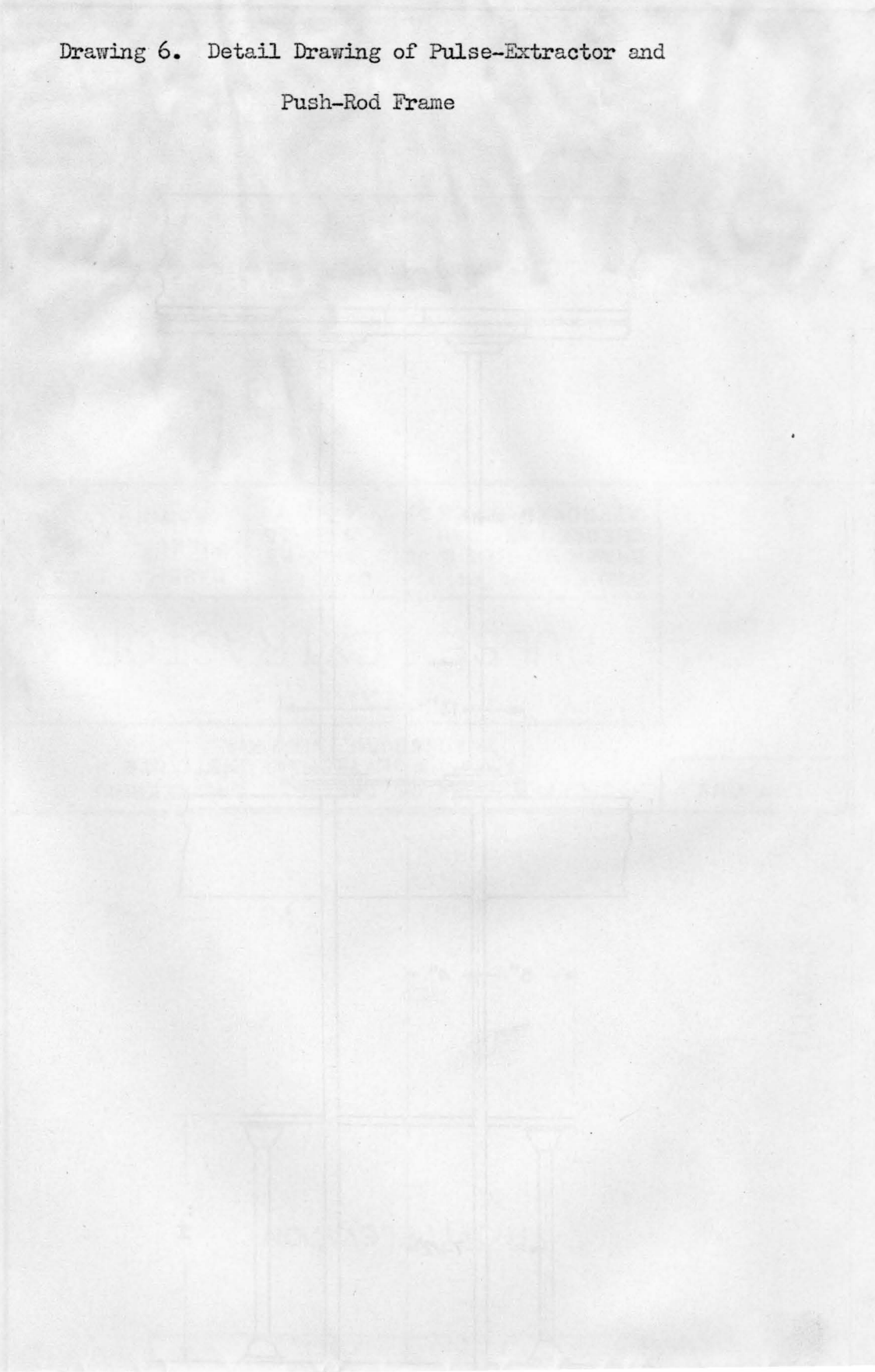
REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA		
	BAFFLE PLATE PULSE EXTRACTOR		
	SCALE: DOUBLE SIZE DRAWN BY: <i>D. Daylord</i> CHECKED BY: <i>D. Daylord</i> APPROVED BY: <i>P. H. W.</i>	DATE 4/15/53 4/15/53 4/16/53	CASE 53 FILE 599 DRAWING 4

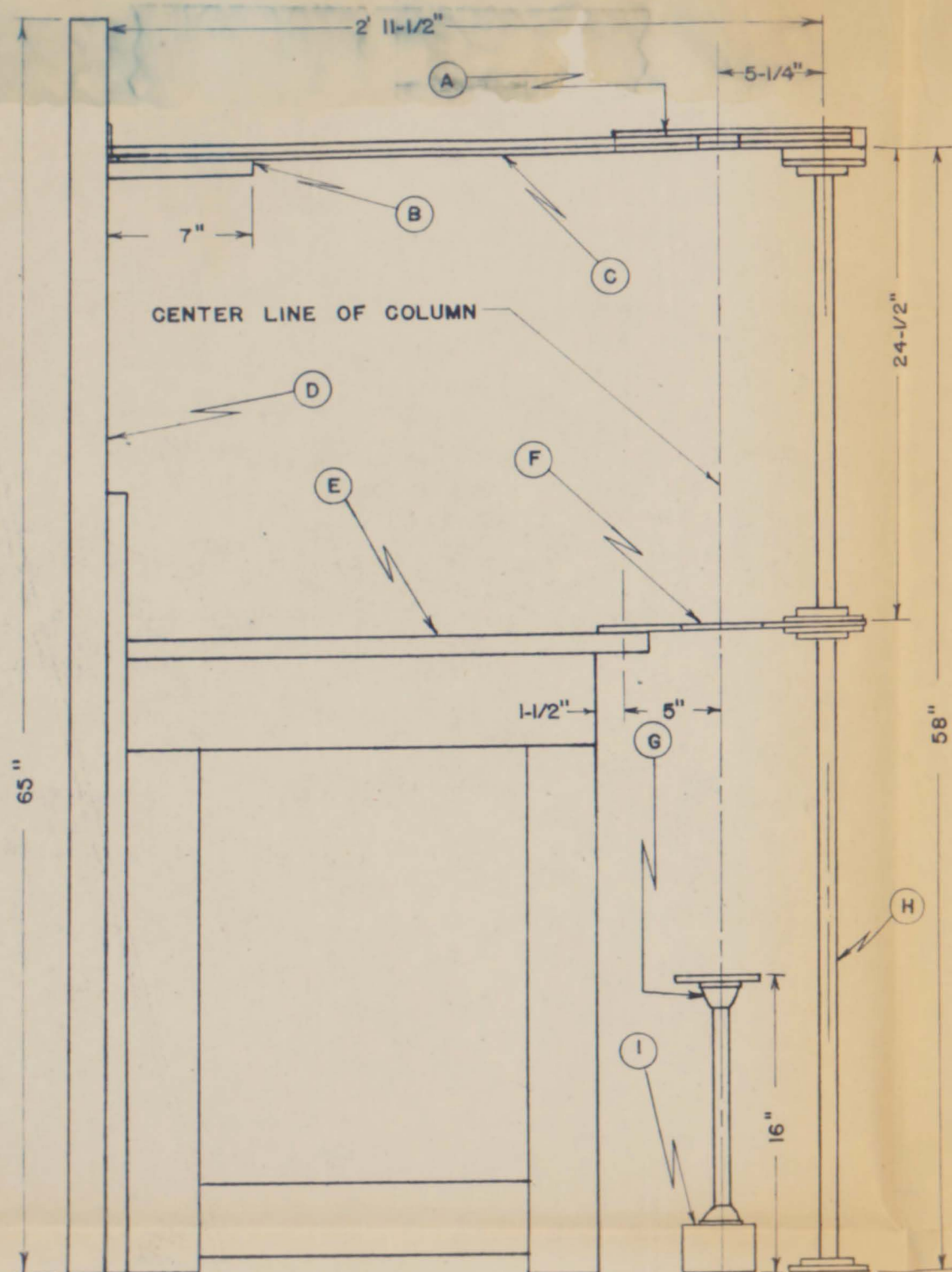


NUMBER REQUIRED: TWO
MATERIAL: ONE, 3/16" COPPER PLATE
ONE, 3/8" STEEL PLATE

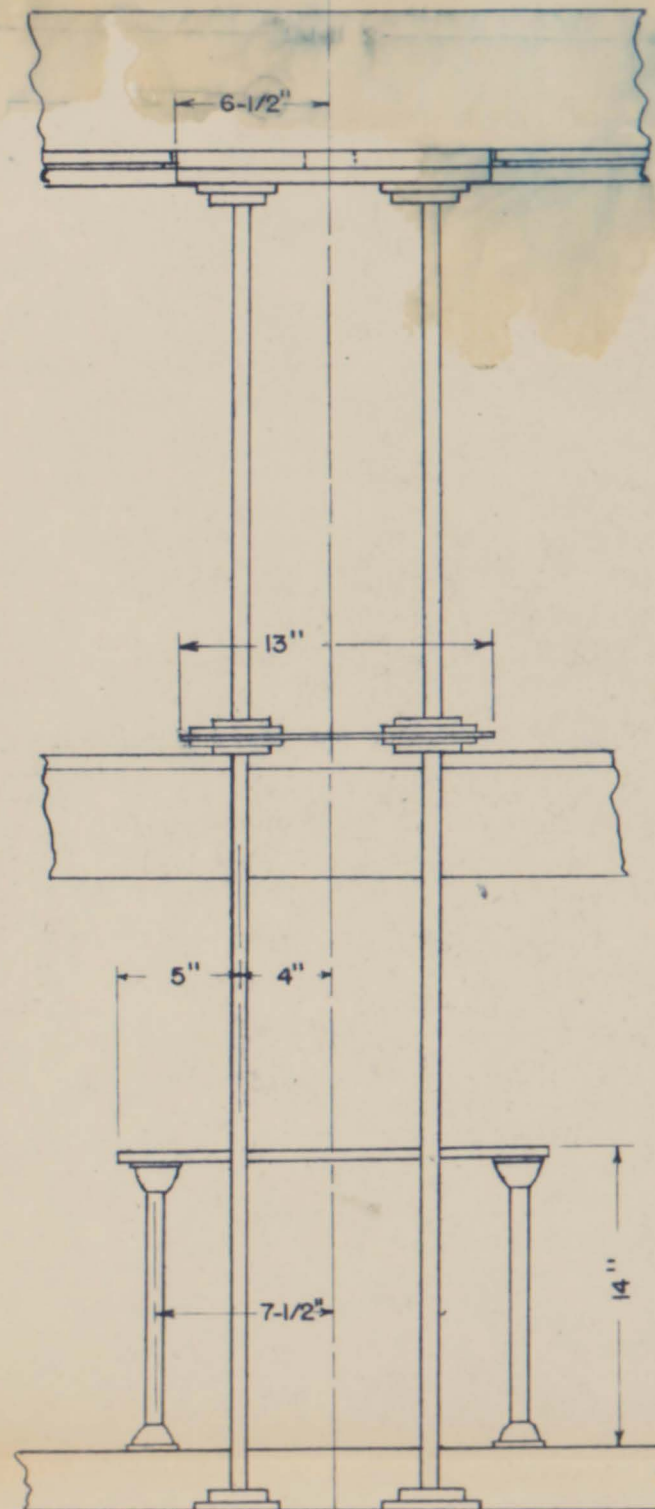
REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA	
	TOP BELLOWS FLANGE PULSE EXTRACTOR	
	SCALE: HALF SIZE DRAWN BY: <i>D. Hayford</i> CHECKED BY: <i>D. Hayford</i> APPROVED BY: <i>AH W</i>	DATE 4/15/53 4/15/53 4/17/53

Drawing 6. Detail Drawing of Pulse-Extractor and
Push-Rod Frame





LEFT SIDE ELEVATION



FRONT ELEVATION

LEGEND

- (A) HORIZONTAL COLUMN SUPPORT
- (B) BOTTLE RACK
- (C) HORIZONTAL SUPPORT FRAME, 3/4 X 3/4 X 1/16-IN. ANGLE IRON
- (D) BOTTLE RACK SUPPORT
- (E) TABLE
- (F) COLUMN BASE PLATE, 13X13X3/8-IN. MILD STEEL
- (G) PUSH-ROD FRAME
- (H) COLUMN FRAME LEGS, 1/2-IN. BLACK IRON PIPE
- (I) FRONT CAM-ASSEMBLY LEG

REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA	
	FRAME PULSE EXTRACTOR	
	SCALE: 1-1/2" = 1'	DATE
	DRAWN BY: S.H.R. & J.E.L.	4-26-53
	CHECKED BY: DAH	4-26-53
	APPROVED BY: <i>P.N. 2/</i>	4-28-53
	CASE: 53	FILE: 599
	DRAWING	6

Pulse-extractor Cam Assembly. The cam assembly was constructed in the Department of Chemical Engineering for this investigation. Details of the cam assembly are shown in Drawing 7, page 66. The bill of materials for this assembly may be found in Table V, page 59.

Pulse-extractor Push-rod Assembly. The push-rod assembly was constructed in the Department of Chemical Engineering for this investigation. Details of this assembly are shown in Drawing 8, page 67. The bill of materials for the push-rod assembly may be found in Table V, page 59.

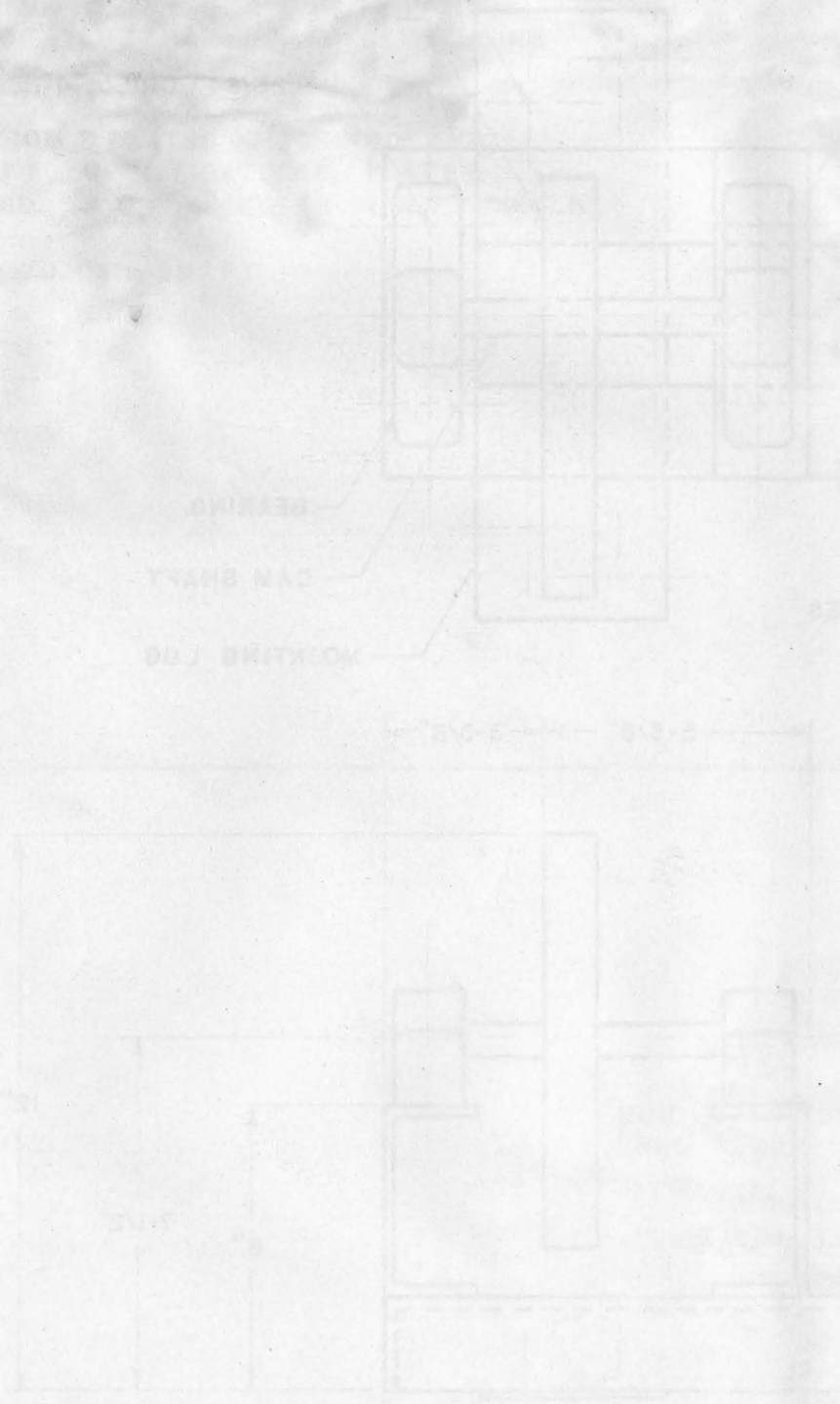
Pulse-extractor Tubing and Storage. The tubing and storage were constructed for this investigation in the Department of Chemical Engineering. The materials required for this section are shown in Table V, page 59.

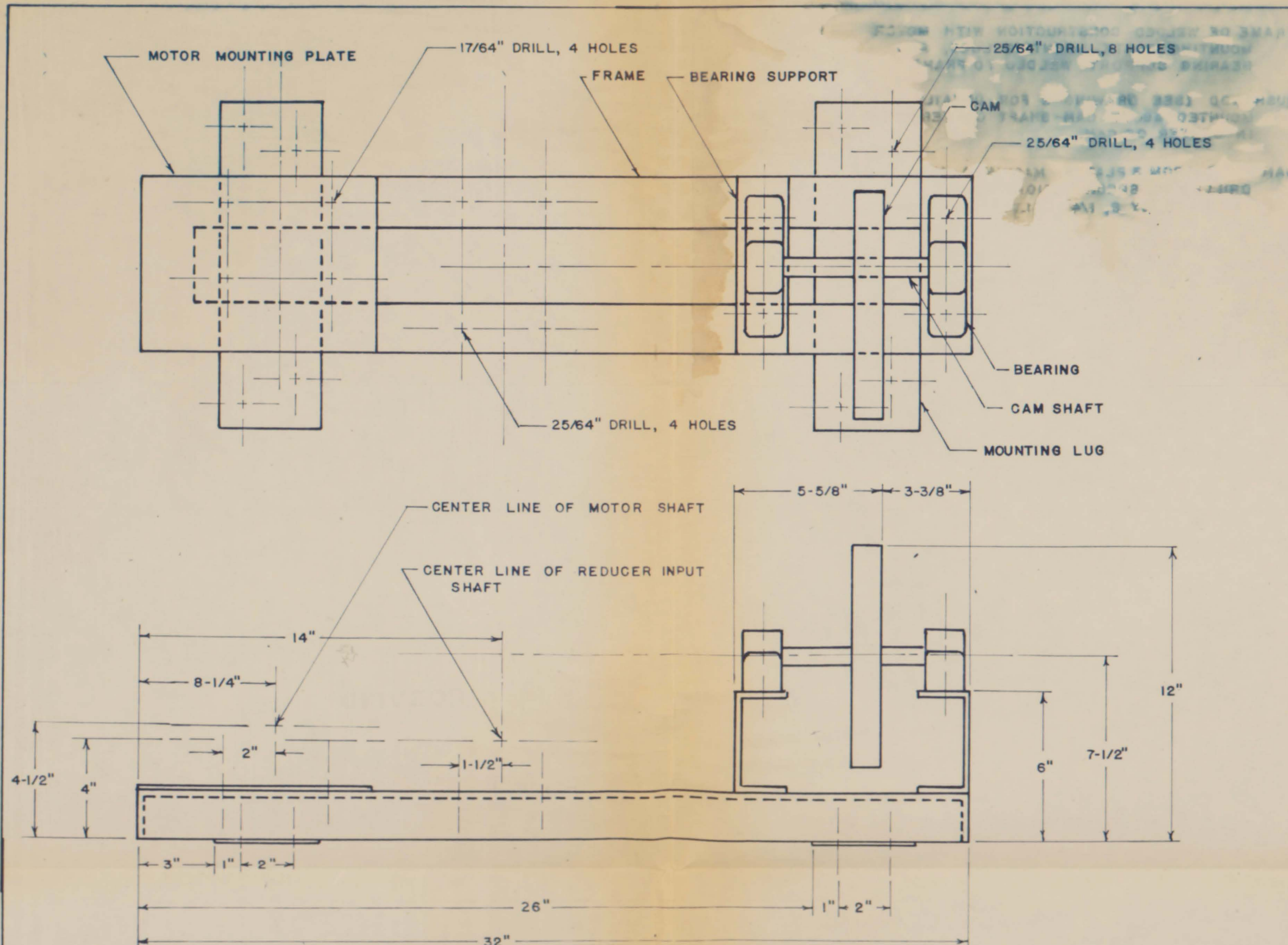
Refractometer. Abbe, A. O. Spencer model 1777, range 1.3000 to 1.7100, sensitivity 0.0002, reads directly in n_D units, serial No 789326721. Obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used to determine the refractive index of samples.

Thermometer. Mercury in glass, range -10 to 120° C. Obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used to measure temperature of samples.

Drawing 7. Detail Drawing of Pulse-Extractor

Cam Assembly



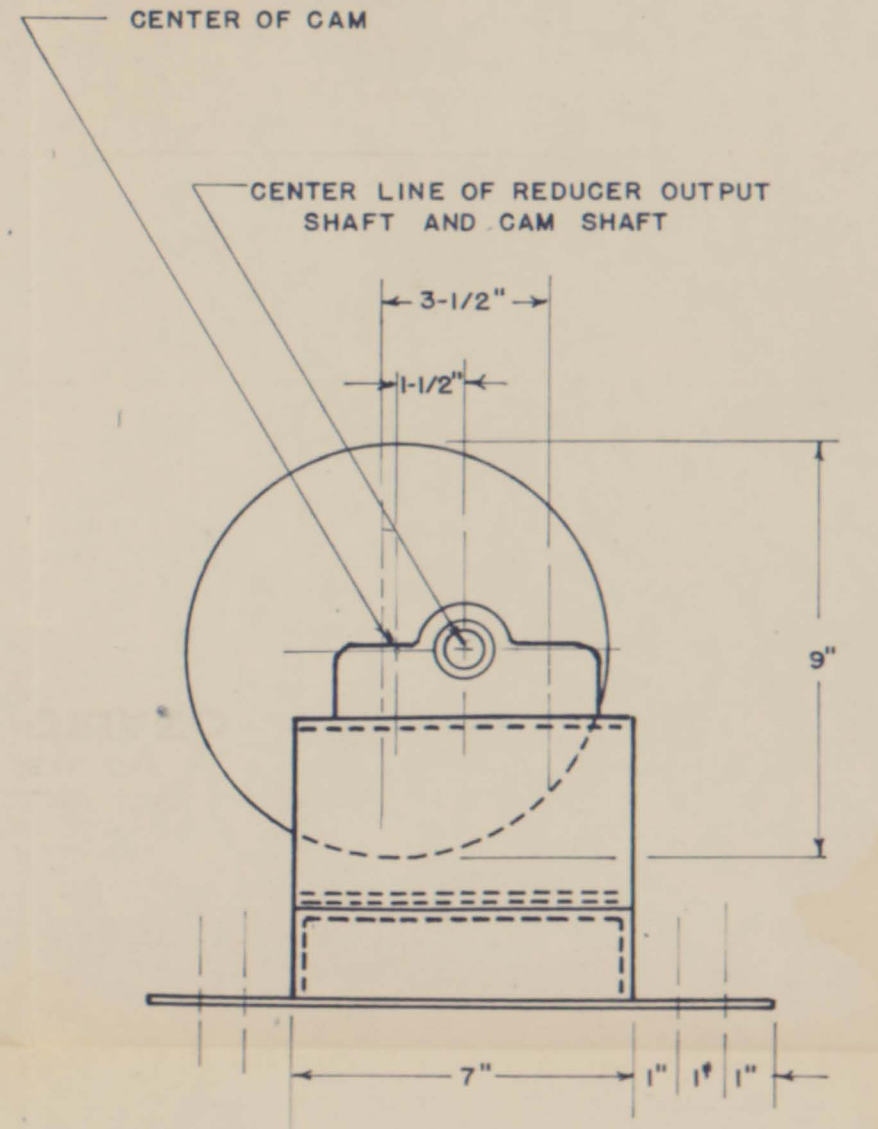


NOTES: FRAME OF WELDED CONSTRUCTION WITH MOTOR MOUNTING PLATE, MOUNTING LUGS, AND BEARING SUPPORT WELDED TO FRAME.

PUSH ROD (SEE DRAWING 8 FOR DETAILS) MOUNTED ABOVE CAM-SHAFT CENTERLINE IN CENTER OF CAM.

CAM MADE FROM 3 PLATES, MACHINED AND DRILLED TO SPECIFICATIONS, PLATES FASTENED BY 8, 1/4" BOLTS IN A 7" CIRCLE.

CAM BRAZED TO CAM SHAFT.

