

DYNAMIC RESPONSE OF A  
PACKED, TUBULAR REACTOR

by

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## I. INTRODUCTION

The tubular flow reactor is by far the most common type presently used in the chemical industry. In spite of its wide usage, the mathematical descriptions used to predict the performance of the tubular flow reactor are frequently inadequate and inaccurate. In general, the flow of reactants through the reactor is assumed to follow a perfect plug flow, or no-mixing model. In reality, the ideal plug flow does not exist.

The true description of the flow through a tubular flow reactor may be thought of as a combination of plug flow, completely mixed or stirred-tank flow, and bypass flow. Through the application of several recently developed analysis procedures, parameters of mixing may be evaluated experimentally.

Notwithstanding recent advances, available data for the dynamic response characteristics of tubular reactors are few. A full study of the dynamic response of industrial and pilot plant scale tubular reactors would be a boon to both academic and industrial research efforts.

The purpose of this investigation was to design and construct a tubular flow reactor capable of handling both liquid and gas phase reactions, both homogeneous and catalyzed, and to demonstrate its ability to produce reliable dynamic response information.

## II. LITERATURE REVIEW

The following section is a review of literature in the fields of fluid flow in packed beds and dynamic analysis of various systems, which is applicable to the investigation of the dynamic response of a packed, tubular reactor.

### Fluid Flow in Packed Beds

There are two main approaches to the theoretical description of flow through packed beds<sup>(3)</sup>. One approach considers the packed column as a tortuous mass of tubes of varying cross-sectional area. The mathematical description of this weird array of plumbing is then developed by applying theories for single, straight pipes to the crooked tubes. The second approach envisions the particles of column packing as individual submerged objects, and their overall effect is estimated by summing up the effects produced by the individual particles.

Reynolds Number. The usual measure for the description of turbulence is the Reynolds Number, which is a function both of the properties of the fluid flowing and the channel through which the fluid flows. The physical significance of the Reynolds Number is that it is the quotient of the inertial forces divided by the viscous forces present. In the case of a packed column, the characteristic dimension

must be a function of particle size and geometry, and of void volume. The hydraulic radius, defined as the cross sectional area divided by the wetted perimeter of the duct, can be expressed in terms of void fraction and a surface function.

$$R_h = \frac{\epsilon}{a}$$

$$R_h = \text{hydraulic radius} \quad (1)$$

$\epsilon$  = volume of voids/volume of bed

$a$  = wetted surface area/volume of bed

The term  $a$  is related to the "specific surface"  $a_v$  (the average surface area to volume ratio of the particles forming the bed) by

$$a = a_v(1-\epsilon) \quad (2)$$

A mean particle diameter,  $D_p$ , may be defined as the diameter of a sphere having the same surface area to volume ratio as the particles:

$$D_p = 6/a_v, \quad (3)$$

and a Reynolds Number is given by

$$Re = D_p G_o / \mu \quad (4)$$

where:  $G_o$  = mass velocity of the fluid based on the cross-sectional area of the empty tube  
 $\mu$  = the viscosity of the fluid  
 $D_p$  = mean particle diameter as defined in (3)

Dispersion of Fluids in a Packed Bed. The two simplest models for dispersion of fluids in a packed bed are the piston or plug flow model and the completely mixed or stirred

tank model. Danckwerts<sup>(8)</sup> points out that many systems do not conform to either of the foregoing assumptions, with the result that calculations based on either model may be incorrect. He explains how distribution functions for residence times can be defined, and how they can be measured for actual systems. His explanations cover both empty and packed tubes, and illustrations are given of the use of distribution functions for the calculation of the efficiency of reactors and blenders. In addition, he describes the use of models to predict the distribution of residence time in large systems. The actual situation of dispersion within a packed column can be described as a combination of plug flow, completely mixed flow, and bypass flow or forward recycle.

Effects of Particle Size on Velocity Distribution.

Schwartz and Smith<sup>(16)</sup> and other investigators<sup>(1, 15)</sup> studied the effects of packing on the variation of flow velocity as a function of radial position. They found that when the ratio of the diameter of the tube to that of the packing was less than about thirty, the axial flow rate varied significantly as a function of radial position, which precluded the assumption of plug flow. This effect was most pronounced approximately one particle diameter from the tube wall, and was explained by the variation in effective packing density caused by the presence of the tube wall. The assumption of plug flow was found to be quite valid when the ratio of tube to particle diameter was greater than thirty.

Axial Dispersion. When radial dispersion effects can be neglected, the deviation from ideal plug flow of the fluid passing through a packed bed may be expressed in terms of axial dispersion. Carter and Bir<sup>(6)</sup> describe a helium tracer technique for the measurement of axial mixing in high pressure tubular reactors. Their equipment included a leak-detector-type mass spectrometer sensitized for helium. Helium was injected at two levels in a reactor through which ethylene was flowing. The axial mixing coefficient was determined from the difference in the helium concentration obtained when the injection point was changed from one level to the other.

Other studies of axial dispersion in packed beds were made by Garberry<sup>(5)</sup> and Converse<sup>(7)</sup>, who developed a technique for determining the effect of the velocity profile on axial dispersion and a correlation based on the use of the Peclet Number.

Temperature Effects. Hawthorn<sup>(9)</sup> presents an analysis of the effects of temperature variations in the radial direction on the values of effective diffusion coefficients. When a condition of laminar flow exists, radial temperature variation may cause an increase or decrease in the effective diffusivity by a factor of two or three, as compared with calculations based on isothermal flow. At Reynolds Numbers greater than 10,000, there is not a significant variation in diffusivity as compared with the constant temperature value. In turbulent flow where the Reynolds Numbers are below 10,000,



there are definitely effects from radial temperature variation, but in this transition region they can only be roughly estimated.

### Dynamic Analysis

Until recently, most analyses of chemical systems were performed when the system under study approached a steady state condition, where there were no major fluctuations of the variables studied with respect to time. Advances in instrumentation and computer technology have allowed the rapid recording of data and speedy calculation of the complex equations associated with transient systems. While the final goal of dynamic response studies is frequently computer control, a very useful facet of dynamic analysis is the mathematical description of the system being studied.

Describing the System. The description of a system, whether it be a complex control system, a distillation column, a chemical reactor, or simply a length of tubing, may be conveniently expressed in terms of frequency response. A plot of frequency response is based on the reaction of the system to a sinusoidally varying input signal. The obvious reason that the sine function waveform is used as an input signal<sup>(19)</sup> is that any curve may be constructed by combining a number of sine waves, and thus the output signal may be fairly easily expressed in terms of the input function.

Three characteristics which completely describe a sine wave are amplitude, frequency, and phase angle. The period is also used, but since it is simply the reciprocal of the frequency, it is not an independent variable. Once the response of a system has been measured for input signals of sine waves of all frequencies, selected values of frequency and amplitude may be combined to produce any desired waveform, and to predict the response of the system to that waveform. Response of a system to a sine wave input of constant frequency and amplitude is measured in terms of the ratio of the amplitude of the output wave to that of the input wave (magnitude ratio or gain) and the difference in phase of the input wave and the output (phase lag).

These characteristic quantities may be conveniently expressed in graphical form in either of two ways. The Bode diagram consists of two curves for each system. One is a log-log plot of gain versus frequency, the other a semi-log plot of phase lag versus frequency. A typical Bode diagram is shown in Figure 1. The Nyquist plot combines the two curves of the Bode diagram on a polar representation, frequency being indicated along the curve. A typical Nyquist plot is presented as Figure 2, page 9.

Experimental Determination of Frequency Response. In some cases, particularly in electrical systems, it is convenient to actually use a sine wave input and to directly measure the output. In fluid flow systems, however, the production of

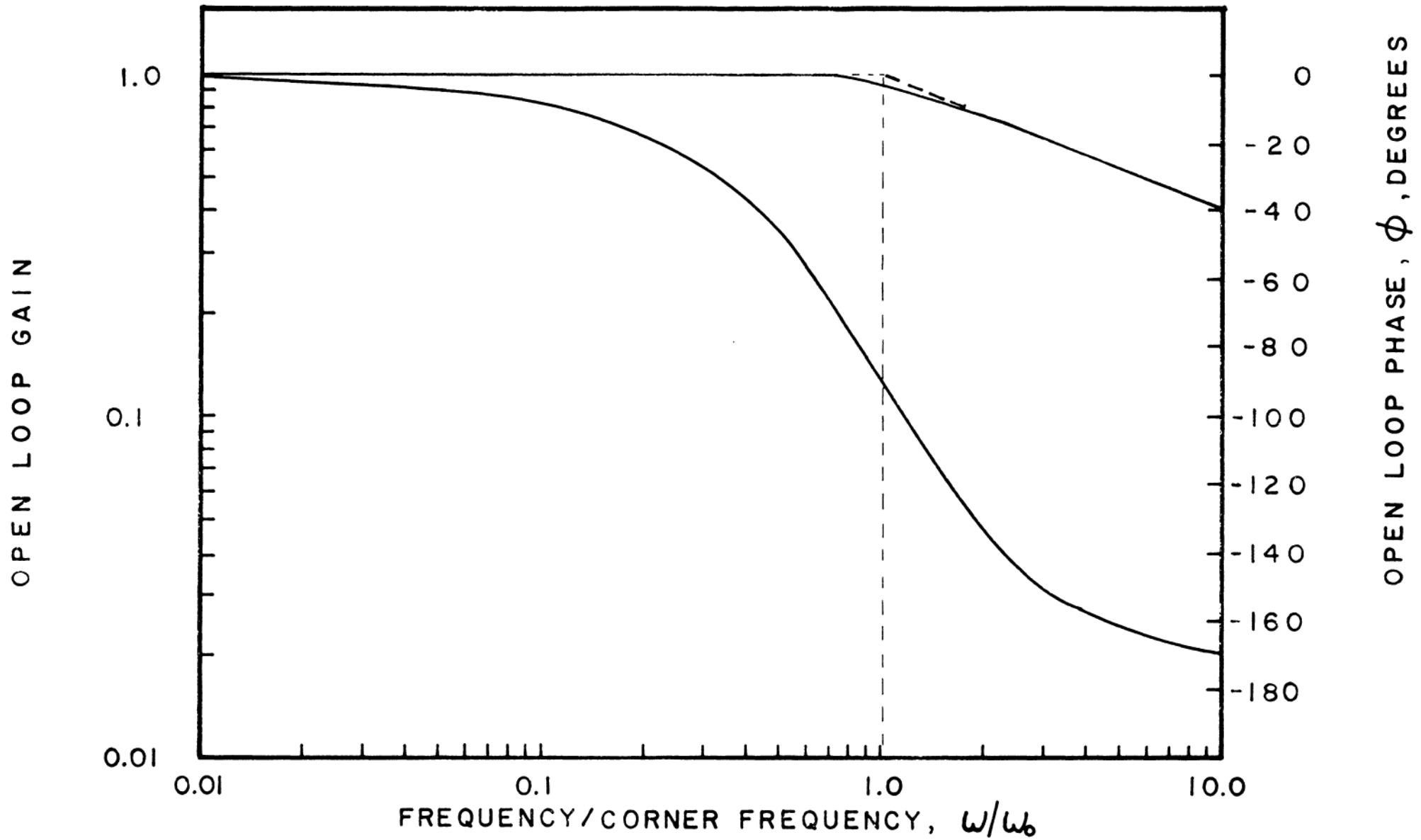


FIGURE 1. TYPICAL BODE DIAGRAM

Chestnut, H. and R. W. Mayer: "Servomechanisms and Regulating System Design," p. 127, John Wiley & Sons, Inc., New York, N.Y., 1961.

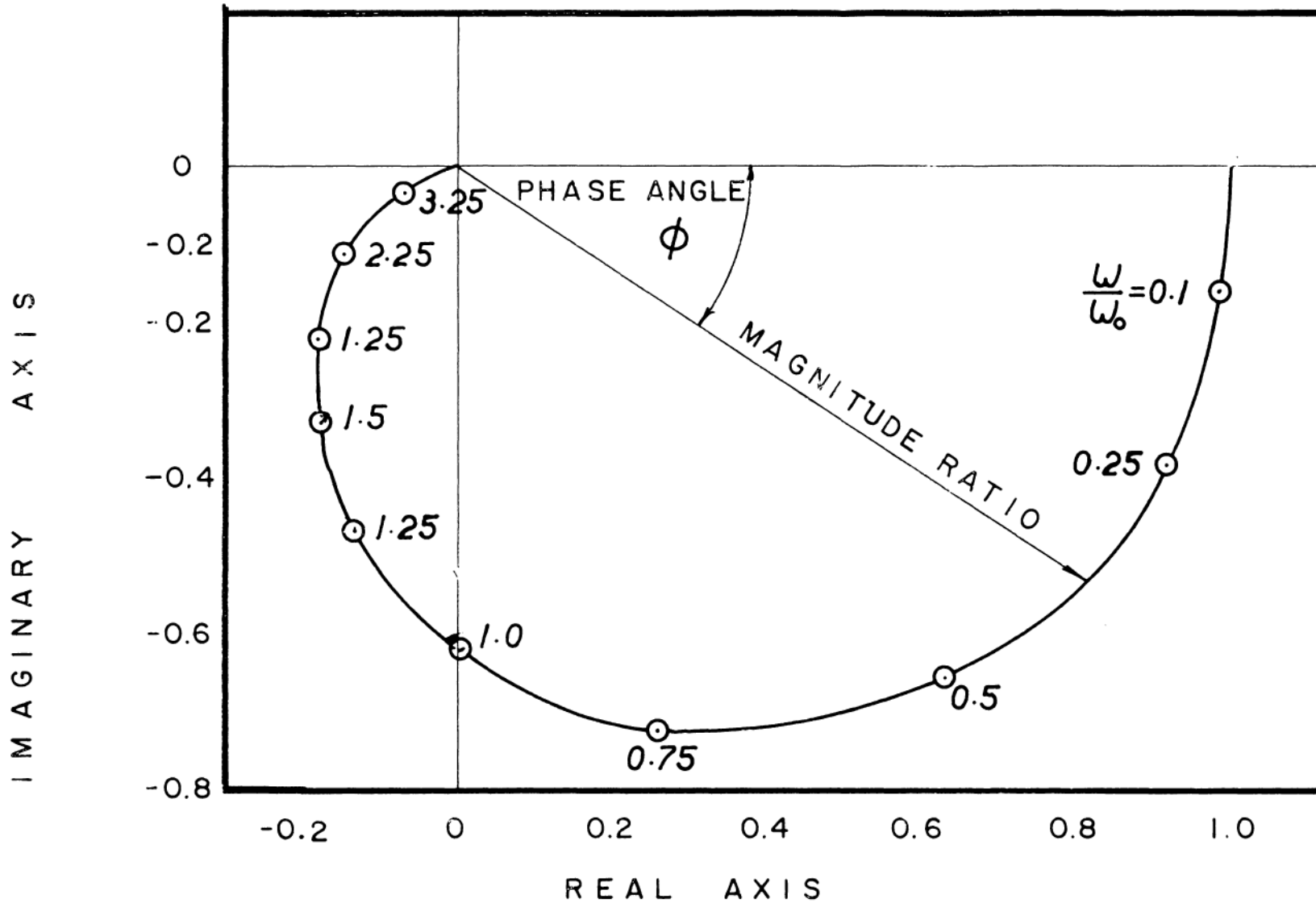


FIGURE 2. TYPICAL NYQUIST PLOT

Chestnut, H. and J. W. Mayer : "Servomechanisms and Regulating System Design," p. 122, John Wiley & Sons, New York, N.Y., 1961

a truly sinusoidal wave input becomes a difficult problem. Pump and valve systems for the simulation of nearly sinusoidal waves have been designed and constructed by several investigators. Kramers and Alberda<sup>(11)</sup> describe such an apparatus, and give illustrations of experimental results obtained in the study of longitudinal diffusion of liquid flow through packed Raschig rings and for backmixing of a liquid flowing over the packing of an absorption column.

#### Frequency Response from other than Sinusoidal Inputs.

In the field of fluid flow systems, it is sometimes more convenient to record responses to inputs other than sinusoidal, and then to calculate from these data corresponding response data for a truly sinusoidal input. Hougen and Walsh<sup>(10)</sup> describe a method for determining frequency response from known-pulse inputs. Teasdale<sup>(18)</sup> demonstrates a similar technique using a step input. An obvious advantage to the use of a step input in a fairly sluggish system is that a reasonably good step change may be introduced into the system manually without the necessity of physically measuring the exact form of the input. The change from initial to final value of the controlled variable may be assumed to follow a perfect step function.

Parametric Sensitivity. Bilous and Amundson<sup>(2)</sup> made rather extensive studies of the response of quasi-isothermal tubular reactors. They found that in some regions of operation small changes in certain variables, such as heat

transfer coefficient at the reactor wall or dilution of the feed, will produce large changes in the effluent. They show analytically that this parametric sensitivity may be predicted by analyzing the frequency response or the transient response of the reactor approximated by local linearization. They show also that, at least theoretically, instabilities may exist in the operation of the reactor, if the feed is partially a recycle stream. These instabilities are based on the transfer function as determined from frequency response data.

Turner<sup>(20)</sup> made a notable attempt to determine the parameters of packed beds by constructing ideal models of two packed beds. The models were so constructed that channeling was eliminated, and his analysis was made by frequency response measurement.

Frequency Response Data from Step Analysis. Teasdale<sup>(18)</sup> proposed a method for the approximate determination of frequency response data from the response of a system to a step input. The output from the step input is approximated by a series of steps functions which are  $\Delta t$  apart in time.  $\Delta t$  is chosen such that  $\omega\Delta t$  is less than one over the range of frequencies studied. ( $\omega$  is the frequency in radians per unit time.) When  $\omega\Delta t$  is greater than one, the results are confused by the sampling frequency.

The first step lags the beginning of the transient portion of the step-response curve by the time  $\Delta t/2$ , the second

step by time  $\frac{3\Delta t}{2}$ , the  $i$ -th step by  $\frac{(2i-1)\Delta t}{2}$ , and so forth. If the transfer function of the system is now denoted by the symbol  $\underline{GH}$ , Teasdale's formula may be written

$$\underline{GH} = \sum_{i=1}^{\infty} (\Delta y)_i \frac{1}{-j\omega \Delta t k_i} \quad (5)$$

where:  $(\Delta y)_i = y_i - y_{i-1}$

$y_i$  = the magnitude of the  $i$ -th step, measured from the base line

$\frac{1}{-j\omega \Delta t k_i}$  = the direction of the  $i$ -th step vector, angle in radians

$\omega$  = the frequency of the hypothetical sine-wave input, (arbitrarily chosen) with  $\omega \Delta t < 1$

$\Delta t$  = the time increment between steps

$k_i$  = the term  $(2i-1)/2$

The interrelation of  $y_i$ ,  $\Delta t$ ,  $k_i$ , and  $\Delta y_i$  is shown in Figure 3. Expressed in rectangular coordinates of the imaginary plane, the expression for the transfer function becomes

$$\underline{GH} = \sum_{i=1}^{\infty} (\Delta y)_i \cos(\omega \Delta t k_i) - j \sum_{i=1}^{\infty} (\Delta y)_i \sin(\omega \Delta t k_i) \quad (6)$$

where:  $j$  = index of the imaginary coordinate.

This expression is of the form

$$\underline{GH} = C - jB \quad (7)$$

where:  $C = \sum_{i=1}^{\infty} (\Delta y)_i \cos(\omega \Delta t k_i)$   
and  $B = \sum_{i=1}^{\infty} (\Delta y)_i \sin(\omega \Delta t k_i)$

The gain of the system is then given by

$$M = |\underline{GH}| = (C^2 + B^2)^{\frac{1}{2}} \quad (8)$$

where:  $M$  = the gain, or magnitude ratio, of the output signal to the input signal

$\underline{GH}$  = the transfer function of the system (a vector quantity)

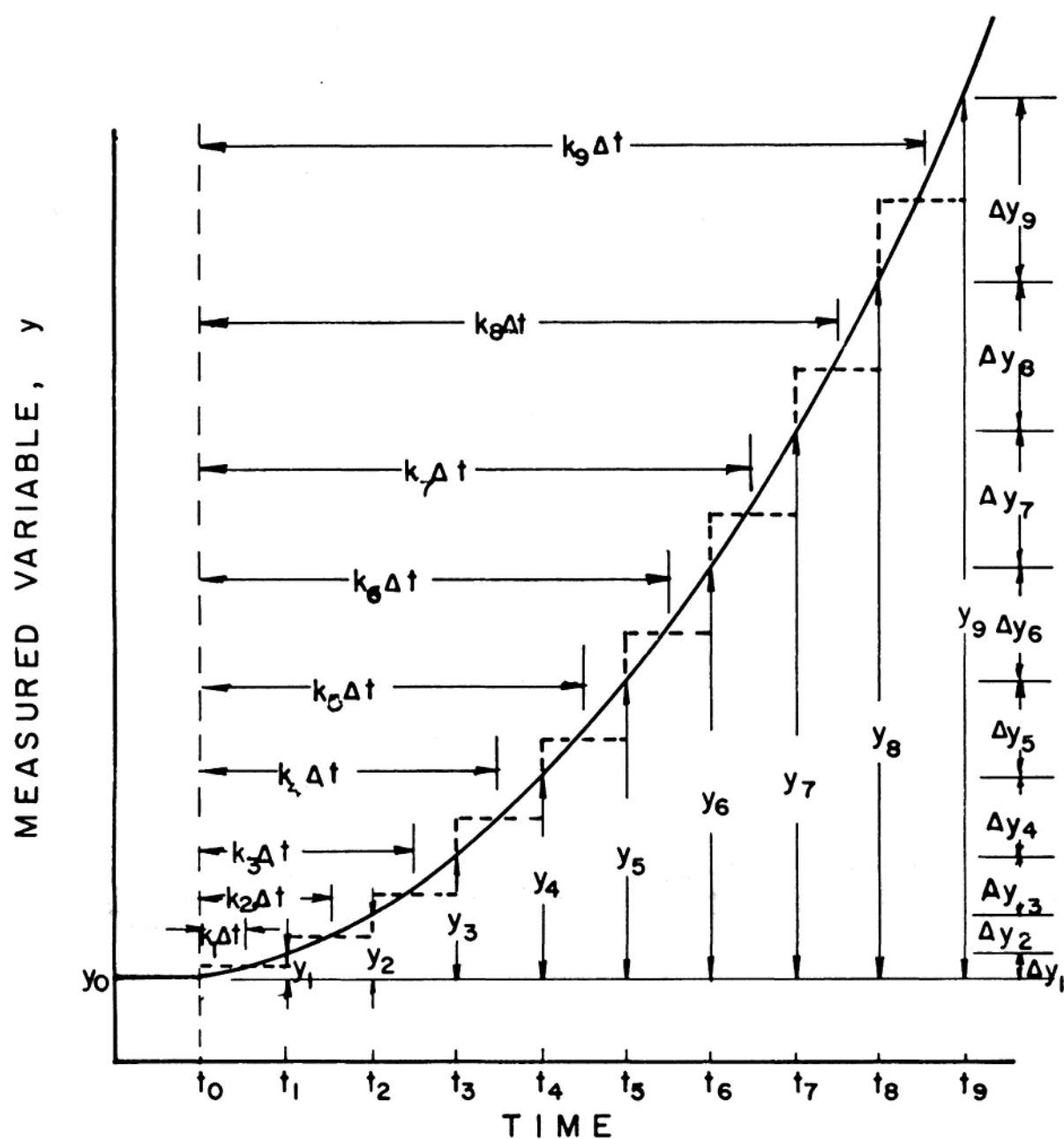


FIGURE 3. APPROXIMATION OF A  
RESPONSE CURVE BY A SERIES  
OF STEP FUNCTIONS

After Caldwell, W.I., Geraldine A. Coon, & L.M. Zoss:  
"Frequency Response for Process Control," p. 373,  
Mc Graw-Hill Book Co., New York, N.Y., 1959.



The phase lag for the system is given by

$$\phi = \text{angle GH} = -\tan^{-1} \left( \frac{B}{C} \right) \quad (9)$$

From the open-loop frequency response characteristics of the system which are determined by the foregoing method, the closed-loop characteristics may be determined mathematically without further experimental data. The closed-loop transfer function of a system is related to its open-loop transfer function by the following equation:

$$M'e^{j\alpha} = \frac{GH}{1+GH} \quad (10)$$

where:  $M'e^{j\alpha}$  = the closed-loop transfer function  
 GH = the open-loop transfer function

The value of the gain, or magnitude ratio, of the closed loop transfer function is given by the expression

$$M' = \left| \frac{GH}{1+GH} \right| = \left| \frac{C - jB}{1+C - jB} \right| = \frac{(C^2 + B^2)^{\frac{1}{2}}}{[(1+C)^2 + B^2]^{\frac{1}{2}}} \quad (11)$$

where: C and B are defined as in equation (7).

