

DISTILLATION COLUMN DYNAMICS--AN EXPERIMENTAL STUDY

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PREFACE

To aid in the development of a dynamic model of a distillation column, a series of experimental runs have been made. These runs were made on both binary and ternary systems. The data obtained have been used to show that the behavior of the column can be accurately described by a first order lumped parameter model. On the basis of this ability to describe the dynamic behavior, the model has been applied to a theoretical feed forward control system. In the feed forward control scheme, the model has shown its ability to control the operation of a distillation column.

I am deeply indebted to Dr. R. N. Maddox for the guidance and advice that he has given me. I would also like to thank Dr. J. H. Erbar for his assistance, especially during the development of the feed forward control system. To Dr. J. B. West, I would like to express my appreciation for his assistance in the analytical work, especially for aid in designing the equipment used in measuring the feed profile. To each of the above and to Dr. J. R. Norton, I extend an additional thank you for the advice they gave as my Doctoral Advisory Committee. My sincere thanks go to my research associate, Mr. J. R. Deam, for the many hours, above those for his own research, which he so willingly gave.

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CHAPTER I

INTRODUCTION

In recent years members of the chemical engineering profession have become increasingly interested in the control of industrial processes. The control of distillation columns has been in the forefront of this increased interest because distilling operations are found in almost every phase of the chemical industry. Due to the widespread application of distillation, industry can realize considerable profits through effective control of columns.

The research in distillation column control has been primarily concerned with determination and prediction of the dynamic behavior of distillation columns. In general, two different paths have been followed in studying distillation column dynamics. One path has been to use a "rigorous" approach. According to this "rigorous" approach, a distillation column is considered to consist of a known number of trays. Each of the trays is assumed to play an equal role in the dynamic behavior of the column and a separate differential equation is written for each component on each tray. All of these differential equations must be solved simultaneously to obtain the dynamic behavior. Obviously, the simultaneous solution of a large number of differential equations is a time consuming task, even with the most modern computer. To add to the computational difficulties, the "rigorous" model should consider the vapor-liquid contacting efficiency, so that the theoretical model will

have practical applications. When all of the factors have been considered, the resulting model is usually too complex to be useful in an operating control scheme.

To avoid the computational difficulties encountered with the "rigorous" model, some researchers have used an alternate approach in determining the dynamic behavior of a distillation column. This approach is to consider the column according to a section concept. According to that concept, the portion of the column that lies between points where either material or energy enters or leaves the column is a section. The advantages of a lumped parameter model over the "rigorous" model are that there are significantly fewer differential equations to be solved and the effects of efficiency do not have to be considered. Efficiency does not have to be considered because a separation parameter is used to describe the separation that occurs in a section, and is assumed to remain constant during the transient period. While the transient response of the distillation column can be represented by the first order lumped parameter model, a dead time must also be included in the model. The dead time accounts for the fact that the column is actually filled with trays, and a significant time is required for the composition change to reach the extremes of the column.

The object of the present project has been to determine if the transient behavior of a distillation column can be accurately represented by the first order lumped parameter model with dead time. To accomplish this objective, experimental data have been obtained for the transient behavior of an Oldershaw distillation column operating as a stripping column. Data were obtained for both binary and ternary systems, and compared with the values predicted by the proposed model. As an

extension of the experimental work, a computational investigation has been conducted to determine the applicability of the lumped parameter model to a feed forward control model.

CHAPTER II

REVIEW OF THE LITERATURE

Prior to 1932 almost no work had been done in developing dynamic models of industrial processes. In 1932 Ivanhoff (16) presented a paper in which he made the first attempt at developing a mathematical model, from a strictly empirical point of view. From the discussion accompanying the article, the results of his experiments appear to have been widely accepted. Several other early authors (1, 41) also approached the development of dynamic models from an empirical point of view. These men correlated statistically the behavior of a process with changes in independent variables and developed an approximate model of the process. Several other researchers (8, 14, 17, 24) approached the problem of developing a dynamic model by constructing small scale plants and then developing a dynamic model from the results obtained. Although these early workers were not interested in the control of distillation columns, but in transient systems in general, their works formed the foundation of modern process dynamics.

In 1947, Marshall and Pigford (22) proposed the first mathematical model of a distillation column. Their model was based on the equilibrium stage as shown in Figure 1. According to the equilibrium tray concept, each tray must be considered individually, and the differential equation that predicts the transient behavior must be written for each component in the form

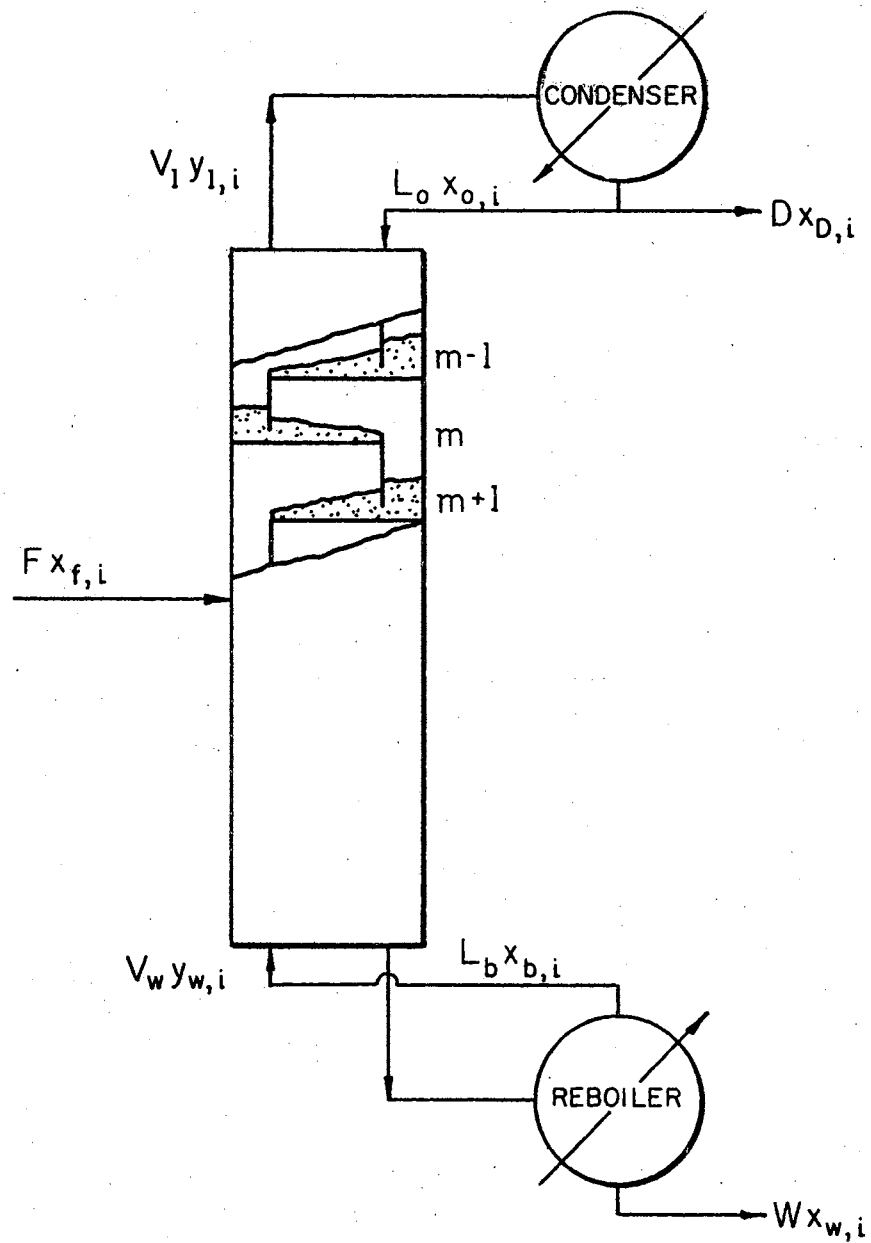


Figure 1. A Schematic Diagram of a Distillation Column Having Trays.

$$\frac{d\delta_{n,n}^L x_n}{dt} + \frac{d\delta_{n,n}^V y_n}{dt} = (L_{n-1} x_{n-1} + V_{n+1} y_{n+1}) - (L_n x_n + V_n y_n) \quad (1)$$

where

$$\frac{d\delta_{n,n}^L x_n}{dt} = \text{change in liquid holdup of a component on tray } n \text{ with time.}$$

$$\frac{d\delta_{n,n}^V y_n}{dt} = \text{change in vapor holdup of a component above tray } n \text{ with time.}$$

$$L_{n-1} x_{n-1} + V_{n+1} y_{n+1} = \text{rate at which a component flows to tray } n.$$

$$L_n x_n + V_n y_n = \text{rate at which a component flows away from tray } n.$$

While this concept is theoretically sound, there are several drawbacks to using it. The most significant of these drawbacks is the vast number of equations that must be solved. This problem is perhaps best illustrated by an example. If the column of interest has n trays and i components, the number of differential equations that must be solved is on the order of n times i . Thus, the model is severely limited in its complexity. Since neither digital nor analog computers were well developed at the time that Marshall and Pigford developed the plate-to-plate model, a rigorous solution of the equations was almost impossible. The difficulty in using the Marshall and Pigford model was compounded further by the fact that the trays are not normally equilibrium ones. Thus, some method of estimating the efficiency, or the approach to equilibrium, of each tray was required. In order to make their model more useful, Marshall and Pigford made the following assumptions:

1. Constant molal overflow
2. Negligible vapor holdup above a tray
3. Approach to equilibrium between the liquid on the tray and the

vapor above the tray could be represented by a straight pseudo equilibrium line.

While these assumptions enabled Marshall and Pigford to obtain an analytical solution to the differential equations, the accuracy of the model was reduced by them. The assumption of negligible vapor holdup is normally a good assumption, but since the assumption of constant molal overflow required that the molar heats of vaporization of the components be equal and the assumption of a straight equilibrium line requires that the concentration of the component be small, the integrated equations are normally too restricted to be useful on actual systems.

Some time after Marshall and Pigford developed their model, Rose and his co-workers (30, 31, 32, 33) applied the basic equation, equation (1), to a batch distillation column. In this application, Rose et al. avoided the assumptions that limited the usefulness of the Marshall and Pigford equations by programming the differential equations on a digital computer. They were, however, confronted by the problem of excessive computer time.

About the same time that Rose et al. were publishing their work, Robinson and Gilliland (29) developed an approximate graphical method for predicting the approach to steady-state of a distillation column. Their method was restricted to cases where the column was upset by a change in the feed composition, and, like previous models, was based on the equilibrium tray concept.

Voetter (37) was perhaps the first to combine experimental data with a theoretical analysis. He compared the equations of Marshall and Pigford with experimental data that he obtained on a sixty tray Oldershaw distillation column. The experimental and the calculated values compared

excellently during the early portion of the transient period, but as the column approached steady-state the experimental and calculated values differed considerably. Voetter's experimental results were for a single section column under the influence of a step change, but he mathematically extended the results to a complete fractionation column under the influence of a frequency response function. In 1957, Wilkinson and Armstrong (39, 40) presented experimental data that were obtained on a five tray four-inch diameter column which was operating on the carbon tetrachloride-benzene binary system. They presented adaptations of the Marshall and Pigford equations and the assumption of a straight equilibrium line was still required. This work was, however, performed on a complete column. In 1961, Armstrong and Wood (2) published experimental as well as calculated results for a twenty-one tray distillation column. The purpose of their work was to determine the effect of changing the reflux rate. At the top of the column the experimental and the calculated values were in good agreement, but at the bottom of the column the experimental and calculated values did not agree well.

In 1961 Baber (3, 4, 5) presented the most extensive experimental and calculational study that has been published. He programmed a series of differential equations that were developed by Lamb and Pigford (18), but were based on the earlier Marshall and Pigford equations, on an analog computer. Baber compared the results obtained on the computer with the experimental data he obtained. The data were obtained on a five tray, single section distillation column. The method of operating the column was to allow the column to come to steady-state at total reflux and then change one of the operating variables; either the reflux rate, the reflux composition, or the vapor rate. The flow rates and

compositions of the various streams were determined before the step change was made and the compositions were measured at intervals throughout the transient period. When the column reached steady-state, the flow rates and the compositions were again measured. For some of the experimental runs, Baber was able to get good agreement between the experimental and the computer values, but for most of the runs he was unable to obtain good agreement.

At the Baltimore meeting of the American Institute of Chemical Engineers, Marr (21) suggested a new concept for predicting the transient behavior of a distillation column. He suggested that in order to get away from the conventional and complicated plate-to-plate model, some parameter which could be used to describe the degree of separation that was occurring in a distillation column should be developed. However, after suggesting the simplified model, Marr complicated it by considering all aspects of the mechanics of construction of the column. Due to these additions, the final model was almost as complex as the plate-to-plate model. Little simplification was actually accomplished. After Marr's work, no further efforts were made to develop a simplified model until Reynolds (28) began his work.

Reynolds envisioned a distillation column as being composed of several sections in which there could be any number of trays. According to the section concept, as shown in Figure 2, a section of a distillation column is that part of the column which lies between the points at which either feed streams enter or product streams leave the column. According to Reynolds, the rate at which mass is transferred from the vapor phase to the liquid phase can be expressed by the equation

$$N_{n,i}^V = -J_{n,i}(y^*-y)_{n,i} \quad (2)$$

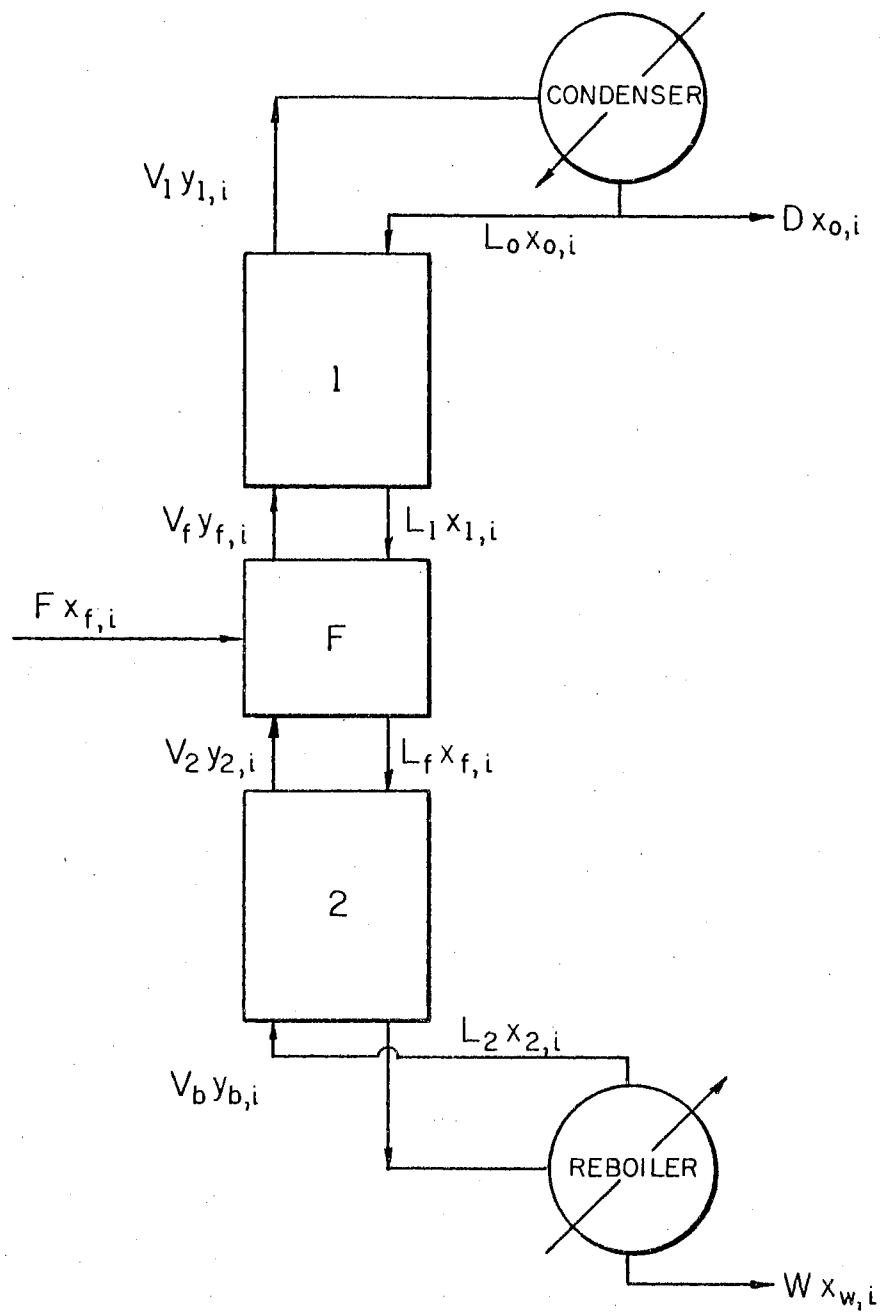


Figure 2. A Distillation Column According to the Section Concept

where $J_{n,i}$ is the parameter which describes the degree of separation occurring in a section and $(y^*-y)_{n,i}$ is the driving force for mass transfer in the section.

The reader will notice that equation (2) is identical in form with the equation that is normally used for mass transfer

$$N = K(y^* - y)$$

There are, however, fundamental differences between equation (2) and the conventional mass transfer equation. In deriving the conventional equation, the coefficient K is related to the diffusivity of the components being transferred. The coefficient in equation (2), however, is not related directly to the physical properties of the component being transferred. The coefficient, $J_{n,i}$, is a parameter that describes the degree of separation occurring in a section and is an empirically determined factor.

Using this idea for the rate of mass transfer in the section, Reynolds developed a set of differential equations for the transient behavior of the liquid and vapor streams leaving the section. In developing the equations, Reynolds made two major assumptions. The first of these assumptions was that $J_{n,i}$ remained constant for small changes of column conditions and the second was that of constant molal overflow throughout the section. After developing the model, Reynolds attempted to prove the model by comparing the values predicted with the model with experimental data, but was unable to obtain good agreement.

In the time since Reynolds completed his work using the lumped parameter model, the research in distillation column dynamics has followed two distinctly different paths. One path has been toward the investigation of the use of lumped parameter models. The other path has been

toward the use of increasingly more complex models.

In the direction of the lumped parameter models, the present author (26, 27) has shown that the model proposed by Reynolds could be used to reproduce the transient behavior of a column. Murrill (25) has shown experimentally that the transient behavior of the composition of the liquid leaving a tray can be accurately represented by an equation of the form

$$x(t) = x_{\infty} [1 - e^{-(t/T)}] + x_0 e^{-(t/T)} \quad (3)$$

Finally, Moczek, et al. (23) have shown theoretically that the transient behavior of the composition of the products from a distillation column can be represented by a simple model using a dead time and two time constants.

In the opposite direction, i.e., toward the use of increasingly more complex models, Hickaba, et al. (15) have shown that by using plate efficiencies and continuous heat balancing the unsteady-state behavior of the composition of the distillation column products can be accurately represented. The model that they used is based on the plate concept and is restricted to binary systems.

Waggoner and Holland (38) developed a theoretical model for the transient behavior of a multicomponent distillation column. They assumed plate efficiencies were known, and used an external material balance to force the column into material balance at the final steady-state.

Following the trend toward increasingly more complex models, Tetlow, Groves, and Holland (35) proposed a method for estimating transient plate efficiencies in a distillation column. Using this method for estimating efficiencies, the same authors (36) presented a transient model in which they considered the hydrodynamics on each tray in the

column. They proposed that the liquid on each tray could be represented by a combination of plug flow, ideal mixing, and bypassing. Independently Duffin and Gamer (10) developed a mathematical model in which they used the Francis Weir formula to determine the holdup on each tray. Neither Holland, et al., nor Duffin and Gamer presented any experimental data to prove their proposed models.

In view of the diverging directions of the effort concerning distillation column dynamics, the present project was undertaken. The objective was to obtain experimental data on distillation column dynamics. These data were to be obtained on both binary and ternary systems. They were to be used to determine if the transient behavior of a distillation column could be represented by a simple lumped parameter model, or if a more complex model was required.

CHAPTER III

EXPERIMENTAL APPARATUS

The experimental apparatus consisted primarily of an Oldershaw column, a reboiler, a vapor-dividing head, and a pressure control system (see Figures 3 and 4). The Oldershaw column was five-eighths of an inch in diameter and contained ten perforated glass plates. Each plate was equipped with a weir to maintain the liquid level on the plate, and downcomers were provided to direct the flow of liquid reflux from plate to plate. The vapor rose through 0.035 inch diameter holes in the plates. The column was enclosed in a Dewar jacket to minimize heat losses. A reflux and holdup measuring trap was connected between the column and the reboiler. The trap was equipped with a sampling valve to permit measurement of column holdup and liquid reflux composition.

The reboiler for the column was essentially a glass pot in which a dip leg was used to permit the withdrawal of a bottom product. A Chromalox C-622 heating element provided heat to the reboiler. The heat input was controlled by a Powerstat. The heating element fitted into a slot in the reboiler so that the heating element did not contact the liquid. The bottoms product was pumped from the reboiler to a combined cooler and receiver. After measurement of the volumetric bottoms rate, the bottoms product was pumped to a storage tank.

A glass feed section, surrounded by a Dewar jacket, was mounted on top of the column. The feed entered the feed plate after being pumped

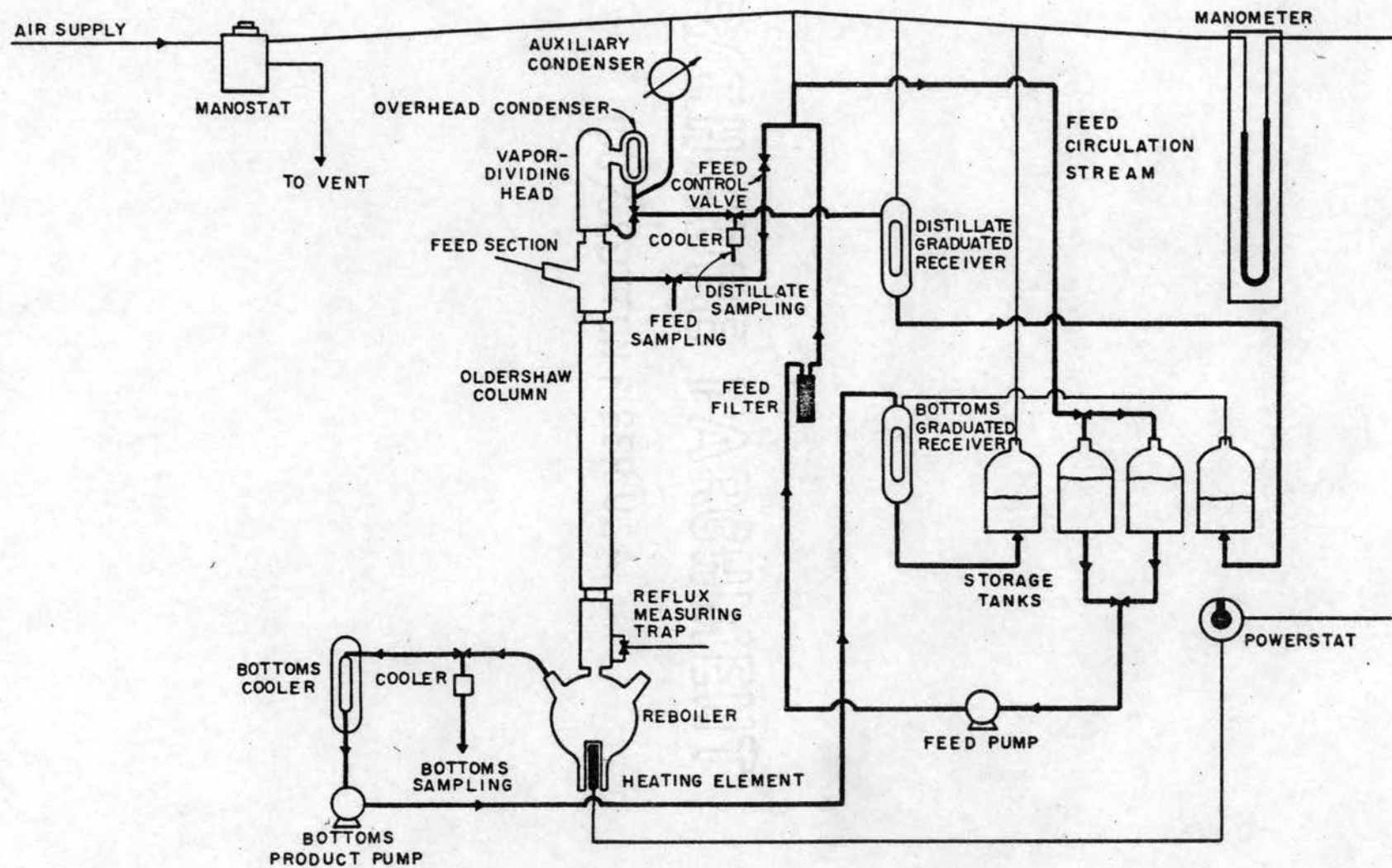


Figure 3. The Experimental Apparatus

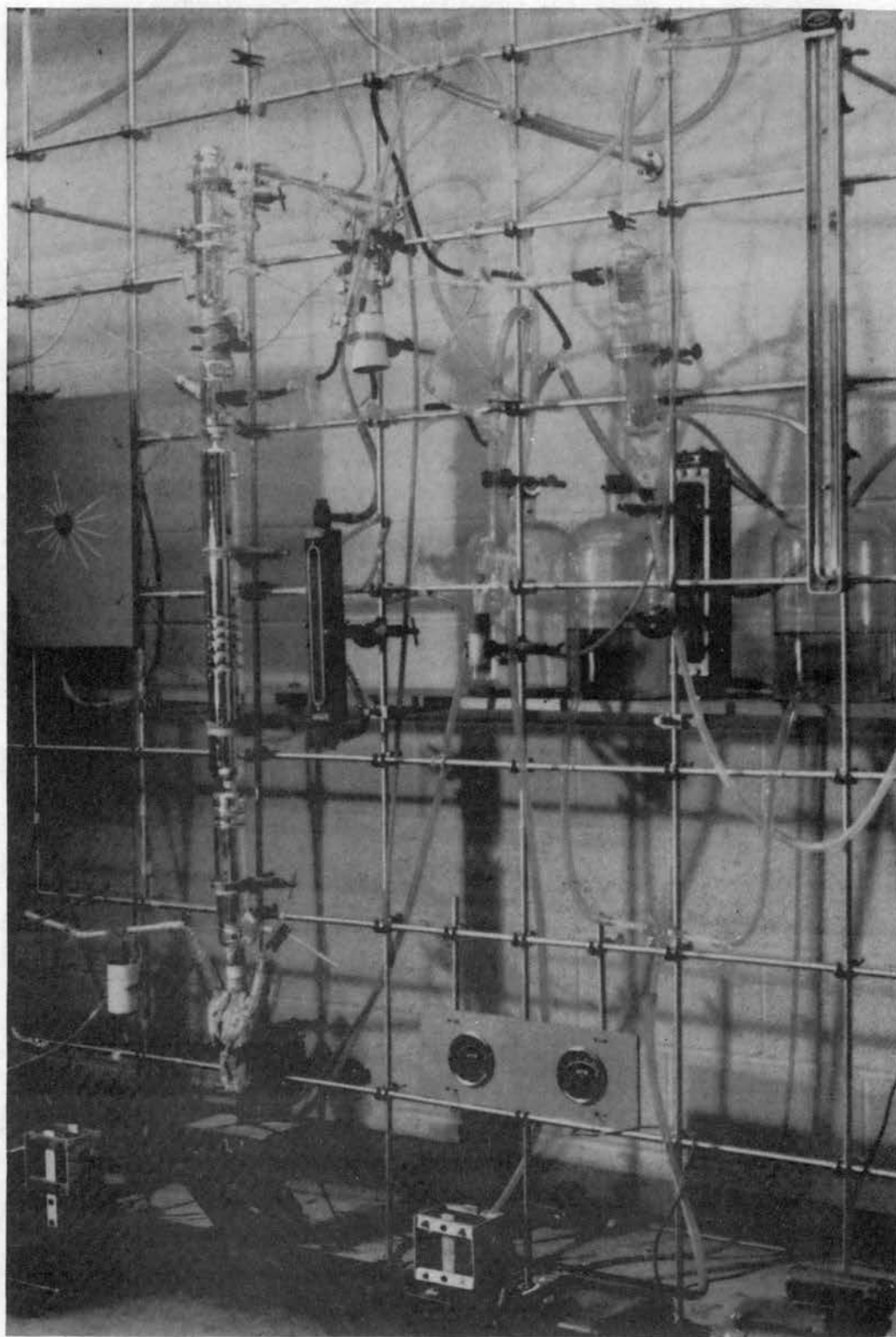


Figure 4. Photograph of the Experimental Apparatus

from feed storage through a constant head tank. A three-way stopcock in the feed line permitted measurements of feed rate and composition.

