AN APPARATUS FOR MEASURING THE RATE OF DIFFUSION OF

GASES THROUGH POROUS SOLIDS AT ELEVATED PRESSURES

by

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ABSTRACT

An apparatus utilizing a constant pressure flow system was developed for measuring the effect of pressure on the diffusion rates, and therefore diffusion coefficients, of binary gas mixtures passing through porous solids.

Hydrogen and nitrogen were employed for testing ceramic porous solids at room temperature, with various pressures from 1 to 14.6 atmospheres absolute. The values obtained for the products of the effective diffusion coefficients and absolute pressures were substantially constant, with a maximum deviation of $\pm 5\%$. It seemed that the diffusion rate of hydrogen increased with pressure, while that of nitrogen decreased. At atmospheric pressure the ratio of diffusion rates (N_{H2}/N_{N2}) was in good agreement with the theoretical value proposed by Hoogschagen i.e. $\sqrt{M_{N2}/M_{H2}}$. However, the experimental diffusion ratio increased with pressure. This behavior might be due to some degree of forced flow present in the diffusion process, although it was not possible to determine a cause for such a flow.

This apparatus is suitable for the study of diffusion rates in the transition region, between Knudsen and ordinary diffusion, by simply changing pressure and hence the mean free path of the gases involved. Forced flow would not be a factor in this region.

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NOMENCLATURE

A	Cross-sectional area of porous sample
A, B,	Molecular species
A_A, A_B	Mole fraction of A in A stream, B stream
C_A, C_B	Molecular concentrations of A, B
C_T	Total Molecular concentration
D_{AB}	Diffusion coefficient of system A-B
D_B	Bulk (ordinary) diffusion coefficient
De	Effective diffusion coefficient
D_K	Knudsen diffusion coefficient
D_T	True ordinary diffusion coefficient
H_H, H_N	Mole fractions of hydrogen in hydrogen stream, in nitrogen stream
k	Boltzman Constant
K	A constant equal to $\frac{RTL}{A}$
M_A, M_B	Molecule weights of A, B
N_A, N_B	Diffusion rates, in moles per unit time
N_{LM}	Log mean of N_A and N_B
P	Absolute pressure
p_A, p_B	Partial pressures
r	Pore radius
T	Temperature
U_A, U_B	Linear velocities of A, B
$\bar{v}$	Mean molecular velocity
V_H^H, V_N^N	Rates of diffusion in mls per unit time
(1)	Inlet

X_H, X_N	Flow rates of hydrogen, nitrogen to diffusion cell
YH, YN	Outlet flow rates of hydrogen, nitrogen from the diffusion cell
β	A proportionality factor of interdiffusion
ϵ_{AB}	Maximum energy of attraction between AB
$\left[\Omega_{AB}^{(1\cdot1)*}(T_{AB}^{*})\right]$	Collision Integral
σ_{AB}	Collision diameter for the unlike molecules A and B
θ	Porosity
ρ, ρ_A, ρ_B	Densities

TABLE OF CONTENTS

LIST OF ILLUSTRATIONS

IN TEXT

LIST OF TABLES

INTRODUCTION

It was desired to develop an apparatus that would allow measurements to be made of the effect of pressure on the diffusion rates, and therefore diffusion coefficients, of binary gas mixtures passing through porous solids. A constant pressure flow system operating in the steady state with counter diffusion of the two gases was preferred for several reasons. These reasons will be discussed in more detail later, but briefly they may be stated as,

(1) The majority of industrial processes in which gas-porous solid contacting is involved operate at essentially constant total pressure with a counter diffusion process occurring, and therefore, it might be worth while to simulate this type of behavior, if it can conveniently be done.

(2) By varying the pressure the mean free path of the gas mixture can be readily changed over wide ranges. This would allow the transition region between Knudsen and ordinary molecular diffusion to be explored more easily, and allow several relationships proposed for diffusion rates in this region to be checked.

(3) Some aspects of counter diffusion under constant total pressure could be investigated at other than 1 atm. absolute pressure.

(4) If the pressure range covered could be made sufficiently large, the variation of the ordinary diffusion

coefficient with pressure could be measured by a new method.

A variety of porous solids would probably be required to achieve all of these objectives. However, a suitable porous solid can usually be found for any specific purpose.

A description of an apparatus of the above type is given in this thesis, together with fairly extensive tests of its performance, using nitrogen-hydrogen mixtures as the test binary pair. These gases were selected because of their ideal nature, and the ease of analysis by thermal conductivity cells. At $20^\circ C$, ideal behavior is obtained for both gases at pressures up to 60 atm.

The apparatus was constructed for a maximum pressure of 300 psia., but was actually operated to a maximum pressure of only 200 psia. at $20^\circ C$. With replacement of flowmeters, the apparatus was capable of being used to 1200 psi.

Some improvements in either measuring technique or diffusion cell design appear to be necessary if the apparatus is to operate in a simpler and more precise manner. However, accurate and reproducible results can be obtained by this method, as demonstrated by the results of diffusion measurements.

THEORY

A.   Diffusion Mechanisms in Porous Solids

The diffusion coefficients of gases at higher
pressures are desirable to know because of the increase
in importance of high pressure catalytic reactions. It
is well known that direct measurements of the diffusion
of gases through porous substances can often provide
valuable information on the rate and the mechanism of the
processes taking place.

Diffusion of a component of a gaseous mixture through
a porous solid may proceed by four possible mechanisms of
transport, namely bulk diffusion, i.e. mean free path of
gas molecules is less than pore size; Knudsen diffusion,
i.e., mean free path of gas molecules is greater than pore
size; surface diffusion in a mobile adsorbed layer, and
forced flow in pores due to the presence of a total
pressure difference.

In general, the transport of gas molecules through a
porous solid, due to bulk diffusion or Knudsen diffusion,
or to both, is of the most interest in this work. The
other two mechanisms may be eliminated from the experiments
if the diffusion is carried out well above the critical
temperature of the gas components, and in the absence of
any total pressure difference.

From a broad mechanistic stand point, molecules are
transported into the inner pore structure of a solid by

their random kinetic motions. If there were no "resistance" to this transport, molecules could diffuse into and through a porous solid with velocities approaching the average molecular velocity. The actual rate of diffusion is many orders of magnitude slower than this due to two causes: (a) collisions with pore walls, and (b) collisions with other molecules.

It is clear that Knudsen diffusion happens when the resistance is caused by molecular collisions with pore walls, i.e. pore size is approximately ten times less than the means free path. In this instance, the diffusion of a particular molecular species at 1 atmosphere in small pores (e. g. less than 400 Å diameter) is independent of the presence or absence of other types of molecules, and depends only on the partial pressure gradient of that particular species. It has been shown that the Knudsen [1][2] diffusion coefficient, D_k, for a molecular species in a pore is a function of pore size and the mean molecular velocity, i.e.

$$D_k = \frac{2}{3} r \bar{v} \qquad (1)$$

where $\bar{v}$ is the average molecular velocity, and r is the pore radius.

For bulk diffusion in a pore, the mean free path λ is much smaller than the pore size, and a molecule within the pore structure will collide with other molecules far more often than with the pore wall. Hence, the bulk

diffusion coefficient D_B will be independent of the pore size and will have the value of the ordinary diffusion coefficient D_T. The simple kinetic theory gives the following approximate formula for the diffusion coefficient for a mixture of two gases of similar mass and molecular diameter,

$$D_B = \frac{1}{3} \bar{v} \lambda \qquad (2)$$

where $\lambda = \frac{0.707}{\pi \sigma^2 C_T} \qquad (3)$

σ = molecular diameter

C_T = total concentration in molecules per C.C.

$\bar{v}$ = average molecular velocity

λ = mean free path of gas

Wheeler[2] has presented several semiempirical, over-all equations for the diffusion coefficient in the transition region between ordinary and Knudsen diffusion, one such equation, for example, is

$$D = \frac{1}{3} \bar{v} \lambda (1 - e^{-\frac{2r}{\lambda}}) \qquad (4)$$

This equation has the property of giving equation (1) when r is less than $\frac{\lambda}{10}$ and of giving equation (2) when r is much greater than λ. The physical problem is concerned with relative probability of a molecule striking the pore wall vs. striking a second molecule. In equation (4) the bracketed expression is exactly the probability that a molecule will have a collision with a second molecule before travelling a distance equal to the pore diameter. As far as can be determined, none of these equations for the

transition region have ever been checked by any experimental data.

These equations (1) (2) and (4) cannot be used at present for calculating the effective diffusion coefficient, but only give a qualitative explanation of the mechanism of gas diffusion through porous solid.

Therefore, for a porous solid the effective diffusion coefficient (D_e), which may be made up of contributions mainly from the discussed two mechanisms, can only be obtained by means of experimental measurements. The effective diffusion coefficient (D_e) is always smaller in value than the true bulk diffusion coefficient (D_T) because of the porosity, pore geometry etc. of the porous solid as stated by Petersen[3] and Cox[4]. The ratio of D_e and D_T is defined as the diffusion ratio for a certain porous solid, and is constant for any gas pair if there is no surface diffusion or forced flow present.

It should be possible to select a porous solid, a gas pair, and a set of conditions of temperature and pressure in order to study the process of bulk diffusion alone, or Knudsen diffusion alone, or a combination of the two in the transition zone.

In particular, as the mean free path varies inversely as the molecular concentration, and therefore absolute pressure, one of the easiest ways to obtain variation in diffusive behavior might be to measure diffusion rates at varying pressures.

B.   Counter Diffusion of Gases at Constant Total Pressure

The theory of diffusion rates in gases was developed as a part of the kinetic theory, and is due principally to Maxwell[5] and to Stefan[6], with later important contributions from O. E. Meyer[7], Sutherland[8], Langevin[9], Chapman[10], Enskog[11], and Jeans[12]. Briefly, for a binary solution of substances A and B which is not of uniform composition, there will be an inter-diffusion of the substances which can be described in terms of the linear velocities of movement of A and B. It is assumed that the drop in concentration of substance A, $-dC_A$, which acts as a driving force for the movement of A, is proportional to the relative linear velocity of A with respect to B, $U_A - U_B$; to the molecular concentrations of the two substances, C_A and C_B; and to the distance dL through which the diffusion occurs. Mathematically, this gives the well-known Maxwell equation,

$$-dC_A = \beta\ C_A\ C_B\ (U_A - U_B)\ dL \tag{5}$$

where β is a proportionality factor of interdiffusion.