$M'$  = Magnitude ratio of closed-loop transfer function

The value of the phase lag for the closed-loop transfer function is defined similarly,

$$\alpha = \text{angle GH} - \text{angle } (1+GH) \quad (12)$$

$$\alpha = \phi - \text{angle } (1+C - jB) \quad (13)$$

$$\alpha = \phi - \tan^{-1} \left( \frac{B}{1+C} \right) \quad (14)$$

Where:  $\alpha$  = closed loop phase angle

$\phi$  = open loop phase angle

Backmixing Model. Levenspiel, <sup>(12)</sup> following Taylor's <sup>(17)</sup> model of eddy diffusivity, considers backmixing as being analogous to molecular diffusion, and presents models describing the axial dispersion. Fick's Law for molecular diffusion is given by the expression

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (15)$$

where; C is concentration,  
x is distance,  
t is time,

and D is the coefficient of molecular diffusion.

The analogous equation representing plug flow with a uniform degree of axial dispersion, or backmixing, is

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (16)$$

where: D is the coefficient of axial or longitudinal dispersion (units are L<sup>2</sup>/t).

C Curve. The tracer input which lends itself most readily to analysis in the determination of the value of D is the pulse input of infinite magnitude and zero duration. The dispersion of the tracer during its passage through the reactor will yield a nearly normal distribution curve as an output, if the degree of backmixing is not great. This form of output is termed the C curve and is illustrated in Figure 4.

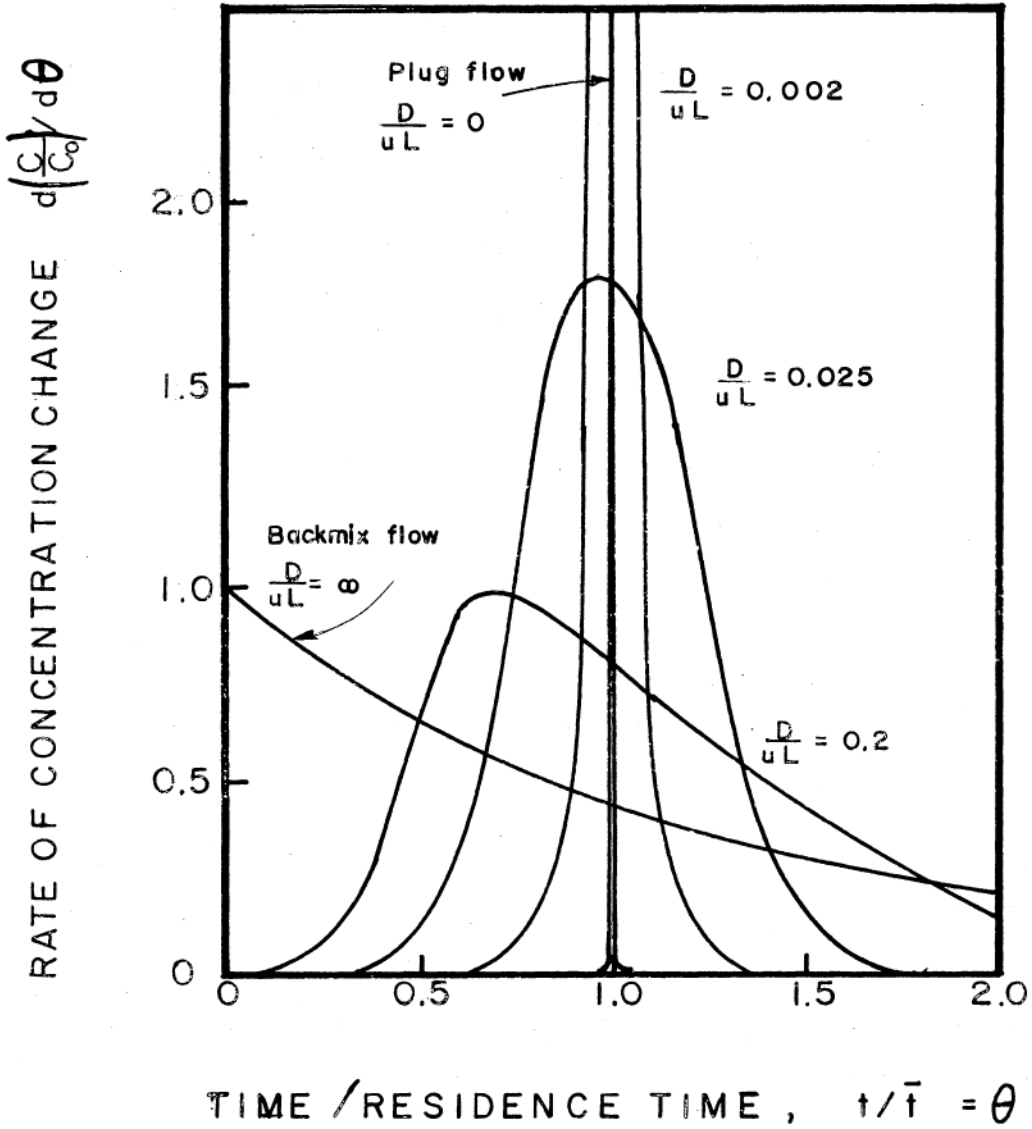


FIGURE 4. "C" CURVES FOR VARIOUS DISPERSION NUMBERS

LEVENSPIEL, O. : "Chemical Reaction Engineering," p.264, John Wiley & Sons, New York, N.Y.

Dispersion Number. The parameter which best describes the dispersion within a reactor is the dimensionless group  $D/uL$  (where  $D$  is the dispersion coefficient,  $u$  is the velocity of the fluid through the reactor, and  $L$  is the length of the reactor under study). This group is termed the reactor dispersion number, and is the reciprocal of the axial Peclet number for mass transfer.

Determination of Dispersion Number. The reactor designed fits the description of a closed vessel (Figure 5) as defined by Levenspiel and Smith<sup>(14)</sup>. They showed the relationship between the variance of the  $C$  curve and the dispersion number to be

$$\sigma^2 = 2 \left( \frac{D}{uL} \right) - 2 \left( \frac{D}{uL} \right)^2 (1 - e^{-uL/D}) \quad (17)$$

Where:  $\sigma^2$  = Variance  
D = Axial Dispersion Coefficient  
u = velocity of fluid  
L = length of vessel studied

The assumptions made for the treatment of closed vessels are that  $D$  is zero at the input and outlet points of the reactor and that  $D$  is a constant throughout the length,  $L$ , of the reactor. A further simplification may be made if the value of the dispersion number is small. The second order and exponential terms may be neglected, and the output function ( $C$  curve) more closely approximates the normal, error, or Gaussian distribution. The variance under these

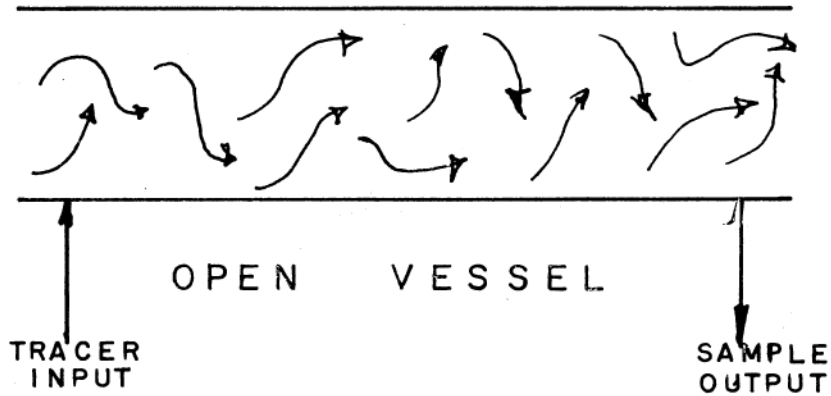
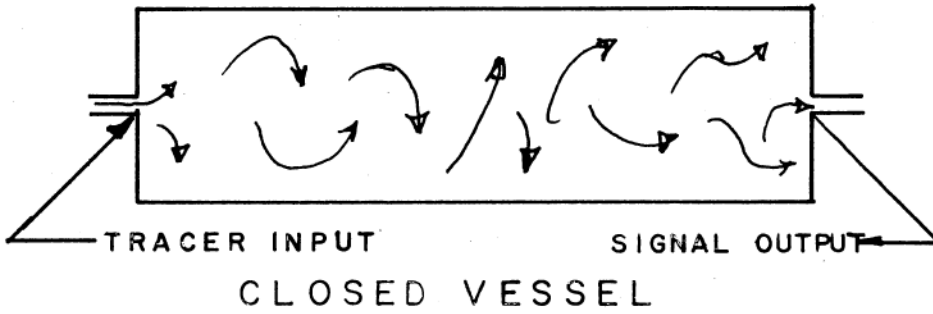


FIGURE 5. CLOSED AND OPEN VESSELS

Levenspiel, O. : "Chemical Reaction Engineering,"  
p. 265, John Wiley & Sons, Inc.,  
New York, N.Y., 1962.

conditions is simply

$$\sigma^2 = 2 \left( \frac{D}{uL} \right) \quad (18)$$

The error involved in this assumption is in the range of five times the value of the dispersion number or:

5 per cent when  $D/uL$  is 0.01

0.5 per cent when  $D/uL$  is 0.001.

Evaluation of Dispersion Number from F Curve. The F curve for a system is the response curve to a step input of unit amplitude. A typical F curve is shown in Figure 6. The C curve is the derivative of the F curve for a given system, and can be calculated from the F curve. If the dispersion number is small (less than about 0.01), the variance of the C curve may be quickly calculated from the points of the F curve. The values of concentration of the F curve are first normalized as percentage points of the concentration gradient involved between initial and final compositions of the sample stream. These values are then plotted on probability paper using time as the independent variable. If the result is a straight line, or approximates a straight line, the time increment intercepted by the straight line representing the F function and the sixteenth and eighty-fourth percentile lines of the probability axis represents two standard deviations. If this time increment is divided by two and then by the holding time of the reactor, the result is a dimensionless standard deviation from which the variance may be calculated directly.

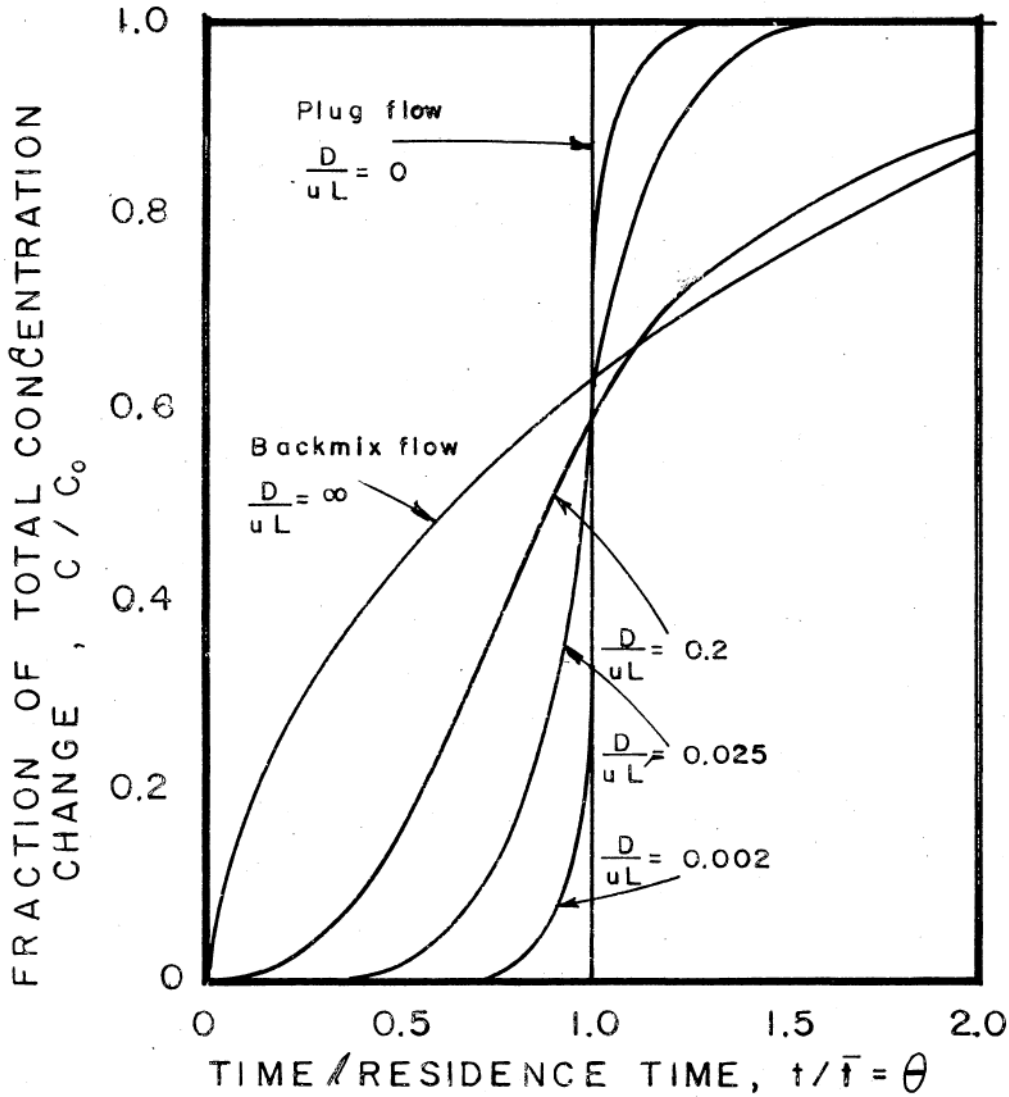


FIGURE 6. "F" CURVES FOR VARIOUS DISPERSION NUMBERS

LEVENSPIEL, O.: "Chemical Reaction Engineering," p. 264, John Wiley & Sons, New York, N.Y., 1962.

$$\sigma^2 = \left( \frac{2\sigma_t}{2\bar{t}} \right)^2 \quad (19)$$

where:  $\bar{t}$  = holding time

The value of the dispersion number may then be calculated by

$$\frac{D}{uL} = \frac{\sigma^2}{2} \quad (20)$$

If the value of the dispersion number so calculated is less than 0.01, this approximation is within five per cent of the actual value, and the method is justified.



### III. EXPERIMENTAL

The following section includes the purpose and plan of the investigation, listings of the materials and apparatus used, the method of procedure, and data collected and results calculated, and sample calculations showing how results were determined.

#### Purpose of Investigation

The purpose of this investigation was to design and construct a tubular flow reactor, capable of handling both liquid and gas phase reactions, both homogeneous and catalyzed, and to demonstrate its ability to produce reliable dynamic response information.

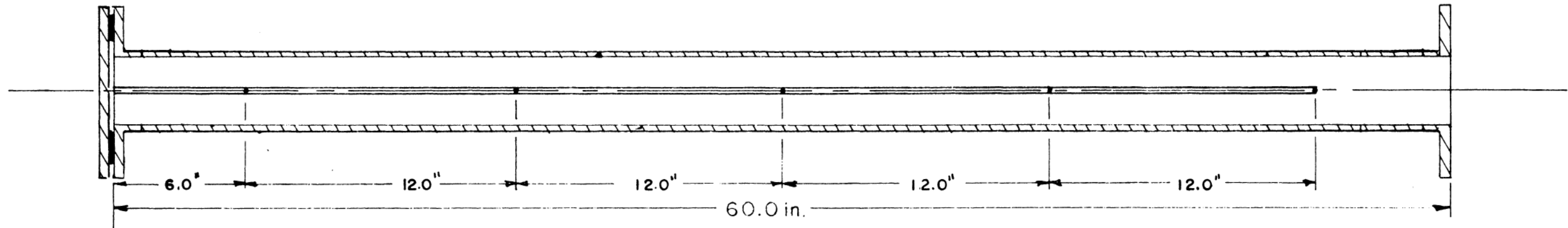
#### Plan of Experimentation

The plan of experimentation was developed in stages. First, a system for reaction was chosen. The reactor was then designed to handle the chosen reaction, but was constructed to be as flexible in its application as was feasible. The analysis system was then chosen, designed, and constructed; again, the flexibility of the equipment was considered.

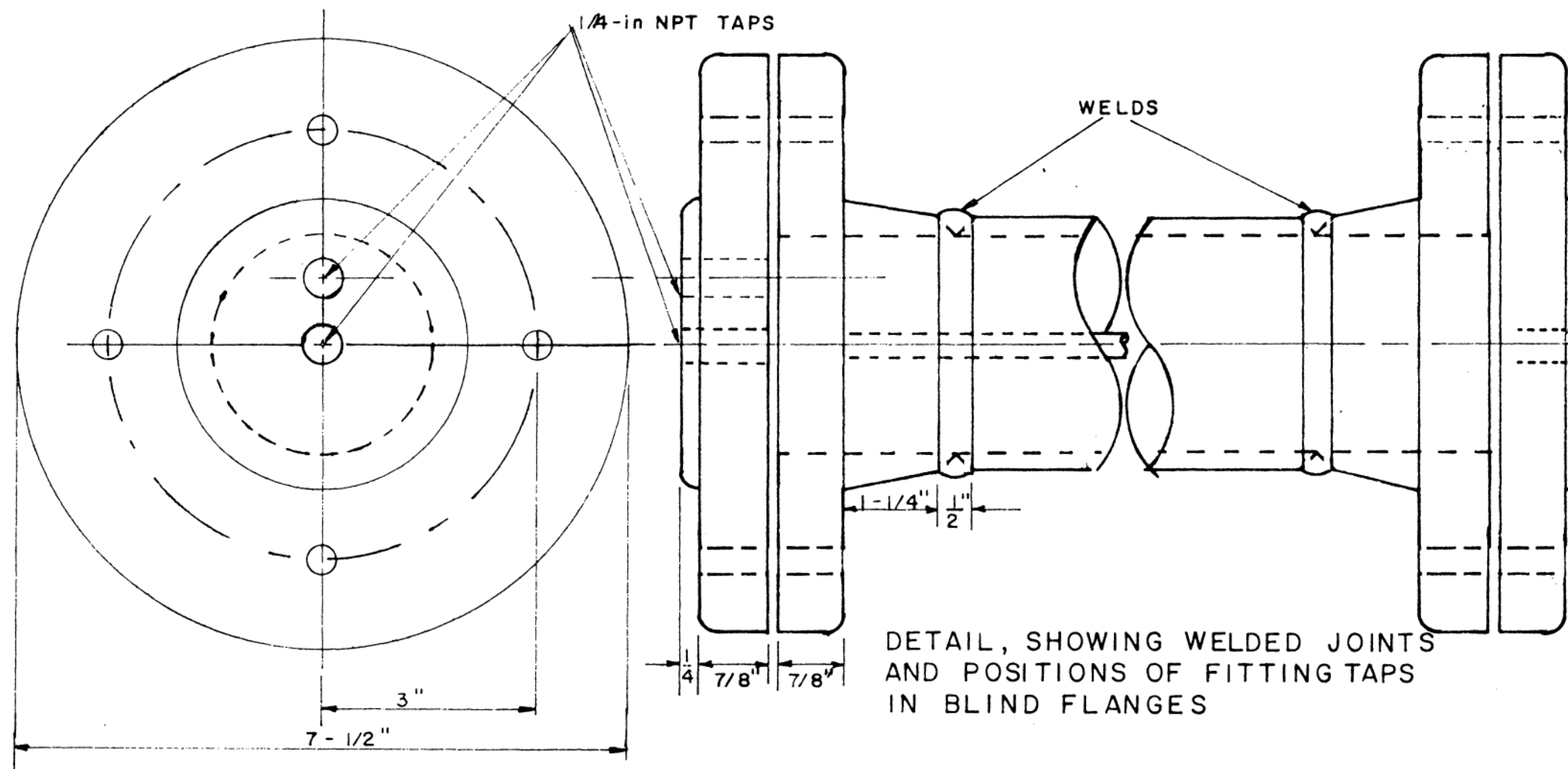
Choice of Reaction System. The system chosen for study was the thermal decomposition of acetaldehyde to methane and carbon monoxide, at temperatures in the range of 300° C. In the preliminary studies made in this thesis for the determination of dynamic response information, the reacting system itself was not actually studied, but rather the mixing of various compositions of carbon monoxide and methane. This system was chosen for several reasons. The reaction system has been widely studied, it is a homogeneous reaction, and its dynamic analysis presents no insurmountable problems.

Reactor Design. The reactor was constructed of type 347 stainless steel. Stainless steel was used to reduce the effects of corrosion under extreme conditions of temperature, and to reduce the possibility of the reactor walls catalyzing an otherwise homogeneous reaction. The reactor was designed to be operated over a range of conditions with regard to temperature and pressure. Design pressure was 150 pounds per square inch, gage, at 70° F., and design temperature was 500° C., at 70 pounds per square inch, gage. Type 347 stainless was used, because welding neck flanges were used on the ends of the reactor to eliminate sources of leakage and to provide easy access to packing or catalyst used in the reactor. The reactor design is shown in Figure 7.

FIGURE 7 Reactor



CUTAWAY VIEW SHOWING POSITIONS OF THERMOCOUPLE JUNCTIONS



DETAIL, SHOWING WELDED JOINTS AND POSITIONS OF FITTING TAPS IN BLIND FLANGES

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# REACTOR

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APPROVED BY:		SHEET NO:

Choice of Analysis System. Since the streams to be analyzed were two component streams, and since carbon monoxide and methane have different thermal conductivities, it was decided to use a thermal conductivity cell for the analysis. The advantages of the thermal conductivity cell over other methods of analysis are its rapid response, and its continuous analysis. Gas chromatography and infrared spectrophotometry were also considered, but thermal conductivity was chosen because of its particular attributes.

Assumptions. Assumptions made during the study were that the gas mixture in the reactor was virtually at a constant temperature from the time it entered the reactor, and that the temperature of the reactor was constant during each test.

Preliminary Studies. The first problems encountered were calibration of equipment and verifying assumptions made. The reactor itself was maintained at constant temperature during the course of the experimentation, as verified by a series of five thermocouples equally spaced within the core of the reactor. Rotameters used for the measurement of the flowrates of the gas streams were supplied by the manufacturer with calibration curves, from which the flow rates of the individual gases could be calculated. The calibration curves supplied were checked by means of a wet-test meter.

Dynamic Studies. Carbon Monoxide was used as the dispersed phase and methane as the continuous phase in the

dynamic studies. From transient data recorded after the introduction of step changes in the composition of the feed stream, dispersion coefficients were determined as functions of Reynolds number, and frequency response data were calculated from the same transient data.

### Materials

The following section is a listing of the materials used in the study of dynamic response of a packed, tubular reactor.

Air. Compressed air from the V.P.I. air lines, delivery pressure 80 psig, used for the calibration of the gas rotameters.

Carbon Monoxide. One size 1-A cylinder of commercial grade carbon monoxide, minimum purity 98.0 per cent. Obtained from the Matheson Company, East Rutherford, New Jersey. Used as a component in the feed to the reactor.

Glycol. Commercial grade ethylene glycol, minimum purity 97 per cent, manufactured by Baker Chemical, New York, N.Y. Used at its boiling point as a temperature standard for the calibration of the thermocouples.

Insulation, Asbestos Cement. 42 square feet of 1/4-inch asbestos cement sheet. Used as an insulating material in the reactor case and the analysis box.

Insulation, Glass Wool. 36 square feet of 6-inch glass wool batting, Manufactured by Johns-Manville Company, New York. Used as insulation in the reactor case.

Methane. One size 1-A cylinder of technical grade methane, minimum purity 95.0 per cent. Obtained from The Matheson Company, East Rutherford, New Jersey. Used as a component in the feed to the reactor.

Nitrogen. One size 1-A cylinder of commercial grade, water pumped nitrogen, minimum purity 99.0 per cent, obtained from the Southern Oxygen Company, Washington, D.C. Used as a reference gas in the thermal conductivity cell.

Packing. Crushed quartz, obtained as 1-2 mesh from the V.P.I. Geology Department, and reduced to 8-14 mesh in the jaw crusher located in the Unit Operations Laboratory of the V.P.I. Chemical Engineering Department. Used as packing in the reactor.

Steam. Steam from the V.P.I. Steam Plant. Delivery pressure 80 psig. Used as the heating medium in the feed preheater.

Water, Distilled. Distilled water obtained from the still located in the Chemical Engineering Department. Used as a primary standard for temperature at freezing and boiling conditions for the calibration of the thermocouples.

Water, Tap. Tap water obtained from the V.P.I. water supply. Used for making ice used in the ice bath to cool distilled water as a temperature standard for the calibration of thermocouples. Also used in connection with the steam system of the apparatus to condense steam escaping with condensate.