A vapor-dividing head directed the flow of the vapor stream to the product condenser. The head was mounted above the feed section and was equipped with a Dewar jacket. The vapor passed through the dividing head into the overhead condenser. A three-way stopcock in the external reflux line directed the flow of condensed vapor back to the column or to the product receiver, or to both. An auxiliary condenser was connected to the overhead condenser. The additional condenser was needed to condense all the overhead vapor. Dry ice was packed around the top of the auxiliary condenser to prevent loss of vapor.

A manostat controlled the pressure in the column and the system at 760 mm Hg. Constant-pressure operation was considered important for several reasons:

1. For comparison purposes, most laboratory data in the literature are reported for one atmosphere pressure.
2. The same pressure for all runs gives these runs a common denominator.
3. Unless the column is operated at constant pressure, there is little reason to assume that the data will be reproducible.

Air was introduced to the system through the manostat when the pressure was less than the desired pressure. When the system pressure was greater than the desired pressure, air and non-condensable vapors were released through the manostat to the atmosphere. Thus, the system pressure was controlled whether the pressure tended to be greater than or less than the desired pressure. Pressure lines connected the manostat to the storage tanks, the product and feed streams, and the column through the

overhead condenser.

The liquid storage system included four eight-liter glass bottles. Two bottles were provided for the feed and one for each of the products.

An F & M Model 500 Programmed High-Temperature Gas Chromatograph was used to determine liquid sample compositions. The chromatograph utilized a thermal conductivity unit with helium as the carrier gas. A Honeywell-Brown recorder recorded the chromatograph response and a Disc integrator measured the area under the peaks. Standard samples were used to calibrate the response from the thermal-conductivity detector of the chromatograph. A regression analysis was used to convert area fractions to weight fractions. The calibration is listed in Appendix B.

Sigmamotor metering pumps were used to pump the feed from feed storage to the feed plate and to pump the bottoms product from the reboiler to the storage tank. Variable-speed Zeromax transmissions, attached to the motors, were used to control the flow rates.

Copper-constantan thermocouples were used to sense column and product stream temperatures. Leads from a thermocouple selector switch were connected to a Leeds and Northrup potentiometer. The thermocouples were calibrated to permit the conversion from millivolts to degrees Fahrenheit.

CHAPTER IV

EXPERIMENTAL PROCEDURE

The start-up procedure consisted first of filling the reboiler with the feed mixture. Powerstat voltage was set to give the desired heat input to the reboiler. The manostat was adjusted to control the system pressure to 760 mm Hg.

The column was then operated at total reflux with the feed and product valves closed until steady state had been reached. The steady-state operation was characterized by constant column temperatures and constant product compositions. Preliminary experiments showed that, at total reflux, approximately ninety minutes were required to achieve product compositions that did not change. The column was usually operated at total reflux for two hours to assure steady-state operation.

Once the total reflux steady-state had been achieved, column operation was changed to total-takeoff of top tray vapor. The bottoms product and feed valves were opened and both pumps were started. The distillate product valve was opened to the position which provided no external reflux. In this manner the column was operated as a non-refluxed stabilizer, or stripping column.

Flow rate measurements were made for the feed and product stream. The feed rate had been determined during total-reflux operation. A stop watch was used to measure the time necessary to collect a volume of feed in a graduated cylinder. Feed rate measurements were continued until a

reproducible feed rate was established. During the run, product rate measurements were made. In a way similar to that for the feed stream, the time necessary to fill the graduated product receivers was recorded. The bottoms product rate could be altered by changing the speed on the pump transmission. There was no provision, other than the Powerstat setting for the heat input, to control the distillate rate.

The column was run at total-takeoff of the top tray vapor until another steady state was obtained. Steady-state operation was defined by the conditions:

1. Constant temperatures in the column
2. Constant product compositions
3. Constant product rates

Usually the time required to satisfy conditions (2) and (3) was thirty minutes. Samples were taken every fifteen minutes for an hour to check for constant product compositions.

When the steady-state condition had been reached the transient run was begun. The procedure for initiating a transient run was to change the tank from which the feed was being drawn. Since the feed had to pass through the feed pump, the feed filter, and the constant head tank, some questions arose as to whether a step change in feed composition was experienced. This question was answered by a series of experimental runs in which the type of change entering the column was measured. The experimentation showed that the departure from a step change was negligible. The runs and the results obtained from them will be discussed later.

After the change was made in the feed tank, samples of distillate and bottoms product were taken at one minute intervals for the first

twelve minutes. For the next eight minutes, samples were taken every two minutes, and for the final fifteen minutes, samples were taken every five minutes.

The sampling procedure was designed to reduce the experimental error as much as possible. To prevent vaporization of the samples, small dry-ice coolers were installed in the sample lines. The coolers were made small to eliminate as much holdup as possible in the sample lines. The sampling procedure was to purge the sample line and then draw the sample. The distillate sample line was equipped with a continuous purge, but this could not be done in the bottoms product sample line.

The samples were taken in one-half dram vials and cooled immediately in a Dewar flask filled with dry-ice. The small sample containers were used to prevent a change in the liquid composition due to a large vapor space above the sample. The samples were kept in the dry-ice filled flask until analyzed. To determine the effectiveness of the small sample containers, a sample was analyzed, resealed, and allowed to set at room temperature for twenty-four hours. After twenty-four hours, the sample was analyzed again. There was no significant difference in the two analyses.

During a transient run, the feed, bottoms, and distillate rates were maintained at the same values that they had at the initial steady-state. This was accomplished by maintaining a constant heating rate in the reboiler and a constant rate of bottoms product removal. Because the heats of vaporization of the components are not equal, the column was not completely in material balance. However, the changes in feed composition were kept small enough that the imbalance was negligible.

Chromatographic Procedure

In order to analyze the samples on the gas chromatograph, the chromatograph had to be started at least two hours before any samples were analyzed. This warm-up period was necessary to eliminate any baseline drift. To analyze the samples, a one micro-liter portion was injected into the chromatograph, with the results of the analysis being recorded on the recorder chart. A sample of the record from a chromatographic analysis is shown in Figure 5. The upper line on the record is the actual response of the chromatograph to the sample and the lower line is the output from the Disc Integrator. The number of up or down strokes the integrator pen makes is proportional to the area under the response curve with the proportionality constant being a function only of the instrument. Thus, the area fraction of each component in a sample can be obtained by dividing the area for the component by the sum of the areas for all of the components in the sample. The chromatograph used in this project was calibrated so that weight fractions could be determined directly from area fractions.

The chromatograph is the limiting factor on the accuracy of the experimental work, and the Disc Integrator is the limiting factor on the chromatograph. The integrator is the limiting factor because the areas under the response curve can be determined only to ± 0.10 square units. Since the areas of the components are normally on the order of ten square units, the resulting error in the area fractions will be on the order of 1.0 weight per cent. This is approximately the error that was obtained in the calibration tests for the chromatograph. The calibration tests are discussed in Appendix B.

After the results were obtained from the chromatograph analyses, they were analyzed further on the I. B. M. 7040 digital computer. The

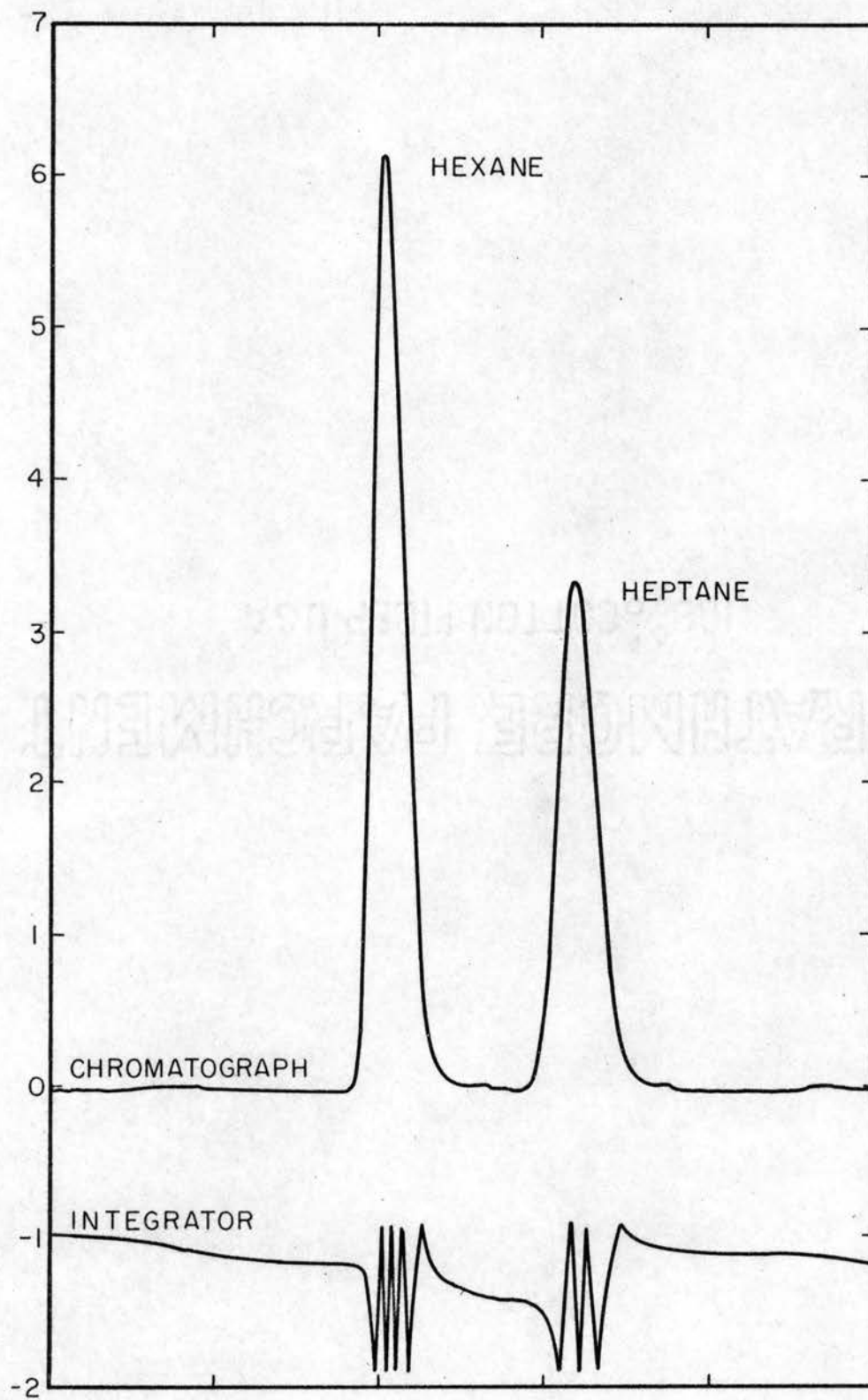


Figure 5. A Sample Chromatograph Record

computer analyses consisted of the following operations: converting the experimental flow rates from a volumetric to a molar basis; performing heat balancing calculations on the column to determine internal stream flow rates and compositions; and using the initial conditions in the column and a program for the lumped parameter model to predict the transient performance of the column. Once the experimental and the calculational work were completed, both sets of data were converted to a reduced form. In the reduced form the compositions were expressed as fraction of total change completed. This fraction was calculated according to the equation

$$w = \frac{x(t) - x_0}{x_\infty - x_0} \quad (4)$$

In this reduced form, the experimental and calculated results were compared to show that the transient performance of the column could be represented with a first order lumped parameter model.

CHAPTER V

DETERMINATION OF TYPE OF CHANGE ENTERING COLUMN

To determine what type of change in feed composition was entering the distillation column a thermal conductivity detector was constructed. The detector was designed to monitor the difference between the rates at which heat was removed from two tungsten heating elements. One of the heating elements was placed in a reference bath while the second element was placed in the feed stream to the distillation column.

The operating principle of the detector is that streams having different compositions will have different thermal conductivities; thus, the rates at which heat is removed from the heating elements will be different. This principle is true only at low flow rates where the removal of heat from the heating element is primarily through conduction. At higher flow rates, convective heat transfer becomes more significant than conductive and the thermal conductivity detector becomes useless.

As the rate of heat removal from the heating element changes, the temperature of the heating element changes. As the thermal conductivity decreases, the temperature of the heating element increases; and conversely, the temperature decreases as the thermal conductivity increases. The change in the temperature of the heating element causes its electrical resistance to change. For tungsten, which was used in the heating elements, the resistance increases with increasing temperature. The changing resistance causes a corresponding change in the current flowing through

the heating element. Since the temperature of the reference element remains unchanged there is a change in the difference in the voltage drops through the two heating elements. The change in the difference in the potentials can be measured with a potentiometer. This measured change can be plotted as a function of time to determine the change in the thermal conductivity of the feed stream. Thus, the change in the composition of the feed stream can be monitored. The equipment used consisted of a matched pair of Gow-Mac tungsten heating elements, a Bristol Dynamaster recording potentiometer, and a F & M variable voltage power supply.

A series of three different types of experiments were performed. The object of the first type was to analyze the change in feed composition in the feed system with the system as it was during the operating runs. The feed system under these conditions is shown in Figure 3. The second type was one in which the constant head tank was removed from the feed system, but the remainder of the system was left unchanged. Finally, in the third type, the feed filter and the feed pump were removed from the feed system, and the tubing for the feed system was arranged to provide a gravity feed. The object of the second type was to measure the effect of the constant head tank on the profile of the change in the feed composition. The object of the third type was to determine the effect of the feed pump and filter on the profile of the change in feed composition.

The results of the three types of experiments are shown in Figure 6. The results of the first type show that the change in the feed composition entering the column is not a step function, but is a ramp function which takes from fifteen to forty-five seconds to enter the column. Considering

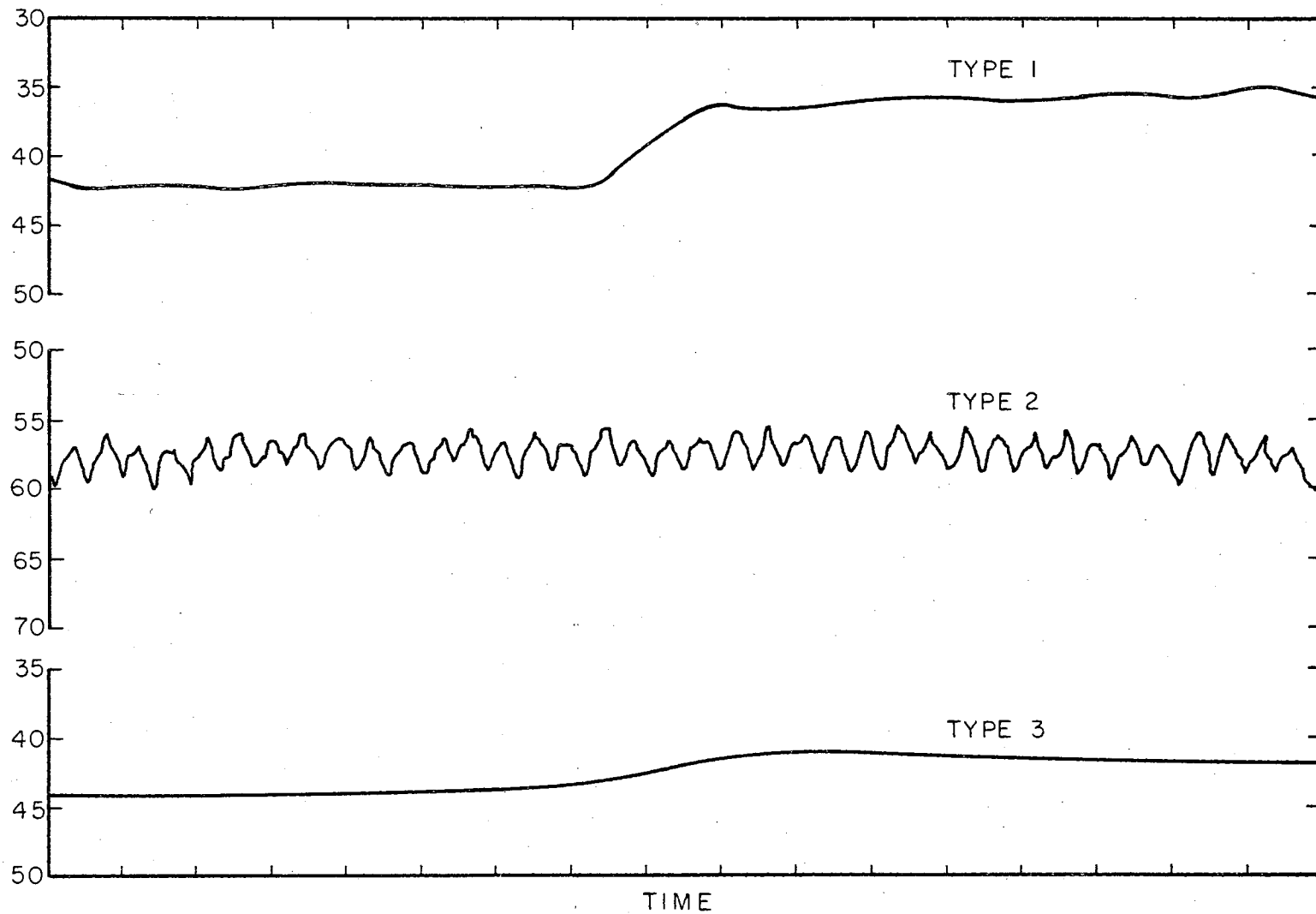


Figure 6. Results of the Feed Profile Determination

that the column requires approximately twenty minutes to change from the initial steady-state to the final steady-state, a forty-five second ramp change in feed composition is less than four per cent of the total time required for the transient change. If only fifteen seconds are required for the change in feed composition to enter the column, the time for the feed change to enter the column is approximately one per cent of the time required for the transient change.

The second type of experiment was performed to determine the effect of the constant head tank on the profile of the change in feed composition. The experiment did not give this information, but it did establish the effectiveness of the constant head tank in removing the pulses due to the peristaltic action of the feed pump. As can be seen in Figure 6, the recorder pen trace is relatively smooth during the first type of experiment, but during the second the trace is wavelike in appearance. The frequency of the wavelike trace is the same as the cycle speed of the pump, ten cycles per minute. Thus, the constant head tank appears to damp out the wavelike action of the pump to the point where it is not detectable.

The results of the third type of experiment indicated that the feed pump, the feed filter, and the constant head tank did not have a significant effect on the profile of the change in feed composition. From the trace shown in Figure 6, the time required for the change in feed composition to enter the column was still approximately fifteen to forty-five seconds even with the gravity feed.

The results of the series of experiments concerning the type of change in feed composition entering the column indicate that the only way that a true step change in feed composition could have been obtained

would have been to install two parallel feed systems. With parallel feeds an instantaneous change from one feed system to the other would give the step change. However, there would exist the possibility of introducing an additional type of perturbation into the column, an undesired change in the feed rate. The change in feed rate would result from the difficulty in matching the flow rates in the two different systems. If there was an additional perturbation introduced by the change in feed rates, it would be impossible to separate the transient response due to the change in feed rate from the transient response due to the change in feed composition. Since the object of the project was to determine the effect of a step change in feed composition and to measure the transient response to the step change, masking of the desired change could make the objective unobtainable.

Comparison of Predicted Results for a Step Change and a Ramp Change in Feed Composition

To determine the effect of a ramp change on the predicted transient behavior of a distillation column, the transient distillation program was modified to include a fifteen second ramp change in feed composition. The modified program was used on eight of the experimental runs and in all cases the difference between the results predicted for the step change and those predicted for the ramp change was less than 0.0001 mole fraction. This difference is much less than is detectable with the analytical equipment used in this project.

Considering the results obtained concerning the profile of the change in feed composition and the effect of a fifteen second ramp change on the predicted transient behavior, the assumption that the column is reacting

to a step change in feed composition appears to be a valid assumption. Thus, the data obtained on the transient performance of the distillation column can be considered to be the result of a step change in feed composition.

CHAPTER VI

DISCUSSION OF RESULTS

The experimental work was designed with two objectives in mind. The first objective was to obtain experimental data on the transient behavior of a distillation column. The other was to determine if the dynamic behavior of the column could be described with a lumped parameter model. The lumped parameter model that was used is derived and discussed in Appendix C.

To accomplish the first objective, twelve experimental runs were made. Nine of the runs were on binary systems and three on ternary systems. Of the nine binary runs, eight were performed with the hexane-heptane system, and one with the benzene-toluene system. The ternary runs were made with the benzene-toluene-p-xylene system. In the binary runs the range of feed compositions was from twenty-five to seventy-five mole per cent of the most volatile component. The range of feed compositions in the ternary runs was from thirty-five to fifty mole per cent benzene, twenty to thirty-five mole per cent toluene, and twenty to forty mole per cent p-xylene. The range of liquid holdups was from twenty-five per cent of the column volume to flooding. The feed rates ranged from 0.036 moles per hour to 0.058 moles per hour. The ratio of the distillate rate to the feed rate ranged from 0.20 to 0.65.

The results of the experimental work are summarized in Tables I, II, and III. Table I contains the feed compositions and column holdups for

TABLE I
SUMMARY OF EXPERIMENTAL RUNS

Run Number	Column Holdup, ml.	Per Cent of Column Volume Filled with Liquid	Initial Feed Composition	Final Feed Composition
105	237	158	0.350	0.431
106	60	40	0.353	0.395
107	40	27	0.337	0.301
109	40	27	0.275	0.404
110	83	55	0.731	0.464
111	63	42	0.675	0.516
115	40	27	0.454-B 0.217-T	0.441-B 0.228-T
116	40	27	0.505-B 0.222-T	0.469-B 0.320-T
117	40	27	0.427-B 0.281-T	0.368-B 0.241-T
130	50	33	0.749	0.627
131	54	36	0.742	0.631
132	126	83	0.540	0.758

TABLE II

SUMMARY OF EXPERIMENTAL FEED RATES AND DISTILLATE-TO-FEED RATIOS

Run	<u>Initial</u>		<u>Final</u>	
	Feed Rate Moles/hr.	D/F	Feed Rate Moles/hr.	D/F
105	.0581	.377	.0594	.380
106	.0489	.438	.0479	.465
107	.0436	.415	.0473	.420
109	.0388	.528	.0399	.432
110	.0408	.409	.0393	.342
111	.0386	.510	.0378	.512
130	.0441	.650	.0432	.608
131	.0465	.543	.0466	.562
132	.0493	.426	.0497	.570
115	.0370	.559	.0416	.548
116	.0360	.505	.0375	.507
117	.0462	.239	.0416	.183

TABLE III
SUMMARY OF PRODUCT COMPOSITIONS

Run	Bottoms Compositions		Distillate Compositions	
	Initial	Final	Initial	Final
105	.159	.248	.704	.776
106	.077	.126	.695	.720
107	.126	.022	.706	.529
109	.008	.006	.551	.674
110	.575	.334	.919	.780
111	.459	.387	.862	.808
130	.464	.278	.909	.850
131	.542	.349	.902	.850
132	.354	.561	.803	.905
115-B	.045	.038	.804	.788
115-T	.345	.403	.141	.162
115-X	.610	.559	.055	.050
116-B	.100	.085	.808	.800
116-T	.376	.437	.143	.154
116-X	.524	.478	.049	.046
117-B	.262	.230	.813	.803
117-T	.342	.266	.136	.127
117-X	.396	.504	.051	.070

the runs, and Table II contains the feed rates and the experimental distillate to feed ratios. The experimental product compositions at the initial and final steady-states are given in Table III. A complete listing of the experimental data appears in Appendix D. The compositions of the product streams are presented in both tabular and graphical form. The graphical presentations are of the compositions plotted as a function of time.

To determine the reproducibility of the transient behavior of the column, two runs were made with feeds of the same composition. The steady-state product compositions were different because the distillate to feed ratios were different. There was a difference in distillate to feed ratios because of difficulty in controlling the flow rate. However, when the compositions are displaced so that the initial steady-state compositions coincide, the transient curves are almost identical. The compositions in Run 130 were displaced by adding the difference between the initial steady-state compositions in Runs 130 and 131 to the measured compositions in Run 130. The results of this reproducibility study are shown in Figure 7.

A series of calculations was made to determine the accuracy of the experimentally determined flow rates. To do this the steady-state product compositions were accepted as being correct, and the distillate and bottoms rates necessary to give these compositions were calculated. The results of these material balance calculations are given in Table IV. The standard deviation of the difference between the experimental and calculated distillate rates at the initial steady-state was 0.002 moles per hour. The mean distillate rate was 0.0215 moles per hour. The binary runs were used to compare the distillate rates because the rates can be

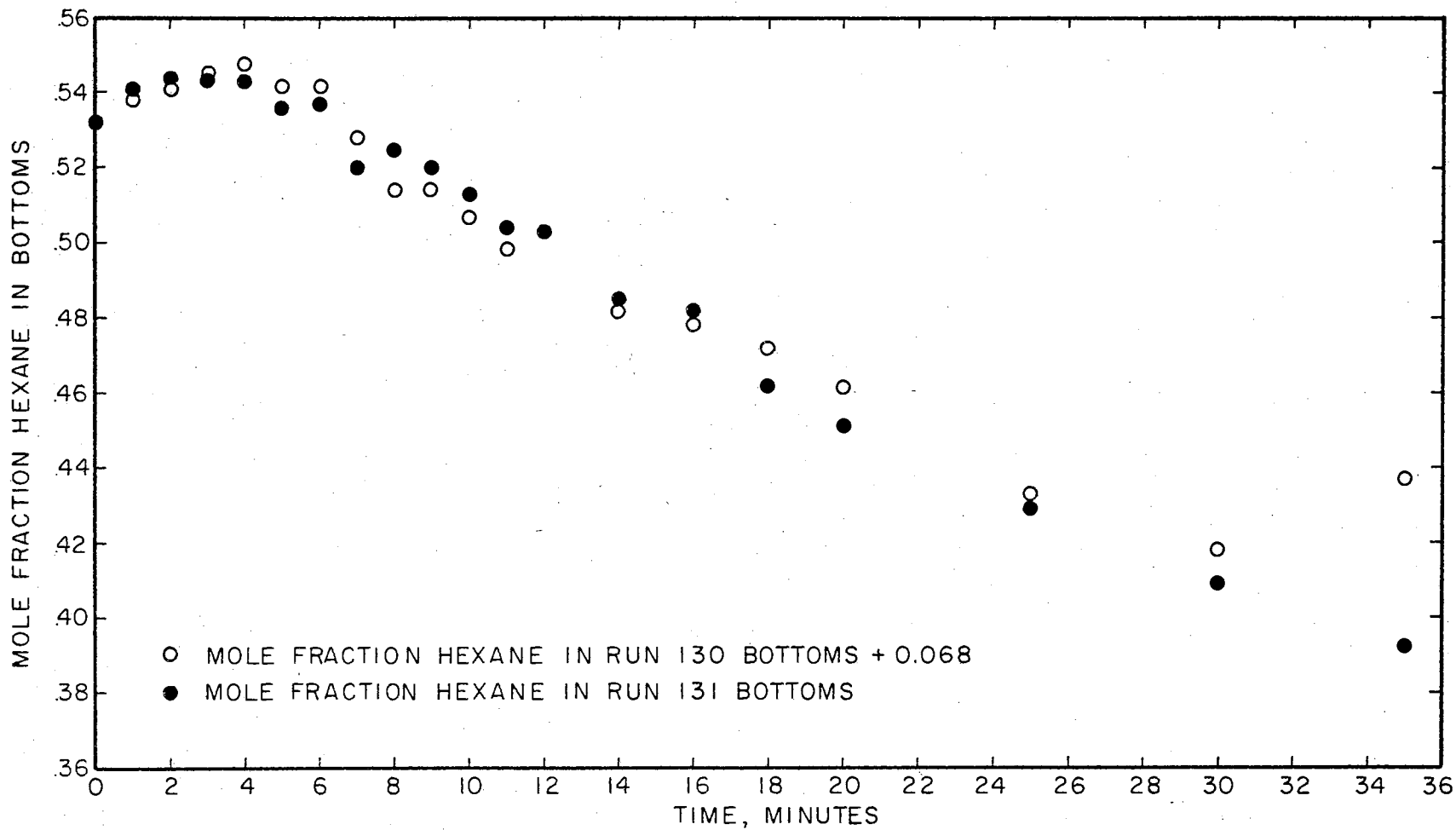


Figure 7. Results of Reproducibility Study

TABLE IV
COMPARISON OF EXPERIMENTAL AND CALCULATED DISTILLATE RATES

Run	<u>Initial Steady-State</u>		<u>Final Steady-State</u>	
	Experimental	Calculated	Experimental	Calculated
105	0.0220	0.0204	0.0216	0.0206
106	0.0214	0.0218	0.0223	0.0217
107	0.0181	0.0159	0.0199	0.0260
109	0.0205	0.0191	0.0212	0.0236
110	0.0167	0.0185	0.0134	0.0114
111	0.0196	0.0207	0.0193	0.0116
130	0.0286	0.0282	0.0263	0.0264
131	0.0253	0.0259	0.0262	0.0262
132	0.0210	0.0204	0.0283	0.0285
115	0.0208	0.0198	0.0228	0.0226
116	0.0179	0.0203	0.0190	0.0209
117	0.0108	0.0138	0.0075	0.0102

calculated exactly for a binary system, but can only be approximated for a multicomponent system.