This basic equation can now be treated to describe the various situations which may arise.[13] In the case of gases,

$$C = \frac{\rho}{M} \tag{6}$$

and ρ density

M   molecular weight

Equation (5) then becomes, using (6) and the ideal gas law,

$$- dP_A' = \frac{1}{RT}\beta \, \frac{P_A \cdot P_B}{M_A \cdot M_B} \, (U_A - U_B) \, dL \tag{7}$$

Define N′ as the number of moles of gas diffusing per time, per unit area in a direction perpendicular to that of the diffusion.

$$N = \frac{\rho U}{M} \tag{8}$$

Applying the ideal-gas law, then

$$- dP_A = \frac{\beta}{RT} \, (N_A' P_B - N_B' P_A) \, dL \tag{9}$$

Since

$$P = P_A + P_B \tag{10}$$

$$- dP_A = \frac{\beta}{RT} \, (N_A' P - N_A' P_A - N_B' P_A) \, dL \tag{11}$$

Let

$$D_{AB} = \frac{R^2 T^2}{\beta P}$$

So that

$$- dP_A = \frac{RT}{D_{AB} P} \, (N_A' P - N_A' P_A - N_B' P_A) \, dL \tag{12}$$

For a steady state diffusion, the total pressure, P, is constant, so are the diffusion rates, N_A and N_B. Integrating Equation (12) for counter diffusion of A and B, i. e. N_A and N_B having opposite signs, then

$$N_A' - N_B' = \frac{D_{AB} P}{RTL} \, \ln \frac{1 - \left(1 - \frac{N_B'}{N_A'}\right)\frac{P_{A2}}{P}}{1 - \left(1 - \frac{N_B'}{N_A'}\right)\frac{P_{A1}}{P}} \tag{13}$$

where the subscripts 1, 2, refer to the ends of the diffusion path. Eqn. (13) was derived by Sherwood and Pigford[14], but its use depends on knowledge of the relation between N_A and N_B.

For gases diffusing through a porous solid, the

D_{AB} obtained from Equation (13) is the effective diffusion coefficient for the specific gas pair and porous solid.

Since

$$N_A - N_B = N_A \left(1 - \frac{N_B}{N_A} \right) \qquad (14)$$

and

$$\frac{PV}{A} = NRT$$

Then

$$\frac{V_A'}{A} = N_A R T \qquad \text{at } P = 1 \text{ atm.} \qquad (15)$$

Here V_A' is the diffusion rate of A, in mls per sec. at 1 atm and diffusion temperature T.

Since V_A', the volume diffused, is not usually measured at the diffusion temperature, but some temperature T_{std}, a temperature correction must be included.

$$V_A' = V_A \frac{T}{T_{std}}$$

and $p_{A_2}/P = A_B$, mole fraction of A in B stream,

$p_{A_1}/P = A_A$ mole fraction of A in A stream.

Substituting these relationships in equation (13) and rearranging

$$D_e \cdot P = \frac{V_A \left(\frac{L}{A} \right) \left(\frac{T}{T_{std}} \right) \left(1 - \frac{N_B}{N_A} \right)}{\ln \frac{1 - \left(1 - \frac{N_B}{N_A} \right) A_B}{1 - \left(1 - \frac{N_B}{N_A} \right) A_A}} \qquad (16)$$

Hoogschagen[15] has shown experimentally that for both Knudsen and ordinary steady state counter diffusion at one atmosphere, $\frac{N_B}{N_A}$ is a constant in a constant total pressure system, and follows the relationship,

$$N_A \sqrt{M_A} + N_B \sqrt{M_B} = 0 \qquad (17)$$

or

$$\frac{N_B}{N_A} = \sqrt{\frac{M_A}{M_B}} \qquad (18)$$

where M_A is the molecular weight of species A,

M_B is the molecular weight of species B.

This equation was derived by Hoogschagen from the Knudsen[16] formula for Knudsen diffusion.

In the case of ordinary diffusion, this derivation was based on an impulse balance for the two diffusing species, and involving several simplifying assumptions. Although this theoretical estimate is only an approximation, the experimental results for the ratio of the diffusion rates for He-O_2, N_2-O_2, CO_2-O_2,[15] H_2-N_2[4] and H_2-NH_3[17] systems at atmospheric pressure (data are shown in the Appendix, Table 8) are in good agreement with this theoretical approach.

It would be desirable to test Equation (18) at higher pressures.

C. Measurement of Diffusion Rates

Two main techniques have been used in the past to measure diffusion rates, and from this information, to calculate diffusion coefficients. These methods, which have been called the Loschmidt method[18], and the Stefan method, are both unsteady state experiments. The Loschmidt method is based on the use of two chambers, each filled with a different gas, which are connected together in some way (by the use of a slide, valve, etc.). The diffusion rate can be calculated from the change in concentration in the chambers with time, and from the dimensions of the apparatus. A number of variations of this type have been used, at various times. Nearly all work on diffusion in dense gases at high pressures has been carried out in this type of cell. In the latter case, a porous plug was usually used to separate the chambers, and concentration changes were followed by means of radioactive tracers. The self-diffusion coefficient, or binary diffusion coefficient at elevated pressures measured in this way has been reported for argon[19], N_2[20], CO_2[20][21][22], N_2-CO_2[23], CH_4[24], CO_2-CH_4[25], and CO_2-C_3H_8[25]. Properly speaking, a tracer technique gives at least a two component mixture, and self-diffusion coefficients measured by this method are really close approximations. Similarly, mixtures containing tracers, and treated as binary

mixtures, are really ternary mixtures. In many cases there have been a wide disagreement among different investigators concerning the effect of pressure on the diffusion coefficient.

The Stefan method is based on the evaporation of liquids from narrow tubes into a flowing gas stream. One of the components must be in the liquid phase, and it is limited,therefore,to fairly narrow ranges of temperature and pressure for any one system. This has been used by only Sage and coworkers, [26],[27],[28], to measure the effect of pressure on diffusion. For n-heptane and n-hexane in binary mixtures with methane, ethane and propane it was found that pressure had a significant influence on the diffusion rate even at pressures below 60 psia, the maximum used.

A constant pressure flow system was used by Wicke and Kallenbach[29] originally to study counter diffusion, particularly when surface flow occurred. Weisz[30] and Cox[4] later used modifications of the method to study effective difusivities of porous solids, and in the latter case, to also measure diffusion coefficients at elevated temperatures. Apparently, a steady state flow system has not been used previous to this work to measure the effect of pressure on the diffusion coefficient.

All correlations which have been presented for the calculation of the binary diffusion coefficient assume

that it varies inversely with the pressure, at least for ideal gases (5), (8), (31), (32), (33). For dense gases, the Enskog theory, (11), (33) which treats the molecules as rigid spheres, has been used to predict behavior of the diffusion coefficient as pressure varies.

Perhaps the best present correlation for the bulk diffusion coefficient of ideal gases is that of Hirschfelder et. al. (33).

$$D_{AB} = 0.0026280 \frac{\sqrt{T^3(M_A + M_B) \big/ 2 M_A \cdot M_B}}{P \sigma_{AB} [\Omega_{AB}^{(1.1)*}(T_{AB}^*)]} \qquad (19)$$

where D_{AB} = diffusion coefficient in $\frac{cm^2}{sec}$

      P = pressure in atmospheres

      T = temperature in ^oK

      T_{AB}^* = $\frac{kT}{\epsilon_{AB}}$

      k = Boltzman constant

      M_A, M_B = Molecular weight of species A and B

      $\sigma_{AB}, \frac{\epsilon_{AB}}{k}$ = Molecular potential energy parameters characteristic of A-B interaction in $\overset{o}{A}$ and ^oK, respectively.

It is well known that for ideal gases the molecular parameters, σ and ϵ, are a function of temperature only. Therefore at constant temperature, Equation (19) may be written in the form:

      D_{AB} = Constant x $\frac{1}{P}$, or

      $D_{AB}P$ = Constant

For an ideal system, this would be the expected result from diffusion rate measurements through porous solids if the pore size of the solid is large enough to eliminate Knudsen diffusion. For the N_2 - H_2 system, this relationship should hold at $20^{\circ}C$ for pressures up to 60 atms.

APPARATUS

A.   Diffusion Apparatus

The apparatus for measuring the effective diffusion coefficient at higher pressures was similar in principle to that used by K. E. Cox for the investigation of the temperature dependence of the diffusion coefficient. A different means of obtaining pressure balance between the two sides of the porous sample, and modification of the thermal conductivity cells were noteworthy changes.

The apparatus, as shown in Figure 1, consisted of two gas trains, one for hydrogen, and the other for nitrogen, which led the two gases to opposite faces of the cylindrical porous solid sample. The hydrogen used was standard commercial grade (99.8%), and the nitrogen was premium quality (99.85%). Both were supplied by the Canadian Liquid Air Company.

Now consider the hydrogen gas stream in detail. The gas from the storage cylinder first was passed through a pressure regulator, and reduced to the desired pressure, before passing through a steel pipe dryer (1" pipe, 24 inches long, packed with anhydrous calcium sulfate). For small flow rates of gas, less than 100 milliliter per minute, a 6 foot long steel capillary, 0.013 inch inside diameter, was inserted before the dryer. This capillary provided a back pressure of approximately 10 pounds per square inch to

smooth out the flow. The hydrogen, after drying, was metered in a ball float glass flowmeter (Matheson types 202, 203 and 204), and was then conducted into one side of the diffusion cell where it contacted the face of the cylindrical porous sample. The nitrogen stream followed the same sequence as the hydrogen. After flowing out of the diffusion cell, the two gas streams, one mainly hydrogen, the other mainly nitrogen, were joined to two branches of a tee and released by a single pressure regulator to the atmosphere. Part of the gas, about 60 milliliters per minute, was taken as sample from each stream before the join. Each gas sample was regulated by a small Ermeto needle valve, and then passed to the modified thermal conductivity cell for analysis.

The differential pressures between the top and bottom chambers of the diffusion cell were measured on an inclined draught gauge (Hays, 12 inch scale, range 0 - 1.0 inch water).

The absolute pressure of the gas in the system was measured by an accurate test gauge of 300 psig scale, and could be read with an accuracy of 0.2 psi. For pressures less than 5 psig. a mercury manometer was used instead of the test gauge.