### Apparatus

The following section is a listing of the apparatus used in the investigation of the dynamic response of a tubular packed reactor.

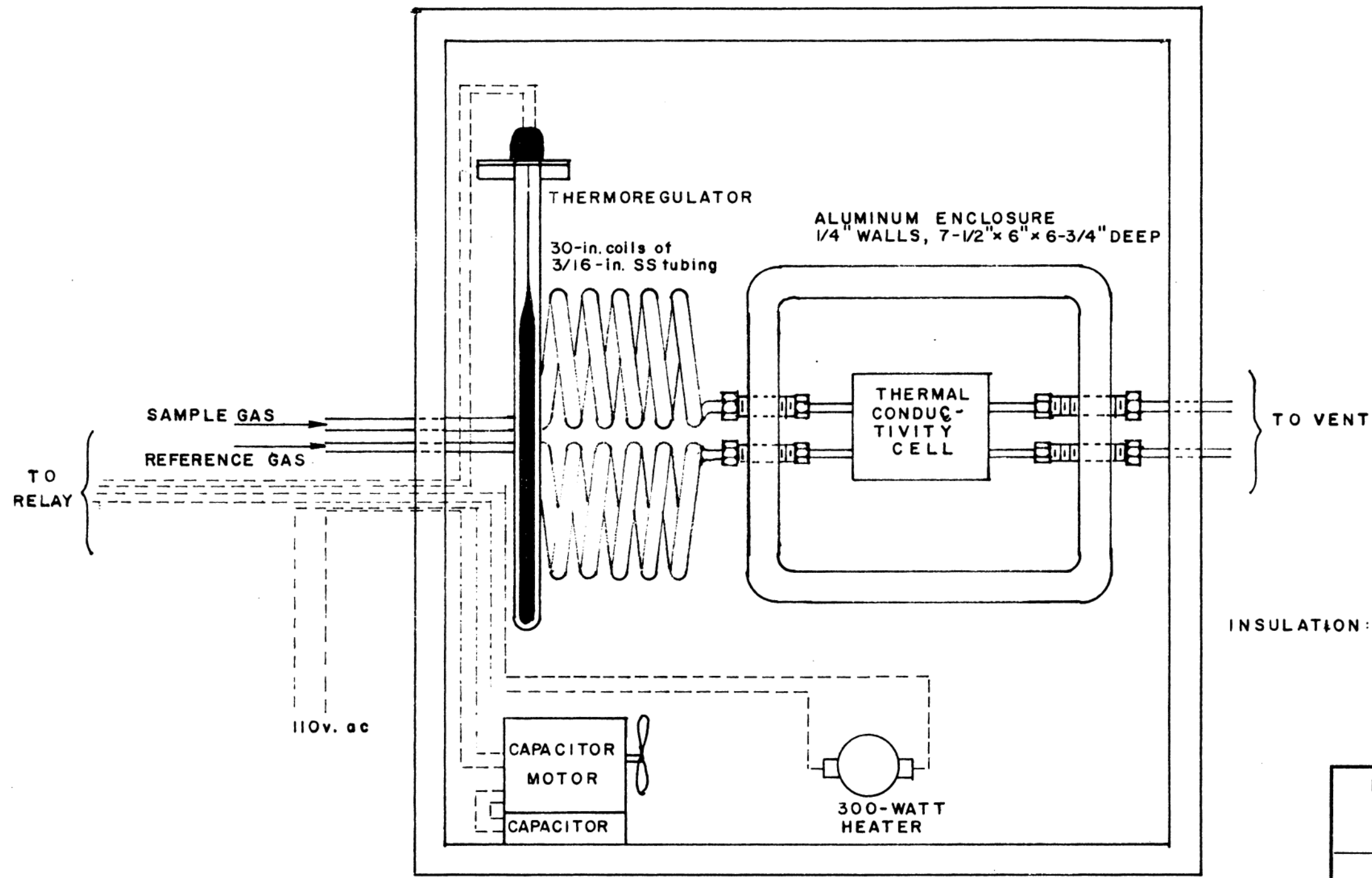
Ammeter. A direct current milliammeter, model B095, catalog number 50-171111 KAKAL, range 0-200 milliamperes. Manufactured by General Electric, Lynn, Mass. Used to indicate the current flow to the thermal conductivity cell circuit from the stabilized direct current power supply.

Bath, Air. A stirred, controlled-temperature air bath was fabricated from a used instrument case, as shown in Figure 8. It was used to contain the analysis system, and was maintained at a temperature of 80°C.

Cell, Thermal Conductivity. A thermal conductivity cell, model 9285 (Pretzel), stainless steel body, hot wire type, tungsten filament. Manufactured by Gow-Mac Instrument Co., Madison, New Jersey. Used in the analysis of the product stream from the reactor.

Electrical System. Electricity was supplied as three-phase alternating current at 110 volts line-to-ground and 220 volts line-to-line. The heater circuit at 220 volts shown in Figure 9, p. 31, was composed of 14 gauge copper wire, and utilized six double pole single throw switches in conjunction with variable transformers to control the heaters individually. The individual heater circuits were fused at ten amperes, and the entire electrical system was





INSULATION: 1/2-in. "Celotex"  
 Fiber Board - all surfaces  
 1/4-in. Asbestos-cement  
 Sheet - bottom only

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## ANALYSIS SYSTEM

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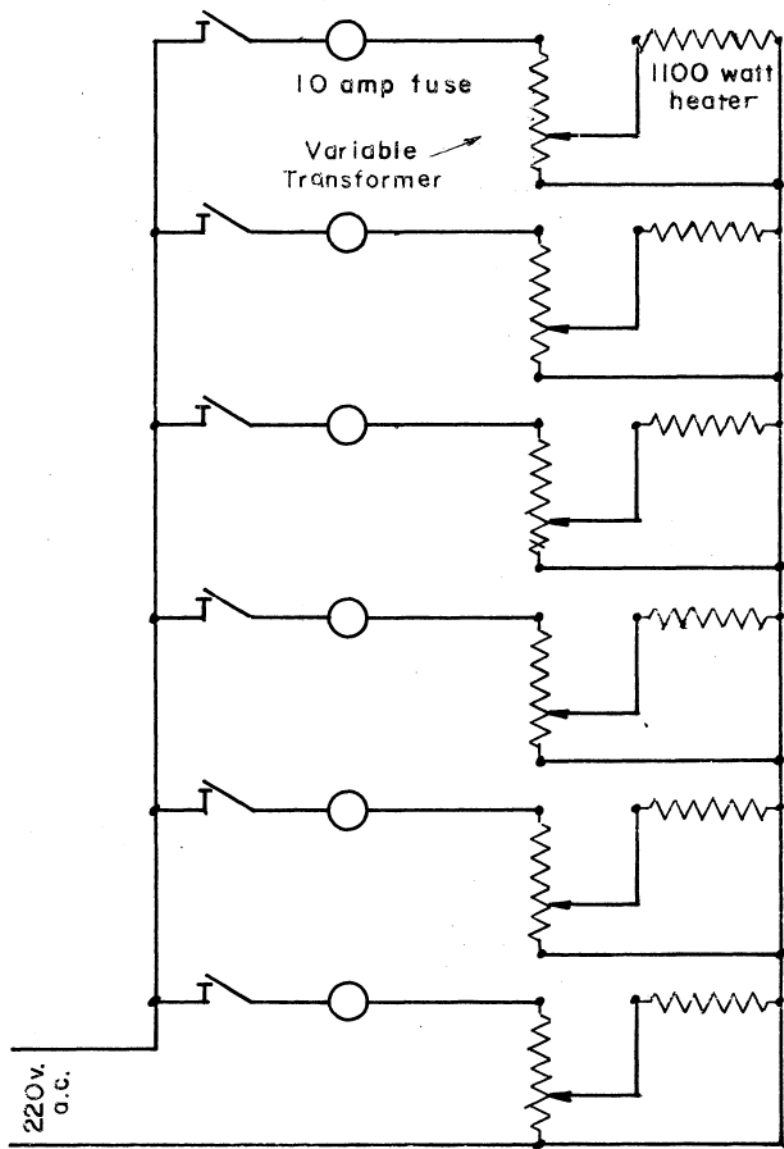


FIGURE 9. SCHEMATIC DIAGRAM OF REACTOR HEATING CIRCUIT

fused at 30 amperes per line. The remainder of the electrical equipment obtained its electrical energy as 110 volts, ac. This was converted into the desired direct current voltages by means of power supplies, in the cases where direct current was required.

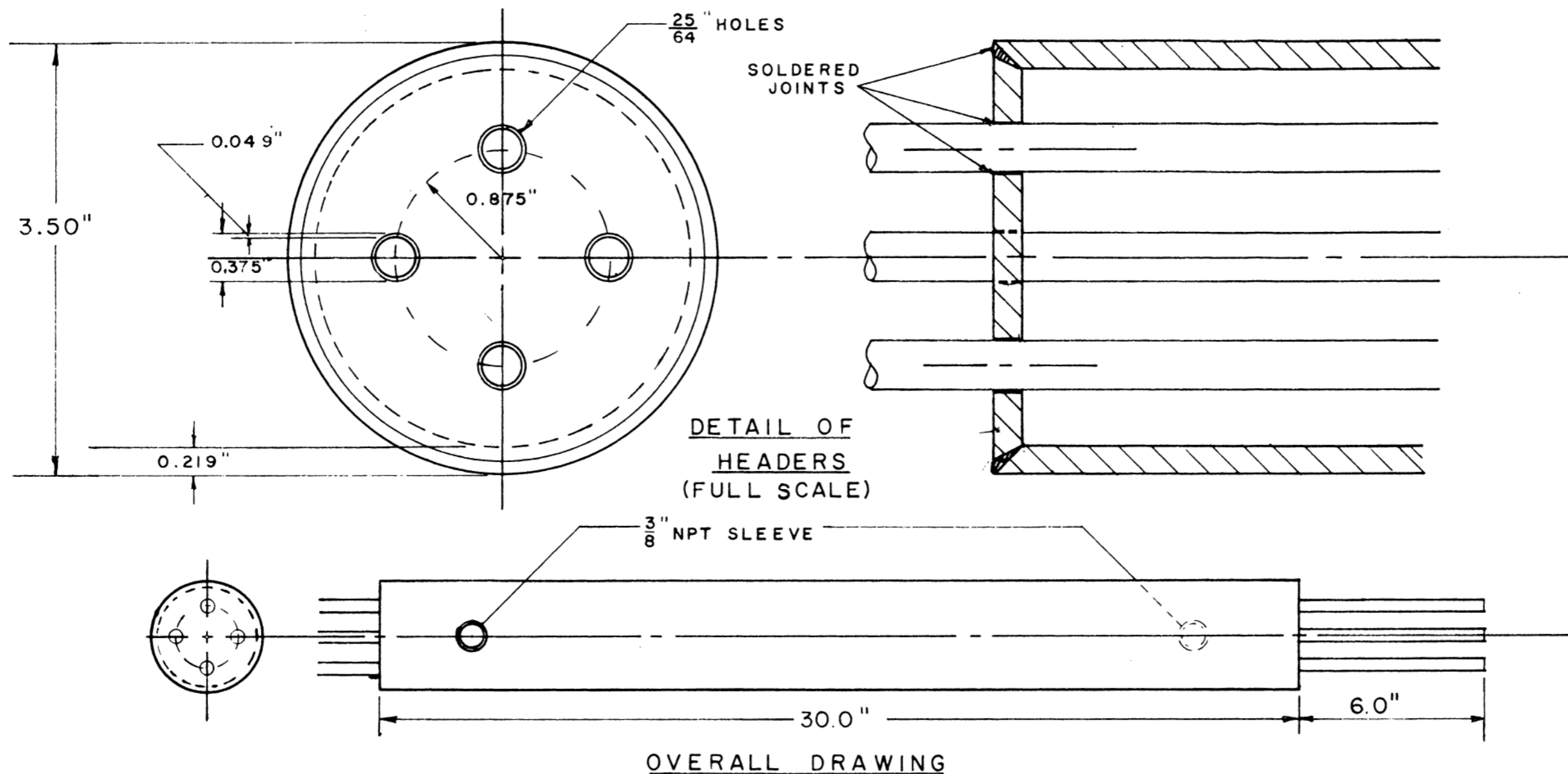
Galvanometer. A galvanometer, catalog number 570-202 range 0-60 mv, manufactured by G-M Laboratories, Chicago. Used in conjunction with the Student potentiometer in the calibration of thermocouples and the temperature recorder.

Heat Exchanger. A Shell-and-tube heat exchanger constructed in the Chemical Engineering Shop, described in Figure 10. Consisted basically of four separate 3/8-inch 316 stainless steel tubes within a 3-inch copper pipe shell. Used to heat four separate streams, either feed or product, steam heated.

Heaters, Cal-rod. Six heaters, cal-rod type, high-temperature nickel-stainless alloy cladding. Power rating was 1000 watts at 220 volts, temperature rating 1400° F., continuous duty. Manufactured by Edwin L. Wiegand, Pittsburgh, Penna. Used to supply heat to the reactor.

Meter, Wet Test. A wet test meter, range 0.250 cubic feet per revolution, indicates to 100,000 cubic feet. Manufactured by Precision Scientific Company, Chicago, Ill. Used for calibrating rotameters.

Potentiometers. Four 3-watt potentiometers, single-turn, with the following catalog numbers, model numbers, and resistances:



MATERIALS OF CONSTRUCTION

SHELL - 3-IN. COPPER PIPE

TUBES - 3/8-IN. TYPE 316 STAINLESS STEEL

JOINTS - SILVER SOLDER

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FEED PREHEATER

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No.	<u>Catalog Number</u>	<u>Model Number</u>	<u>Resistance</u>
2	140-6206	58C1-100	100 ohms
1	5936	58C1- 30	30 ohms
1	141-6148	58C1- 1	1 ohm

Manufactured by the Clarostat Manufacturing Company, Dover, New Hampshire. Used to adjust the sensitivity of the potential recorder, balance the resistances of the bridge of the thermal conductivity cell, control the bridge current, and to control the output of the unstabilized power supply.

Potentiometer, Student. A Fisher Student Potentiometer, type S., manufactured by Fisher Scientific Company. Used to calibrate thermocouples and the temperature recorder.

Power Supply, Stabilized. The power supply used to provide the bridge current of the analysis circuit was a model QM 6.3-0.64 Sorensen Power Supply. Input voltage and current ratings were 115 volts and 0.15 amperes, respectively, with a frequency range of 50 to 60 cycles per second. The output was a stabilized d.c. voltage of up to 6.3 volts with a maximum ripple of 1.0 millivolt at currents up to 0.64 amperes. Manufactured by Sorensen, New York.

Power Supply. A power supply to provide 26 volts d.c. for the relay in the air bath temperature control circuit was constructed as a full wave, bridge-type selenium diode rectifier circuit. A diagram of the circuit of the power supply is shown in Figure 11.

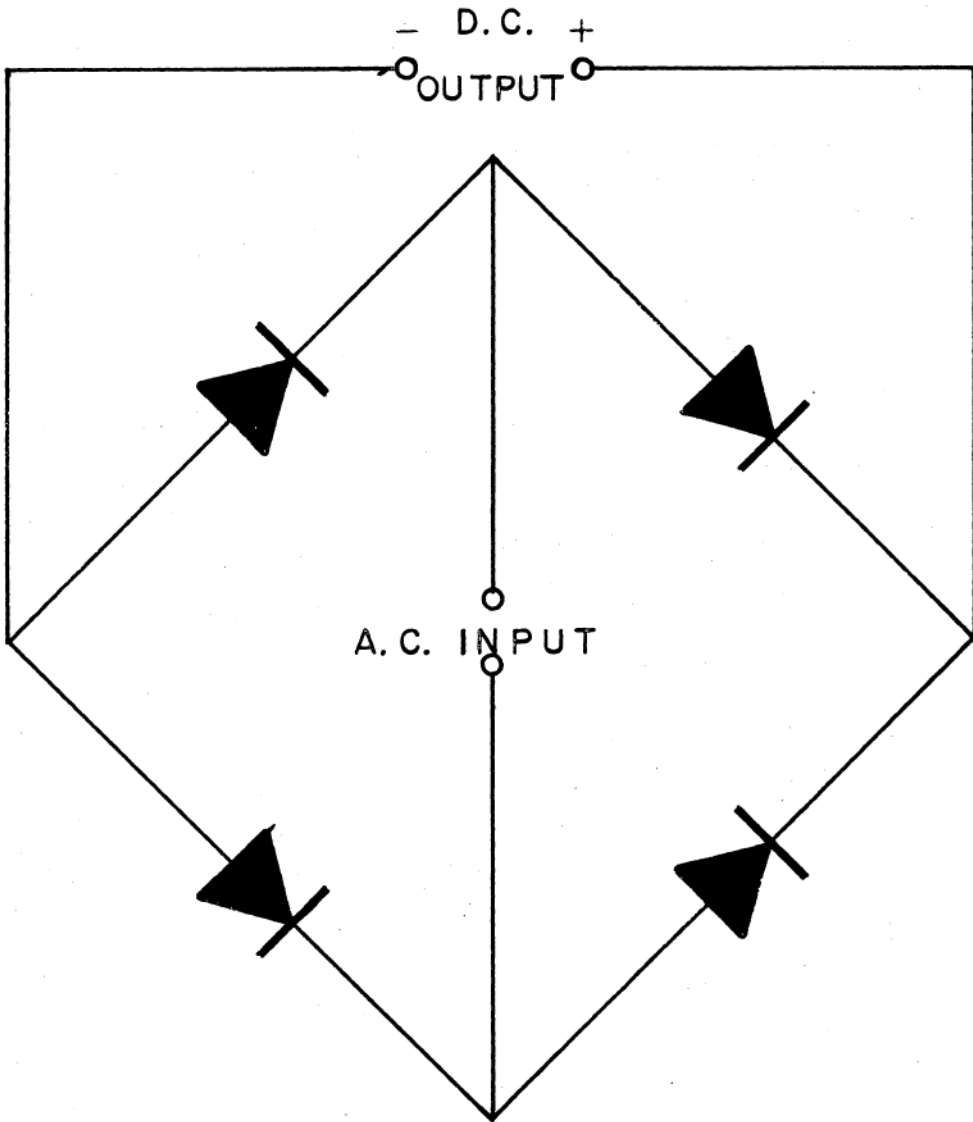


FIGURE II. FULL-WAVE DIODE BRIDGE RECTIFIER CIRCUIT

Pipe and Fittings, Steel. 3/8-inch Sch 40, and 1/2-inch Sch 40 obtained from Chemical Engineering Department, V.P.I. Used for water and steam lines.

Tubing, Polyethylene. 3/8-inch, obtained from Chemical Engineering Department, V.P.I. Used for gas lines.

Tubing, Stainless Steel. 3/8-inch, 0.049-in. wall; 1/4-inch, 0.035-in. wall; 3/16-inch, 0.025-in. wall; and assorted fittings. Obtained from Whitehead Metals Company, Baltimore, Md. Used for gas lines.

Reactor. A tubular flow reactor was constructed of type 347 stainless steel. The reactor consisted of a five-foot length of 3-inch schedule 40 pipe, fitted with welding-neck flanges on both ends. The ends were closed with blind flanges, which were drilled and tapped to accommodate the desired fittings for feed and outlet streams. Steel supplied by Whitehead Metals Company, Baltimore, Md. Reactor described in detail in Figure 7, p. 24.

Recorder, Temperature. A six-point Micromax temperature recorder, range 0 - 500° C. Manufactured by Leeds and Northrup, Philadelphia, Pa. Used to record the reactor temperature at points along the axis of the reactor.

Recorder, Voltage. A potential recorder, range 1 - 10 millivolts, model G-10. Manufactured by Varian Associates, Palo Alto, California. Used to measure the signal from the thermal conductivity cell circuit.

Regulators. Three two-stage regulators, catalog number 6, first stage 0-3000 psig, second stage 0-100 psig. Manufactured by the Matheson Company, East Rutherford, New Jersey. Used to regulate the flow of gases used for feed and reference gas for the thermal conductivity cell.

Relay. An electrical relay, Model KCP11, plate circuit 5000 ohms, activating signal 26 volts d.c. Manufactured by Potter and Brumfield, Princeton, Indiana. Used to control the current flow to the heater within the air bath, activated by the thermoregulator.

Rotameter. One rotameter, model WS/1157/6 with tube number 02-F-1/8-12-5/70. Manufactured by Fischer-Porter Co.; used to meter the flow of Nitrogen used as a reference gas in the analysis cell.

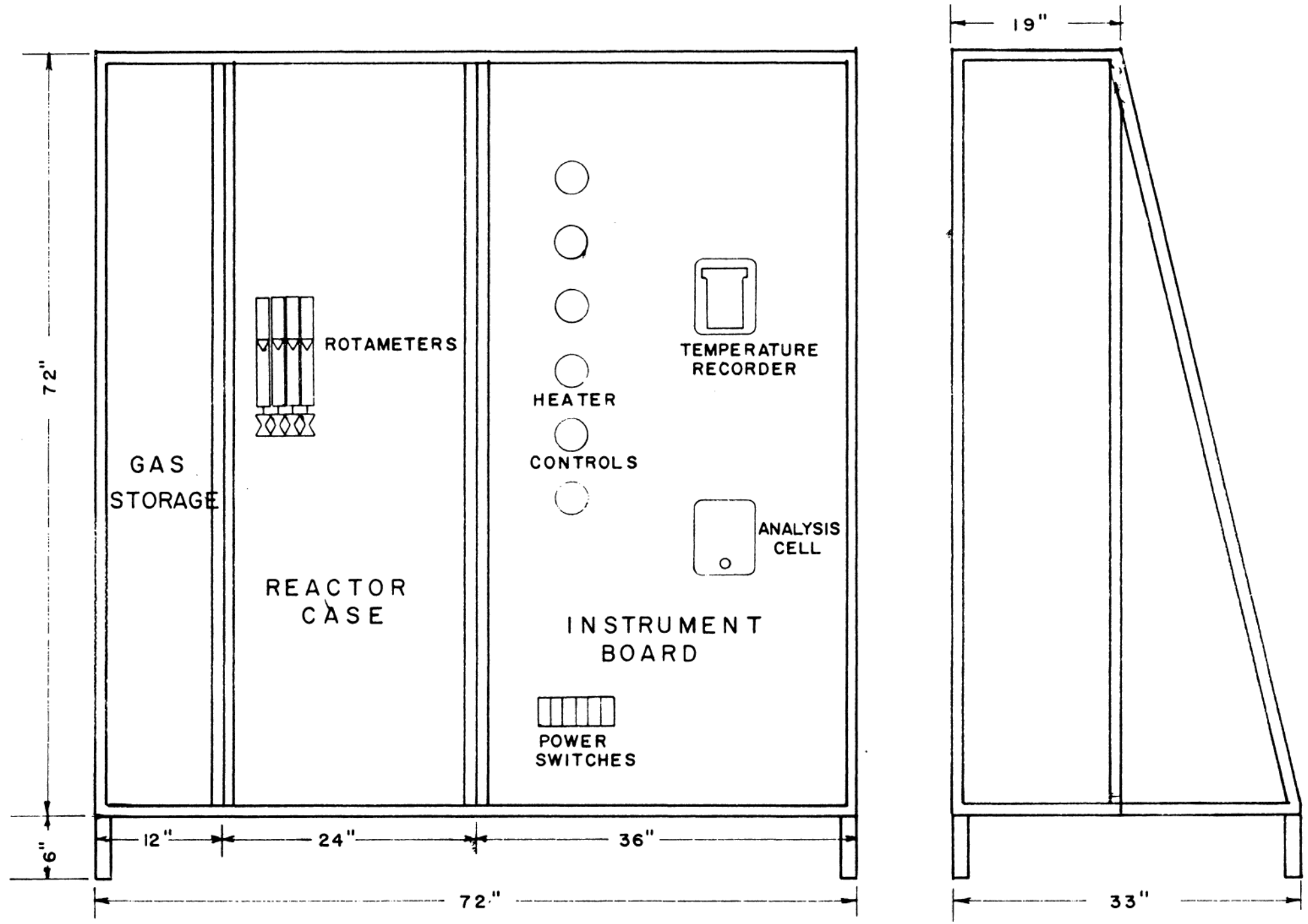
Rotameters. Three model 625P - 802 rotameters, manufactured by the Matheson Company, East Rutherford, New Jersey. Used to meter the flows of the components of the feed and the sample portion of the product stream.

Stand, Equipment. An equipment stand was constructed of 1-inch angle iron, 1-1/4 inch pipe, and 3/4-inch Fir plywood. The construction details are given in Figure 12. Used to support the equipment used in the investigation.