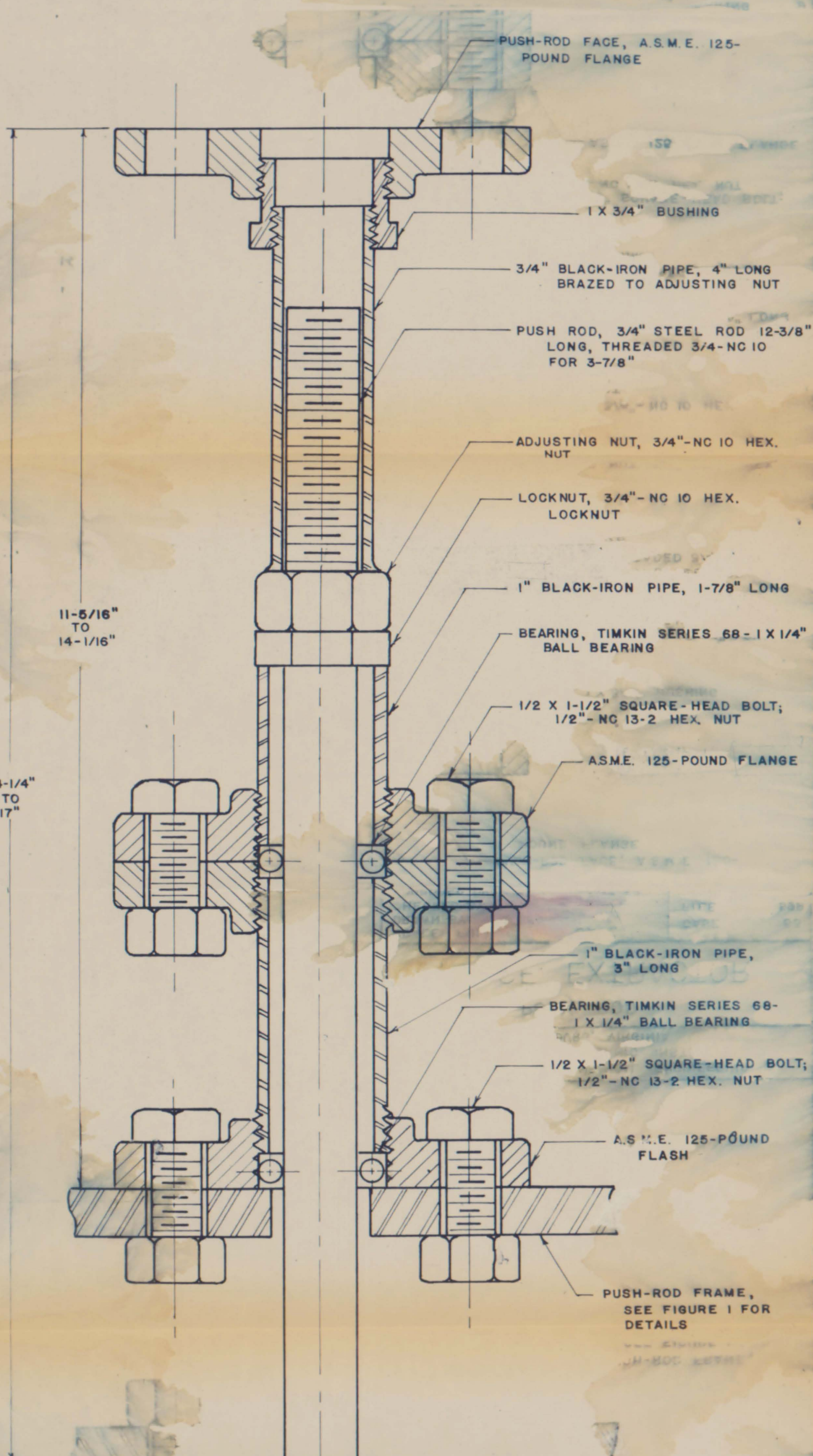


PART	NUMBER REQUIRED	MATERIAL
CAM	1	3/8" STEEL PLATE
CAM SHAFT	1	3/4" STEEL ROD
BEARING	2	PURCHASED
BEARING SUPPORT	2	4 X 2 X 1/4" STEEL CHANNEL
FRAME	1	2 X 2 X 1/4" ANGLE IRON
MOUNTING LUG	2	1/8" STEEL PLATE
MOTOR MOUNTING PLATE	1	1/8" STEEL PLATE

REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA		
	CAM ASSEMBLY PULSE EXTRACTOR		
	SCALE: 3" = 1'	DATE 4-16-53	CASE 53
	DRAWN BY: <i>DAM</i>	4-16-53	FILE 599
	CHECKED BY: <i>DAM</i>	4-17-53	DRAWING 7
	APPROVED BY: <i>PXW</i>		

Drawing 8. Detail Drawing of Pulse-Extractor

Push-Rod Assembly



11-5/16"
TO
14-1/16"

14-1/4"
TO
17"

REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA	
	PUSH ROD PULSE EXTRACTOR	
	SCALE: FULL	DATE
	DRAWN BY: <i>DWH</i>	4-16-53
	CHECKED BY: <i>DWH</i>	4-16-53
	APPROVED BY: <i>PXV</i>	4-17-53
	CASE	53
	FILE	599
	DRAWING	8

✓ Timer. Electric "Precision Time It," graduated in 1/10 of a second, continuous with 4 digits, 115 volts, 60 cycle, 5 watts. Obtained from Precision Scientific Company, Chicago, Illinois. Used for timing tests.

7490ⁿ
Tools. Various hand tools, welding equipment, vises, drills and lathes were used in the construction of the pulse extractor and its auxiliary apparatus. Available in the Department of Chemical Engineering, Virginia Polytechnic Institute, Blacksburg, Virginia.

✓ Westphal Balance. Specific gravity, stainless steel, reinman-type plummet. Obtained from the Fisher Scientific Company, Silver Spring, Maryland. Used to determine the specific gravity of samples.

Method of Procedure

The method of procedure followed in the performance of this investigation consisted of the construction of the pulse extractor, method of analysis, determination of refractive-index data, operation of the extractor, and testing procedure.

Construction of the Column Frame. The column frame refers to the supporting structure for the pulse extractor. This frame is shown in detail in Drawing 6, page 64. An integral part of the column frame was a standard laboratory table 39 inches high. This table contained a bottle rack 26 inches above the table top and an additional shelf eight inches above the floor. This table was used to support the column frame and most of the auxiliary apparatus used in this investigation. The terms "left" and "right" refer to directions as observed by a person facing the front of the laboratory table.

The base plate, upon which the column actually rested, was a steel plate $13\frac{1}{2} \times 12$ inches cut from a $\frac{3}{8}$ -inch steel plate. One end of the plate overlapped the front edge of the table by $1\frac{1}{2}$ inches. This edge of the base plate was fastened to the table with three $\frac{1}{2}$ -inch bolts. The other edge of the base plate was supported by a pipe frame consisting of sections of $\frac{1}{2}$ -inch pipe with $\frac{1}{2}$ -inch flanges bolted to the base plate and anchored

to the floor. It was found necessary to anchor the table to the floor to prevent its movement relative to the frame legs.

To hold the extractor and bellows in place, six $17/64$ -inch holes on a $6-1/4$ -inch center were drilled in the center of the 12×12 -inch section of the base plate. Concentric to the bolt circle, a hole for the bellows, four inches in diameter, was cut.

The frame also contained a wooden horizontal column support to prevent lateral movement of the column. The column support was a 12×12 -inch square, with a two-inch diameter hole cut in the center. This support was constructed so that it could be dismantled to allow the removal or installation of the pulse extractor. The front of the column support was held in position, 26 inches above the base plates by $1/2$ -inch pipe legs concentric to the legs holding the base plate. The back was supported by $3/4 \times 3/4 \times 1/16$ -inch angle iron braces that extended back to and were bolted to the bottle rack of the laboratory table.

Construction of the Column Flanges. The flanges that closed the column top and the side outlet of the tee, were constructed for this equipment. Details of these flanges may be seen in Drawings 2 and 3, pages 60 and 61, respectively. These flanges differed in dimensions; but they were, however, constructed following a similar procedure. For brevity, only the procedure followed in constructing the column bottom flange will be given. The circular flange was cut to a diameter of $7-1/2$ inches from

a 3/16-inch steel plate. The rough flange was machined to 7-5/16 inches in diameter. After machining, six 17/64-inch holes were drilled on a 6-1/4 inch bolt circle. Two 5/8-inch holes were then drilled on centers 2-5/8 inches apart on a diameter for the bulkhead union. The bulkhead unions were inserted into position and brazed to the flange.

Construction of Bellows Flanges. A cross section of the bellows, with its three flanges, is shown in Drawing 1, page 58. Details of the top bellows flanges are seen in Drawing 5, page 63. The bellows, as received from the manufacturer, was open on both ends. Before use it was necessary to seal off one end and to provide some means of fastening the other end of the bellows to the extractor base plate. The bellows was originally sent to the Industrial Engineering Machine Shop, Virginia Polytechnic Institute, to be fitted with copper flanges. The top flange was "doughnut" shaped with an inside diameter of 6 inches and an outside diameter of 7-1/2 inches. Six 17/64-inch holes were drilled on a 6-1/4-inch circle. The bottom flange was simply a 5-1/2-inch circle cut from 3/16-inch copper sheet. This flange was designed as a blank flange used to seal the bottom of the bellows. Both flanges were silver soldered to the bellows in this shop.

When the bellows was installed on the column, leaks were noticed along the silver-soldered joints. In addition to this leakage, the copper top flange was too soft to allow a tight seal

to be made against the gasket. The copper was warped even when a gasket material as soft as rubber was used. To stop the leaks both flanges were soft-soldered over the silver-soldered joints. To reinforce the top copper flange, a steel flange was made. This steel flange had identical dimensions to the copper flange except that it was cut from 3/8-inch steel plate. After machining and drilling, the finished flange was soft-soldered to the copper bellows flange.

Construction of Baffle Plates. Eight baffle plates were constructed, the details of which are shown in Drawing 4, page 62. These 2-1/16-inch plates were cut and machined from 1/8-inch steel sheet. After machining, one plate was selected as a standard. This plate was ruled, center punched, and drilled. One hundred sixteen 3/32-inch holes were drilled on a 1/8-inch triangular pattern. After this plate had been completed it was used as a template for drilling the other plates.

The plates were then mounted upon a 1/4-inch aluminum rod that had been threaded 1/4-NC 20. The plates were placed on the rod at four-inch intervals and held in place by steel 1/4-NC 20 square nuts.

Assembly of Column. The column was assembled in the following steps: Positioning of bellows and pyrex tee, positioning of extraction column, installation of baffle plates, installation of column top flange, and installation of column bottom flange.

Positioning of Bellows and Pyrex Tee. Because the bellows and pyrex tee were fastened back to back on the base plate as illustrated in Drawing 1, page 58, they had to be positioned together. The silicone rubber bellows gasket was placed on top of the bellows, and the bolts were forced up through the bellows flange, the gasket, and the base plate. The asbestos gasket for the pyrex tee was then placed on the base plate. Finally, the pyrex tee with its flange was lowered over the bolts. It was quite difficult to align the bellows, base plate, and pyrex flange in order to insert the bolts through the entire "sandwich." However, when all the bolts were through, nuts were placed on the bolts and they were tightened until both gaskets were sufficiently compressed to be liquid tight.

Positioning of Extraction Column. The flange was cut off one end of a four-foot section of Corning standard two-inch pipe. This section of pipe, now 45 inches long, formed the extraction column. The pipe was placed on top of the pyrex tee with the cut end up. The extraction column was then bolted into place, using a Corning standard two-inch flange set.

Installation of Baffle Plates. The plates, assembled on the rod, were inserted into the column. Because of the flare near the bottom of the pyrex column, the plates could

not pass all the way through the column. As a result, the bottom plate could not be installed lower than three inches above the bottom of the pyrex pipe. At this point the diameter of the column was too small to allow further insertion of the plates. As the plates were a tight fit, some difficulty was encountered in installing and removing them.

Before the column top flange was fastened into place, the top of the rod that held the baffle plates in place was cut off flush with the top of the gasket. It was necessary that this rod be the correct length to prevent any movement of the plates during the pulsation operation. If the rod was too long, a tight seal at the top of the column could not be obtained.

Installation of Column Top Flange. The flared end of the top of the column was cut off so that the plates could be inserted into the column. Without this flare, however, the standard end flanges for pyrex pipe could not be used. To seal the top of the column, the following method was used. A detail of this seal may be found in Drawing 1, page 53.

About one-half dozen turns of friction tape were wrapped on top of itself around the top of the column. The Corning pyrex flange and the column top flange were fastened together with friction tape between the two flanges. As the two flanges were forced together by tightening the bolts,

the friction tape was pulled along the pipe so that it pressed firmly against the top gasket. The pressure of the tape against the top gasket sealed this gasket to the column top flange, and also sealed the tape to the bottom of the gasket. However, because of the tapered shape of the Corning standard flange, the tape was also forced against the glass, making a water-tight seal. This type of seal was found to be enough to stop all leaks except at water flow rates approaching 20,000 pounds per hour-square foot.

Installation of Column Bottom Flange. The internal connections for the heavy-phase outlet and the light-phase inlet had to be fastened to the column bottom flange before it was inserted into the column. Details of the column bottom flange and these connections are shown in Drawing 1, page 58. The light-phase inlet consisted of an eight-inch length of $3/8$ -inch copper tubing, ending in a brass elbow pointing downward. The heavy-phase outlet consisted of a 14-inch length of $3/8$ -inch copper tubing bent down toward the bellows, and finished off with an eight-inch length of $1/2$ -inch tygon tubing. The tygon tubing was fastened to the end of the copper tubing with two $1/2$ -inch tubing clamps. The tygon tubing extended down to the bottom of the bellows. It was used because the copper tubing would interfere with the

flexing of the bellows. The column bottom flange was held to the side outlet of the tee by means of $1/4 \times 2$ -inch bolts fastened through a standard four-inch Corning flange.

Construction of Cam Assembly. The cam assembly consisted of the eccentric cam, a worm-gear reducer, an electric motor, and the framework required to support these parts. The cam assembly fits underneath of, and drives the push rod. The cam assembly must be strong enough to withstand the large stresses developed by the cam as it moves. The gear reducer selected had a reduction ratio of 80 to 1. This ratio was sufficient when coupled to a one-quarter horse power motor, to drive the cam under all conditions except severe binding of the moving metal parts.

The construction of the cam assembly may be broken down into the construction of the cam-assembly frame, the construction of the cam, and the final assembly.

Construction of the Cam-Assembly Frame. The cam-assembly frame was constructed from $2 \times 2 \times 1/4$ -inch angle iron welded into a 7×21 -inch rectangle as shown in Drawing 7, page 66. Pieces of $1/8$ -inch sheet steel were welded to the finished frame to form the motor mounting plate and the mounting lugs. Two pieces of $2 \times 4 \times 1/4$ -inch steel channels each 7 inches long were then welded near one end of the frame 9 inches apart to support the bearings. The finished frame was drilled to the dimensions shown in the drawing for mounting the frame, the motor, the gear reducer, and the bearings.

Construction of Cam. The cam was constructed from three circular sections, ten inches in diameter, of 3/8-inch steel plates bolted together. The plates were bolted together with eight 1/4 x 2-inch bolts located in a 7-1/2-inch bolt circle. After the machining was finished, a 3/4-inch hole was drilled in the cam 1-1/2 inches eccentric to the center. A 3/4-inch shaft 11 inches long was centered through the cam and welded into position.

Final Cam Assembly. After the cam and frame were constructed, the final assembly took place as shown in Drawing 7, page 66. The motor and gear reducer were securely bolted to the frame in their respective positions.

The cam bearings were aligned with respect to the gear-reducer output shaft and bolted into place. The axial trueness of the bearings was tested by placing a drop of oil on the bottom seat and slowly rotating the cam shaft with the bearing caps removed. Adjustments of the bearing alignment were continued until the bearing deposited a thin film of oil around the shaft. After final adjustment, the bearing mounting bolts were tightened. An additional check was then made to make sure that the bearings had not accidentally shifted while being tightened. As the bearing alignment was still satisfactory, the bearing caps were placed in position and tightened.

The cam shaft and the output shaft of the gear reducer were connected by means of a ball-type flexible coupling. Holes, $3/8$ inch in diameter were started in the two shafts to give the points of the set screws something to "bite" into. At first it was thought that a key and keyway would be required in addition to these holes, but experience had proven that this was unnecessary.

The cam assembly was fastened to two wooden legs by means of $1/2$ -inch lag screws. These wooden legs consisted of 2 x 4-inch boards, 24 inches long, anchored to the floor with the four-inch side down.

Construction of the Push-Rod Frame. The push-rod frame consisted of a horizontal plate supporting the push-rod bearing, and two vertical legs supporting the horizontal plate. Details of this frame are shown in Drawing 6, page 64. The plate was cut from $1/2$ x 4-inch steel strip and drilled with a $17/64$ -inch drill for the adjusting brackets and flanges and with a $17/32$ -inch drill for the push-rod bearing. The two $1/2$ -inch pipe legs, 14 inches long, were mounted on the plate by means of flanges. The other end of the legs were fastened to the same wooden block as the front of the cam-assembly frame. As the push-rod bearing fits rigidly on the push-rod frame, the entire frame had to be moved in order to center the push-rod. To accomplish this centering, two sections of $1/4$ -inch threaded rod were attached to the legs

of the pulse-extractor frame and to the horizontal plate of the push-rod frame. By adjusting the nuts on these rods, the push-rod frame could be moved about one inch in any direction in the horizontal plane.

Construction of a Push-Rod Assembly. The push-rod assembly, illustrated in Drawing 8, page 67, consists of the adjustable push-rod and its bearing. The construction of this assembly will be considered in three sections. These three sections are the construction of the push rod, the construction of the push-rod bearing, and the final assembly of the unit.

Construction of the Push Rod. The push rod consisted of a $3/4$ -inch shaft and an adjustable face. For the construction of the rod a 14-inch length of $3/4$ -inch circular stock was threaded for about four inches. The push-rod face consisted of a one inch, 125 pound, A.S.M.E. pipe flange. This pipe flange was attached to a $1 \times 3/4$ -inch bushing and a 4-inch length of $3/4$ -inch black iron pipe. A heavy series $3/4$ -NC 10-2 hexagonal nut was brazed to the pipe. Another nut was taken and cut to a $3/8$ -inch thickness. This cut section of the nut was threaded onto the push rod, followed by the push-rod face. The length of the push rod from the face to the butt end could be varied about 4 inches by turning the face relative to the push rod. The thin section of a nut was used as a lock nut.

Construction of the Push-Rod Bearing. The push-rod bearing consisted of a four-inch length of one-inch pipe containing a set of ball bearings at each end as shown in Drawing 8, page 67. The pipe was drilled and tapped in the center for a 1/8-inch National Pipe thread to receive a grease cup. At each end of this section of pipe a one-inch A.S.M.E. 125-pound flange was fastened. The ball-bearing sets were of such size that they would just pass through the one-inch flange, but they could be kept from passing through the flange by inserting a piece of one-inch pipe into the flange. The top ball-bearing set is held in place by another flange, blocked by a piece of pipe, that is bolted to the first flange. The short blocking section of the pipe was cut off flush with the top of the second flange. The bottom ball-bearing set is held in place by the frame that the push-rod assembly sits on.

Assembly of Push-Rod Unit. The completed push-rod bearing was fastened to its frame by means of four 1/2-inch bolts as shown in Drawing 8, page 67. The hole cut in the frame was just large enough to allow the push rod to pass through, and served to hold the bottom ball-bearing set in place. The bearing was accurately aligned with both the cam and the center of the column.

Before the push rod was inserted into its bearing (this must be done before the bellows is in place) a 1-1/2-inch length of one-inch pipe was placed over the bearing. This section of pipe limited the downward travel of the push rod. With its stroke limited in this fashion, the push rod never lost contact with the bellows.

Construction of Storage Section. The storage section consisted of four 5-gallon carboys mounted in pairs. The carboys were covered with hardware cloth as a safety measure. Each carboy was stoppered with a two-hole Number 8 rubber stopper. The carboys were placed on the laboratory table, two on the top level, and two on the shelf underneath. They were fastened down with wire, passing over the top of the carboys, and turnbuckles. In addition to securing the carboys, the wire held the tops in place under the operating pressure of 20 pounds per square inch. Through one of the holes in the stopper a piece of 3/8-inch copper tubing was introduced that extended to the bottom of the carboy. This section of tubing was joined to the extraction apparatus, and was used to transfer liquids. Through the other hole in the rubber stopper, a section of 3/8-inch copper tubing was inserted that barely extended through the stopper. This section of tubing was used as an air vent and an air pressure line.

Tubing. Before the copper tubing was used to connect the various parts of the extraction apparatus together, it was necessary to bring the water and air lines to the extractor site. Take-offs

from both the air and water lines were available near the extractor. A 3/8-inch water line was laid from the take-off point to the front right-hand edge of the table, divided, and closed by two gate valves. A 1/4-inch air line was laid from the take-off point to the front edge of the table just to the left of the water line. This line was connected to an air-reducing valve. Beyond this valve the line was split into four sections, and each section was closed with a globe valve. On the equipment side of these globe valves, each line was equipped with a pressure gauge and an air vent.

Copper tubing and brass fittings were used to connect the storage section to the pumps, the pumps to the extractor, the carboys to the drains, the column air vent to the column, and the water lines to the solvent carboys. This tubing was placed by standard methods and no particular arrangement was desired.

Analytical Procedure. The analytical procedure consisted of the determination of the specific gravity of the samples at 30° C, the determination of the specific gravity of samples at room temperature, and the determination of the refractive index of samples.

Determination of Specific Gravity. The specific gravity of various samples was determined with a Westphal balance. Standard techniques were used in these determinations (82). For the tests at a constant temperature of 30° C the balance

was located above a constant temperature water bath that had been adjusted to this temperature.

During testing for steady-state conditions the specific gravities of various samples were determined. An identical procedure was followed, except that no attempt was made to control the temperature for these determinations. However, since these tests were made in a relatively short time, no appreciable change in temperature should have been encountered.

Determination of Refractive Index. Samples, the refractive index of which was desired, were taken in culture tubes, sealed, and stored until such time as the refractometer was used. The refractive index of these samples was taken using standard techniques⁽⁸³⁾.

Determination of Refractive Index Data. There was no data available in the literature showing the relationship between the refractive index and the concentration of acetone in the respective phases for the system carbon tetrachloride-acetone-water. However, data were available for analyzing the saturated phases from this system by means of the specific gravity. To prepare refractive index data, samples of carbon tetrachloride, acetone, and water were mixed and analyzed by measuring the specific gravity.

Into 250-milliliter, glass-stoppered erlenmeyer flasks, approximately 100 grams of carbon tetrachloride and 100 grams of water were added. To these flasks, acetone was then added in

amounts varying from zero to 50 grams. The flasks were closed, shaken, and allowed to stand at room temperature. The flasks were shaken at frequent intervals for about 14 hours. After settling, the two phases were separated by means of a separatory funnel. The specific gravity at $30^{\circ}/4^{\circ}$ C and the refractive index were obtained for each sample by the procedure mentioned above.

Preparation of Column for Operation. The preparation of the extraction column for operation involved several steps. These steps consisted of the preparation of the feed, the adjustment of the pulse, and the starting and filling of the extractor. All references to equipment and valves in the discussion that follows will refer to Drawing 9, page 36.

Preparation of Feed. During the experimental tests, a feed consisting of approximately 10 weight per cent acetone in carbon tetrachloride was used. This composition was prepared by mixing, on the basis of weight, the required quantities of acetone and carbon tetrachloride. As the raffinate was continually reused and as there was no satisfactory method of analysis available at the extractor site, the feed composition actually varied widely from the desired value. The mixture of carbon tetrachloride in acetone, prepared in batches of five gallons, was placed in one of the five-gallon cans in which the carbon tetrachloride was purchased. A high-speed propeller-type mixer was used to

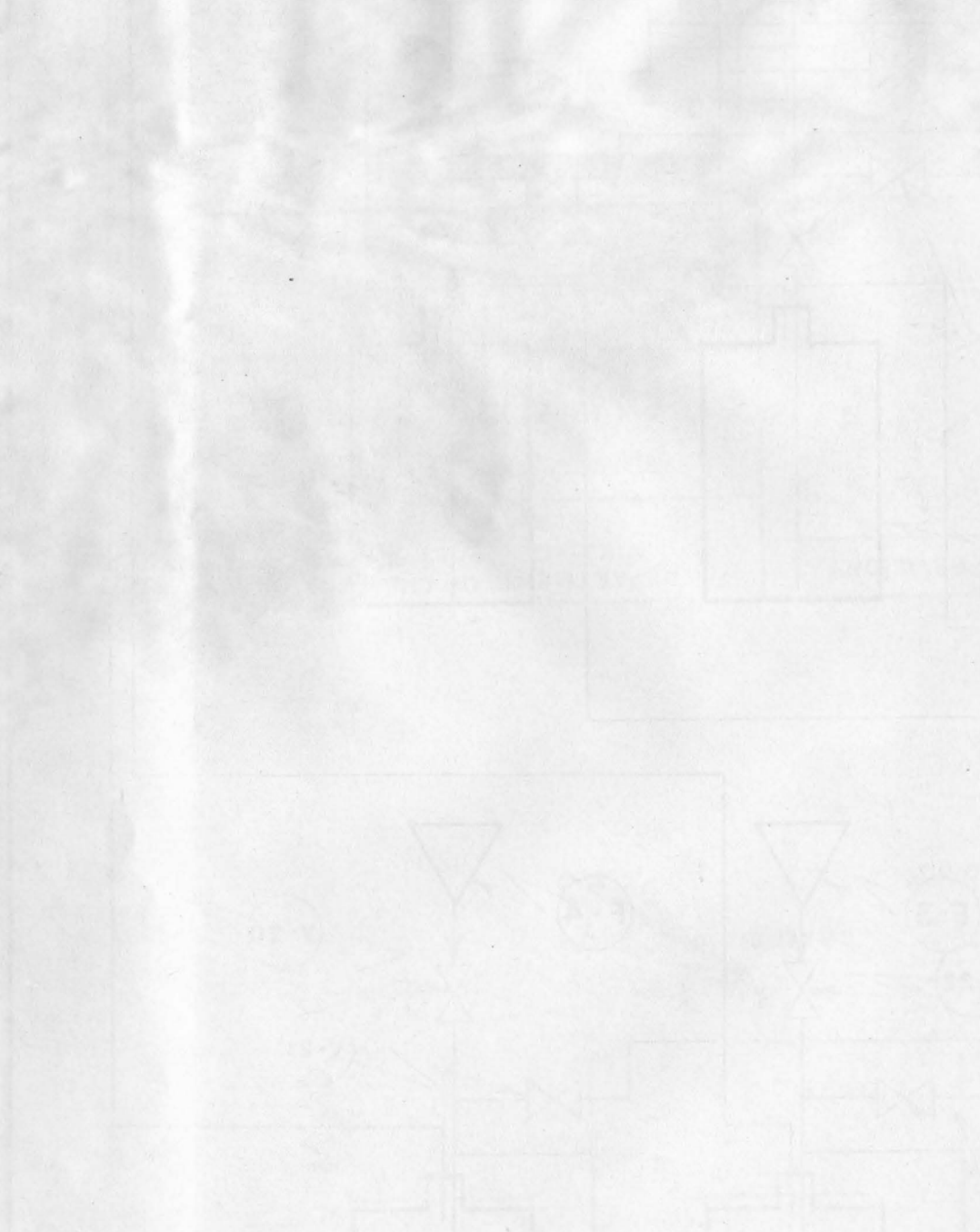
agitate the feed. The mixture was agitated in the presence of a slight excess of water for at least 20 minutes. Usually, two batches of feed were prepared at the same time.