The second objective of the experimental work was to determine how well the transient behavior of the column could be described with the lumped parameter model. The accomplishment of this objective is best discussed when considered in two parts: the ability of the model to predict the final steady-state compositions, and the ability of the model to predict the transient response curve in reduced dimensions.

The ability to predict the final steady-state product compositions was determined by finding the difference between the experimental final steady-state compositions and those predicted with the model. There are four methods which could be used to predict the final steady-state compositions. These are as follows: use the experimentally determined flow rates at both initial and final steady-states; use the experimentally determined values for the initial steady-state and assume that they are valid at both steady-states; use the calculated rates for both steady-states; or use the calculated rates at the initial steady-state and assume that they apply at both steady-states. Three of the four possible methods of correlation have been used in determining the ability of the model to predict the final steady-state compositions. The method assuming that the experimental initial steady-state flow rates were valid at both steady-states was not used. It was not used because if one of the experimental values is valid, both should be assumed to be valid.

The stream flow rates are necessary for the prediction of the final steady-state compositions because the column must be in an over-all material balance at the steady-state. The initial steady-state conditions are used to evaluate the separation parameter $J_{n,i}$. The dynamic model

and the predicted final steady-state compositions are highly dependent on $J_{n,i}$. Thus, an error in the flow rates causes an error in $J_{n,i}$ and an error in the predicted final steady-state compositions.

The results obtained using the experimentally determined flow rates are given in Table V. For some of the runs there is excellent agreement between the theoretical and the experimental steady-state compositions. The agreement appears to be best when the difference between the experimental and calculated distillate flow rates and the change in flow rates from one steady-state to the other are small. When either, or both, of these are large, there is poor agreement between experimental and calculated final steady-state compositions.

The calculational method that used the calculated flow rates at the initial steady-state and assumed that those rates applied during the transient period was accurate only when the experimental flow rates at both steady-states were approximately equal. The results of the calculations also appear in Table V. In general, the final product compositions obtained with the second method of calculation do not agree with the experimental compositions as well as those predicted with the experimental flow rates. The results indicate that the flow rates were not held constant during the transient period as was intended.

The best comparison between experimental and calculated final steady-state compositions was obtained when the calculated flow rates for both the initial and final steady-states were used. These results are given in Table V. The differences between the calculated and experimental final steady-state compositions are plotted as a function of the experimental composition change in Figure 8.

On the basis of the calculations of the final steady-state

TABLE V

COMPARISON OF CALCULATED AND EXPERIMENTAL FINAL STEADY-STATE COMPOSITIONS

Run	Bottoms Composition					Distillate Composition				
	Experimental		Calculated			Experimental		Calculated		
	Initial	Final	a	b	c	Initial	Final	a	b	c
	<u>Binary Runs</u>									
105	.159	.248	.233	.257	.257	.704	.776	.756	.755	.755
106	.077	.126	.102	.122	.116	.695	.720	.730	.732	.743
107	.126	.022	.144	.087	.012	.706	.529	.625	.676	.516
109	.008	.006	.078	.106	.053	.551	.674	.701	.703	.649
110	.575	.334	.230	.148	.294	.919	.780	.812	.806	.821
111	.459	.387	.275	.271	.400	.862	.808	.765	.762	.795
130	.464	.278	.260	.237	.269	.909	.850	.847	.838	.846
131	.542	.349	.379	.381	.378	.902	.850	.828	.830	.829
132	.354	.561	.556	.643	.553	.803	.905	.910	.921	.910
	<u>Ternary Runs</u>									
115-B	.045	.038	.061	.000	.000	.804	.788	.744	.721	.668
115-T	.345	.403	.407	.477	.449	.141	.162	.203	.199	.238
115-X	.610	.559	.532	.523	.551	.055	.050	.053	.081	.095
116-B	.100	.085	.164	.103	.102	.808	.800	.766	.770	.759
116-T	.376	.437	.445	.443	.454	.143	.154	.195	.194	.202
116-X	.524	.478	.391	.454	.444	.049	.046	.039	.036	.029
117-B	.262	.230	.273	.186	.221	.813	.803	.776	.803	.830
117-T	.342	.266	.268	.293	.287	.136	.127	.155	.124	.108
117-X	.396	.507	.459	.522	.492	.051	.070	.069	.073	.066

- Compositions calculated with experimental flow rates.
- Compositions calculated by determining the theoretical initial steady-state flow rates, assuming them to be valid during the transient period.
- Compositions calculated by determining the theoretical initial and final steady-state flow rates, and using both for calculations.

compositions and the comparisons between the calculated and experimental compositions, the lumped parameter model appears to predict the final steady-state compositions well if the flow rates are known accurately. From the results shown in Figure 8, the predictive ability of the model appears to be relatively good for changes in composition up to twenty-five mole per cent. The model has not been tested for changes above twenty-five mole per cent.

Three binary and two ternary runs have been selected to illustrate the ability of the lumped parameter model to predict the transient response curve. The results of the comparisons of the experimental and calculated transient response are shown in Figures 9 through 15. The number of illustrations of the ability of the model to predict the transient response was limited to five runs because if the model can be used successfully in those cases, it can be assumed to be equally valid for the other runs. This statement is based on a consideration of the equations used in the lumped parameter model and on the fact that the behavior of the column will be of the same order in all runs.

Consider the dynamic equations that can be written for a stripping column which is being analyzed with a lumped parameter model. Such a column is shown in Figure 16. There is a first order holdup in the distillate stream, the reboiler is assumed to be a first order holdup, and the holdup in the feed section is assumed to be negligible.

The differential equation for the composition of the distillate product can be written

$$\delta^D \frac{dy_{D,i}}{dt} = f_1 = D(y_{2,i} - y_{D,i}) \quad (5)$$

Since the vapor leaving the feed section is assumed to be in equilibrium

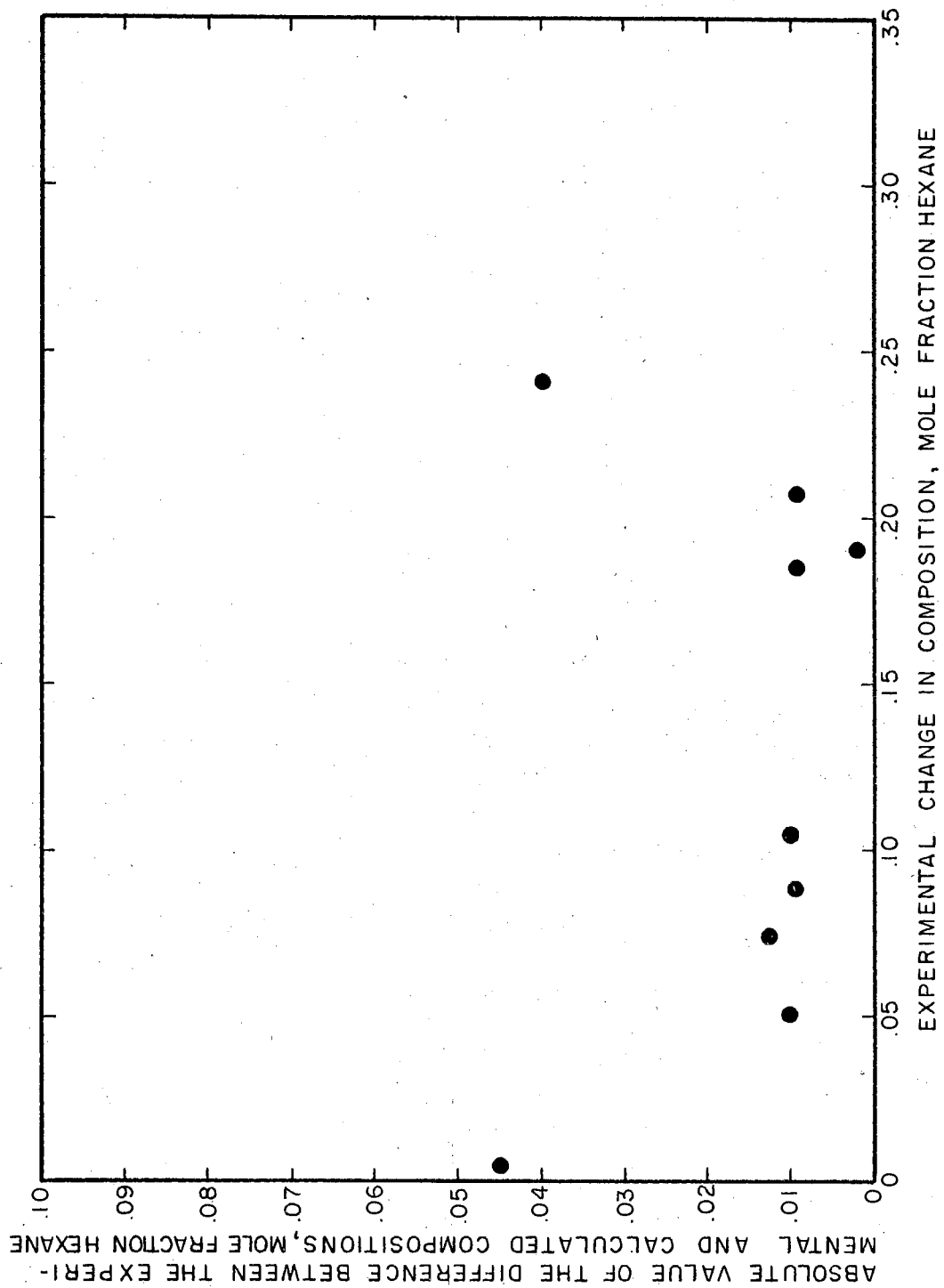


Figure 8. Comparison of Predicted and Experimental Changes in Compositions for Binary Runs