Since the apparatus was designed for measuring the diffusion rates at elevated pressures, all equipment used under high pressure was assembled and connected by
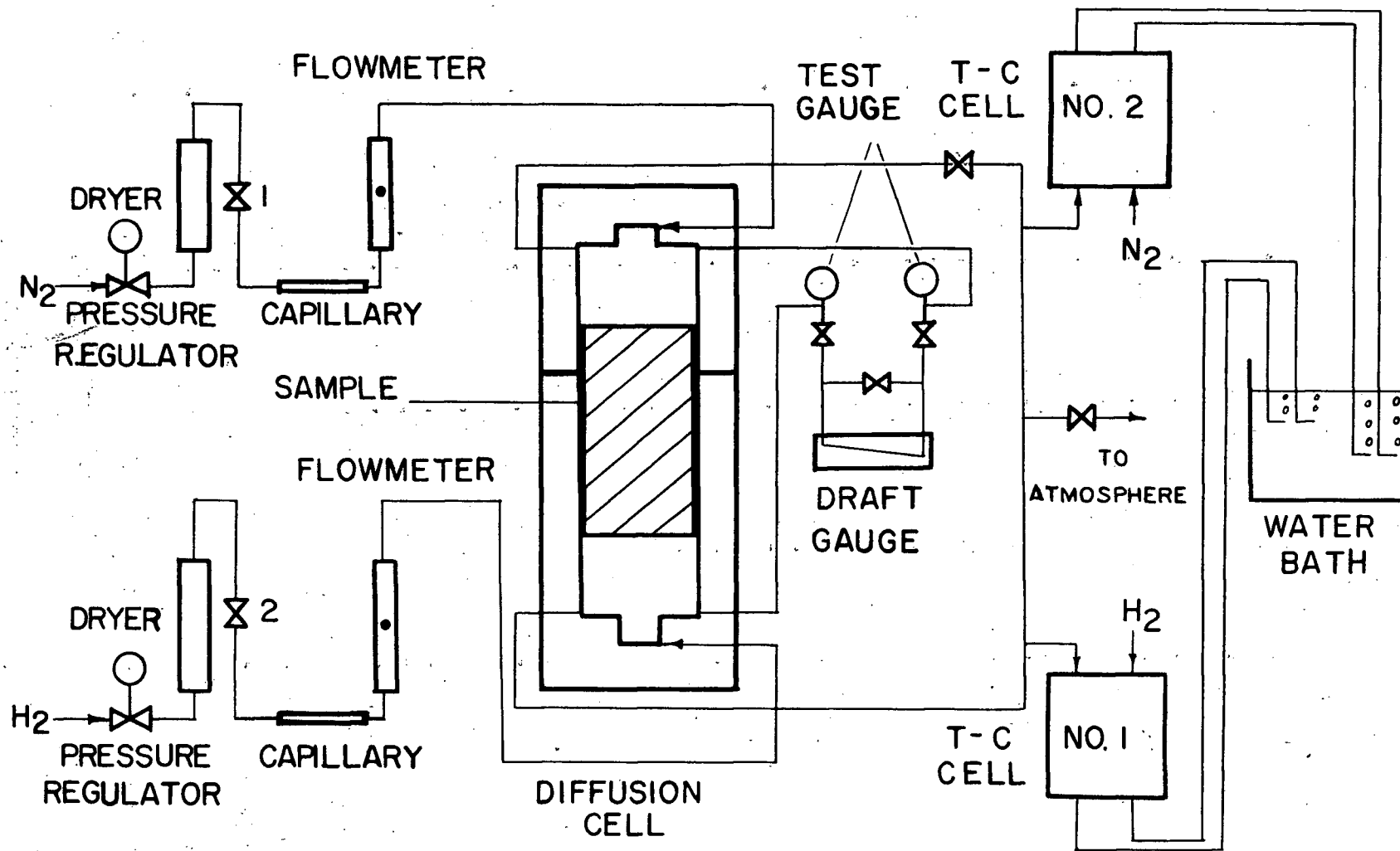
Figure 1. Apparatus for Measurement of Diffusion Rate

3/8" standard stainless steel tubing and Ermeto type
fittings. All equipment was tested to a pressure of
at least 300 psi.

B.   Modification of Thermal Conductivity Cells. (T-C Cell)

The thermal conductivity cells employed were two Gow-
Mac, Model NIS, recorder type, with four filament chambers
for each cell. These cells were of the "diffusion type"
with the filaments being located in a diffusion passage.
This construction was supposed to make the cell response
independent of gas flow rate. As shown in the Appendix,
Figure 15, this was true only if the flow was above 100
mls per minute, at lower flows the cell was very sensitive
to flow rate.

If the differential pressure between the two sides of
porous sample was to be adequately controlled by mixing
the two streams after sampling, then the gas sample could
be only a small part of the outlet flow (about 300-500 mls
per min. each). The millivolt output apparently was a
single function of gas composition in the Gow-Mac cells
only if the gas pressure, as well as the flow rate was
maintained constant all the time. From this point of
view, the T-C cells were modified to give a constant flow
rate and pressure over the filaments even if the gas
sample rate varied. As shown in Figure 2, this was
achieved by "leaking" a fraction of the sample over the
filament and out through a capillary. A constant flow
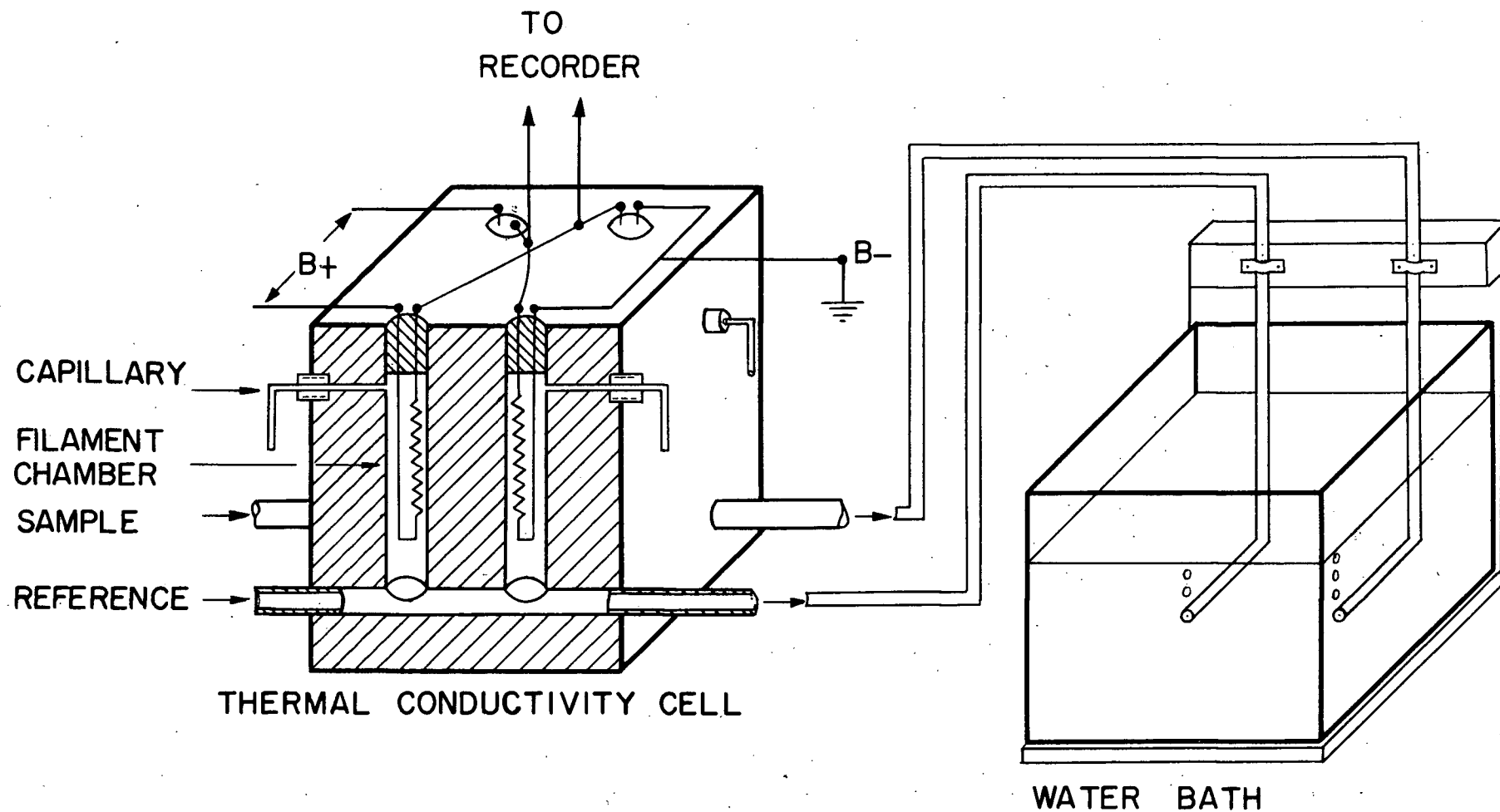
Figure 2. Modification of Thermal Conductivity Cell