Standard Cell. A Fisher "EPLAB" student standard cell was used to calibrate the student potentiometer before and during use in the calibration of thermocouples and the temperature recorder. Manufactured by Fisher Scientific, Pittsburgh, Pa.



FIGURE 12 Equipment Stand



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**EQUIPMENT  
 STAND**

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Thermocouples. Thermocouples were made up of 18 gauge iron and constantan wire and calibrated individually. Used to measure the temperature of the reactor along its axis.

Thermoregulator. A thermoregulator, mercury in glass, adjustable set point. Range up to 400°F. Model 15-182-5. Manufactured by Fisher Scientific Company, Chicago, Ill. Used to control the temperature of the air bath containing the thermal conductivity cell circuit.

Timer. A Precision "Time-It" timer, range 0 - 10,000.0 seconds in 0.1 second increments. 115 volts, 60 cycles, 5 watts. Manufactured by Precision Scientific Company, Pittsburgh, Pa. Used to time volume measurements on the wet test meter during the calibration of the gas rotameters.

Transformers. Six variable transformers, catalog number VTSN, rated at 10.0 amperes at 120 volts, single phase alternating current. Manufactured by Ohmite, Skokie, Ill., and supplied through Allied Electronics. Used to control the voltage to the reactor heaters.

### Method of Procedure

The following section describes the procedures used in the investigation, and covers design, construction, and testing of the apparatus.

Design Considerations. The reactor was designed to be as flexible as possible with respect to reaction systems. The factors of volume, temperature and pressure of the reaction, corrosion, adaptibility to both gaseous and liquid systems, and provision for both homogeneous and catalytic reactions were considered. The main body of the reactor was constructed of type 347 stainless steel, basically a five foot length of three-inch schedule 40 pipe, fitted with welding neck flanges. Blind flanges fitted with the necessary tubing connectors completed the reactor proper. A thermowell equipped with five thermocouples spaced at one foot intervals starting six inches from the bottom of the column provided a measurement of the temperature distribution through the center of the column. Heat was provided by six calrod-type heaters wrapped around the column, and the entire assembly was well insulated with rock wool insulation at least six inches thick. Safe operating conditions for the column ranged from 150 pounds per square inch, gage, at a temperature of 70°F to 70 pounds per square inch, gage, at temperatures in the range of 500° C.

A feed preheater provided the capability of preheating three separate feed streams to a temperature of up to 160°C. This same heat exchanger was also used to partially cool the product stream prior to analysis. The feed preheater was a shell-and-tube heat exchanger using steam at pressures up to 50 pounds per square inch, gage, on the shell side and having four separate stainless steel tubes through which the various feed and product streams were passed for heating and cooling, respectively.

Calibration of Instruments. The calibration curves supplied with the Matheson rotameters were checked against measurements made with a wet test meter. Air was allowed to flow through the rotameters at various flow rates as indicated by the position of the rotameter float. For each rotameter setting, the volume of air passed during a measured time period was measured with the wet test meter.

The thermocouples used to measure the reactor temperature were calibrated at 0° C (ice bath), 98° C (boiling water bath) and 195° C (boiling ethylene glycol bath.) The calibration curves were extrapolated into the region studied. The temperature recorder was found to have a constant error, reading approximately five degrees below the actual temperature, when equipped with the thermocouples used. The adjustment made on the temperature reading was to simply add five degrees, C. to the indicated reading.

Controlled Conditions. The reactor was maintained at a constant temperature of 333° C., during the entire test period. The pressure also remained constant at 707 mm of mercury, absolute.

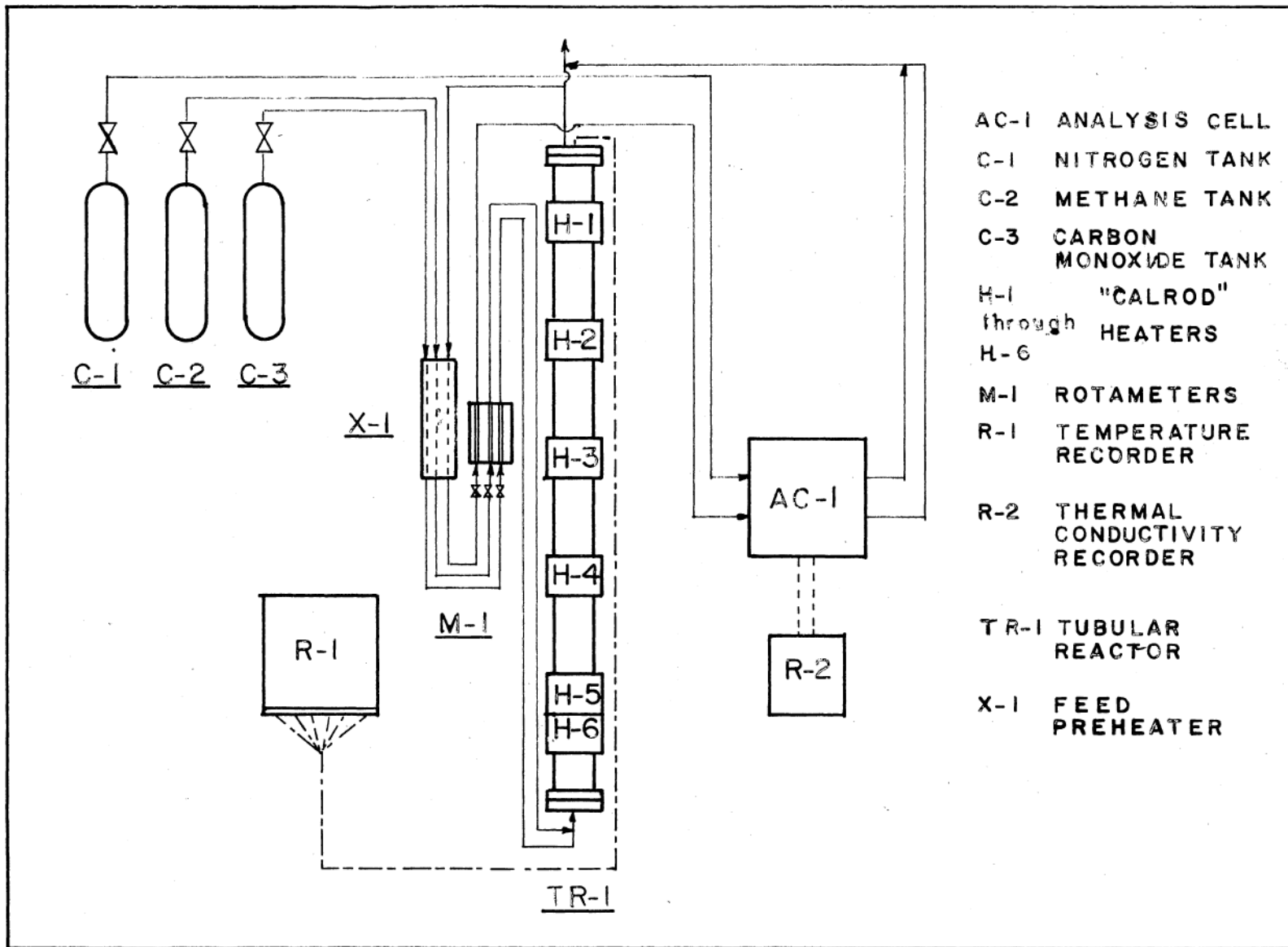
Variables Studied. The feed composition and flow rate were the variables under study. The feed was composed of mixtures of carbon monoxide and methane, and the composition range was from zero to 80 per cent carbon monoxide. The range of flow rates was from 0.458 to 2.190 cubic feet of gas measured at 54° C., and 707 millimeters of mercury, absolute.

The volume of each gas was metered prior to their being mixed at the entrance to the reactor, and from these data the flow rates and compositions of the feed stream were calculated. The composition of the product stream was continuously monitored by the thermal conductivity cell circuit. A measured sample was allowed to pass through the analysis circuit.

An overall flow diagram for the reactor system is given in Figure 13.

Calibration of the Analysis System. For all flow rates studied, it was observed that the magnitude of the output step was a function of composition change only, and seemed to be independent of the flow rate of gas through the analysis cell. Based on this observation, a correlation

FIGURE 13. OVERALL FLOW DIAGRAM FOR  
REACTOR SYSTEM



between composition change and potential change of the output signal in terms of scale units was made.

### Analysis System

The thermal conductivity of the product gas stream was chosen as the measured variable for analysis. Use of a thermal conductivity cell in conjunction with a potential recorder allowed a constant analysis of the product stream, and provided a record of the bridge potential as a function of time.

The bridge circuit used is shown in Figure 14. Nitrogen was used as a reference gas, and a stabilized power supply provided a constant voltage source.

In order to provide conditions of constant temperature, the analysis cell was enclosed in a heavy aluminum box which in turn was enclosed in a cabinet with a thermostatic temperature controller. Air in the cabinet was heated by a small screw-base heater. The air was kept stirred by means of a capacitor-motor driven fan, and the heater circuit was controlled by a mercury-in-glass thermoregulator. The thermoregulator was rated to provide a maximum fluctuation of 0.05 degree, C. from the set-point.

Both the reference gas and the product gas were heated to the air bath temperature by passing them through 30-inch coils of 3/16 inch tubing prior to analysis in the thermal conductivity cell.

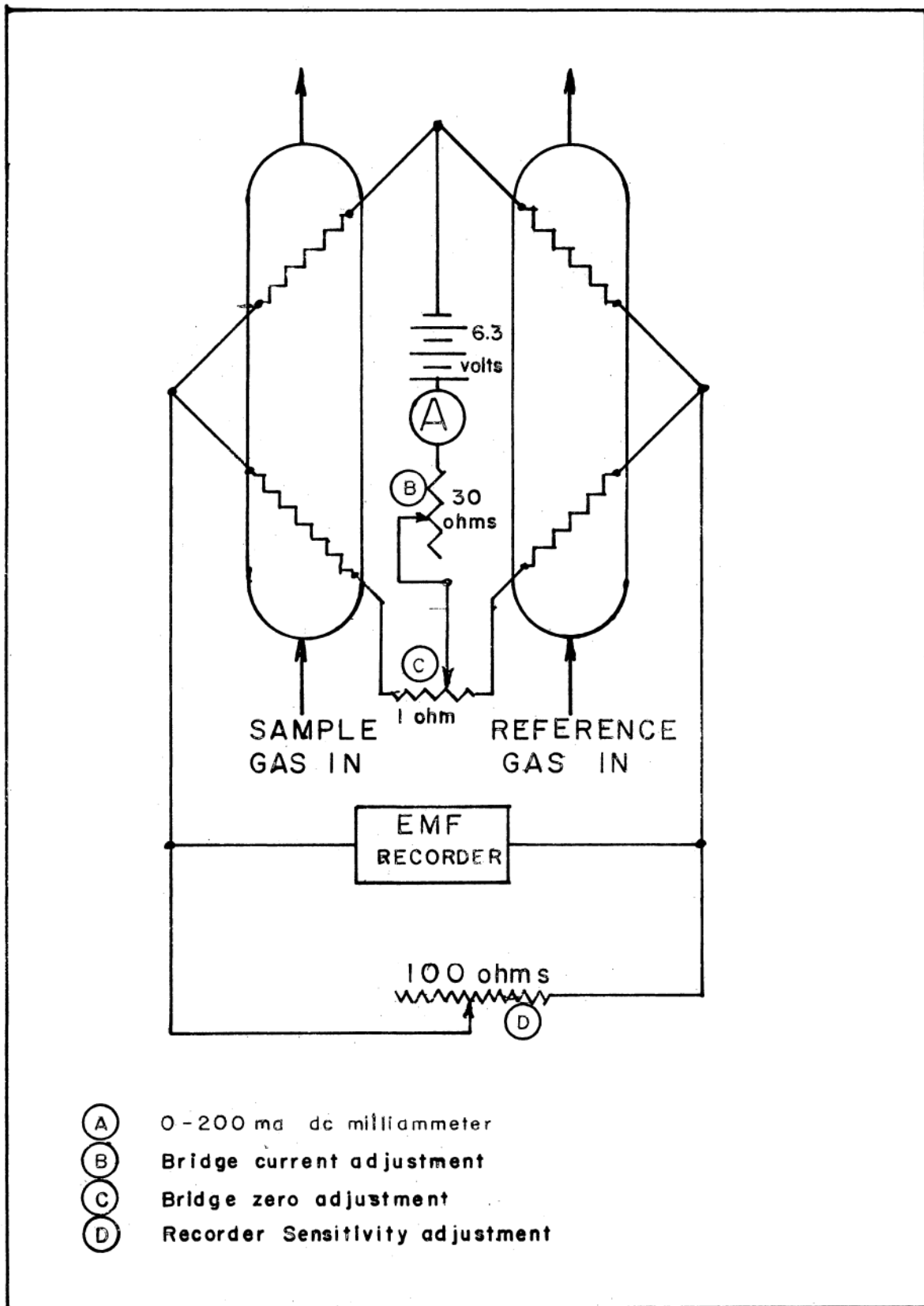


FIGURE 14. SCHEMATIC DIAGRAM OF ANALYSIS CIRCUIT



Step Change Analysis. The actual variable studied was the transient response of the system to a step change in composition of the feed stream at a constant flow rate. This function was produced by simply adding a given flow rate of one gas to given flow rate of the other gas. One gas was allowed to flow through the reactor and the analysis system, until the analysis system had reached a steady reading. The flow rate of this base gas was recorded and maintained constant throughout the run. The second gas was added to the first, and its flow rate quickly adjusted to the value which would produce the desired total flow rate. The recorder measuring the output from the thermal conductivity cell reacted to the advance in flow rate by transposing the base line being traced by a small degree, thus giving a visual record of when the step change was introduced. The run was allowed to continue until no further change was observed in the reading of the output from the thermal conductivity cell.

The output curves were used for the determination of the frequency response of the reactor, and for the calculation of the dispersion numbers for the reactor as functions of flow parameters.

### Data and Results

The following section is a listing of the data observed and the results calculated for the investigation of the dynamic response characteristics of a tubular packed reactor.

Rotameter Calibration. The gas rotameters metering methane and carbon monoxide were calibrated using air at 76° F., and 707 millimeters of mercury, absolute, pressure. Since the variations from the chart supplied with the instruments were less than two per cent in all cases and reasonably within the range of the accuracy of chart reading, the calibration from the chart was used for the calculation of calibration curves for the metering of carbon monoxide and methane. A simple density correction was applied in the calculation. The check of the calibration curve supplied against measurements made using a wet-test meter appears in Table I, and the corrected calibration curves for the metering of methane and carbon monoxide appear in tabular form as Table II, p. 49, and in graphical form as Figure 15, p. 50.

### Correlation of Concentration Change and Output Signal.

The output signal from the thermal conductivity cell was a potential which was not a linear function of the concentration change which it represented. It was therefore necessary to develop a correlation between the output potential and the concentration change causing it and

TABLE I.

CALIBRATION OF GAS ROTAMETERS WITH AIR

ROTAMETER READING	TIME OF TEST	WET TEST METER READING		VOLUME OF AIR PASSED	MEASURED FLOW RATE	FLOW RATE FROM CHART *	PER CENT DIFFERENCE
		START	FINISH				
CM	MINUTES	CU FT	CU FT	CU FT	CU FT/HR	CU FT/HR	PER CENT
1.0	40.61	0.0000	0.0650	0.0650	0.096	0.095	1.56
2.0	32.78	0.0685	0.2040	0.1355	0.248	0.244	1.64
3.0	28.45	0.2100	0.4250	0.2150	0.453	0.449	0.88
4.0	22.16	0.4300	0.6765	0.2465	0.668	0.673	0.75
5.0	20.02	0.7100	1.0015	0.2915	0.873	0.876	0.34
6.0	18.97	0.0200	0.3525	0.3325	1.052	1.047	0.47
7.0	15.25	0.3800	0.6815	0.3015	1.187	1.183	0.34
8.0	15.22	0.7000	1.0370	0.3370	1.327	1.322	0.38
9.0	15.10	0.0800	0.4415	0.3615	1.435	1.439	0.28
10.0	15.60	0.4700	0.8705	0.4005	1.540	1.543	0.20
11.0	15.33	0.9000	1.3170	0.4170	1.630	1.641	0.68
12.0	15.22	0.3400	0.7825	0.4425	1.742	1.733	0.52
13.0	10.31	0.8000	1.1110	0.3110	1.809	1.821	0.66
14.0	10.42	0.1500	0.4770	0.3270	1.880	1.894	0.75
15.0	10.56	0.4900	0.8375	0.3475	1.972	1.956	0.81

TEMPERATURE = 76 DEGREES, F.

PRESSURE = 707 MM MERCURY, ABSOLUTE

\* CALIBRATION CHART SUPPLIED WITH ROTAMETERS BY MATHESON CORP.,  
EAST RUTHERFORD, NEW JERSEY

TABLE II.

CORRECTED CALIBRATION OF GAS ROTAMETERS  
FOR CARBON MONOXIDE AND METHANE

ROTAMETER READING	FLOW RATE OF CARBON MONOXIDE	FLOW RATE OF METHANE
CM	CU FT/HR	CU FT/HR
1.0	0.089	0.067
2.0	0.230	0.174
3.0	0.423	0.320
4.0	0.633	0.480
5.0	0.825	0.625
6.0	0.986	0.747
7.0	1.115	0.844
8.0	1.246	0.943
9.0	1.353	1.025
10.0	1.453	1.102
11.0	1.546	1.171
12.0	1.632	1.237
13.0	1.714	1.290
14.0	1.783	1.341
15.0	1.840	1.407

TEMPERATURE = 54 DEGREES, C.

PRESSURE = 707 MM MERCURY, ABSOLUTE

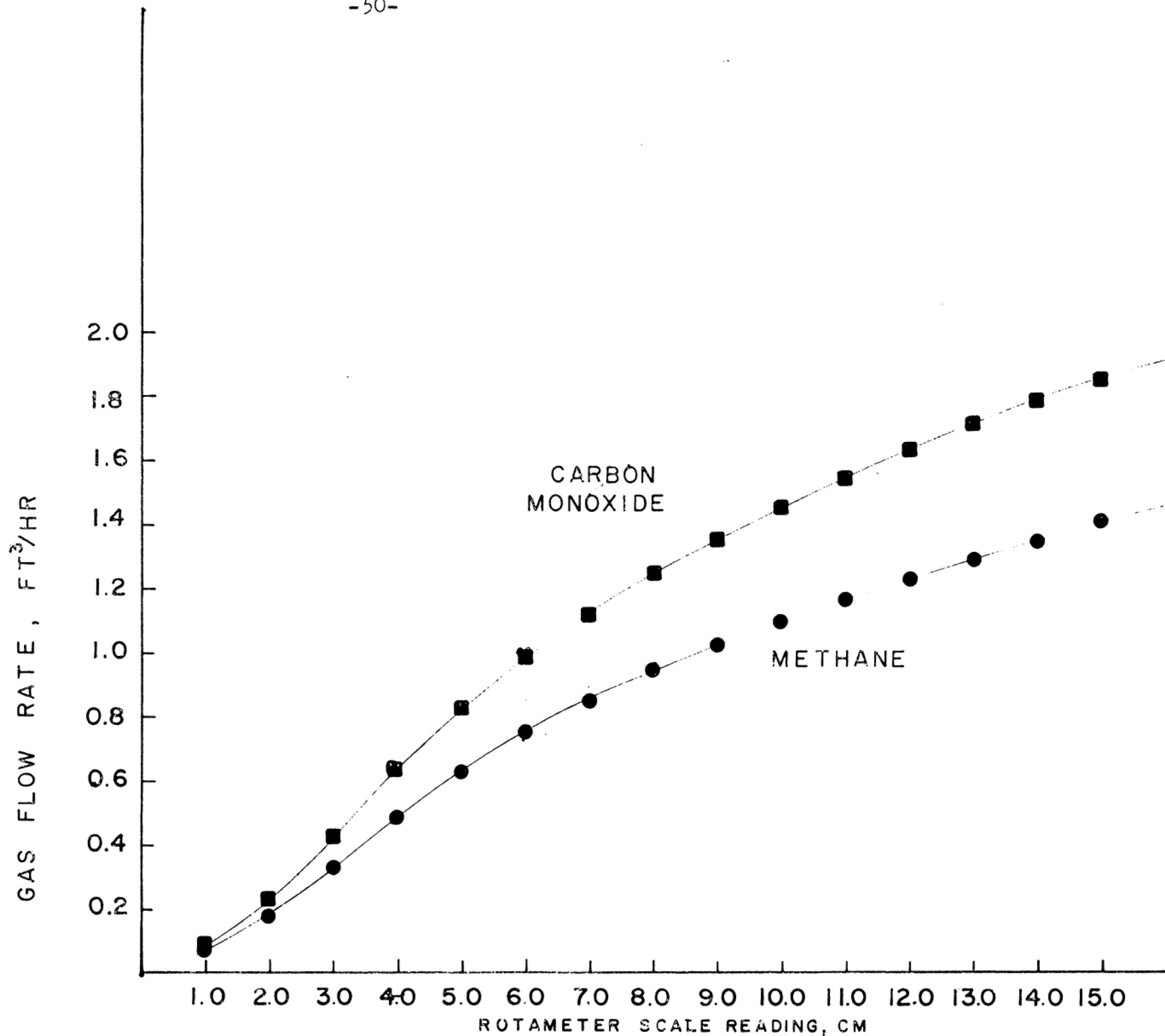


FIGURE 15. CALIBRATION CURVES FOR ROTAMETERS METERING CARBON MONOXIDE AND METHANE (MEASURED AT 54° C., & 707 mm MERCURY, ABSOLUTE).

exponential function fitted the data with a correlation coefficient of 0.95, and was deemed sufficient for this investigation. Tabular and graphical representations of this correlation are given in Table III, and Figure 16, page 53.

Physical Constants. A listing of physical properties of the gases and packing material used is given in Table IV, p. 54.

Dispersion Number. The dispersion number as a function of Reynolds Number was chosen as the parameter by which to evaluate the system. The data and results for 13 tests are given individually in Tables V through XVII, p. 55 through 67. A summary of the important results is given in Table XVIII, p. 68, and a plot of Reynolds number versus Dispersion Number appears as Figure 17, p. 69.

Frequency Response. For control purposes, the frequency response of a system is the logical choice of mathematical descriptions. Since the method of calculation used was tedious and repetitious, the IBM 1620 computer was utilized to perform the calculations. Table XIX, p. 70, is a listing of the FORTRAN language program used to calculate frequency response characteristics as a function of the frequency of the mathematically imposed input signal. A sample calculation of one set of frequency response data is given in Table XX, p. 71. Tables XXI through XXIV, pages 72 through 86, are tabular presentations of the frequency response characteristics for each test of open loop gain, open

TABLE III.