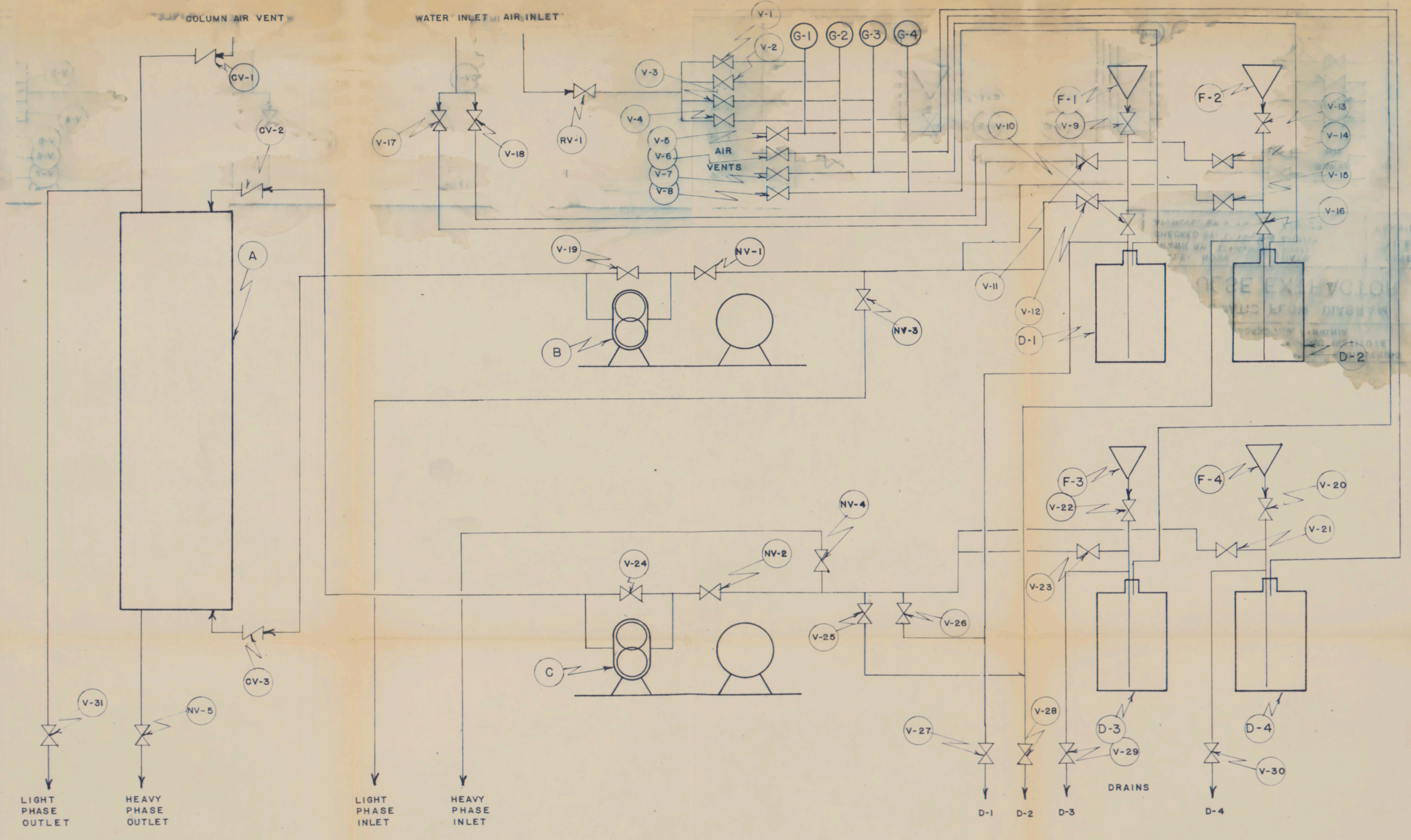
After mixing, the feed was placed in the feed carboys. Referring to Drawing 9, page 86, the feed carboys are D-3 and D-4. The other two carboys, D-1 and D-2, are for the light phase if some solvent other than water were to be used. Before filling the carboys, valves V-6 and V-22 are opened, and valves V-2, V-23, and V-29 are closed if carboy D-3 is to be filled. If D-4 is to be filled, the corresponding valves are V-5 and V-20, and V-1, V-20, and V-30. After filling, valve V-22 for D-3, and V-23 for D-4 are closed.

Adjustment of Pulse. The frequency of the pulse was adjusted by changing the pulleys on the cam-assembly motor and gear reducer. The motor turned at 1720 revolutions per minute and a fixed speed-reduction ratio of 30 to 1 was built into the gear reducer. Thus, any cam shaft speed could be obtained by selecting pulleys of the appropriate size. For this investigation, pulleys of two and four inches, and three and three inches were used, resulting in cam-shaft speeds of 10.5 and 21 revolutions per minute.

The amplitude of the pulse was adjusted by turning the push-rod face relative to the remainder of the push rod. Before adjusting, however, the column must be filled in order

Drawing 9. Schematic Flow Diagram for
Pulse Extractor





LEGEND

- A PULSE EXTRACTOR
- B LIGHT PHASE PUMP
- C HEAVY PHASE PUMP
- D-1 & D-2 LIGHT PHASE STORAGE
- D-3 & D-4 HEAVY PHASE STORAGE
- F-1 TO F-4 FUNNELS FOR FILLING STORAGE
- G-1 TO G-4 PRESSURE GAGES
- CV-1 TO CV-3 CHECK VALVES
- NV-1 TO NV-5 FLOW CONTROL (NEEDLE) VALVES
- RV-1 PRESSURE REDUCER
- V-1 TO V-31 VALVES

REVISIONS	DEPARTMENT OF CHEMICAL ENGINEERING VIRGINIA POLYTECHNIC INSTITUTE BLACKSBURG, VIRGINIA	
	SCHEMATIC FLOW DIAGRAM PULSE EXTRACTOR	
	SCALE: NONE	DATE: 4/11/53
	DRAWN BY: D. Hayford	FILE 599
	CHECKED BY: D. Hayford	4/11/53
	APPROVED BY: PHW	4/11/53
		DRAWING 9

to be able to measure the amplitude of the pulse. To fill the column, valves NV-1, NV-2, NV-4, NV-5, V-9, V-10, and V-11 were closed, or checked to see that they were closed, and valve V-31 was opened. After the cam assembly motor was started, valves V-17, V-10, V-12, and NV-1 were opened to allow water to fill the extractor. On the outside of the top disengaging section of the extractor, a paper scale was fastened to measure the movement of the liquid. As soon as the water reached the height of this scale it was stopped by closing valve NV-1. The amplitude was adjusted by turning the push-rod face. When the desired amplitude was obtained, the push-rod length was fixed by tightening the lock nut. After adjustment the column was drained by opening valve NV-5.

Starting Column Operation. After the adjustment of the pulse and preparation of the feed, the extractor was ready to start operation. The first step in putting the extractor into operation was to start the cam. Next, the water flow was started by opening valve NV-1. Assume that feed from carboy D-3 was to be used first. To cause the feed to flow into the column, valve V-2 was opened and valve V-6 was closed. These two valves were adjusted until the pressure in the system as indicated on gauge G-2 was equal to 20 pounds per square inch. Valves V-24 and V-23 were opened wide and the feed flow rate was adjusted by valve NV-2. Fairly accurate

preliminary adjustments of the two flow rates could be obtained by observing the rise of water in the extractor and the bubbles of feed flowing down through the top disengaging section. Any product removed before steady-state conditions were reached were, of course, collected. For this purpose galvanized pails were used.

Operation of Column. The column was now ready for operation. In this section the technique to be used during the operation, and the operations involved in changing from one feed carboy to another feed carboy, will be covered.

Operational Technique. The flow rates were checked by collecting the proper phase output for a definite period of time and weighing the amount collected. Any adjustments in the heavy-phase flow rate were made by adjusting valve V-2. Any adjustments in the light-phase flow rate were made with valve V-1. The interface in the top disengaging section was fixed by the conditions of operation. However, the interface in the lower disengaging section must be maintained at a constant level by adjusting valve NV-5. The heavy-phase flow rate was found to be extremely sensitive to variations in the air pressure. Both flow rates are, of course, affected by any appreciable rate of change of the interface. The operation of the extractor was very simple. Very little attention to the valves, once steady-state conditions had been reached, were found to be necessary.

Changing Feed Carboys. When the contents of one feed carboy were nearly exhausted it was necessary to change to the other feed carboy. For this illustration it will be assumed that feed from D-3 was being used, and that it was desired to change to D-4. Valves V-20, V-21, and V-30 were closed. Valves V-1 and V-5 were opened and closed respectively until the air pressure as indicated on gauge G-1 was 20 pounds per square inch. After the air pressure was adjusted satisfactorily, valve V-21 was opened. After a moment valve V-23 was closed. The extractor was now receiving feed from carboy D-4. Adjustment in the feed flow rate was still made by valve NV-2. Carboy D-3 was then drained by opening valve V-29, released by closing valve V-2, and vented by opening valve V-6.

Experimental Tests. Three important aspects of the experimental testing procedure will be covered in this section. These three aspects are the determination of steady-state conditions, the procedure followed in the efficiency tests, and the procedure followed in the limiting flow rate tests.

Steady-state Conditions. For the purpose of this investigation "steady-state conditions" was defined as that condition obtained within the extractor, when for three successive five-minute readings, commencing not before two complete liquid changes in the extractor, there was no change in

either flow rate or in the density of (recorded to the third place after the decimal) the raffinate. An alternate criteria, that was also used, was five successive readings with no change in either flow rate and with a change in the specific gravity of the raffinate of no more than one in the third place after the decimal. After the flow rates were adjusted to the desired values, samples of the raffinate were taken every five minutes for a specific gravity determination. The flow rates were also measured at the same time. As soon as the test for steady-state conditions was passed, the testing period was started.

Efficiency Testing Procedure. During the efficiency testing period, adjustments of the interface only were allowed. For a period of fifteen minutes (this time was later changed to ten minutes for some of the tests with a proportional decrease in the other time intervals) the following operations were performed: Collection of the extract, collection of the raffinate, and a sample of the feed every five minutes. The feed sample was taken by opening valve NV-4 and collecting the liquid in a culture tube. After completion of the test another sample of the feed was taken, and the collected raffinate and extract were weighed. After weighing, two samples of each phase were taken. Any remarks of unusual features occurring during the operation of the extractor were, of course, recorded.

Flow Rate Testing Procedure. Tests to determine the limiting flow rates of the extractor were performed by a procedure very similar to that described above. In the limiting flow rate testing, however, there was no testing for steady-state conditions. As soon as the flow rates were adjusted visually and the interface showed a tendency to remain constant, the test period was started. These tests were of varied length, ranging from one minute at the high flow rates to five minutes at the lower flow rates. During the limiting flow rate tests, the only data recorded, besides the amplitude and frequency of the pulse, were the flow rates of both phases.

Completion of Tests. As soon as a series of tests were completed, the column was drained and cleaned. To stop the liquids flowing into the extractor, valves NV-1 and NV-2 were closed. The cam motor was kept running until all the heavy phase had worked down into the bottom of the column. After this, the cam was stopped. The extractor was completely drained through NV-5. After draining, valve NV-1 was opened wide and water was allowed to flow through the extractor. This water rate was sufficient to fill the extractor and to drain through the extractor overflow outlet even though valve NV-5 was left open. As soon as the water leaving the column was clear, valve NV-1 was closed and the column again drained through valve NV-5. Feed carboys were emptied, released, and vented as described previously.

Data and Results

The data and results of this investigation are presented in the following sections.

Refractive-index Data. The refractive-index and distribution data obtained during the experimental portion of this investigation are presented in Table VI, page 93. In Figure 3, page 94, the relation between the concentration of acetone in and the specific gravity of the saturated phases is shown. In Figure 4, page 95, the relation between the concentration of acetone in and the refractive index of the saturated phases is shown. In Figure 5, page 96, the experimentally determined distribution data are graphically presented. These data are similar to and should be compared to the data presented in Figure 2, page 21.

Pulse-Extractor Operating Data. In Table VII, page 97, the operational data for the pulse extractor are presented.

Material Balance. As an aid in the evaluation of the accuracy of the experimental technique, a summary of the acetone material balances is shown in Table VIII, page 98. The average value of the discrepancy for all of these tests was 4.3 per cent.

Stage Efficiencies. The stage efficiencies for the various experimental tests are presented in Table IX, page 99. Included in this table, also, are the operational parameters that were studied. These parameters are the pulse number (defined as the

TABLE VI

Distribution and Refractive-index Data for the SystemCarbon Tetrachloride-Acetone-Water at 22° C

Carbon tetrachloride phase			Water phase		
Acetone concentration	Specific gravity	Refractive index	Acetone concentration	Specific gravity	Refractive index
wt %	30°/4° C	n_D^{20}	wt %	30°/4° C	n_D^{20}
0.00 ^a	1.5768	1.4628	0.00 ^a	0.9946	1.3320
0.00 ^b	1.5764	1.4594	0.00 ^b	0.9952	1.3330
0.49	1.5698	1.4588	2.87	0.9913	1.3341
1.73	1.5513	1.4568	5.98	0.9872	1.3365
2.38	1.5418	1.4558	8.14	0.9844	1.3376
5.53	1.4947	1.4503	14.83	0.9758	1.3419
8.68	1.4480	1.4446	17.79	0.9709	1.3449
12.81	1.3966	1.4377	21.93	0.9643	1.3479
17.39	1.3419	1.4302	26.88	0.9566	1.3508

a. Analysis of original component.

b. Analysis of original component saturated with other phase.

Figure 3. Specific Gravity of Equilibrium
Solutions for the System Carbon
Tetrachloride-Acetone-Water

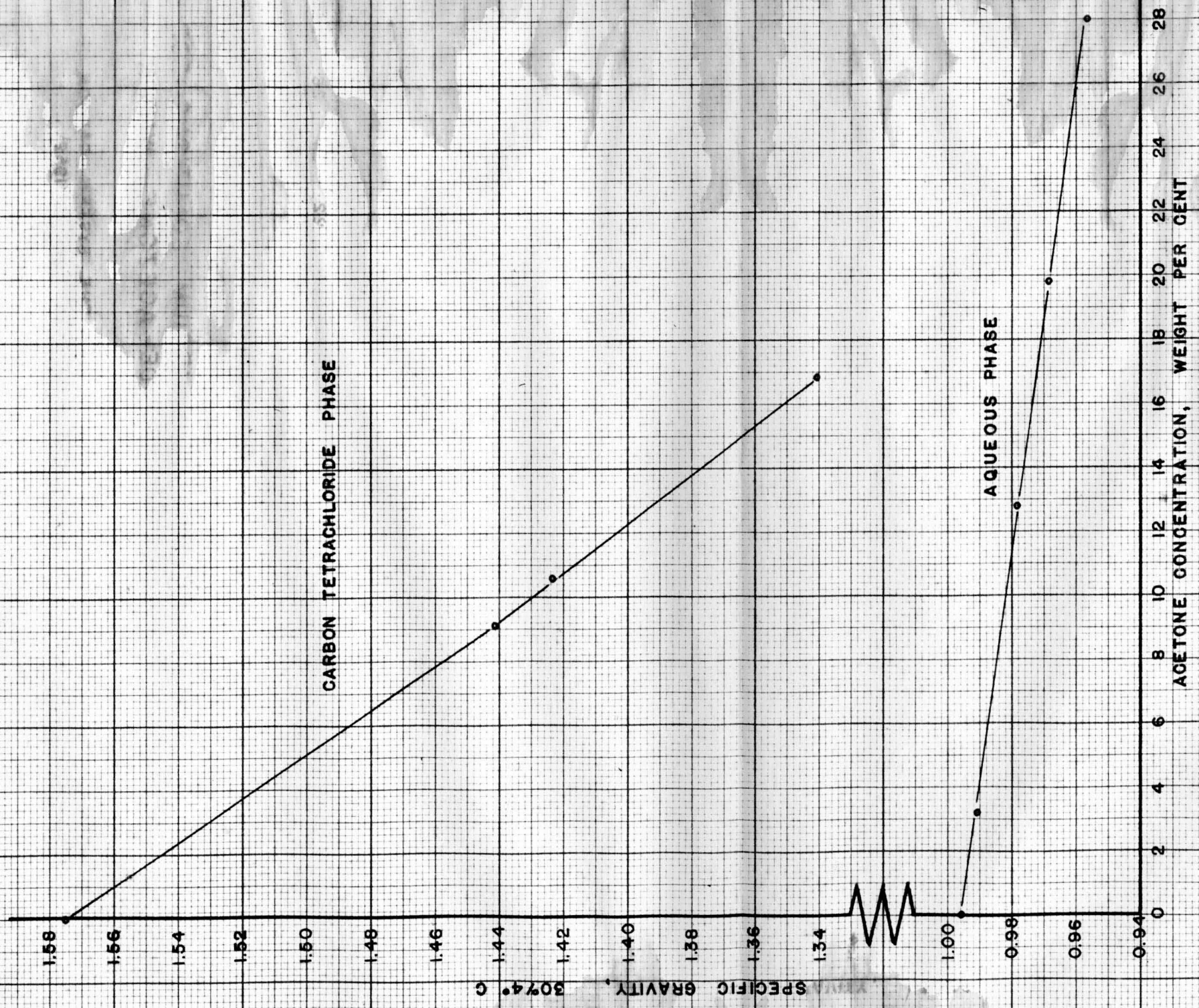


FIGURE 3. SPECIFIC GRAVITY OF EQUILIBRIUM SOLUTIONS FOR THE SYSTEM CARBON TETRACHLORIDE - ACETONE - WATER

BUCHANAN, R. H.: LIQUID-LIQUID EQUILIBRIUM DATA FOR THE SYSTEM CARBON TETRACHLORIDE - ACETONE - WATER, IND. ENG. CHEM., 44, 2449 (1952).

Figure 4. Refractive Index of Equilibrium Solutions for
the System Carbon Tetrachloride-Acetone-Water

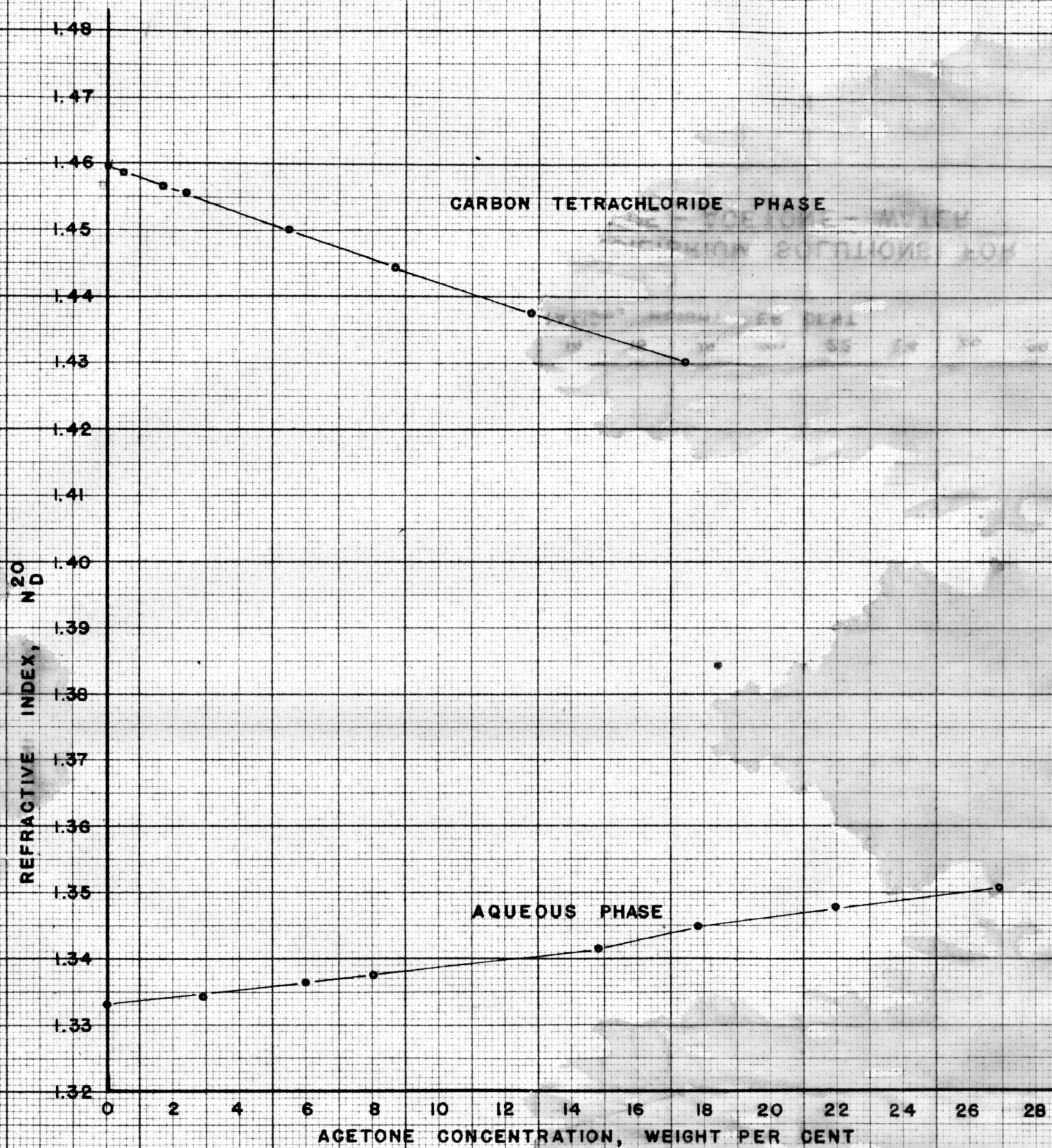


FIGURE 4. REFRACTIVE INDEX OF EQUILIBRIUM SOLUTIONS FOR THE SYSTEM CARBON TETRACHLORIDE - ACETONE - WATER

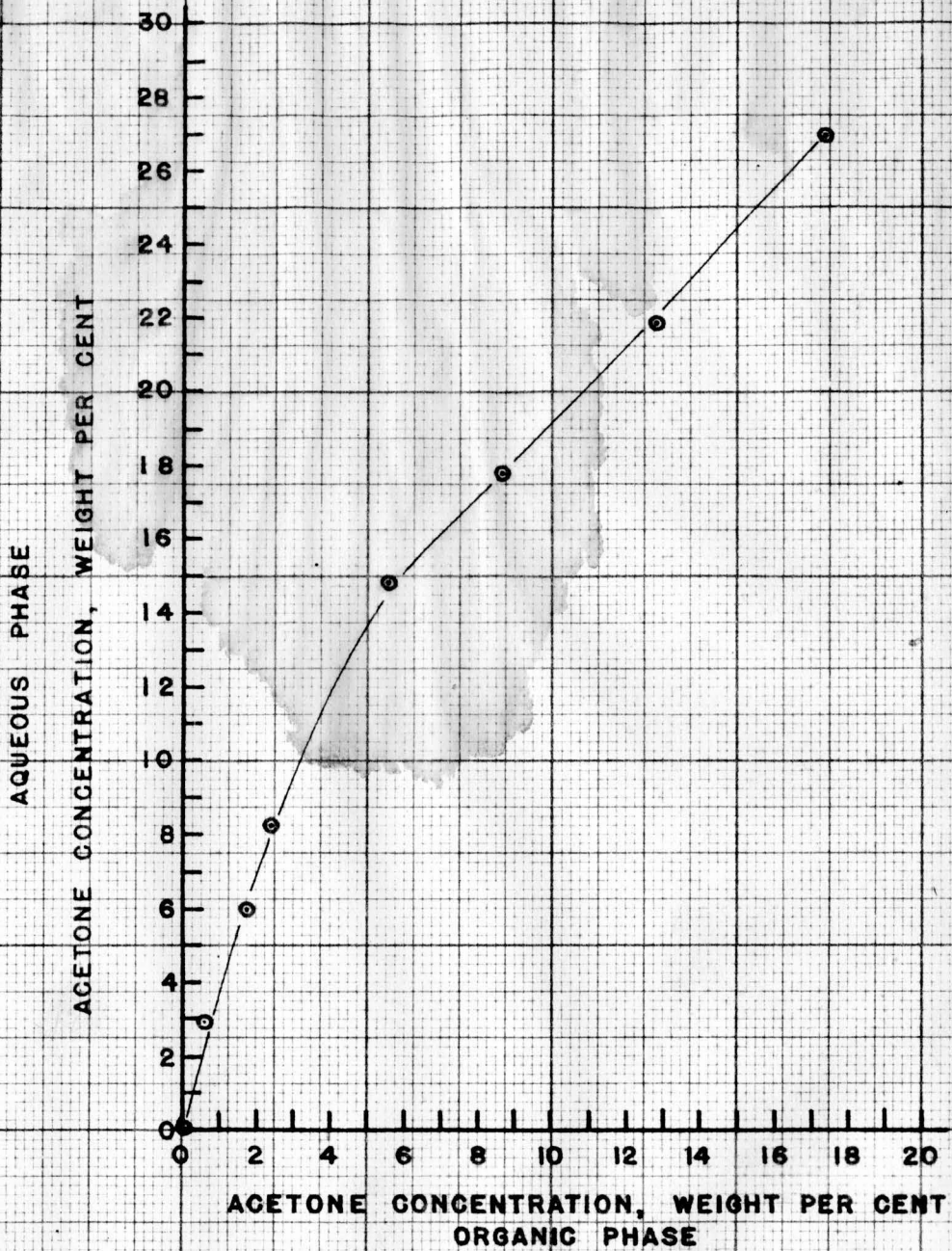


FIGURE 5. DISTRIBUTION OF ACETONE BETWEEN CARBON TETRACHLORIDE AND WATER AT 22° C

TABLE VII

Operational Data for a Two-inch Experimental Pulse
Extraction Column Extracting Acetone from Carbon
Tetrachloride with Water

(The table content is extremely faint and illegible due to the quality of the scan. It appears to be a multi-column table with approximately 8 columns and 10 rows of data.)