17a
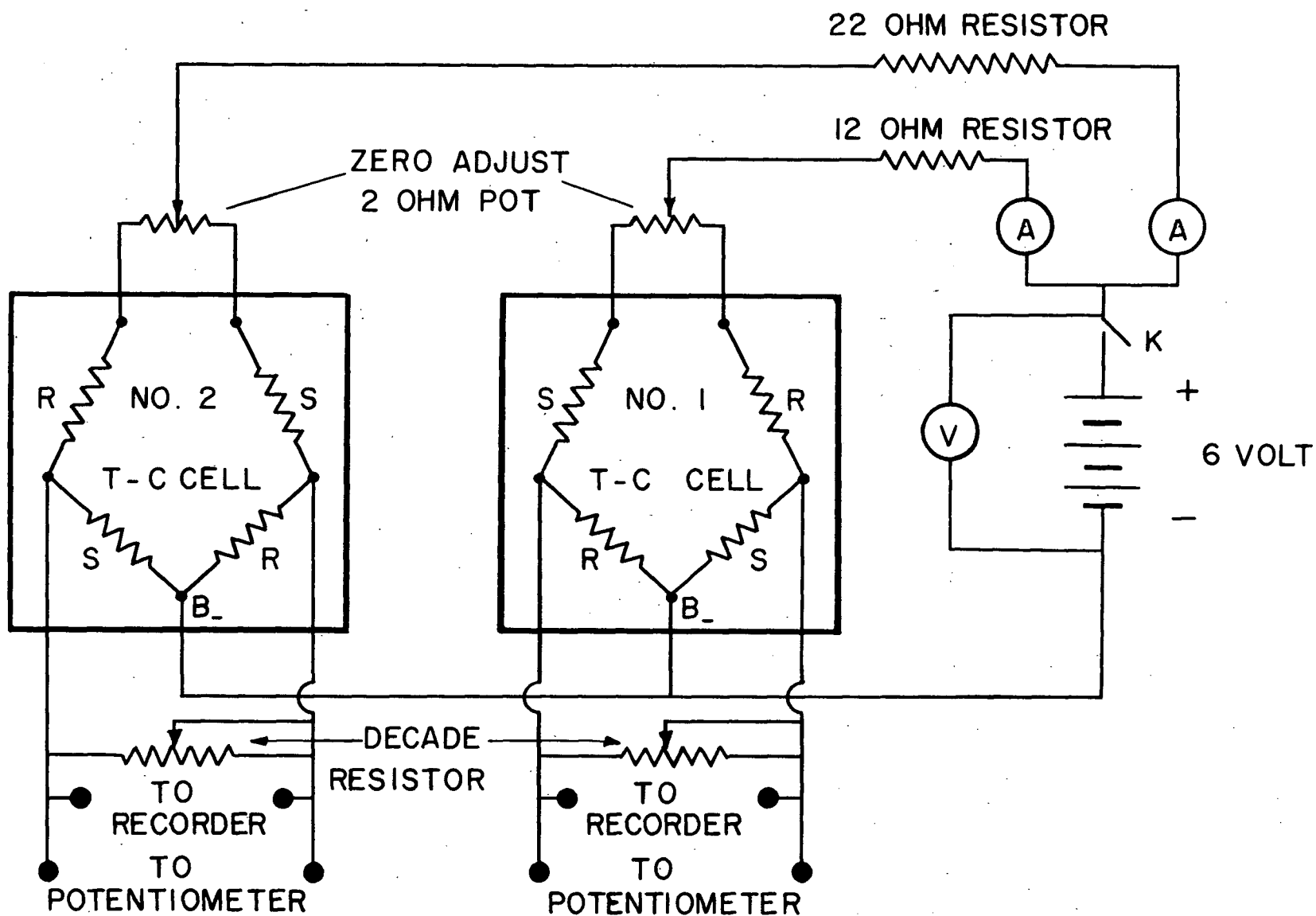
Figure 3. Wiring Diagram of Thermal Conductivity cells

rate was attained through the capillary by keeping the sample flow under a constant pressure by bubbling it out through a water bath maintained at a constant level.

For each sample or reference filament chamber, a capillary 1 1/2 inch long and 0.013 inch inside diameter welded to a 1/4 inch pipe plug was screwed into the brass block of the T-C cell. A hole was drilled to join the filament chamber to the plug and hence capillary. The pressures of sample gas were maintained at 2 inches water head for nitrogen and 1 inch for hydrogen.

This arrangement worked very well, giving reproducible readings for both streams, without having to regulate the sample flow except in a qualitiative sense so that about the same rate of bubbling was used at all times.

The wiring diagram for the thermal conductivity cell is shown in Figure 3. A Varian Associates G-10 recorder as well as a Leeds and Northrup Portable precision Potentiometer were used to measure the output millivolts from the thermal conductivity cell bridge. The filament current was provided by a 6 volt storage battery. One voltmeter and two milliammeters were connected in the wiring system to indicate the voltages and milliamperes respectively for the filament currents of the two cells. For the detection of N_2 in hydrogen, a filament current of 280 ma was used, and for detecting H_2 in nitrogen 124 ma. Also a decade resistor was used for regulating

the output millivolt signal of each thermal conductivity
cell to the potentiometer or the Varian recorder.

C.   Diffusion Cell

The diffusion cell was a cylindrical bomb 6 inches
high and 4 inches in diameter as shown in Figure 4, 5 and 6.
It consisted, essentially, of two cylindrical pieces, the
top one 2.5 inches high and the bottom one 3.5 inches high,
each machined from mild steel bar stock. The inside sample
chamber diameter was 1 3/32 inches which was just the
diameter of the porous sample when covered with a Gooch
rubber sleeve. This sample chamber was 1 1/2 inches deep
in the top half and 2 1/2 inches deep in the bottom
half.

In the top half, were six vertical holes of 7/16
inch diameter drilled for placing socket head cap screws.
The upper piece also had three gas ducts 3/16 inch in
diameter, used as a gas entrance, a gas outlet, and a
connection to the differential draught gauge. Two of
the ducts were drilled from the lower face of the top
piece vertically up into the solid and turned a 90 degree
angle to the end of the chamber, as shown in Figure 4.
One duct entered along the axis of the sample chamber
through a small extension, 1/2 inch deep and 1/4 inch in
diameter which was machined further into the steel from
the end of the sample chamber.

The face of the bottom piece had two grooves
machined in it. The inner one was for a standard high

PLAN VIEW OF BOTTOM PIECE

$1\frac{17}{32}"D$

$1\frac{5}{16}"D$

$1\frac{1}{8}"D$

$1\frac{3}{32}"D$

$\frac{7}{16}"D$ 6 HOLES
FOR SOCKET
HEAD CAP
SCREWS

$\frac{5}{16}"D$

$\frac{3}{16}"D$

6 GAS DUCTS

A A

SECTION B-B
GAS DUCT DETAIL

ADAPTER ETC., NOT SHOWN
IN THESE VIEWS

SECTION C-C
GAS DUCT DETAIL

TOP
PIECE

DIFFUSION
CHAMBER

O-RING

ADAPTER

BOTTOM
PIECE

$\frac{1}{2}"$

$2\frac{1}{2}"$

$\frac{1}{2}"$

$3\frac{1}{2}"$

$2\frac{1}{2}"$

$\frac{1}{2}"$

$\frac{1}{4}"$

$1\frac{3}{32}"$

4"

SECTION A-A
BOTH CELL PIECE

FIGURE 4   DIFFUSION CELL

SAMPLE

2.5"

3.5"

4"