CALCULATION OF THE CORRELATION BETWEEN THE CHANGE  
IN MOLE FRACTION OF CARBON MONOXIDE IN THE FEED  
STREAM AND THE CHANGE IN THE SCALE READING OF  
THE OUTPUT FROM THE THERMAL CONDUCTIVITY CELL

TEST NO.	ROTA- METER READING FOR CO	ROTA- METER READING FOR CH4	FLOW RATE OF CO	FLOW RATE OF CH4	MOLE FRAC- TION OF CO, X	CHANGE IN SCALE READING E	LOG E	LOG X
	CM	CM	CU FT HR	CU FT HR		SCALE UNITS		
1053	05.95	08.45	0.975	0.985	0.500	0.292	-0.5346	-0.3010
1043	04.65	06.30	0.755	0.781	0.495	0.279	-0.5544	-0.3054
1012	01.70	02.70	0.180	0.271	0.435	0.211	-0.6757	-0.3615
1032	03.20	05.30	0.465	0.663	0.418	0.224	-0.6498	-0.3788
1054	07.60	06.40	1.205	0.790	0.682	0.378	-0.4225	-0.1662
1033	03.80	04.50	0.735	0.555	0.515	0.287	-0.5421	-0.2882
1044	05.55	04.95	0.915	0.615	0.597	0.357	-0.4473	-0.2240
1035	05.35	02.35	0.880	0.218	0.815	0.573	-0.2418	-0.0888
1055	11.40	03.85	1.580	0.460	0.778	0.573	-0.2418	-0.1090
1124	02.85	02.70	0.390	0.271	0.580	0.354	-0.4510	-0.2366
1151	03.85	14.90	0.605	1.400	0.296	0.155	-0.8097	-0.5287
1134	04.60	03.70	0.755	0.435	0.636	0.384	-0.4157	-0.1965
1141	02.45	12.05	0.310	1.240	0.179	0.092	-1.0362	-0.7471
1115	02.70	01.25	0.360	0.089	0.773	0.553	-0.2673	-0.1118
1114	02.30	02.10	0.285	0.180	0.585	0.353	-0.4522	-0.2328
1122	02.20	03.60	0.265	0.415	0.472	0.195	-0.7100	-0.3242
1113	01.90	02.30	0.210	0.210	0.500	0.258	-0.5884	-0.2749
1142	03.80	07.75	0.600	0.925	0.646	0.203	-0.6925	-0.1898
1152	04.70	11.30	0.770	1.185	0.394	0.205	-0.6882	-0.4045
1245	07.90	02.85	1.235	0.295	0.812	0.586	-0.2321	-0.0904
1231	01.90	07.40	0.215	0.890	0.198	0.100	-1.0000	-0.7033
1223	02.50	03.10	0.325	0.335	0.448	0.270	-0.5686	-0.3487
1211	01.00	03.10	0.089	0.335	0.207	0.111	-0.9547	-0.6840
1225	03.50	02.00	0.535	0.174	0.753	0.496	-0.3045	-0.1232
1221	01.30	04.30	0.125	0.525	0.207	0.125	-0.9031	-0.6840

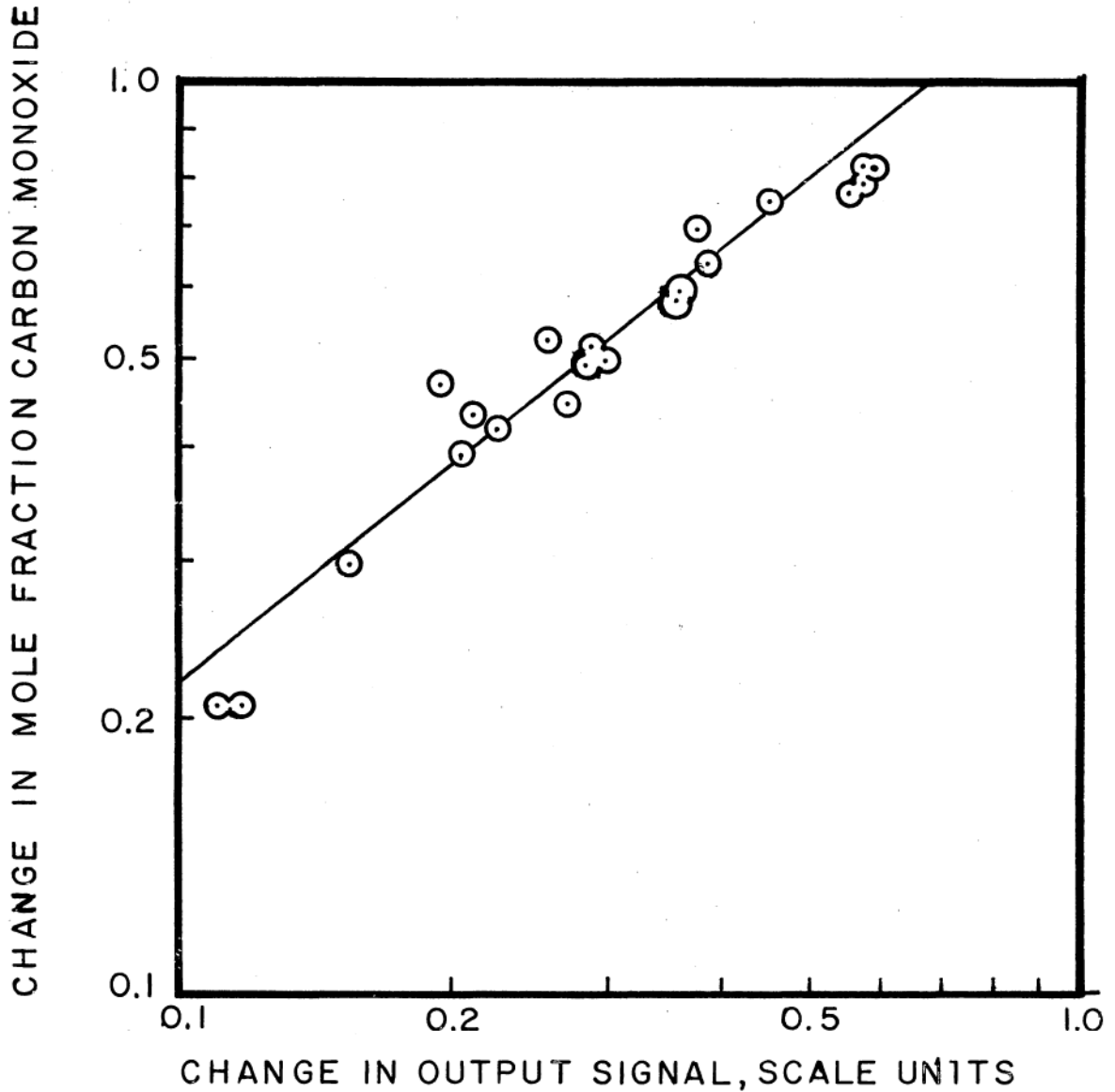


FIGURE 16. LEAST SQUARES RELATIONSHIP BETWEEN CHANGE IN SCALE READING OF OUTPUT SIGNAL FROM THERMAL CONDUCTIVITY CELL AND THE CHANGE IN COMPOSITION OF THE FEED STREAM TO THE REACTOR



TABLE IV.

PHYSICAL PROPERTIES OF MATERIALS  
USED IN INVESTIGATION

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VISCOSITIES OF PURE COMPONENT GASES AT 333 DEGREES, C. AND  
ONE ATMOSPHERE (1)

---

METHANE	0.0192 CENTIPOISES
CARBON MONOXIDE	0.0292 CENTIPOISES

---

<u>U. S. SIEVE SERIES</u>	<u>SIEVE NO.</u>	<u>SIEVE OPENING</u>	(2)
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	8	0.0937 INCHES	
	10	0.0787 INCHES	
	(11	0.0724) INCHES	
	12	0.0661 INCHES	
	14	0.0555 INCHES	

<u>VOID FRACTION OF PACKING MATERIAL</u>	<u>0.486</u>
--	--------------

1. PERRY, J. H. (EDITOR) CHEMICAL ENGINEER-S HANDBOOK, P. 371,  
MCGRAW-HILL, NEW YORK, N. Y.
2. IBID, P. 963.

TABLE V.  
 DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
 DISPERSION NUMBER FOR FLOW RATE OF 0.458 CUBIC FEET  
 OF FEED PER HOUR AND A FEED CONSISTING OF 0.776  
 MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 1115)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.090	0.000	0.000	0.000	0	0
.092	0.002	0.011	0.011	5	25
.095	0.005	0.024	0.013	10	100
.097	0.007	0.030	0.006	15	225
.100	0.010	0.040	0.010	20	400
.106	0.016	0.059	0.019	25	625
.114	0.024	0.081	0.022	30	900
.123	0.033	0.105	0.024	35	1225
.133	0.043	0.129	0.024	40	1600
.151	0.061	0.171	0.042	45	2025
.169	0.079	0.209	0.038	50	2500
.189	0.096	0.244	0.035	55	3025
.208	0.118	0.289	0.045	60	3600
.230	0.140	0.330	0.041	65	4225
.251	0.161	0.370	0.040	70	4900
.275	0.185	0.413	0.043	75	5625
.300	0.210	0.457	0.044	80	6400
.325	0.235	0.500	0.043	85	7225
.350	0.260	0.542	0.042	90	8100
.375	0.285	0.582	0.040	95	9025
.400	0.310	0.622	0.040	100	10000
.423	0.333	0.660	0.038	105	11025
.446	0.356	0.695	0.035	110	12100
.467	0.377	0.728	0.033	115	13225
.488	0.398	0.760	0.032	120	14400
.507	0.417	0.790	0.030	125	15625
.522	0.432	0.813	0.023	130	16900
.541	0.451	0.841	0.028	135	18225
.557	0.467	0.863	0.022	140	19600
.570	0.480	0.882	0.019	145	21025
.581	0.491	0.900	0.018	150	22500
.591	0.501	0.914	0.014	155	24025
.600	0.510	0.927	0.013	160	25600
.609	0.519	0.940	0.013	165	27225
.617	0.527	0.952	0.012	170	28900
.621	0.531	0.957	0.005	175	30625
.623	0.533	0.960	0.003	180	32400
.626	0.536	0.965	0.005	185	34225
.630	0.540	0.970	0.005	190	36100
.631	0.541	0.971	0.001	195	38025
.633	0.543	0.974	0.003	200	40000
.635	0.545	0.978	0.004	205	42025
.637	0.547	0.981	0.003	210	44100
.638	0.548	0.982	0.001	215	46225
.640	0.550	0.984	0.002	220	48400
.641	0.551	0.986	0.002	225	50625
.642	0.552	0.985	0.001	230	52900
.643	0.553	0.989	0.002	235	55225
.644	0.554	0.990	0.001	240	57600
.645	0.555	0.992	0.002	245	60025
.646	0.556	0.993	0.001	250	62500
.647	0.557	0.995	0.002	255	65025
.648	0.558	0.996	0.001	260	67600
.648	0.558	0.997	0.001	265	70225
.649	0.559	0.997	0.000	270	72900
.649	0.559	0.998	0.001	275	75625
.649	0.559	0.999	0.001	280	78400
.650	0.560	1.000	0.001	285	81225

VARIANCE = 12560.15 - 10245.49 = 2314.66 SEC

TABLE VI.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 0.540 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.815  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 2015)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.102	0.000	0.000	0.000	0	0
.104	0.002	0.005	0.005	5	25
.108	0.006	0.014	0.009	10	100
.111	0.009	0.020	0.006	15	225
.114	0.012	0.028	0.008	20	400
.117	0.015	0.034	0.006	25	625
.123	0.021	0.047	0.013	30	900
.130	0.028	0.068	0.021	35	1225
.140	0.038	0.098	0.030	40	1600
.151	0.049	0.117	0.019	45	2025
.166	0.064	0.155	0.027	50	2500
.180	0.078	0.190	0.046	55	3025
.197	0.095	0.230	0.040	60	3600
.215	0.113	0.274	0.044	65	4225
.231	0.129	0.312	0.038	70	4900
.251	0.149	0.363	0.051	75	5625
.270	0.168	0.409	0.046	80	6400
.290	0.188	0.459	0.050	85	7225
.310	0.208	0.506	0.047	90	8100
.331	0.229	0.557	0.051	95	9025
.352	0.250	0.606	0.049	100	10000
.370	0.268	0.639	0.033	105	11025
.390	0.288	0.677	0.038	110	12100
.412	0.310	0.714	0.037	115	13225
.430	0.328	0.746	0.032	120	14400
.450	0.348	0.776	0.030	125	15625
.469	0.367	0.804	0.028	130	16900
.488	0.386	0.833	0.029	135	18225
.506	0.404	0.858	0.025	140	19600
.520	0.418	0.878	0.020	145	21025
.534	0.432	0.899	0.021	150	22500
.547	0.445	0.909	0.010	155	24025
.558	0.456	0.921	0.012	160	25600
.567	0.465	0.931	0.010	165	27225
.576	0.474	0.940	0.009	170	28900
.583	0.481	0.946	0.006	175	30625
.591	0.489	0.955	0.009	180	32400
.600	0.498	0.963	0.009	185	34225
.604	0.502	0.967	0.004	190	36100
.610	0.508	0.973	0.006	195	38025
.615	0.513	0.977	0.004	200	40000
.620	0.518	0.981	0.004	205	42025
.621	0.519	0.983	0.002	210	44100
.624	0.522	0.985	0.002	215	46225
.626	0.524	0.986	0.001	220	48400
.629	0.527	0.987	0.001	225	50625
.630	0.528	0.988	0.001	230	52900
.631	0.529	0.989	0.001	235	55225
.632	0.530	0.990	0.001	240	57600
.633	0.531	0.991	0.001	245	60025
.633	0.531	0.992	0.001	250	62500
.634	0.532	0.993	0.001	255	65025
.635	0.553	0.994	0.001	260	67600
.636	0.534	0.995	0.001	265	70225
.636	0.534	0.996	0.001	270	72900
.637	0.535	0.997	0.001	275	75625
.638	0.536	0.998	0.001	280	78400
.639	0.537	0.999	0.001	285	81225
.640	0.538	1.000	0.001	290	84100

$$\text{VARIANCE} = 10823.15 - 8561.80 = 2261.35 \text{ SEC}$$

TABLE VII.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 0.452 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.796  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 3015)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.057	0.000	0.000	0.000	0	0
.058	0.001	0.007	0.007	5	25
.061	0.004	0.020	0.013	10	100
.065	0.008	0.034	0.014	15	225
.068	0.011	0.046	0.012	20	400
.070	0.013	0.050	0.004	25	625
.075	0.018	0.065	0.015	30	900
.085	0.028	0.094	0.029	35	1225
.093	0.036	0.115	0.021	40	1600
.106	0.049	0.146	0.031	45	2025
.120	0.063	0.179	0.033	50	2500
.135	0.078	0.211	0.032	55	3025
.150	0.093	0.244	0.033	60	3600
.170	0.113	0.286	0.042	65	4225
.190	0.133	0.325	0.039	70	4900
.209	0.152	0.358	0.033	75	5625
.231	0.174	0.403	0.045	80	6400
.250	0.193	0.437	0.034	85	7225
.271	0.214	0.475	0.038	90	8100
.298	0.241	0.522	0.047	95	9025
.320	0.263	0.559	0.037	100	10000
.340	0.283	0.593	0.034	105	11025
.361	0.304	0.629	0.036	110	12100
.383	0.326	0.665	0.036	115	13225
.403	0.346	0.698	0.033	120	14400
.424	0.367	0.730	0.032	125	15625
.440	0.383	0.754	0.034	130	16900
.489	0.432	0.832	0.022	140	19600
.504	0.447	0.855	0.023	145	21325
.460	0.403	0.787	0.033	135	18225
.516	0.459	0.873	0.318	150	22500
.527	0.470	0.891	0.018	155	24025
.536	0.479	0.904	0.013	160	25600
.545	0.488	0.917	0.013	165	27225
.553	0.496	0.930	0.013	170	28900
.560	0.503	0.940	0.010	175	30625
.567	0.510	0.950	0.010	180	32400
.572	0.515	0.956	0.006	185	34225
.574	0.517	0.961	0.005	190	36100
.577	0.520	0.966	0.005	195	38025
.580	0.523	0.969	0.003	200	40000
.582	0.525	0.972	0.003	205	42025
.586	0.529	0.979	0.003	215	46225
.584	0.527	0.976	0.004	210	44100
.588	0.531	0.980	0.001	220	48400
.590	0.533	0.984	0.004	225	50625
.592	0.535	0.985	0.001	230	52900
.594	0.537	0.989	0.004	235	55225
.596	0.539	0.990	0.001	240	57600
.597	0.540	0.992	0.002	245	60025
.598	0.541	0.993	0.00.	250	62500
.599	0.542	0.995	0.002	255	65025
.600	0.543	0.997	0.002	260	67600
.601	0.544	0.998	0.001	265	70225
.602	0.545	1.000	0.002	270	72900

$$\text{VARIANCE} = 11449.32 - 9378.95 = 2070.37 \text{ SEC}$$

TABLE VIII.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 0.720 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.743  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 1225)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.049	0.000	0.000	0.000	0	0
.051	0.002	0.012	0.012	5	25
.056	0.007	0.033	0.021	10	100
.058	0.009	0.040	0.007	15	225
.061	0.012	0.051	0.011	20	400
.070	0.021	0.080	0.029	25	625
.085	0.036	0.122	0.042	30	900
.101	0.052	0.166	0.044	35	1225
.126	0.077	0.229	0.063	40	1600
.157	0.108	0.295	0.066	45	2025
.191	0.142	0.367	0.072	50	2500
.225	0.176	0.437	0.070	55	3025
.263	0.214	0.510	0.073	60	3600
.300	0.251	0.580	0.070	65	4225
.330	0.281	0.635	0.055	70	4900
.361	0.312	0.690	0.055	75	5625
.395	0.346	0.749	0.059	80	6400
.418	0.369	0.790	0.041	85	7225
.443	0.394	0.830	0.040	90	8100
.462	0.413	0.864	0.034	95	9025
.481	0.432	0.895	0.031	100	10000
.494	0.445	0.916	0.021	105	11025
.508	0.459	0.939	0.023	110	12100
.517	0.468	0.954	0.015	115	13225
.524	0.475	0.967	0.013	120	14400
.529	0.480	0.975	0.008	125	15625
.530	0.481	0.977	0.002	130	16900
.532	0.483	0.979	0.002	135	18225
.534	0.485	0.981	0.002	140	19600
.535	0.486	0.982	0.001	145	21025
.536	0.487	0.983	0.001	150	22500
.537	0.488	0.984	0.001	155	24025
.538	0.489	0.986	0.002	160	25600
.539	0.490	0.988	0.002	165	27225
.540	0.491	0.989	0.001	170	28900
.540	0.491	0.991	0.002	175	30625
.541	0.492	0.992	0.001	180	32400
.541	0.492	0.993	0.001	185	34225
.541	0.492	0.993	0.000	190	36100
.542	0.493	0.994	0.000	195	38025
.542	0.493	0.995	0.001	200	40000
.543	0.494	0.996	0.001	205	42025
.543	0.494	0.996	0.000	210	44100
.543	0.494	0.996	0.000	215	46225
.543	0.494	0.996	0.000	220	48400
.543	0.494	0.996	0.000	225	50625
.543	0.494	0.996	0.000	230	52900
.544	0.495	0.997	0.001	235	55225
.544	0.495	0.998	0.001	240	57600
.544	0.495	0.998	0.000	245	60025
.545	0.496	1.000	0.002	250	62500

$$\text{VARIANCE} = 5244.52 - 4254.30 = 990.22 \text{ SEC}$$

TABLE IX.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 1.115 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.789  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 1035)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.040	0.000	0.000	0.000	0	0
.044	0.004	0.019	0.019	5	25
.049	0.009	0.036	0.017	10	100
.053	0.013	0.048	0.012	15	225
.074	0.034	0.120	0.072	20	400
.110	0.070	0.165	0.045	25	625
.157	0.117	0.280	0.115	30	900
.222	0.182	0.400	0.120	35	1225
.291	0.251	0.517	0.117	40	1600
.369	0.329	0.642	0.125	45	2025
.411	0.371	0.705	0.063	50	2500
.470	0.430	0.795	0.090	55	3025
.514	0.474	0.858	0.063	60	3600
.547	0.507	0.907	0.049	65	4225
.575	0.535	0.946	0.039	70	4900
.595	0.555	0.974	0.028	75	5625
.607	0.567	0.991	0.017	80	6400
.612	0.572	0.999	0.008	85	7225
.613	0.573	1.000	0.001	90	8100

VARIANCE = 2139.50 - 1848.14 = 291.36 SEC

TABLE X.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 1.110 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.798  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 2135)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME)
SCALE UNITS	SCALE UNITS			SEC	(SEC)
.089	0.004	0.019	0.019	5	25
.092	0.007	0.030	0.011	10	100
.100	0.015	0.055	0.025	15	225
.112	0.027	0.088	0.033	20	400
.142	0.057	0.158	0.070	25	625
.178	0.093	0.235	0.077	30	900
.198	0.113	0.275	0.040	35	1225
.228	0.143	0.332	0.057	40	1600
.290	0.205	0.454	0.122	45	2025
.355	0.270	0.551	0.974	50	2500
.420	0.345	0.668	0.117	55	3025
.475	0.390	0.738	0.070	60	3600
.523	0.438	0.811	0.073	65	4225
.562	0.477	0.867	0.056	70	4900
.590	0.505	0.908	0.041	75	5625
.612	0.527	0.937	0.029	80	6400
.628	0.543	0.961	0.024	85	7225
.637	0.552	0.975	0.014	90	8100
.643	0.558	0.983	0.008	95	9025
.647	0.562	0.987	0.004	100	10000
.648	0.563	0.989	0.002	105	11025
.649	0.564	0.991	0.002	110	12100
.651	0.566	0.992	0.001	115	13225
.652	0.567	0.994	0.002	120	14400
.653	0.568	0.995	0.001	125	15625
.653	0.568	0.995	0.000	130	16900
.653	0.568	0.996	0.001	135	18225
.654	0.569	0.997	0.001	140	19600
.654	0.569	0.998	0.001	145	21025
.654	0.569	0.998	0.000	150	22500
.654	0.569	0.999	0.001	155	24025
.655	0.570	1.000	0.001	160	25600

$$\text{VARIANCE} = 2797.55 - 2512.01 = 467.54 \text{ SEC}$$

TABLE XI.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 1.105 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.801  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 3035)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.094	0.000	0.000	0.000	0	0
.099	0.005	0.011	0.011	5	25
.103	0.009	0.023	0.010	10	100
.116	0.022	0.053	0.032	15	225
.132	0.038	0.091	0.038	20	400
.175	0.081	0.194	0.103	25	625
.223	0.129	0.306	0.112	30	900
.290	0.196	0.468	0.162	35	1225
.360	0.266	0.625	0.157	40	1600
.425	0.331	0.735	0.110	45	2025
.475	0.381	0.808	0.073	50	2500
.519	0.425	0.855	0.047	55	3025
.555	0.461	0.908	0.053	60	3600
.585	0.491	0.938	0.030	65	4225
.610	0.516	0.961	0.023	70	4900
.625	0.531	0.974	0.013	75	5625
.639	0.545	0.984	0.010	80	6400
.645	0.551	0.988	0.004	85	7225
.649	0.555	0.990	0.002	90	8100
.651	0.557	0.991	0.001	95	9025
.652	0.558	0.993	0.002	100	10000
.654	0.560	0.994	0.001	105	11025
.655	0.561	0.995	0.001	110	12100
.656	0.562	0.996	0.001	115	13225
.657	0.563	0.997	0.001	120	14400
.658	0.564	0.998	0.001	125	15625
.659	0.565	0.999	0.001	130	16900
.660	0.566	1.000	0.001	135	18225

VARIANCE = 1928.92 - 1651.20 = 277.72 SEC



TABLE XII.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 1.625 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.760  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 1245)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCENTRATION CHANGE	CONCENTRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.042	0.000	0.000	0.000	0	0
.045	0.003	0.015	0.015	5	25
.051	0.009	0.035	0.020	10	100
.070	0.029	0.090	0.055	15	225
.108	0.066	0.174	0.084	20	400
.172	0.130	0.300	0.126	25	625
.252	0.210	0.439	0.139	30	900
.346	0.304	0.591	0.152	35	1225
.430	0.388	0.720	0.129	40	1600
.504	0.462	0.827	0.107	45	2025
.550	0.006	0.891	0.064	50	2500
.586	0.544	0.942	0.051	55	3025
.610	0.568	0.975	0.033	60	3600
.620	0.578	0.989	0.014	65	4225
.624	0.582	0.993	0.004	70	4900
.624	0.582	0.994	0.001	75	5625
.625	0.583	0.995	0.001	80	6400
.626	0.584	0.996	0.001	85	7225
.626	0.584	0.997	0.001	90	8100
.627	0.585	0.998	0.001	95	9025
.627	0.585	0.999	0.001	100	10000
.628	0.586	1.000	0.001	105	11025

VARIANCE = 1430.70 - 1239.04 = 191.66 SEC

TABLE XIII.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 1.635 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.755  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 2245)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.081	0.000	0.000	0.000	0	0
.083	0.002	0.011	0.011	5	25
.089	0.008	0.033	0.022	10	100
.100	0.019	0.065	0.032	15	225
.130	0.049	0.104	0.039	20	400
.190	0.109	0.265	0.161	25	625
.280	0.199	0.428	0.162	30	900
.380	0.299	0.591	0.164	35	1225
.465	0.384	0.721	0.130	40	1600
.525	0.454	0.824	0.103	45	2025
.585	0.504	0.896	0.072	50	2500
.615	0.534	0.938	0.042	55	3025
.635	0.554	0.966	0.028	60	3600
.647	0.566	0.983	0.017	65	4225
.651	0.570	0.988	0.005	70	4900
.653	0.572	0.991	0.003	75	5625
.655	0.574	0.994	0.003	80	6400
.657	0.567	0.995	0.001	85	7225
.658	0.577	0.996	0.001	90	8100
.658	0.577	0.997	0.001	95	9025
.658	0.577	0.998	0.001	100	10000
.659	0.578	1.000	0.002	105	11025

VARIANCE = 1450.71 - 1302.13 = 148.58 SEC

TABLE XIV.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 1.615 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.765  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 3045)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCEN- TRATION CHANGE	CONCEN- TRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.090	0.000	0.000	0.000	0	0
.094	0.004	0.018	0.018	5	25
.103	0.013	0.048	0.030	10	100
.132	0.042	0.124	0.076	15	225
.175	0.085	0.216	0.092	20	400
.263	0.173	0.384	0.168	25	625
.365	0.275	0.555	0.171	30	900
.460	0.370	0.703	0.148	35	1225
.530	0.440	0.809	0.106	40	1600
.590	0.500	0.894	0.085	45	2025
.621	0.531	0.938	0.044	50	2500
.640	0.550	0.964	0.026	55	3025
.653	0.563	0.983	0.019	60	3600
.659	0.569	0.992	0.009	65	4225
.660	0.570	0.993	0.001	70	4900
.660	0.570	0.993	0.000	75	5625
.660	0.570	0.994	0.001	80	6400
.661	0.571	0.994	0.000	85	7225
.661	0.571	0.995	0.001	90	8100
.662	0.572	0.995	0.000	95	9025
.663	0.573	0.996	0.001	100	10000
.663	0.573	0.997	0.001	105	11025
.664	0.574	0.998	0.001	110	12100
.665	0.575	1.000	0.002	115	13225

VARIANCE = 1213.32 - 1029.45 = 183.87 SEC

TABLE XV.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND DISPERSION NUMBER FOR FLOW RATE OF 2.190 CUBIC FEET OF FEED PER HOUR AND A FEED CONSISTING OF 0.722 MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 1055)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCENTRATION CHANGE	CONCENTRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.040	0.000	0.000	0.000	0	0
.045	0.005	0.023	0.023	5	25
.070	0.030	0.095	0.072	10	100
.133	0.093	0.234	0.139	15	225
.235	0.195	0.422	0.188	20	400
.375	0.335	0.652	0.230	25	625
.475	0.435	0.803	0.151	30	900
.541	0.501	0.899	0.096	35	1225
.580	0.540	0.955	0.056	40	1600
.597	0.557	0.978	0.023	45	2025
.610	0.570	0.997	0.019	50	2500
.612	0.572	0.998	0.001	55	3025
.613	0.573	1.000	0.002	60	3600

$$\text{VARIANCE} = 705.40 - 611.08 = 94.32 \text{ SEC}$$

TABLE XVI.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND DISPERSION NUMBER FOR FLOW RATE OF 2.155 CUBIC FEET OF FEED PER HOUR AND A FEED CONSISTING OF 0.733 MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 2055)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCENTRATION CHANGE	CONCENTRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.087	0.000	0.000	0.000	0	0
.098	0.011	0.042	0.042	5	25
.141	0.054	0.146	0.104	10	100
.225	0.138	0.320	0.174	15	225
.325	0.238	0.494	0.174	20	400
.425	0.338	0.654	0.160	25	625
.530	0.443	0.881	0.157	30	900
.600	0.513	0.912	0.101	35	1225
.633	0.546	0.956	0.046	40	1600
.648	0.561	0.980	0.022	45	2025
.653	0.566	0.986	0.006	50	2500
.656	0.569	0.991	0.005	55	3025
.658	0.571	0.992	0.001	60	3600
.660	0.573	0.994	0.002	65	4225
.660	0.573	0.995	0.001	70	4900
.660	0.573	0.995	0.000	75	5625
.661	0.574	0.997	0.002	80	6400
.661	0.574	0.997	0.000	85	7225
.662	0.575	0.998	0.001	90	8100
.662	0.575	0.999	0.001	95	9025
.663	0.576	1.000	0.001	100	10000

VARIANCE = 690.34 - 561.45 = 128.89 SEC

TABLE XVII.