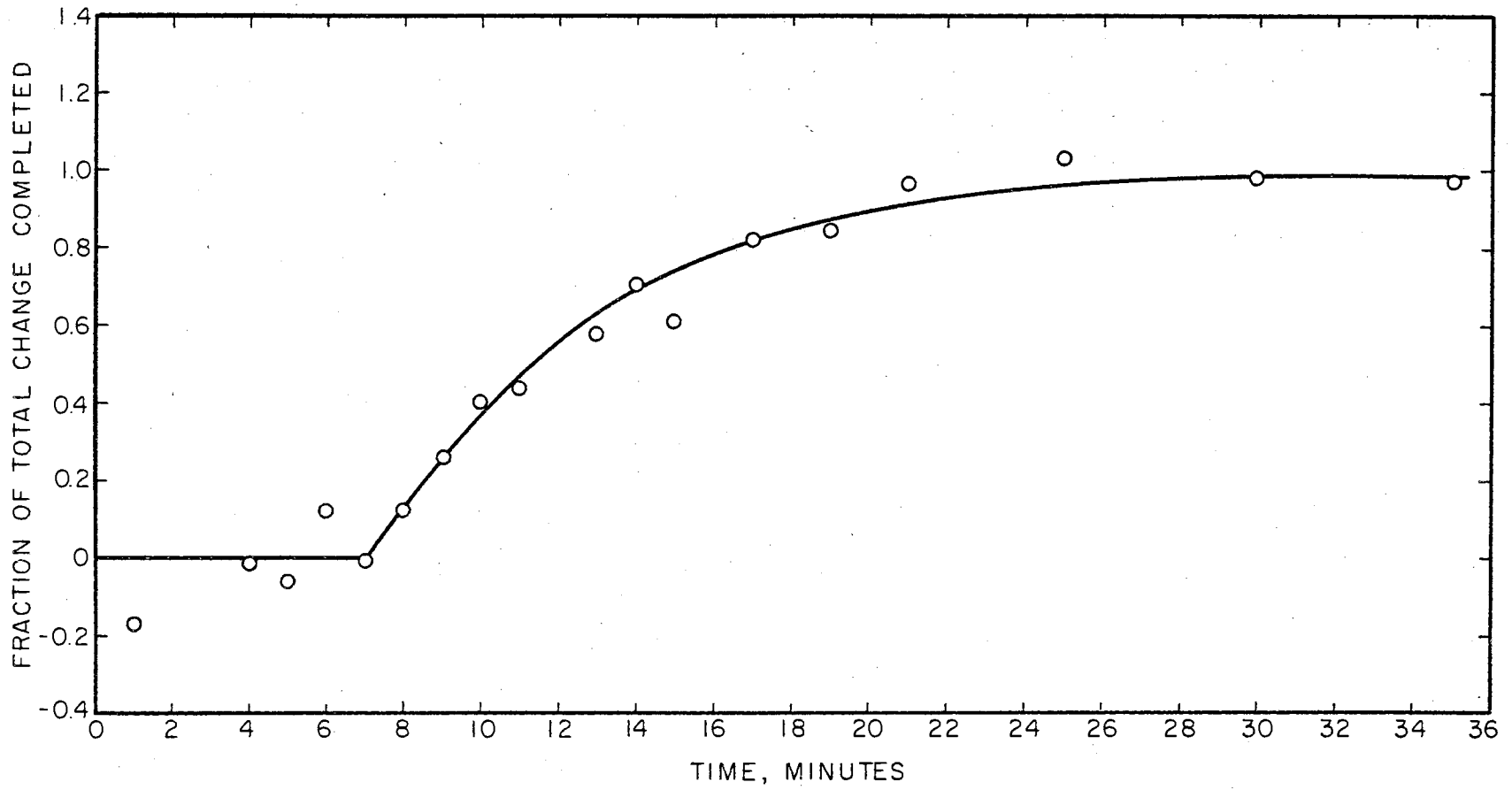


Figure 9. Transient Response Curve for Run 105

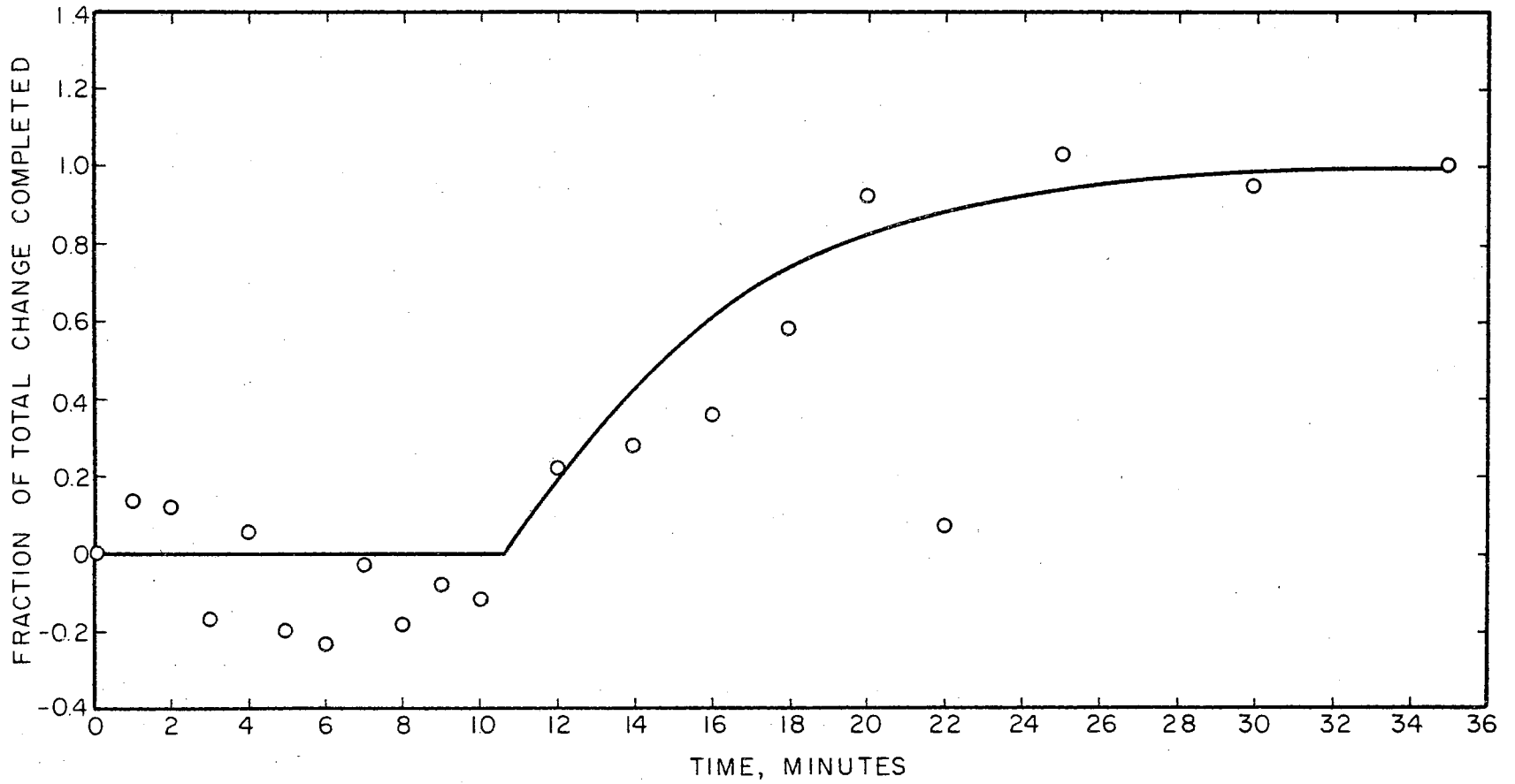


Figure 10. Transient Response Curve for Run 106

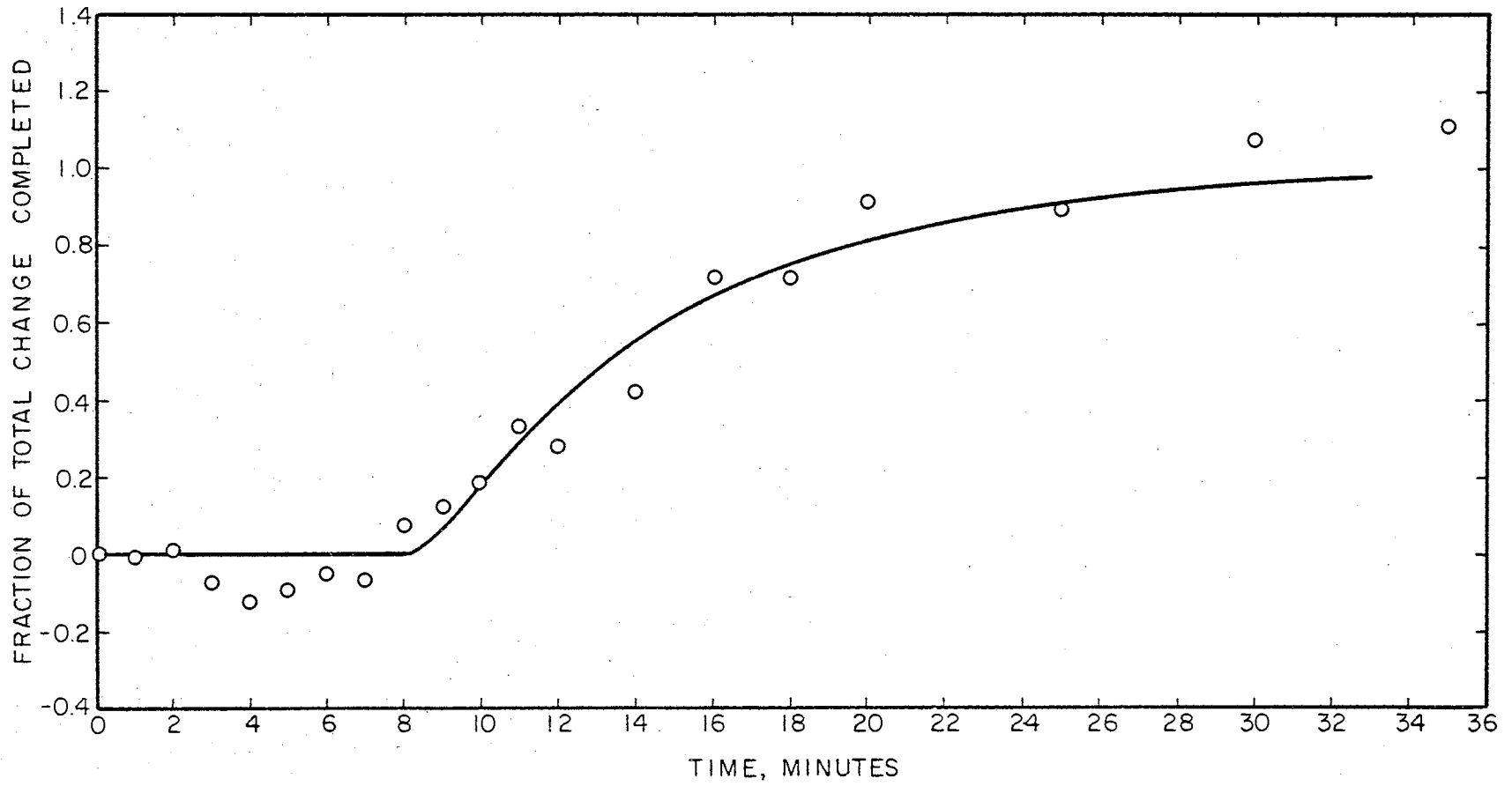


Figure 11. Transient Response Curve for Run 107

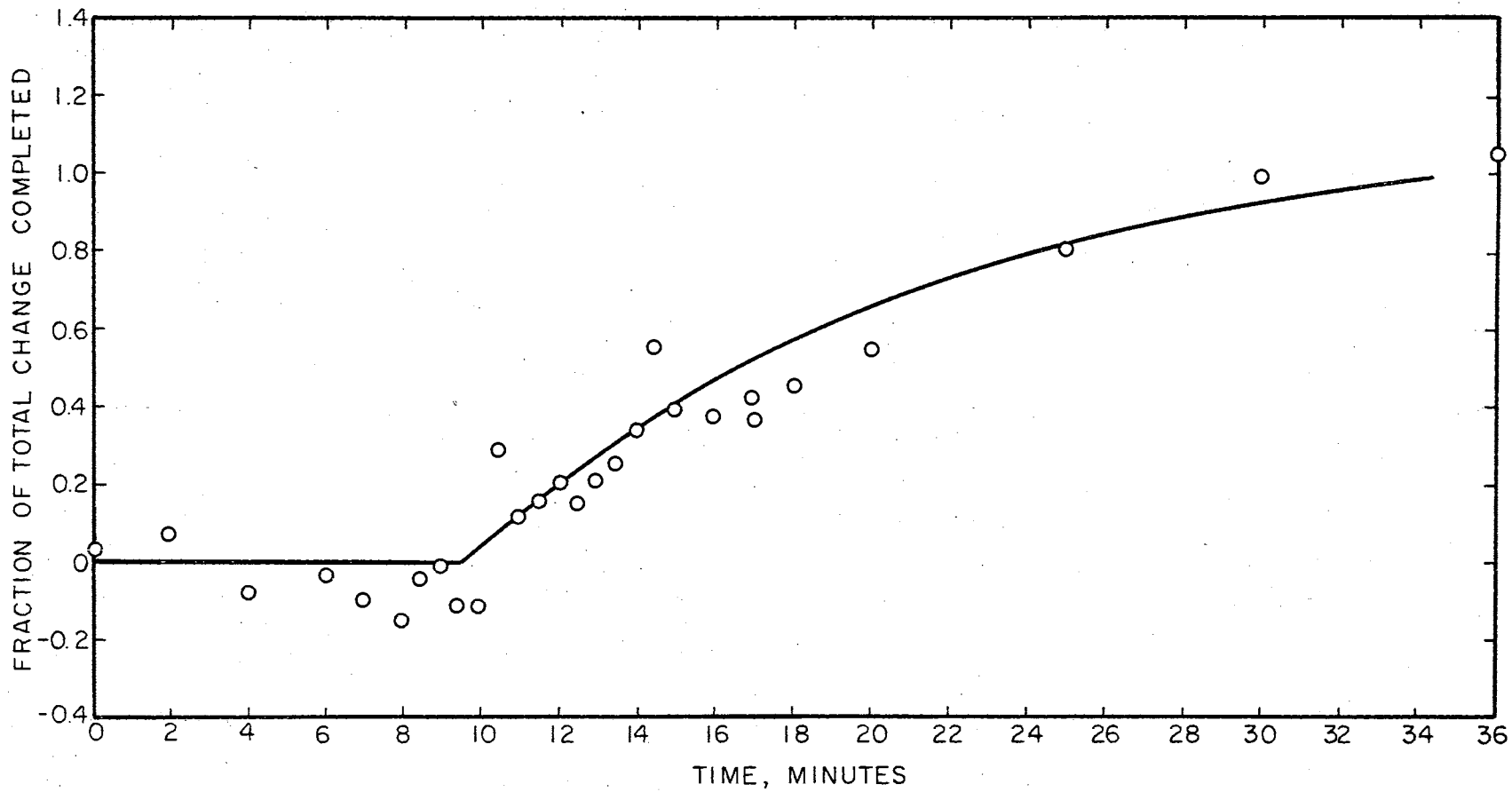


Figure 12. Transient Response Curve for Toluene in Run 116

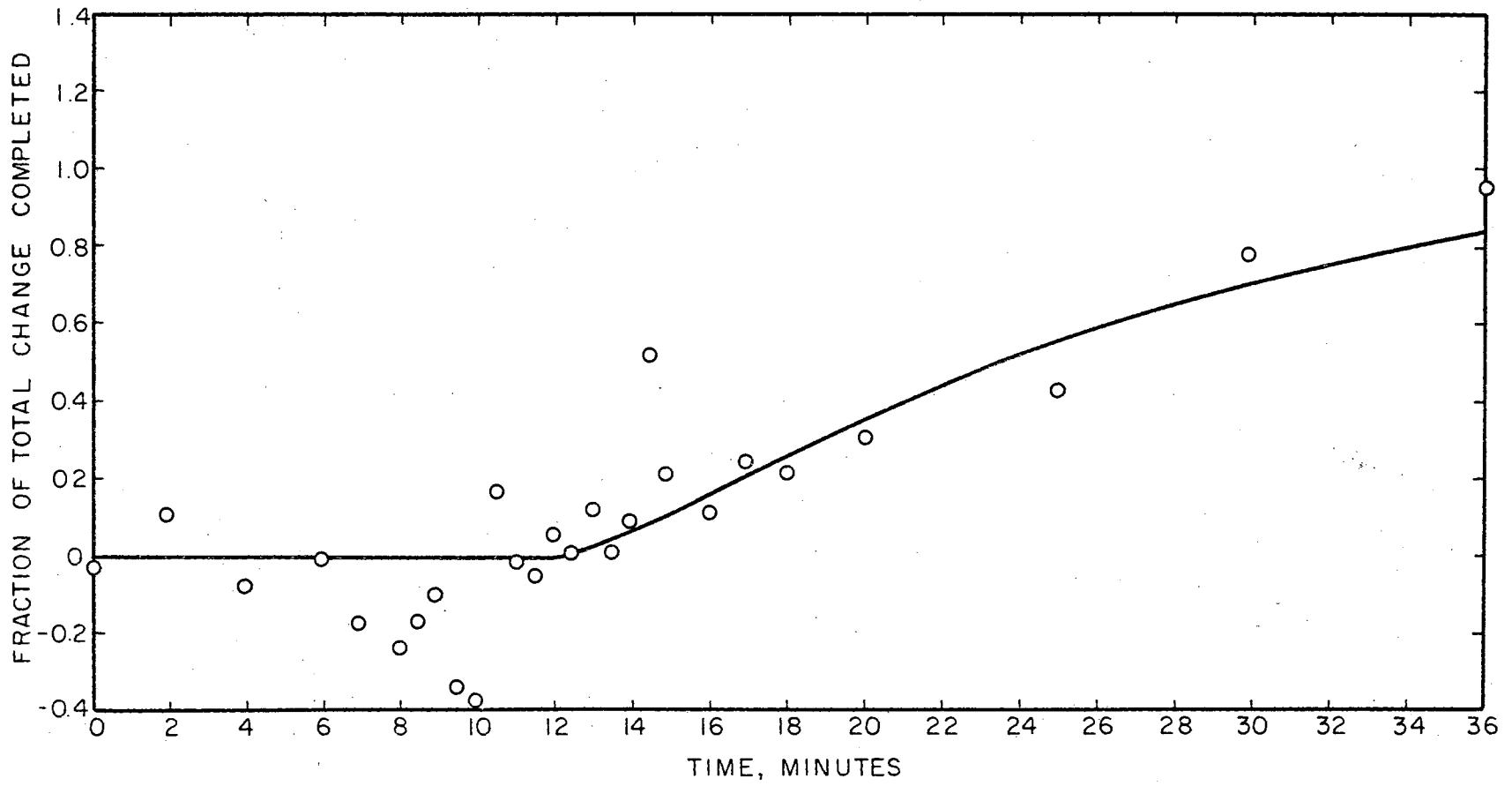


Figure 13. Transient Response Curve for Xylene in Run 116

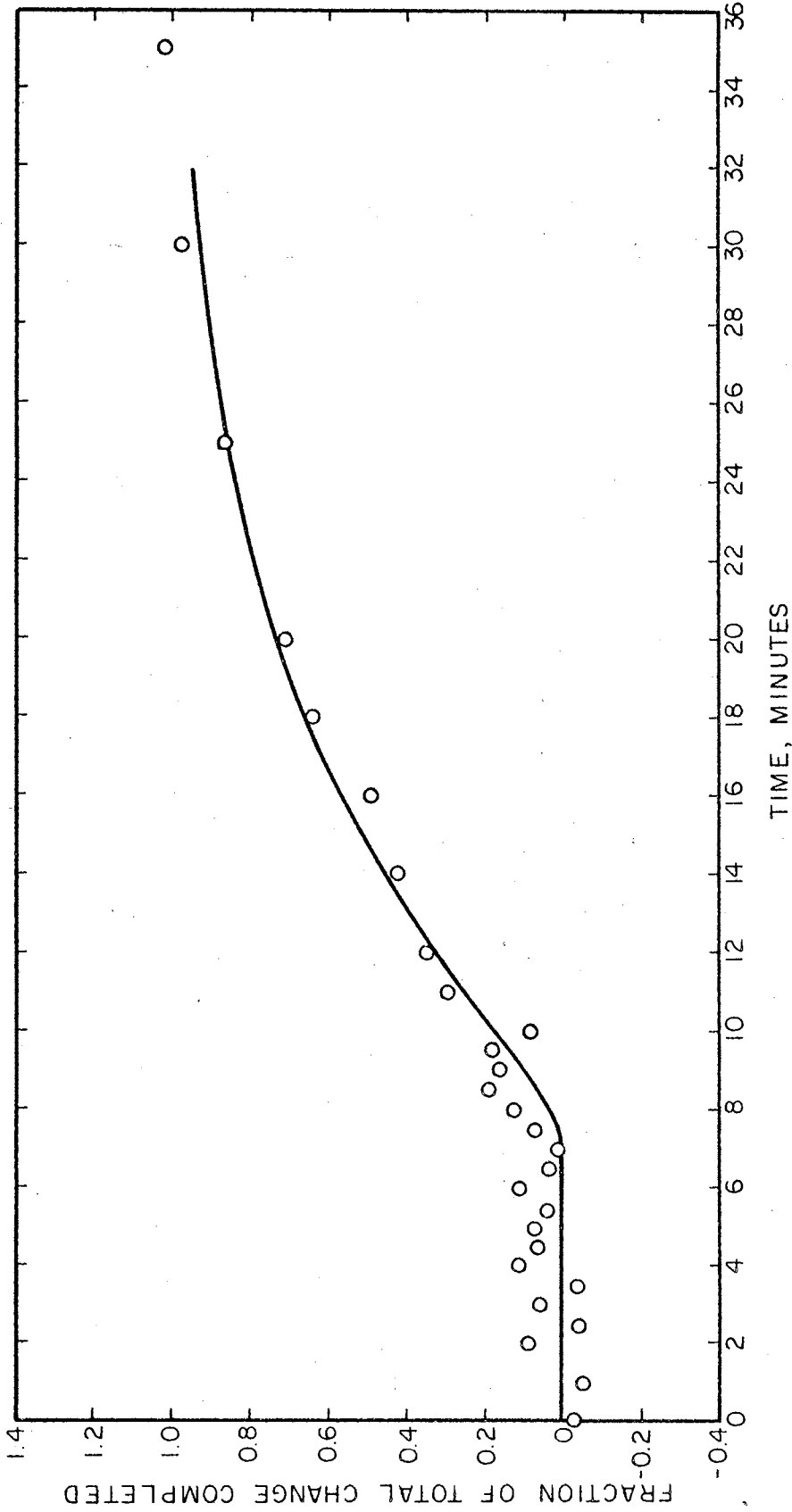


Figure 14. Transient Response Curve for Toluene in Run 117

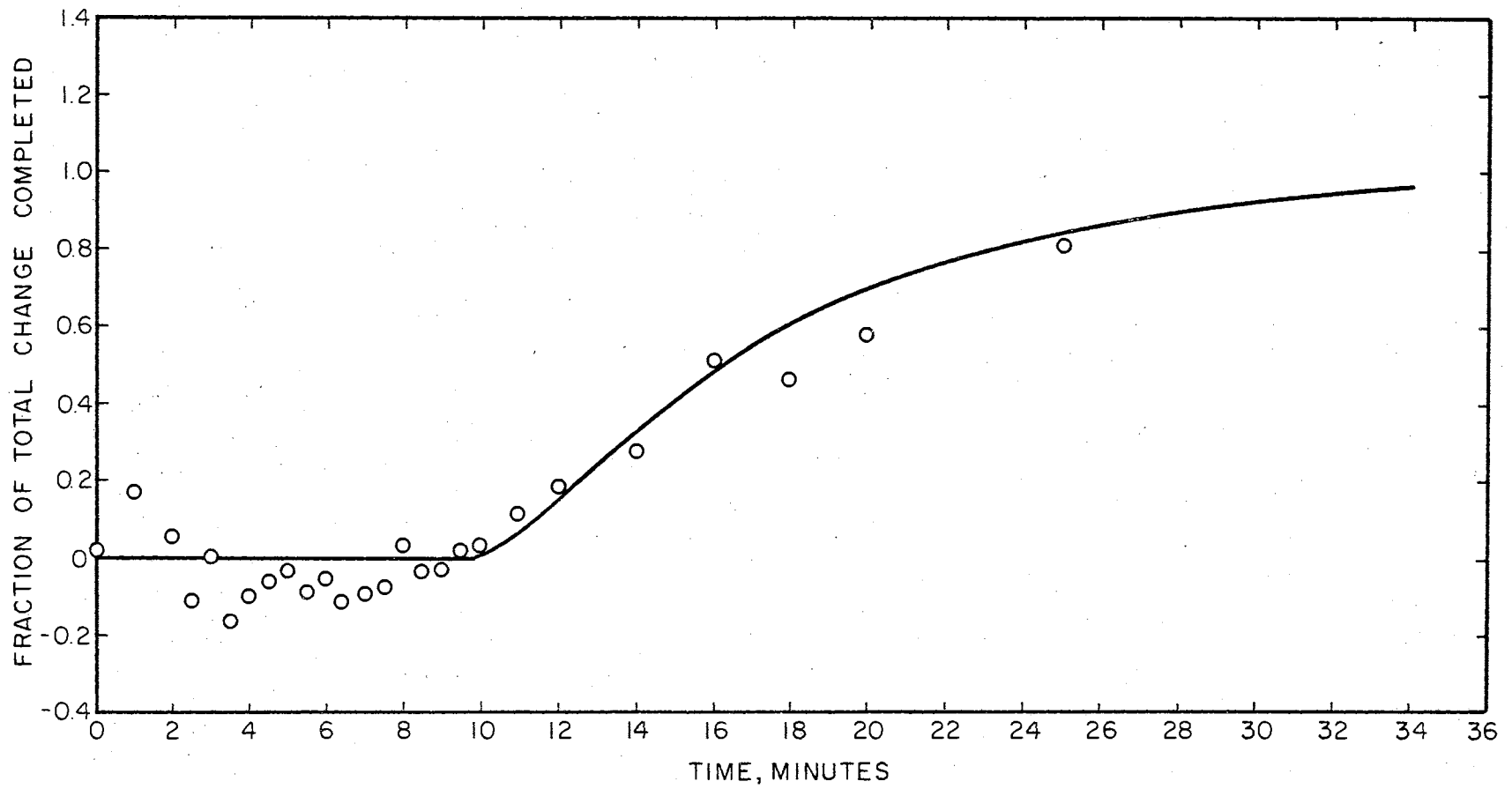


Figure 15. Transient Response Curve for Xylene in Run 117

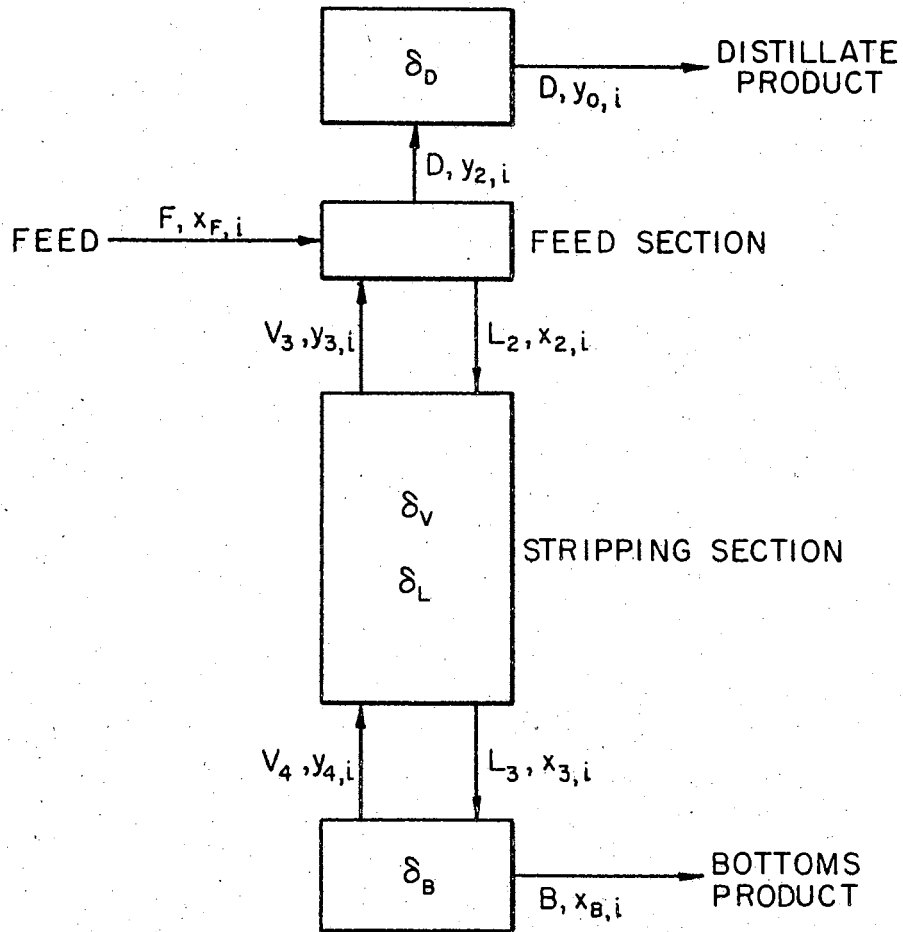


Figure 16. Schematic Diagram of a Stripping Column

with the liquid leaving the feed section, the vapor composition can be expressed as

$$\frac{dy_{2,i}}{dt} = f_2 = \frac{1}{(D + K_{2,i}L_2)} (Fx_{f,i} + V_3y_{3,i}) \quad (6)$$

Using the proposed dynamic model for the column, the compositions of the vapor and liquid streams leaving the stripping section can be written

$$\delta^V \frac{dy_{3,i}}{dt} = f_3 = [V_4y_{4,i} - V_3y_{3,i} + J_i(K_{2,i}x_{2,i} - y_{4,i})] \quad (7)$$

$$\delta^L \frac{dx_{3,i}}{dt} = f_4 = [L_2x_{2,i} - L_3x_{3,i} - J_i(K_{2,i}x_{2,i} - y_{4,i})] \quad (8)$$

Finally, the composition of the bottoms product is

$$\delta^B \frac{dx_{B,i}}{dt} = f_5 = [L_3x_{3,i} - (B + K_{4,i}V_4)x_{B,i}] \quad (9)$$

Since the model predicts accurately the final compositions of the product streams, the functions f_1 , f_2 , f_3 , f_4 , and f_5 can be considered to be correct. Thus, the dynamic behavior of the column can be described by the equations

$$\frac{dy_{D,i}}{dt} = f_1 \frac{1}{\delta^D} \quad (10)$$

$$\frac{dy_{2,i}}{dt} = f_2 \quad (11)$$

$$\frac{dy_{3,i}}{dt} = \frac{f_3}{\delta^V} \quad (12)$$

$$\frac{dx_{3,i}}{dt} = \frac{f_4}{\delta^L} \quad (13)$$

$$\frac{dx_{B,i}}{dt} = \frac{f_5}{\delta^B} \quad (14)$$

where the parameters δ^D , δ^V , δ^L , and δ^B are holdup terms which are

not functions of time. Since the functions f_1 , f_2 , f_3 , f_4 and f_5 control the final compositions of the streams, the holdup terms merely serve as time constants. Consequently, if the holdup terms can be adjusted so as to predict the transient response curve for several of the runs, the ability of the model to reproduce the curve can be extended to all of the runs.

If the lumped parameter model is to be used for control purposes or in the design of control systems, there should be some information on how the measured column holdup compares with the value of the lumped parameter that is required to reproduce the transient response curve. An effort was made to make this comparison with the experimental data obtained in this project. The effort was not wholly successful in that only order of magnitude comparisons were possible.

The lack of success in comparing the holdups was due to the difficulty in determining the reboiler holdup. The reboiler holdup is significant because it is approximately five times the column holdup. This difference in sizes results in the effect of the column holdup being masked by the reboiler holdup. Since the reboiler can be accurately represented by a first order equation, no difficulties would have arisen if the holdup was accurately known. However, the holdup of approximately five-hundred milliliters was known only to plus or minus one-hundred milliliters. This inaccuracy was due to the difficulty in determining and controlling the product flow rates.

Due to the effect of the reboiler only order of magnitude comparisons between the holdups were made. For runs where the column holdup was close to flooding, a large value of the lumped parameter was required (seventy per cent of the column volume for Run 105), and for runs with a

low holdup, a smaller value of the lumped parameter was required (twenty-five per cent of the column volume for Run 106 and fifty per cent for Run 107).

In Run 105 the column was incipient to flooding. The liquid holdup in Runs 106 and 107 were forty and twenty-seven per cent of the column volume, respectively. No definite conclusions can be made from these comparisons, but a justifiable preliminary one seems to be that the value of the lumped parameter required to reproduce the transient response curve is approximately the same as the column volume.

To complete the analysis of the dynamic behavior of the column, the dead time was estimated. The dead time is the period between the time a perturbation enters the column and the time the bottoms product begins to respond to the perturbation. The distillate was found to respond almost instantaneously when a perturbation entered the column. Thus, the dead time was determined by finding the difference between the time of the beginning of the distillate response and the beginning of the bottoms response. An accurate determination of the dead time was not possible because of the errors in the sample analyses and errors in the determination of the times for the beginning of the responses. However, they were estimated to be approximately three minutes.

Deam (8) has commented that fractionation occurs between the column and the condenser with the equipment used in this project. The effect of the fractionation is to invalidate the assumption that the distillate is in equilibrium with the liquid leaving the feed section. The distillate will be richer in the more volatile component than the vapor leaving the feed section.

To consider the effect of fractionation between the column and the

condenser, assume that:

1. The fractionation that occurs between the column and the condenser can be described by the lumped parameter model.
2. The separation parameter, J_D , can be obtained from the initial steady-state operating conditions according to

$$J_D = \frac{Dy_D - V_F y_f}{y_D - y_f} \quad (15)$$

where the section between the column and the condenser can be shown schematically as in Figure 17.

3. The parameter J_D does not change during the transient period.
4. The flow rates in the section between the column and the condenser do not change significantly during the transient period.

Using the lumped parameter model and these assumptions, the dynamic behavior of the distillate composition can be expressed as

$$\delta \frac{V}{D} \frac{dy_D}{dt} = V_f y_f - Dy_D + J_D (y_D - y_f) \quad (16)$$

At a steady-state operating condition, the derivative of the distillate composition with respect to time is zero. Thus,

$$V_f y_f - Dy_D + J_D (y_D - y_f) = 0 \quad (17)$$

Equation (17) can be rearranged to give

$$\frac{y_D}{y_f} = \frac{V_o - J_D}{D - J_D} \quad (18)$$

Thus, if the above assumptions are true, the ratio y_D/y_f is a constant. If, in addition, the lumped parameter model is valid for the column itself, the predicted steady-state values for the distillate composition will be correct. Consequently, even though fractionation occurs between the column and the condenser, the effect on the difference between the

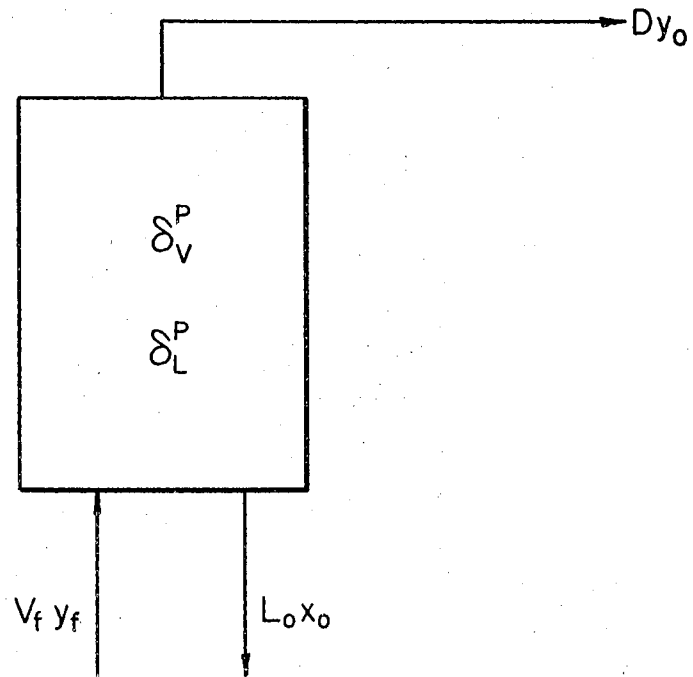


Figure 17. Schematic Representation of Holdup Between Column and Condenser

predicted and experimental final steady-state distillate compositions should be small. This conclusion is supported by the experimental results in which the final steady-state compositions were accurately predicted by the lumped parameter model.

According to the lumped parameter model, the holdup between the column and the condenser acts as a first order time constant. The magnitude of this time constant cannot be determined; however, its effect will be to delay the response of the distillate composition to the upset in the column operation. This effect was observed in the experimental data when the distillate did not respond instantaneously, as was predicted by the lumped parameter model with no holdup.

The experimental results obtained have shown that the dynamic behavior of a distillation column can be adequately described by a lumped parameter model. The ability of the model to predict the final steady-state compositions is dependent of the magnitude of the change in column operating conditions and on the accuracy with which the flow rates are known at the final steady-state. This prediction of the final steady-state compositions has been shown to be highly dependent on stream flow rates. The ability of the model to reproduce the transient response curve is determined by a dead time and four first order holdup terms. The transient response curve, which is determined by the equipment and the holdups in the equipment, has been reproduced for five of the runs. On the basis of these five comparisons and a study of the dynamic equations for transient curve has been extended to all of the runs. Study of the experimental data for all of the runs shows that this is a warranted extension.

CHAPTER VII

DEVELOPMENT OF A FEED FORWARD CONTROL MODEL

In recent years the control of distillation columns has received increasing attention from chemical engineers. The classical approach to the control problem has been to use what is known as feed back control. In feed back control no action is taken to correct an upset in column operations until the upset is noticed in the product streams. When an upset is detected, action is taken to return the system to the desired operating condition. This control scheme works well on systems where there are few upsets. However, if there are frequent upsets in column operation, a substantial amount of time is spent at undesired operating conditions. For such systems, the control scheme known as feed forward control should be used.

In principle, feed forward control is preferable to feed back control because disturbances in the quality of the products from a distillation column can be minimized. According to the principles of feed forward control, a disturbance is detected before it enters the column and corrective action is begun as the disturbance enters the column. The operating variables are manipulated so that the product does not change from the desired value. The advantages of feed forward control are obvious. First, the column can be operated with less margin for error. This allows production of more specification product from a given amount of feed. Second, the column can be operated at the desired

condition at all times. Thus, there will be no production of poor quality product which must be either up-graded or recycled through the column.

While in principle feed forward control appears to be the answer to all quality control problems, there are numerous drawbacks. These drawbacks are: requirement of in-plant computers; requirement of a reliable dynamic model of a distillation column; and requirement of a control system which has a small response time. The necessity of having a control system which has a low response time is independent of the other two requirements and will not be discussed further. The other two requirements are closely linked and must be considered together.

The type of in-plant computer that is required is determined primarily by the dynamic model that is used. When a disturbance is detected, the corrective action must be begun as the upset enters the column. Thus, the combination of computing speed and model simplicity must be such that the initial corrective action is available immediately. If a dynamic model that is based on the plate concept is used, a computer approximately the size of an I. B. M. 7094 is required. Such a computer is seldom, if ever, installed as an in-plant computer. On the otherhand, if a lumped parameter model based on the section concept is used, a much smaller computer can be used. Conceivably, a computer the size of an I. B. M. 1620 would suffice. Since the lumped parameter model has been shown to accurately describe the transient behavior of a distillation column, it appears to be sufficient for the feed forward model.

The feed forward model that is discussed here will be for a stripping column in which the feed section holdup is assumed to be negligible. Such a column is shown in Figure 18. For the feed section, the heat and material balances can be written

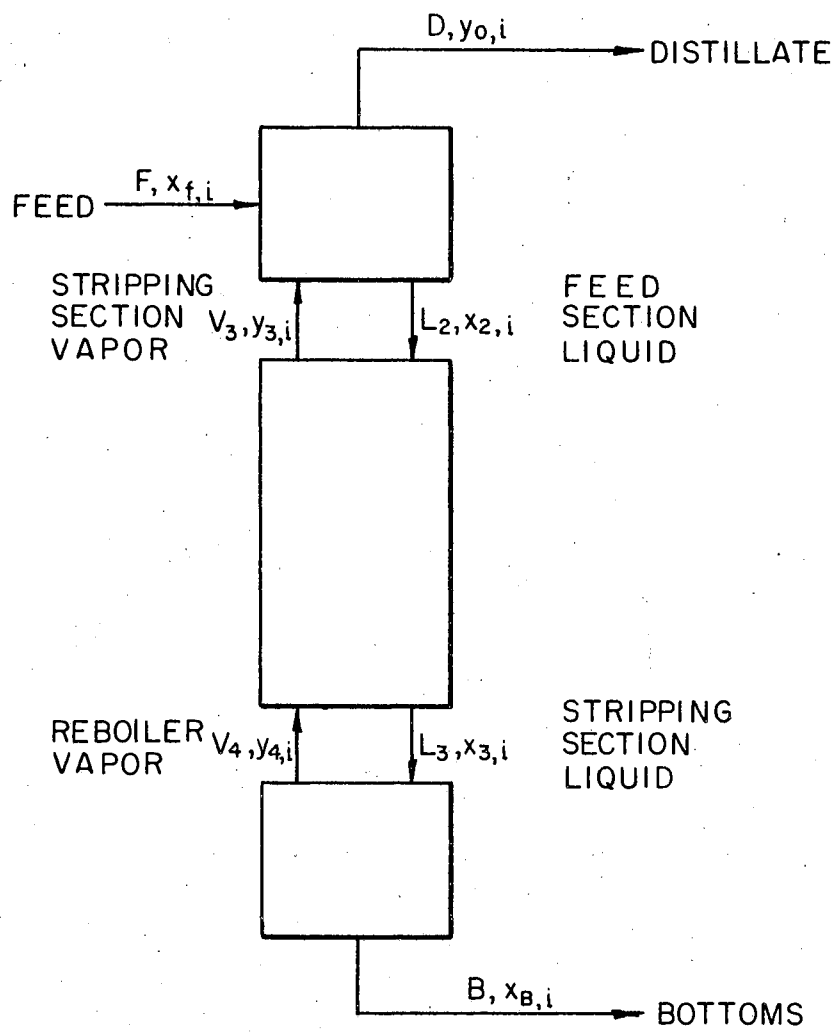


Figure 18. Stripping Column

$$V_3 + F = D + L_2 \quad (19)$$

$$V_3 y_{3,i} + F x_{f,i} = D y_{D,i} + L_2 x_{2,i} \quad (i = 1, 2, \dots, N) \quad (20)$$

$$V_3 H_3 + F h_f = D H_D + L_2 h_2 \quad (21)$$

Since holdup in the feed section has been assumed to be negligible, equations (19), (20), and (21) are valid at all times. In a stripping column, the distillate is in equilibrium with the liquid leaving the feed section if the feed section is assumed to be a theoretical stage. Thus, the distillate and the liquid leaving the feed section are related according to the equation

$$y_{D,i} = K_{2,i} x_{2,i} \quad (i = 1, 2, \dots, N) \quad (22)$$

Assuming that the column is at a thermal steady-state at all times, the following equations can be written for the stripping section at any time

$$L_2 + V_4 = L_3 + V_3 \quad (23)$$

$$L_2 h_2 + V_4 H_4 = L_3 h_3 + V_3 H_3 \quad (24)$$

Since the stripping section has appreciable holdup, it will not necessarily be at steady-state with respect to the component flow rates. Consequently, a dynamic model must be used to determine the compositions of the streams leaving the stripping section. Accordingly, the compositions can be expressed as

$$x_{3,i} = \int_0^t \left(\frac{dx_{3,i}}{dt} \right) dt + x_{3,i}(0) \quad (i = 1, N) \quad (25)$$

$$y_{3,i} = \int_0^t \left(\frac{dy_{3,i}}{dt} \right) dt + y_{3,i}(0) \quad (i = 1, N) \quad (26)$$

Using the first order lumped parameter model, the derivatives of the

liquid and vapor compositions can be written

$$\frac{dx_{3,i}}{dt} = \frac{1}{\delta L} [L_2 x_{2,i} - L_3 x_{3,i} - J_i (K_{2,i} x_{2,i} - y_{4,i})] \quad (i = 1, N) \quad (29)$$

$$\frac{dy_{3,i}}{dt} = \frac{1}{\delta V} [V_4 y_{4,i} - V_3 y_{3,i} + J_i (K_{2,i} x_{2,i} - y_{4,i})] \quad (i = 1, N) \quad (28)$$

Since the reboiler is assumed to have appreciable holdup, the heat and material balance equations can be written

$$L_3 = B + V_4 \quad (29)$$

$$x_{B,i} = \int_0^t \left(\frac{dx_{B,i}}{dt} \right) dt + x_{B,i}(0) \quad (i = 1, N) \quad (30)$$

$$\frac{dx_{B,i}}{dt} = \frac{1}{\delta B} [L_3 x_{3,i} - (B x_{B,i} - V_4 y_{4,i})] \quad (i = 1, N) \quad (31)$$

$$Q_R + L_3 h_3 = B h_B + V_4 H_4 \quad (32)$$

Due to the assumption that the reboiler is an equilibrium stage, the compositions of the bottoms product and that of the vapor leaving the reboiler are related according to the equation

$$y_{4,i} = K_{4,i} x_{B,i} \quad (i = 1, N) \quad (33)$$

The enthalpies of the vapor and liquid streams are assumed to be ideal; i.e., there is no enthalpy of mixing. Thus, the liquid enthalpies are calculated according to the equation

$$h_j = \sum_{i=1}^N x_{j,i} h_i(T_j) \quad (j = 1, 2, 3, 4) \quad (34)$$

where the pure component liquid enthalpies are calculated

$$h_i(T_j) = a_i + b_i T_j + c_i T_j^2 + d_i T_j^3 \quad (i = 1, N) \quad (35)$$

The vapor enthalpies are calculated according to the equation

$$H_j = \sum_{i=1}^N y_{j,i} H_i(T_j) \quad (j = 2, 3, 4) \quad (36)$$

where the pure component vapor enthalpies are calculated with

$$H_i(T_j) = A_i + B_i T_j + C_i T_j^2 + D_i T_j^3 \quad (i = 1, N) \quad (37)$$

The vapor liquid equilibrium coefficients are assumed to be functions of temperature only. Thus, the coefficients can be expressed

$$K_{j,i} = \alpha_i + \beta_i T_j + \gamma_i T_j^2 + \delta_i T_j^3 \quad (i = 1, N) \quad (38)$$

Stream compositions are assumed to be normalized so that the sums of the mole fractions are equal to unity for each stream. With the exception of the feed stream, all liquid streams are assumed to be at their bubble points. Thus, for the liquid stream,

$$\sum_{i=1}^N K_{j,i} x_{j,i} = 1 \quad (j = 2, 3, 4) \quad (39)$$

Likewise, all vapor streams are assumed to be at their dew points so that

$$\sum_{i=1}^N \frac{y_{j,i}}{K_{j,i}} = 1 \quad (j = 2, 3, 4) \quad (40)$$

Illustrative Example. To illustrate the behavior of the feed forward model, an example problem has been solved for a stripping column. The column is assumed to be subjected to a step change in feed composition. In this example the variable that is to be controlled is the composition of the lightest component in the bottoms product. The example is analogous to an operation where an absorbed gas is being stripped from an absorber oil and the amount of gas in the lean oil is to be controlled. In the example the system being considered is a ternary one. The initial operating conditions and the properties of the components are given in Table VI.