TOP
DUCTS

BOTTOM
DUCTS
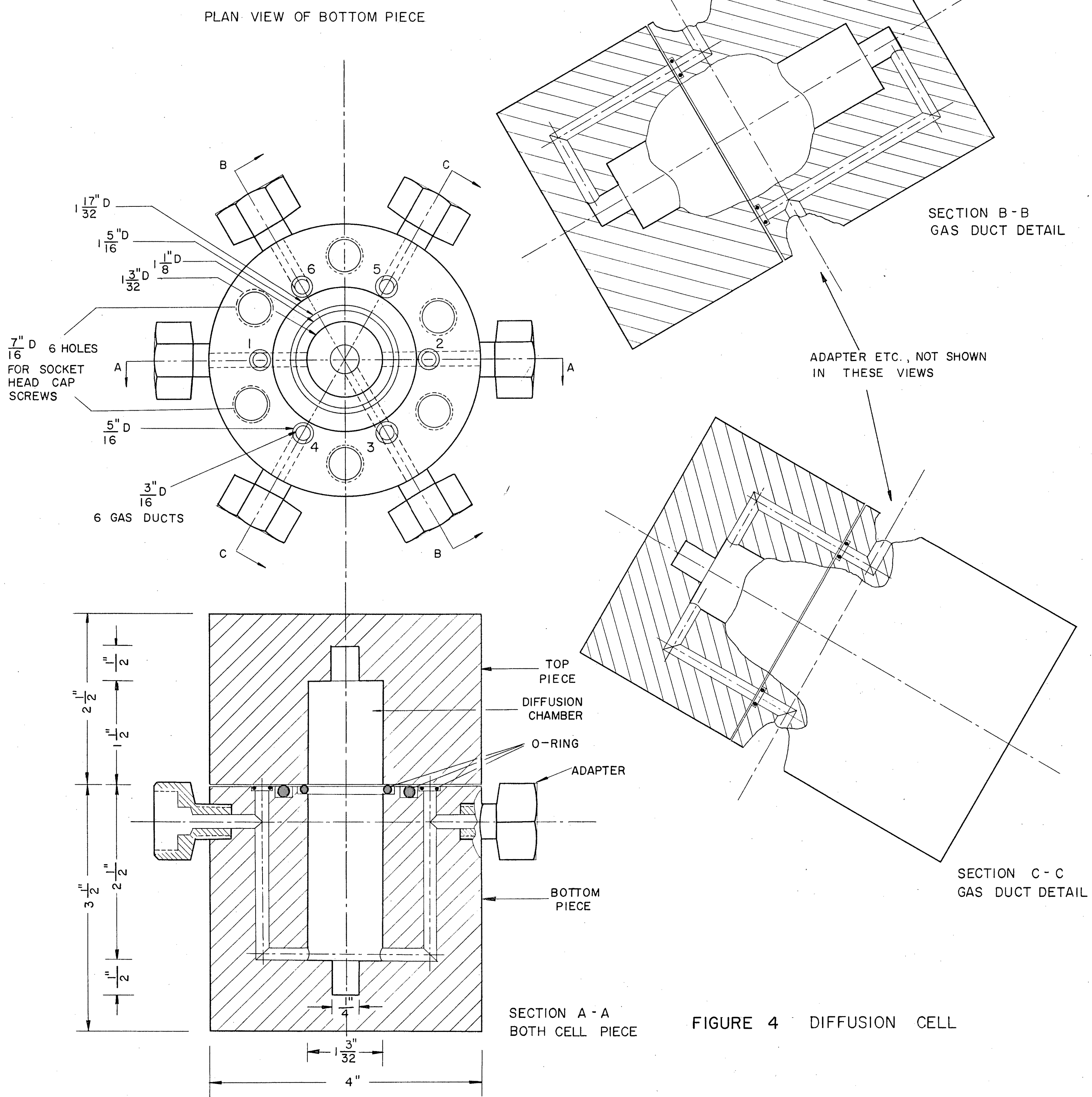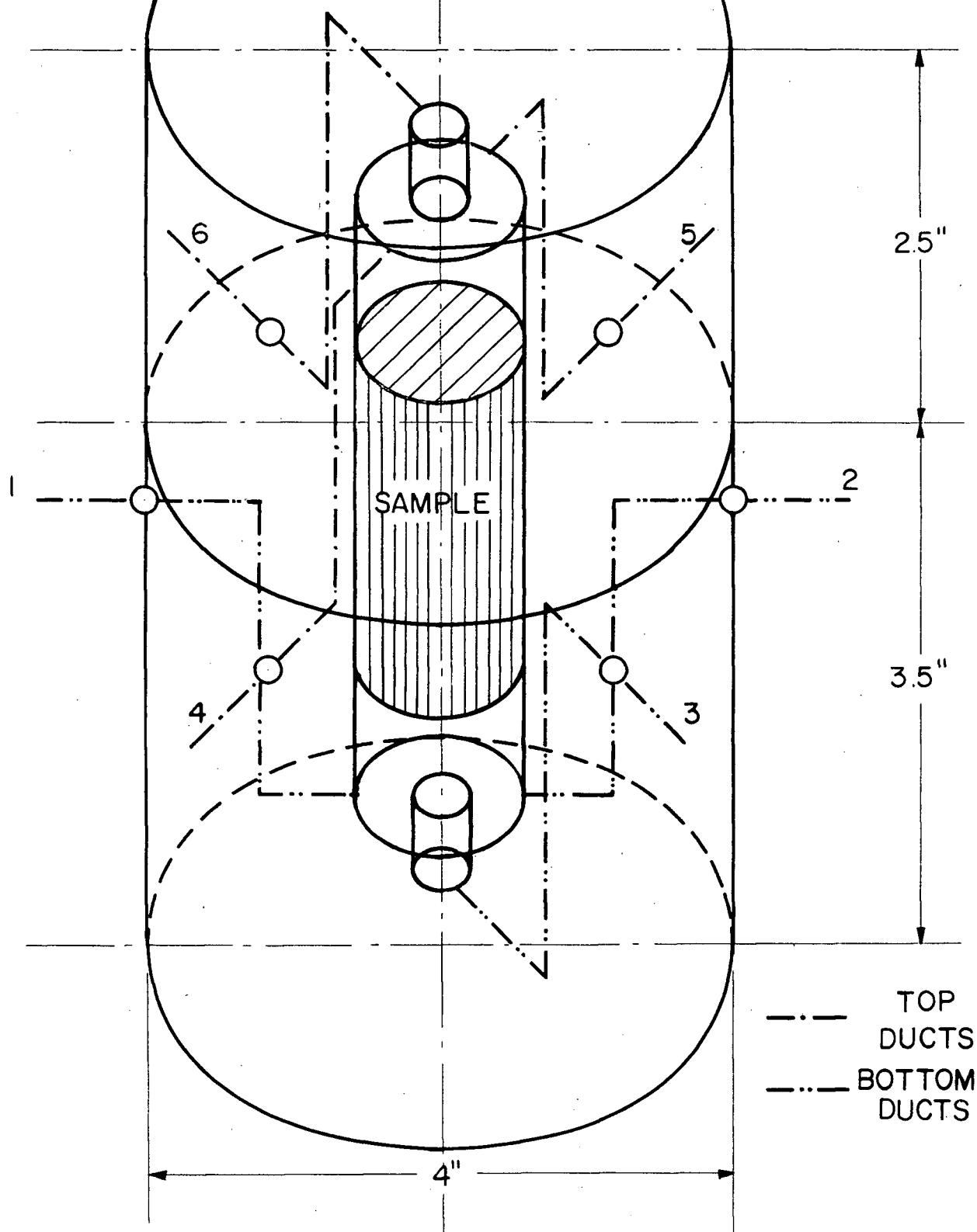
Figure 4a. Isometric Sketch of Diffusion Cell, Showing Gas
Duct Arrangement
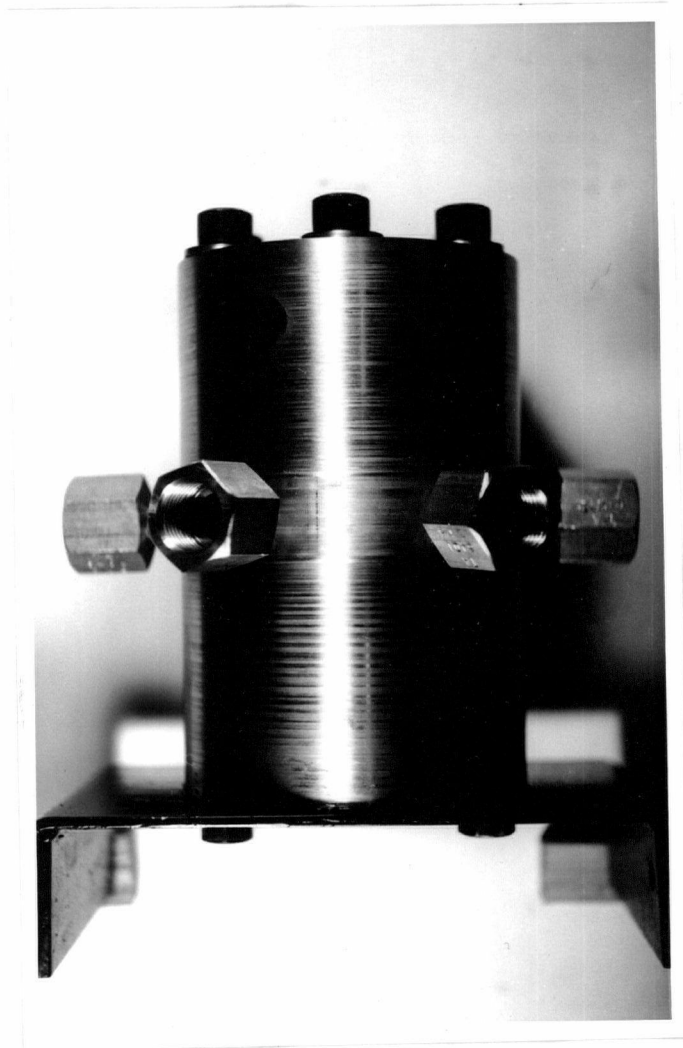
Figure 5. Diffusion Cell - Side View
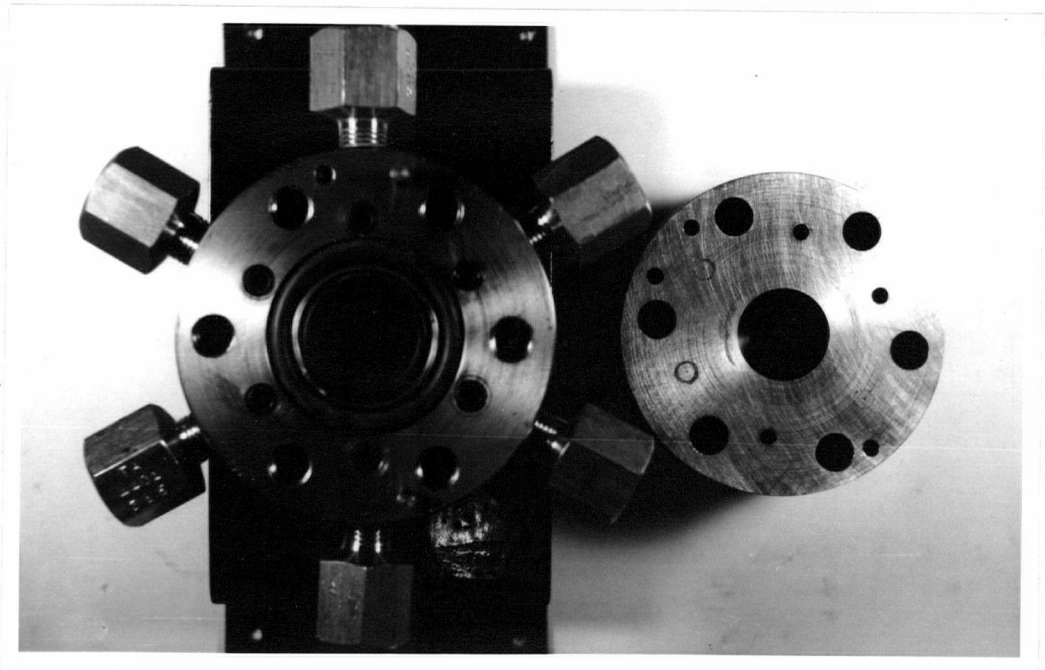
Figure 6.   Diffusion Cell - Top View

pressure O-ring which could hold the porous sample in a proper position. In the outer groove was placed an O-ring for sealing the two halves of the cell from leakage. There were also six holes of 7/16 inch diameter for the cap screws. The gas ducts, which were identical in the top and bottom sections, were sealed by small o-rings.

D. Porous samples

The porous samples used in this study were the Selas 03 and 01 supplied by the Selas Corporation of America in the form of rods of approximately 1 inch diameter and 6 inches length. Samples 03-A and 03-B, named No. 1 and No. 2 were cut from the same rod (Number 03-3) with lengths approximately 2 inches and 1 1/4 inches, respectively. Sample 01-A, named No. 3, was cut into a 2 inch cylinder from a longer rod (Number 01-1). The two end faces of a sample were machined to smoothness. From the manufacturer's literature and from other work[4] the Selas samples were known to be microporous synthetic ceramic rod, used primarily as bacteriological filters. The Selas 01 has a much greater porosity, and a larger pore size, than the 03 grade. The characteristics of the samples are given in Table 1.

TABLE I

Characteristics of Sample

No.	Selas.No	Length L, cm	X-sectional Area, A. cm^2	Porosity Θ	Pore Size Average Microns	D_T/D_e This work	D_T/D_e Cox's work
1	03-A	5.019	5.376	0.286[a]	1.31[a]	10.66	11.40
2	03-B	3.157	5.376	0.286	1.31	12.86	11.40
3	01-A	5.065	5.391	0.590[b]	4.5[b]	3.97	2.61

Data for Porosity and Pore Size were taken from Cox's work.

a. For Sample 03-2

b. For Sample 01-1

EXPERIMENTAL PROCEDURES

A.   Calibration of Test Gauge

The test gauge used for indicating the total pressure
in the diffusion cell was calibrated with an Ashcroft
gauge tester. The results, as shown in Table 2 showed
that most of the readings were very accurate, and the
maximum deviation was only 0.3 psig.

B.   Calibration of flowmeters

The calibration of the Matheson Universal flowmeters
(sizes 202, 203 and 204) could be calculated by knowing
the specific gravity and viscosity of the gas under the
conditions of flow. Other methods, such as rising soap
bubble method[4], and a wet test meter were also used to
check the calibration. Since the calibration curves,
showing tube reading against flow rate, for each flow
meter are a function of temperature, pressure and gas
properties, each flow meter had to be calibrated for
the entire range of working conditions. The flow rates
were expressed in terms of millimeters per minute
(or mls/min) at standard conditions, i.e. $70^{\circ}F$ $(21.1^{\circ}C)$
and atmospheric pressure.

