To the Graduate Council:

I am submitting herewith a thesis written by John Mark Williams entitled "The Solar Powered Distillation of an Ethanol-Water Mixture." I recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Chemical Engineering.

J. J. Perona

J. J. Perona, Major Professor

We have read this thesis
and recommend its acceptance:

Charles F. Moore

Edward S. Clark

Accepted for the Council:

L. Evans Roth

Vice Chancellor
Graduate Studies and Research

THE SOLAR POWERED DISTILLATION
OF AN ETHANOL-WATER MIXTURE

A Thesis

Presented for the

Master of Science

Degree

The University of Tennessee, Knoxville

John Mark Williams

December 1980

ACKNOWLEDGEMENTS

The author appreciates the patience and guidance of his research advisor, Dr. J. J. Perona. The author is also grateful to Dr. H. F. Johnson and the Department of Chemical and Metallurgical Engineering at The University of Tennessee for financial aid in the form of a graduate assistantship.

Sincere thanks are extended to Dr. J. M. Schreyer for his direction and encouragement, and to Mr. V. A. Emert for his help in constructing the equipment. Finally, the author wishes to acknowledge his loving wife who continually gave to him her support throughout the work.

ABSTRACT

The purpose of this study was to determine the H.T.U. under different operating conditions for a packed column operating at less than atmospheric pressure and to evaluate the viability of using one form of solar energy to provide the heat input to the reboiler for such a system. The majority of the experimental effort was focused on determining the effect that the operating pressure and steady state reboiler and distillate concentrations had on the height of a transfer unit.

Data obtained from the experiments indicate that the $(H.T.U.)_{OG}$ decreased for an increase in either the operating pressure or the initial feed concentration. The values of the $(H.T.U.)_{OG}$ obtained in this investigation varied from 0.71 feet to 1.24 feet for a packed height of 5.5 feet in an 8-inch internal diameter tower using 0.25-inch Intalox saddles at total reflux.

The unique nature of the heat source used in this investigation required that the column be operated at less than atmospheric pressure. Two of the difficulties that were encountered when performing a vacuum distillation were: (1) significant product withdrawal by the vaccum pump when small air leaks existed and (2) air pockets preventing the transfer of fluids due to the location and type of valves located throughout the system.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

NOMENCLATURE

a = surface area of packing per unit of packed volume, sq. ft. per cu. ft.

d = differential operator

G = superficial vapor rate, lb. moles/(hr.)(sq. ft.)

H.T.U. = height of a transfer unit

K_G = overall gas mass transfer coefficient, lb. moles per hour per sq. ft. (mole fraction)

L = superficial liquid rate, lb. moles/(hr.)(sq. ft.)

m = slope of equilibrium curve

N = number of transfer units

x = mole fraction of diffusing component in liquid

x* = mole fraction of diffusing component in liquid in equilibrium with a vapor of mole fraction, y

y = mole fraction of diffusing component in gas

y* = mole fraction of diffusing component in equilibrium with a liquid of mole fraction x

Z = height of column, or packed section, ft.

Subscripts:

1 and 2 = terminal conditions

G = gas

L = liquid

OG = overall transfer measured in gas units

OL = overall transfer measured in liquid units

p = product

w = waste

CHAPTER I

INTRODUCTION

Increasing dependence on petroleum imports from politically unstable regions of the world has led to a transition in our economy which includes the use of alternative fuel sources. Fuel grade ethanol, produced from agricultural feedstocks, is rapidly becoming economically competitive with petroleum-derived fuels. The distillation process for the separation of this fuel from a dilute aqueous solution is very energy intensive. It has been argued, with some success, that more energy is required to produce a quantity of alcohol than the alcohol contains.

One of the possible ways to make the overall energy balance more attractive is to use solar energy as the heat source for the distillation process. When conventional solar panels are used, the relatively low temperatures produced require that the distillation be carried out under less than atmospheric pressure. One of the advantages of performing a vacuum distillation is that the azeotrope is raised to a level where it may not affect the distillation process.

The degree of separation that will be achieved in such a distillation will be a function of the following: (1) liquid and vapor rates (mass velocity); (2) reflux ratio; (3) shape, size, and material of tower packing; (4) degree of wetting of packing; (5) packed height;

(6) ratio of tower diameter to packing size; (7) effectiveness of liquid distribution over tower area; (8) operating pressure; and (9) physical properties of materials being fractionated. These, in turn, will depend on the quantity of heat available and the amount of product desired at a specified purity. The United States Department of Energy wished to determine if a solar-powered distillation is possible and, if so, its operational characteristics.

I. PREVIOUS WORK

Equilibria for an ethanol-water system at the desired operating pressures was determined by Bebe, Coulter, Lindsay, and Baker, et al.[1] Information enabling the estimation of the height of a transfer unit (H.T.U.) was scarce. Colburn and Pigford[2] offered suggestions relating liquid and gas velocities, temperature, composition, pressure, reflux ratio, packing, and column dimensions to the height of a transfer unit.

Using a packing material similar to that chosen for this investigation, Furnas and Taylor[3] found that the height of a transfer unit (H.T.U.) varied from 1.65 to 13.3 feet for an ethanol-water system operated at atmospheric pressure. Because of the inherent transients to be expected when using solar energy as the heat source for a reboiler, the H.T.U.'s for different operating conditions were central to this investigation.

II. PURPOSE OF THE INVESTIGATION

The experimentation of this investigation was designed to determine the H.T.U. under different operating conditions for a packed column operating at less than atmospheric pressure and to evaluate the viability of using one form of solar energy to provide the heat input to the reboiler for such a system.

Where appropriate, predicted values were compared with experimental results. The purpose of this work was to contribute to the body of knowledge concerning the prediction and evaluation of the H.T.U. for a packed column operating at less than atmospheric pressure under different operating conditions.

III. SCOPE OF THE INVESTIGATION

The distillation of an ethanol-water solution in a packed column operated at less than atmospheric pressure was performed, and the determination of the H.T.U. under different operating conditions was evaluated. This included the design, purchase, and, where necessary, the fabrication of all equipment to perform the desired experiments. The fabrication and operation of the equipment was conducted at Oak Ridge National Laboratory to facilitate the construction and evaluation of the overall system as a producer of fuel-grade ethanol.

CHAPTER II

THEORY

I. HEIGHT OF A TRANSFER UNIT

The mode of operation for a packed tower used for distillation is characterized by a rising vapor stream passing counter-currently against a falling liquid reflux stream. At any point in the tower the concentration of the lower boiling component in the vapor at the vapor-liquid interface is greater than that in the rising vapor stream. The resultant diffusion of the lower boiling component into the vapor stream causes the concentration of the lower boiling component to increase continuously as the vapor passes upward, rather than in steps as in a plate column.

The quantity of the lower boiling component that is transferred to the vapor is G dy and may be described, using overall mass transfer coefficients, as

$$G\,dy = K_G a(y^{*}-y)dZ \qquad (1)$$

where $y^{*}-y$ is the overall gas phase concentration driving force. Equation (1) may be rearranged to read

$$\frac{dy}{y^{*}-y} = \frac{K_G a dZ}{G} \qquad (2)$$

where the left-hand portion is a measure of the difficulty of the separation. Chilton and Colburn[4] proposed that this term in its integrated form represents the number of overall gas transfer units N_{OG}.

$$N_{OG} = \int_{Y_1}^{Y_2} \frac{dy}{y^*-y} \quad . \tag{3}$$

The Height of a Transfer Unit (H.T.U.) is then expressed as the height of the tower divided by the number of transfer units:

$$(H.T.U.)_{OG} = \frac{Z}{N_{OG}} = \frac{Z}{\int_{Y_1}^{Y_2} \frac{dy}{(y^*-y)}} \quad . \tag{4}$$

Equation (4) provides a means of obtaining a true measure of a finite separation while operating in infinitesimal steps. It also allows the evaluation of the mass transfer coefficient K_Ga, which can be compared to that obtained in other operations such as gas absorption.

The fundamental relations involved in the H.T.U.'s have been presented by Colburn.[5]

$$(H.T.U.)_{OG} = \frac{Z}{\int_{y_1}^{y_2} \frac{dy}{y^*-y}} \tag{5}$$

$$(H.T.U.)_{OL} = \frac{Z}{\int_{x_1}^{x_2} \frac{dx}{x-x^*}} \tag{6}$$

II. PROCESS CONTROL

Although the equipment investigated was operated at total reflux, the area of process control will be briefly addressed in order to facilitate any future investigations on equipment of this type.

The number of variables that can be controlled in the operation of the equipment investigated is limited by the necessity to satisfy the laws of conservation of mass and energy and the conditions of thermodynamic equilibrium between the phases. If the distillation column is to be operated at a constant pressure with two product streams, two independent compositions can be specified. If the column is also desired to be operated with the minimum amount of heat input for a given separation, then the feed location will be determined by the number of transfer units and the feed composition. The variables chosen to regulate will be determined by the specifications of the products and the nature of the input disturbance.

The three classes of input disturbances that may affect the control of a distillation column are changes in (1) feed (composition, rate, thermal quality), (2) service supplies such as cooling water or steam, and (3) the required output stream concentrations or flow rates.[6] Unlike most distillation columns, the equipment investigated falls into the second category due to the fluctuations of heat input to the reboiler.

A common method of composition control is by the inferential measurement of either temperature or vapor pressure. The unique relationship between temperature and composition for a binary mixture allows a temperature detector to be normally placed in the top of the column, providing an indirect measurement for changes in composition. As can be seen from the equilibrium data shown in Appendix A, the variation in temperature at high ethanol concentrations is minimal, unlike that found at low concentrations. This would suggest the possibility of locating a temperature detector in the lower section of the distillation tower.

CHAPTER III

EXPERIMENTAL APPARATUS

A discussion of the equipment used in this study can be divided into two major categories: (1) equipment development, and (2) details of equipment. The first section of this chapter describes the problems that arose in the implementation of the original design and the resultant modifications; the second section describes the equipment that was used in this investigation.

I. EQUIPMENT DEVELOPMENT

The development of an operational system required modifications and refinements to be performed in three areas: (1) a proportional flow controller; (2) valve placement; and (3) air leakage.