DATA USED FOR THE CALCULATION OF RESIDENCE TIME AND  
DISPERSION NUMBER FOR FLOW RATE OF 2.055 CUBIC FEET  
OF FEED PER HOUR AND A FEED CONSISTING OF 0.752  
MOLE FRACTION OF CARBON MONOXIDE IN METHANE (TEST 3155)

CHART READING	CHANGE IN CHART READING FROM START	FRACTION OF TOTAL CONCENTRATION CHANGE	CONCENTRATION CHANGE PER FIVE SECONDS	TIME BEYOND DEAD TIME	(TIME) <sup>2</sup>
SCALE UNITS	SCALE UNITS			SEC	(SEC) <sup>2</sup>
.082	0.000	0.000	0.000	0	0
.088	0.006	0.027	0.027	5	25
.100	0.018	0.064	0.037	10	100
.145	0.063	0.172	0.108	15	225
.235	0.153	0.351	0.179	20	400
.385	0.303	0.605	0.254	25	625
.485	0.403	0.728	0.153	30	900
.570	0.488	0.884	0.126	35	1225
.615	0.533	0.948	0.064	40	1600
.634	0.552	0.975	0.027	45	2025
.645	0.563	0.990	0.015	50	2500
.647	0.565	0.993	0.003	55	3025
.648	0.566	0.995	0.002	60	3600
.649	0.567	0.996	0.001	65	4225
.649	0.567	0.997	0.001	70	4900
.650	0.568	0.998	0.001	75	5625
.650	0.568	0.999	0.001	80	6400
.651	0.569	1.000	0.001	85	7225

VARIANCE = 790.30 - 688.54 = 101.76 SEC

TABLE XVIII.

SUMMARY OF DISPERSION NUMBERS  
REYNOLDS NUMBERS AND SCHMIDT NUMBERS

TEST NO.	FLOW RATE	MOLE FRAC-TION CH <sub>4</sub>	RESI-DENCE TIME	DISPERSION NUMBER	REYNOLDS NUMBER	SCHMIDT NUMBER
	CFH		SEC			
1115	0.4580	0.7760	398.1	0.009760	0.0599	0.9101
1225	0.7200	0.7430	266.8	0.006974	0.0891	0.9133
1035	1.1150	0.7890	194.4	0.003849	0.1228	0.9089
1245	1.6250	0.7600	142.6	0.004656	0.1669	0.9116
1055	2.1900	0.7220	110.2	0.003903	0.2151	0.9153
2015	0.5400	0.8150	364.8	0.008225	0.0656	0.9065
2225	0.7220	0.7540	268.0	0.006761	0.0887	0.9122
2135	1.1050	0.8010	194.6	0.006070	0.1228	0.9078
2245	1.6350	0.7550	143.2	0.004340	0.1662	0.9121
2055	2.1550	0.7330	120.4	0.005280	0.1972	0.9142
3015	0.7030	0.7900	412.6	0.008564	0.0579	0.9083
3125	0.7030	0.2100	254.8	0.009101	0.0937	0.9088
3035	1.1100	0.7980	191.3	0.005277	0.1249	0.9081
3045	1.6150	0.7650	130.5	0.005352	0.1825	0.9112
3155	2.0550	0.7520	108.6	0.004200	0.2189	0.9124