The two 204 flowmeters, used for calibrating the
thermal conductivity cells, gave a flow rate ranging
from 100 to 5000 mls/min, and were simply calibrated by
the Matheson calculating method. Cox[4] checked these
flowmeters by using a wet test meter, and proved that

TABLE 2

Calibration of Test Gauge

Standard Pressure psig	Gauge Pressure psig	Deviation psig
5.0	5.0	0
10.0	10.3	+0.3
15.0	15.2	+0.2
20.0	20.0	0
25.0	25.0	0
30.0	30.0	0
50.0	50.0	0
75.0	75.2	+0.2
100.0	100.0	0
125.0	125.0	0
150.0	149.7	-0.3
200.0	200.0	0

these calculated calibrations were accurate enough to apply without further experimental measurements. But the two 202 flowmeters, with flow rates ranging from 5 to 100 mls/min, had to be calibrated by the rising soap bubble meter. The calibration curves obtained from the Matheson calculation, a nd the rising soap bubble meter are shown in the appendix, Figures 16 and 17. Evidently, the Matheson calibration for the 202 flowmeter, had a discrepancy of 20-50%, and was not recommended.

For diffusion runs, flowmeter 203-1 was employed for the hydrogen stream and flowmeter 203-2 for nitrogen (A sapphire float was used if the last number is 1, and a steel float when the last number is 2). Both were calibrated by the Matheson calculating method, and also by use of the wet test meter at room temperature under various pressures of 1.068, 1.2, 1.34, 1.68, 2.02, 3.04, 4.376, 6.1, 7.8, 9.5, 11.2 and 14.6 atmospheres. Several calibration curves are shown in the Appendix, Figures 18, 19 and 20. It appeared that, for the nitrogen stream, the Matheson results have a positive discrepancy of the order of 1-5% from that of the wet test meter. Usually the magnitude of the discrepancies increased with pressure and had an average deviation of +4%. For hydrogen flow, the discrepancies were even worse (up to 10 or 20%), and more irregular. Therefore, the calibration results from the wet test meter were

accepted for measuring the flow rates in the diffusion runs. It was estimated that these wet test meter calibrations were accurate to 1%.

Although the average temperature was about 21°C, it varied during experiments from 20°-25°C. Therefore, the flow rate obtained from the calibration curves should be corrected for temperature to the working condition. Also, as mentioned before, the pressure gauge might give 0.3 psig errors in readings. The error in recording temperatures and total pressures would affect the calibration of the flowmeters. The magnitude of this effect could be estimated from the Matheson calculations, because, although the Matheson calculation was not satisfactory at elevated pressures, the calculated calibration curves usually were parallel and proportional to those of the wet test meter. As shown in the Appendix, Table 9, the calibrations of hydrogen flowmeter (203-1) and nitrogen flowmeter (203-2) were all calculated at six conditions: 70°F, 150 psig; 70°F, 15 psig, etc. It was found that the effect of 5°F in temperature or 0.3 psig in pressure on the flow rate was of the order of about 1%. This proved that the possible error in recording experimental temperatures and pressures, i. e. less than 0.5°F and 0.3 psig, would not affect the calibrations of the flowmeters significantly.

C.   Calibration of the Thermal Conductivity Cells (T-C Cells)

Before modification, the two thermal conductivity cells were calibrated in the same manner as that described by Cox[4]. No. 1 T-C cell was calibrated for detecting the percentage of nitrogen in the hydrogen stream. A constant flow of pure hydrogen gas was taken from a gas cylinder to the reference side as reference gas. No. 2 T-C cell was used for detecting the percentage of hydrogen in nitrogen, and pure nitrogen was used as reference gas. The calibration results, millivolts against composition of gas mixture, are shown in the Appendix, Figures 21 and 22.

Since the thermal conductivity of hydrogen is about seven times that of nitrogen, the No. 2 T-C cell is more sensitive than No. 1. Usually the No. 1 T-C cell took about two hours to warm up and attain a steady zero point due to the particular behavior of hydrogen. The other cell would reach its steady zero point within 30 minutes.

The calibration procedure of the modified T-C cells is stated briefly as follows:

(1) Start pure reference gas through both paths of the cell at a flow rate of 60-65 mls/min (measured at standard state) for each. Approximately 30 mls/min of the gas was evolved from the two capillaries and the rest of it, 30-35 mls/min, bubbled out under a one inch water head for the No. 1 T-C cell and a two inch head for the No. 2 T-C cell.

(2) The cells were now switched on. After waiting a few minutes until the cell was free of air, the filament current was adjusted to 280 milliampers for No. 1 cell and 124 for No. 2 cell.

(3) The 2 ohm pot attached to the cell was regulated until a straight zero line was obtained on the recorder chart. Thus the zero point of the cell was determined.

(4) With the reference gas running as before, a gas mixture of known composition was passed through the sample path. The mixture was made by taking the two gas streams, hydrogen and nitrogen at constant known flow rates into a tee branch mixer. Although the mixture might have a very high flow rate only about 60-65 mls/min of it was taken into the cell for analysis. The composition of the mixture was calculated from the known flow rates.

(5) The output millivolts were measured on the potentiometer after a constant reading had been obtained on the millivolt recorder. The sensitivity control resistor was set at 900 ohms for No. 1 T-C cell, and 100 ohms for No. 2 cell.

In this way a series of points, mole fraction (percentage of gas by volume) against millivolts were obtained with high accuracy. The calibration results are shown in Figures 7 and 8.
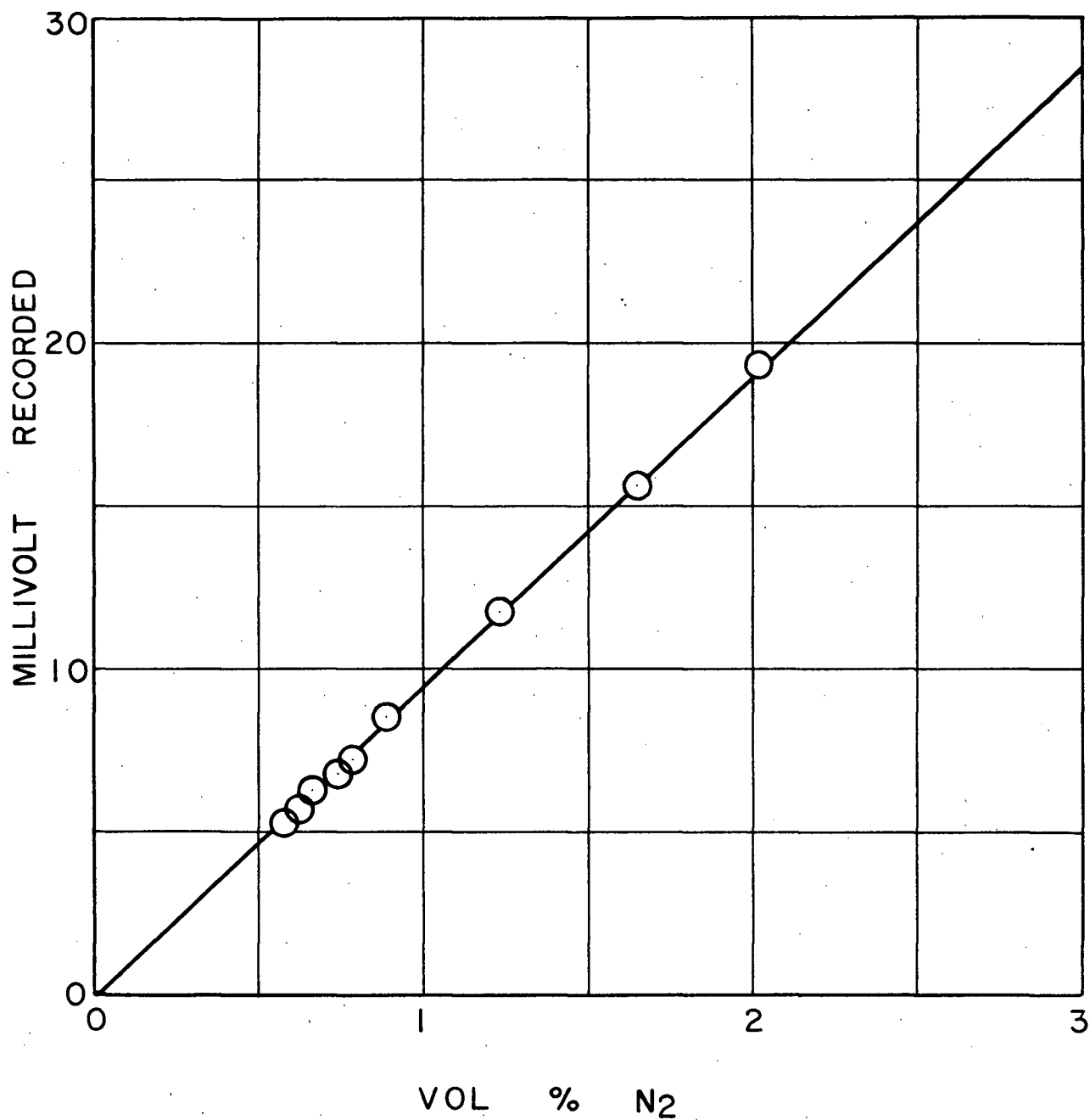
Figure 7. Calibration plot of Modified No-1
Thermal Conductivity Cell
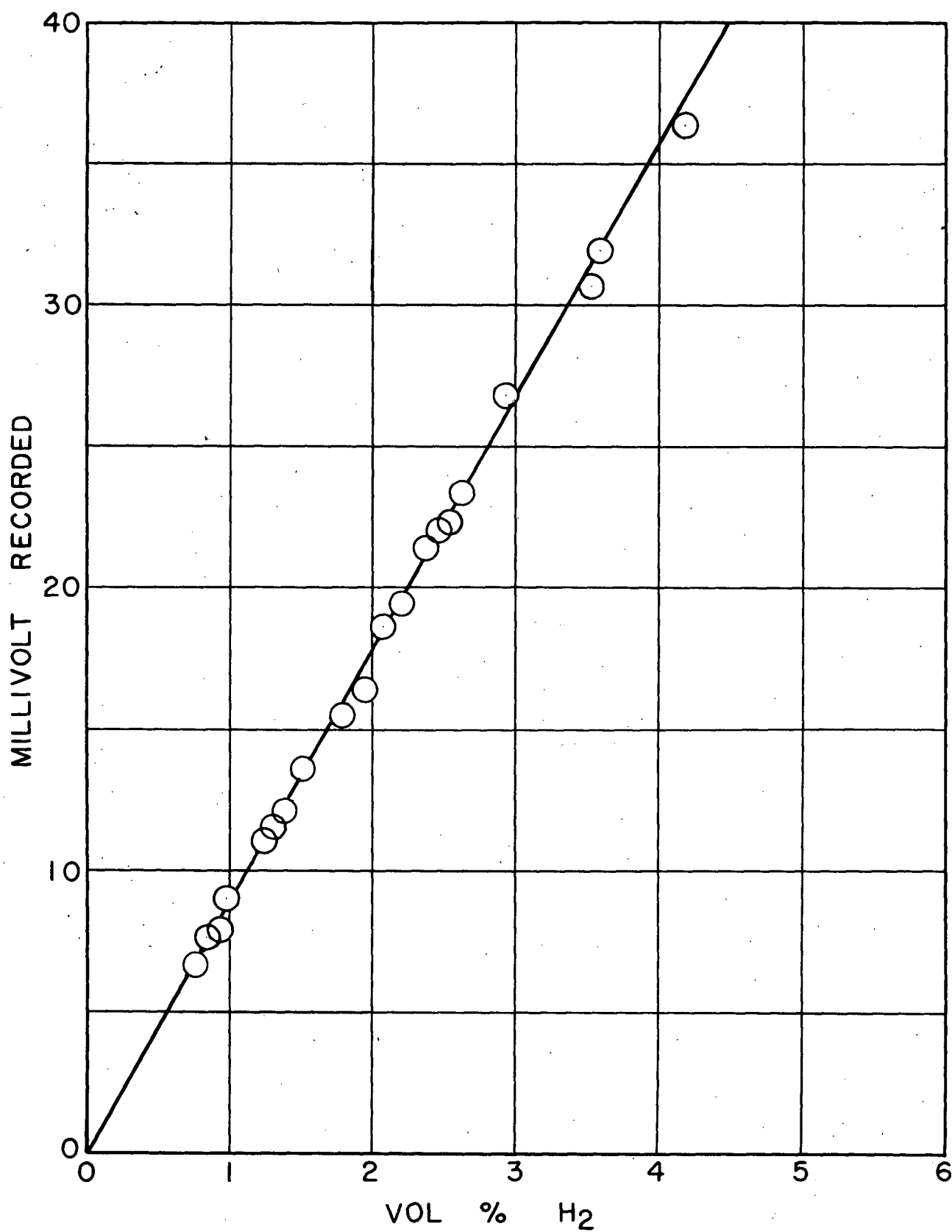
Figure 8. Calibration plot of Modified NO.2
Thermal Conductivity Cell