TABLE VII

Operational Data for a Two-inch Experimental Pulse Extraction Column
Extracting Acetone from Carbon Tetrachloride with Water

Test No	Pulse amplitude ft	Pulse frequency min ⁻¹	Phase	Flow rates		Refractive index n _D ²⁰	Acetone concentration wt %
				gm/10 min	lb hr-ft ²		
1	0.083	10.5	Raffinate	1092 ^c	442	1.4588	0.49
			Extract	3286 ^c	1332	1.3407	12.62
			Feed	—	610	1.4196	27.84 ^a
			Solvent	—	1164	—	0.00
2	0.133	10.5	Raffinate	976 ^c	395	1.4588	0.49
			Extract	3437 ^c	1390	1.3343	3.16
			Feed	—	441	1.4411	10.78
			Solvent	—	1346	—	0.00
3	0.167	10.5	Raffinate	1204 ^c	487	1.4592	0.18
			Extract	3324 ^c	1346	— ^b	4.33 ^b
			Feed	—	545	1.4410	10.81
			Solvent	—	1288	—	0.00
4	0.167	10.5	Raffinate	1098	676	1.4567	1.81
			Extract	2308	1400	1.3372	7.31
			Feed	—	772	1.4357	14.08
			Solvent	—	1298	—	0.00
5	0.167	10.5	Raffinate	5538	2240	1.4528	4.48
			Extract	3054	1236	1.3452	18.22
			Feed	—	2451	1.4378	12.78
			Solvent	—	1011	—	0.00
6	0.167	10.5	Raffinate	4444	2570	1.4552	2.80
			Extract	2274	1380	1.3412	13.33
			Feed	—	2729	1.4449	8.49
			Solvent	—	1196	—	0.00
7	0.167	10.5	Raffinate	1828	1110	1.4578	1.10
			Extract	5884 ^d	7140	1.3339	2.30
			Feed	—	1271	1.4363	13.70
			Solvent	—	6976	—	0.00
8	0.167	10.5	Raffinate	2523	1530	1.4568	1.74
			Extract	3300	2000	1.3368	6.49
			Feed	—	1658	1.4436	9.29
			Solvent	—	1870	—	0.00
9	0.167	10.5	Raffinate	8306	5040	1.4577	1.18
			Extract	7013 ^d	8520	1.3370	6.96
			Feed	—	5602	1.4405	11.10
			Solvent	—	7926	—	0.00
10	0.233	10.5	Raffinate	1332	839	1.4589	0.40
			Extract	2564	1560	1.3348	3.78
			Feed	—	899	1.4481	6.96
			Solvent	—	1501	—	0.00
11	0.233	10.5	Raffinate	899	545	1.4589	0.40
			Extract	4043	2450	1.3347	3.63
			Feed	—	628	1.4363	13.70
			Solvent	—	2361	—	0.00
12	0.233	10.5	Raffinate	1096	664	1.4593	0.12
			Extract	5408	3286	1.3346	3.50
			Feed	—	1191	1.4477	14.20
			Solvent	—	3171	—	0.00
13	0.250	10.5	Raffinate	1848	1122	1.4588	0.49
			Extract	2523	1533	1.3352	4.29
			Feed	—	1191	1.4477	6.26
			Solvent	—	1487	—	0.00
14	0.075	21.0	Raffinate	867	526	1.4587	0.53
			Extract	2601	1568	1.3348	3.78
			Feed	—	584	1.4417	10.42
			Solvent	—	1509	—	0.00
15	0.121	21.0	Raffinate	1411	855	1.4585	0.66
			Extract	2969	1803	1.3363	5.70
			Feed	—	953	1.4409	10.86
			Solvent	—	1700	—	0.00

- a. Calculated from the material balance. This value tends to agree with the value of the refractive index for this sample that is above the limit of available calibration data.
- b. Calculated from the material balance. The original samples contaminated before the value of the refractive index was determined.
- c. Flow rates expressed in grams per 15 minutes.
- d. Flow rates expressed in grams per five minutes.

TABLE VIII

Summary of Acetone Material Balances for a Two-inch
Experimental Pulse Extractor Extracting Acetone
from Carbon Tetrachloride with Water

Test No	Acetone out	Acetone in	Discrepancy	Discrepancy
	lb/hr-sq ft	lb/hr-sq ft	lb/hr-sq ft	per cent ^a
1	170.2	170.2	— ^b	— ^b
2	45.9	47.5	1.6	3.4
3	59.2	59.2	— ^b	— ^b
4	114.6	108.4	6.2	5.6
5	327.2	312.6	14.6	4.6
6	255.8	231.3	24.5	10.2
7	176.4	173.3	3.1	1.8
8	156.2	154.7	1.5	1.0
9	653.6	621.0	32.6	5.1
10	62.4	62.5	0.1	0.2
11	91.6	85.8	5.8	6.7
12	116.0	108.8	7.2	6.4
13	71.3	74.6	3.3	4.5
14	62.1	60.8	1.3	2.1
15	108.7	103.9	4.8	4.5

a. Per cent of mean of acetone in and acetone out.

b. Assumed equal in calculations.

TABLE IX

Stage Efficiency for a Two-inch Experimental Pulse Extraction
Column Extracting Acetone from Carbon Tetrachloride
with Water.

Test No	Pulse number ^a	Flow rates		Acetone Relative ^b in feed phase	Acetone wt %	Stage efficiency per cent
		Heavy phase	Light phase			
	ft/min	lb/hr-sq ft	ft			
1	0.92	440	1164	0.33	27.84	23.8
2	1.42	393	1346	0.28	10.78	18.8
3	1.75	486	1288	0.36	10.81	23.8
4	1.75	664	1298	0.48	14.08	16.3
5	1.75	2138	1011	1.82	12.78	25.0
6	1.75	2498	1196	1.86	8.49	23.8
7	1.75	1098	6976	0.16	13.70	12.5
8	1.75	1503	1870	0.77	9.29	15.0
9	1.75	4981	7926	0.59	11.10	18.8
10	2.42	836	1501	0.54	6.96	21.3
11	2.42	542	2361	0.22	13.70	17.5
12	2.42	663	3171	0.20	14.20	23.8
13	2.67	1116	1467	0.73	6.26	21.3
14	1.58	523	1509	0.34	10.42	20.0
15	2.50	849	1700	0.47	10.86	21.3

- a. The pulse number is defined as the product of the amplitude and frequency of the pulse.
 b. The relative flow rate is equal to the raffinate-phase flow rate divided by the extract-phase flow rate.

UNIVERSITY OF CALIFORNIA
 LIBRARY
 1955

product of the pulse amplitude and the pulse frequency), the carrier solvent flow rate, the solvent flow rate, the relative (raffinate to extract) flow rate, and the concentration of acetone in the feed. An equation was proposed to explain the relation between these parameters and the stage efficiency. The constants in this equation were evaluated by a statistical analysis, the results of which are shown in Table X, page 101.

The equation as calculated for this investigation is presented below:

$$y = 12.44 + 3.70x_1 + 0.00503x_2 - 0.00302x_3 + 0.371x_4 - 6.93x_5$$

where:

- y = stage efficiency, per cent
- x₁ = pulse number, feet per minute
- x₂ = carrier solvent flow rate, pounds per hour-square foot
- x₃ = solvent flow rate, pounds per hour-square foot
- x₄ = concentration of acetone in feed, weight per cent
- x₅ = relative flow rate, dimensionless.

Using this equation and the experimentally determined values of x₁, x₂, x₃, x₄, and x₅ the value of y was calculated for each test. This calculated value of y is compared to the experimentally determined value of y in Table XI and in Figure 6, pages 102 and 103, respectively.

Limiting Liquid Flow Rates. The limiting liquid flow rates for the pulse extractor are shown in Table XII, page 104. In Figure 7, page 105, the limiting liquid flow rates are shown graphically for a pulse frequency of 10.5 strokes per minute and an amplitude of 0.25

TABLE X
Results of the Analysis of the Two-Block Experimental
Pulse-Extension Efficiency Tests

Variable	Units	Range of variable covered	Regression coefficients	Significance level	per cent
x ₁ Pulse number ^b	ft/min	0.92 to 2.67	3.70	4.7	4.7
x ₂ Heavy-phase flow rate	lb/hr-sq ft	393 to 4981	0.00503	0.4	0.4
x ₃ Light-phase flow rate	lb/hr-sq ft	1011 to 7926	-0.00302	0.1	0.1
x ₄ Solute concentration	wt %	6.26 to 27.34	0.371	4.2	4.2
x ₅ Relative flow rate ^c		0.16 to 1.86	-6.93	0.8	0.8
y Stage efficiency	per cent	12.5 to 25.0	---	---	---

a. Constants in the equation $y = 12.44 + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4 + b_5x_5$.
 b. Defined as the product of the pulse amplitude and the pulse frequency.
 c. Raffinate-phase flow rate to the extract-phase flow rate.

TABLE XI

Comparison of Actual and Calculated Stage Efficiencies
for a Two-inch Experimental Pulse Extractor

Test No	Efficiency calculated per cent	Efficiency observed per cent	Discrepancy per cent
1	22.6	23.8	5.0
2	17.7	18.8	5.8
3	19.0	23.8	20.2
4	18.8	16.3	15.3
5	22.2	25.0	11.2
6	20.1	23.8	15.5
7	12.3	12.5	1.6
8	18.9	15.0	26.0
9	19.9	18.8	5.9
10	20.0	21.3	6.1
11	19.0	17.5	8.6
12	20.5	23.8	13.9
13	20.8	21.3	2.4
14	18.8	20.0	6.0
15	21.6	21.3	1.4

REGRESSION EQUATION:

$$Y = 12.44 + 3.70X_1 + 0.00503X_2 - 0.00302X_3 + 0.371X_4 - 6.93X_5$$

WHERE:

- Y = STAGE EFFICIENCY, PER CENT
- X₁ = PULSE NUMBER (PRODUCT OF FREQUENCY AND AMPLITUDE), FEET PER MINUTE
- X₂ = CARBON TETRACHLORIDE FLOW RATE, LB/HR-SQ FT
- X₃ = WATER FLOW RATE, LB/HR-SQ FT
- X₄ = ACETONE CONCENTRATION IN FEED, WT PER CENT
- X₅ = RELATIVE (RAFFINATE / EXTRACT) FLOW RATE.

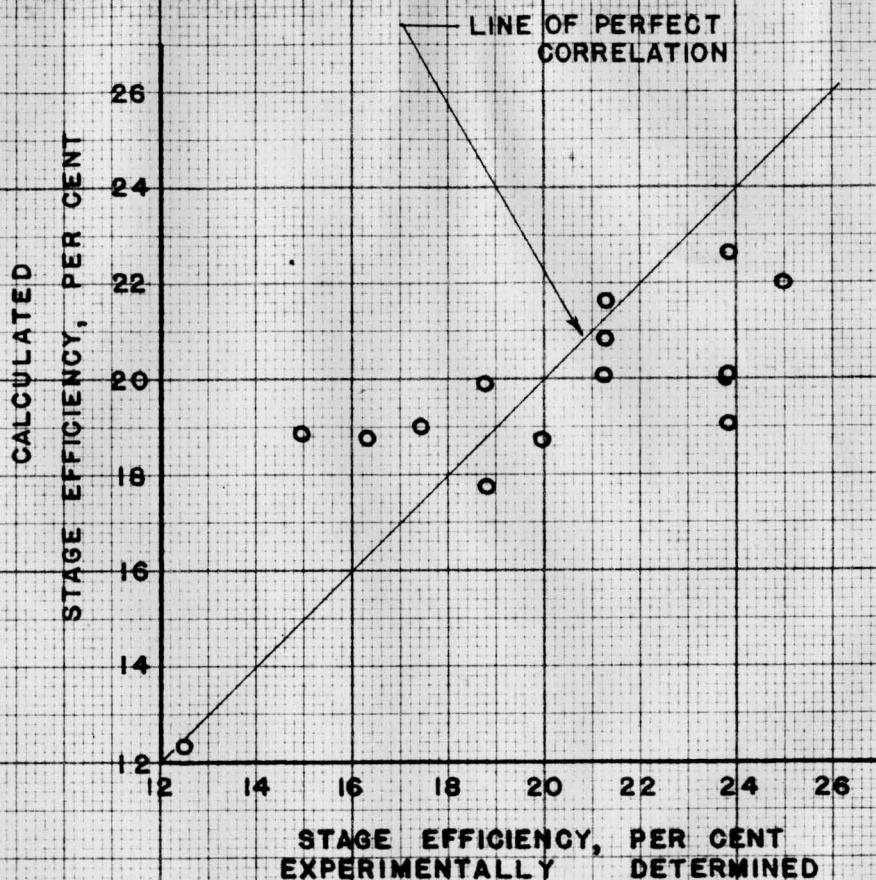


FIGURE 6. COMPARISON OF EXPERIMENTALLY DETERMINED AND CALCULATED STAGE EFFICIENCIES

TABLE XII

Limiting Liquid Flow Rates in an Experimental
Two-inch Pulse Extractor Using the System
of Carbon Tetrachloride and Water

TABLE XII

Limiting Liquid Flow Rates in an Experimental Two-inch Pulse Extractor
Using the System of Carbon Tetrachloride and Water

Test No	Pulse amplitude ft	Pulse frequency min ⁻¹	Heavy-phase flow rate lb/hr-sq ft	Light-phase flow rate lb/hr-sq ft	Limiting condition
1	0.25	10.5	2,010	920	Dumping
2	0.25	10.5	4,730	1,860	Dumping
3	0.25	10.5	5,860	2,330	Dumping
4	0.25	10.5	6,820	3,210	Dumping
5	0.25	10.5	9,370	3,950	Dumping
6	0.25	10.5	9,980	4,880	Dumping
7	0.25	10.5	11,800	5,580	Dumping
8	0.25	10.5	13,020	6,170	Dumping
9	0.25	10.5	15,580	6,940	Dumping
10	0.25	10.5	16,000	7,960	Dumping
11	0.25	10.5	18,600	9,010	Dumping
12	0.25	10.5	19,720	9,980	Dumping
13	0.25	10.5	20,380	10,310	Heavy-phase flooding
14	0.25	10.5	19,870	11,070	Heavy-phase flooding
15	0.25	10.5	20,060	12,100	Heavy-phase flooding
16	0.25	10.5	19,980	12,890	Heavy-phase flooding
17	0.25	10.5	19,400	14,020	Heavy-phase flooding
18	0.25	10.5	19,360	16,210	Heavy-phase flooding
19	0.25	10.5	18,530	17,960	Heavy-phase flooding
20	0.25	10.5	17,610	21,640	Both phases flooding
21	0.25	10.5	189	920	Blowing
22	0.25	10.5	215	1,860	Blowing
23	0.25	10.5	237	2,330	Blowing
24	0.25	10.5	263	3,950	Blowing
25	0.25	10.5	297	5,580	Blowing
26	0.25	10.5	316	6,940	Blowing
27	0.25	10.5	480	10,310	Blowing
28	0.25	10.5	610	12,890	Blowing
29	0.25	10.5	890	16,210	Blowing
30	0.25	10.5	1,020	21,640	Light-phase flooding
31	0.125	10.5	4,090	1,720	Dumping
32	0.125	10.5	6,010	2,830	Dumping
33	0.125	10.5	8,730	4,010	Dumping
34	0.125	10.5	10,680	5,270	Dumping
35	0.125	10.5	12,990	6,460	Dumping
36	0.125	10.5	14,500	7,800	Dumping
37	0.125	10.5	15,860	8,470	Heavy-phase flooding
38	0.125	10.5	15,470	9,880	Heavy-phase flooding
39	0.125	10.5	15,310	13,020	Heavy-phase flooding
40	0.125	10.5	15,930	16,890	Heavy-phase flooding
41	0.125	10.5	15,460	22,870	Both phases flooding
42	0.125	10.5	163	1,720	Blowing
43	0.125	10.5	179	4,010	Blowing
44	0.125	10.5	303	6,460	Blowing
45	0.125	10.5	337	8,470	Blowing
46	0.125	10.5	680	13,020	Blowing
47	0.125	10.5	1,060	22,870	Light-phase flooding
48	0.125	21.0	4,320	1,060	Dumping
49	0.125	21.0	7,150	1,980	Dumping
50	0.125	21.0	9,850	3,080	Dumping
51	0.125	21.0	12,220	4,230	Dumping
52	0.125	21.0	16,080	5,370	Dumping
53	0.125	21.0	19,000	7,100	Dumping
54	0.125	21.0	21,070	9,120	Heavy-phase flooding
55	0.125	21.0	20,830	11,360	Heavy-phase flooding
56	0.125	21.0	19,180	15,110	Heavy-phase flooding
57	0.125	21.0	17,870	18,240	Heavy-phase flooding
58	0.125	21.0	15,730	20,860	Both phases flooding
59	0.125	21.0	356	1,060	Blowing
60	0.125	21.0	473	1,980	Blowing
61	0.125	21.0	507	3,080	Blowing
62	0.125	21.0	670	4,230	Blowing
63	0.125	21.0	660	5,370	Blowing
64	0.125	21.0	820	7,100	Blowing
65	0.125	21.0	1,160	9,120	Blowing
66	0.125	21.0	1,630	11,360	Blowing
67	0.125	21.0	2,160	15,110	Blowing
68	0.125	21.0	2,370	18,240	Blowing
69	0.125	21.0	2,220	20,860	Light-phase flooding

HEAVY PHASE: CARBON TETRACHLORIDE
LIGHT PHASE: WATER
PULSE FREQUENCY: 10.5 STROKES PER MINUTE
PULSE AMPLITUDE: 0.25 FEET
PLATES: EIGHT SPACED AT FOUR-INCH INTERVALS,
DRILLED 3/32-INCH HOLES (116) TO 2.59 PER
CENT FREE AREA

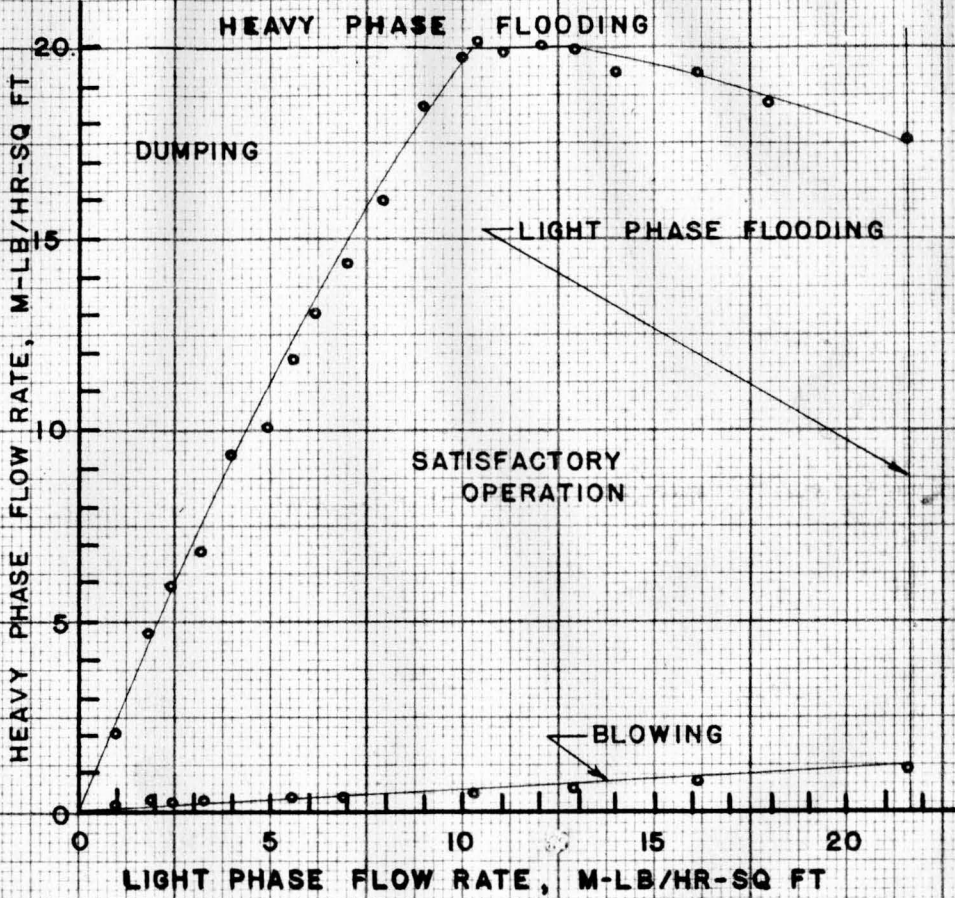


FIGURE 7. LIMITING LIQUID FLOW RATES IN
A TWO-INCH PULSE EXTRACTOR

feet per stroke. In Figures 8 and 9, pages 107 and 108, similar data are shown except that the pulse frequency is 10.5 and 21 strokes per minute, respectively. The pulse amplitude for both of these figures is 0.125 feet per stroke.