The first step in the calculational procedure is to determine the values of the lumped parameter J_1 from the initial operating conditions.

TABLE VI

OPERATING CONDITIONS FOR FEED FORWARD EXAMPLE

System: Benzene-Toluene-Xylene

Stream Flow Rates

<u>Stream</u>	<u>Flow Rate, moles/hr.</u>
Feed	0.0462
Distillate Product	0.0138
Bottoms Product	0.0324
Feed Section Liquid	0.0639
Stripping Section Liquid	0.0625
Stripping Section Vapor	0.0316
Reboiler Vapor	0.0301

Stream Compositions

<u>Component</u>	<u>Feed</u>	<u>Distillate</u>	<u>Bottoms</u>	<u>Feed Liquid</u>	<u>Stripping Liquid</u>	<u>Stripping Vapor</u>	<u>Reboiler Vapor</u>
Benzene	.427	.813	.262	.555	.401	.855	.550
Toluene	.281	.136	.342	.233	.321	.120	.299
Xylene	.292	.051	.396	.212	.278	.025	.151

Second Feed Composition

Benzene	.368
Toluene	.241
Xylene	.391

Once the lumped parameters have been determined, the feed composition is changed to its new value in a step manner. The column is then restored to heat balance. This operation is permissible because the column is always in heat balance.

New values for $x_{3,i}$ and $y_{3,i}$ ($i = 1, N$) at a differential time increment away from time zero are calculated. These calculations are made with equations (25) and (26) according to the equations

$$x_{3,i} \Big|_{t+\Delta t} = \exp \left[\ln x_{3,i} \Big|_t + \frac{\Delta t}{x_{3,i}} \left(\frac{dx_{3,i}}{dt} \right) \Big|_t \right] \quad (41)$$

$$y_{3,i} \Big|_{t+\Delta t} = \exp \left[\ln y_{3,i} \Big|_t + \frac{\Delta t}{y_{3,i}} \left(\frac{dy_{3,i}}{dt} \right) \Big|_t \right] \quad (42)$$

Equations (41) and (42) are discussed in Appendix E.

The new values of $x_{3,i}$ and $y_{3,i}$ are then used to restore the column to heat balance. The vapor boilup rate is changed so as to bring the bottoms composition back to the desired value. Finally, using the new vapor boilup rate, the column is again restored to heat balance. This procedure of calculating values of $x_{3,i}$ and $y_{3,i}$, heat balancing, adjusting the vapor rate, and heat balancing again is repeated until the column reaches steady-state. The distillate rates that are required to maintain the desired bottoms product composition during the transient period are shown in Figure 19.

The above method of determining the feed forward control of a distillation column is termed the direct calculation method. This method is significantly different from the method that would be used if the dynamic model were based on the plate concept. If a plate model was used, a set of n differential equations would have to be solved for each plate. All of the equations for all of the plates would have to be

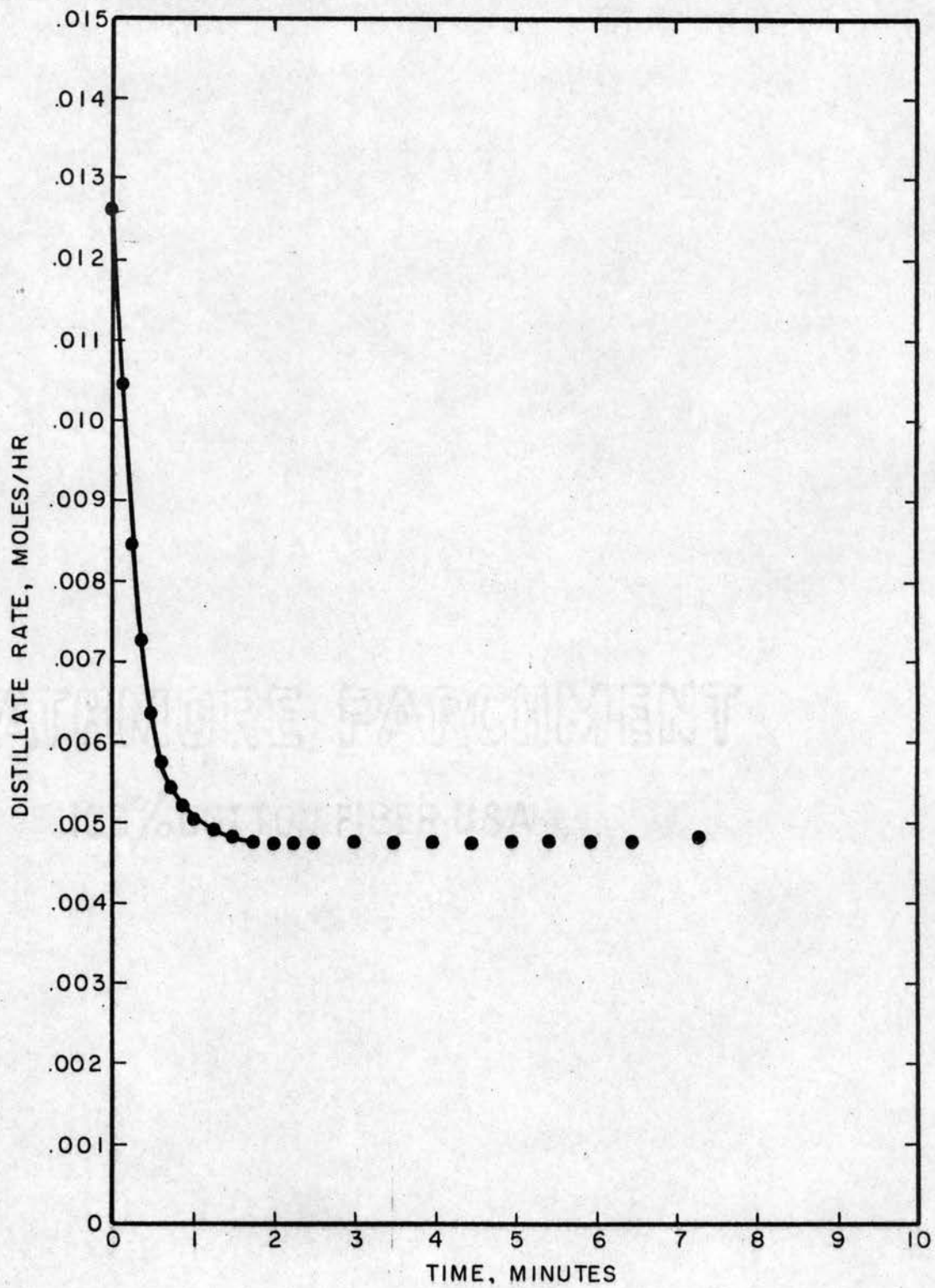


Figure 19. Distillate Rate Response for Feed Forward Control Model

solved simultaneously. These simultaneous solutions would probably be accomplished with a matrix inversion method.

The direct calculation method would not be applicable to solution with a plate method because of the large number of plates. Even a matrix solution for a plate model is so time consuming that a complete solution has never been obtained for a feed forward control system using the plate method. Luyben and Gerster (20) proposed a model using the plate model, but the solution they obtained for a feed forward control scheme was based on an external material balance scheme. Shinskey (34) and Luyben (19) have both presented recent articles on the feed forward control of distillation columns. These articles were based on external material balance control.

In the external material balance control scheme, the feed forward procedure is based on the steady-state behavior of the column rather than on column dynamics. When an upset is detected a new steady-state solution is obtained. This new steady-state solution is one which will give the desired product quality. The operating variables are then changed to the values determined from the steady-state solution. In this manner the column is restored to the desired operating condition. While this method of feed forward control will return the column to the desired steady-state, there is a period during which the column is not producing the desired product. This period is unavoidable with a feed forward control system using the over-all column control scheme. The period of off-quality operation does not exist when a model based on column dynamics is used.

There are no experimental data with which to test the proposed feed forward control model. Without these data no absolute evaluation is

possible, but existing theoretical and experimental work concerning column dynamics can be used to obtain some preliminary conclusions about the model. Since the lumped parameter model reproduces the transient performance of the distillation column when the column is subjected to a step change in feed composition and when the column is subjected to a change in the feed rate, the model should be valid when the flow rates and the feed composition are changed simultaneously. Considering this applicability of the transient model along with the ability of the feed forward model to maintain a constant bottoms product composition, the apparent conclusion is that the proposed feed forward model will be useful in the control of distillation columns. The extent of this utility will have to be proven by application of the model, but on the basis of the preliminary work the model appears to describe the measures necessary to control a distillation column with a feed forward control system.

CHAPTER VIII

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

The objectives of this research project were to obtain experimental data on the dynamic behavior of a distillation column and to determine if that behavior could be predicted with a first order lumped parameter model with dead time. These objectives have been accomplished by performing a series of twelve experimental runs on an Oldershaw distillation column that was operated as a stripping column. The principal sources of error in the experimental data were found to be the integrator on the gas chromatograph and methods of measuring the product flow rates. Within the limits of the accuracy of the analytical equipment, the experimental results were shown to be reproducible in separate runs.

Experimental data have been used to show that the dynamic behavior of a distillation column can be accurately predicted with a first order lumped parameter model with dead time. The model has been shown to accurately predict the final steady-state compositions of the product streams if the stream flow rates are known accurately, and to accurately predict the transient response curve for the column.

On the basis of the ability of the lumped parameter model to predict the dynamic behavior of the distillation column, a feed forward model has been developed for predictive control of a column. The feed forward model is completely theoretical in that no experimental data have been

presented to show its accuracy. However, since the lumped parameter model does accurately describe the dynamic behavior of a distillation column, and since the feed forward model does, on the basis of a calculated example, control the column products to the desired specifications, the feed forward model has been deemed worthy of further investigation. This further investigation should consist of using the control model in a control scheme for an operating column. With the data obtained, the utility of the feed forward model can be accurately determined.

Recommendations

For future studies concerning the dynamic behavior of distillation columns, several changes are recommended for the experimental apparatus. The reboiler and the condenser should be equipped with sight gages so that the liquid holdup in each can be measured during experimental runs. Feed and product streams should be monitored and controlled so that the flow rates can be accurately known and easily controlled to the desired rates. If a gas chromatograph is used for an analytical instrument, it should be equipped with a digital integrator or some other highly reproducible type of integrator. The mole fractions obtained with the gas chromatograph should be reproducible to an accuracy better than one-tenth of one mole per cent. Finally, future experimental work should be concerned with multicomponent systems; i.e., systems of three or more components. Work in multicomponent systems is recommended because there are no published experimental data for systems having more than two components.

NOMENCLATURE

Major Symbols

English Letters

- B = bottoms product flow rate, moles/hour.
- D = distillate flow rate, moles/hour.
- F = feed flow rate, moles/hour.
- f_n^L = fraction of the section that is filled with liquid.
- f_n^V = $(1 - f_n^L)$ = fraction of the section that is filled with vapor.
- h = specific enthalpy of liquid, BTU/lb.-mole.
- H = specific enthalpy of vapor, BTU/lb.-mole.
- J = parameter which describes the degree of separation occurring in the section, moles/hour-mole fraction.
- K = vapor liquid equilibrium coefficient.
- L = liquid flow rate, moles/hour.
- N = net rate of mass transfer between phases, moles/hour.
- Q_r = reboiler heat duty, BTU/hour.
- S = cross-sectional area of the column, sq. ft.
- V = vapor flow rate, moles/hour.
- w = reduced composition.
- x = liquid composition, mole fraction.
- y = vapor composition, mole fraction.
- z = height of the section, ft.

Greek Letters

- δ = holdup in a section or on a tray, moles.
 ρ = molar density, moles/cu. ft.
 ω = acentric factor

Subscripts

- b = bottoms.
d = distillate.
f = feed section.
i = component number.
m = subsection number.
n = section number.

Superscripts

- * = equilibrium value.
L = liquid phase.
o = initial condition.
V = vapor phase.

Groups

- $\frac{d}{dt}$ = total derivative with respect to time.
 $\frac{\partial}{\partial t}$ = partial derivative with respect to time.
 $\frac{\partial}{\partial z}$ = partial derivative with respect to distance.

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APPENDIX A
METHODS OF CALCULATION

METHODS OF CALCULATION

Equilibrium Values

Equilibrium values for benzene, toluene, and p-xylene were calculated from a method presented by Edmister (11).

$$K_i = \frac{1}{\theta_i} \frac{P_i^o}{P} \frac{\gamma_i^L}{\gamma_i^V} \quad (43)$$

γ_i^V , the vapor activity coefficient, was assumed to be unity because of the low system pressure.

The liquid activity coefficient, γ_i^L , was calculated from the Scatchard-Hildebrand equation,

$$\ln \gamma_i^L = \frac{V_i^L}{RT} (\delta_m - \delta_i)^2 \quad (44)$$

V_i^L in equation (44) is the molar liquid volume of component i. The molar liquid volume was determined from the equation

$$V_i^L = \frac{RT_{C_i} V_{R_i}}{P_{C_i}} \quad (45)$$

V_{R_i} , the reduced volume for component i, was found from the following relation,

$$V_{R_i} = V_i^* (5.7 + 3.0 T_p) \quad (46)$$

V_i^* is the reduced expansion factor, and is related to the acentric factor, ω , by the following equation,

$$V_i^* = 0.01361 - 0.00328\omega - 0.0244\omega^2 + 0.0599\omega^3 - 0.0308\omega^4 \quad (47)$$

δ_m is the liquid volume average solubility parameter for the entire mixture,

$$\delta_m = \frac{\sum x_i V_i^L \delta_i}{\sum x_i V_i^L} \quad (48)$$

The imperfection pressure correction, θ , is designed to permit evaluation of correction factors to be applied to the ideal K-value. The imperfection pressure correction in generalized form is

$$\ln \theta_1 = \frac{1}{T_R} \left[\frac{BP_C}{RT_C} - \frac{V^L P_C}{RT_C} \right] (P_r - P_r^O) \quad (49)$$

P_r^O is the reduced vapor pressure. The term $\frac{BP_C}{RT_C}$ is a reduced second virial coefficient and is a function of the reduced temperature and acentric factor,

$$\begin{aligned} \frac{BP_C}{RT_C} = & (0.1445 + 0.073\omega) - (0.330 - 0.46\omega)/T_r - (0.1385 + \\ & 0.50\omega)/T_r^2 - (0.0121 + 0.097\omega)/T_r^3 - (0.0073\omega)/T_r^8 \end{aligned} \quad (50)$$

Vapor pressures were calculated from the Antoine equation

$$\log P^O = A - B/(C + T) \quad (51)$$

A, B, and C are experimentally determined coefficients.

Ideal K-values were used to check the K-values that were calculated by the above method. Ideal K-values were calculated from the vapor pressure and the system pressure,

$$K = \frac{P^O}{P} \quad (52)$$

The K-values from the two methods agreed within one per cent over the entire composition range.

Heats of Vaporization

The Clausius-Clapeyron equation was used to calculate heats of vaporization,

$$\frac{dP}{dT} = \frac{H_{\text{vap}}}{T(V^G - V^L)} \quad (53)$$

The vapor pressure data needed for equation (53) were obtained via the Antoine equation, equation (51). Differentiation of equation (51) gives

$$\frac{dP}{dT} = \frac{2.303BP}{(C + T)^2} \quad (54)$$

Equation (53) and equation (54) were combined to obtain an expression for the heat of vaporization

$$H_{\text{vap}} = PT \left[\frac{2.303B}{(C + T)^2} \right] (V^G - V^L) \quad (55)$$

The liquid molar volume is much smaller than the gas molar volume and can therefore be neglected. The molar gas volume was calculated from

$$V^G = \frac{RT}{P} + B^0 \quad (56)$$

B^0 is the second virial coefficient.

The resulting equation used for the calculation of the heat of vaporization was

$$H_{\text{vap}} = PT \left[\frac{2.303B}{(C + T)^2} \right] \left[\frac{RT}{P} - B^0 \right] \quad (57)$$

The Kistiakowsky equation was used to check the values calculated from the above method. The Kistiakowsky equation evaluated the heat of vaporization at the normal boiling point,

$$H_{\text{vap}} = (7.58 + 4.571 \log T_B) T_B \quad (58)$$

The heats of vaporization at temperatures other than the normal boiling point were found using the Watson equation,

$$\frac{H_{\text{vap}}}{(H_{\text{vap}})_{T_L}} = \left[\frac{T_C - T}{T_C - T_B} \right]^{0.38} \quad (59)$$

The results obtained from the Kistiakowsky-Watson method agreed with the results calculated via the Clapeyron equation within less than one per cent.

APPENDIX B

CHROMATOGRAPH CALIBRATION

CHROMATOGRAPH CALIBRATION

Liquid sample compositions were determined by the use of an F & M Model 500 Programmed High-Temperature Gas Chromatograph. The output from the chromatograph consisted of curves traced from the thermal-conductivity detector response. The areas under the peaks of these curves are proportional to the amount of each component in the sample.

The area fraction for each component can easily be calculated. However, the area fraction by itself is not a common indication of composition. Compositions are generally reported on the basis of mole fraction or weight fraction. The purpose of the calibration was to convert the area fractions obtained from the chromatograph analyses to a more meaningful weight-fraction basis.

The procedure used in calibrating the gas chromatograph consisted first in the preparation of standard samples. The amount of each component in the sample was carefully weighed using an analytical balance. The weight of each component was determined to the nearest one-tenth of a milligram. Nine samples were prepared to cover the full range of weight fractions for each component. The compositions of the standard sample are presented in Table VII.

The samples were refrigerated before analysis to prevent loss by evaporation. Multiple analyses were made for each sample to make the results as accurate as possible. The areas under the peaks of the response curves were measured, and the area fractions for the components were calculated. The chromatograph results are shown in Table VIII.

TABLE VII
COMPOSITION OF STANDARD SAMPLES

Sample	Benzene	Weight Fraction	
		Toluene	p-Xylene
A	0.10391	0.87180	0.02428
B	0.62039	0.07129	0.30832
C	0.64855	0.16785	0.18359
D	0.12000	0.84568	0.03437
E	0.49378	0.35773	0.14847
F	0.22236	0.23943	0.44789
G	0.14428	0.24125	0.61446
H	0.11820	0.12071	0.76109
I	0.31268	0.12754	0.65010

TABLE VIII
ANALYSIS OF CHROMATOGRAPH RESULTS

Sample	<u>Benzene</u>		<u>Toluene</u>		<u>p-Xylene</u>	
	Mean Area Fraction	Standard Deviation	Mean Area Fraction	Standard Deviation	Mean Area Fraction	Standard Deviation
A	0.09330	0.004848	0.89488	0.007350	0.01323	0.004322
B	0.68731	0.013999	0.04309	0.006697	0.26958	0.008952
C	0.69230	0.009316	0.16786	0.002411	0.13984	0.008041
D	0.11404	0.003269	0.86281	0.007293	0.02316	0.004439
E	0.49385	0.013862	0.35755	0.019822	0.14860	0.008637
F	0.22149	0.011027	0.23776	0.006067	0.43232	0.005950
G	0.14668	0.003678	0.23753	0.002361	0.61579	0.003182
H	0.11808	0.005021	0.10996	0.010055	0.77196	0.012386
I	0.32992	0.006312	0.11758	0.005335	0.66093	0.014166

The next step in the calibration procedure involved the correlation of area fractions with weight fractions. The first method attempted was internal normalization. By this method the weight fraction is expressed as

$$w_{f_i} = \frac{f_i A_i}{\sum_{j=1}^N f_i A_j} \quad (60)$$

w_{f_i} and A_{f_i} are the weight and area fractions respectively, and f_i represents the standardization coefficient for component "i." The results predicted from this type of correlation did not agree with the standard sample analyses.

The next attempt to arrive at a correlation involved the use of a linear model

$$w_{f_i} = a_i + b_i A_i \quad (61)$$

where a_i and b_i are regression coefficients. The results obtained with this method were much better than those of the first method. Most of the error associated with the prediction of weight fractions from area fractions were due to the error in the determination of the data points. The results are presented in Table IX.

The other method of correlation that was tried was a quadratic model,

$$w_{f_i} = a_i + b_i A_{f_i} + c_i A_{f_i}^2 \quad (62)$$

The results, shown in Table IX, obtained from this method were not significantly better than those obtained from the linear model.

A comparison of the correlations indicated that the linear model, equation (61), best represented the data. The linear model was used to convert area fractions determined via chromatograph analyses to weight

TABLE IX
REGRESSION COEFFICIENTS AND STANDARD ERROR

Linear Model

Component	Regression Coefficients		Standard Error of Estimate
	a	b	
Benzene	0.01671	0.90922	0.01500
Toluene	0.01622	0.95599	0.00951
p-Xylene	0.02463	0.96051	0.01717

Quadratic Model

Component	Regression Coefficients			Standard Error of Estimate
	a	b	c	
Benzene	0.00195	0.01032	-0.15692	0.01419
Toluene	0.02552	0.88481	0.07053	0.00882
p-Xylene	0.01292	1.09518	-0.17977	0.01437

fractions. Then the mole fractions were calculated from the weight fractions.

The evaluation of the error in the predicted weight fractions provides an estimate of the accuracy of the reported compositions. The difference, or error, between the true weight fraction and the predicted weight fraction can be considered as a sum of several errors. These errors are

1. Error in weighing the standard samples.
2. Experimental, or random, error.
3. Error in measurement of areas from the curves on the chromatogram.
4. Error associated with lack of fit of the linear model.

The first error can be neglected, since the accuracy of the weighing measurements was carried out to the nearest tenth of a milligram. The total error is then given by the standard error of the estimate, which is presented in Table IX. The standard error of the estimate was calculated as follows. The measured values of the weight fractions, that is, the true weight fractions, were regressed as a function of the calculated area fraction, according to the linear model. Then the linear model was used to calculate weight fractions, which were then compared with the true weight fractions. The differences were treated in the same manner as the conventional statistical method used to determine the standard deviation. The second and third types of errors were estimated by the standard deviation given in Table VIII. A comparison of Tables VIII and IX illustrates that the standard error of the estimate, representing the total error, is not significantly greater than the standard deviation, representing the error in the determination of the data points. Thus, the conclusion was that most of the error associated with the prediction

of weight fractions from area-fraction data was due to the error in the determination of the area fraction. The error appears to be within two area per cent over the entire composition range.

APPENDIX C

DEVELOPMENT OF THE LUMPED PARAMETER MODEL

DEVELOPMENT OF THE LUMPED PARAMETER MODEL

Assuming that the basic idea of the model developed by Reynolds (28) is valid; i.e., that the net rate at which mass is transferred from the vapor phase to the liquid phase in a section can be represented by equation (2), a material balance can be made on the vapor stream entering and leaving a section. Since the general law of conservation of matter applies; i.e., that the difference between the input and the output is the accumulation, the following equations expressing the input, the output, and the accumulation are valid:

$$\text{input} = V_{n+1} y_{n+1,i} \quad (63)$$

$$\text{output} = V_{n+1} y_{n+1,i} + \frac{\partial (V_n y_{n,i})}{\partial z} dz + N_{n,i}^V \quad (64)$$

$$\text{accumulation} = \frac{\partial (\delta_n^V y_{n,i})}{\partial t} \quad (65)$$

$$\delta_n^V = f_n^V e_n^V S z \quad (66)$$

The symbols appearing, in all equations are defined in the Nomenclature. The terms in equation (64) may need further explanation than is supplied by the equation itself. Thus, in order to further the meaning of the equation, the explanation that follows is presented. The output that is calculated in equation (64) is the sum of all of the material that leaves the vapor phase, either by flow or by mass transfer. The rate at which a component flows from the section is equal to the flow rate in $(V_{n+1} y_{n+1,i})$ plus the increase in the stream flow rate that occurs in

the section $\frac{\partial (V_n^y y_{n,i})}{\partial z} dz$. The material transferred from the vapor phase is represented by the term $N_{n,i}^V$. If equations (63), (64), and (65) are substituted into a material balance, the resulting equation is

$$\frac{\partial (\delta_n^V y_{n,i})}{\partial t} = - \frac{\partial (V_n^y y_{n,i})}{\partial z} dz - N_{n,i}^V \quad (67)$$

A partial differential equation that represents the behavior of the composition of the vapor stream passing through the column can now be obtained by substituting equation (2) into equation (67). The partial differential equation that results from this substitution is

$$\frac{\partial (\delta_n^V y_{n,i})}{\partial t} = - \frac{\partial (V_n^y y_{n,i})}{\partial z} dz + J_{n,i} (y^* - y)_{n,i} \quad (68)$$

A partial differential equation for the liquid stream that is analogous to equation (68) for the vapor stream can be obtained by making a material balance on the liquid stream passing through the section. In this material balance the input, output, and accumulation are represented by the equations

$$\text{input} = L_{n-1} x_{n-1,i} \quad (69)$$

$$\text{output} = L_{n-1} x_{n-1,i} + \frac{\partial L_{n-1} x_{n-1,i}}{\partial z} dz + N_i^L \quad (70)$$

$$\text{accumulation} = \frac{\partial (\delta_n^L x_{n,i})}{\partial t} \quad (71)$$

$$\delta_n^L = f_n^L e_n^L S z \quad (72)$$

The terms that appear in equation (70) are like those that appeared in equation (64) except they apply to the liquid phase rather than the vapor phase. The equation that results from the substitution of equations (69), (70), and (71) into a material balance equation is

$$\frac{\partial \delta_n^L x_{n,i}}{\partial t} = - \frac{\partial L_n^L x_{n,i}}{\partial z} dz - N_i^L \quad (73)$$

Equation (73), which is similar to equation (66), is a partial differential equation that represents the behavior of the composition of the liquid stream passing through a section of a distillation column.