During the operations, it was found that more
attention had to be paid to the zero point of No. 1
cell in which hydrogen was used as reference.

D.   Measurement of the Effective Diffusion Coefficient of
      Porous Solids and Rate of Diffusion at Various
      Pressures.

Runs at various pressures, from 1.068 to 14.6
atmospheres, were all done in the same manner. To start
a run the apparatus was assembled and tested to be sure
it was free of leakage. The thermal conductivity cells
were then switched on for warming and zeroing over a two
hour period. The gas pressure regulators and needle
valves were then set to give steady flowmeter readings
at the required flow rates and pressures. The operating
conditions of the diffusion runs could be easily
adjusted to the required ones by turning needle valves
1 or 2 and the outlet reducing valve. The draft gauge
was set to zero differential pressure across the porous
solid by adjusting the needle valve which was placed in
the nitrogen line between the diffusion bomb and the tee
joint to the outlet regulator.

When conditions were steady, a constant reading was
obtained on the conductivity cell recording chart. The
output millivolts of the two T-C cells were then
accurately measured on the potentiometer connected in
parallel with the recorder.

For each run the following data were recorded:

Date of run

Run number

Number of sample

Flowmeter numbers, flowmeter readings and flow rates for hydrogen and nitrogen gas

Absolute pressure

Ambient temperature

Output millivolts of the two T-C cells

From these data it was possible to calculate the gas compositions after diffusion, and thus the rates of hydrogen and nitrogen diffusion (in milliliters per minute per square centimeter of the sample area). From the rates of diffusion the effective diffusion coefficient, D_e, for the sample tested was calculated. Complete recorded data for several runs (Table 10) and a sample calculation are given in the Appendix.

RESULTS

The rates of diffusion of the gas pair, hydrogen and nitrogen were measured at a temperature of 20°-25°C for total pressures from 1 to 14.6 atms absolute, and with different arrangements of the gas ducts in the diffusion bomb. In arrangement I, the pressure tap was located opposite to the gas outlet and the gas was conducted into each diffusion chamber from the small extension as shown in Figure 4 and 4a. In arrangement II, the pressure tap was now the previous gas inlet, so the gas inlet and outlet now became opposite to each other. For arrangement III, gas ducts were used in the same way as in arrangement II, but a thick paper baffle 1/4 or 7/8 inch high was inserted vertically between the inlet and outlet taps and coincided with a diameter of each chamber. Therefore, the gas which came into the diffusion chamber must flow over the baffle and be well mixed before going out of the cell.

Generally, the diffusion results are expressed in terms of

V_H rate of diffusion of hydrogen through a sample, mls/min (at 21°C and 1 atm)

V_N rate of diffusion of nitrogen through a sample mls/min (at 21°C and 1 atm)

$\dfrac{V_H}{V_N}$ ratio of rates of diffusion, dimensionless.

De.P the product of effective diffusion coefficient and pressure $(cm^2/sec)(atm)$

For convenience in comparison, the rates of diffusion and the effective diffusion coefficients were all corrected to the corresponding values at standard room temperature, i. e. $21.1^{\circ}C$ $(70^{\circ}F)$. The temperature dependence of the diffusion coefficient was calculated from the Hirschfelder et. al. equation (19). The diffusion coefficient for H_2-N_2 increases about 0.5% with an increase of $1^{\circ}C$ in temperature.

The results of diffusion runs at one atmosphere are shown in Table 3. The values of individual runs and their averages are all tabulated. The ratio of the diffusion rates of hydrogen for sample 2 and 1, as shown by the first two sets of results, is $\frac{10.57}{7.0} = 1.52$, which is in good agreement with the inverse ratio of the two sample lengths, i. e. $\frac{5.019}{3.175} = 1.59$. The error of 4.4% might be caused by inaccuracies in the flowmeter calibrations for Run B1-B4 of sample 1, or by minor differences in the two samples although they were cut from the same rod. Therefore, effective diffusion coefficients calculated, were taken to be independent of sample length. For sample 1, in the steel bomb, (Runs B1-B4) the ratios of the rates of diffusion for hydrogen and nitrogen gas are close enough to the theoretical value of 3.74 [4] at one atmosphere to confirm the absence of any appreciable forced flow. At 1.068 atmospheres, the ratios for sample 2 and 3 are higher than 3.74, and lie in the range of

3.8 to 4.0.

The ratios of diffusion rates for Young's and Kiss's cell are in good agreement with one another having values of 3.87 and 3.84 respectively at 1.068 atmospheres. The cell constant of Kiss's cell (which used the same porous sample as that in Cox's work) determined from these data checks with that of Cox. The temperature dependence of the gas pair NH_3-H_2 investigated by Young[35] has been found to agree with that obtained by Kiss[17], although Young used a different porous sample with the cell constant determined from Runs 48, 49 and 50.

Results for higher pressures are presented here in three series.

Series I: Sample 2 was tested with arrangement I and gas flow rates ranging from 200-1600 mls per minute. The average results are shown in table 4. For individual runs, the results are shown in Appendix, Table 4 a. The D_e.P product, and ratio of diffusion rates against absolute pressure are plotted in Figures 9 and 10.

Series II: This set of experiments was intended to show the effect of pore size of the solid, if any, as sample 3 has nearly 4 times the pore diameter and twice the porosity of sample 2. The average diffusion results for sample 3(01-A) with arrangement I and various pressures are shown in Table 5 and Figure 11. It can be seen that the D_e.P product does not vary with pressure.

Series III: These measurements were carried out to investigate the effect of cell arrangement. The average diffusion results for sample 2 with cell arrangement II and III are shown in Tables 6, 7 and 7a, and in Figures 12, 13 and 13a respectively. The D_e.P products of sample 2 with arrangement III (Table 7d in the Appendix) have a standard deviation of 2.35% with a corresponding mean deviation of 1.84%.

The average results given for each series were obtained by averaging all the diffusion runs made, which varied in number from 2 to 6 at each set of conditions.

TABLE 3

Diffusion Results at one Atmosphere and 21.1°C

(1)   Sample I, Arrangement I, Nitrogen in the top chamber

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21.1°C	atm P
B1	6.928	1.963	3.53	.0617	1
B2	7.039	1.920	3.67	.0622	1
B3	7.029	1.795	3.92	.0611	1
B4	6.990	1.869	3.74	.0617	1
Average	7.0	1.887	3.71	.0618	

(2)   Sample 2, Arrangement I, Nitrogen in the top chamber

Run No.	V_H		$\dfrac{V_H}{V_N}$	De. P at 21.1°C	atm P
4	10.72		(3.74)*	.0590	1.068
5	10.43		"	.0582	1.068
6	10.43		"	.0583	1.068
10	10.75		"	.0594	1.068
11	10.36		"	.0574	1.068
12	10.73		"	.0591	1.068
Average	10.57			.0584	

(3)   Sample 2, Arrangement I, Hydrogen in the top chamber

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21.1°C	atm P
73	10.22	2.542	4.015	.0574	1.068
74	10.22	2.620	3.904	.0562	"
75	10.10	2.646	3.820	.0588	"
Average	10.18	2.609	3.913	.0574	

(4)   Sample 3, Arrangement I, Hydrogen in the top chamber

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21.1°C	atm P
93	24.90	5.055	4.925**	.2028	1.068
94	22.66	4.787	4.70**	.1907	1.068
95	22.72	5.984	3.80	.2134	1.068
Average	23.43	5.275		.2025	

(5)   Kiss's diffusion cell (same as Cox's) Sample 03-2

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21.1°C	atm P
	10.01	2.653	3.92		1.068
	10.02	2.660	3.76		1.068
Average	10.01	2.657	3.84		

TABLE 3 (cont'd)

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	$\dfrac{atm}{P}$
(6) Young's diffusion cell, Sample 015				
40	21.40	5.576	3.840	1.068
49	20.84	5.300	3.932	1.068
50	20.52	5.338	3.844	1.068
Average	20.92	5.405	3.87	

*Assumed value $= \sqrt{\dfrac{M_{N_2}}{M_{H_2}}}$

**High hydrogen rate.

TABLE 4

Series I

Average Diffusion Results at Elevated Pressure

Sample 2 Arrangement I

Pressures P_T. atm	V_H	V_H	$\dfrac{V_H}{V_N}$	$De.P_T$ at $21^\circ C$	Note
1.068	10.57		$(3.74)^1$	(.0584)	
1.20	11.08		(4.05)	(.0598)	
1.34	11.18	2.572	4.42	.0599	
1.681	11.55		(4.46)	(.0615)	
2.02	11.89	2.564	4.673	.0614	
2.71	12.21		(4.82)	(.0632)	
3.04	12.37	2.615	4.83	.0649	
4.376	12.77	2.470	5.019	.0633	
6.1	12.92	2.511	5.14	.0638	
7.8	13.01	2.583	5.03	.0647	
9.503	13.69*	2.691	5.09	.0648	
11.204	13.42	2.553	5.343	.0633	
14.605	13.85	2.544	5.434	.0644	
6.1	12.71	2.418	5.24	.0640	(H_2in top)

*Very high flow rate of nitrogen (800-1200 mls/min)

[1]Results in parentheses based on interpolated values for ratio of diffusion rates.