HEAVY PHASE: CARBON TETRACHLORIDE
LIGHT PHASE: WATER
PULSE FREQUENCY: 10.5 STROKES PER MINUTE
PULSE AMPLITUDE: 0.125 FEET
PLATES: EIGHT SPACED AT FOUR-INCH INTERVALS,
DRILLED 3/32-INCH HOLES (116) TO 2.59 PER
CENT FREE AREA

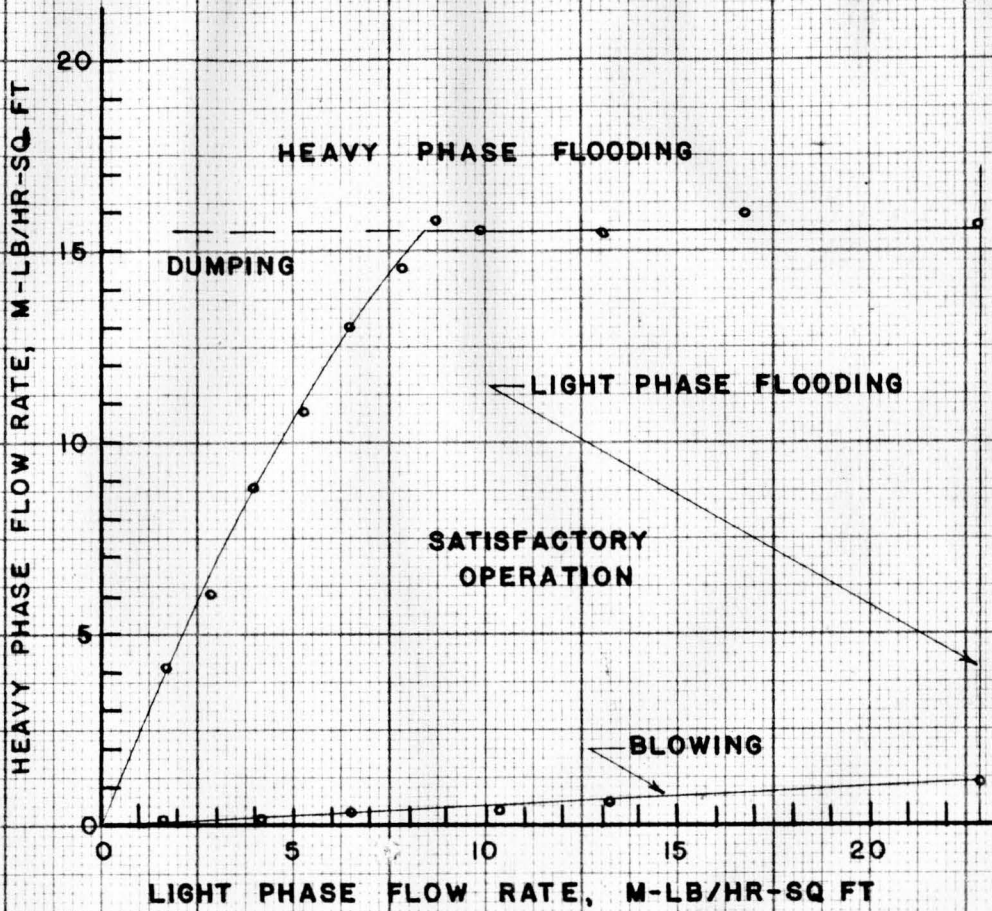


FIGURE 8. LIMITING LIQUID FLOW RATES IN
A TWO-INCH PULSE EXTRACTOR

HEAVY PHASE: CARBON TETRACHLORIDE
LIGHT PHASE: WATER
PULSE FREQUENCY: 21.0 STROKES PER MINUTE
PULSE AMPLITUDE: 0.125 FEET
PLATES: EIGHT AT FOUR-INCH INTERVALS, DRILLED
3/32-INCH HOLES (116) TO 2.59 PER CENT FREE AREA

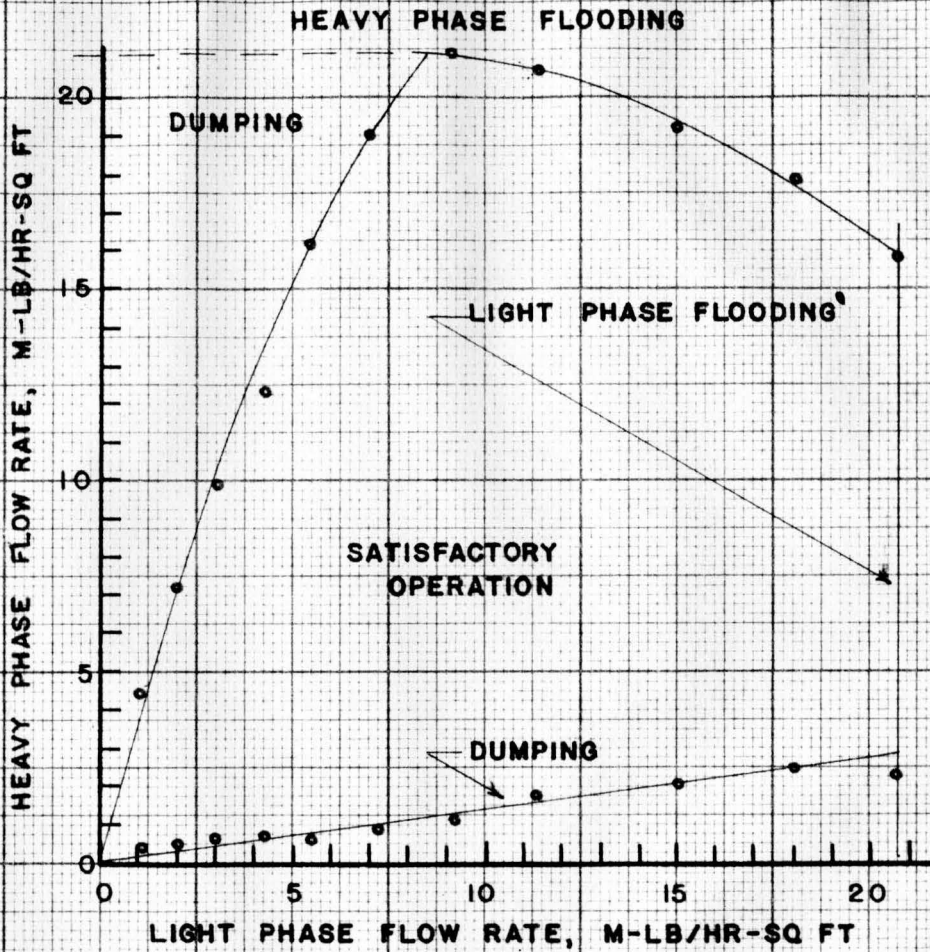


FIGURE 9. LIMITING LIQUID FLOW RATES IN A TWO-INCH PULSE EXTRACTOR

Sample Calculations

In this section examples of the calculations used in this investigation are presented.

Acetone Material Balance. As a check on the experimental accuracy, an acetone material balance was calculated for each test. The summary of these balances are contained in Table VIII, page 96. This calculation is illustrated using data from the second experimental test given in Table VII, page 97.

The acetone leaving the extractor or the "acetone out" was calculated as shown below:

$$A_0 = R x_1 + E y_2$$

where:

- A_0 = acetone leaving the extractor, pounds per hour-square foot
- R = raffinate phase flow rate, pounds per hour-square foot
- E = extract phase flow rate, pounds per hour-square foot
- x_1 = concentration of acetone in the raffinate phase, weight fraction
- y_2 = concentration of acetone in extract phase, weight fraction

$$A_0 = (395 \times 0.0049) + (1390 \times 0.0316)$$

$$A_0 = 45.9 \text{ lb/hr-sq ft.}$$

The acetone entering the extractor or the "acetone in" was calculated as shown below:

$$A_1 = \frac{x_2}{1-x_2} R(1-x_1)$$

where:

- A_1 = acetone entering the extractor, pounds per hour-square foot
- R = raffinate phase flow rate, pounds per hour-square foot
- x_1 = concentration of acetone in raffinate phase, weight fraction
- x_2 = concentration of acetone in feed, weight fraction

$$A_1 = \frac{0.1078}{1-0.1078} 395 (1-0.049)$$

$$A_1 = 47.5 \text{ lb/hr-sq ft.}$$

The discrepancy is, of course, the absolute difference between the acetone in and the acetone out. The discrepancy is more often expressed as a per cent of the mean of the acetone in and the acetone out. This has been done in Table VIII, page 96.

Calculation of the Stage Efficiency. Before the stage efficiency was calculated the number of theoretical stages had to be calculated. The number of theoretical stages was calculated by the graphical method introduced by Hunter and Nash^(33, 34). This method is more familiar in the form presented by Elgin⁽²³⁾ and Bull and Coli⁽¹¹⁾. A slight modification was made by the author. This modification consisted of the substitution of a distribution diagram for the tie lines used by Hunter and Nash^(33, 34) and the conjugate line used by Elgin⁽²³⁾ and Bull and Coli⁽¹¹⁾. As both the tie lines

and the conjugate line are obtained from the distribution diagram no real change in theory has been made.

The graphical solution for the number of theoretical stages for the second experimental test presented in Table VII, page 97, is shown in Figure 10, page 112. The distribution data, that is also used, is shown in Figure 5, page 96.

The stage efficiency was calculated as shown below:

$$y = \frac{N_s}{N_p} \times 100$$

where:

- y = stage efficiency, per cent
- N_s = number of theoretical stages, dimensionless
- N_p = number of actual stages, dimensionless

$$y = \frac{1.5}{8} \times 100$$

$$y = 18.8 \text{ per cent.}$$

Analysis of Stage Efficiency. The results presented in Table IX, page 99, showing the variation of the stage efficiency with the operating parameters of the pulse extractor, are too interrelated to show any significant trends. This lack of a definite trend was caused by the masking effect of all five operating parameters varying at once. To try to eliminate the masking effect of these variations, a statistical analysis of the data was used. This analysis consisted of proposing an equation expressing the relation between stage efficiency and the column parameters, determining the constants in this equation by the method of least squares, and

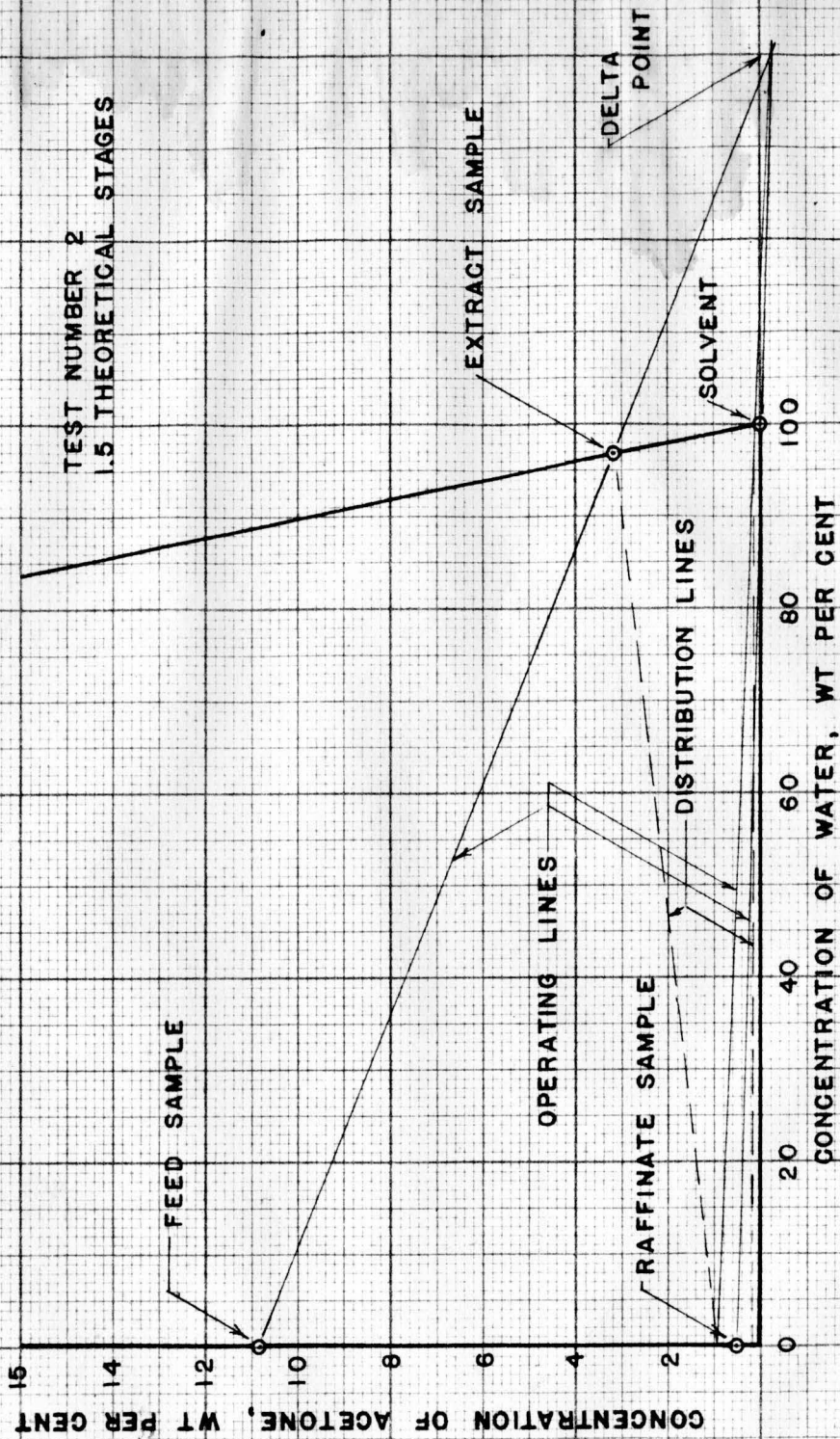


FIGURE 10. GRAPHICAL SOLUTION FOR THE NUMBER OF THEORETICAL STAGES

determining if these constants differ significantly from zero (zero would indicate that the variable has no effect on the stage efficiency).

The proposed multiple regression equation was of the following form:

$$y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4 + b_5x_5$$

where:

- y = stage efficiency, per cent
- x₁ = pulse number (product of the pulse amplitude and pulse frequency), feet per minute
- x₂ = carrier solvent flow rate, pounds per hour-square foot
- x₃ = solvent flow rate, pounds per hour-square foot
- x₄ = concentration of acetone in feed, weight per cent
- x₅ = relative (raffinate to extract) flow rate, dimensionless
- b₀ = y intercept, per cent
- b_i's = regression coefficients.

To perform the desired analysis the following operations were performed: Calculation of the sums of products, determination of the constants, determination of the significance level, and calculation of the final form of the equation.

For each of the parameters, represented by x , a corresponding X term was defined. The latter X term was used in the computational work to simplify the calculations. The X terms were defined as follows:

$$\begin{aligned} X_1 &= x_1 \times 10^{-1} & X_2 &= x_2 \times 10^{-4} & X_3 &= x_3 \times 10^{-4} \\ X_4 &= x_4 \times 10^{-1} & X_5 &= x_5 \times 10^0 \end{aligned}$$

Calculation of the Sum of Products. The value of the sums of products of X_1, X_2, X_3, X_4, X_5 , and y , with each other and with themselves, were required.

The sum of products, of any variable, X_i , with any other variable, X_j , is given by the equation below:

$$S_{ij} = \sum (X_i - \bar{X}_i)(X_j - \bar{X}_j)$$

where:

- S_{ij} = sum of products of X_i and X_j
- X_i = any reading in the X_i column
- \bar{X}_i = the mean value of the X_i column
- X_j = any reading in the X_j column
- \bar{X}_j = the mean value of the X_j column.

For computational simplicity a modified form of the above equation, which is given below, was used. This calculation is illustrated by using data from the columns of the modified pulse number, X_1 , and the modified carbon tetrachloride flow rate, X_2 :

$$S_{1j} = \sum X_1 X_j - \frac{1}{n} \left(\sum X_1 \right) \left(\sum X_j \right)$$

where:

S_{1j} = sum of products of X_1 and X_j

$X_1 X_j$ = the product of any number in the X_1 column with the number in the X_j column on the same row

n = number of individual values in any column

X_1 = any reading in the X_1 column

X_j = any reading in the X_j column

$$S_{12} = \sum X_1 X_2 - \frac{1}{15} \left(\sum X_1 \right) \left(\sum X_2 \right)$$

$$S_{12} = 4.22557 - 1/15(34.3)(1.873)$$

$$S_{12} = -0.05735.$$

By a similar procedure S_{11} , S_{13} , S_{14} , S_{15} , S_{17} , S_{22} , S_{23} ,

S_{24} , S_{25} , S_{27} , S_{33} , S_{34} , S_{35} , S_{37} , S_{44} , S_{45} , S_{47} , S_{55} , S_{57} ,

and S_{yy} were calculated.

Matrices. In order to determine the value of the constants in the proposed regression correlation it was necessary to handle a type of equation called a matrix equation. The laws of matrix algebra are similar to, but vary from, the laws of ordinary algebra⁽⁸⁰⁾. A matrix may be defined as an array of quantities. An example of a simple matrix is as follows:

$$\begin{bmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{bmatrix}$$

The quantities, " a_{ij} " are called the elements of the matrix. The vertical lines of the matrix are called columns. The horizontal lines of the matrix are called rows. The first subscript refers to the row, and the second subscript refers to the column. If a_{21} equals a_{12} the matrix is called symmetric matrix. Square, symmetric matrices and matrices involving only a single column were involved in this investigation. Two matrices are equal when, and only when, the corresponding elements are equal.

The calculations for this investigation were handled following methods outlined by Duncan⁽⁸⁵⁾ and Fisher⁽⁸¹⁾.

Determination of Regression Coefficients. The matrices

that were important in this investigation are the B, C, G, I and S matrices defined as follows:

I and S matrices defined as follows:

$$S = \begin{bmatrix} s_{11} & s_{12} & s_{13} & s_{14} & s_{15} \\ s_{21} & s_{22} & s_{23} & s_{24} & s_{25} \\ s_{31} & s_{32} & s_{33} & s_{34} & s_{35} \\ s_{41} & s_{42} & s_{43} & s_{44} & s_{45} \\ s_{51} & s_{52} & s_{53} & s_{54} & s_{55} \end{bmatrix} =$$

$$\begin{bmatrix} 4.617300 & -0.057350 & 0.008630 & -2.352500 & 0.224000 \\ -0.057350 & 0.203957 & 0.214521 & -0.181159 & 0.422766 \\ 0.008630 & 0.214521 & 0.637814 & 0.018643 & -0.433639 \\ -2.352500 & -0.181159 & 0.018643 & 3.518790 & -0.994450 \\ 0.224000 & 0.422766 & -0.433639 & -0.994450 & 3.953800 \end{bmatrix}$$

$$C = \begin{bmatrix} c_{11} & c_{12} & c_{13} & c_{14} & c_{15} \\ c_{21} & c_{22} & c_{23} & c_{24} & c_{25} \\ c_{31} & c_{32} & c_{33} & c_{34} & c_{35} \\ c_{41} & c_{42} & c_{43} & c_{44} & c_{45} \\ c_{51} & c_{52} & c_{53} & c_{54} & c_{55} \end{bmatrix}$$

$$B = \begin{bmatrix} b_1 \\ b_2 \\ b_3 \\ b_4 \\ b_5 \end{bmatrix} \quad I = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{bmatrix}$$

$$G = \begin{bmatrix} g_1 \\ g_2 \\ g_3 \\ g_4 \\ g_5 \end{bmatrix} = \begin{bmatrix} s_{1Y} \\ s_{2Y} \\ s_{3Y} \\ s_{4Y} \\ s_{5Y} \end{bmatrix} = \begin{bmatrix} 0.8000 \\ 0.0039 \\ -5.3622 \\ 3.0244 \\ 3.9538 \end{bmatrix}$$

The I matrix serves the same purpose in matrix algebra as the numeral "one" serves in ordinary algebra.

Thus, the values of the elements in the \underline{H} and \underline{Q} matrices only were unknown. The \underline{Q} matrix is defined as the inverse of the \underline{H} matrix and is obtained from the latter matrix by a process called inversion. Inversion consists of the solution of the equation stating that the product of the \underline{H} and \underline{Q} matrices equals the \underline{I} matrix. The solution of this equation was accomplished by the abbreviated Doolittle⁽⁸¹⁾ method. In this method the \underline{H} matrix is factored into the product of two triangular matrices, \underline{D}^t and \underline{E} . A triangular matrix is one that has zeroes in all the spaces below the principal diagonal (the diagonal extending from the upper-left to the lower-right corner). In addition the \underline{E} matrix has ones along this diagonal. The factorization was carried out so that \underline{H} premultiplied by \underline{D}^t , the transpose of \underline{D} , gave the \underline{E} matrix. The transpose of a matrix is the matrix obtained by interchanging the ij and the ji element and is denoted by a prime mark. The \underline{D} and \underline{E} matrices were inverted and the desired \underline{Q} matrix was obtained as the product of the transpose of the inverse of \underline{D} premultiplied by the inverse of \underline{E} . The inversion was accomplished most easily in this fashion because the inversion of the factor matrices, \underline{D} and \underline{E} , is simplified by the unique properties of triangular matrices. An example of this method of calculation is given by Anderson and Bancroft⁽⁸¹⁾.

Upon solving these equations by this method, the C matrix shown below was obtained:

$$C = \begin{bmatrix} 0.376955 & 1.054782 & -0.445026 & 0.276637 & -0.113370 \\ 1.054782 & 26.170640 & -11.469804 & 1.022746 & -3.858801 \\ -0.445026 & -11.469804 & 6.729198 & -0.389029 & 1.891826 \\ 0.276637 & 1.022746 & -0.389029 & 0.512916 & -0.038692 \\ -0.113370 & -3.858801 & 1.891826 & -0.038692 & 0.869710 \end{bmatrix}$$

The B matrix was solved by making use of the relation that B equals GC. After first setting up the G and the C matrix, they were multiplied together. The result was the B matrix shown below:

$$B = \begin{bmatrix} 3.080 \\ 50.286 \\ -30.181 \\ 3.709 \\ -6.928 \end{bmatrix} = \begin{bmatrix} b_1 \\ b_2 \\ b_3 \\ b_4 \\ b_5 \end{bmatrix}$$

In the B matrix the value of each element is the regression coefficient of the corresponding X term.

Determination of Significance Level. A significance level is defined as the probability of concluding that a particular regression coefficient is significant when, in fact, there is no real trend to the data. In other words, it is the probability that an estimate of the true regression will be large enough by chance to be called significant when the true regression being estimated is nil. To determine this level, we calculate a statistic called "t." This statistic is defined as the regression coefficient divided by its standard error. Thus, if the regression coefficient

greatly exceeds its standard error it means that there is a very low probability that this large value of "t" was obtained by a chance trend of random errors. Tables of the probability of obtaining various values of "t" are available in statistical text books⁽⁸⁴⁾.

Before the "t" test can be applied it was necessary to calculate the error variance of the regression equation. The y values vary around the mean y value of the data. The amount of this variation is expressed by the term " S_{yy} ." Now, the value of S_{yy} may be thought of as being composed of two terms. The first term $S_{\hat{y}}$ expresses the variation of y values given by the regression equation about the mean y value. This is called the regression sum of squares. The other term expresses the variance of the observed y values about the y values given by the regression equation. It thus measures, in a sense, the error or failure of the regression equation to match the observations. This term is called the error sum of squares. The error variance is defined as the error sum of squares divided by an appropriate number. The appropriate number is dependent upon the number of experimental tests.

The error sum of squares was calculated as follows:

$$S_e = S_{yy} - S_r = S_{yy} - \sum b_i S_{iy}$$

where:

- S_e = error sum of squares
- S_{yy} = total sum of squares
- S_r = regression sum of squares
- S_{iy} = sum of products of X_i and y
- b_i = regression coefficient

$$S_e = 191.31 - \left[\begin{array}{l} 0.80(3.08) + 0.0039(50.286) - \\ 5.362(30.161) + 0.0224(3.709) - \\ 3.9538(6.928) \end{array} \right]$$

$$S_e = 43.00.$$

The error variance of the regression was calculated from the following equation:

$$s_e^2 = \frac{S_e}{n - 1 - m}$$

where:

- s_e^2 = error variance
- S_e = error sum of squares
- n = total number of observations
- m = number of variables studied

$$s_e^2 = \frac{43.00}{15 - 1 - 5}$$

$$s_e^2 = 4.78.$$

After calculating the error variance, it was then possible to calculate the standard error of any regression coefficient. This calculation was performed as shown below. The results for X_1 , the pulse number, will be used.

$$s_{bi} = \sqrt{c_{ii} s_e^2}$$

where:

s_{bi} = standard error of the i th regression coefficient

c_{ii} = the element from the C matrix at the i th row and the i th column

s_e^2 = error variance

$$s_{b1} = \sqrt{0.376955 \times 4.78}$$

$$s_{b1} = 1.342$$

When the standard error of the regression coefficient was available the value of " t " was calculated as shown below:

$$t = \frac{b_1}{s_{b1}}$$

where:

t = a statistic

b_1 = regression coefficient

s_{b1} = standard error of the regression coefficient

$$t_1 = \frac{3.08}{1.342}$$

$$t_1 = 2.30$$

From a table of the probability distribution of the " t " statistic the probability that a " t " of 2.30 or greater could be obtained by chance errors was read. This value was entered in Table X, page 101, under the appropriate heading.

This last series of calculations was repeated until the significance level of all the regression coefficients had been obtained.

Calculations of Final Equation Form. Before the regression equation was available in a useful form, the value of b_0 must be calculated; and the values of the b 's must be changed to correspond to the x 's instead of to the X 's. The latter calculation merely involved changing the decimal point to take into consideration the difference between the respective x 's and the X 's. The adjusted constants are as follows:

$$\begin{aligned} b_1 &= 0.308 & b_2 &= 0.00503 & b_3 &= -0.00302 \\ b_4 &= 0.371 & b_5 &= -6.923 \end{aligned}$$

The value of b_1 was further modified to change the units of x_1 from inches per minute to feet per minute. The changed b_1 value was 3.70.

The constant, " b_0 ," was calculated by making use of the following relation:

$$b_0 = \bar{y} - b_1\bar{x}_1 - b_2\bar{x}_2 - b_3\bar{x}_3 - b_4\bar{x}_4 - b_5\bar{x}_5$$

where:

- b_0 = intercept constant, per cent
- \bar{y} = mean value of stage efficiency, per cent
- b_1 = regression coefficient of pulse number on stage efficiency, 3.70
- \bar{x}_1 = mean value of pulse number, feet per minute
- b_2 = regression coefficient of heavy phase flow rate on stage efficiency, 0.00503
- \bar{x}_2 = mean value of heavy phase flow rate, pound per hour-square foot
- b_3 = regression coefficient of light phase flow rate on stage efficiency, -0.00302
- \bar{x}_3 = mean value of light phase flow rate, pounds per hour-square foot
- b_4 = regression coefficient of acetone concentration in feed on stage efficiency, 0.371
- \bar{x}_4 = mean value of acetone concentration in feed, weight per cent
- b_5 = regression coefficient of relative flow rate on stage efficiency, -6.93
- \bar{x}_5 = mean value of relative flow rate, dimensionless

$$b_0 = 20.20 - 0.308(22.87) - 0.00503(1248) + 0.00302(2386) - 0.371(1209) + 6.928(0.410)$$

$$b_0 = 12.44 \text{ per cent.}$$

19
IV. DISCUSSION

The results obtained during this investigation are discussed, recommendations for future work are presented, and the limitations imposed upon the investigation are stated in this section.