Since the partial differential equations obtained for the liquid and vapor streams leaving a section of the column cannot, under normal circumstances, be integrated exactly some simplifications of the equations are necessary. First, the partial derivatives $\frac{\partial L_n^L x_{n,i}}{\partial z}$ and $\frac{\partial V_n^V y_{n,i}}{\partial z}$ can be replaced by the approximations

$$\frac{\partial (V_n^V y_{n,i})}{\partial z} = \frac{(V_n^V y_{n,i} - V_{n+1}^V y_{n+1,i})}{\Delta z} \quad (74)$$

$$\frac{\partial (L_n^L x_{n,i})}{\partial z} = \frac{(L_n^L x_{n,i} - L_{n-1}^L x_{n-1,i})}{\Delta z} \quad (75)$$

In order for the approximations represented by equations (74) and (75) to be valid, the change in height Δz must be small. Since the groups $\delta_n^V y_{n,i}$ and $\delta_n^L x_{n,i}$ are now functions of time only, the partial derivatives with respect to time can be replaced with total derivatives. Using this change from partial to total derivatives, the approximations of equations (74) and (75), the assumption of constant molal holdup in a section, and the assumption of constant rate of interphase mass transfer in a section, equations (68) and (73) can be rewritten in the forms

$$\delta_n^V \frac{dy_{n,i}}{dt} = - (V_n^V y_{n,i} - V_{n+1}^V y_{n+1,i}) + J_{n,i} (y^* - y)_{n,i} \quad (76)$$

$$\delta_n^L \frac{dx_{n,i}}{dt} = - (L_n^L x_{n,i} - L_{n-1}^L x_{n-1,i}) - N_{n,i}^L \quad (77)$$

Up to this point no attempt has been made to relate the net rate at which mass is transferred from the liquid phase with the net rate at

which mass is transferred from the vapor phase; this relationship will now be shown. For this purpose, consider a section of a column that is small enough that the molal holdups of the respective phases is negligible in comparison to the flow rates of the liquid and vapor streams. For such a section, equations (76) and (77) can be written

$$-(V_n y_{n,i} - V_{n+1} y_{n+1,i}) + J_{n,i} (y^* - y)_{n,i} = 0 \quad (78)$$

$$-(L_n x_{n,i} - L_{n-1} x_{n-1,i}) - N_{n,i}^L = 0 \quad (79)$$

Also for such a section, even under transient conditions, the over-all material balance can be written

$$L_n x_{n,i} + V_n y_{n,i} - (L_{n-1} x_{n-1,i} + V_{n+1} y_{n+1,i}) = 0 \quad (80)$$

Summing equations (78) and (79) and the subsequent use of equation (80) yields the equation

$$N_{n,i}^L = J_{n,i} (y^* - y)_{n,i} \quad (81)$$

Now by using equation (81), equation (77) can be rewritten

$$\delta_n^L \frac{dx_{n,i}}{t} = -(L_n x_{n,i} - L_{n-1} x_{n-1,i}) - J_{n,i} (y^* - y)_{n,i} \quad (82)$$

Equation (76) and (82) are valid for any system which meets the assumptions that were made in deriving them; however, they cannot be used in this present form because no method for evaluating $J_{n,i}$, the parameter that describes the degree of separation which occurs in the column, has been developed. In order to evaluate $J_{n,i}$ the assumption that it remains constant for small changes in column conditions must be made. In addition since the function $J_{n,i} (y^* - y)_{n,i}$, which represents the net rate of mass transfer between phases, is based on passing streams which cannot be measured, some method of approximating the driving force for mass transfer must be developed. Reynolds attempted to use both the

driving force at the top of the section and the average driving force. He defined the average driving force as the arithmetic average of the driving forces at the top and bottom of the column. Reynolds did not get good results with either the driving force at the top of the column or the average driving force. In order to obtain a new method for representing the driving force for mass transfer, the reasoning which is described below was used.

If a section of the column is considered to be subdivided into an infinite number of subsections (as shown in Figure 20), the driving force for one of these infinitesimal subsections can be represented by the equation

$$(y^* - y)_{m,i} = (Kx)_{m-1,i} - y_{m+1,i} \quad (83)$$

If the number of subsections for which the driving force applies is increased to two, then the driving force for the two subsections m and $m-1$ can be approximated by the equation

$$(y^* - y)_{\left. \begin{matrix} m \\ m-1 \end{matrix} \right\}, i} = (Kx)_{m-2,i} - y_{m+1,i} \quad (84)$$

If a similar line of reasoning is used to extend the interval for which the driving force applies to the entire section, the driving force for the section can be approximated by the equation

$$(y^* - y)_{n,i} = (Kx)_{n-1,i} - y_{n+1,i} \quad (85)$$

Thus, equation (2) can be rewritten in the form

$$N_{n,i}^V = -J_{n,i} [(Kx)_{n-1,i} - y_{n+1,i}] \quad (86)$$

Using equation (86), equations (76) and (82) can be rewritten in the form

$$S_n^V \frac{dy_{n,i}}{t} = -[V_n y_{n,i} - V_{n+1} y_{n+1,i}] + J_{n,i} [(Kx)_{n-1,i} - y_{n+1,i}] \quad (87)$$

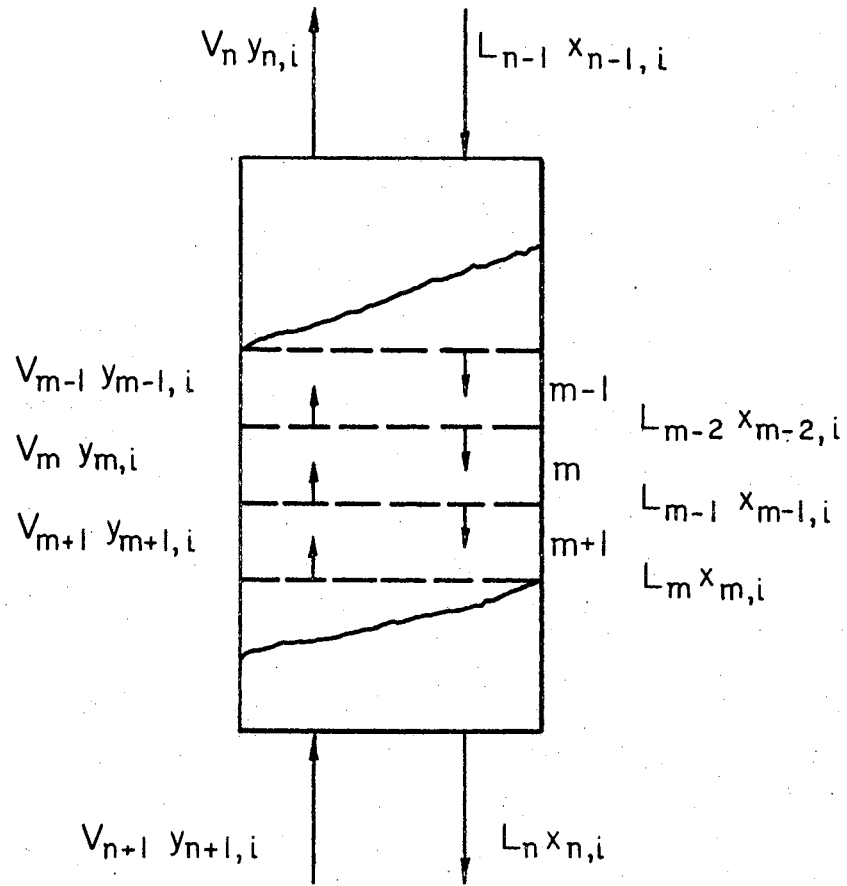


Figure 20. A Section of a Distillation Column Divided into Subsections

$$\delta_n^L \frac{dx_{n,i}}{t} = -[L_n x_{n,i} - L_{n-1} x_{n-1,i}] - J_{n,i} [(Kx)_{n-1,i} - y_{n+1,i}] \quad (88)$$

Now by assuming that $J_{n,i}$ is constant for small changes in column conditions, a solution to the transient behavior of the compositions of the vapor and liquid streams leaving the column can be obtained by integrating equations (87) and (88). In most cases the integration cannot be performed analytically and either numerical or graphical techniques must be used. Regardless of which technique is used, the value of $J_{n,i}$ must be obtained. Equation (87) must be equal to zero at steady-state since by definition $\frac{dy_{n,i}}{dt} = 0$ at steady-state. Thus, by using the initial conditions in the column, a value for $J_{n,i}$ can be calculated using the equation

$$J_{n,i} = \frac{(V_n y_{n,i} - V_{n+1} y_{n+1,i})}{(Kx)_{n-1,i} - y_{n+1,i}} \quad (89)$$

Likewise, since the time derivative for the liquid phase must be zero at steady-state, equation (88) can be rearranged so that the initial liquid flow rates can be used to obtain $J_{n,i}$. Thus, a value for $J_{n,i}$ can also be obtained using the initial conditions and the equation

$$J_{n,i} = \frac{-(L_n x_{n,i} - L_{n-1} x_{n-1,i})}{(Kx)_{n-1,i} - y_{n+1,i}} \quad (90)$$

Equations (89) and (90) reveal that the only information that is required to obtain a value for $J_{n,i}$ is that information which is normally obtained from a computer solution. Likewise, equations (87) and (88) reveal that the only information, other than that which can be obtained from a computer solution, that is required to use the model, is the liquid holdup. In addition, the data required to use the model are normally obtained in the design of a distillation column. Since the

vapor and liquid holdup terms in equations (87) and (88) do not appear in equations (89) and (90), a logical conclusion would be that they do not have any effect on the final steady-state values, but only serve as time constants. A numerical integration of equations (87) and (88) using several different holdups has shown this conclusion to be valid. A solution to the same problem was also obtained with a plate-to-plate model (3). The results of both of these solutions, along with a plot of experimental data (3), are shown in Figure 21. The experiments from which the data were obtained are discussed elsewhere (3, 4, 26). As can be seen in Figure 21, the transient solution obtained with the present model is one which has a first order time constant. Also, the solution can be made to coincide with the experimental data by merely changing the holdup, while the plate-to-plate model does not follow the experimental data at all. The purpose of this comparison has not been to point out the fact that the plate-to-plate model does not approximate the curves, because it does in many cases, but to point out that the present model can be made to follow the experimental data by merely changing the holdup term.

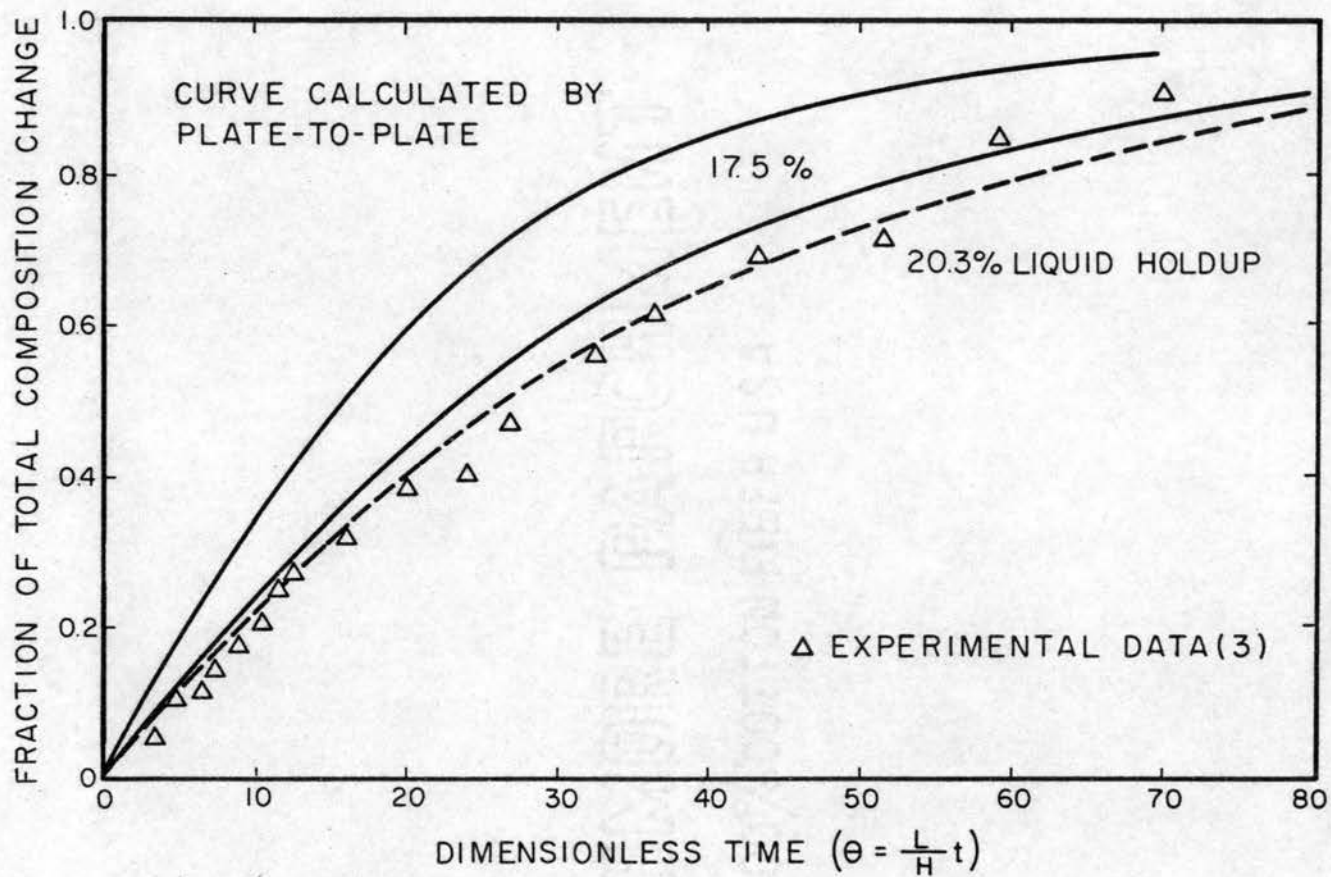


Figure 21. Effect of Holdup on the Transient Behavior of a Distillation Column

APPENDIX D

EXPERIMENTAL DATA

EXPERIMENTAL DATA FOR RUN 105

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.058092	0.058092
Distillate Rate, moles/hr.	0.021983	0.021983
Bottoms Rate, moles/hr.	0.036109	0.036109
Feed Composition, m. f.*	0.350	0.431
Column Holdup, ml. liquid	-----	237.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
1.00	0.706	0.144
4.00	0.702	0.158
5.00	0.732	0.153
6.00	0.734	0.165
7.00	0.741	0.158
8.00	0.746	0.170
9.00	0.763	0.183
10.00	0.756	0.195
11.00	0.768	0.198
12.00	0.769	0.208
13.00	0.775	0.211
14.00	0.761	0.222
15.00	0.767	0.214
17.00	0.763	0.232
19.00	0.774	0.234
21.00	0.773	0.245
25.00	0.763	0.251
30.00	0.774	0.246
35.00	0.778	0.246

EXPERIMENTAL DATA FOR RUN 106

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.048889	0.047863
Distillate Rate, moles/hr.	0.021386	0.022320
Bottoms Rate, moles/hr.	0.027053	0.025543
Feed Composition, m. F.*	0.353	0.395
Column Holdup, ml. liquid	-----	60.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.695	0.078
1.00	0.685	0.084
2.00	0.698	0.083
3.00	0.687	0.069
4.00	0.682	0.080
5.00	0.697	0.067
6.00	0.713	0.066
7.00	0.704	0.076
8.00	0.718	0.068
9.00	0.726	0.073
10.00	0.721	0.072
12.00	0.734	0.088
14.00	0.720	0.091
16.00	0.723	0.095
18.00	0.728	0.106
20.00	0.698	0.122
22.00	0.719	0.081
25.00	0.733	0.128
30.00	0.723	0.124
35.00	0.719	0.126

EXPERIMENTAL DATA FOR RUN 107

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.043578	0.047323
Distillate Rate, moles/hr.	0.018127	0.019872
Bottoms Rate, moles/hr.	0.025451	0.027451
Feed Composition, m. f.*	0.337	0.301
Column Holdup, ml. liquid	-----	40.0

*Compositions are given in mole fraction hexane.

Product Stream Compositions.

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.708	0.126
1.00	0.706	0.127
2.00	0.701	0.126
3.00	0.704	0.134
4.00	0.709	0.139
5.00	0.710	0.136
6.00	0.696	0.132
7.00	0.674	0.133
8.00	0.647	0.118
9.00	0.613	0.114
10.00	0.558	0.107
11.00	0.539	0.091
12.00	0.526	0.097
14.00	0.521	0.082
16.00	0.524	0.052
18.00	0.520	0.052
20.00	0.525	0.032
25.00	0.530	0.033
30.00	0.594	0.014
35.00	0.533	0.008

EXPERIMENTAL DATA FOR RUN 109

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.038794	0.039894
Distillate Rate, moles/hr.	0.020482	0.021247
Bottoms Rate, moles/hr.	0.018312	0.018647
Feed Composition, m. f.*	0.275	0.404
Column Holdup, ml. liquid	-----	40.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.550	0.003
1.00	0.551	0.007
2.00	0.535	
3.00	0.565	0.017
4.00	0.562	0.009
5.00	0.459	0.039
6.00	0.575	0.003
7.00	0.568	0.002
8.00	0.570	0.045
9.00	0.615	0.012
10.00	0.615	0.008
11.00	0.625	0.014
12.00	0.648	0.003
14.00	0.645	0.014
16.00	0.652	0.005
18.00	0.663	0.004
20.00	0.678	0.003
25.00	0.664	0.005
30.00	0.680	0.006
35.00	0.688	0.009

EXPERIMENTAL DATA FOR RUN 110

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.040774	0.039276
Distillate Rate, moles/hr.	0.016710	0.013400
Bottoms Rate, moles/hr.	0.024064	0.025876
Feed Composition, m. f.*	0.731	0.464
Column Holdup, ml. liquid	-----	83.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.925	0.559
1.00	0.896	0.573
2.00	0.917	0.583
3.00	0.918	0.571
4.00	0.917	0.575
5.00	0.918	0.574
6.00	0.913	0.559
7.00	0.908	0.576
8.00	0.907	0.575
9.00	0.898	0.570
10.00	0.890	0.551
11.00	0.861	0.547
12.00	0.859	0.529
14.00	0.830	0.512
16.00	0.805	0.492
18.00	0.801	0.473
20.00	0.794	0.456
25.00	0.769	0.417
30.00	0.778	0.398
35.00	0.783	0.364
45.00	0.779	0.334

EXPERIMENTAL DATA FOR RUN 111

Benzene-Toluene

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.03856	0.03776
Distillate Rate, moles/hr.	0.01965	0.01932
Bottoms Rate, moles/hr.	0.01891	0.01844
Feed Composition, m. f.*	0.675	0.516
Column Holdup, ml. liquid	-----	63.0

*Compositions are given as mole fraction benzene.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.857	0.470
1.00	0.857	0.461
2.00	0.864	0.455
3.00	0.869	0.445
4.00	0.872	0.456
5.00	0.841	0.474
6.00	0.862	0.454
7.00	0.849	0.463
8.00	0.863	0.464
9.00	0.862	0.464
10.00	0.836	0.462
11.00	0.838	0.425
12.00	0.825	0.458
14.00	0.861	0.455
16.00	0.813	0.453
18.00	0.809	0.437
20.00	0.811	0.414
25.00	0.812	0.396
30.00	0.807	0.384
35.00	0.803	0.389

EXPERIMENTAL DATA FOR RUN 115

Benzene-Toluene-Xylene

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.03704	0.04160
Distillate Rate, moles/hr.	0.02071	0.02279
Bottoms Rate, moles/hr.	0.01633	0.01881
Feed Composition, m. f.		
Toluene	0.217	0.296
Xylene	0.329	0.263
Column Holdup, ml. liquid	-----	40.0

Product Stream Compositions

<u>Time, min.</u>	<u>Toluene</u>		<u>Xylene</u>	
	<u>Distillate Composition</u>	<u>Bottoms Composition</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.132	0.379	0.050	0.521
4.00	0.109	0.346	0.053	0.610
6.00	0.137	0.347	0.059	0.607
7.00	0.138	0.343	0.063	0.610
7.50	0.145	0.343	0.050	0.607
8.00	0.146	0.353	0.047	0.605
8.50	0.156	0.356	0.050	0.600
9.00	0.148	0.358	0.040	0.594
9.50	0.151	0.364	0.044	0.601
10.00	0.150	0.312	0.040	0.600
10.50	0.151	---	0.047	---
11.00	0.145	0.347	0.044	0.597
11.50	0.164	0.348	0.048	0.613
12.00	0.166	0.362	0.050	0.595
12.50	0.160	---	0.050	---
13.00	0.158	0.343	0.040	0.603
13.50	0.172	---	0.063	---
14.00	0.165	0.366	0.066	0.597
14.50	0.163	0.358	0.069	0.597
15.00	0.168	0.370	0.070	0.595
15.50	---	0.382	---	0.574
16.00	0.175	0.384	0.041	0.577
17.00	0.175	0.385	0.038	0.584
18.00	---	0.390	---	0.571
20.00	---	0.398	---	0.572
25.00	---	0.403	---	0.558
30.00	0.158	0.380	0.050	0.561
35.00	0.154	0.405	0.060	0.557

EXPERIMENTAL DATA FOR RUN 116

Benzene-Toluene-Xylene

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.03602	0.03746
Distillate Rate, moles/hr.	0.01818	0.01899
Bottoms Rate, moles/hr.	0.01784	0.01847
Feed Composition, m. f.		
Toluene	0.222	0.320
Xylene	0.272	0.201
Column Holdup, ml. liquid	-----	40.0

Product Stream Compositions

<u>Time, min.</u>	<u>Toluene</u>		<u>Xylene</u>	
	<u>Distillate Composition</u>	<u>Bottoms Composition</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.120	0.379	0.060	0.525
2.00	0.123	0.381	0.055	0.519
4.00	0.120	0.371	0.053	0.527
6.00	0.126	0.374	0.055	0.524
7.00	0.131	0.371	0.051	0.532
7.50	0.135	---	0.049	---
8.00	0.143	0.367	0.052	0.535
8.50	0.137	0.374	0.046	0.532
9.00	0.141	0.376	0.049	0.529
9.50	0.140	0.370	0.048	0.539
10.00	0.145	0.370	0.048	0.541
10.50	0.138	0.394	0.052	0.517
11.00	0.147	0.384	0.046	0.524
11.50	0.143	0.386	0.049	0.526
12.00	0.128	0.389	0.048	0.522
12.50	0.148	0.386	0.047	0.523
13.00	0.167	0.389	0.051	0.519
13.50	0.145	0.392	0.045	0.523
14.00	0.152	0.397	0.046	0.520
14.50	0.151	0.410	0.038	0.500
15.00	0.149	0.400	0.044	0.514
16.00	0.159	0.399	0.049	0.519
17.00	0.148	0.400	0.045	0.513
18.00	0.140	0.404	0.048	0.514
20.00	0.153	0.409	0.050	0.510
25.00	0.151	0.425	0.043	0.504
30.00	0.159	0.437	0.042	0.488
36.00	0.153	0.438	0.049	0.481

EXPERIMENTAL DATA FOR RUN 117

Benzene-Toluene-Xylene

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.04616	0.04156
Distillate Rate, moles/hr.	0.01102	0.00752
Bottoms Rate, moles/hr.	0.03514	0.03464
Feed Composition, m. f.		
Toluene	0.281	0.241
Xylene	0.292	0.391
Column Holdup, ml. liquid	---	40.0

Product Stream Compositions

<u>Time, min.</u>	<u>Toluene</u>		<u>Xylene</u>	
	<u>Distillate Composition</u>	<u>Bottoms Composition</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.139	0.343	0.044	0.398
1.00	0.131	0.346	0.058	0.415
2.00	0.137	0.335	0.048	0.402
2.50	0.136	0.345	0.050	0.385
3.00	0.134	0.337	0.058	0.397
3.50	0.136	0.344	0.052	0.379
4.00	0.140	0.341	0.054	0.386
4.50	0.133	0.336	0.051	0.390
5.00	0.143	0.336	0.050	0.393
5.50	0.134	0.339	0.051	0.387
6.00	---	0.333	---	0.390
6.50	---	0.339	---	0.385
7.00	0.144	0.340	0.058	0.386
7.50	0.125	0.336	0.058	0.388
8.00	0.132	0.332	0.061	0.400
8.50	0.129	0.328	0.056	0.393
9.00	0.126	0.329	0.060	0.393
9.50	0.126	0.328	0.055	0.399
10.00	0.126	0.325	0.054	0.400
11.00	0.124	0.319	0.064	0.409
12.00	0.127	0.316	0.063	0.416
14.00	0.135	0.310	0.090	0.430
16.00	0.100	0.304	0.066	0.452
18.00	0.117	0.293	0.071	0.446
20.00	0.126	0.287	0.068	0.460
25.00	0.126	0.276	0.073	0.484
30.00	0.130	0.267	0.071	0.497
35.00	0.125	0.264	0.069	0.512

EXPERIMENTAL DATA FOR RUN 130

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.044055	0.043201
Distillate Rate, moles/hr.	0.028644	0.026289
Bottoms Rate, moles/hr.	0.015411	0.016912
Feed Composition, m. f.*	0.749	0.627
Column Holdup, ml. liquid	-----	50.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.909	0.464
1.00	0.909	0.470
2.00	0.909	0.473
3.00	0.905	0.477
4.00	0.895	0.479
5.00	0.880	0.474
6.00	0.871	0.474
7.00	0.861	0.460
8.00	0.856	0.446
9.00	0.856	0.446
10.00	0.852	0.439
11.00	0.847	0.430
12.00	0.849	0.435
14.00	0.853	0.414
16.00	0.852	0.410
18.00	0.836	0.405
20.00	0.849	0.394
25.00	0.851	0.365
30.00	0.846	0.350
35.00	0.850	0.369

EXPERIMENTAL DATA FOR RUN 131

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, Moles/hr.	0.046544	0.046607
Distillate Rate, moles/hr.	0.025274	0.026219
Bottoms Rate, moles/hr.	0.021270	0.020388
Feed Composition, m. f.*	0.742	0.631
Column Holdup, ml. liquid	-----	54.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.902	0.532
1.00	0.897	0.541
2.00	0.897	0.544
3.00	0.902	0.543
4.00	0.893	0.543
5.00	0.883	0.536
6.00	0.870	0.537
7.00	0.868	0.520
8.00	0.866	0.525
9.00	0.862	0.520
10.00	0.854	0.513
11.00	0.852	0.504
12.00	0.853	0.503
14.00	0.851	0.485
16.00	0.849	0.482
18.00	0.853	0.462
20.00	0.853	0.451
25.00	0.851	0.429
30.00	0.849	0.409
35.00	0.845	0.392

EXPERIMENTAL DATA FOR RUN 132

Hexane-Heptane

<u>Parameter</u>	<u>Initial Steady-State</u>	<u>Final Steady-State</u>
Feed Rate, moles/hr.	0.049322	0.049736
Distillate Rate, moles/hr.	0.021020	0.021389
Bottoms Rate, moles/hr.	0.028302	0.028347
Feed Composition, m.f.*	0.540	0.758
Column Holdup, ml. liquid	-----	126.0

*Compositions are given as mole fraction hexane.