Proportional Flow Controller

During the early stages of development, a proportional flow controller was located beneath the condenser with the desire to maintain a constant reflux ratio regardless of the quantity of condensate. Problems in its performance, as well as those in other areas, led to its eventual replacement with a reflux drum and to the operation of the system at total reflux. The main problems in its successful operation are briefly discussed in order that future attempts to use this type of equipment may avoid the same problem areas. Schematics of this piece of equipment are shown in Figures 1 and 2 as originally developed by Wilkinson and Beatty.[6]

ORNL-DWG 80-11911

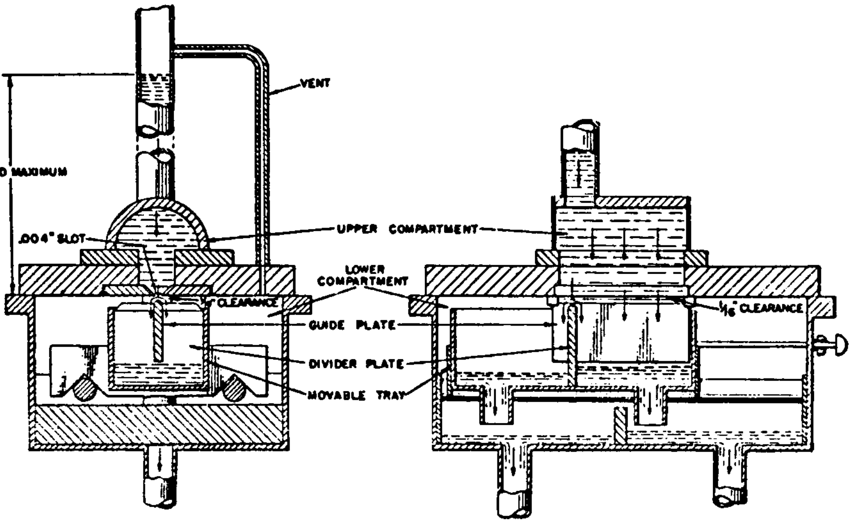

FIGURE 1

CROSS SECTION OF FLOW CONTROLLER

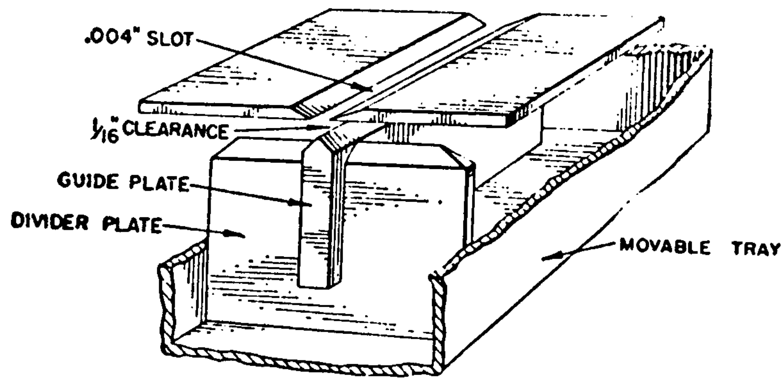

FIGURE 2

FLOW CONTROLLER

Aluminum was chosen as the material of construction because of its relatively soft nature, making it easy to fabricate. After several weeks of testing, a loosely held granular substance began forming on the inner surfaces of the flow controller. Small amounts of this corrosion would routinely break loose from the uppermost surfaces and block a segment of the thin slot through which all of the liquid passed. This blockage would alter the relative flow rates of the two bottom streams or occasionally stop one of them entirely.

The guide plate was intended to collect the descending liquid as a uniform falling film. In practice the alcohol-water solution tended to form irregular rivulets which changed with the flow rate, causing unpredictable results. Because of the previously mentioned corrosion that was occurring, the aluminum guide plate was replaced with one constructed of polished stainless steel which, along with changes in the clearances, vastly improved the sheeting action at all but low flow rates.

Due to the numerous seams in this piece of equipment and the high pressure differential encountered due to the vacuum operation of the system, air leaks were a significant problem. These leaks did not affect the flow controller's operation but had to be sealed in order to prevent problems arising in other areas.

Valve Placement

At the same time the proportional flow controller was replaced with a reflux drum, needle valves were located immediately below it in an attempt to manually control the reflux ratio. This location and the type of valve chosen led to the formation of air pockets in the lines immediately beneath their location. Because the condensate was returned by gravity to the column, any air pockets effectively prevented flow through the reflux line. By relocating the valves and using gate valves when possible, the air pockets were eliminated.

Air Leakage

After the initial hours of the system's operation, an ethanol-water solution was found to have contaminated the oil in the vacuum pump. Under conditions of high vacuum operation, the rate of condensate loss was significant. A cold trap was built and installed on the reflux drum at the point where the vacuum was applied to the system, but proved to be ineffective because of the low gas density. The oil-filled vacuum pump was replaced with a vane-type centrifugal pump, but the alcohol was found to leach the binder out of the carbon vanes, causing the pump to have a major breakdown. The next pump chosen was a dual metal bellows pump constructed of stainless steel which performed without any additional problems. Because a significant quantity of ethanol-water vapor was still being lost through the vacuum pump, a small shell-and-tube condenser was located on the positive side of the pump where the vapor could be easily condensed at atmospheric pressure.

The experiments that were conducted on the equipment investigated assumed steady state operating conditions. The rate of condensate that was being removed by the vacuum pump was significant enough so that the validity of any measurements assuming steady state behavior could be questioned. In order to eliminate any condensate being removed by the vacuum pump, an airtight system would have been required. A pressure of approximately 30 pounds per square inch gauge was placed on the system, the insulation removed, and all joints and valves thoroughly checked for leaks. This procedure lowered the air leakage rate to a level where the amount of ethanol-water solution withdrawn by the vacuum pump did not significantly affect calculations based on steady state operation.

II. EQUIPMENT DESCRIPTION

A commercially available Freon-loaded solar collector system, developed by Dr. J. M. Schreyer of the Oak Ridge National Laboratory, was used to provide the heat to the reboiler. A schematic diagram of the apparatus is shown in Figure 3, and a photograph of the equipment is shown in Figure 4. Operating temperatures of 200°F have been recorded using these panels, but more often a temperature range from 120-160°F is encountered. When the column was not operated, the hot Freon gas was routed to a heat exchanger where hot water was produced for later usage or, if desired, passed through a space heater.

The tower used in this apparatus has an 8-inch internal diameter and was packed to a depth of 5.5 feet. Stainless steel was the material of construction because of its availability and resistance to corrosion. The uninsulated tower is shown in Figure 5 with one of the liquid distributors being displayed. Figure 6 shows the construction