Discussion of Results

The discussion of results deals with the experimental errors, the performance and design of the extractor, the limiting liquid flow rates, the analysis of stage efficiency, and the evaluation of the pulse extractor.

Experimental Errors. It is the author's opinion that the two greatest causes of error in experimental liquid-liquid extraction studies are the improper mixing of the feed and failure to obtain a steady-state condition before starting the testing period. This opinion is based upon experience gained from reading student reports (32) about operating a two-inch laboratory spray column. After a study of the experimental procedure, additional possible errors are recognized. Among these additional errors the following should be mentioned: Errors in weighing the phase flow rates, errors connected with sampling and analysis, faulty refractive-index data, and errors in the calculations.

Steady-state Conditions. The method of calculation upon which the stage efficiency is based assumes that the extractor is operating at a "steady-state condition." The steady-state condition is defined by Colburn and Pigford⁽¹⁷⁾ as "that state in which there is no change in composition of either phase at any position with time." For this investigation the steady-state condition was defined as that condition obtained within the extractor, when for three successive five-minute readings, commencing not before two complete liquid changes in the extractor, there was no change in either the flow rate or in the density of (recorded to the third place after the decimal) the raffinate phase. Alternate criteria, that were also used, were five successive readings with no change in either flow rate and with a change in the specific gravity of the raffinate phase of no more than ± 0.001 units.

Colburn and Pigford⁽¹⁷⁾ present data on the time required to approach to within 99 plus per cent of the steady-state condition. The factors they considered important in determining the time required to reach this condition were the number of theoretical stages present in the extractor and the relative flow rate. For the range of relative flow rates used in this investigation, from 1.75 to 2.20 complete changes in the column liquid holdup were required before the steady-state condition was reached. Colburn and Pigford's data assume that the extractor is operating

with no change in the composition or flow rate of any feed stream. They further assume that the extractor is filled with material of the same composition as the feed streams. In this investigation the column was drained and refilled with the proper feed before each test.

The liquid holdup in the pulse extractor was 444 cubic inches. This holdup was divided between the sections as follows: 114 cubic inches contained in the bellows, 176 cubic inches contained in the bottom tee, and 154 cubic inches contained in the column, itself. Of this volume, the heavy phase occupied about 190 cubic inches, leaving 254 cubic inches for the light phase. Assuming that no solute was present in the extractor, the heavy-phase holdup was 9.15 pounds and the light-phase holdup was 10.8 pounds. If any acetone had been present, the holdup would have been less because of the decrease in density of the phases as acetone is introduced. For the lowest flow rates used during this investigation a holdup time of 1.1 and 0.53 hours was obtained for the heavy phase and the light phase, respectively. Holdup time is the time required for one complete change of the liquid in the extractor. For the highest flow rates used in this investigation the holdup time was 0.09 and 0.07 hour for the heavy phase and the light phase, respectively. The average values of the time required for one complete liquid change are 0.36 and 0.22 hour, respectively, or about 20 minutes.

A question that naturally arises is whether or not the criteria for the steady-state condition selected for this investigation are really capable in detecting any significant changes during the operation of the extractor. The data given below to support the affirmative answer to this question were not included in this report. During Test Number 12, the holdup time was 0.69 and 0.16 hour for the heavy phase and light phase, respectively. Starting 1.75 hours after the extractor was filled the following values for the specific gravity of the raffinate phase were obtained: 1.580, 1.582, 1.583, 1.584, 1.586, 1.588, 1.588, 1.590, 1.589, 1.588, 1.590, 1.590, 1.590, and after the testing period ended, 1.590. During the period of approach to the steady-state condition the specific gravity of the raffinate phase changed about 0.01 unit. This change in the specific gravity is equivalent to a concentration change of about 1.0 per cent acetone. For Test Number 5 the liquid holdup was 0.18 hour based on the heavy phase. During this test, an original higher flow rate was changed to the value equivalent to a holdup of 0.18 hour, because of unsatisfactory plate operation. The specific gravity of the raffinate phase samples (commencing 0.67 hour after the extractor was filled) was as follows: 1.576, 1.548, 1.527, 1.522, and 1.518. At this time the flow rate was reduced. The specific gravity readings continued as follows: 1.513, 1.518, 1.520, 1.525, 1.527,

1.529, 1.529, 1.530, 1.529, 1.530, 1.530, and after the testing period, 1.530. The fast response to the change in the flow rate for this test should be noted.

Finally, it will be mentioned that the criteria for the steady-state condition used for this investigation were stricter than the criteria mentioned in the literature⁽¹⁷⁾. It was thus believed that the procedure employed for testing for the presence of the steady-state condition was adequate to detect any significant trends in the extractor performance. It is also believed, conversely, that no appreciable error was caused by taking the experimental data during a condition of transient operation. It is, of course, impossible to estimate the exact limits of this error. This error may be considered one of the factors contributing to the random experimental error for this investigation.

Lagrange Preparation of the Feed. In the analysis of a three-component system by physical methods, two different physical properties must be fixed in order to determine the exact composition. Thus, for example, in the system carbon tetrachloride-acetone-water if the refractive index and the density of a sample were known the exact composition of this sample could be found (assuming, of course, that data expressing the relation between the density, index of refraction, and composition were available). For this investigation the physical property determined, in addition to the refractive index, was knowing (or really assuming)

that the respective phases were saturated with the third component. A raffinate phase sample with a refractive index of, say, 1.4588 contains 0.49 per cent acetone if, and only if, the sample is also saturated with water. If each phase is not saturated with the third component the refractive index reading, no matter how accurate, would not give the true composition. It is the author's belief that the error caused by a slight undersaturation or oversaturation with the third component of each sample far overshadows any inaccuracies in determining the refractive index.

In the raffinate-phase and the extract-phase samples very little trouble with saturation should have been encountered. The close and intimate contact of these phases with each other should certainly result in their being saturated with the third component. The feed sample, however, was taken before it had been contacted with solvent in the extractor. Unless some definite precautions were taken to see that the feed was, it probably would not be, saturated with water. In this investigation the feed was prepared by mechanical mixing in the presence of an excess of water.

The magnitude of the error caused by unsaturation may be illustrated by a comparison with the data in Table VI, page 93. The carbon tetrachloride used in this investigation had a refractive index of 1.4628. When the carbon tetrachloride was saturated with water, however, the refractive index was reduced to 1.4594, a difference of 0.0034. This difference is equivalent

to an error of about four weight per cent acetone. For the extract phase the refractive indices for the water saturated and unsaturated with carbon tetrachloride were 1.3330 and 1.3320. This difference is equivalent to about 2.5 per cent acetone.

Other Experimental Errors. It is believed that no large errors were made in the weighings, the sampling, and in measuring the refractive index of the various samples. It is also believed that the timed periods were held to within plus or minus 15 seconds, and that the weights recorded were within plus or minus ten grams of the true weight. Both of these estimates are conservative, since an attempt was made to hold these readings to within such narrower limits. The accuracy of the sampling and analytical procedures may be shown by a comparison of the tests numbered 2 and 3, 9 and 13, and 14 and 15 in Table VII, page 97. For each of these pairs of tests an identical feed was used. The feed compositions determined, and listed in this table, were 10.78 and 10.81 for tests 2 and 3, 6.96 and 6.26 for tests 9 and 13, and 10.42 and 10.86 for tests 14 and 15, all in weight per cent of acetone. The differences between the members of any pair of readings include all the errors connected with sampling, storage of the samples, and in determining the refractive

index. It is certainly felt that the errors contained in these samples are negligible compared to the other errors present in this investigation.

Acetone Material Balance. The acetone material balance furnishes a check on the total accuracy of the measured flow rates and compositions. If, as mentioned previously, the errors in determining the flow rates and compositions of the extract and raffinate phases are considered negligible, then the acetone balance serves as a check on the accuracy of the feed composition. If the feed sample were not saturated with water, it would tend to make the "acetone in" less than the "acetone out." For 12 out of 13 tests contained in Table VIII, page 98, the acetone in was less than the acetone out. This consistent difference tends to add credence to the theory that the principal experimental error was in the measured feed composition. It appears definite that this error was large enough to overshadow any other errors that may affect the material balance. No other known source of error would cause such a trend in the acetone balance. Examining the discrepancy values closer, it is seen that three of these values are contained in the group from zero to two per cent; two, from two to four per cent; five, from four to six per cent; and three, over six per cent. The average of all discrepancy values is 4.3 per cent. This average discrepancy value may be taken as an indication of the accuracy of the measured experimental data.

Refractive Index Data. The refractive index data determinations were based upon a method of analysis presented in the literature⁽¹⁰⁾. This method of analysis consisted of determining the specific gravity of a sample at $30^{\circ}/4^{\circ}$ C. The refractive index data were determined by measuring the refractive index of samples containing known amounts of acetone. These samples were prepared by mixing carbon tetrachloride, acetone, and water and allowing equilibrium to be reached between the two phases. Thus both the refractive index data and distribution data were obtained at the same time. The agreement between the experimentally determined distribution data and the data presented in the literature should serve as a check on the overall accuracy of both sets of data. That there is a close check may be seen by comparing points from Table II, page 18, to the distribution curve shown in Figure 5, page 96. Except for one point that appears out of line, the curve in Figure 5 is bounded by the distribution data from the literature at 30° C and 20.6° C. This is as expected because the experimental distribution data were obtained at about 22° C. The one point that is out of line in Figure 5 is also the only point that does not lie on a straight line on the refractive index versus acetone content chart, Figure 4, page 95. In Figure 4 and Figure 5 the curves were drawn through this point. In the charts used in the calculations this point was rejected and a smooth curve drawn through the remaining points.

Calculations. The calculations in an experimental investigation are often overlooked as a potential source of errors. It is believed, of course, that there are no mathematical errors present in the calculations. However a large error could easily be obtained if the calculations were such that they did not give the desired information. The following remarks are considered important enough to mention in this discussion.

During the calculation of both the acetone balance and the stage efficiency it was assumed that the carrier solvent was immiscible with the extract phase and that the solvent was immiscible with the raffinate phase. The average raffinate phase contained 0.86 per cent acetone and, if saturated, 0.00 per cent water. The average extract phase contained about six per cent acetone and, if saturated, 0.26 per cent carbon tetrachloride. Any errors introduced by the assumption of immiscible phases are certainly negligible when compared to the much larger experimental errors.

The methods of calculating the number of theoretical stages for an extraction process have become quite standardized. The method used in this investigation is the method recommended by Hunter and Nash^(33, 34), Elgin⁽²²⁾, and Bull and Coli⁽¹¹⁾. The method of interpolating to find the last fractional stage in this method is, in the author's opinion, open to question.

This interpolating was done in a similar manner for all tests; so that, any errors introduced by this calculation are at least consistent. As no better method of calculation has appeared this error must be tolerated.

The author has been asked several times during the preparation of this work why the results are presented in terms of the stage efficiency rather than in terms of the H.E.T.S. The stage efficiency was selected to avoid having to arbitrarily select a height upon which to base the H.E.T.S. If the H.E.T.S. were based upon the entire extractor height it would make the extractor appear unduly inefficient. For example, if the height of the extractor were extended in order to obtain more separation, a lot of the height would not be duplicated for the larger installation. The total column height may be considered as composed of two parts. These parts are the active height and the auxiliary height. For a larger installation only the active height is increased. The question is just what is active height and what is not. If just the height occupied by the baffle plates were taken as the height upon which to base the H.E.T.S. the converse argument is true. To avoid arbitrarily selecting an intermediate height on which to base the H.E.T.S. the stage efficiency was used. The eight baffles used in the pulse column can present no basis for argument. In mentioning stage efficiency, however, it is often wrongly assumed that the stage efficiency is a property

of the stage, itself, and not a property of the entire extractor. Because in the pulse extractor used in this investigation at certain flow rates a stage efficiency of, say, 20 per cent was obtained, it does not mean that five additional plates would result in the addition of one theoretical stage to the extractor.

In calculating the stage efficiency the use of four different graphical steps was involved. These steps were the determination of the concentration of acetone from the specific gravity of a sample, the determination of the concentration of acetone from the refractive index of a sample, the determination of equilibrium concentrations from the distribution chart, and the graphical solution of the Hunter and Nash plot. In this report the steps are represented by Figures 3, 4, 5, and 10, pages 94, 95, 96, and 112, respectively. For inclusion in this report, however, these figures were reduced in scale. For the actual calculations enlarged versions of these charts were used. The exact scale used in these enlarged charts was as follows: Concentration of acetone for Figures 3, 4, and 5, one inch equals 0.20 per cent; concentration of acetone for Figure 10, one inch equals 0.40 per cent; concentration of water for Figure 10, one inch equals 10.0 per cent; refractive index for Figure 4, one inch equals 0.002 unit; and specific gravity for Figure 3, one inch equals 0.002 unit.

Summary of Experimental Errors. After considering the nature of the possible experimental errors in this investigation it is believed that the principal error was caused by the under-saturation of the feed with water for the pulse extractor. This error resulted in erroneous low values for the feed compositions. It is further believed that the acetone balance is a direct measure of this error. If this is true, 4.3 per cent can be taken as a good indication of the accuracy of the experimental values. It should be noticed that a low value of the feed concentration causes errors on the safe side. That is, if greater acetone concentrations were used in the calculations, greater stage efficiencies would have been obtained.

Design Features and Operations of the Pulse Extractor. Several qualitative observations and notes on the design and operation of the pulse extractor are considered important enough to be included in this discussion.

Flow Characteristics of the Pulse Extractor. The exact flow pattern of the phases during operation of the pulse extractor was probably a unique feature of this extractor. This flow pattern was found to be remarkably stable. That is, it varied only slightly with large changes in flow rates and agitation. As the heavy-phase flow rate increased, the amount of the heavy-phase holdup on each plate also increased. The

violence, or rapidity, of the flow pattern changed as the pulse amplitude and the pulse frequency were increased. Except for these two changes, the flow pattern was similar during all experimental tests.

An attempt will be made to describe the flow pattern for this extractor. Following each pulse, the heavy phase and the light phase are allowed to settle. After settling each plate is covered with a layer of the heavy phase. Similarly, underneath each plate there is a layer of the light phase. During the upward part of the pulse, the light phase immediately underneath each plate was forced up through the plate. Upon contacting the heavy phase on the top of the plate the light phase was dispersed into drops. As only a relatively thin film of the heavy phase is present on each plate, the time of passage of the light phase through the heavy phase is quite short. In the return half of the stroke the entire heavy phase on each plate was dropped through the plate. The downward motion of the phases imparted a violently swirling motion to the highly dispersed drops of the heavy phase. Settling of the heavy phase took place during the pause before the upward half of the next stroke. Visually, it appeared as if the principal agitation took place during the downward half of the pulse.

In the pulse extractor there is no continuous or discontinuous phase in the sense that the word is usually used in

extraction. That is, in the pulse extractor there are two principal interfaces, one in the top disengaging section and one in the bottom disengaging section. In between these two interfaces are located the stages, with the interface present in each stage. In many aspects, however, the heavy phase acts as if it were a dispersed phase passing through the "continuous" light phase. Two important similarities between the heavy phase of the pulse extractor and the dispersed phase of a spray tower will be mentioned. The first similarity involves the flow patterns during flooding. Flooding, in an extractor in which one phase is dispersed, is accompanied by a carry-over of the dispersed phase with the continuous phase. This is true no matter which phase is causing the flooding. In an extractor in which there is no dispersed or continuous phase, flooding is accompanied by the carry-over of the phase not causing the flooding. In the pulse extractor, no matter which phase is causing the flooding condition, the net result of flooding is a carry-over of the heavy phase with the light. The second similarity involves the flow patterns during increased agitation. In extractors in which one phase is dispersed, the dispersed phase moves through a relatively motionless fluid. Any agitation applied to the extractor causes an increased dispersion and motion of the dispersed phase. The additional agitation has relatively little effect upon

the continuous phase. In the pulse extractor an increase in agitation seems to affect only the flow pattern of the heavy phase. In the pulse extractor an increase in agitation results in an increase in the dispersion of the heavy phase on the downward part of the stroke. In extreme cases the heavy phase was so finely dispersed that complete settling had not been obtained before the next pulse. After a few pulses at such conditions, the entire column was filled with a cloudy dispersion of the heavy phase in the light phase. The extract phase carried out of the extractor droplets of the feed. The raffinate phase remained clear, however, and was unaffected by the increased agitation. The extract, after removal from the extractor, settled into two layers. For the pulse extractor used in this investigation mechanical carry-over of the feed with the extract phase occurred at a pulse amplitude of 0.217 and 0.133 feet and a frequency of 21 strokes per minute.

Bellows. In the design of a pulse extractor considerable thought must be given to the means of pulsating the liquid phases. Two methods that might be used are, one, a constant volume pump modified to permit pulsing and, two, a bellows equipped with a suitable driving mechanism. In at least one pulse extractor⁽⁵⁰⁾ in which a pump was used as the means of applying pulsation trouble was encountered from contamination

of the pump's lubricants and from leaks around the shaft seals. It can be stated that no such difficulties were encountered when using a bellows.

Pulsations. For a given frequency and amplitude the nature of the pulse can still differ depending upon the time distribution of the motion. The pulse may operate, for example, with simple harmonic motion with the desired frequency and amplitude. Alternately, the pulse may consist of simple harmonic motion interrupted at each half stroke to allow settling. These two methods are, undoubtedly, the methods that would first occur to a designer. The construction of a mechanism to apply this type of motion to the bellows is extremely difficult. For this investigation a simple harmonic motion interrupted once during each stroke was used. The retracted, or bottom portion, of the simple harmonic pulse was cut off to allow settling. The position of the cut-off point varied with the amplitude of the pulse. In fact, the pulse amplitude was selected by varying the cut off. The motion of the push rod during the experimental studies was up-down-pause and repeat. If the cut-off point were eliminated the motion of the push rod would have been simple harmonic motion with a natural amplitude of about three inches and a frequency depending upon the rotational speed of the cam shaft.

The nature of the time distribution of the pulse used in this investigation had one severe disadvantage. At the start of the upward stroke the acceleration, and hence the force, applied to the push rod approached infinity (except for the deformation of the steel frame, cam, and other structures it would have been infinite). This terrific original force is extremely undesirable in a commercial model, as it causes injurious vibrations and requires the use of heavier and stronger parts.

Column Holdup. The total liquid holdup was about three times the holdup of the active portion of the pulse extractor. This condition is undesirable in commercial extractors because of the large solvent inventories and equipment sizes required. This large holdup is also undesirable in an experimental extractor because of the greater time required for the steady-state condition to be reached. A great deal of this holdup could be eliminated by changing the design of the bottom disengaging section and eliminating the tee.

The exact column holdup is about 20 pounds. When operating at a frequency and amplitude of 10.5 strokes per minute and 0.25 feet per stroke the holdup amounts to about 2.5 per cent of the hourly capacity.

Limiting Liquid Flow Rates. For an extractor of any given dimensions only certain liquid flow rates can be used. These

liquid flow rates form the limits at which the extractor may be operated and are, hence, an important design factor. In fact, the cross sectional area of an extractor is fixed by the necessity of keeping the liquid flow rates between the limiting liquid flow rates for that particular type of equipment. Tests were made to determine the effect of the pulse frequency, the pulse amplitude, the light phase flow rate, and the heavy phase flow rate on the limiting-liquid flow rates for the pulse extractor.

Definitions. During the operation of the pulse extractor four conditions were recognized as being examples of unsatisfactory column operation. These conditions were called heavy-phase flooding, light-phase flooding, dumping and blowing. As these terms are, to a large extent, arbitrary they will be defined.

Heavy-phase flooding refers to that condition in which the heavy phase is being added to the extractor faster than it is being moved down the extractor by the pulsating motion of the liquids. During heavy-phase flooding, the heavy phase fills the top disengaging section and begins to flow out of the light-phase outlet line.

Dumping refers to that condition in which the heavy phase fails to cease flowing during the pauses between the strokes. During dumping only partial, at best, coalescence of the heavy phase takes place. The operation of the extractor during

dumping may be compared to the operation of a perforated plate extractor with slight additional agitation.

Blowing refers to that condition in which the heavy phase fails to maintain a layer on the top of the plate during the upward part of the pulse. That is, the heavy-phase layer is not thick enough to remain on top of the plate and disperse the light phase but is simply "blown" up with the light phase.

Light-phase flooding refers to that condition when the light phase is being introduced so fast that it carries the heavy phase up through the extractor and out the light-phase outlet. Both light-phase and heavy-phase flooding are similar in that the heavy phase is eventually forced out the extract line. The two types of flooding are similar, also, in that the heavy phase is being introduced faster than it can pass through the extractor. In light-phase flooding, however, there is no downward motion of the heavy phase. Heavy-phase flooding can take place only at high heavy-phase flow rates. Light-phase flooding is essentially unaffected by the heavy-phase flow rate. During this investigation no condition that could properly be called light-phase flooding was encountered. The term "light-phase flooding" was extended to apply to that condition present when the high light-phase flow rates appeared to endanger the physical construction of the column. During the normal operation of the extractor an air cushion was present at the top of the column. At high light-phase flow rates this

air cushion was reduced to a dangerous extent between the strokes. Then, during the upward portion of the stroke, the entire force of the moving liquids was absorbed by the column top flange. During one test the column top flange was actually forced off the column. Leaks around the column top flange and the pyrex pipe gaskets were encountered in the tests near the light-phase flooding flow rate.

Limitations on Study. Before any quantitative discussion is begun the following points should be mentioned. The limiting flow rates were determined using the two component system carbon tetrachloride and water. Thus much larger density and interfacial tension differences were present than if any acetone had been present.

Although the flooding velocities place actual limits on the operation of the extractor, the dumping and blowing velocities do not. That is, the column can be operated in the dumping and blowing region; but the plates will not operate in a manner defined as satisfactory operation. It may be true that although the column plates are not operating "satisfactorily" the column itself may be operating quite efficiently.

The limiting flow rates were measured by operating the column at what was visually defined as satisfactory plate operation. Thus, allowances for the subjective error must be made in interpreting the data on the limiting liquid flow rates.

Blowing. At low heavy-phase flow rates there was not enough of the heavy phase on each plate to disperse the light phase as it was forced through the plates. This condition was called blowing. The heavy-phase flow rate at various light-phase flow rates necessary to stop this phenomenon are shown graphically in Figures 7, 8, and 9, pages 105, 107, and 108. The data used to plot these figures are contained in Table XII, page 104.

It was found that the heavy-phase flow rate necessary to stop blowing was a function of the light-phase flow rate and the pulse frequency. For all three tests there was a linear relation between the heavy-phase flow rate necessary to eliminate blowing and the light-phase flow rate. For the two tests at pulse frequencies of 10.5 strokes per minute this linear relation started at the origin and increased till about 1,000 pounds per hour-square foot of the heavy phase were required at a light-phase flow rate of 20,000 pounds per hour-square foot. When the frequency was increased to 21 strokes per minute the slope of the line increased until about 2,750 pounds per hour-square foot of the heavy phase was required at a light-phase flow rate of 20,000 pounds per hour-square foot. This was almost a threefold increase in the heavy phase velocity needed to stop blowing for a twofold increase in

the frequency. The pulse amplitude was found to have no effect on the phenomenon of blowing.

Light Phase Flooding. The light-phase flooding flow rate was found to be equal to about 22,000 pounds per hour-square foot. The rather surprising result was obtained that this rate was independent of both the pulse frequency and the pulse amplitude. The determination of the light-phase flooding flow rate was not very reproducible. Because of this the slight differences between the three tests were not considered important.

Dumping. If the extractor were operating with a light-phase flow rate of zero and a finite heavy-phase flow rate, there would be a slight downward motion of the phases, during the settling period, caused by the raffinate-phase draw off. This downward movement of the phases results in the heavy-phase flowing or dumping through the plates. During dumping, the heavy-phase contents of a stage were lost. This loss was accompanied by the formation of relatively large drops or even continuous streams. During a condition of dumping, then, there is low interfacial area between the two phases. The application of power to the extractor when dumping, resulted in only a slight increase in the interfacial area because most of the heavy phase had already passed the plates.

Poor extraction would be expected for a pulse extractor operating in the dumping region.

If a sufficient upward movement were imparted to the phases, the dumping of the plates was stopped. This slight upward movement of the phases was accomplished by introducing a certain amount of the light phase. For every heavy-phase velocity the light-phase velocity must exceed a certain minimum value if dumping is to be avoided. The plot of the light-phase velocity necessary to stop the dumping of the heavy phase results in the dumping limit curve. In Figures 7, 8, and 9, pages 105, 107, and 108, examples of the dumping limit curves are shown.

The dumping limit curve is a smooth curve, slightly concave inward, that could nearly be represented by a straight line. The curve was similar in appearance for all three tests. The exact position of the curve was found to vary slightly with the pulse frequency and probably not at all with the pulse amplitude. At higher pulse frequencies a lower light-phase flow rate should be required to stop dumping. At the higher frequencies less of the heavy phase is held on each plate. This probably indicates that the dominant factors determining the boundary of the dumping region are the heavy-phase liquid holdup on each plate and the heavy-phase flow rate.