Product Stream Compositions

<u>Time, min.</u>	<u>Distillate Composition</u>	<u>Bottoms Composition</u>
0.00	0.803	0.353
1.00	0.803	0.353
2.00	0.804	0.360
3.00	0.812	0.356
4.00	0.844	0.350
5.00	0.864	0.354
6.00	0.876	0.359
7.00	0.890	0.371
8.00	0.888	0.388
9.00	0.891	0.389
10.00	0.902	0.405
11.00	0.906	0.424
12.00	0.907	0.424
14.00	0.907	0.488
16.00	0.908	0.460
18.00	0.910	0.471
20.00	0.908	0.487
25.00	0.912	0.509
30.00	0.906	0.529
35.00	0.904	0.546

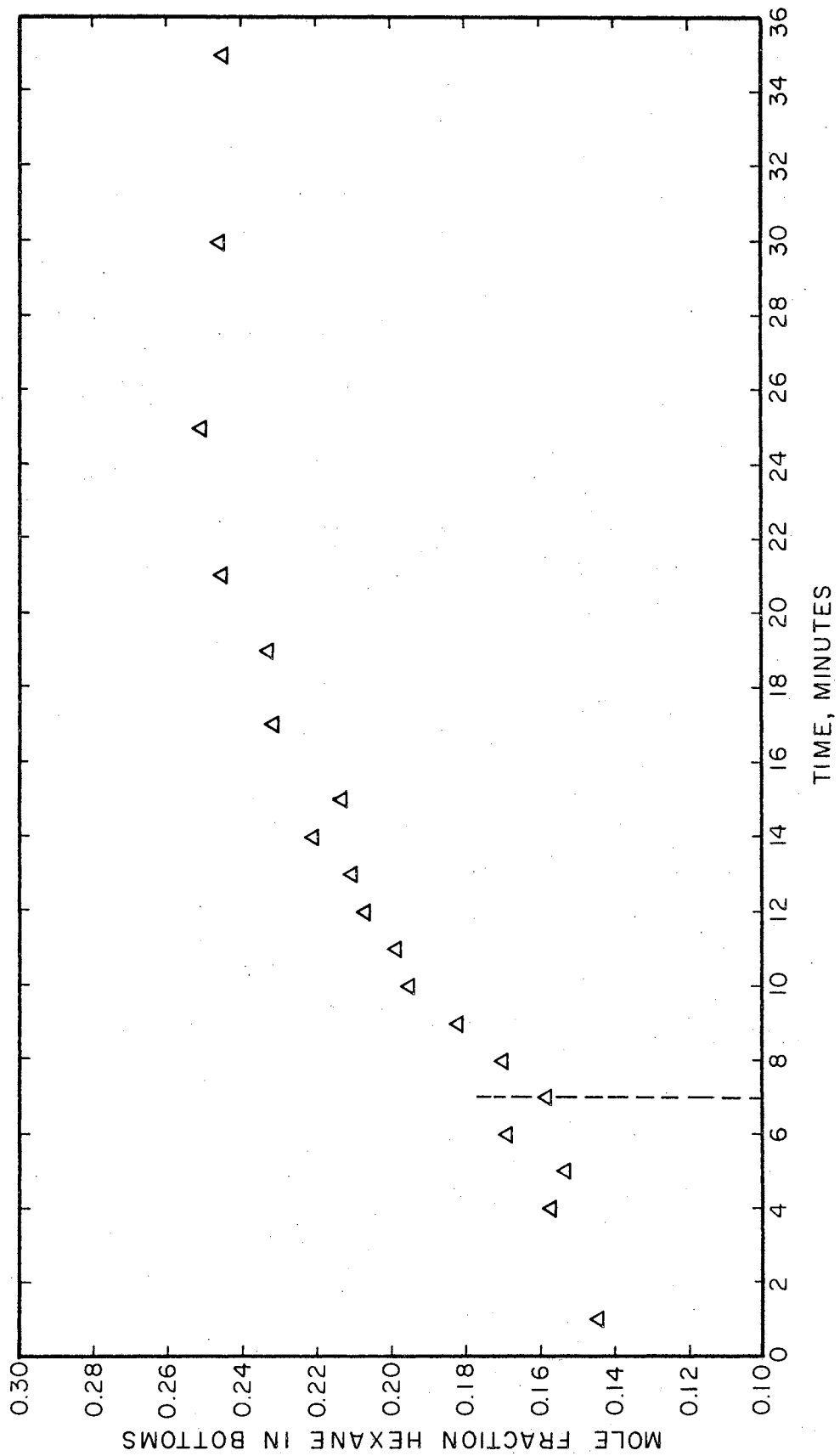


Figure 22. Experimental Data for Run 105

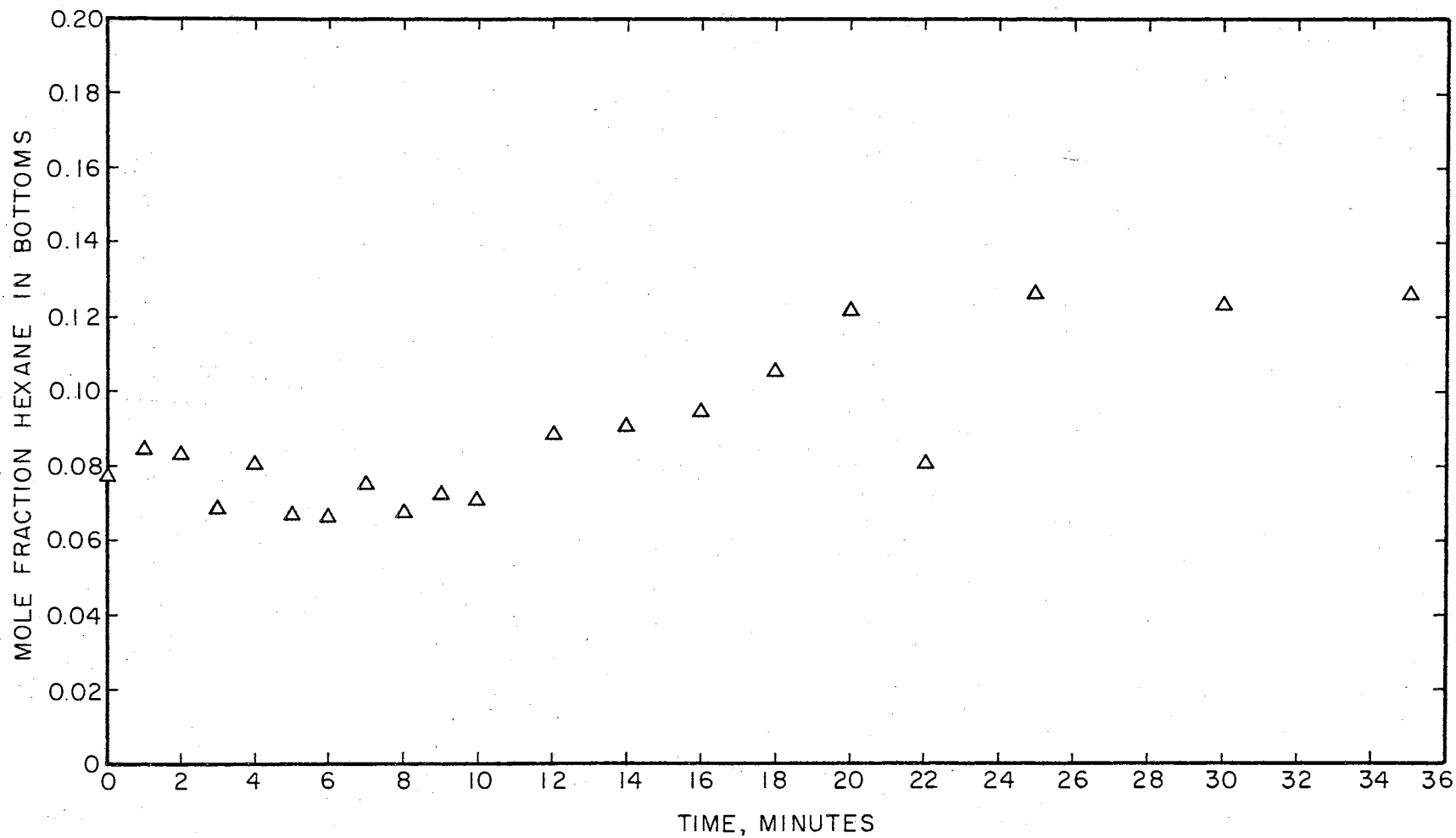


Figure 23. Experimental Data for Run 106

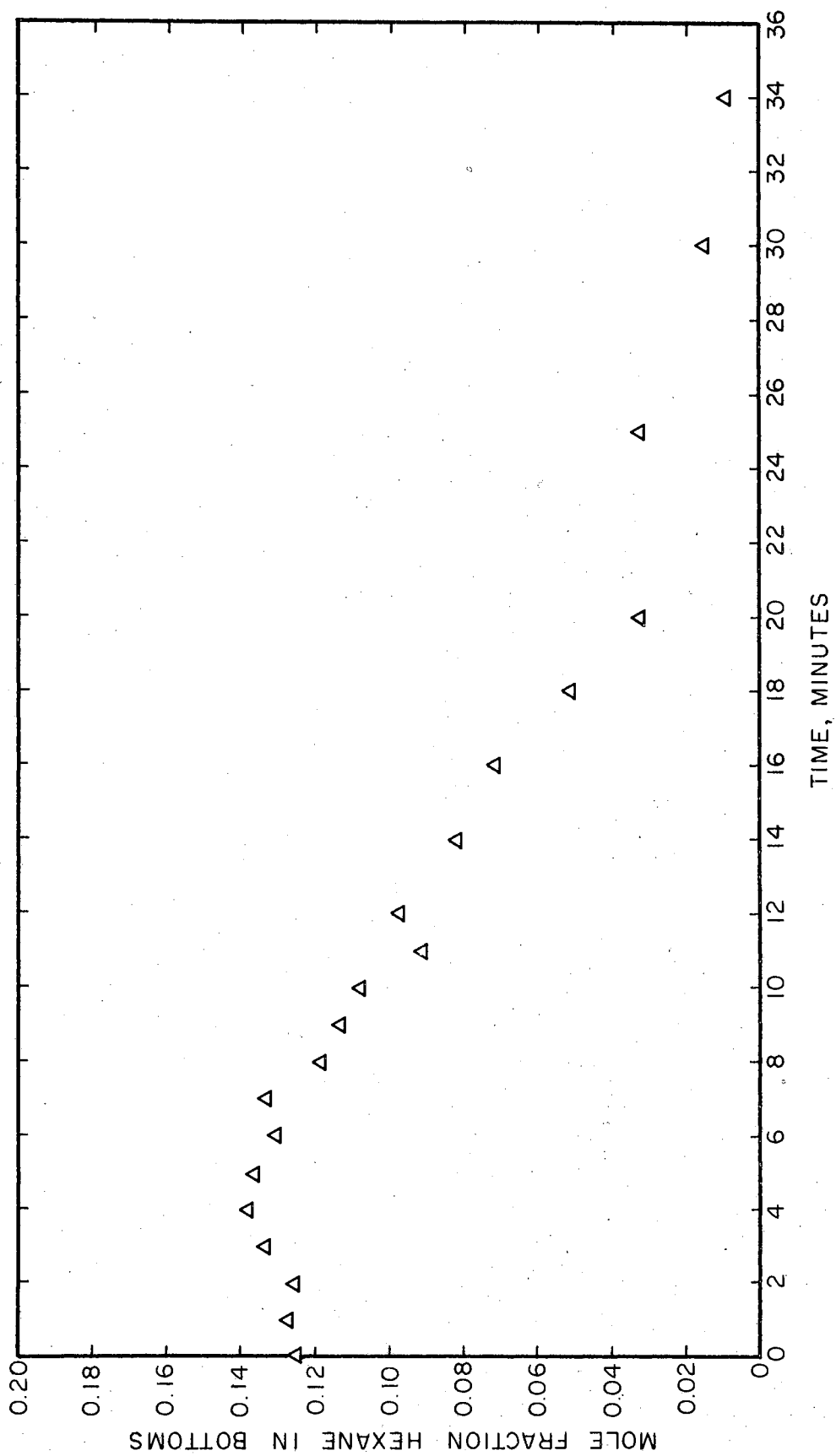


Figure 24. Experimental Data for Run 107

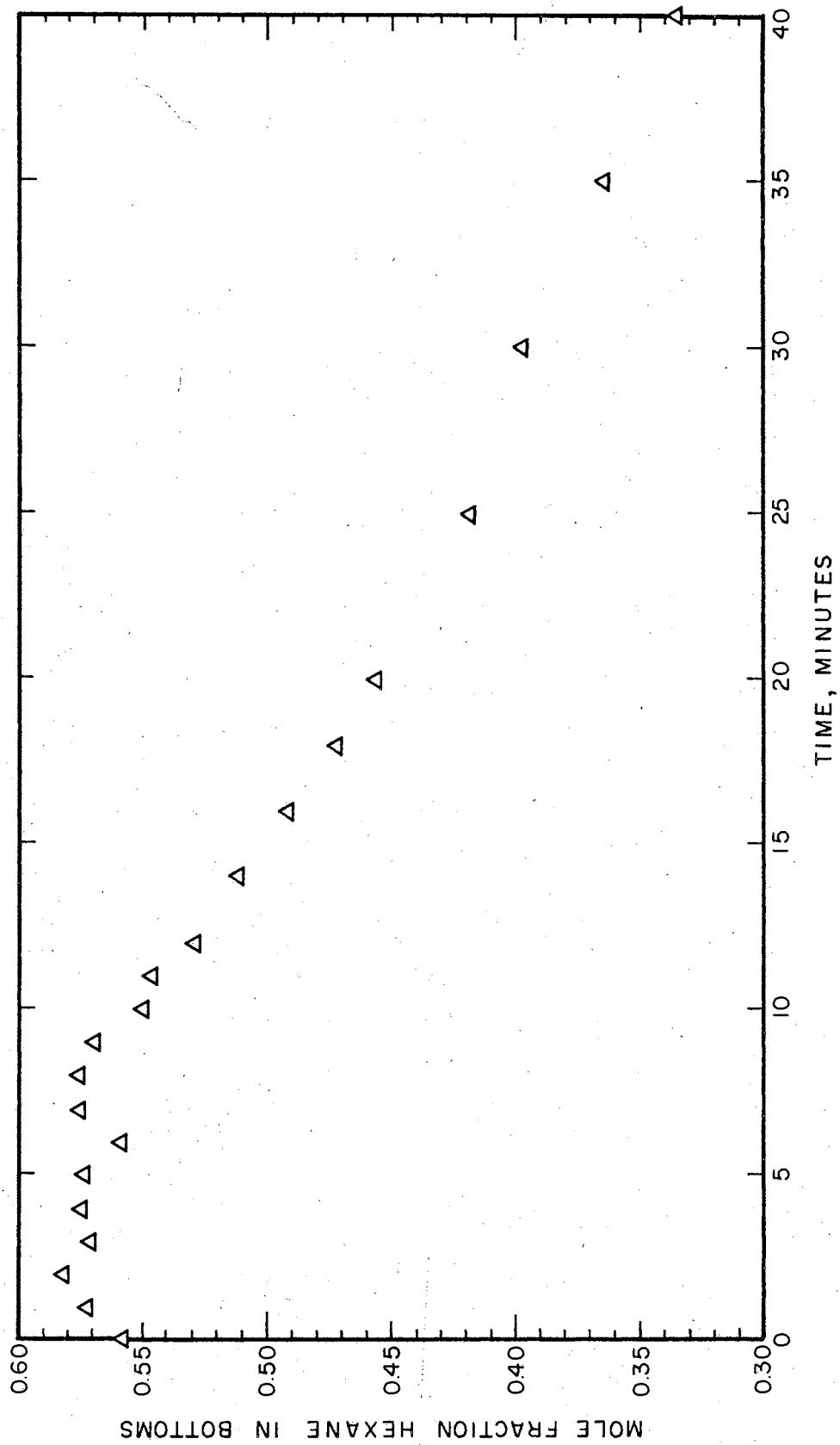


Figure 25. Experimental Data for Run 110

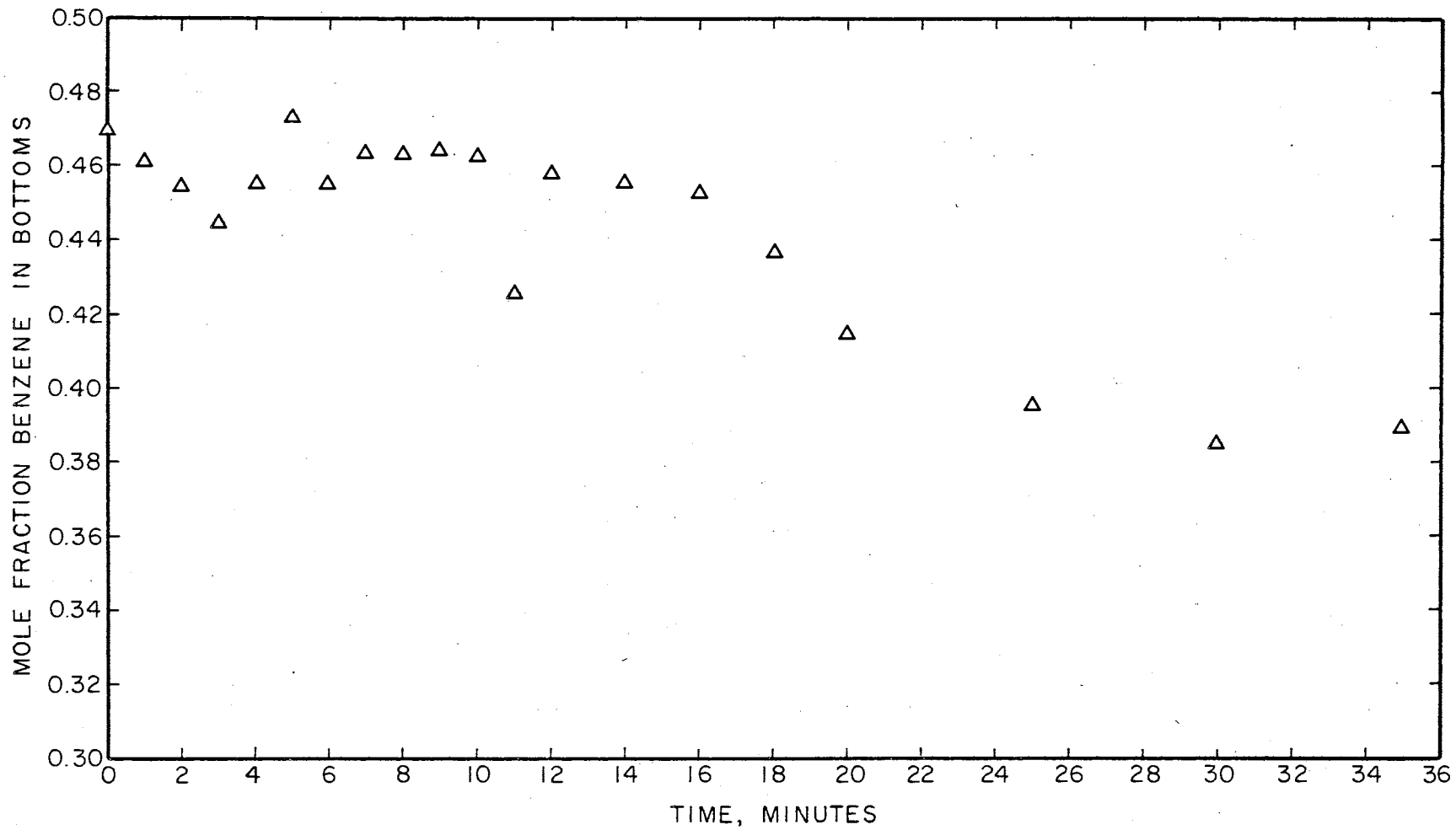


Figure 26. Experimental Data for Run III

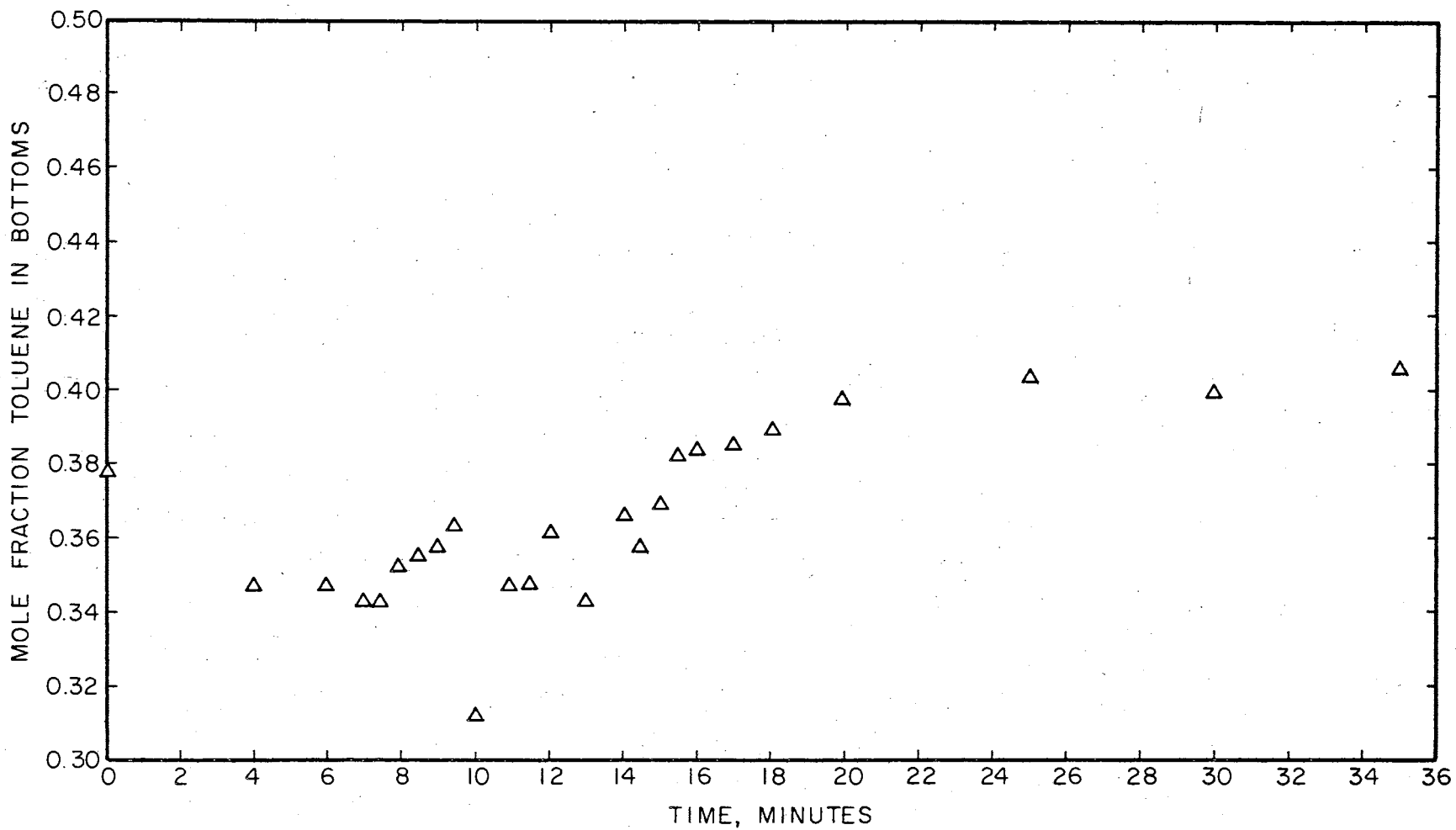


Figure 27. Experimental Data for Run 115

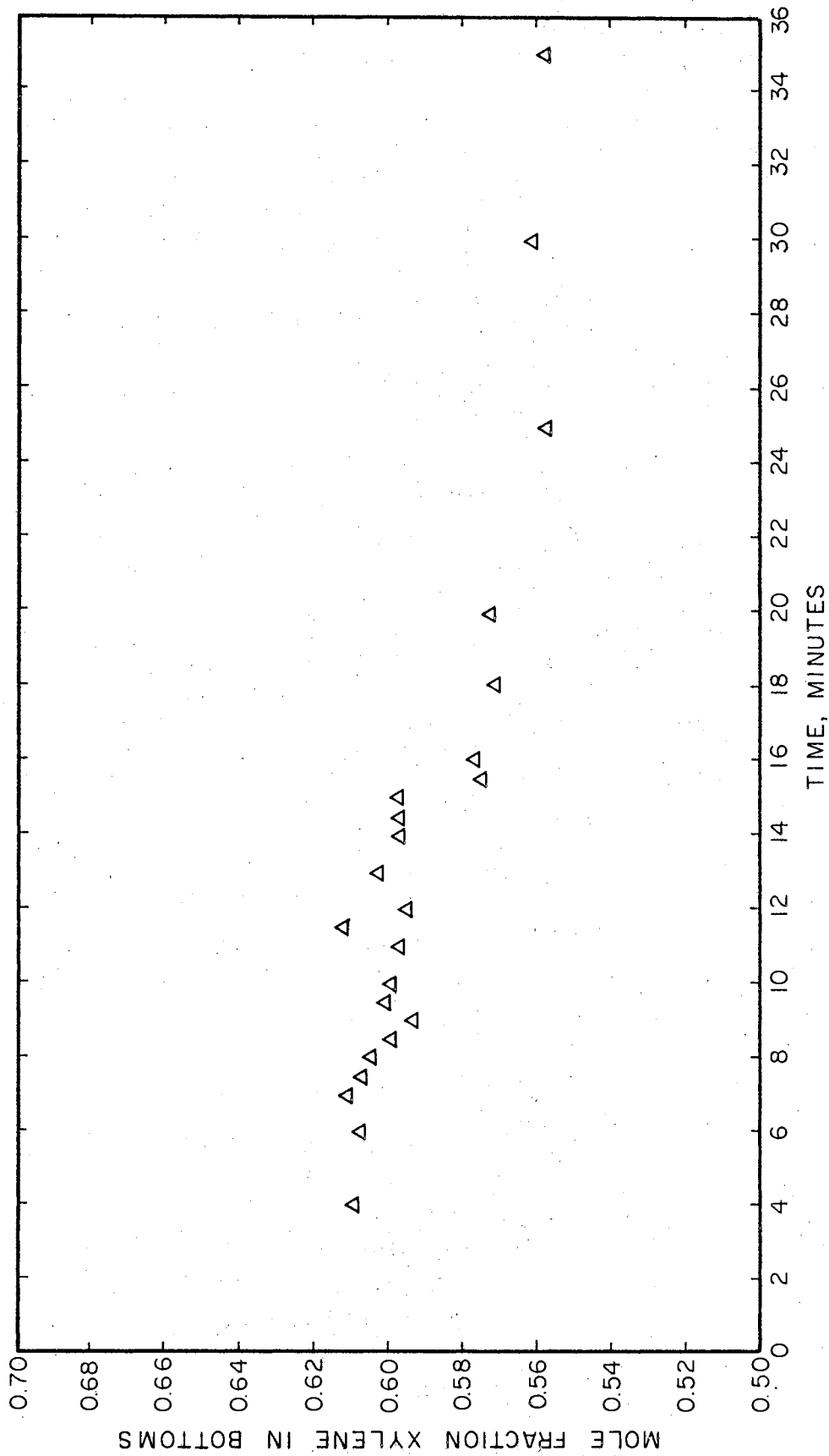


Figure 28. Experimental Data for Run 115

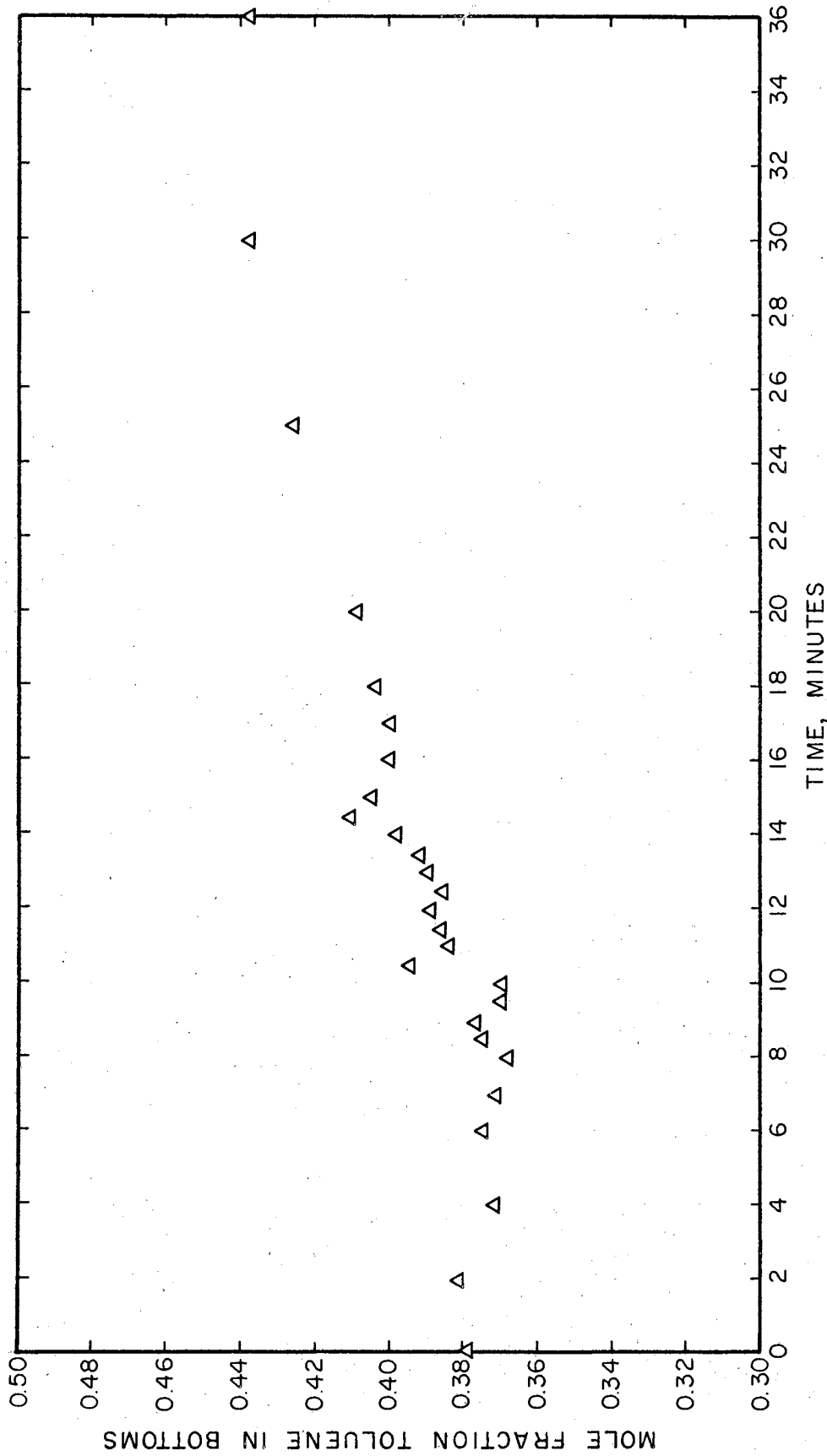


Figure 29. Experimental Data for Run 116

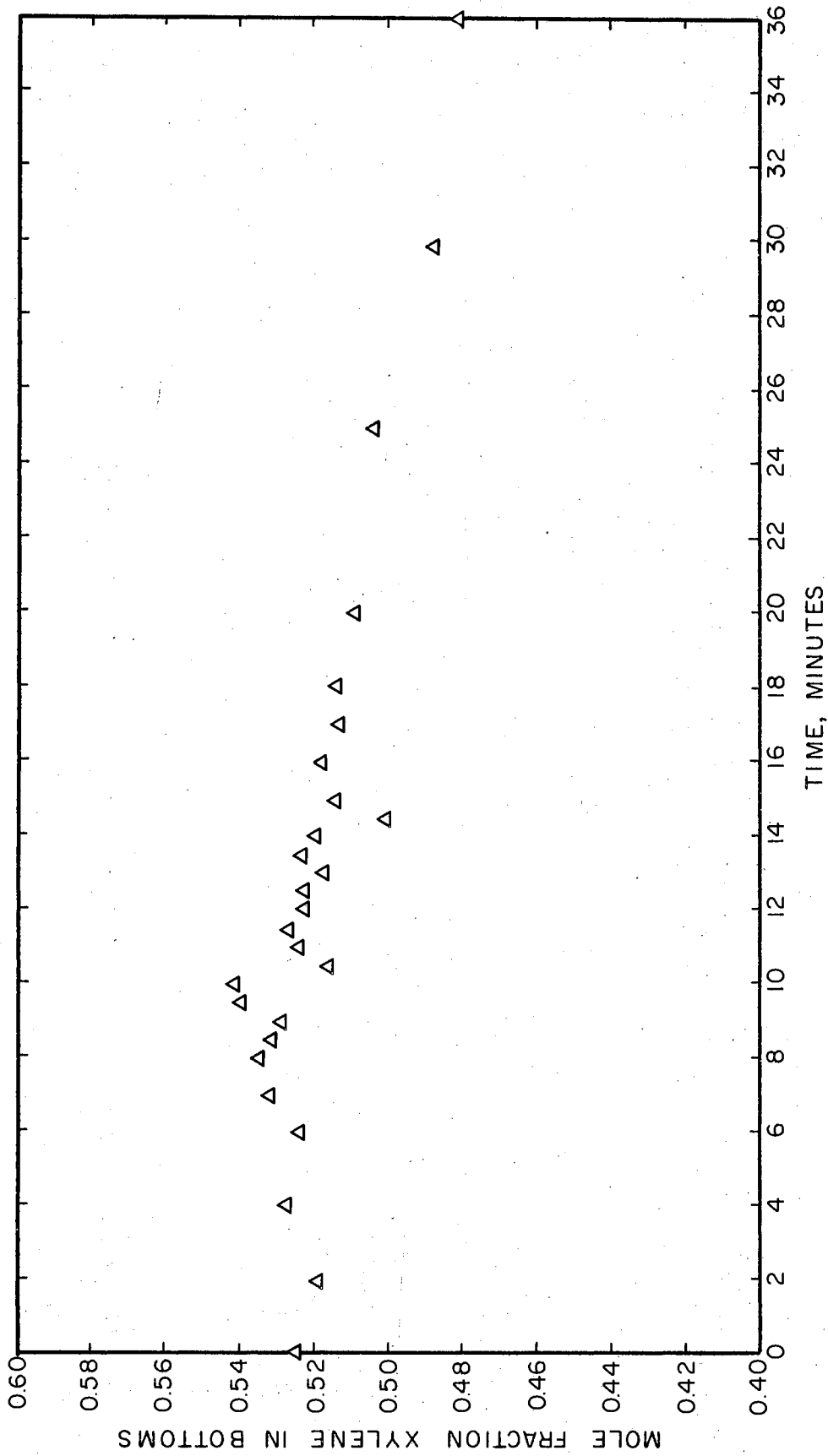


Figure 30. Experimental Data for Run 116

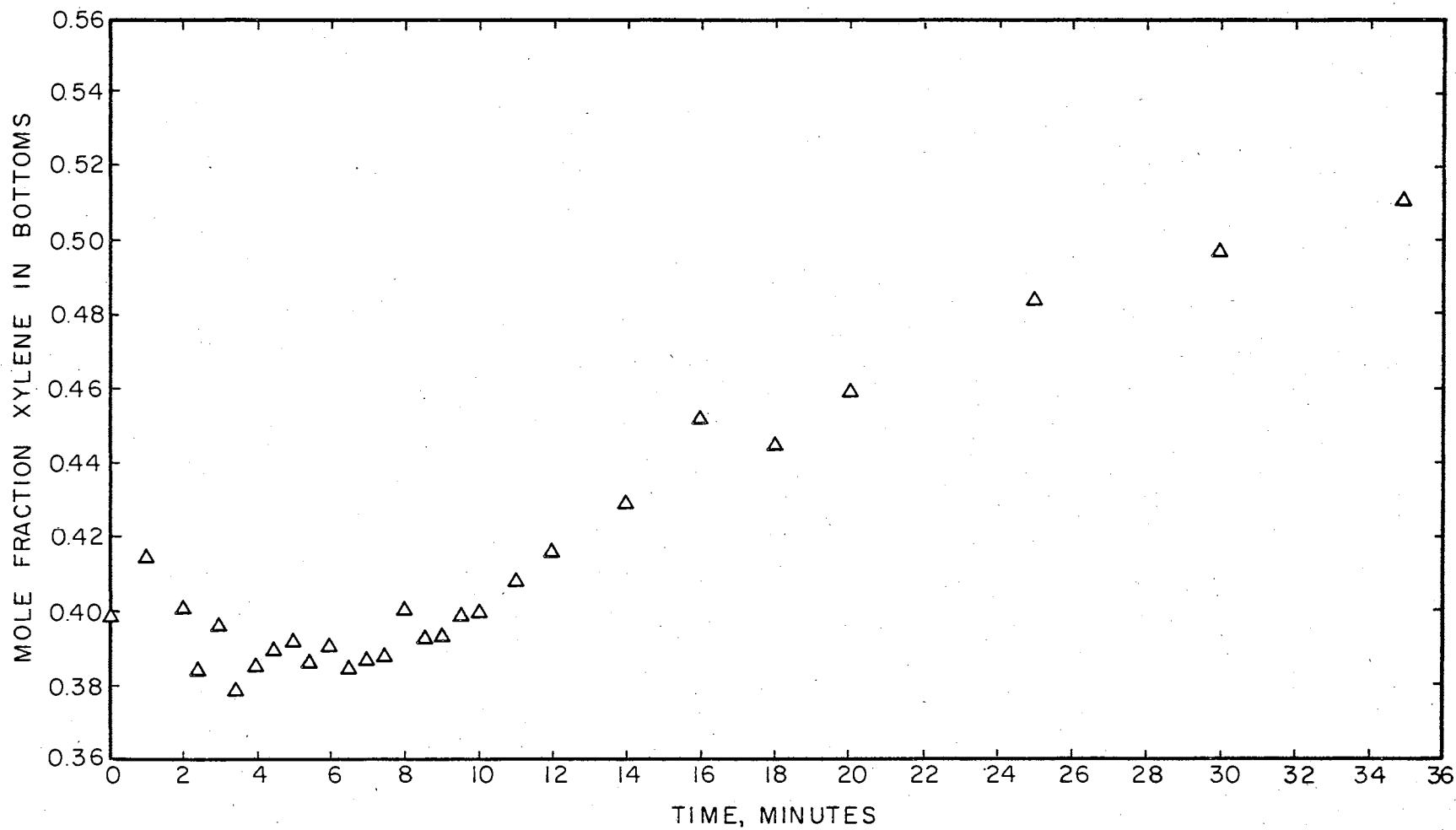


Figure 31. Experimental Data for Run 117

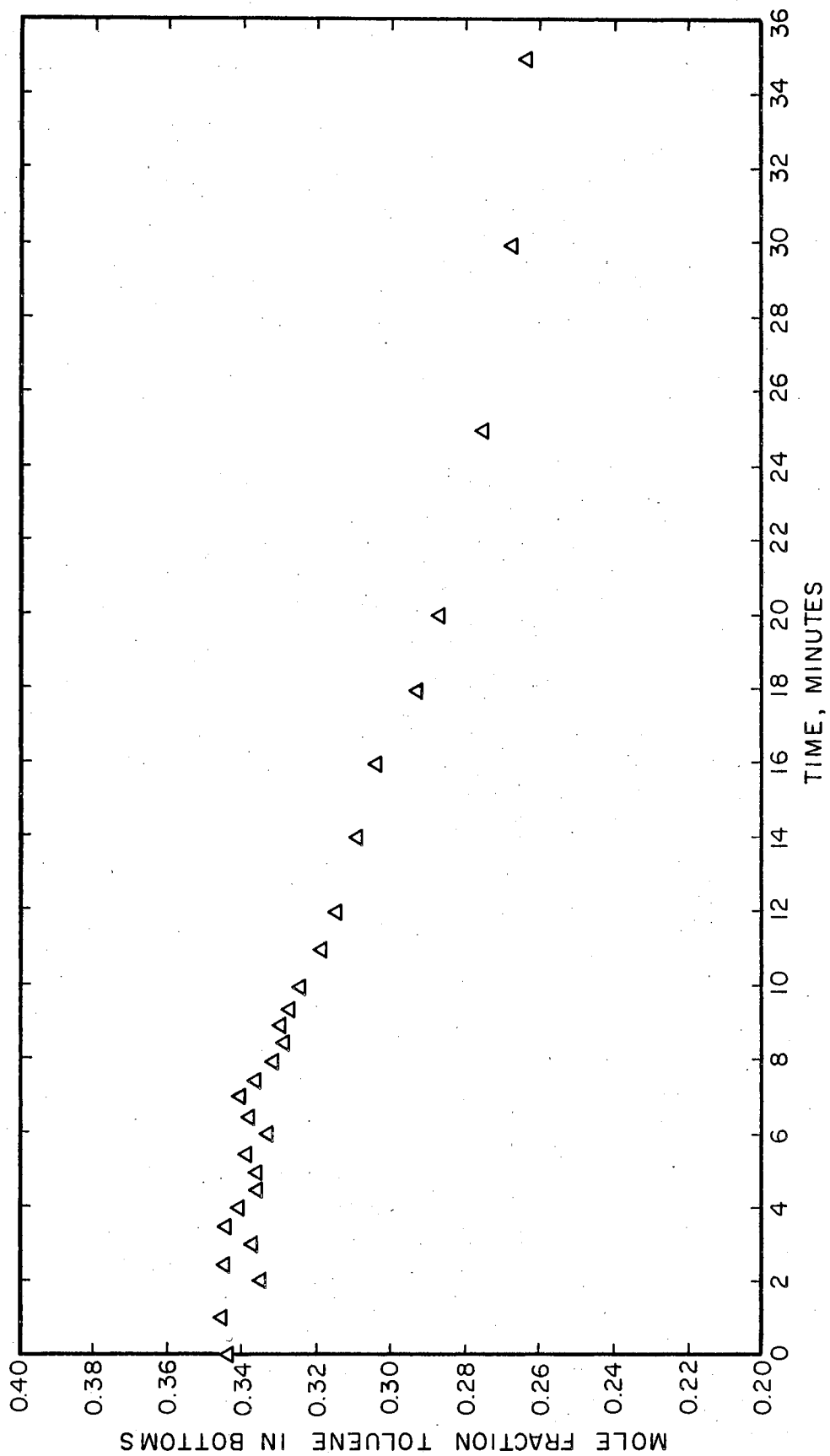


Figure 32. Experimental Data for Run 117

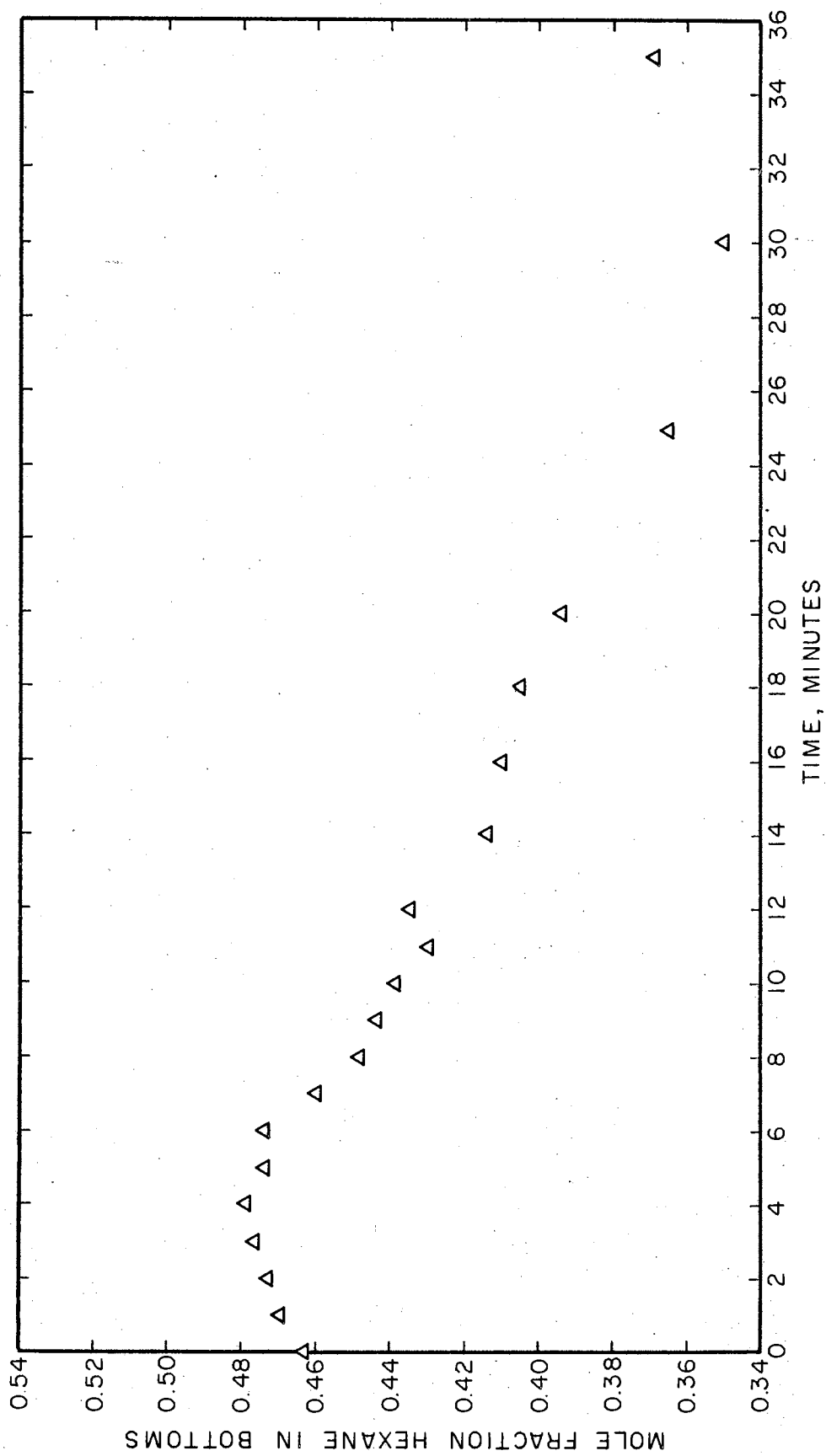


Figure 33. Experimental Data for Run 130

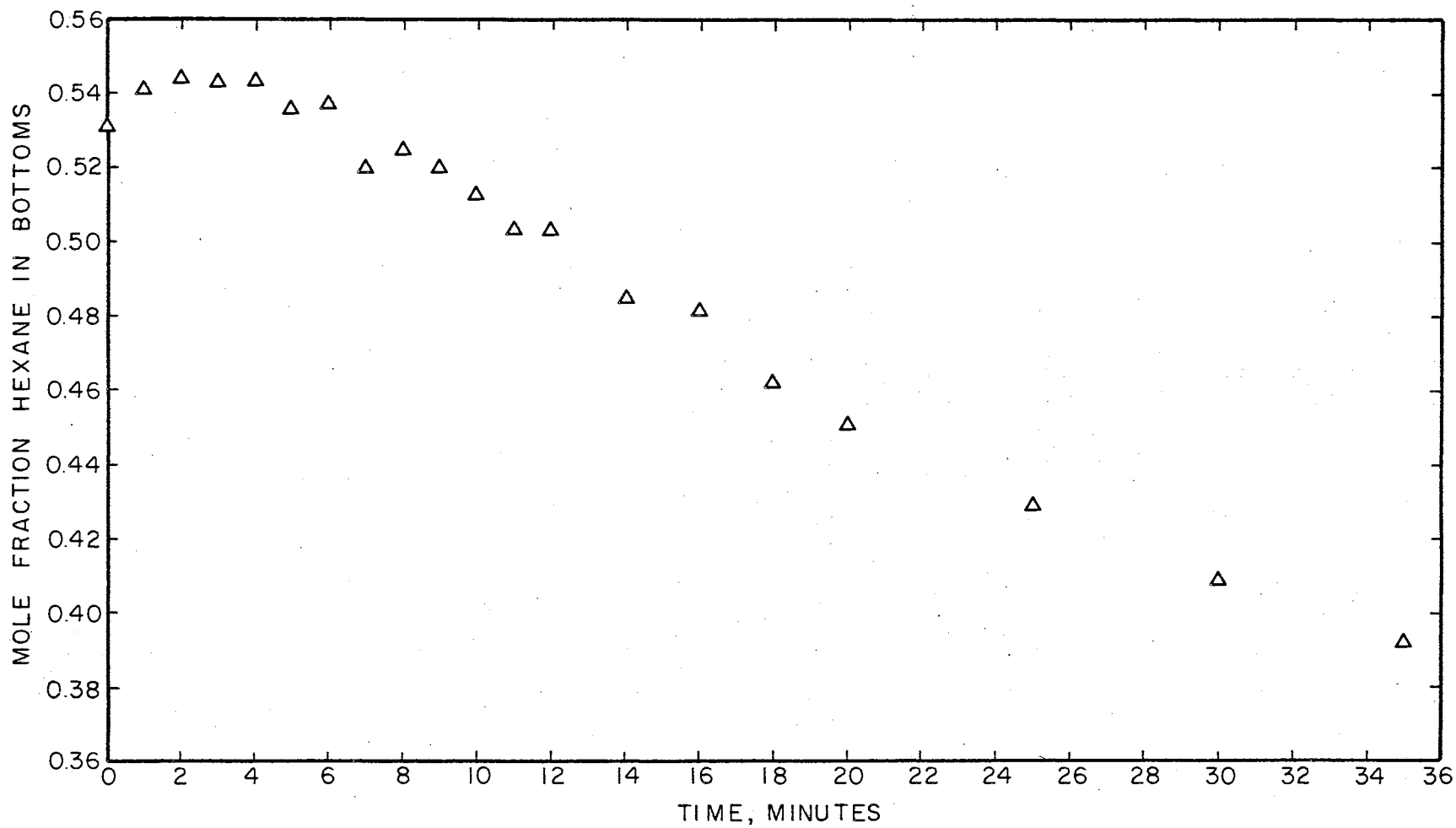


Figure 34. Experimental Data for Run 131

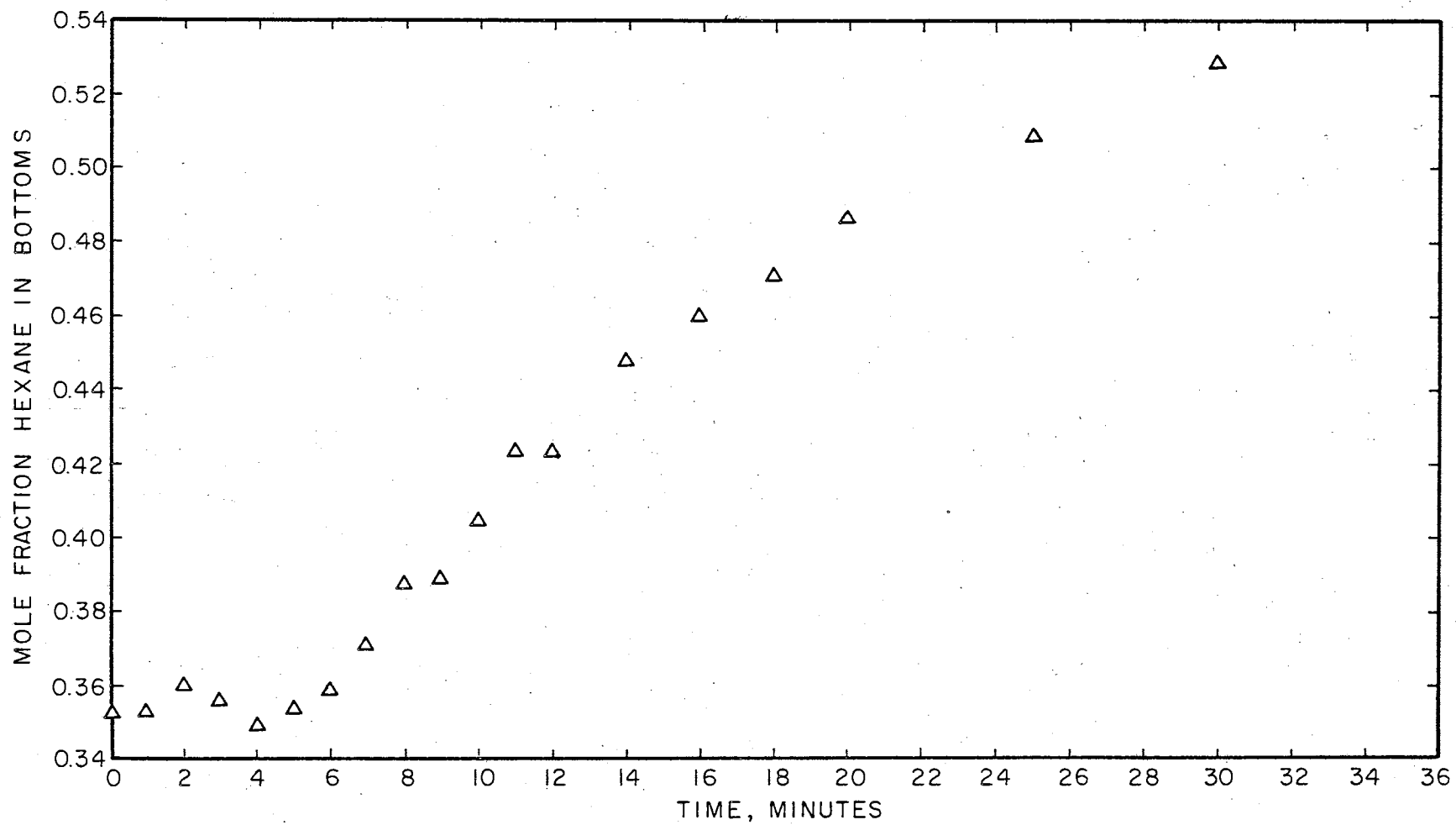


Figure 35. Experimental Data for Run 132

APPENDIX E

DERIVATION OF LOGARITHMIC INTEGRATION FORMULA

DERIVATION OF LOGARITHMIC INTEGRATION FORMULA

One of the most difficult differential equations to solve with numerical methods is the equation

$$\frac{dx}{dt} = -Kx \quad (91)$$

which is the differential form of the equation

$$x(t) = x_0 e^{-t/T} \quad (92)$$

Numerical solution can be difficult because the rate of change of x is proportional to x and for small values of the independent variable t , Δx is usually large. To obtain an accurate solution of equation (1), the size of the differential time increment is usually made very small. This does improve the accuracy of the solution, but it also increases the number of incremental steps that are required. If a digital computer is being used, the computing time is increased proportionally to the decrease in the size of the time increment.