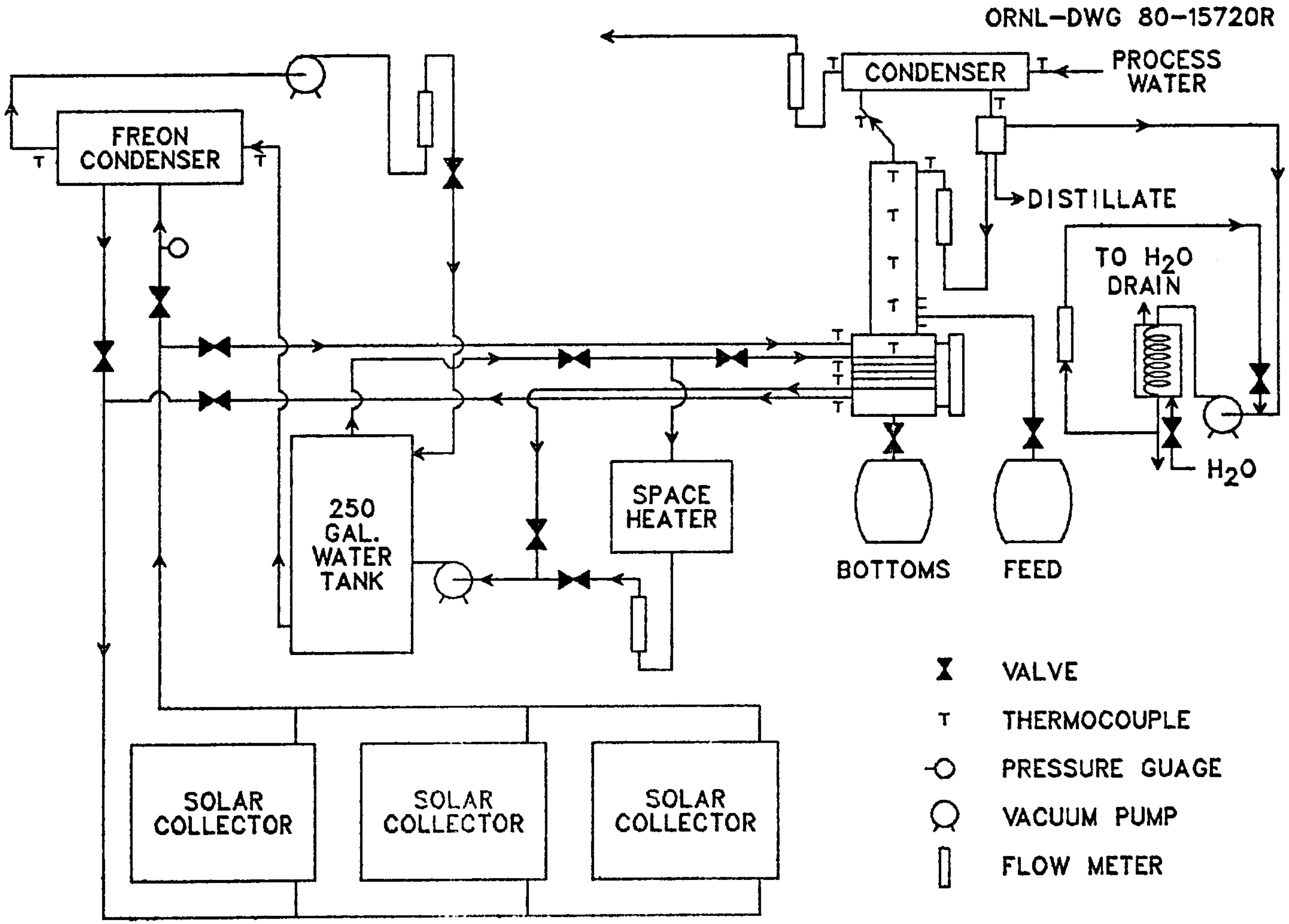

FIGURE 3

FLOW AND EQUIPMENT SCHEMATIC

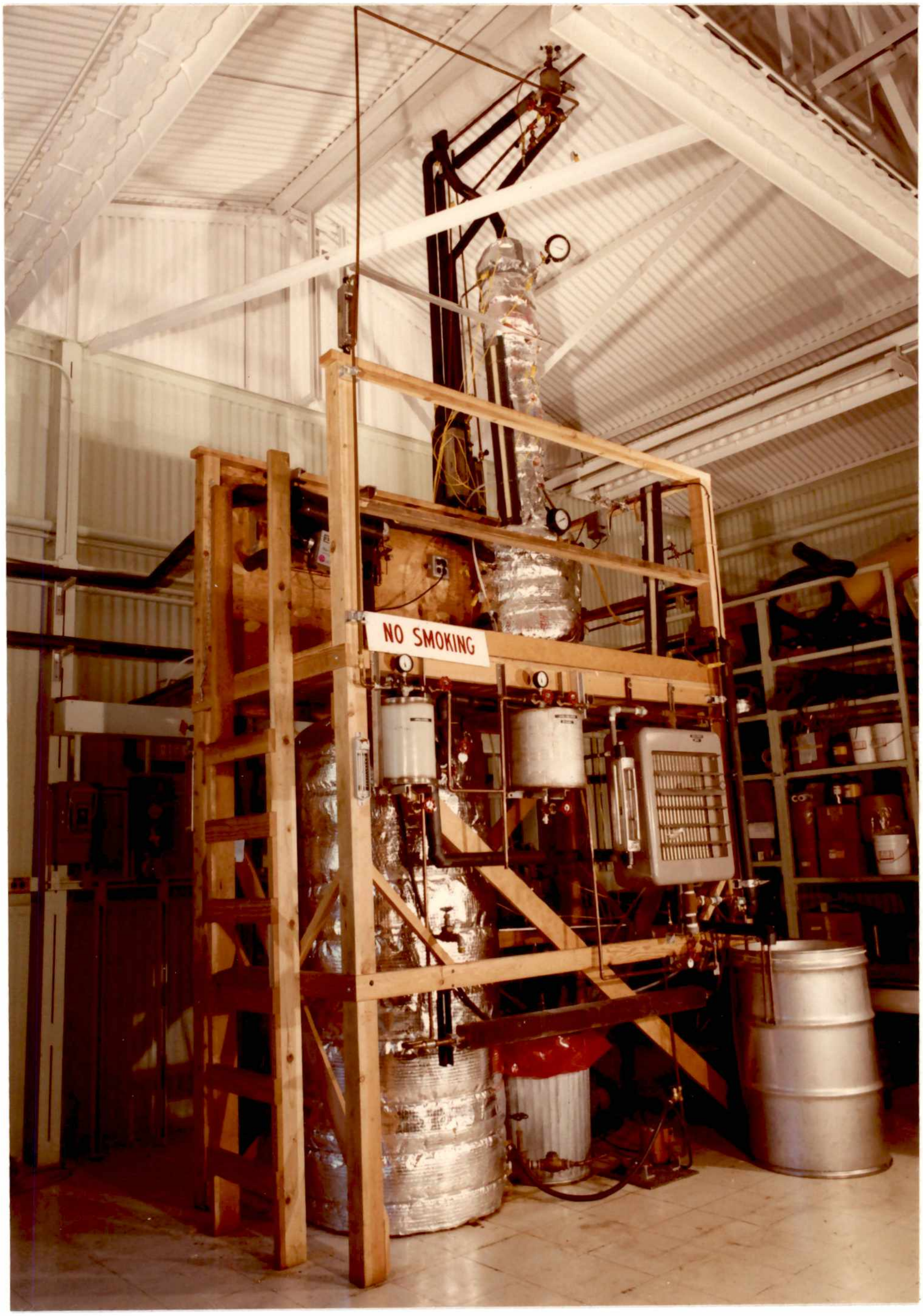

FIGURE 4

OVERVIEW OF COMPLETED FACILITY

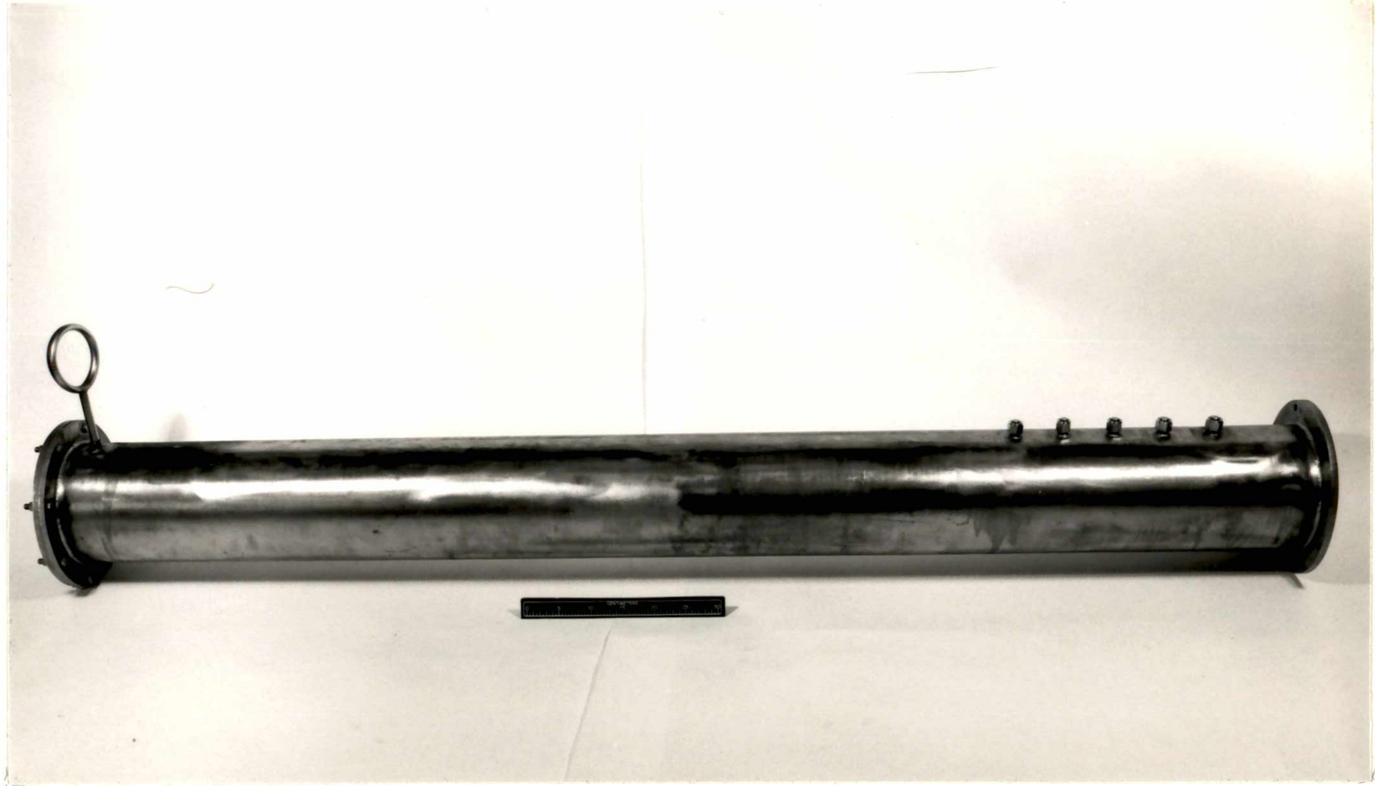

FIGURE 5

DISTILLATION TOWER

ORNL-DWG 80-17387

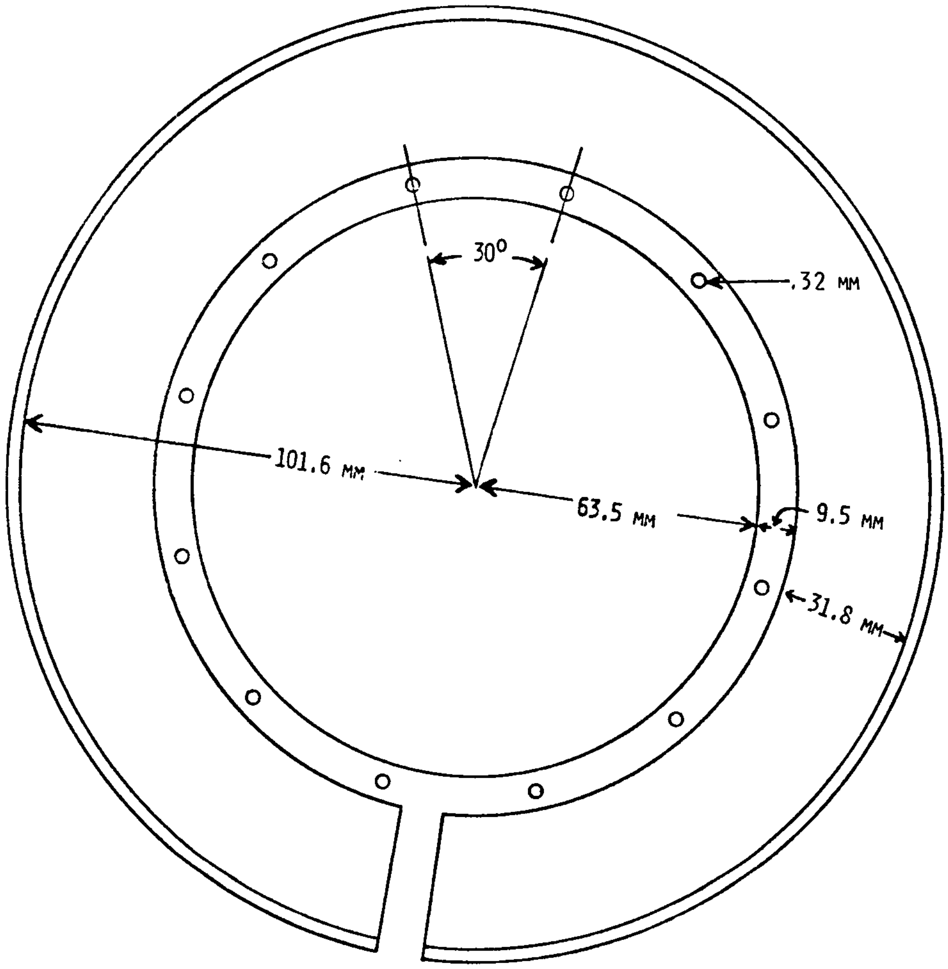

FIGURE 6

REFLUX DISTRIBUTOR

of the reflux distributor, which was also fabricated from stainless steel. A commercial packing of 0.25-inch ceramic Intalox saddles was chosen because of its high surface area per unit volume. Pressure gauges, flowmeters and thermocouples were located throughout the system, as shown in Figure 3, in order to facilitate system operation and data evaluation.