Heavy Phase Flooding. From Figures 7, 8, and 9, pages 105, 107, and 108, it appears as if the heavy-phase flooding flow rate is affected by three variables: The frequency, amplitude, and light-phase flow rate. The effects of the frequency and amplitude on the heavy-phase flooding flow rate are seen by the different values of the maximum point for the three different test series. The effect of the light-phase flow rate is seen in the decrease in the heavy-phase flooding velocity as the light-phase flow rate increases. Oddly enough this latter effect was unnoticed for the test at a pulse frequency of 10.5 strokes per minute and an amplitude of 0.125 feet.

For the tests at 10.5 strokes per minute, a plateau was obtained at the top of the heavy-phase flooding curve. This seemed to indicate that over certain ranges the heavy-phase flooding velocity was not a function of the light-phase flow rate. In this range the flooding velocity depended only upon the amount of liquid that could be moved down the column by the pulse. In Figures 7 and 9, pages 105 and 108, an essentially equal maximum value for the heavy-phase flooding velocity is seen. In both of the tests an equal pulse number of 2.6 feet per minute was used. For one test, however, the pulse number was made up of the product of 10.5 strokes per minute and 0.250 feet per stroke. For the other test the

pulse number was made up of the product of 21.0 strokes per minute and 0.125 feet per stroke. The pulse number is a measure of the amount of liquid moved in the extractor. For a pulse number of 2.6 feet per minute it is found that 156 cubic feet of the combined phases per hour-square foot was moved by the bellows. This is equivalent to 5,500 pounds of the heavy phase per hour-square foot. This value was only about one-fourth of the total heavy-phase flow rate through the extractor. The ratio of the heavy-phase flow rate to the heavy phase moved by the bellows is even higher for the other test series. Although the maximum flow rate and the amount of liquid moved by the bellows seemed to be related, the exact nature of this function is unknown.

High values of the light-phase flow rate reduced the value of the heavy-phase flooding velocity slightly for the tests at a pulse number of 2.6 feet per minute. This lowering was, however, more pronounced for the test at 21 strokes per minute. It is believed that this phenomenon was caused by an effective decrease in the pulse frequency caused by a fast moving light-phase flow rate. This effective decrease in the pulse frequency would affect the test using the greater frequency most. It was further believed that if the light-phase flow rate could be increased sufficiently that the

test with a pulse number of 1.3 feet per minute would show a similar decrease in the heavy phase flooding velocity.

Analysis of Stage Efficiency. Fifteen experimental tests were made to determine the effect of the flow rates and agitation on the stage efficiency. The data from these tests are presented in Table VII, page 97, and the results deduced from these tests are presented in Table IX, page 99. A discussion of this data and the statistical analysis made of this data will be presented in this section.

Reasons for the Multiple Regression Analysis. The stage efficiency was a function of a great many variables. During the investigation most of these variables remained constant. However, large changes, both uncontrolled and intentional, in the pulse frequency, the pulse amplitude, the light-phase flow rate, the heavy-phase flow rate, and the feed concentration were encountered. The large and apparently random changes in these variables made it difficult to estimate from the results which variables affected the stage efficiency and to what extent. By glancing at Table IX, page 99, it is obvious that there are no readily apparent trends in the results.

From this table, also, it is seen that while the carrier-solvent flow rate varied twelvefold, the solvent flow rate varied sevenfold, the concentration of acetone in the feed varied fourfold, and the pulse number varied threefold the

stage efficiency changed from 12.5 to 25.0 per cent, a two-fold variation. Relatively large changes in the operating conditions were accompanied by relatively small changes in the stage efficiency.

Because of the overlapping variations in the column conditions and the relatively small change in the stage efficiency with large changes in the column parameters, it was essential that the data be analyzed by some method not so open to subjective errors as the usual graphical interpretation of data. For the purpose of analyzing the experimental data a multiple regression equation was used. The statistical analysis was used as a mathematical tool to aid in the correlation of the results. This method of correlation was selected as being the most satisfactory considering the two factors mentioned previously.

Purpose of the Multiple Regression Analysis. Because of its very nature, the multiple regression analysis, like other mathematical devices, cannot reveal anything that is not already implicit in the data. In simple cases the experimenter's judgment would lead him to the same conclusions as would be arrived at by the application of statistics. The value of the latter would merely be to form an objective test for conclusions not liable to bias on the part of the observer.

As the complexity of the data increases, however, it becomes more and more difficult to arrive at a reliable conclusion by unaided judgment. In such cases some form of statistical treatment is extremely helpful.

In this study it was desired to determine the effect on the stage efficiency of varying independently each of the four experimental parameters. As stated before nothing was added to the data, the data were merely analyzed so that the effect of each variable could be studied independently of the other variables. Because of limitations in the equipment and procedure, the effect of the variables could not be separated except by a treatment such as used.

Assumptions in the Analysis. The statistical analysis used in this report is based upon three assumptions. These assumptions are that the experimental errors are random, homogeneous, and independent⁽³²⁾. The first assumption requires that the errors inherent in the experiment vary randomly. It is generally assumed in work of this nature that the errors are random unless there is some reason to doubt this. The second assumption requires that each experimental test be open to the same source of errors. As no change was made in the procedure or the experimental apparatus during this investigation, no reason was seen for doubting the validity of this assumption.

The third assumption requires that the errors in one test be unaffected by the errors in the other tests.

Nature of the Regression Equation. The form of the regression equation must be selected by the experimenter. The statistical analysis merely determines the constants in the selected equation and gives some measure of their probable accuracy. Any type of equation may be assumed. The simplest equation to assume is, of course, an equation having linear variations. Any variable desired may be included in the equation.

It was finally decided to use a linear regression equation with the following terms: Pulse number (defined as the product of the pulse frequency and amplitude), heavy-phase flow rate, light-phase flow rate, acetone concentration, relative flow rate (defined as the raffinate-phase flow rate divided by the extract-phase flow rate), and as the dependent variable, the stage efficiency.

The pulse number was selected to measure the relative agitation applied to the extractor. It would have been desirable to retain the pulse amplitude and the pulse frequency as separate variables. This was not possible, however, because of the lack of tests at varying pulse frequencies. The heavy-phase and light-phase flow rates were included because of the well known effect of flow rates on extraction efficiency.

The pulse number and the relative flow rate were both thought to increase the stage efficiency. However, no idea of the possible relation was known. The concentration of acetone was thought to have little or no effect on the stage efficiency. For these three variables, then, there was no reason to pick any function other than a linear variation with stage efficiency. That the flow rates affect extraction in an exponential function is well known. The nature of this function is, however, a subject of great disagreement. Also, curved relations often approximate straight lines over short ranges such as encountered in this work.

The final assumed form of the regression equation was as follows:

$$y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4 + b_5x_5$$

where:

- y = stage efficiency, per cent
- x_1 = pulse number (product of the pulse amplitude and pulse frequency), feet per minute
- x_2 = carrier solvent flow rate, pounds per hour-square foot
- x_3 = solvent flow rate, pounds per hour-square foot
- x_4 = concentration of acetone in feed, weight per cent
- x_5 = relative (raffinate to extract) flow rate, dimensionless
- b_0 = y intercept, per cent
- b 's = regression coefficients.

The constants for this equation were obtained by the method of least squares. The completed multiple regression equation is shown below:

$$y = 12.44 + 3.70x_1 + 0.00503x_2 - 0.00302x_3 + 0.371x_4 - 6.93x_5$$

where:

- y = stage efficiency, per cent
- x₁ = pulse number, feet per minute
- x₂ = carrier solvent flow rate, pounds per hour-square foot
- x₃ = solvent flow rate, pounds per hour-square foot
- x₄ = concentration of acetone in feed, weight per cent
- x₅ = relative flow rate, dimensionless.

Significance Level. An advantage to determining the constants in the above equation by statistical methods, is that their significance may be easily determined. For example, consider constant "b₁." This constant is called the regression coefficient of the pulse number on the stage efficiency. This coefficient shows the relation that a unit change in the pulse number would have on the stage efficiency, if all the other variables remained constant. This constant, "b₁," was determined from data on only 15 tests and is quite subject to a chance trend of experimental error. The true relation between the pulse number and the stage efficiency is represented by "B₁," the value of b₁ determined from an infinite number of tests. The value of B₁ can never be determined. The value can, however, be approximated on the basis of a limited number of tests. This approximation is the constant b₁.

An important criterion in a statistical analysis is the significance level. The significance level may be defined as the probability that the given value of b_1 is obtained even though B_1 equals zero. If B_1 equals zero the pulse number has no effect on the stage efficiency. The calculated significance level for b_1 is 0.047. That is, there is a probability of 0.047 that the pulse number has no effect on the stage efficiency, and conversely a probability of 0.953 that the pulse number has an effect on the stage efficiency. Continuing the analysis further, it is possible to say that the probability is 0.80 that the true value of B_1 lies in the interval from 1.63 to 5.76. The midpoint of this interval is 3.70 the value of b_1 and the best available estimate of B_1 .

These results given below are summarized in Table X, page 101. The significance level for each of the other regression coefficients are as follows: Heavy-phase flow rate 0.004, Light-phase flow rate 0.001, acetone concentration in feed 0.042, and relative flow rate 0.008. The range of values that has a probability of 0.80 of covering the true value of the regression coefficients are as follows: Heavy-phase flow rate 0.00351 to 0.00665, light-phase flow rate -0.00229 to -0.00375, acetone concentration in the feed 0.171 to 0.571, and relative flow rate -4.32 to -9.54.

Prediction Equation. Using the equation discussed in the previous section it is possible to calculate the most probable value of the stage efficiency for any given values of the experimental parameters. This calculation has been performed using the values of the experimental parameters from the 15 tests. These results are compared to the experimentally determined stage efficiencies in Table XI, page 102, and Figure 6, page 103. From Table XI, it is seen that the average discrepancy between the calculated and experimentally determined stage efficiencies was 9.7 per cent. The maximum discrepancy was also found to be 26.0 per cent.

There appears to be a slight trend to these results. That is, the equation predicts accurately in the middle region. For values of the stage efficiency above about 22 per cent the equation predicts values too low and for values of the stage efficiency below about 15 per cent the equation predicts values too high. By changing the assumed linear functions to more complex functions it should be possible to reduce the variation between the calculated and actual stage efficiencies even further. It was felt that with the limited data available this more exact treatment was not warranted. The equation in its present form gives good prediction of experimental values, but it is undoubtedly possible to derive an equation that will give even more accurate predictions.

Effect of the Pulse Number on Stage Efficiency. The pulse number was defined as the product of the pulse amplitude and the pulse frequency for this investigation. The pulse number is a measure of the amount of agitation applied to the extractor. It would be desirable to factor the pulse number to consider the effect of the pulse frequency and the pulse amplitude separately. Enough data were not available to allow this to be done.

As the pulse number expresses the relative amount of agitation, it is logical to expect an increase in the stage efficiency with an increase in the pulse number. No data in the literature were available on the effect of increased agitation on the stage efficiency of a pulse extractor. Oldshue and Rushton⁽⁴⁷⁾ report that as the power input to a multi-stage mixer column is increased an increase in the stage efficiency is obtained. For a twofold increase in power input, the increase in efficiency varied anywhere from 10 to 40 per cent, depending upon the other conditions. In the Feick⁽²⁵⁾ and the Scheibel^(54, 55) extractors an increase in the power input resulted in an increase in the extraction efficiency. For all three of the extractors the increase of stage efficiency with power input was a straight line function at low values of the power input.

The regression coefficient determined in this investigation for the pulse number on the stage efficiency is 3.70. That is, an increase in one unit for the pulse number would increase the stage efficiency 3.70 per cent. During the experimental tests the pulse number was varied between 0.92 and 2.67 feet per minute. This variation in the pulse number caused a maximum change in the stage efficiency of 6.45 per cent. From the statistical analysis it can be stated that there is a probability of 0.933 that the pulse number has an effect on the stage efficiency. It can also be stated that there is a probability of 0.80 that the true value of the constant expressing the relation between the stage efficiency and the pulse number is in the range from 1.63 to 5.76. The midpoint of this range and the most probable value of B_1 is 3.70.

A twofold increase in the pulse number, say from 1.0 to 2.0, results in an increase in efficiency from about 20.0 to 23.7 per cent an effective increase of about 20 per cent. It appears as if there would be an optimum pulse number for industrial operation based, not on the stage efficiency, but on the cost of the power required to operate the pulse extractor. What this optimum pulse number would be, would depend greatly on the use to which the pulse extractor is put and the cost of power at the plant site.

Effect of the Heavy-Phase Flow Rate on Stage Efficiency.

For the only other pulse extractor, for which literature⁽⁵⁰⁾ was available, an increase in the heavy-phase flow rate increased the stage efficiency. For a light-phase flow rate equal to about 0.7 of the heavy-phase flow rate a stage efficiency of 15.7 per cent was found. When the heavy-phase flow rate was doubled at a constant light-phase flow rate the stage efficiency increased to 20.2 per cent. The complete operational results for this extractor are shown in Table IV, page 46.

For the Scheibel^(54, 55), Feick⁽²⁵⁾ and Oldshue and Rushton⁽⁴⁷⁾ agitated extractors it was found that the flow rates had only a slight effect on the stage efficiency. The degree of agitation in these three extractors was a much more important factor. If the heavy phase is considered equivalent to a dispersed phase, the extractor may be compared to data in the literature on an unagitated perforated plate column. Pyle, Colburn and Duffy⁽⁸⁶⁾ stated that in a perforated plate column the stage efficiency increased only slightly with large increases in the dispersed phase flow rate.

In this investigation the regression coefficient of the heavy-phase flow rate on the stage efficiency was found to be 0.00503. That is, for every increase in the heavy-phase flow rate of 1,000 pounds per hour-square foot the stage

efficiency increases 5.03 per cent. It was also found that there was a probability of 0.996 that the heavy-phase flow rate has an effect on the stage efficiency. It can be further stated that there is a probability of 0.80 that the true value expressing this relation lies within the range of values from 0.00351 to 0.00655. The mean of this range and the most probable value of the coefficient is 0.00503. The regression coefficient for the relative flow rate (which is defined as the raffinate-phase flow rate over the extract-phase flow rate) was -6.93. That is, as the relative flow rate increased the stage efficiency decreased. The heavy-phase flow rate cannot be considered independently of the relative flow rate. The effect of the heavy-phase flow rate on the stage efficiency, taking into consideration the relative flow rate, is shown in Figure 11, page 163. As the light-phase flow rate increases, the effect of the relative flow rate decreases. This results in a lower efficiency for any given heavy-phase flow rate but a faster rate of increase in the stage efficiency for the same change in the heavy-phase flow rate. All three curves shown in this figure are similar, and if extended far enough the curves would take the same form. The data available covers only the range shown on the curve. When using a correlation of the type used in this investigation it is better not to extrapolate beyond the experimental limits. Up to the

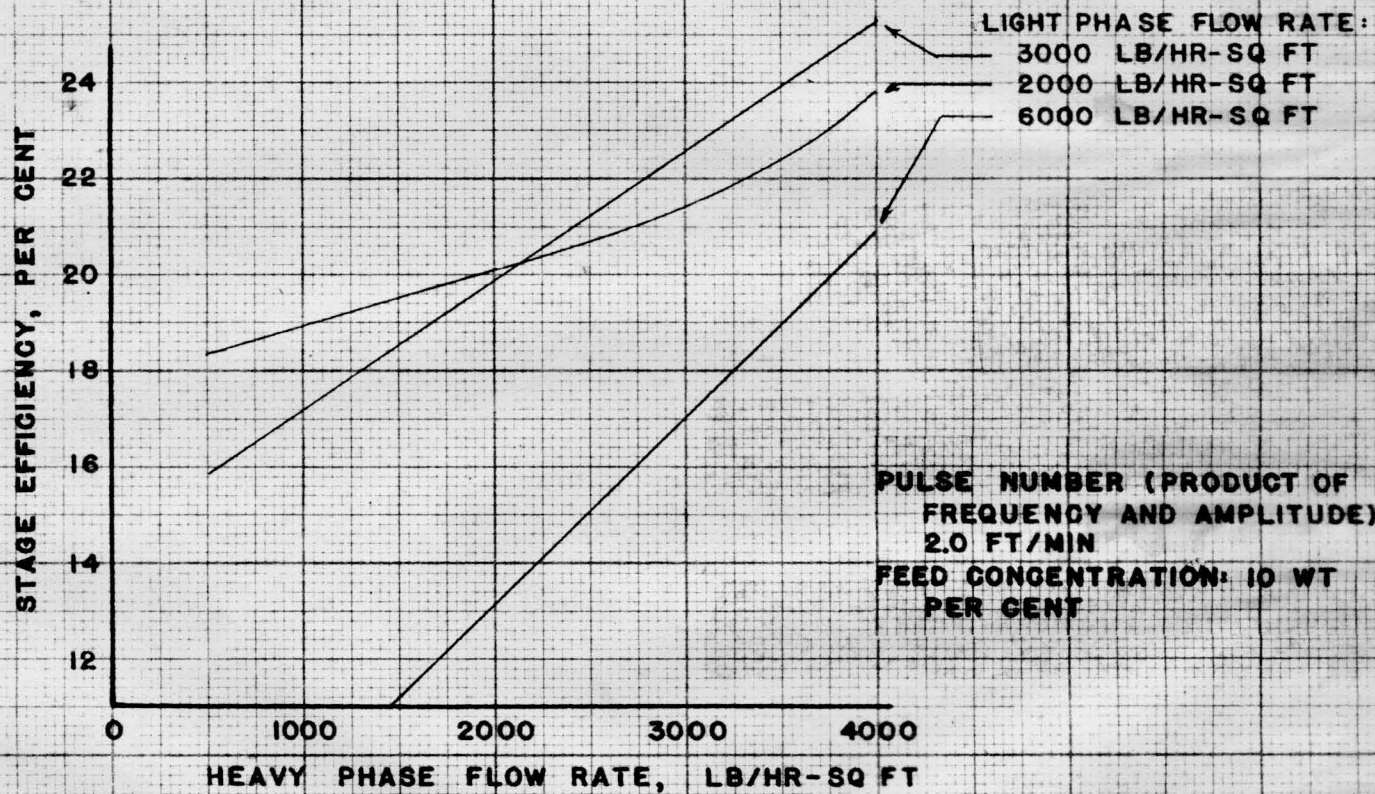


FIGURE III. EFFECT OF THE HEAVY PHASE FLOW RATE ON THE STAGE EFFICIENCY OF A TWO-INCH EXPERIMENTAL PULSE EXTRACTOR EXTRACTING ACETONE FROM CARBON TETRACHLORIDE WITH WATER

limit covered in this experiment, and probably beyond, it is desirable to use as high a heavy-phase flow rate as possible to increase the stage efficiency.

Effect of the Light-Phase Flow Rate on Stage Efficiency.

There was available in the literature no data on the effect of the light-phase flow rate on the stage efficiency in a pulse extractor. For the other agitated extractors mentioned previously, the flow rates are virtually without effect on the stage efficiency.

For this investigation the regression coefficient of the light-phase flow rate on stage efficiency was found to be -0.00302 . This coefficient expresses the change in stage efficiency for a unit change in the light-phase flow rate. This means that an increase in the light-phase flow rate was accompanied by a decrease in the stage efficiency. It was found that there is a probability of 0.999 that the true value of the regression coefficient is not zero. Similarly, there is a probability of 0.80 that the true value of the regression coefficient will lie between -0.00229 and -0.00375 . In a manner analogous to the heavy-phase flow rate, this regression coefficient is modified by the relative flow rate.

The regression coefficient of the relative flow rate on the stage efficiency is -6.93 . This means that a unit change in the relative flow rate causes a change of 6.93 in

the stage efficiency in the opposite direction. The probability that the relative flow rate has an effect on the stage efficiency is 0.992. The interval that has a probability of 0.80 of containing the true value of this regression coefficient is from -4.32 to -9.54.

For a constant pulse number, acetone concentration in the feed, and heavy-phase flow rate the effect of the light-phase flow rate is given by the following equation:

$$y = k - 0.00302x_3 - \frac{n}{x_3}$$

where:

- y = stage efficiency, per cent
- k = a constant
- x_3 = light-phase flow rate, pounds per hour-square foot
- n = 6.93 times the heavy-phase flow rate, pounds per hour-square foot.

Plots of this equation for several different values of the heavy-phase flow rate are shown in Figure 12, page 166. It is seen that an increase in the light-phase flow rate increases the stage efficiency until the value of the heavy-phase flow rate is reached. Above this flow rate, a further increase in the light-phase flow rate decreases the value of the stage efficiency. From this curve it can be seen that no matter what value of the heavy-phase flow rate is used an equal light-phase flow rate should be used for maximum efficiency.

PULSE NUMBER (PRODUCT OF FREQUENCY
AND AMPLITUDE): 2.0 FT/MIN
FEED CONCENTRATION: 10 WT PER CENT

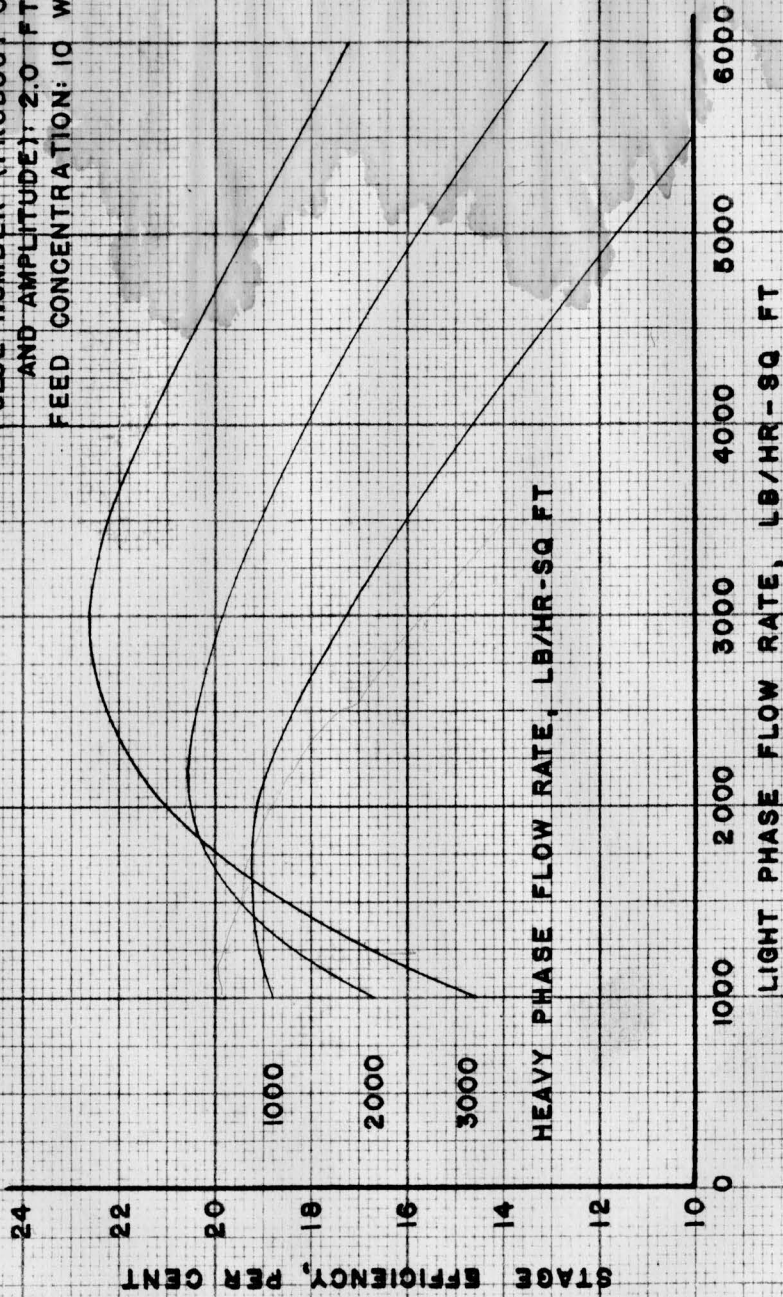


FIGURE 12. EFFECT OF THE LIGHT PHASE FLOW RATE ON THE STAGE EFFICIENCY OF A TWO-INCH EXPERIMENTAL PULSE EXTRACTOR EXTRACTING ACETONE FROM CARBON TETRACHLORIDE WITH WATER

Effect of Acetone Concentration on Stage Efficiency. The effect of the solute concentration in the feed on the stage efficiency has received a great deal of speculation in the literature. Pyle, Colburn and Duffy⁽⁸⁶⁾ stated that an increased solute concentration seemed to increase the stage efficiency. The other variables in their work were not constant enough, however, to state more than that there was an apparent trend. The same effect has been commented on by Feick⁽²⁵⁾ and Sherwood⁽⁵⁹⁾. Both of these authors also point out that the stage efficiency is increased by using a system with high mutual solubility of the solvents. That is, better stage efficiencies would be expected for an extractor using solvents such as water-methyl-ethyl-ketone and water-butanol than for solvents such as water-carbon tetrachloride and water-benzene.

In this investigation the regression coefficients of the acetone concentration in the feed on the stage efficiency was found to be 0.371. That is, if the acetone content of the feed is increased by ten per cent the stage efficiency is increased by 3.71 per cent. The probability was 0.958 that there was a relation between the stage efficiency and the acetone concentration. The limit of values that had a probability of 0.80 of covering the true value of this regression coefficient was 0.171 to 0.571.