A simple technique that greatly improves the accuracy of the numerical solution of equation (90) consists of rewriting the equation according to

$$\frac{1}{x} \frac{dx}{dt} = -K \quad (93)$$

The numerical solution of equation (93) is much more accurate than that of equation (91). Since the technique linearizes equation (91), longer time steps can be used and a solution is obtained much more easily.

Consider that the equation

$$\frac{dx}{dt} = f(x,t) \quad (94)$$

is to be solved numerically, that the solution is exponential in form, and that a steady-state solution is reached at a very large time. First rewrite the equation according to

$$\frac{1}{x} \frac{dx}{dt} = \frac{d \ln x}{dt} = \frac{1}{x} f(x,t) \quad (95)$$

Using the Euler First Order Method, equation (94) can be integrated numerically according to

$$\ln x \Big|_{t+\Delta t} = \ln x \Big|_t + \Delta t \left[\frac{1}{x} f(x,t) \right] \Big|_t \quad (96)$$

The value of the dependent variable x can be obtained according to the equation

$$x \Big|_{t+\Delta t} = \exp \left[\ln x \Big|_t + \Delta t \left[\frac{1}{x} f(x,t) \right] \Big|_t \right] \quad (97)$$

Equations (41) and (42) are identical in form to equation (97).

Since the differential equations that describe the dynamic behavior of a distillation column are similar to equation (94), the possibility of using the linearization technique in the feed forward control system was investigated. To determine the utility of the linearization technique, it was compared with Ball's method as described by Burman (7). Both methods were incorporated into identical control programs and run on the same problem. There was no difference in the results. Since the same time increment was used in both solutions, the linearization technique was concluded to be as accurate as Ball's method. While there was no difference in the results, the computing times required to obtain a solution were quite different. Ball's method required considerably more computer time than the linearization method. On the basis of this comparison of the results and the computing times, the linearization

technique was selected for incorporation into the feed forward control scheme.

VITA

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Thesis: DISTILLATION COLUMN DYNAMICS--AN EXPERIMENTAL STUDY

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