One of the design goals was that the reboiler be as small as possible in order to minimize the start-up time but still be able to contain a heat exchanger with sufficient surface area. A constraint in the design was that all Freon condensing internally be able to flow by gravity back to the collectors. In order to maximize the efficiency of the collectors, a low temperature across the heat exchanger was also desired. All of these constraints led to the design and construction of the reboiler shown in Figure 7 which is also shown unassembled in Figure 8. It consisted of approximately 350 feet of 0.5-inch internal diameter copper tubing wound in a series of pancake coils and joined together with short pieces of tubing. A plug was inserted in the center of the coils to reduce the amount of liquid held by the reboiler to 19.7 liters.

The solar collector system consisted of 12 separate collectors housed in three insulated aluminum cases. Each collector had an area of 10 square feet for a combined surface area of 120 square feet. The collectors were constructed using a steel panel with steel tubes attached through which the liquid Freon flowed that extracted heat from the collector plate. The face of each collector was covered by an aluminum frame over which an envelope of 0.1-millimeter du Pont Tedlar

ORNL-DWG 80-17388

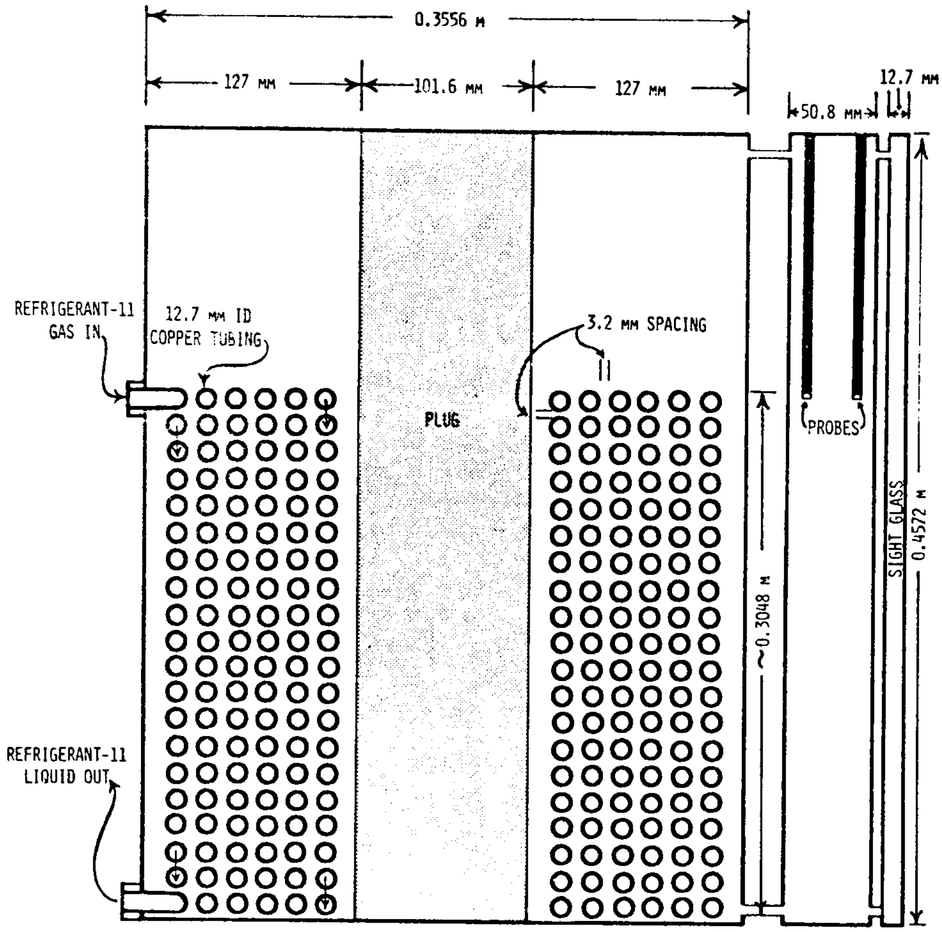

FIGURE 7

SCHEMATIC OF REBOILER

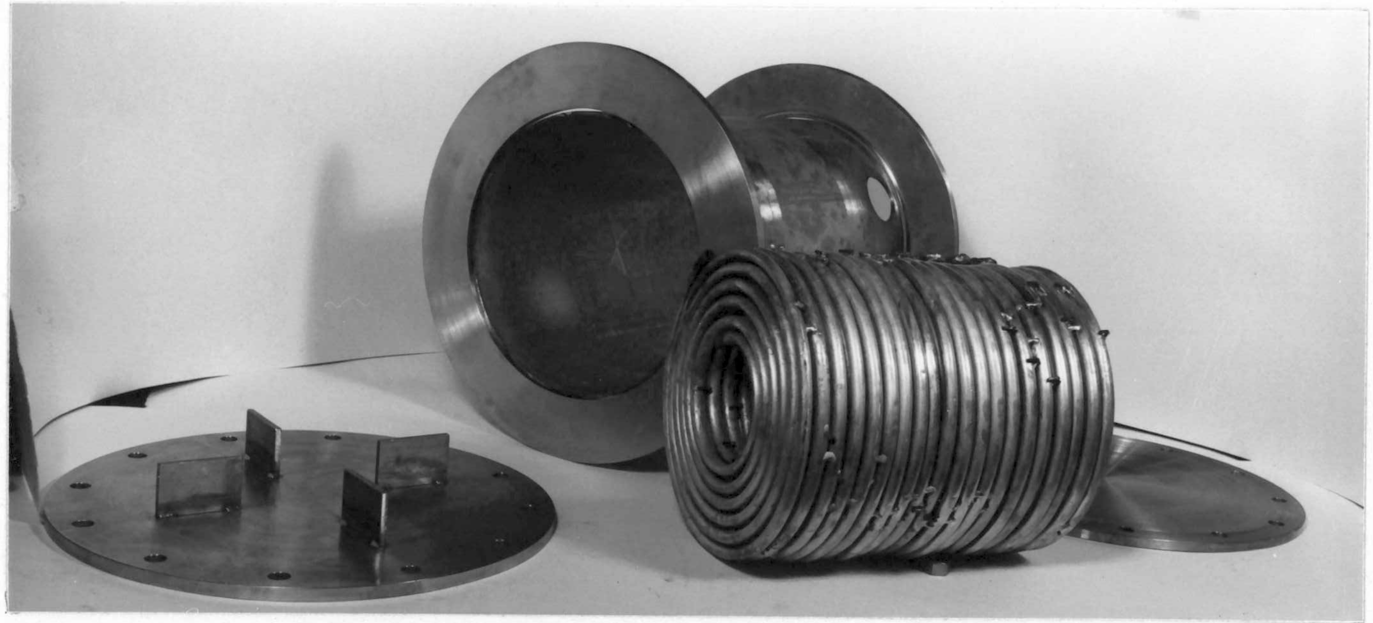

FIGURE 8

UNASSEMBLED REBOILER

(polyvinyl fluoride) was thermally shrunk to provide double glazing. Panels of this type have been shown to provide an average of 200 Btu/hr-ft^2 when operated at relatively low temperatures (100-110°F).

The solar collector system, shown in Figure 9, was separated from the reboiler by approximately 55 feet of one-inch piping. Heat losses were reduced by the use of three inches of fiber glass on all external piping, one inch of foam rubber on internal piping, and approximately six inches of fiber glass on the distillation tower and reboiler. The collectors were installed facing due south at a tilt from the horizontal of 60°.

The majority of the equipment described in this section was located on a platform approximately seven feet above the floor. Due to the fact that an access road was located between the solar panels and the building in which the equipment was located, the piping which brought in the vaporized Freon was required to have a specified ground clearance. The Freon condensers had to be located above these pipes in order for the liquid Freon to flow by gravity back to the collectors. These requirements led to the equipment being located as shown in Figure 4, page 14.