Comparison of Stage Efficiency. The only other pulse extractor for which information was available was the 1-1/2-inch pulse extractor located at North Carolina State (50). The only tests performed on this extractor were made at a constant pulse number with varying liquid flow rates. A summary of these tests are presented in Table IV, page 46. The stage efficiency for this extractor varied from 15.7 to 24.2 per cent. This is almost the exact range of stage efficiencies that were found in this investigation. No closer comparison of the results could be made because of the large differences in constructional and operational features of the two extractors.

Recommendations

On the basis of this investigation the following recommendations are made for future studies on pulse extraction.

Elimination of the Pulse Number. During this investigation the product of the pulse amplitude and the pulse frequency was treated as a single variable. It is recommended that additional tests be performed to determine the effect of the pulse amplitude and the pulse frequency on stage efficiency separately. It is recommended that the pulse frequency be studied in steps of five strokes per minute up to 30 strokes per minute. The same range of amplitudes used in this investigation should be used.

Extention of Correlation Equation. The correlation equation derived during this investigation was obtained at relatively low flow rates. It is recommended that additional experimental tests be performed to determine the effect of the flow rates on the stage efficiency over a much wider range. If possible this range should be extended up to the limiting liquid flow rates of approximately 20,000 pounds per hour-square foot for each phase.

Plate Spacing. It is recommended that experimental tests be performed to determine the effect of the plate spacing and the number of plates on the stage efficiency. The optimum plate spacing for the most efficient extraction should be determined; so that the arbitrarily selected plate spacing of four inches can be improved

upon. It is believed that the optimum plate spacing will be less than four inches. Thus, decreasements of 0.5 inch could be made in the plate spacing down to about one inch. If the effect of the number of plates on the stage efficiency were known, it would be possible to extend the experimental data on the pulse extractor to columns of various heights. This amounts to determining the end effects for the pulse extractor.

Plate Design. It is recommended that experimental tests be performed to determine the effect of the plate design on stage efficiency. Two of the more important design features that should be studied are the hole size and the plate free area. It is recommended that hole sizes from $1/64$ to $1/8$ inch in steps of $1/64$ of an inch be investigated. The plate free area should be investigated over as wide a range as the plates can be constructed. Besides the perforated baffles used in this investigation other types of plates should be studied. Two other possibilities are the bubble-cap plate and the Koch baffle.

Packing. It is recommended that experimental tests be performed to determine the effect on the stage efficiency of placing fine wire-mesh packing between the stages and in the disengaging space to induce coalescence. The effect of this packing would probably become increasingly important as the plate spacing is decreased. Alternately, the baffle plates may be removed and replaced entirely by packing. The column would then consist of

alternately packed and unpacked layers. The extreme example of this type of packed extractor is the continually packed pulse column.

Demonstration Systems. It is recommended that as soon as the characteristics of the pulse extractor are adequately determined using the system carbon tetrachloride-acetone-water other demonstration systems should be used. The system used is expected to affect the stage efficiency of the pulse extractor. With a different system a different plate design and arrangement for the most efficient extraction would probably be required. It is recommended that a system be used in which water is the heavy phase. An example of a system of this type that might be used is the system benzene-acetone-water.

Solvent Storage. It is recommended that the available organic-phase storage be increased from the present ten gallons to at least 50 gallons and preferably to 100 gallons. This would allow the extractor to be operated continuously at equivalent flow rates to those used in commercial equipment. For one hour of operation, approximately 400 to 500 pounds of solvent are required at flow rates approaching the flooding velocity (20,000 pounds per hour-square foot).

Feed Composition. It is recommended that the feed be analyzed before the experimental test is started. Thus the feed could be adjusted, if needed, to the desired concentration. This would

eliminate one of the variables affecting the stage efficiency. A feed of about 10 per cent acetone in carbon tetrachloride is recommended for the experimental tests.

Flow Rates. It is recommended that proportioning feed pumps be installed on the equipment. With the use of this type of pump, accurate control of the feed and solvent rates would be possible. The selected pumps should have a capacity of at least two gallons per minute.

Limitations

The limitations placed on experimental work in this investigation are given in the following sections.

Pulse Extractor. For this investigation a vertical two-inch pulse extraction column was constructed. This column contained eight horizontal baffle plates spaced at four-inch intervals. Each plate contained 116 holes $3/32$ of an inch in diameter (2.59 per cent free area) spaced on $1/8$ -inch triangular centers. The column was supported on a pyrex 4 x 2 x 4-inch tee. The side outlet of the tee was closed and used for the introduction of the solvent and the removal of the raffinate phase. The bottom of the tee was connected to a 6-inch syphon bellows. A reciprocating motion was imparted to the bellows by means of a push rod driven by an eccentric cam. No change in the design of the extractor was made during this investigation.

System. The demonstration system used in this investigation was carbon tetrachloride-acetone-water. The direction of transfer of the solute was from the carbon tetrachloride phase into the water phase. The refractive index data for this system used as a means of analysis was determined as a part of this investigation. This data was determined at 22° C, the approximate temperature of the extraction apparatus during the experimental tests.

Variables Studied. Fifteen experimental tests were made. The effect of the pulse number, the heavy-phase flow rate, the light-phase flow rate, and the solute concentration on the stage efficiency was studied. In this investigation the range of each of the variables investigated was as follows: Pulse number (product of pulse amplitude and frequency) from 0.92 to 2.67 feet per minute, heavy-phase flow rate from 393 to 4981 pounds per hour-square foot, light-phase flow rate from 1011 to 7926 pounds per hour-square foot, acetone concentration from 6.26 to 27.84 per cent, and relative (raffinate-phase to extract-phase) flow rate from 0.16 to 1.86. For all of these tests the maximum variation of the stage efficiency was from 12.5 to 25.0 per cent.

Limiting Liquid Flow Rates. Three tests were made to determine the effect of the pulse amplitude and the pulse frequency on the limiting liquid flow rates. The pulse frequency and amplitude used in these tests were 10.5 and 0.25, 10.5 and 0.125, and 21.0 and 0.125 units of strokes per minute and feet per stroke, respectively. The condition of heavy-phase flooding, light-phase flooding, dumping, and blowing were studied.

19
V. CONCLUSIONS

Extraction, using the demonstration system carbon tetrachloride-acetone-water, was performed in a two-inch experimental pulse extractor. The pulse extractor contained eight horizontal baffle plates spaced at four-inch intervals. One hundred sixteen $3/32$ -inch holes (2.59 per cent free area) were drilled in each plate on a $1/8$ -inch triangular lattice. The pulses were applied to the liquids by means of a bellows actuated by a push rod driven by an eccentric cam. Extraction was from the carbon tetrachloride phase into the water phase. This investigation led to the following conclusions.

1. The stage efficiency of the extractor may be correlated by the following equation:

$$y = 12.44 + 3.70x_1 + 0.00503x_2 + 0.00502x_3 + 0.371x_4 - 6.93x_5$$

where:

- y = stage efficiency, per cent
- x_1 = pulse number, feet per minute
- x_2 = carrier solvent flow rate, pounds per hour-square foot
- x_3 = solvent flow rate, pounds per hour-square foot
- x_4 = concentration of acetone in feed, weight per cent
- x_5 = relative flow rate, dimensionless.

The range of variables over which this equation is applicable is as follows: Pulse number from 0.92 to 2.67 feet per minute, heavy-phase flow rate from 393 to 4981 pounds per hour-square foot, light-phase flow rate from 1011 to 7926 pounds per hour-square foot, solute concentration in feed from 6.26 to 27.84 weight per cent, relative flow rate from 0.16 to 1.86, and stage efficiency from 12.5 to 25.0 per cent.

2. The light-phase flooding velocity is $22,000 \pm 1,000$ pounds per hour-square foot. This velocity is independent of the degree of agitation, as shown by an equal flooding velocity for tests performed with a pulse frequency and a pulse amplitude of 10.5 and 0.25, 10.5 and 0.125, and 21.0 and 0.125 units of strokes per minute and inches per stroke, respectively.

3. The heavy-phase flooding velocity is $20,500 \pm 500$ pounds per hour-square foot at a maximum. This velocity is a function of the pulse number. As the pulse number is decreased from 2.6 feet per minute to 1.3 feet per minute the heavy-phase flooding velocity is decreased to $15,500 \pm 500$ pounds per hour-square foot.

4. The refractive index of solutions of acetone in carbon tetrachloride saturated with water may be expressed by the following equation:

$$n_D^{20} = 1.4594 - 0.0016x$$

where:

$$n_D^{20} = \text{refractive index of carbon tetrachloride phase}$$
$$x = \text{acetone concentration, weight per cent.}$$

This equation applies from acetone concentrations of zero to 17.39 per cent.

5. The refractive index of solutions of acetone in water saturated with carbon tetrachloride may be expressed by the following equation:

$$n_D^{20} = 1.3330 + 0.00063y$$

where:

$$n_D^{20} = \text{refractive index of water phase}$$
$$y = \text{acetone concentration, weight per cent.}$$

This equation applies from acetone concentrations of zero to 26.88 per cent.

21
VI. SUMMARY

The purpose of this investigation was to design, construct and operate a liquid-liquid pulse extractor, and to determine the effect of the flow rates and frequency and amplitude of the pulses on extraction efficiency. The limiting liquid flow rates for the extractor were also determined.

An examination of the literature was made of available information on pulse extraction. Although, a great deal of literature was available on the topic of liquid-liquid extraction, detailed information on only one pulse extractor could be found. The results reported on this extractor were comparable with the results obtained during the investigation.

A two-inch experimental pulse-type extractor was constructed. The extractor was equipped with eight baffle plates, each spaced at four-inch intervals. The plates were drilled with 116 holes $3/32$ inch in diameter, 2.59 per cent free area. The liquids were pulsed by means of bellows driven by a push rod, operated from an eccentric cam. For the satisfactory operation of the extractor various auxiliary apparatus were required, important among these were the supporting framework, the push-rod bearing, the reduction train for the cam, the storage carbonyls, and the tubing and valves.

Distribution and refractive-index data were determined for the demonstration system carbon tetrachloride-acetone-water. Fifteen experimental tests were made extracting acetone from carbon tetrachloride with water to determine the effect of the various operating conditions on the stage efficiency. During the experimental tests the following variables were studied: Pulse number (product of the pulse amplitude and the pulse frequency) from 0.92 to 2.67 feet per minute, heavy-phase flow rate from 393 to 4981 pounds per hour-square foot, light-phase flow rate from 1011 to 7926 pounds per hour-square foot, acetone concentration in the feed from 6.26 to 27.84 weight per cent, and relative (ratio raffinate-phase flow rate to extract-phase flow rate) from 0.16 to 1.86.

The experimental results were correlated by the following equation:

$$y = 12.44 + 3.70x_1 + 0.00503x_2 - 0.00302x_3 + 0.371x_4 - 6.93x_5$$

where:

- y = stage efficiency, per cent
- x_1 = pulse number, feet per minute
- x_2 = carrier solvent flow rate, pounds per hour-square foot
- x_3 = solvent flow rate, pounds per hour-square foot
- x_4 = concentration of acetone in feed, weight per cent
- x_5 = relative flow rate, dimensionless.

The average discrepancy between the experimentally determined and calculated stage efficiencies was 9.7 per cent using this equation.

The maximum deviation that was found was 26.0 per cent.

Three tests were made to determine the effect of the pulse amplitude and the pulse frequency on the limiting liquid flow rates. The pulse frequency and amplitude used in these tests were 10.5 and 0.25, 10.5 and 0.125, and 21.0 and 0.125 units of strokes per minute and feet per stroke, respectively. The conditions of heavy-phase flooding, light-phase flooding, dumping, and blowing were studied.

22

VII. BIBLIOGRAPHY

1. Atkins, G. T., Jr. (to Standard Oil Development Co.):
Separation of Aromatic and Paraffin Constituents of
Petroleum Oils, U. S. Patent 2,274,030 (Feb. 24, 1942).
2. Bachman, I.: Tie Lines in Ternary Liquid Systems, Ind. Eng.
Chem., Anal. Ed., 12, 38 (1940).
3. Benedict, M.: Multistage Separation Processes, Trans. Am.
Inst. Chem. Engr., 43, 41 (1947).
4. Blanding, P. H. and J. C. Elgin: Limiting Flow in Liquid-
liquid Extraction Columns, Trans. Am. Inst. Chem. Engr.,
38, 305 (1942).
5. Border, L. E.: Solutizer, A New Principle Applied to Gasoline
Sweetening, Chem. Met. Engr., 47, 776 (1940).
6. Brancher, A. V., T. G. Hunter, and A. W. Nash: Tie Lines in
Two-liquid Phase Systems, Ind. Eng. Chem., Anal. Ed.,
12, 35 (1940).
7. Brown, G. G. et al.: "Unit Operations," pp. 297-302. John
Wiley and Sons, Inc., New York, N. Y., 1950.
8. *ibid*, pp. 302-319.
9. *ibid*, pp. 525-540.

10. Buchman, R. H.: Liquid-liquid Equilibrium Data for the System Carbon-Tetrachloride-Acetone-Water, *Ind. Eng. Chem.*, 44, 2449 (1952).
11. Bull, F. W. and G. J. Colli: Graphical Methods as Applied to Extraction Problems, Virginia Polytechnic Institute, Blacksburg, Virginia, Engineering Experiment Station, Bulletin No 72 (1949).
12. Campbell, S. A.: Distribution in Hydrocarbon Solvent Systems, *Ind. Eng. Chem.*, 40, 103 (1948).
13. Coahran, J. M.: Extraction Apparatus Suitable for Ether Extraction of Acetic Acid from Pyroligneous Acid Liquor, U. S. Patent 1,845,128 (Feb 16, 1932).
14. Cohen, R. M. and G. H. Beyer: Institute for Atomic Research, Iowa State College; Program, Am. Inst. of Chem. Engr. National Meeting, Biloxi, Miss., March 8-11, 1953.
15. Colburn, A. P.: The Simplified Calculation of Diffusional Processes, General Consideration of Two-Film Resistances, *Trans. Am. Inst. Chem. Engr.*, 35, 211 (1939).
16. _____: Simplified Calculation of Diffusional Processes, *Ind. Eng. Chem.*, 33, 459 (1941).
17. _____ and R. L. Pigford: General Theory of Diffusional Operations, "Chemical Engineers' Handbook" (J. H. Perry, Editor), pp. 523-559. McGraw-Hill Book Co., Inc., New York, N. Y., 1950. 3rd ed.

18. Comings, E. W. and S. W. Briggs: Liquid-liquid Extraction in Wetted-wall and Packed Columns, Trans. Am. Inst. Chem. Engr., 38, 143 (1942).
19. Cornish, R. E., R. C. Archibald, Elisabeth A. Murphy, and H. M. Evans: Fractional Distribution between Immiscible Solvents, Ind. Eng. Chem., 26, 397 (1934).
20. Elgin, J. C.: Design and Applications of Liquid-liquid Extraction, Chem. Met. Eng., 49, 110-16 (1942).
21. _____ and H. C. Foust: Countercurrent Flow of Particles Through Moving Continuous Fluids, Ind. Eng. Chem., 42, 1127 (1950).
22. _____ and R. Wynkoop: Solvent Extraction, "Chemical Engineers' Handbook" (J. H. Perry, Editor), pp. 714-718. McGraw-Hill Book Co., Inc., New York, N. Y., 1950. 3rd ed.
23. *ibid*, pp. 729-46.
24. *ibid*, pp. 747-52.
25. Feick, G. and H. M. Anderson: Performance of a Packed Liquid-Liquid Extraction Column Under Controlled Agitation, Ind. Eng. Chem., 44, 404-9 (1952).
26. Glasstone, S.: "Textbook of Physical Chemistry," pp. 790-812. D. Van Nostrand Co., Inc., New York, N. Y., 1946. 2nd ed.
27. Gloyer, S. W.: Furans in Vegetable Oil Refining, Ind. Eng. Chem., 40, 228 (1948).

28. Goos, A. W. and A. A. Reiter: New Products from Wood Carbonization, *Ind. Eng. Chem.*, 38, 132 (1946).
29. Gordon, J. J. and J. H. Zeigler (to Eastman Kodak Co.): Countercurrent Extraction Apparatus, U. S. Patent 2,258,982 (Oct. 14, 1941).
30. Hand, D. B.: The Distribution of a Consolute Liquid between Two Immiscible Phases, *J. Phys. Chem.*, 34, 1961 (1930).
31. Harrington, D. J. (to Standard Oil Development Co.): Tower for Countercurrent Washing of Mixed Naphtha or Similar Operations, U. S. Patent 1,943,822 (Jan. 16, 1934).
32. Hayford, D. A., et al.: Unpublished experimental data gathered under the author's direction, Unit Operations Laboratory, Department of Chemical Engineering, Virginia Polytechnic Institute, Blacksburg, Va., August 1952.
33. Hunter, T. G. and A. W. Nash: The Application of Physicochemical Principles to the Design of Liquid-liquid Contact Equipment, Part I, *J. Soc. Chem. Ind.*, 51, 285T (1932).
34. _____: The Application of Physicochemical Principles to the Design of Liquid-liquid Contact Equipment, Part II, *J. Soc. Chem. Ind.*, 53, 95T (1934).
35. Kenyon, R. L., S. W. Glover, and C. C. Georgian: Solvent Extraction of Vegetable Oils with Purfural, *Ind. Eng. Chem.*, 40, 1162 (1948).

36. Kiersted, W., Jr. (to Texas Co.): Treatment of Hydrocarbon Oils with Solvents such as Furfural, U. S. Patent 2,176,429 (Oct. 17, 1939).
37. King, C. C. and N. L. B. Dickinson (to M. W. Kellogg Co.): Separation of Organic Compounds, U. S. Patent 2,552,564 (May 15, 1951).
39. Koch, F. C.: High Capacity Distilling Tower, U. S. Patent 2,401,569 (June 4, 1946).
40. Maloney, J. O. and A. E. Schubert: The Application of Rectangular Coordinate Methods to Solvent Extraction Design, Trans. Am. Inst. Chem. Engr., 36, 741 (1940).
41. McCloud, D. M.: Personal Communication, April 1, 1953, Oak Ridge, Tenn.
42. McConnell, E. B. (to the Standard Oil Co. (Ohio)): Apparatus for Extracting Petroleum, U. S. Patent 2,091,645 (Aug. 31, 1937).
43. Messner, H. P., C. A. Stokes, C. M. Hunter, and G. M. Harrow: Continuous Countercurrent Dehydration, Ind. Eng. Chem., 36, 917 (1944).
44. Morello, V. S. and N. Poffenberger: Commercial Extraction Equipment, Ind. Eng. Chem., 42, 1021-35 (1950).
45. Murdock, D. G. and M. Cucksey: Removal of Phenol from Gasworks Ammonical Liquors, Trans. Inst. Chem. Engr. (London), 24, 90 (1946).

46. Murphy, H. F.: Personal Communication, March 4, 1953,
Blacksburg, Va.
47. Oldshue, J. Y. and J. H. Rushton: Continuous Extraction in
a Multistage Mixer Column, Chem. Engr. Progress,
48, 297-306 (1952).
48. Othmer, D. F. (to Eastman Kodak Co.): Countercurrent Liquid
Extraction Apparatus Suitable for Extracting Acetic Acid
with Ethyl Acetate, U. S. Patent 2,000,606 (May 7, 1934).
49. _____ and F. E. Tobias: Liquid-liquid Extraction Data,
Ind. Eng. Chem., 34, 690 (1942).
50. Pike, F. P., D. S. Arnold, and C. A. Plank: The Performance
of Contactors for Liquid-liquid Extraction. Unpublished
Progress Report, Department of Engineering Research,
North Carolina State College, Raleigh, N. C., July 1, 1952.
51. Podbielinak, W. J.: Apparatus for the Centrifugal Extraction
of Materials, U. S. Patent 2,003,308 (June 4, 1935).
52. Rogers, M. C. and E. W. Thiele: Bubble-cap Column as a Liquid-
Liquid Contact Apparatus, Ind. Eng. Chem., 29, 529 (1937).
53. Row, S. B., J. H. Koffelt, and J. R. Withrow: Characteristics
and Performance of a Nine-inch Liquid-liquid Extraction
Column, Trans. Am. Inst. Chem. Engr., 37, 559 (1941).
54. Scheibel, E. G.: Fractional Liquid Extraction, Part I, Ind.
Eng. Chem., 44, 681 (1948).

55. Scheibel, E. G.: Fractional Liquid Extraction, Part II, Ind. Eng. Chem., 44, 771 (1948).
56. _____: Extraction Apparatus, U. S. Patent 2,493,265 (Jan. 3, 1950).
57. _____ and A. E. Karr: Semi-commercial Multistage Extraction Column, Ind. Eng. Chem., 42, 1048 (1950).
58. Sherwood, T. K. and R. L. Pigford: "Absorption and Extraction," p. 394. McGraw-Hill Book Co., Inc., New York, N. Y., 1952. 2nd ed.
59. *ibid*, p. 441.
60. *ibid*, p. 438.
61. Skrzec, A. E.: Mass Transfer Studies in Countercurrent Liquid-Liquid Extraction Systems. Unpublished Ph. D. Thesis, Library, Virginia Polytechnic Institute, Blacksburg, Va., (1952).
62. Smith, A. S.: Solutropes, Ind. Eng. Chem., 42, 1206 (1950).
63. Standard Oil Development Co.: Apparatus for the Countercurrent Contacting of Liquids of Different Gravities, Brit. Patent 584,892 (Feb. 5, 1947).
64. Sutherland, D. G.: The Extraction of Acetone from an Acetone-Water Mixture in a Vertical Spray Type Column Using Trichloroethane as the Extracting Solvent. Unpublished B. S. Thesis, Library, Virginia Polytechnic Institute, Blacksburg, Va.

65. Thompson, D.: Personal Communication, Jan. 14, 1953,
Blacksburg, Va.
66. Treybal, R. E.: Solvent Extraction, Ind. Eng. Chem., 44,
53-63 (1952).
67. _____: Solvent Extraction, Ind. Eng. Chem., 45,
58-67 (1953).
68. _____: "Liquid Extraction," pp 1-4. McGraw-Hill
Book Co., Inc., New York, N. Y. 1951.
69. *ibid*, p. 38.
70. *ibid*, pp. 89-91.
71. *ibid*, pp. 125-202.
72. *ibid*, pp. 257-89.
73. *ibid*, pp. 290-342.
74. _____ and L. T. Works: Countercurrent Liquid-liquid
Extraction in a Wetted-wall Tower, Trans. Am. Inst. Chem.
Engr., 38, 203 (1942).
75. Van Dijk, W. J. D.: Process and Apparatus for Intimately
Contacting Fluids, U. S. Patent 2,011,186 (Aug. 13, 1935).
76. Varteressian, K. A. and M. R. Fenske: Liquid-liquid Extraction,
Ind. Eng. Chem., 28, 1353 (1936).
77. _____: Graphical Design Methods
Applied to the Fractional Extraction of Methylcyclohexane
and n-Heptane Mixtures, Ind. Eng. Chem., 29, 270 (1937).

78. Vold, R. E. and M. J. Vold: *Physical Methods of Organic Chemistry*, pp. 297-326. Interscience Publishers, Inc., New York, N. Y., 1949, 2nd ed.
79. Wen Hon Li: *Liquid-Liquid Extraction in a Pulsed, Perforated Plate Column*. Unpublished Ph. D. Thesis, Georgia Institute of Technology, Atlanta, Ga.; original unavailable.

Addenda

80. Anderson, R. L. and T. A. Bancroft: "Statistical Theory in Research," p. 168-172. McGraw-Hill Book Co., Inc., New York, N. Y. 1952.
81. *ibid*, pp. 196-203.
82. Bauer, N.: Determination of Density, "Physical Methods of Organic Chemistry" (A. Weissberger, Editor) p.253. Interscience Publishers Inc., New York, N. Y. 1949. 2nd ed.
83. _____ and K. Fajans: Refractometry, "Physical Methods of Organic Chemistry" (A. Weissberger, Editor) p. 653. Interscience Publishers Inc., New York, N. Y. 1949. 2nd ed.
84. Davies, O. L.: "Statistical Methods in Research and Production," p. 270. Oliver and Boyd, Edinburgh, England 1949. 2nd ed.
85. Duncan, D. B.: Personal communication, April 20, 1953, Blacksburg, Virginia.
86. Pyle, C., A. P. Colburn, and H. R. Duffy: Factors Controlling Efficiency and Capacity of Sieve-plate Extraction Towers, *Ind. Eng. Chem.*, 42, 1042 (1950).

16

VIII. ACKNOWLEDGMENTS

The author wishes to express his deepest gratitude to Dr. P. H. Watkins for his limitless patience and guidance throughout the course of this investigation. The author wishes, further, to express his sincere appreciation to his associate, Mr. S. H. Roberts, for his aid in the performance of this investigation.

The author also wishes to thank Professor Fred W. Bull who suggested the topic and who helped in the design and construction, Dr. D. B. Duncan of the Statistics Department who aided in the statistical analysis of the data, and Mr. M. A. Price who helped in the construction of the pulse extractor. The author thanks the entire staff and graduate body of the Department of Chemical Engineering for their helpful comments and criticisms.

Finally, the author wishes to acknowledge his deepest debt to his wife whose encouragement, understanding, and typing made this paper possible.

**The vita has been removed from
the scanned document**