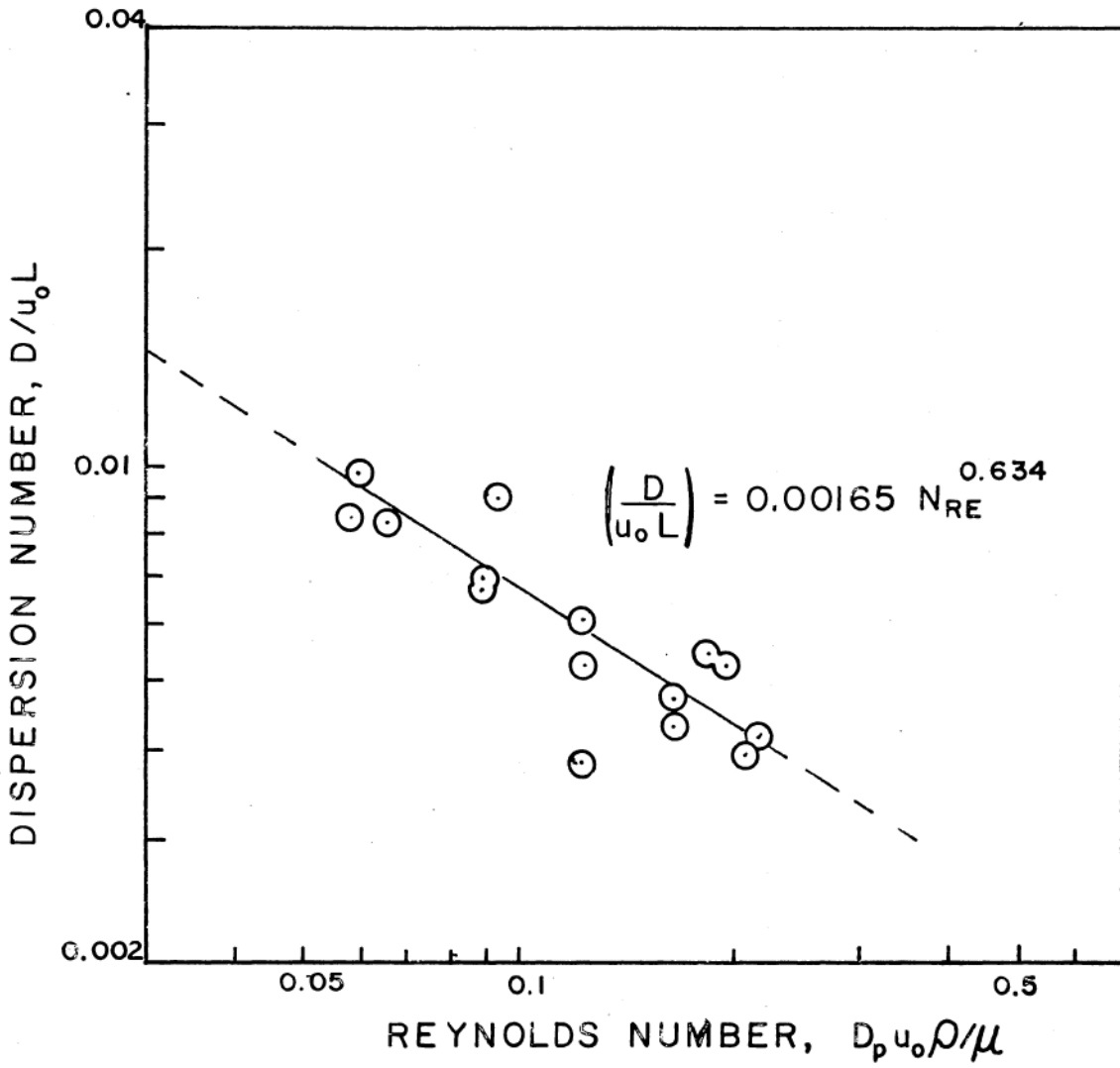


FIGURE 17. CORRELATION OF DISPERSION NUMBER WITH REYNOLDS NUMBER



TABLE XIX.  
 1620 FORTRAN PROGRAM  
 FOR THE DETERMINATION OF  
 FREQUENCY RESPONSE OF A PACKED  
 TUBULAR REACTOR

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C C J. M. WASHINGTON, DEPARTMENT OF CHEMICAL ENGINEERING
    DIMENSION ARDSU(50), AMDU(50)
    1 READ 2, KCODE
      PUNCH 61, KCODE
111 READ 2, IT,L
      READ 4, OMEGA, DELT, AMAX
      PUNCH 6
      PUNCH 11
      READ 4, AZR
      DO 19 I=1,L
        READ 4, ASU
19  AMDU(I)=((ASU-AZR)/(AMAX-AZR))*0.7981
        M=L-1
        DO 70 J=1,M
          70 ARDSU(J)=AMDU(J+1)-AMDU(J)
22  OMDLT=OMEGA*DELT
      R=0.0
      B=0.0
      C=0.0
      DO 35 K=1,M
        AK=(2.0*(R+1.0)-1.0)/2.0
        ASIN=SIN(OMDLT*AK)
        TERMB=ARDSU(K)*ASIN
        ACOS=COS(OMDLT*AK)
        TERMC=ARDSU(K)*ACOS
        B=B+TERMB
        C=C+TERMC
35  R=R+1.0
      GAIN=SQRT(B*B+C*C)
      PHASE=-(ATAN(B/C))*57.29577
      CLGAN=GAIN/SQRT((1.0+C)*(1.0+C)+B*B)
      CLPHS=PHASE+ATAN(B/(1.0+C))*57.29577
      PUNCH 41,B,C,OMEGA,GAIN,PHASE,CLGAN,CLPHS
      IF(OMEGA-99.9999) 73, 50, 50
73  IF(GAIN-0.02) 50, 50, 74
74  IF(SENSE SWITCH 1) 151, 50
151 OMEGA=1.5*OMEGA
      GO TO 22
50  PUNCH 51, KCODE
      GO TO 1
61  FORMAT (7HCODE = ,15)
    2  FORMAT(I4,I4)
    4  FORMAT(F8.4,F8.4,F8.4)
    6  FORMAT(10X31HFROM STEP TO FREQUENCY RESPONSE)
11  FORMAT (21X3HOME,6X6HO.L.GN,4X6HO.L.PH,4X6HC.L.GN,4X6HC.L.PH)
41  FORMAT (F7.3,2X,F7.3,2X,F8.4,2X,F8.4,2X,F8.3,2X,F8.4,2X,F8.3)
51  FORMAT (20X,14HEND OF CODE = 15//)
59  END
  
```

TABLE XX.

SAMPLE CALCULATION OF FREQUENCY RESPONSE OF REACTOR  
 AT A FLOW RATE OF 1.1625 CUBIC FEET PER MINUTE  
 AND 0.760 MOLE FRACTION OF CARBON MONOXIDE IN METHANE  
 INPUT FREQUENCY EQUALS 0.10 RADIAN PER MINUTE

SCALE READING	SCALE READING DIFFER- ENCE	FRACTION OF TOTAL CONCEN- TRATION CHANGE	DELTA Y(I)	(2I-1)/2 K	$K\omega\Delta T$	COSINE $K\omega\Delta T$	SINE $K\omega\Delta T$
SCALE UNITS	SCALE UNITS				RADIANS		
.042	0.000	0.000	0.000	0.000	0.0000	1.0000	0.0000
.045	0.003	0.015	0.015	0.500	0.0042	1.0000	0.0042
.051	0.009	0.035	0.020	01.500	0.0125	0.9999	0.0125
.071	0.029	0.090	0.055	02.500	0.0208	0.9998	0.0208
.108	0.066	0.174	0.084	03.500	0.0291	0.9996	0.0291
.172	0.130	0.300	0.126	04.500	0.0375	0.9993	0.0375
.252	0.210	0.439	0.139	05.500	0.0468	0.9989	0.0468
.346	0.304	0.591	0.152	06.500	0.0551	0.9985	0.0550
.430	0.388	0.720	0.129	07.500	0.0635	0.9980	0.0640
.504	0.462	0.827	0.107	08.500	0.0718	0.9974	0.0718
.550	0.508	0.891	0.064	09.500	0.0801	0.9968	0.0801
.586	0.544	0.942	0.051	10.500	0.0885	0.9960	0.0884
.610	0.568	0.975	0.033	11.500	0.0968	0.9953	0.0967
.620	0.578	0.989	0.014	12.500	0.1051	0.9944	0.1049
.624	0.582	0.993	0.004	13.500	0.1135	0.9936	0.1133
.624	0.582	0.993	0.000	14.500	0.1218	0.9926	0.1215
.625	0.583	0.995	0.002	15.500	0.1301	0.9915	0.1297
.626	0.584	0.996	0.001	16.500	0.1384	0.9905	0.1380
.626	0.584	0.996	0.000	17.500	0.1468	0.9892	0.1463
.627	0.585	0.998	0.002	18.500	0.1551	0.9880	0.1544
.627	0.585	0.998	0.000	19.500	0.1634	0.9867	0.1627
.628	0.586	1.000	0.002	20.500	0.1718	0.9853	0.1710

TABLE XXI.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.776  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 0.458 CUBIC FEET PER MINUTE  
 (TEST 1115)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.155	.964	.1000	.9765	-9.176	.4956	-4.642
.231	.944	.1500	.9725	-13.760	.4966	-6.976
.339	.901	.2250	.9637	-20.626	.4988	-10.506
.484	.810	.3375	.9440	-30.891	.5037	-15.901
.650	.624	.5062	.9011	-46.172	.5151	-24.356
.755	.294	.7593	.8114	-68.687	.5411	-38.411
.629	-.122	1.1390	.6409	79.009	.5934	114.641
.223	-.307	1.7085	.3798	36.060	.5217	53.947
.022	-.125	2.5628	.1273	-10.017	.1455	-11.468
.001	-.035	3.8443	.0359	-2.549	.0373	-2.645
.003	-.005	5.7665	.0100	-58.411	.0100	-58.904

TABLE XXII.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.743  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 0.720 CUBIC FEET PER MINUTE  
 (TEST 1225)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.103	.991	.1000	.9970	-5.965	.4999	-2.986
.154	.983	.1500	.9954	-8.945	.5003	-4.482
.230	.964	.2250	.9918	-13.410	.5019	-6.732
.337	.923	.3375	.9837	-20.091	.5036	-10.128
.463	.836	.5062	.9660	-30.055	.5087	-15.293
.624	.658	.7593	.9281	-44.823	.5206	-23.291
.781	.340	1.1390	.8519	-66.470	.5492	-36.234
.706	-.096	1.7085	.7130	62.182	.6219	120.219
.294	-.391	2.5628	.4894	36.974	.7235	62.773
.079	-.192	3.8443	.2083	-22.341	.2568	-27.943
.069	-.018	5.7665	.0716	-75.384	.0728	-79.424
.010	.026	8.6497	.0284	-20.617	.0276	-20.059
.006	.017	12.9746	.0188	-20.396	.0185	-20.026

TABLE XXIII.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.789  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 1.115 CUBIC FEET PER MINUTE  
 (TEST 1035)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.067	.997	.1000	.9995	-3.857	.5001	-1.929
.100	.994	.1500	.9990	-5.786	.5004	-2.894
.150	.986	.2250	.9979	-8.678	.5009	-4.343
.224	.969	.3375	.9954	-13.017	.5020	-6.523
.330	.932	.5062	.9896	-19.523	.5047	-9.812
.477	.852	.7593	.9769	-29.274	.5107	-14.811
.657	.683	1.1390	.9487	-43.877	.5248	-22.545
.809	.365	1.7085	.8880	-65.694	.5594	-35.039
.756	-.107	2.5628	.7639	81.913	.6529	122.190
.308	-.443	3.8443	.5398	34.819	.8483	63.791
.109	-.218	5.7665	.2444	-26.615	.3097	-34.592
.099	-.007	8.6497	.0998	-85.713	.1000	-91.441
.024	.028	12.9746	.0372	-40.953	.0362	-39.591
.012	.057	19.4619	.0593	-12.516	.0561	-11.819
.060	-.010	29.1929	.0609	80.334	.0614	83.806
.006	.067	43.7893	.0678	5.829	.0635	5.459
.076	-.044	65.6840	.0886	-60.034	.0924	-64.627
.059	.027	98.5261	.0656	64.759	.0637	61.454

TABLE XXIV.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.760  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 1.625 CUBIC FEET PER MINUTE  
 (TEST 1245)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.054	.998	.1000	.9997	-3.115	.5001	-1.557
.081	.996	.1500	.9994	-4.672	.5002	-2.337
.121	.991	.2250	.9986	-7.008	.5006	-3.506
.181	.980	.3375	.9970	-10.511	.5013	-5.263
.269	.955	.5062	.9932	-15.763	.5030	-7.908
.394	.902	.7593	.9849	-23.632	.5069	-11.906
.560	.787	1.1390	.9666	-35.405	.5159	-18.012
.739	.558	1.7085	.9269	-52.968	.5373	-27.566
.829	.160	2.5628	.8448	-79.032	.5922	-43.482
.611	-.317	3.8443	.6894	62.357	.7523	104.443
.048	-.433	5.7665	.4366	6.416	.7685	11.344
.138	-.055	8.6497	.1497	-68.175	.1568	-76.548
.003	.032	12.9746	.0326	6.641	.0316	6.432
.001	.016	19.4619	.0169	-5.788	.0166	-5.691

TABLE XXV.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.722  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 2.190 CUBIC FEET PER MINUTE  
 (TEST 1055)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.037	.999	.1000	.9998	-2.122	.5000	-1.061
.055	.998	.1500	.9997	-3.183	.5001	-1.591
.083	.995	.2250	.9993	-4.774	.5002	-2.388
.124	.990	.3375	.9985	-7.161	.5006	-3.583
.185	.979	.5062	.9966	-10.741	.5013	-5.379
.275	.953	.7593	.9924	-16.108	.5030	-8.084
.402	.897	1.1390	.9850	-24.147	.5069	-12.178
.567	.776	1.7085	.9623	-36.172	.5158	-18.445
.743	.538	2.5628	.9173	-54.091	.5370	-28.306
.812	.135	3.8443	.8241	-80.568	.5962	-44.956
.569	-.314	5.7665	.6500	-61.100	.7293	100.782
.054	-.388	8.6497	.3925	-8.041	.6396	13.174
.138	-.068	12.9748	.1510	-66.258	.1590	-74.632
.029	.019	19.4619	.0356	-56.478	.0349	-54.807
.044	-.004	29.1929	.0465	-84.119	.0446	-86.665
.019	-.038	43.7893	.0434	-26.699	.0451	-27.862
.058	.307	65.6840	.3134	10.701	.2393	8.153
.026	.023	98.5261	.0353	48.064	.0345	46.591

TABLE XXVI.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.815  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 0.540 CUBIC FEET PER MINUTE  
 (TEST 2015)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.145	.979	.1000	.9901	-8.457	.4988	-4.249
.216	.962	.1500	.9867	-12.682	.4997	-6.384
.318	.925	.2250	.9790	-19.011	.5016	-9.607
.458	.845	.3375	.9621	-28.474	.5058	-14.517
.625	.681	.5062	.9250	-42.565	.5156	-22.151
.757	.379	.7593	.8471	-63.350	.5382	-34.597
.695	-.040	1.1390	.6963	86.678	.5876	122.597
.325	-.316	1.7085	.4534	45.795	.5989	71.222
.008	-.180	2.5628	.1804	-2.557	.2200	-3.120
.008	-.054	3.8443	.0556	-8.988	.0588	-9.516
.010	.000	5.7665	.0103	89.243	.0103	88.649



TABLE XXVII.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.752  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 0.702 CUBIC FEET PER MINUTE  
 (TEST 2225)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.108	.992	.1000	.9986	-6.210	.5003	-3.107
.161	.983	.1500	.9969	-9.314	.5008	-4.664
.239	.963	.2250	.9932	-13.964	.5020	-7.005
.351	.919	.3375	.9848	-20.923	.5045	-10.542
.502	.825	.5062	.9663	-31.307	.5103	-15.928
.674	.635	.7593	.9265	-46.708	.5237	-24.297
.790	.299	1.1390	.8457	-69.267	.5559	-37.938
.683	-.140	1.7085	.6979	78.358	.6356	116.864
.254	-.394	2.5628	.4696	32.819	.7152	55.629
.082	-.178	3.8443	.1970	-24.766	.2387	-30.506
.070	-.002	5.7665	.0708	-88.308	.0708	-92.368
.004	.008	8.6497	.0094	25.275	.0093	25.045

TABLE XXVIII.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.801  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 1.105 CUBIC FEET PER MINUTE  
 (TEST 2135)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.079	.996	.1000	.9993	-4.550	.5002	-2.276
.118	.991	.1500	.9985	-6.825	.5005	-3.415
.177	.980	.2250	.9967	-10.237	.5011	-5.126
.262	.957	.3375	.9927	-15.351	.5026	-7.703
.384	.905	.5062	.9838	-23.012	.5060	-11.601
.545	.794	.7593	.9641	-34.469	.5139	-17.559
.721	.573	1.1390	.9218	-51.553	.5325	-26.901
.813	.189	1.7085	.8355	-76.913	.5798	-42.526
.614	-.280	2.5628	.6752	65.463	.7136	105.947
.061	-.414	3.8443	.4194	8.418	.7129	14.409
.134	-.003	5.7665	.1346	-88.422	.1339	-96.114
.071	-.002	8.6497	.0714	87.985	.0714	92.077
.091	-.011	12.9746	.0918	-82.560	.0926	-87.829
.093	-.010	19.4619	.0945	83.351	.0951	88.776
.030	.026	29.1929	.0408	-48.840	.0397	-47.124
.020	.005	43.7893	.0214	-75.644	.0213	-74.461
.058	.060	65.6840	.0840	-44.052	.0791	-40.897
.024	.037	98.5261	.0443	-33.158	.0427	-31.819

TABLE XXIX.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.755  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 1.635 CUBIC FEET PER MINUTE  
 (TEST 2245)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.055	.996	.1000	.9983	-3.173	.4997	-1.588
.082	.994	.1500	.9980	-4.760	.4999	-2.382
.123	.989	.2250	.9973	-7.140	.5003	-3.575
.185	.978	.3375	.9958	-10.710	.5011	-5.366
.274	.953	.5062	.9923	-16.062	.5030	-8.061
.401	.898	.7593	.9846	-24.080	.5072	-12.135
.569	.781	1.1390	.9675	-36.080	.5171	-18.348
.752	.547	1.7085	.9302	-53.983	.5407	-28.045
.841	.140	2.5628	.8529	-80.529	.6018	-44.110
.615	-.346	3.8443	.7066	60.596	.7873	103.903
.038	-.476	5.7665	.4776	4.661	.9091	9.899
.211	-.058	8.6497	.2199	-74.538	.2279	-87.229
.001	.080	12.9746	.0804	1.211	.0744	1.121
.028	-.014	19.4619	.0322	62.718	.0327	64.386
.004	-.015	29.1929	.0161	16.061	.0164	16.322

TABLE XXX.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.733  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 2.155 CUBIC FEET PER MINUTE  
 (TEST 2055)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.042	.998	.1000	.9997	-2.425	.5000	-1.212
.063	.997	.1500	.9995	-3.637	.5001	-1.819
.094	.994	.2250	.9989	-5.456	.5002	-2.729
.141	.987	.3375	.9975	-8.182	.5006	-4.096
.211	.971	.5062	.9945	-12.270	.5015	-6.151
.311	.937	.7593	.9879	-18.390	.5034	-9.251
.449	.862	1.1390	.9732	-27.538	.5078	-13.960
.619	.708	1.7085	.9416	-41.168	.5180	-21.230
.770	.420	2.5628	.8774	-61.399	.5431	-32.919
.755	-.021	3.8443	.7557	88.407	.6111	126.063
.362	-.403	5.7665	.5427	41.954	.7774	73.272
.144	-.210	8.6497	.2551	-34.518	.3177	-44.892
.000	.109	12.9746	.1094	.059	.0986	.053
.092	.057	19.4619	.1091	-58.374	.1028	-53.351
.021	-.167	29.1929	.1691	7.283	.2032	8.759
.064	.137	43.7893	.1520	24.971	.1334	21.742
.174	.064	65.6840	.1860	69.572	.1724	60.275
.161	.006	98.5261	.1619	87.535	.1587	78.407

TABLE XXXI.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.796  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 0.452 CUBIC FEET PER MINUTE  
 (TEST 3015)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.164	.970	.1000	.9845	-9.599	.4978	-4.837
.243	.949	.1500	.9801	-14.396	.4989	-7.271
.356	.902	.2250	.9702	-21.586	.5013	-10.958
.507	.801	.3375	.9461	-32.347	.5067	-16.615
.673	.597	.5062	.9002	-48.413	.5192	-25.559
.761	.243	.7593	.7996	-72.239	.5482	-40.764
.580	-.176	1.1390	.6072	73.069	.6027	108.281
.135	-.280	1.7085	.3112	25.824	.4248	36.490
.007	-.050	2.5628	.0515	-8.703	.0543	-9.174
.048	-.019	3.8443	.0524	67.962	.0534	70.803
.010	-.020	5.7665	.0230	-28.240	.0235	-28.878
.057	.070	8.6497	.0906	-39.193	.0845	-36.130
.021	.059	12.9746	.0635	19.355	.0999	18.216
.093	.045	19.4619	.1040	-63.992	.0991	-58.882
.043	.106	29.1929	.1153	-22.040	.1041	-19.801
.034	.115	43.7893	.1201	16.718	.1077	14.942
.083	-.000	65.6840	.0837	89.639	.0834	94.428
.070	-.063	98.5261	.0953	47.991	.1015	52.317

TABLE XXXII.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.760  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 0.723 CUBIC FEET PER MINUTE  
 (TEST 3025)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.102	.989	.1000	.9952	-5.933	.4994	-2.973
.153	.981	.1500	.9934	-8.695	.4998	-4.462
.228	.962	.2250	.9893	-13.329	.5007	-6.700
.334	.921	.3375	.9803	-19.947	.5026	-10.073
.477	.834	.5062	.9609	-29.767	.5070	-15.187
.641	.660	.7593	.9206	-44.172	.5172	-23.046
.765	.557	1.1390	.8451	-64.947	.5421	-35.530
.719	-.061	1.7085	.7216	65.145	.6102	122.591
.397	-.400	2.5628	.5240	40.087	.7621	69.481
.088	-.225	3.8443	.2421	-21.483	.3109	-28.012
.090	-.003	5.7665	.0906	-87.973	.0905	-93.167
.032	.032	8.6497	.0463	-44.866	.0448	-43.054
.004	.022	12.9746	.0230	-11.485	.0225	-11.228
.026	.013	19.4619	.0300	-63.773	.0296	-62.249
.008	.014	29.1929	.0164	30.209	.0162	29.741

TABLE XXXIII.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.798  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 1.110 CUBIC FEET PER MINUTE  
 (TEST 3035)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.067	.997	.1000	.9994	-3.854	.5001	-1.927
.100	.993	.1500	.9987	-5.781	.5003	-2.892
.150	.985	.2250	.9972	-8.670	.5007	-4.341
.223	.968	.3375	.9939	-12.999	.5016	-6.519
.328	.929	.5062	.9863	-19.478	.5038	-9.806
.472	.846	.7593	.9697	-29.150	.5087	-14.803
.642	.677	1.1390	.9340	-43.502	.5199	-22.531
.777	.369	1.7085	.8607	-64.550	.5465	-34.981
.722	-.060	2.5628	.7252	-85.189	.6120	122.768
.346	-.377	3.8443	.5125	-42.480	.7199	71.574
.058	-.240	5.7665	.2476	-13.752	.3251	-18.186
.107	-.006	8.6497	.1081	-86.316	.1083	-92.521
.032	.024	12.9746	.0408	-52.319	.0398	-50.514
.021	.020	19.4619	.0293	-46.870	.0289	-45.659
.007	.024	29.1929	.0251	16.480	.0245	16.080
.005	-.022	43.7893	.0234	-13.136	.0240	-13.446
.070	-.048	65.6840	.0854	-55.546	.0895	-59.782
.023	.007	98.5261	.0245	71.498	.0243	70.176

TABLE XXXIV.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.765  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 1.615 CUBIC FEET PER MINUTE  
 (TEST 3045)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.049	.998	.1000	.9997	-2.821	.5000	-1.410
.073	.996	.1500	.9994	-4.231	.5001	-2.116
.110	.992	.2250	.9987	-6.346	.5004	-3.175
.164	.983	.3375	.9971	-9.517	.5010	-4.765
.244	.962	.5062	.9935	-14.268	.5022	-7.157
.359	.917	.7593	.9856	-21.373	.5051	-10.764
.512	.821	1.1390	.9683	-31.969	.5117	-16.248
.689	.627	1.7085	.9320	-47.678	.5273	-24.729
.812	.282	2.5628	.8602	-70.804	.5665	-36.457
.704	-.187	3.8443	.7294	75.095	.6781	116.043
.210	-.457	5.7665	.5033	24.674	.8649	45.841
.154	-.167	8.6497	.2282	-42.622	.2697	-53.145
.037	.060	12.9746	.0713	31.702	.0672	29.676
.041	-.010	19.4619	.0431	75.641	.0436	78.265
.002	.013	29.1929	.0134	-11.223	.0132	-11.075



TABLE XXXV.

FREQUENCY RESPONSE OF PACKED, TUBULAR REACTOR  
 CALCULATED FROM THE TRANSIENT RESPONSE CURVE  
 RESULTING FROM THE INPUT OF A STEP CHANGE IN  
 THE FEED COMPOSITION FROM ZERO TO 0.752  
 MOLE FRACTION CARBON MONOXIDE IN METHANE AT A  
 FLOW RATE OF 2.055 CUBIC FEET PER MINUTE  
 (TEST 3155)

TERM B	TERM C	OMEGA	OPEN LOOP GAIN	OPEN LOOP PHASE	CLOSED LOOP GAIN	CLOSED LOOP PHASE
		RADIANS PER MIN		DEGREES		DEGREES
.039	.999	.1000	.9998	-2.261	.5000	-1.131
.059	.997	.1500	.9996	-3.392	.5001	-1.696
.088	.995	.2250	.9993	-5.089	.5003	-2.545
.132	.989	.3375	.9984	-7.633	.5007	-3.819
.197	.976	.5062	.9964	-11.447	.5016	-5.733
.292	.947	.7593	.9921	-17.163	.5036	-8.616
.426	.885	1.1390	.9824	-25.721	.5083	-12.976
.598	.752	1.7085	.9611	-38.503	.5191	-19.647
.772	.492	2.5628	.9161	-57.316	.5452	-30.133
.824	.062	3.6443	.8264	-85.636	.6145	-47.649
.529	-.400	5.7665	.6640	-52.871	.8304	94.334
.055	-.409	8.6497	.4130	-7.737	.6961	-13.114
.172	.005	12.9746	.1730	88.041	.1694	78.288
.036	.068	19.4619	.0771	-28.150	.0722	-26.197
.053	-.017	29.1929	.0560	-72.138	.0569	-75.245
.036	-.062	43.7893	.0722	-30.342	.0770	-32.572
.164	.268	65.6840	.3322	29.686	.2557	22.410
.066	-.002	98.5261	.0667	-87.710	.0667	-91.536

loop phase lag, closed loop gain, and closed loop phase lag, as functions of the frequency of the input signal. A summary of the control points of the reactor system, i.e. the corner frequency of the open loop gain, the frequency at which a phase lag of  $-180^\circ$  occurs (open loop and closed loop phase lags were so similar as to defy separation), and the frequency at which the maximum point of the closed loop gain curve occurs, is given in Table XXXVI, p. 88. Plots of open loop and closed loop frequency response are presented in Figure 18 and 19, p. 89 and 90. Three similar tests were summarized on these figures to indicate the accuracy and reproducibility of dynamic response data.

TABLE XXXVI.

SUMMARY OF CORNER FREQUENCIES AND FREQUENCIES OF 180 DEGREES PHASE LAG OF OPEN LOOP FREQUENCY RESPONSE CURVES FOR PACKED TUBULAR REACTOR

TEST NUMBER	FLOW RATE	MOLE FRACTION CO	RESIDENCE TIME	CORNER FREQUENCY	FREQUENCY OF 180 PHASE LAG
	CFH		SEC	CPM	CPM
1115	0.458	0.776	396.7	1.175	1.401
1225	0.720	0.743	266.7	1.360	1.538
1035	1.115	0.789	150.5	1.517	1.690
1245	1.625	0.760	142.7	1.716	1.777
1055	2.190	0.722	110.2	1.878	1.971
2015	0.540	0.815	365.0	1.204	1.424
2225	0.702	0.752		1.336	1.523
2135	1.105	0.801	194.6	1.468	1.614
2245	1.635	0.755	143.6	1.610	1.770
2055	2.155	0.733	116.2	1.641	1.831
3015	0.452	0.796	404.3	1.057	1.396
3125	0.723	0.760		1.343	1.530
3035	1.110	0.798	189.1	1.527	1.717
3045	1.615	0.765	130.6	1.648	1.815
3155	2.055	0.752	108.7	1.749	1.896

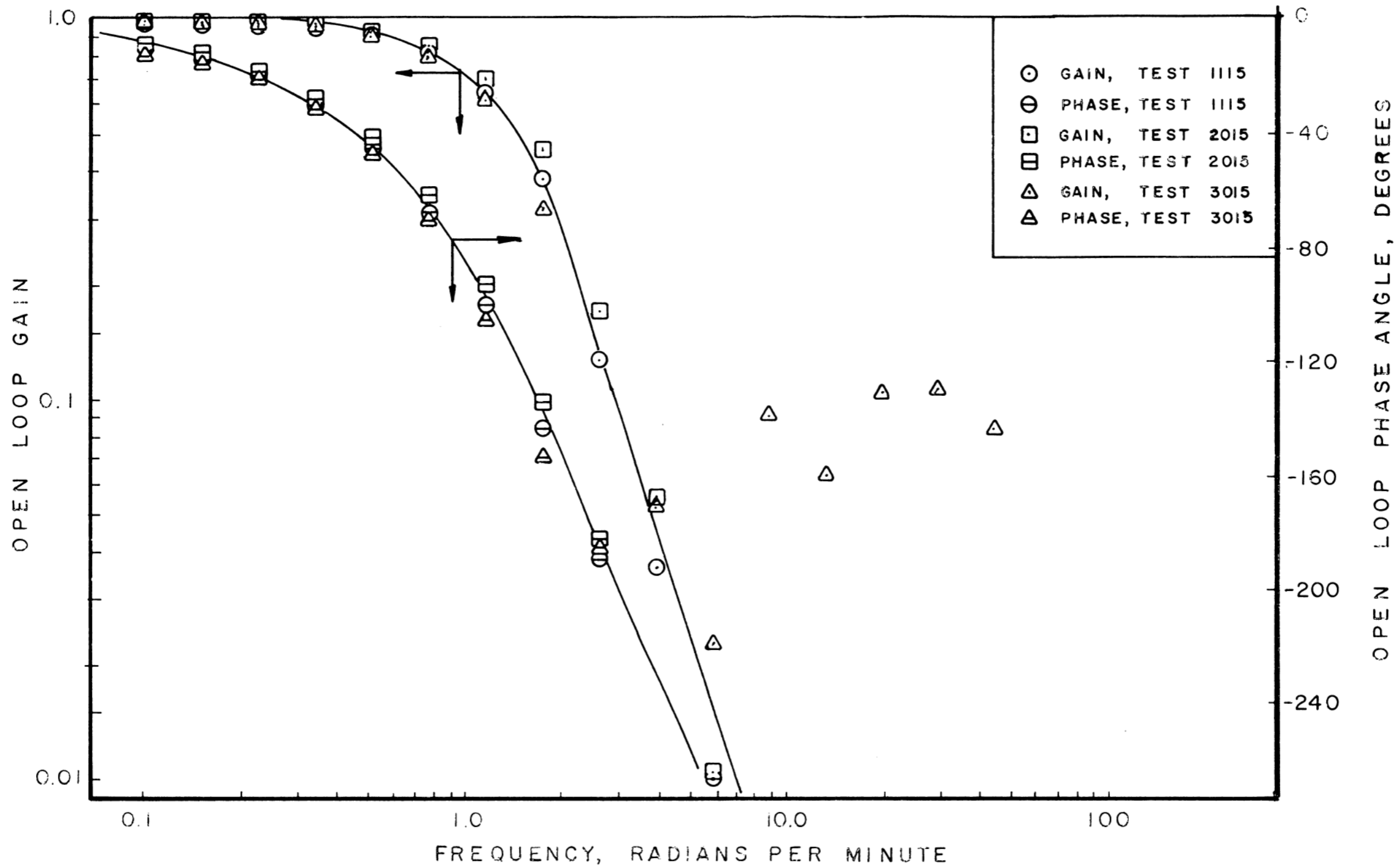


FIGURE 18. FREQUENCY RESPONSE OF A PACKED, TUBULAR REACTOR. OPEN LOOP CHARACTERISTICS FOR FLOW RATES OF APPROXIMATELY 0.5 (ft<sup>3</sup>/hr) OF A MIXTURE CONTAINING 0.8 MOLE FRACTION CARBON MONOXIDE AND 0.2 MOLE FRACTION METHANE

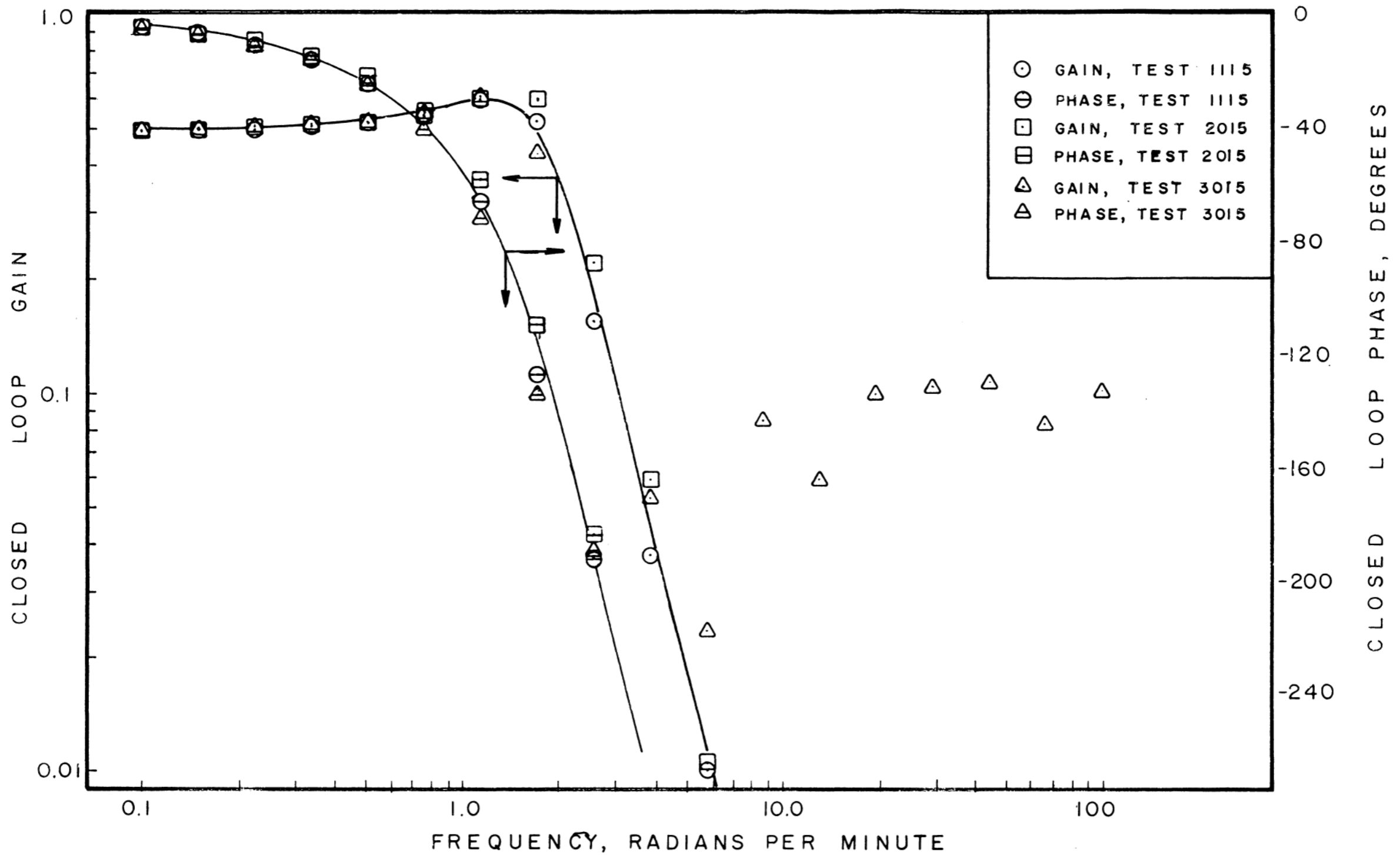


FIGURE 19. FREQUENCY RESPONSE OF A PACKED, TUBULAR REACTOR. CLOSED LOOP CHARACTERISTICS FOR FLOW RATES OF APPROXIMATELY 0.5 (ft<sup>3</sup>/hr) OF A MIXTURE CONSISTING OF 0.8 MOLE FRACTION CARBON MONOXIDE AND 0.2 MOLE FRACTION METHANE

Sample Calculations

The following section contains samples of all calculations made in the study of the dynamic response characteristics of a packed tubular reactor.

Dynamic Residence Time. The transient portion of the response to a step input signal was divided into five second increments, beginning at the point where the first variation from the zero point of the output signal was recorded. The section of the output signal between the time of introduction of the step change in the feed composition and the beginning of the transient portion of the response curve was defined as the mean residence of the gas in the reactor beyond the dead time. Its value was calculated using the data listed in Tables V through XVII as follows:

$$\bar{T} = \frac{\sum t_i (\Delta y_i)}{\sum \Delta y_i}$$

Where:  $\bar{T}$  = the dynamic residence time

$t_i$  = the time beyond "dead time"

$\Delta y_i$  = the fraction of the total concentration change accomplished in the i-th five-second time increment beyond the introduction of the step change

Substituting  $\sum t_i (\Delta y_i)$  and  $\sum \Delta y_i$  from Table VII, page 57.

$$\bar{T} = \frac{96.845}{1.000}$$

$$\bar{T} = 96.845 \text{ seconds}$$

Total Residence Time. The total residence time of the gas in the reactor was determined by adding the value of the dead time to the value of dynamic residence time as follows,

$$\bar{t} = t_d + \tau$$

where:  $\bar{t}$  = Total residence time

$t_d$  = Dead time

$\tau$  = Dynamic residence time

Substituting data from Table XVIII, page 68; for test number 3015:

$$\bar{t} = 307.5 + 96.8 = 404.3 \text{ seconds}$$

Determination of Gas Velocity. The average linear velocity of the gas through the reactor was obtained by simply dividing the length of the reactor column by the residence time of the gas, as follows:

$$u = \frac{L}{\bar{t}}$$

where:  $u$  = average linear velocity of the gas through the reactor

$L$  = the length of the reactor

$\bar{t}$  = the total residence time of the gas in the reactor

Substituting total residence time from Table XVIII, page 68, for test 3015 and the length of the reactor as shown in Figure 7, page 24.

$$u = \frac{5.0}{404.3} = 0.0124 \text{ feet/second}$$

Void Fraction of Bed. The void fraction of the packing in the bed was determined by filling a 1000 milliliter graduated cylinder with the packing material, and then adding measured portions of water until the meniscus reached the 1000 milliliter graduation. The void fraction was then simply calculated by dividing the volume of water added by the total volume of the bed, as follows; from data recorded in Table IV, p. 54 .

$$\epsilon = \frac{V_w}{V_b} = \frac{486}{1000} = 0.486$$

where:  $V_w$  = volume of water added, milliliters  
 $V_b$  = total volume of the bed, milliliters  
 $\epsilon$  = void fraction of the packed bed

Mean Particle Diameter. The mean particle diameter of the packing is given by the expression

$$D_p = \frac{6(1 - \epsilon)}{a}$$

where:  $D_p$  = the mean particle diameter  
 $\epsilon$  = the void fraction of the packed bed  
 $a$  = the surface area per unit volume of the packed bed

Assuming the packing to be made up of spherical particles of 11 mesh, the value of  $a$  was calculated to be 512 square feet per cubic foot of packing, and the mean particle diameter was calculated to be

$$D_p = \frac{6(1 - 0.486)}{512} = 0.00603 \text{ feet}$$



Determination of the Density of the Gas Mixture. The reactor conditions were 333°C. at a pressure of 707 millimeters of Mercury, absolute. Under these conditions, the ideal gas law was assumed to be valid for predicting the density of the gas mixture of methane and carbon monoxide. The molecular weight of the mixture was assumed to be a linear function of the composition,

$$M_{\text{mix}} = X_{\text{CO}} M_{\text{CO}} + X_{\text{CH}_4} M_{\text{CH}_4}$$

$M_{\text{CO}}$  = molecular weight of carbon monoxide

$M_{\text{CH}_4}$  = molecular weight of methane

$X_{\text{CO}}$  = mole fraction of carbon monoxide

$X_{\text{CH}_4}$  = mole fraction of methane

At the reactor conditions one cubic foot of volume is occupied by 0.001167 pound moles of gas, according to the ideal gas law. The density of the gas mixture is consequently given by the expression

$$\rho = n \left( X_{\text{CO}} M_{\text{CO}} + X_{\text{CH}_4} M_{\text{CH}_4} \right)$$

where:  $n$  = the number of pound moles of gas occupying one cubic foot of volume at reactor conditions.

$\rho$  = Density of gas mixture

Substituting data from Table XVIII, test 3015, p. 68, the following result was obtained:

$$\rho = 0.001167 \left[ (0.796) (28) + (0.204) (16) \right]$$

$\rho = 0.0299$  pounds-mass/cubic foot of gas mixture

Determination of Viscosity of Gas Mixture. The viscosity of the gas mixture was calculated by assuming a linear relation of viscosity to composition, based on the viscosities of the pure components. The viscosity of the gas mixture was calculated from the following expression:

$$\mu = 6.72 \times 10^{-4} \left( X_{CO} \mu_{CO} + X_{CH_4} \mu_{CH_4} \right)$$

where:  $\mu$  = viscosity of the gas mixture, lb-mass/foot-sec

$X_{CO}$  = mole fraction of carbon monoxide

$\mu_{CO}$  = viscosity of carbon monoxide at reactor conditions, cp

$X_{CH_4}$  = mole fraction of methane

$\mu_{CH_4}$  = viscosity of methane at reactor conditions, cp