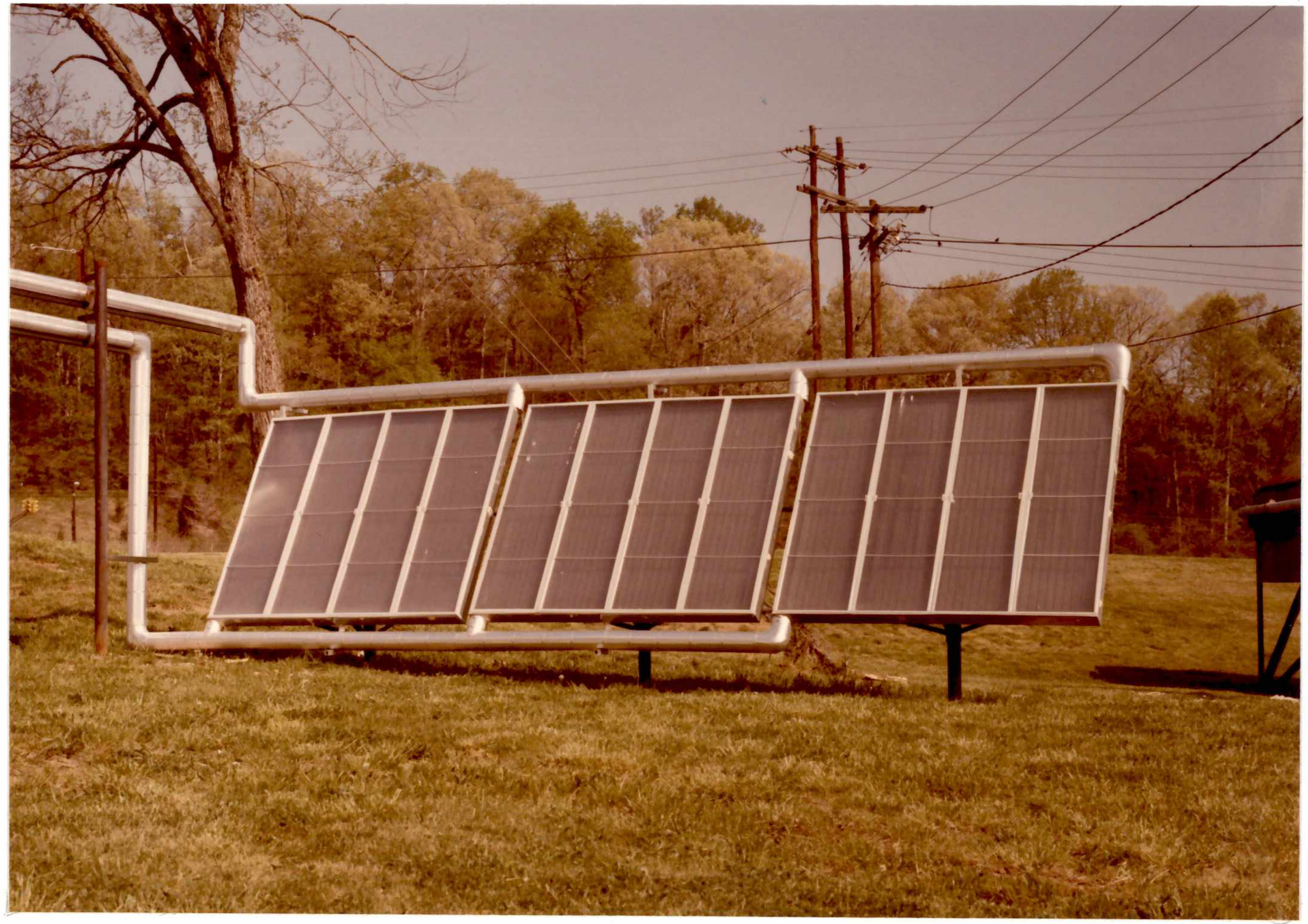

FIGURE 9

SOLAR COLLECTORS

CHAPTER IV

EXPERIMENTAL PROCEDURE

The experimental procedure used in this study can be broken down into four major categories: (1) operation of equipment; (2) calculation of the H.T.U.; (3) mass balance on the system's operation; and (4) energy requirements of the system's operation.

I. OPERATION OF EQUIPMENT

The primary steps involved in the operation of the equipment investigated were (1) feed and equipment preparation; (2) analysis of system's operation; and (3) sample collection and analysis procedure.

Feed and Equipment Preparation

The ethanol-water feed for the system's operation was prepared by combining the required quantities of distilled water and 190 proof alcohol in order to produce a 19.2 liter mixture of the desired concentration. For each run the solution was analyzed immediately before its introduction to the system. The ethanol concentration of the feed and of all other solutions used in this investigation was measured by means of hydrometers which were accurate to a specific gravity reading of ± 0.0005. The concentration-density data used was taken from Chemical Engineers' Handbook, 5th ed. (New York: McGraw-Hill, 1973).

The preparation of the equipment for each run began by allowing process water to pass through the condenser at the relatively high flow rate of 15.8 l/min. A high flow rate was desired so that the condensate would have a low vapor pressure, in order to maintain a low ethanol concentration of any gases withdrawn from the system by the vacuum pump. Cooling water was then allowed to pass through a shell-and-tube condenser located on the exhaust side of the vacuum pump. This condensed, at atmospheric pressure, any ethanol removed from the system by the vacuum pump. Next the vacuum pump was turned on and the bleed valve adjusted so that the desired operating pressure of the system was maintained. Figure 10 is a schematic of the vacuum pump, condenser and piping that was used to control the operating pressure of the equipment investigated.

The ethanol-water solution was allowed to enter the system at that time (through the feed distributor) until the coils inside the reboiler were fully covered. Valves on the Freon lines were adjusted so that all of the vaporized Freon would pass directly from the solar collectors to the heat exchanger inside the reboiler, and the condensed Freon returned directly to the collectors.

Analysis of System's Operation

The flowmeter located on the reflux line and thermocouples located throughout the system indicated when the first overhead product was condensed and returned to the column as reflux. At that time and at approximate 30 minute intervals during the remainder of the experimental run, data were recorded from flowmeters, pressure gauges and thermocouples in order to determine the approximate steady state operating conditions for each experimental run. Samples were taken of the distillate and from the reboiler immediately after the data

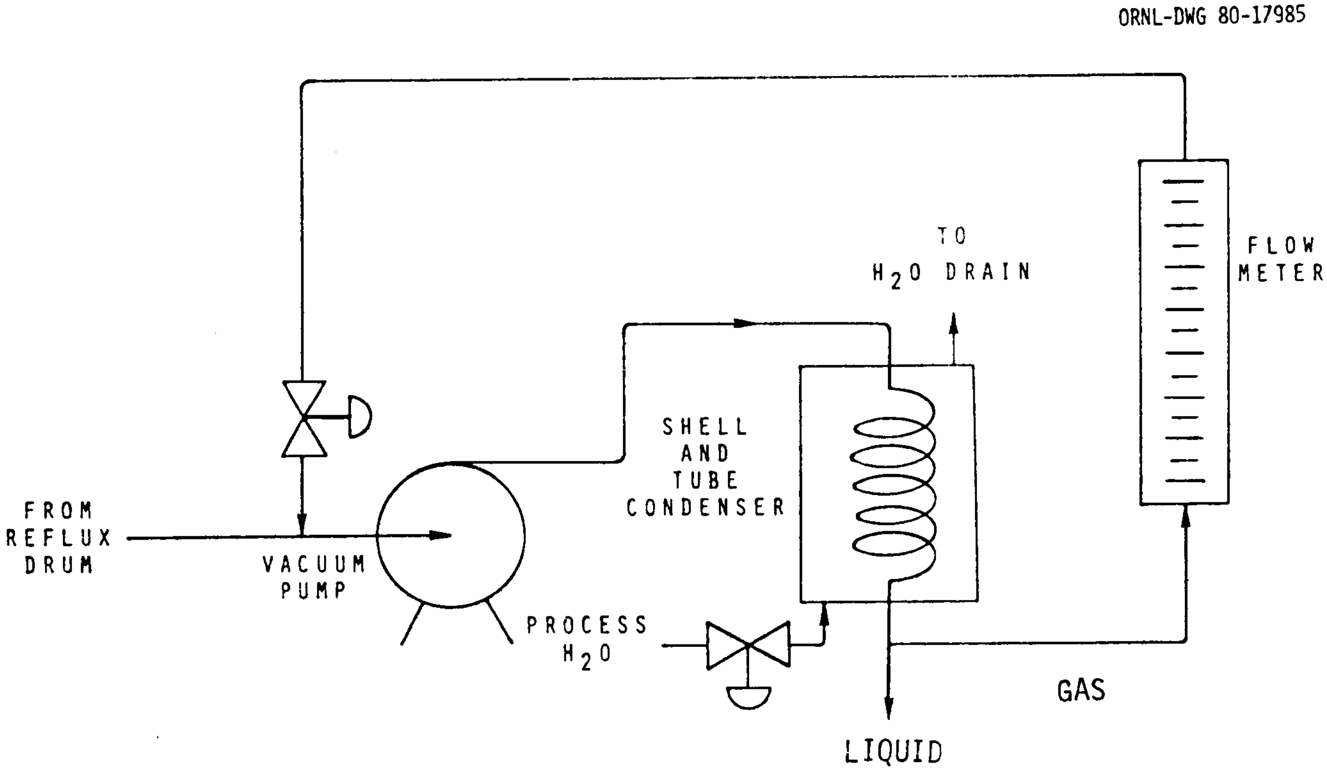

FIGURE 10

SCHEMATIC OF THE VACUUM SYSTEM

observations in order to determine the respective ethanol concentrations. This process was continued until the ethanol concentrations appeared to have reached steady state values.

Sample Collection and Analysis Procedure

At approximate 30 minute intervals, 300 milliliter samples were withdrawn from the reflux drum and reboiler. As Figure 11 indicates, a receiving flask was inserted under the reflux drum and the pressure inside the flask allowed to come to equilibrium with that of the operational system. A valve on the reflux line was then closed so that the condensate was temporarily routed to the receiving flask. After approximately one minute, when sufficient sample had been collected, the valve on the reflux line was opened and the condensate returned to the column.

Samples taken from the reboiler were removed by using a separate vacuum source lower in pressure than that of the operating system. Figure 12 shows the equipment that was used and the location of the sample probe used to withdraw the liquid from the reboiler. Samples withdrawn from the reflux drum varied in temperature from 20°C to 25°C and were analyzed immediately, whereas samples withdrawn from the reboiler varied from 44°C to 73°C and had to be lowered in temperature before their specific gravity measurements could be located in the tables used. Reboiler samples were allowed to pass immediately after collection through an all-glass heat exchanger, which lowered their temperatures to approximately 25°C. After both samples were analyzed

ORNL-DWG 80-17983

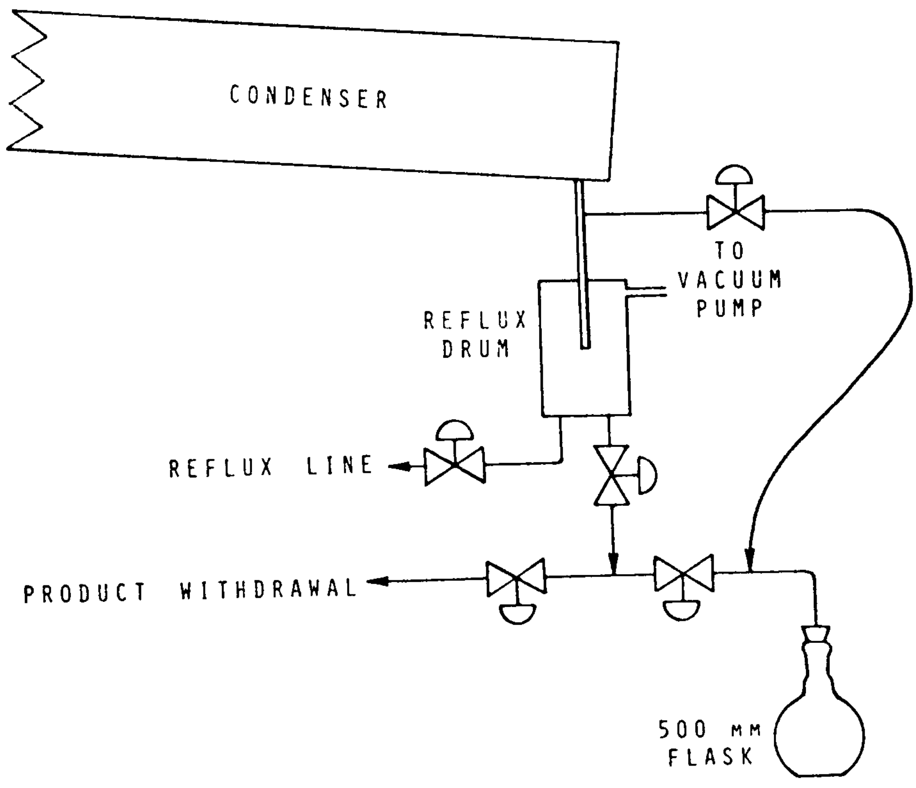

FIGURE 11

SCHEMATIC OF THE DISTILLATE SAMPLING SYSTEM

ORNL-DWG 80-17984

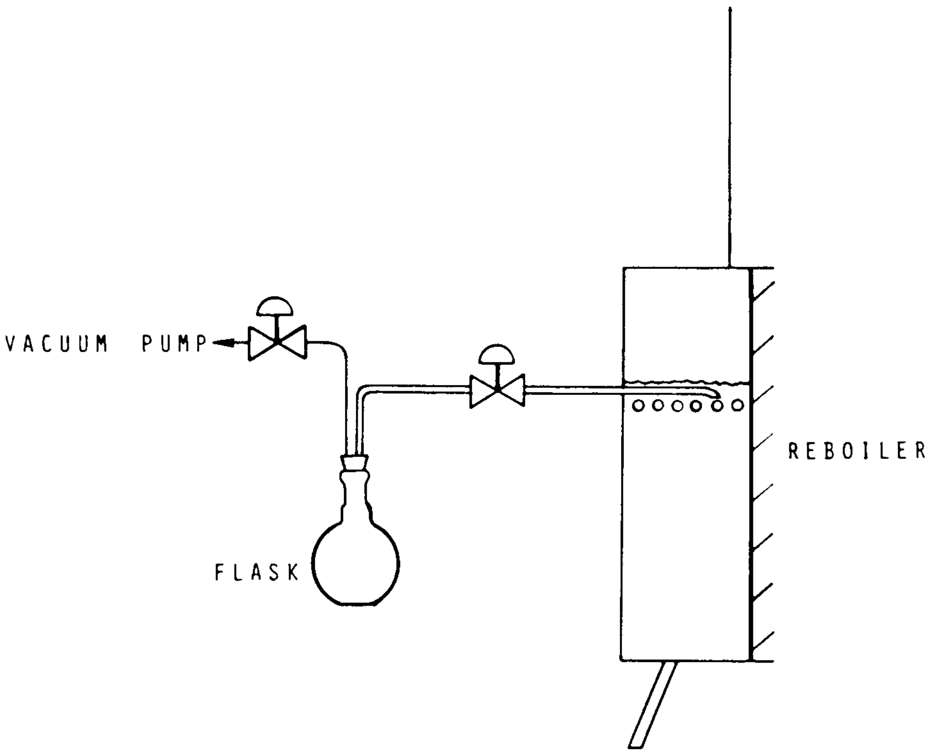

FIGURE 12

SCHEMATIC OF THE REBOILER SAMPLING SYSTEM they were reintroduced to the system at the approximate location at which they were withdrawn.

Temperatures were recorded to the nearest .1°C and specific gravity measurements to the nearest 0.0025 in all determinations of alcohol content in the distillate and reboiler. Tables containing the raw data are located in Appendix B.

II. CALCULATION OF THE H.T.U.

This section discusses the calculation of the $(H.T.U.)_{OG}$ in two sections. The first section contains a description of the procedure, while the second section details the actual calculations as performed for this study.

General Description

A number of values of x between x_w and x_p were chosen and corresponding values of y were obtained from the stripping and rectifying lines. Because the system being investigated was operated at total reflux, the stripping and rectifying lines were coincidental with the x=y diagonal. At the same values of x, the values of y^* were obtained from the equilibrium curve. A plot is then made of $1/(y^*-y)$ versus y and a graphical intergration of the area under the curve between the limits of y_1 (reboiler composition) and y_2 (distillate composition) gave the total number of transfer units.

Description of Calculations

The number of points chosen between x_w and x_p varied according to the specific number of data points given by the equilibrium curve at that pressure. In this investigation the H.T.U. was calculated at total reflux, facilitating steady state operation and eliminating the uncertainties in knowing the exact reflux ratio. Under this condition the values of y were equal to the values of x chosen. A computer program was devised that calculated the area under the curve of $1/(y^*-y)$ versus y by forming a trapezoid using each two successive points along the $1/(y^*-y)$ curve. This method was felt to be sufficiently accurate because of the numerous points along the $1/(y^*-y)$ curve. Appendix C contains the program used and a sample calculation.

III. MASS BALANCE ON SYSTEM'S OPERATION

The equipment being investigated in this study was operated at total reflux with all samples being returned to the system after analysis of the ethanol concentration. If the system was perfectly airtight, and the initial feed solution was void of any dissolved air, then negligible material would have been lost from the system. The rate of gas passing through the system due to leaks in the system was calculated to be 51.5, 48.7, and 36.8 cc/min., at pressures of 95, 190, and 380 mm Hg respectively. The amount of dissolved air in the initial feed concentration was assumed to be negligible compared to the amount leaked into the system.

Assuming that all of the air entering the system was removed by the vacuum pump, the amount of alcohol withdrawn was proportional to the ratio of the vapor pressure of the alcohol solution (at the point where the vacuum is drawn on the system) to the system's operational pressure.

The amounts of alcohol solution lost at pressures of 95, 190, and 380 mm Hg were approximately 5.0, 2.4, and .9 ml/hr. respectively. After an experimental run, the alcohol solution in the reboiler was remeasured after approximately six hours of operation and the amount of alcohol lost was verified to be less than 1 percent of that in the original solution.

IV. ENERGY REQUIREMENTS OF SYSTEM'S OPERATION

The unique nature of the heat source used in this investigation made it difficult to determine the amount of energy being delivered to the reboiler. During the course of an experimental run, the temperature, pressure, and velocity of the vaporized Freon changed continuously. All attempts to measure these parameters were found to critically impair the natural circulation of the Freon system.

The water-cooled Freon condenser was utilized in an attempt to approximate the conditions of the experimental runs. For each operational pressure of 95 mm Hg, 190 mm Hg, and 380 mm Hg, there were corresponding Freon operating pressure ranges of 110-26.5, 26.5-35.0, and 43.0-60.5 pounds per square inch gauge respectively. After adjusting the valves on the Freon line so that all of the vaporized Freon went directly to the water-cooled condenser, the water flow rate was adjusted to yield a specific Freon operating pressure. At this pressure setting of the Freon system, the water flow rate and temperature differential were recorded. The water flow rate was readjusted and the procedure repeated to yield several different Freon operating pressures.

Figure 13 shows a plot of the energy transferred to the water against various operational pressures for the Freon system. The Freon operational pressure ranges for the distillation conditions of 95 mm Hg, 190 mm Hg, and 380 mm Hg are also shown in Figure 13. Because the water-cooled condenser could not exactly duplicate the conditions under which the reboiler operated, Figure 13 is intended to represent an approximation to the energy that was being delivered to the distillation column under various operating conditions.

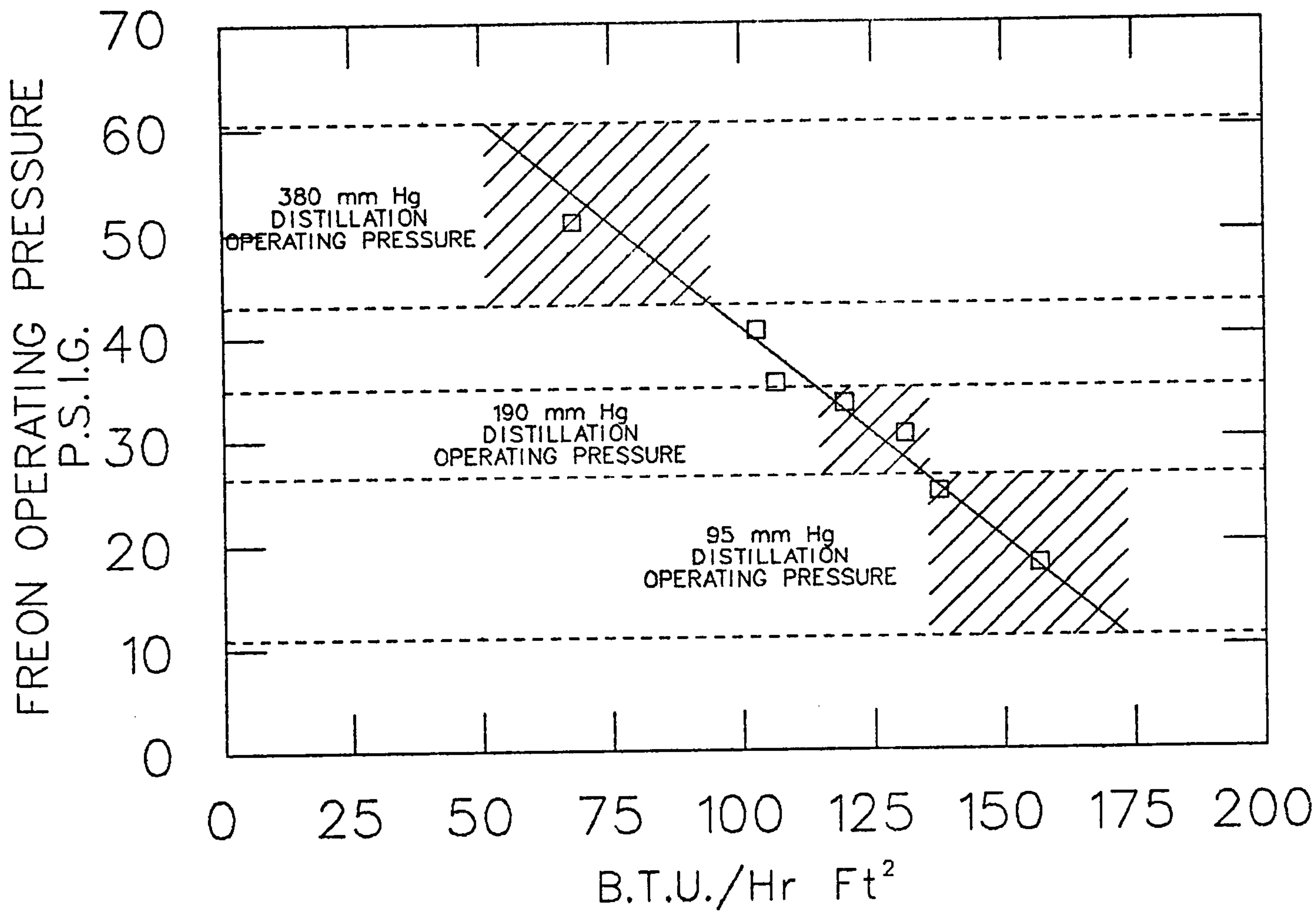

FIGURE 13

ENERGY OUTPUT OF SOLAR COLLECTORS FOR VARIOUS FREON OPERATING PRESSURES

CHAPTER V

RESULTS AND DISCUSSION OF RESULTS

Presented in this chapter are the experimental results and the conditions under which the results were obtained. Also included in this chapter are a discussion of the results and a comparison with those obtained in previous investigations.