Substituting mole fractions from Table XVIII, p. 68, for test 3015 and viscosity data from Table IV, p. 54.

$$\begin{aligned} &= 6.72 \times 10^{-4} \left[ (0.796)(0.0292) + (0.204)(0.0192) \right] \\ &= 1.83 \times 10^{-5} \text{ lb-mass/ft-sec} \end{aligned}$$

Determination of Reynolds Number. The Reynolds number for fluid flow in a packed bed is given by the expression:

$$N_{Re} = \frac{D_p u \rho}{\mu}$$

Where:  $N_{Re}$  = Reynolds Number

Substituting the values for the terms as calculated for test 3015, from data in Table XVIII, page 68;

$$N_{Re} = \frac{(0.00603)(0.0124)(0.486)(0.0299)}{(1.45 \times 10^{-5})}$$

$$N_{Re} = 0.0750$$

Determination of the Dispersion Number. The dispersion number was determined from the variance of the rate of change of the concentration of the product stream with respect to time (C curve). The relationship between dispersion number and the variance of C curve is as follows:

$$\sigma^2 = \frac{2D}{u_0 L} + 2 \left( \frac{D}{u_0 L} \right)^2 (1 - \exp(u_0 L/D))$$

When the value of  $D/u_0 L$  is less than 0.01, the second order and exponential terms become insignificant, and the dispersion number becomes simply

$$\frac{D}{u_0 L} = \frac{\sigma^2}{2}$$

The variance term is defined by the expression

$$\sigma^2 = \frac{\sigma_t^2}{\bar{t}^2}$$

where:  $\sigma^2$  = variance in dimensionless terms

$\sigma_t^2$  = variance as a function of time

$\bar{t}$  = holding time

The variance as a function of time is defined by the expression

$$\sigma_t^2 = \left[ \frac{t_i^2 (\Delta y_i)}{\Delta y_i} \right] - \left[ \frac{t_i (\Delta y_i)}{\Delta y_i} \right]^2$$

Substituting data from Table VII, page 57, Test 1115,

$$\sigma_t^2 = 11449.3250 - 9378.9540$$

$$\sigma_t^2 = 2070.37$$

The dispersion number may now be readily obtained, using the total residence time from Table VI, page 56.

$$\frac{D}{u_o L} = \frac{\sigma^2}{2} = \frac{\sigma_t^2}{2\bar{t}^2}$$

$$\frac{D}{u_o L} = \frac{2070.37}{2(404.35)^2} = 0.00633$$

Determination of Schmidt Number. The Schmidt number for the carbon monoxide - methane system was calculated by use of the Gilliland equation,

$$D = 0.0166 \frac{T^{3/2}}{P(V_1 + V_2)^2} \sqrt{\frac{1}{M_1} + \frac{1}{M_2}}$$

where:  $D$  = diffusivity of the gases in each other, for equimolar counter-diffusion square feet per hour

$T$  = temperature,  $^{\circ}K$

$P$  = pressure in atmospheres

$M_1, M_2$  = molecular weights of the gases

$V_1, V_2$  = molecular volumes at the normal boiling points ml/gm-mole

Using the value of the diffusivity determined above, the Schmidt number is readily calculated, using the density and viscosity values determined during the calculation of Reynolds number.

$$N_{Sc} = \frac{\mu}{D\rho}$$

For Test 1245, the following determination was made, using data from Table IV, p. 54, and Table XIX:

$$D = 0.0166 \frac{(333+273)}{\frac{707}{760} (29.6 + 22.2)} \sqrt{\frac{1}{16} + \frac{1}{28}}$$

$$D = 2.43 \text{ ft}^2/\text{hr}$$

$$N_{Sc} = \frac{(1.83 \times 10^{-5})(3600)}{(0.0299)(2.43)} = 0.907$$

Determination of Frequency Response. The frequency response of the system was calculated from the transient portion of the output signal plot. Teasdale's<sup>(18)</sup> method of the addition of vectors was used, and the actual calculations were performed by the IBM 1620 computer. The FORTRAN program used to program the computer is given in Table XIX, p. 70. The calculation of a single point, representing the response of the system under the conditions of test 1245, to an impressed signal with a frequency of 0.1 radians per minute is given in Table XXXVI.

The transfer function of the system was calculated from points of the output signal by the following series of operations:

$$y_i = \frac{C_i}{C_n} = \left( \frac{E_i - E_o}{E_n - E_o} \right) 0.798$$

where:  $C_i$  = Mole Fraction CO at the end of  $i$  time periods

$C_n$  = Mole Fraction Carbon monoxide at steady state condition after step change introduction

$E_i$  = Chart reading of output signal at the end of  $i$  time periods

$E_0$  = Chart reading corresponding to zero mole fraction carbon monoxide

$E_n$  = Chart reading corresponding to steady state concentration of carbon monoxide

$y_i$  = Fraction of total concentration change after  $i$  time periods

The change in the value of  $y$  in the  $i$ -th time period is given by

$$\Delta y_i = y_i - y_{i-1}$$

where:  $\Delta y_i$  = the change of  $y$  during the  $i$ -th time period

The transfer function of the system is given by the expression

$$GH = y_i / \underline{-\omega \Delta t k_i}$$

where:  $GH$  = the transfer function of the system

$\omega$  = the frequency of the input signal, radians per min

$$k_i = (2i-1)/2$$

$\Delta t$  = the time increment between measurements of output signal, 5.0 seconds

If the polar equation is transformed into rectangular coordinates with one real and one imaginary axis, the following expression is obtained:

$$GH = \sum \Delta y_i \cos(\omega \Delta t k_i) - j \sum \Delta y_i \sin(\omega \Delta t k_i)$$

If the terms  $\underline{B}$  and  $\underline{C}$  are now defined as

$$B = \sum \Delta y_i \sin(\omega \Delta t k_i)$$

then  $GH = C - jB$ ,

and the following characteristics of the frequency response may be calculated:

Open Loop Gain  $M = \sqrt{B^2 + C^2}$

Open Loop Phase Lag  $\phi = -\tan^{-1} \left( \frac{B}{C} \right)$

Closed Loop Gain  $M' = \sqrt{\frac{C^2 + B^2}{(1+C)^2 + B^2}}$

Closed Loop Phase  $\alpha = \phi - \tan^{-1} \left( \frac{B}{1+C} \right)$

Where:  $M$  = open loop gain  
 $\phi$  = open loop phase lag  
 $M'$  = closed loop gain  
 $\alpha$  = closed loop phase lag

For test 1245, at a frequency of 0.100 radians per minute, the following results were calculated from data in Table XX, p. 71.

$B = 0.054$

$C = 0.998$

$M = \sqrt{(0.054)^2 + (0.998)^2} = 0.997$

$\phi = -\tan^{-1} (0.054/0.998) = -3.115 \text{ degrees}$

$M' = \sqrt{\frac{(0.054)^2 + (0.998)^2}{(1+0.998)^2 + (0.054)^2}} = 0.5001$

$\alpha = -3.115 - \tan^{-1} \left( \frac{0.054}{1+0.998} \right) = -3.087 \text{ degrees}$

Determination of Corner Frequency. The corner frequency of frequency response was determined graphically from the plot of open loop gain versus frequency. The slope of the constant rate of decrease in gain was extended back to the gain datum, as shown in Figure 18, p. 89. The corner frequency for the average of Tests 1115, 2015, and 3015 was found to be

$\omega_0 = 1.3 \text{ radians per minute}$

#### IV. DISCUSSION

The following section contains a discussion of the procedures used in the investigation, a discussion of the results obtained, a listing of the limitations imposed upon the applicability of the results, and recommendations for further studies.

##### Discussion of Procedures

A brief discussion of the procedures followed in the study is presented in the following paragraphs.

Reactor Temperature Control. The temperature of the reactor was maintained constant at approximately 333° C. As mentioned in the Method of Procedure section, the allowable variation at measured-temperature points was two degrees C. The cal-rod type heaters used were wrapped around the reactor tube, each heater completing three wraps. Two of the heater elements were positioned near the extreme base of the reactor to provide the necessary heat to increase the temperature of the feed stream from 54° C to 333° C in as short a distance as possible. This arrangement was apparently satisfactory, because the first temperature measurement, made in the center of the tube, six inches from the gas inlet, indicated the control temperature and maintained it without fluctuation even in the cases when feed flow rates



in the range of five cubic feet per hour were being used. The quartz packing greatly extended the heat transfer area of the reactor, and the problem of temperature control would be much more difficult in an empty tube. It would be interesting to measure temperatures along the reactor wall, to see how they compare with those measured along the axis of the tube.

Introduction of Step Change. The step change in the feed composition was effected by the addition of carbon monoxide to a stream of methane. No particular advantage would have been obtained by using an initial concentration of carbon monoxide other than zero, because the only added factor would be the self-diffusion of carbon monoxide in itself. The step change was made by opening the needle valve which controlled the carbon monoxide flow rate and adjusting the flow to a predetermined value as quickly as possible. In practice, about two seconds were required to make the change. For this reason, the assumption of a perfect step input was fairly well justified. The sampling frequency utilized in the transformation of the continuous output from the thermal conductivity cell into discrete increments for numerical analysis was one sample every five seconds. For very accurate work, it would be preferable to measure the shape of the input wave. The sampling frequency should also be increased to depress the error inherent in the approximation of a continuous function by linear interpolation between measured points.

Analysis of Gas Stream. The thermal conductivity cell was quite sufficient for the analysis made. Its response was well within the limits of error set by the sampling frequency and the imperfection of the input signal. The cell had a time constant of only one second. This value amounts to less than one per cent of the total residence time of the gas for the tests made in the study. The flow rate of the gas through the thermal conductivity cell determined the output signal, but the magnitude of the output step response was independent of the flow rate of gas through the cell. This fact greatly simplified the analysis of the data, eliminating the need for adjusting each output curve for the effect of flow rate. The relationship between the output signal and the composition was not linear with respect to the mole fraction of carbon monoxide, but an exponential relationship was determined which allowed a mathematical statement of the correlation of the scale reading change with the composition change. This relationship was very useful in the determination of frequency response, because a digital computer was used in the calculation of frequency response.

Accuracy of Analysis. The first attempt at the calculation of the dispersion number as a function of Reynolds number was made by plotting the normalized, corrected transient composition versus time curve on probability paper. The points of the curve did not lie on a completely straight line,

but the slope of the line between 16 and 84 per cent of the total composition change could be fairly well represented by a straight line. The time interval intercepted by the straight line and the 16 and 84 per cent lines represented two standard deviations. From the value of the standard deviation, the dispersion number could be evaluated fairly easily. The method seemed ideal, until it was observed that slight errors in the estimation of the slope of the curve could represent as much as thirty per cent of the total range of dispersion numbers studied. This error was deemed unacceptable, and the variance was determined numerically.

The accuracy of the analysis leans heavily on the degree of accuracy with which the discrete points can be read from the output signal recording. Obtaining three significant figures involved the estimation of the position, between lines ten units apart, of a line two units wide, to an accuracy of one unit.

Temperature Control of the Analysis Cell. The thermal conductivity cell was maintained at a constant temperature of 80° C by the use of an air bath whose temperature was controlled by a thermoregulator with a sensitivity of plus or minus 0.05° C. The air in the bath was kept stirred by means of a small fan driven by a capacitor run electric motor. The thermal conductivity cell was contained within an aluminum box with 3/8-inch walls, which acted as a heat sink to provide a very stable temperature. The gas streams

fed to the analysis cell were brought to cell temperature by passing them through 30 inches of 3/16-inch tubing within the air bath prior to their entrance into the thermal conductivity cell.

Calculation of Dispersion Number. The composition changes as calculated from the output signal from the thermal conductivity cell were normalized as fractions of the total composition change at five second intervals in the transient region of the output signal. Because the time increments were of equal length, the normalization of the time axis with respect to the total residence time was reserved until after the calculation of the variance of the  $C$  curve. This procedure allowed the terms  $\underline{t}_i$  and  $\underline{t}_i^2$  to be listed once and used in each of the dispersion number determinations. The variance in dimensionless units was calculated by dividing the variance obtained above by the square of the total residence time.

### Discussion of Results

The following section is a discussion of the results obtained, including their reproducibility and comparison with published information.

Dispersion Number. The dispersion number of a system is a function of the velocity of the fluid through the channel, the length of the section of channel being studied, and the dispersion coefficient. The dispersion coefficient

is called eddy diffusivity by some authors, and is analogous to diffusivity, the driving force being fluctuations in the flow velocity, rather than concentration gradients as in diffusion. A correlation was made by a least squares analysis of the plot of log Reynolds number versus log dispersion number. The range covered was dispersion numbers from 0.00976 to 0.00390 at Reynolds numbers from 0.0599 to 0.2151, respectively. The slope of the correlation line, Figure 17, p. 69, was 0.643.

Levenspiel<sup>(13)</sup> presented a generalized chart of Reynolds number versus dispersion number. The slope of the predicted correlation in the range of Reynolds numbers studied was minus one, but the experimental data shown were presented in the form of hazy regions as functions of the Schmidt number. The hazy area representing Schmidt numbers of 0.2 lay on the line with a slope of minus one, but the apparent slope of the correlation line decreased (approached zero) with increasing Schmidt number. The highest indicated values of Schmidt number shown were in the range of 1.5. Inference was made from the chart that Schmidt numbers between 0.2 and 1.5 would produce slopes of correlation between minus one and zero, respectively. The results obtained in the investigation demonstrated this trend. A portion of Levenspiel's correlation chart is shown in Figure 20, p. 107, with the results of this investigation superimposed to demonstrate the degree of agreement.

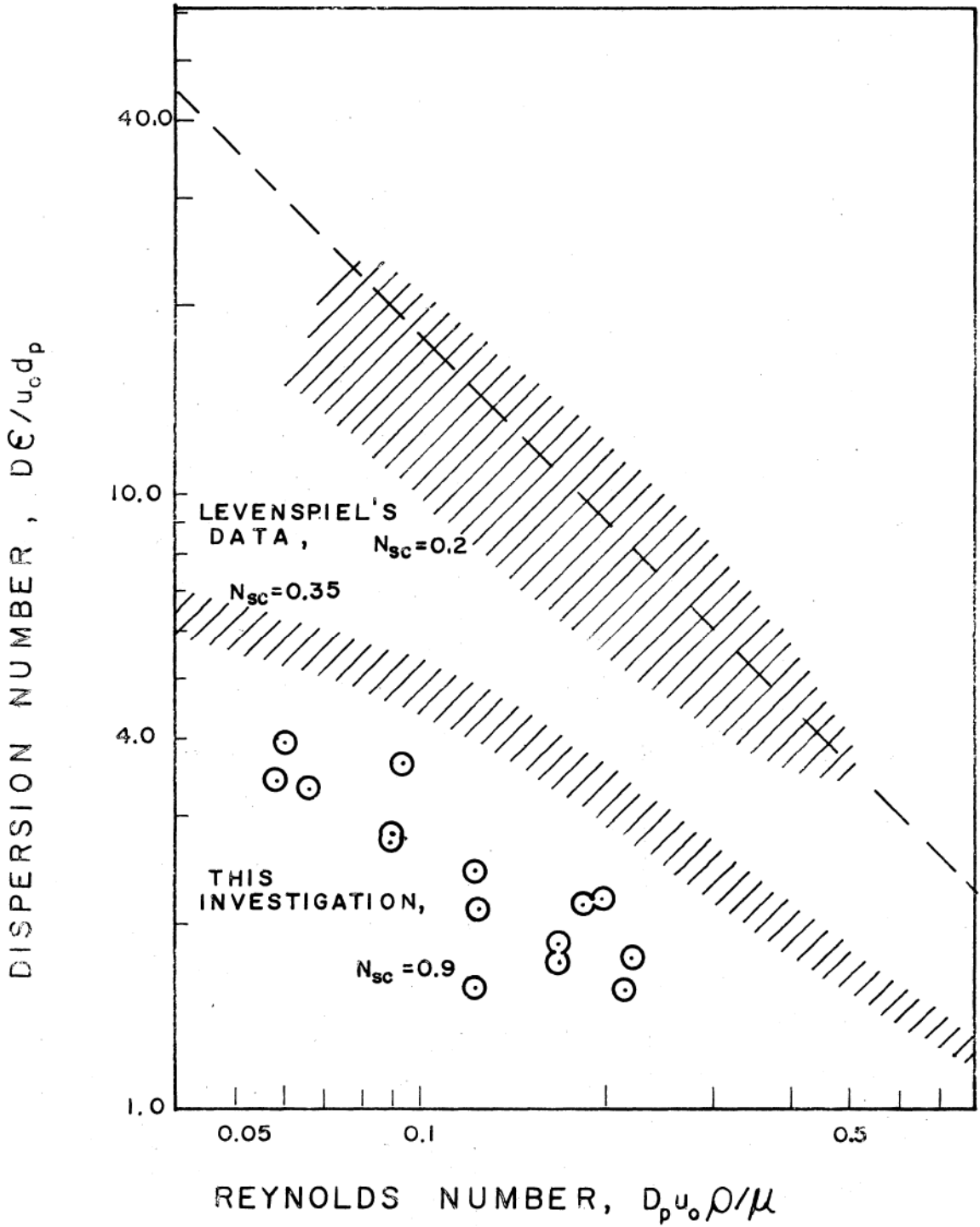


FIGURE 20. COMPARISON OF RESULTS

Levenspiel, O.: "Chemical Reaction Engineering," p.275, John Wiley & Sons, New York, N.Y., 1962.

Effect of Schmidt Number. Because the range of value of Schmidt numbers studied was only from 0.907 to 0.918, no significance can be attached to variation of dispersion number with Schmidt number. Changing the conditions of the reactor would produce only insignificant changes in the Schmidt number for the existing gas mixture, but the use of other gas mixtures could alter its value.

Effect of Reynolds Number. Although the results indicated that the dispersion number decreased with increasing Reynolds number, the quantitative aspects of the correlation are marred by the relatively small range of Reynolds numbers studied. In order to completely describe the system, it would be advisable to increase the range of Reynolds numbers studied. This could be accomplished by increasing the flow rate of the gas stream, increasing the pressure of the reactor, or by using other components in the feed stream.

Frequency Response. For the series of frequency response curves calculated, the general characteristics were those of a second order, overdamped system. The slope of the open-loop gain curve was in the range of 40 decibels per decade in its constant rate of descent portion. The phase lag plots demonstrated typical second order lags, coupled with the effects of dead time. The second order lag will approach  $-180^\circ$  as a minimum point, but the dead time curve will produce an overall curve similar to those calculated. Typical of the response curves calculated are

those shown in Figure 18, p 89. In all cases studied, the determining factor in the control of the reactor system would be the gain characteristic, the corner frequency being less than the frequency at which  $180^\circ$  of phase lag occurs. It bears mentioning, however, that the phase margin (difference between the phase lag at the corner frequency and the phase lag of  $180^\circ$ ) is quite small in some cases. Under the conditions which cause these small values of phase margin, the system would be extremely difficult to control.

Reproducibility of Frequency Response Data. The frequency response data were reproducible to within five to eight per cent. The major problem in reproducing or of even accurately obtaining dynamic data hinges on the accurate conversion of continuous output data into discrete points. The spacing of the sample points is also of interest. In this investigation a sampling interval of five seconds was used. This fact automatically invalidated any frequency response information which was calculated for frequencies greater than twelve radians per minute, based on the limiting conditions of  $\omega \Delta t$  equal to one. Decreasing the sampling interval to two seconds would allow frequencies up to thirty radians per minute. It is also advantageous to determine frequency response at a number of frequencies near the corner frequency, in order to decrease errors introduced by extrapolation of data points.



### Recommendations

Based on the results obtained from the investigation of the dynamic response characteristics of a packed tubular reactor at  $333^{\circ}$  C and a pressure of 707 millimeters of mercury, absolute, to step changes in the feed composition from zero to approximately 0.80 mole fraction of carbon monoxide in methane, at flow rates of from 0.45 to 2.2 cubic feet per hour, the following recommendations are made.

1. It is recommended that the work begun in this thesis be extended to cover the determination of dispersion numbers in the transition and turbulent flow regimes, over a range of Reynolds numbers from 1.0 to 100.

2. It is recommended that the effect of Schmidt number on the dispersion number be studied, using Schmidt numbers from 0.2 to 1.5. Since the Schmidt number of a given gas system is relatively independent of temperature and pressure, it will be necessary to choose systems other than the one studied.

3. It is recommended that additional thermal conductivity analysis cells be used to analyze the feed stream to the reactor and perhaps intermediate streams, with the purpose of increasing the accuracy of the measurement of dispersion.

### Limitations

The following limitations are imposed on the applicability of the results and conclusions of this investigation.

Reactor. The reactor was a five foot length of three-inch schedule 40 pipe, packed with crushed quartz of eight to 14 mesh.

Operating Conditions. The study was made at a reactor temperature of  $333^{\circ}$  C and a pressure of 707 millimeters of mercury, absolute. Flow rates of feed ranged from 0.45 to 2.2 cubic feet per hour, measured at  $54^{\circ}$  C and 707 millimeters of mercury absolute.

Sampling. A sample stream from the product stream of the reactor was passed through a thermal conductivity cell at  $80^{\circ}$  C, and the output from the thermal conductivity cell was recorded on a millivolt recorder.

Analysis of Data. The continuous data were transformed into a series of discrete data points by visual measurement, at five second time increments. Calculation of dispersion numbers was performed on a desk calculator, and an IBM 1620 computer was utilized for the calculation of frequency response.

Assumptions. It was assumed that a constant temperature existed within the reactor, and also within the analysis cell.

## V. CONCLUSIONS

Based on the studies made of the dynamic response characteristics of a packed tubular reactor to step changes in the mole fraction of carbon monoxide in methane used as the feed stream to the reactor, at a temperature of 333° C and a pressure of 707 millimeters of mercury, absolute, the following conclusions were reached.

The reactor system with its accompanying thermal conductivity produces dynamic response data with an accuracy of plus or minus five per cent. The greatest deviation of reproducibility was measured to be eight per cent, based on the calculation of frequency response.

The correlation between Reynolds number and dispersion number was found to be

$$\frac{D}{u_o L} = 0.00165 \left( \frac{D_p u_o \rho}{\mu} \right)^{-0.643}$$

in the range of Reynolds numbers from 0.06 to 0.22.

## VI. SUMMARY

An investigation was undertaken to design, construct, and prove suitable for dynamic response studies a tubular flow reactor.

The reactor was designed and constructed of stainless steel to provide maximum flexibility in its application to various reaction systems, and was provided with a temperature control system capable of maintaining a constant temperature of  $333^{\circ}$  C. A thermal conductivity cell was used for the analysis of the composition of the product stream from the reactor, and the composition of the feed was calculated from the readings of calibrated rotameters in the feed component lines. The reactor was packed with crushed quartz of 8 to 14 mesh.

No reaction was studied, but the dynamic characteristics of frequency response and dispersion number were determined from the analysis of the response of the system to step changes in the composition of a feed stream consisting of carbon monoxide in methane.

Data were found to be reproducible to within eight per cent, and the relationship between Reynolds number and dispersion number for Reynolds numbers from 0.587 to 0.2190 was found to be

$$\frac{D}{u_o L} = 0.00165 \left( \frac{D_p u_o \rho}{\mu} \right)^{-0.643}$$

where  $D$  = dispersion coefficient  
 $u_o$  = superficial gas velocity, based on the empty cross sectional area of the reactor  
 $L$  = length of the test section of the reactor  
 $D_p$  = average diameter of packing particles  
 $\rho$  = density of the gas mixture

at a Schmidt number of 0.907 to 0.915

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## ABSTRACT

An investigation was undertaken to design, construct, and prove suitable for dynamic response studies, a tubular flow reactor.

The reactor was designed and constructed of stainless steel to provide maximum flexibility in its application to various reaction systems, and was provided with a temperature control system capable of maintaining a constant temperature of up to  $333^{\circ}$  C. A thermal conductivity cell was used for the analysis of the composition of the product stream from the reactor, and the composition of the feed stream was calculated from the readings of calibrated rotameters in the feed component lines. The reactor was packed with crushed quartz of 8 to 14 mesh.

No reaction was studied, but the dynamic characteristics of frequency response and dispersion number were determined from the analysis of the response of the system to step changes in the composition of the feed stream, which consisted of carbon monoxide in methane.

Data were found to be reproducible to within eight per cent, and the relationship between Reynolds number and dispersion number for Reynolds numbers from 0.0587 to 0.2190 and Schmidt numbers of 0.907 to 0.915 was found to be

$$\frac{D}{u_0 L} = 0.00165 \left( \frac{D_p u_0 \rho}{\mu} \right)^{-0.543}$$

Where:

- D = dispersion coefficient
- $u_0$  = superficial gas velocity, based on the empty cross-sectional area of the reactor
- L = length of the test section of the reactor
- $D_p$  = average particle diameter of packing
- $\rho$  = density of gas mixture
- $\mu$  = viscosity of gas mixture