I. RESULTS

The majority of the experimental effort was focused on determining the effect that the operating pressure and steady state reboiler and distillate concentrations had on the height of a transfer unit. Six experimental runs were performed at total reflux with the equipment to be investigated. At each pressure of 95 millimeters mercury, 190 millimeters mercury, and 380 millimeters mercury, the initial feed concentration was approximately 13 percent and 18 percent by weight ethanol, respectively, for two separate runs. The test duration varied with each run and lasted until concentrations in the reboiler and of the distillate were approximately constant. Table 1 presents the results of each run and the conditions under which the data was obtained.

II. DISCUSSION OF THE RESULTS

As can be seen from Table 1, the $(H.T.U.)_{OG}$ decreased for an increase in either the operating pressure or the initial feed concentration. The following discussion attempts to explain why each of these variables had an effect on the $(H.T.U.)_{OG}$.

TABLE 1

RESULTS AND CONDITIONS UNDER WHICH OBTAINED

Operating Pressure mm Hg (± 5)	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.	N_{Sc}	$\frac{(H.T.U.)}{(N_{Sc})^{.5}}$
95	12.8	1.236	85.5	.6	215	47.0	21.5	581.9	.0512
95	17.7	.877	90.7	4.1	200-295	44.5	14.5	708.9	.0329
190	12.9	.837	90.5	2.1	0-110	60.0	29.5	464.7	.0388
190	17.2	.747	91.5	4.7	130-160	59.5	31.0	517.8	.0328
380	13.5	.814	90.4	1.5	110-170	78.5	59.0	338.2	.0443
380	18.2	.711	91.5	6.3	110-170	72.5	50.0	456.1	.0333

For a given initial feed concentration, an increase in the operating pressure results in a decrease in the $(H.T.U.)_{OG}$. Most empirical formulas for the calculation of the $(H.T.U.)_{OG}$ or $(H.T.U.)_{OL}$ contain the Schmidt number, just as the Prandtl number is often encountered in problems of heat transfer. The Schmidt number, $N_{Sc} = \mu/\rho D$, is the ratio of the kinematic viscosity to the diffusivity. The temperatures encountered throughout the system were found to increase for a rise in the operating pressure, but temperature is known to have little effect on values of H_G because the Schmidt number for gases is essentially independent of temperature and pressure. Because the terms involved in the Schmidt number for the liquid are temperature sensitive, its value will decrease as a result of a rise in the pressure and temperature in the system. This occurrence is shown in Table 1 where values of the Schmidt number are listed for each of the sample runs.

Furnas and Taylor[3] evaluated a packed tower similar to the one being investigated using an ethanol-water mixture at one atmosphere pressure. They concluded from their experimental data that a large part of the resistance to transfer was in the liquid film. Pigford and Colburn[2] indicate for this situation that a rise in temperature will cause a decrease in values of H_L and may be estimated by assuming the quotient $(H.T.U.)_{OG}/(N_{Sc})^{.5}$ is equal to a constant. As shown in Table 1, the data obtained in this investigation supports this empirical formula indicating that a rise in temperature and pressure will lead to a decrease in the value of the $(H.T.U.)_{OG}$.

The relationship between various H.T.U.'s was shown to be

$$(H.T.U.)_{OG} = (H.T.U.)_G + (H.T.U.)_L \frac{mG}{L} \quad .$$

If part of the resistance is in the liquid film, a decrease in the slope of the equilibrium curve with increasing concentration will also cause the $(H.T.U.)_{OG}$ to decrease. This would explain why experimentally determined $(H.T.U.)_{OG}$'s decreased for an increase in the system's ethanol concentration.

Variations in the liquid and vapor rates encountered in this investigation were found to have a negligible effect on the $(H.T.U.)_{OG}$. As can be seen from the data in Table 2, the concentrations of ethanol increased and decreased gradually in the distillate and reboiler respectively during the experimental run. These concentrations were not found to fluctuate as the liquid and vapor rates rose and fell due to intermittent cloud cover.

The values of the $(H.T.U.)_{OG}$ obtained in this investigation varied from 0.69 feet to 1.24 feet for a packed height of 5.5 feet in an 8-inch internal diameter tower using 0.25-inch Intalox saddles at total reflux. Values of the $(H.T.U.)_{OG}$ obtained by Furnas and Taylor[3] for an ethanol-water system at 1 atmosphere and at total reflux varied from 0.90 feet to 1.76 feet using 0.50-inch saddles in a 12-inch internal diameter tower packed to a depth of 9.66 feet. The similarity in values of $(H.T.U.)_{OG}$ obtained in both cases indicates that the resistance to transfer was also similar in both cases.

TABLE 2

REPRESENTATIVE SAMPLE RUN AT 190 mm Hg AND INITIAL FEED CONCENTRATION OF 12.9 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
10:35	205	12.9	-	-	-	1st	56.5	28.0
11:00	180	12.9	-	-	-	110-130	58.0	29.5
11:40	184	12.9	-	-	-	120-150	60.0	32.0
11:45	-	12.9	-	84.7	3.2	-	-	-
12:10	188	12.9	-	-	-	150-180	61.5	33.0
12:15	-	12.9	-	86.9	1.7	-	-	-
12:40	188	12.9	-	-	-	140-160	62.0	33.5
12:45	-	12.9	-	88.7	2.0	-	-	-
1:10	185	12.9	-	-	-	95-110	60.5	31.5
1:15	-	12.9	-	90.1	2.1	-	-	-
1:40	186	12.9	-	-	-	0-110	60.0	31.0
1:45	-	-	.837	90.5	2.1	-	-	-

CHAPTER VI

CONCLUSIONS

The following conclusions were drawn from the results of the present investigation:

(1) The $(H.T.U.)_{OG}$ decreased for an increase in either the operating pressure or the initial feed concentration.

(2) Variations in the liquid and vapor rates observed in this investigation were found to have a negligible effect on the $(H.T.U.)_{OG}$.

(3) The energy produced by the solar collectors decreased for a rise in the operating pressure of the Freon system.

(4) Temperature variations at high ethanol concentrations were minimal, unlike those found at low concentrations.

(5) The relative location of valves and piping was significant because of the systems reliance on gravity to transfer liquids.

(6) When the system was operated at low pressures, small air leaks resulted in significant product removal by the vacuum pump.

LIST OF REFERENCES

LIST OF REFERENCES

[1] Beebe, A. H., Coulter, K. E., Lindsay, R. A., and Baker, E. M., Industrial and Engineering Chemistry 34, p. 1501 (1942).

[2] Colburn, A. P., and Pigford, R. L., Chemical Engineers Handbook, Third Edition, New York, McGraw-Hill Book Co. (1950).

[3] Furnas, C. C., and Taylor, M. L., American Institute of Chemical Engineers Transactions 36, p. 135 (1940).

[4] Chilton, T. H., and Colburn, A. P., Industrial and Engineering Chemistry 29, p. 447 (1937).

[5] Colburn, A. P., American Institute of Chemical Engineers Transactions 35, p. 211 (1939).

[6] Shinskey, Francis G., Energy Conservation Through Control, New York: Academic Press, 1978.

[7] Wilkinson, William R. and Beatty, Harold A., Industrial and Engineering Chemistry 18, p. 725 (1946).

APPENDIXES

APPENDIX A

APPENDIX A

EQUILIBRIUM DATA

Table 3 is the equilibrium data for ethanol-water as obtained by Bebe, Coulter, Lindsay, and Baker[1].

TABLE 3

EQUILIBRIUM DATA FOR ETHANOL-WATER

Pressure = 95 mm Hg			Pressure = 190 mm Hg			Pressure = 380 mm Hg		
X	Y	TEMP.	X	Y	TEMP.	X	Y	TEMP.
0.0035	0.0205	50.60	0.0160	0.1460	62.00	0.0160	0.1470	78.10
0.0045	0.0275	50.30	0.0370	0.2755	60.00	0.0315	0.2505	76.00
0.0175	0.1315	48.10	0.0650	0.3650	57.20	0.0600	0.3765	72.40
0.0585	0.3050	45.10	0.0900	0.4125	55.30	0.0855	0.4300	69.30
0.0680	0.3615	43.10	0.1580	0.5015	52.20	0.1465	0.5005	67.70
0.0935	0.4110	41.70	0.2090	0.5455	53.00	0.2060	0.5415	67.50
0.1650	0.5200	40.50	0.2385	0.5650	52.40	0.2360	0.5600	67.10
0.2125	0.5455	40.00	0.3535	0.6045	50.10	0.3495	0.5945	65.30
0.2410	0.5675	39.30	0.4705	0.6445	49.80	0.4675	0.6410	64.70
0.3615	0.6060	37.50	0.4970	0.6540	48.90	0.4875	0.6425	64.30
0.4740	0.6505	35.90	0.5805	0.6925	50.50	0.5800	0.6890	64.40
0.4985	0.6555	36.20	0.6525	0.7260	48.50	0.6525	0.7250	64.20
0.5815	0.6970	37.20	0.7000	0.7550	49.80	0.7000	0.7495	63.80
0.6460	0.7290	36.50	0.7200	0.7685	48.70	0.7175	0.7680	63.20
0.6540	0.7310	36.00	0.7895	0.8152	50.20	0.7890	0.8111	63.80
0.7230	0.7760	34.60	0.8416	0.8502	49.10	0.8420	0.8488	62.70
0.7900	0.8200	37.30	0.8735	0.8790	49.00	0.8749	0.8768	62.50
0.8370	0.8520	36.20	0.8970	0.8990	49.80	0.8967	0.8973	63.60
0.8731	0.8817	36.70	0.9485	0.9466	49.50	0.9485	0.9440	63.50
0.8830	0.8885	36.30	0.9600	0.9580	47.60	0.9727	0.9692	63.00
0.8880	0.8930	36.00	0.9719	0.9700	50.30			
0.8973	0.9012	37.00	0.9812	0.9798	48.60			
0.9489	0.9502	36.30						
0.9707	0.9715	37.40						
0.9825	0.9835	35.40						

APPENDIX B

APPENDIX B

ORIGINAL DATA

Tables 4 through 9 present the original data as obtained during the experimental runs.

TABLE 4

DATA OBTAINED DURING RUN AT 95 mm Hg AND INITIAL FEED CONCENTRATION OF 12.8 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
10:05	93	12.8	-	-	-	1st	40.0	13.0
10:30	93	12.8	-	-	-	140	41.5	14.0
10:45	95	12.8	-	-	-	150	43.0	15.5
10:55	95	12.8	-	-	-	170	43.5	16.5
11:00	-	12.8	-	77.0	4.0	-	-	-
11:40	100	12.8	-	-	-	200	46.0	20.0
11:45	-	12.8	-	85.6	1.5	-	-	-
12:10	99	12.8	-	-	-	215	46.5	21.0
12:15	-	12.8	-	84.9	.3	-	-	-
12:40	99	12.8	-	-	-	215	47.0	21.5
12:45	-	12.8	1.236	85.9	.2	-	-	-

TABLE 5

DATA OBTAINED DURING RUN AT 95 mm Hg AND INITIAL FEED CONCENTRATION OF 17.7 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
9:50	94	17.7	-	-	-	1st	36.5	11.0
10:25	100	17.7	-	-	-	160-170	41.5	15.0
10:55	100	17.7	-	-	-	205-215	43.5	17.5
11:00	-	17.7	-	83.0	6.0	-	-	-
11:45	98	17.7	-	-	-	250-260	44.0	17.5
11:50	-	17.7	-	89.7	5.0	-	-	-
12:15	92	17.7	-	-	-	195-205	43.0	13.5
12:20	-	17.7	-	91.0	5.5	-	-	-
1:00	98	17.7	-	-	-	180-195	43.0	26.0
1:05	-	17.7	-	91.4	3.9	-	-	-
1:25	103	17.7	-	-	-	200-205	45.0	26.5
1:30	102	17.7	-	-	-	205-215	45.0	24.5
1:35	-	17.7	-	90.4	3.3	-	-	-
1:50	96	17.7	-	-	-	200-295	44.5	14.5
1:55	-	17.7	.877	90.3	5.1	-	-	-

TABLE 6

DATA OBTAINED DURING RUN AT 190 mm Hg AND INITIAL FEED CONCENTRATION OF 12.9 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
10:35	205	12.9	-	-	-	1st	56.5	28.0
11:00	180	12.9	-	-	-	110-130	58.0	29.5
11:40	184	12.9	-	-	-	120-150	60.0	32.0
11:45	-	12.9	-	84.7	3.2	-	-	-
12:10	188	12.9	-	-	-	150-180	61.5	33.0
12:15	-	12.9	-	86.9	1.7	-	-	-
12:40	188	12.9	-	-	-	140-160	62.0	33.5
12:45	-	12.9	-	88.7	2.0	-	-	-
1:10	185	12.9	-	-	-	95-110	60.5	31.5
1:15	-	12.9	-	90.1	2.1	-	-	-
1:40	186	12.9	-	-	-	0-110	60.0	31.0
1:45	-	12.9	.837	90.5	2.1	-	-	-

TABLE 7

DATA OBTAINED DURING RUN AT 190 mm Hg AND INITIAL FEED CONCENTRATION OF 17.2 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
10:21	195	17.2	-	-	-	1st	56.0	26.5
10:32	195	17.2	-	-	-	95-120	56.0	26.5
10:36	-	17.2	-	75.5	10.5	-	-	-
11:55	195	17.2	-	-	-	140-160	59.5	34.0
12:00	-	17.2	-	86.0	4.5	-	-	-
12:25	195	17.2	-	-	-	140-160	60.5	34.0
12:30	-	17.2	-	90.0	4.5	-	-	-
1:10	190	17.2	-	-	-	130-150	60.0	31.5
1:15	-	17.2	-	90.8	4.2	-	-	-
1:45	194	17.2	-	-	-	140-170	59.0	35.0
1:50	-	17.2	-	91.3	4.7	-	-	-
2:15	194	17.2	-	-	-	160-195	60.0	32.0
2:20	-	17.2	-	91.6	4.6	-	-	-
2:45	190	17.2	-	-	-	130-160	59.5	29.5
2:50	-	17.2	.747	91.6	4.7	-	-	-

TABLE 8

DATA OBTAINED DURING RUN AT 380 mm Hg AND INITIAL FEED CONCENTRATION OF 13.5 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
11:55	378	13.5	-	-	-	1st	72.5	48.0
12:10	375	13.5	-	-	-	70-85	73.0	50.0
12:25	384	13.5	-	-	-	95-110	76.5	52.5
12:30	-	13.5	-	80.0	5.5	-	-	-
12:55	389	13.5	-	-	-	130-170	76.5	55.5
1:00	-	13.5	-	83.0	4.0	-	-	-
1:25	386	13.5	-	-	-	95-130	77.0	59.0
1:30	-	13.5	-	85.5	2.5	-	-	-
1:55	386	13.5	-	-	-	120-180	79.0	59.0
2:00	-	13.5	-	86.5	1.4	-	-	-
2:25	381	13.5	-	-	-	120-195	78.5	59.5
2:30	-	13.5	-	89.0	.6	-	-	-
2:55	381	13.5	-	-	-	110-170	78.5	59.0
3:00	-	13.5	.814	90.4	.6	-	-	-

TABLE 9

DATA OBTAINED DURING RUN AT 380 mm Hg AND INITIAL FEED CONCENTRATION OF 18.2 PERCENT

Time	Operating Pressure mm Hg	Initial Feed Concentration wt. %	H.T.U. ft.	Distillate Concentration wt. %	Reboiler Concentration wt. %	Reflux Rate ml/min.	Reboiler Temperature °C	Freon Pressure P.S.I.G.
11:08	379	18.2	-	-	-	1st	69.5	43.5
11:55	381	18.2	-	-	-	110-170	71.5	48.0
12:00	-	18.2	-	83.0	8.0	-	-	-
12:25	381	18.2	-	-	-	110-180	72.5	49.5
12:30	-	18.2	-	85.5	6.5	-	-	-
12:55	381	18.2	-	-	-	120-170	72.5	50.0
1:00	-	18.2	-	88.0	6.3	-	-	-
1:25	383	18.2	-	-	-	110-180	73.0	50.5
1:30	-	18.2	-	90.1	6.3	-	-	-
1:55	383	18.2	-	-	-	140-180	72.5	50.5
2:00	-	18.2	-	90.9	6.3	-	-	-
2:25	381	18.2	-	-	-	105-170	72.5	50.0
2:30	-	18.2	-	91.3	6.3	-	-	-
2:55	383	18.2	-	-	-	110-170	72.5	50.0
3:00	-	18.2	.711	91.5	6.3	-	-	-

APPENDIX C

APPENDIX C

CALCULATION OF THE H.T.U.

The following is the computer program used to calculate the height of the transfer unit from the experimental data and a sample run of the program.

```
      G1 = 0.0
      READ (5,5) X1,Y1
    5 FORMAT (F6.4,3X,F7.4)
      Z1 = (1/(Y1-X1))
      WRITE (6,10)
   10 FORMAT ('1',16X,23HVAPOR-LIQUID EQUILIBRIA)
      WRITE (6,15)
   15 FORMAT (/,23X,1HX,9X,1HY,15X,1HY,8X,2HY*,4X,8H1/(Y*-Y),/)
      WRITE (6,20) X1,Y1,X1,Y1,Z1
   20 FORMAT (20X,F6.4,3X,F7.4,10X,F6.4,3X,F7.4,3X,F7.4)
   25 READ (5,30) X2,Y2
   30 FORMAT (F6.4,3X,F7.4)
      Z2 = (1/(Y2-X2))
      IF (X2 .EQ. .9999) GO TO 50
      WRITE (6,35) X2,Y2,X2,Y2,Z2
   35 FORMAT (20X,F6.4,3X,F7.4,10X,F6.4,3X,F7.4,3X,F7.4)
      C = ABS(X1-X2)
      IF (Z1 .LE. Z2) GO TO 40
      D = Z2*C
      GO TO 45
   40 D = Z1*C
   45 E = ABS(Z1-Z2)
      F = (.5*C*E)
      G = D+F
      G1 = G1+G
      X1 = X2
      Y1 = Y2
      Z1 = Z2
      GO TO 25
   50 WRITE (6,55) G1
   55 FORMAT (/,20X,8HN.T.U. =,2X,F6.4)
      STOP
      END
```

VAPOR-LIQUID EQUILIBRIA

X	Y	Y	Y*	1/(Y*-Y)
0.0024	0.0128	0.0024	0.0128	96.1539
0.0035	0.0205	0.0035	0.0205	58.8235
0.0045	0.0275	0.0045	0.0275	43.4783
0.0175	0.1315	0.0175	0.1315	8.7719
0.0585	0.3050	0.0585	0.3050	4.0568
0.0680	0.3615	0.0680	0.3615	3.4072
0.0935	0.4110	0.0935	0.4110	3.1496
0.1650	0.5200	0.1650	0.5200	2.8169
0.2125	0.5455	0.2125	0.5455	3.0030
0.2410	0.5675	0.2410	0.5675	3.0628
0.3615	0.6060	0.3615	0.6060	4.0900
0.4740	0.6505	0.4740	0.6505	5.6657
0.4985	0.6555	0.4985	0.6555	6.3694
0.5815	0.6970	0.5815	0.6970	8.6580
0.6460	0.7290	0.6460	0.7290	12.0482
0.6540	0.7310	0.6540	0.7310	12.9870
0.6975	0.7594	0.6975	0.7594	16.1551

N.T.U. = 4.4486

VITA

John Mark Williams was born in Hixson, Tennessee on October 22, 1953. He attended elementary school in that city and was graduated from Hixson High School in 1971. He entered Middle Tennessee State University and received the Bachelor of Science degree in Chemistry in 1975.

The author began employment with the Chattanooga-Hamilton County Air Pollution Control Bureau in the summer of 1975. In August 1978, he entered the Graduate School of The University of Tennessee, Knoxville, and received the Master of Science degree in Chemical Engineering in December 1980. He will be employed by the du Pont Company in Victoria, Texas.

He is an associate member of the American Institute of Chemical Engineers and a member of the American Chemical Society.

The author is married to the former Debra Mason of Lebanon, Tennessee.