TABLE 5

Series II

. Average Diffusion Results for Sample 31-A

	Arrangement I			
Pressure atm.abs.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21.1°C
1.068	23.09	5.275	4.49	.2025
1.34	25.1	5.046	4.91	.2085
1.68	25.67	4.67	5.51	.2051
3.04	27.5	4.48	6.26	.2033

TABLE 6

Series III

Average Diffusion Results for Sample 21-D

	Arrangement II			
Pressure	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P
1.34	10.41	2.175	4.788	.0538
2.02	11.17	1.965	5.668	.0533
3.04	11.32	1.953	5.798	.0537
4.376	11.35	1.955	5.804	.0539
11.204	11.99	1.683	7.142	.0531

TABLE 7

Results for Sample 2, Arrangement III with 1/4" Baffles

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De.P 21.1°C	P atm	T_{rm} $^\circ$F
117	10.74	2.461	4.364	.0572	1.2	70
118	10.81	2.425	4.458	.0570	1.2	70
119	11.32	2.412	4.71	.0583	1.68	71
120	11.43	2.345	4.873	.0581	3.04	71
122	11.65	2.377	4.901	.0598	3.04	71
123	11.85	2.288	5.18	.0585	4.376	71
124	12.03	2.031	5.923	.0575	4.376	71
126	11.72	1.903	6.159	.0593	11.204	

TABLE 7a

Average Diffusion Results for Sample 2 - 3

Arrangement III, with 7/8" baffle

Pressure	V_H	V_N	$\dfrac{V_H}{V_N}$	De.P 21.1°C
1.34	11.33	2.455	4.617	.0581
2.02	11.71	2.55	4.607	.0604
3.04	11.61	2.45	4.747	.0595
4.376	11.59	2.441	4.749	.0584
7.80	11.51	2.317	4.970	.0574
11.204	11.67	2.330	4.954	.0579

Figure 9. Diffusion Results for Sample 2, Arrangement I. De. P VS. P

Figure 10. Diffusion Results for Sample 2, Arrangement I. Ratio of Diffusion Rate VS. Pressure
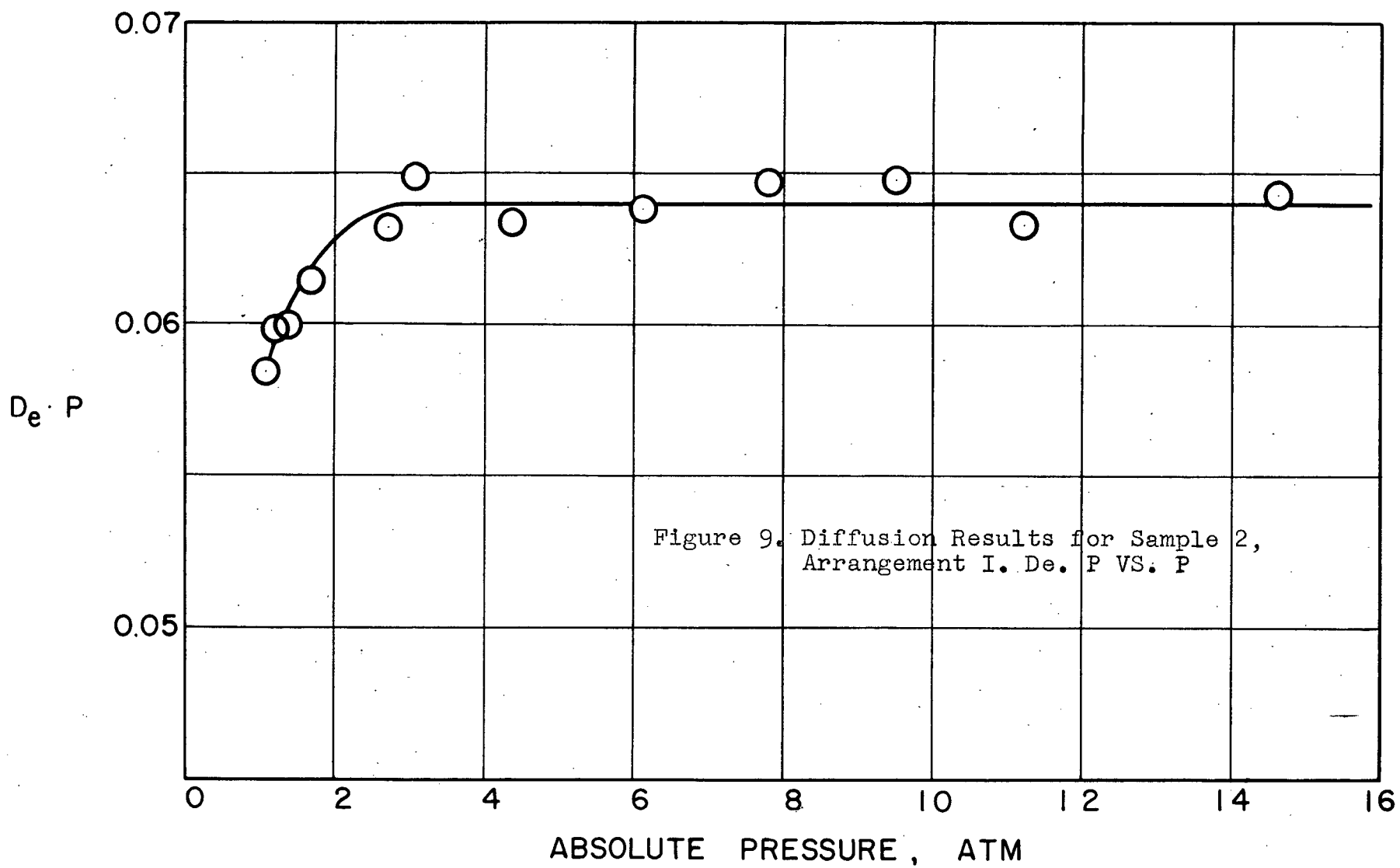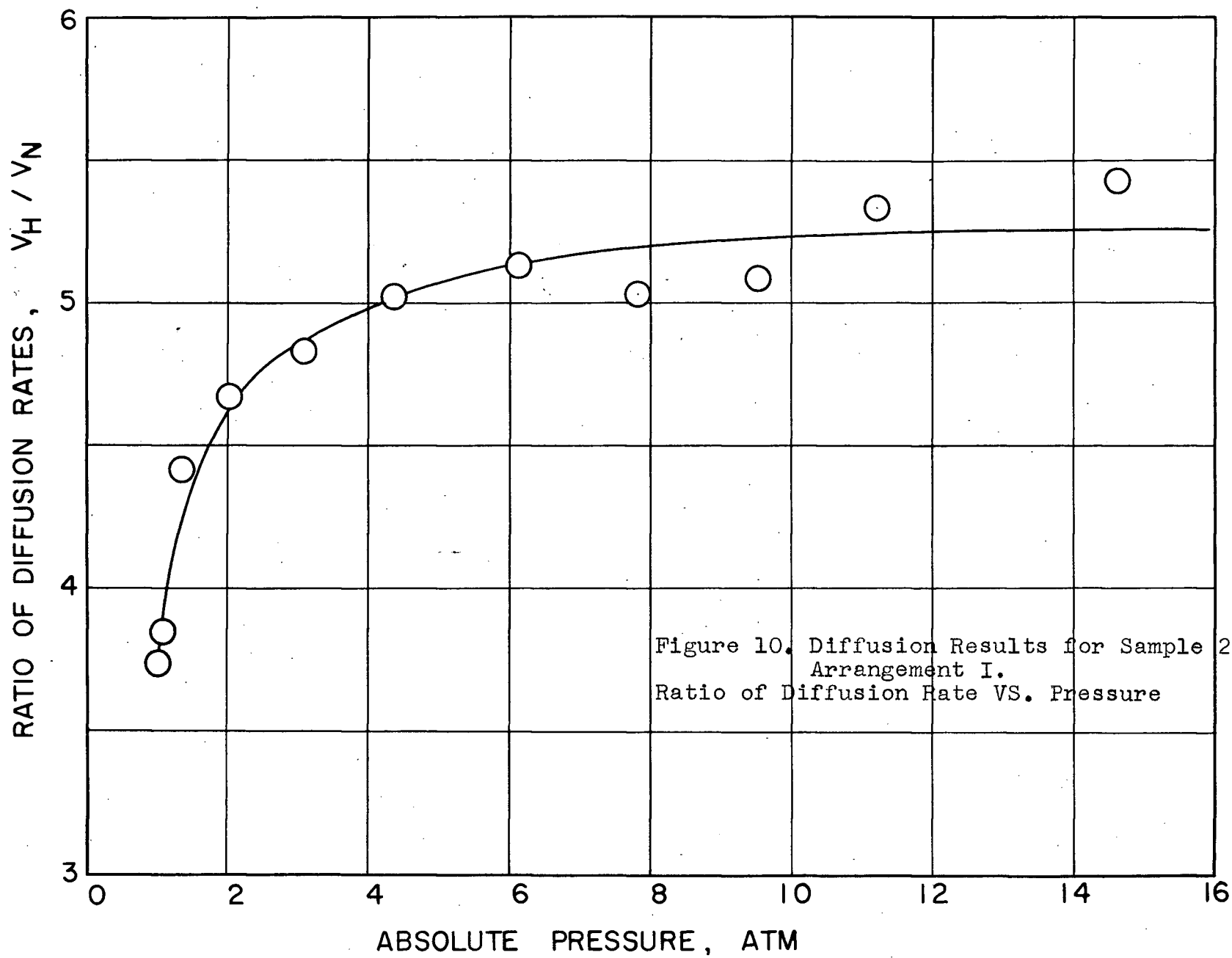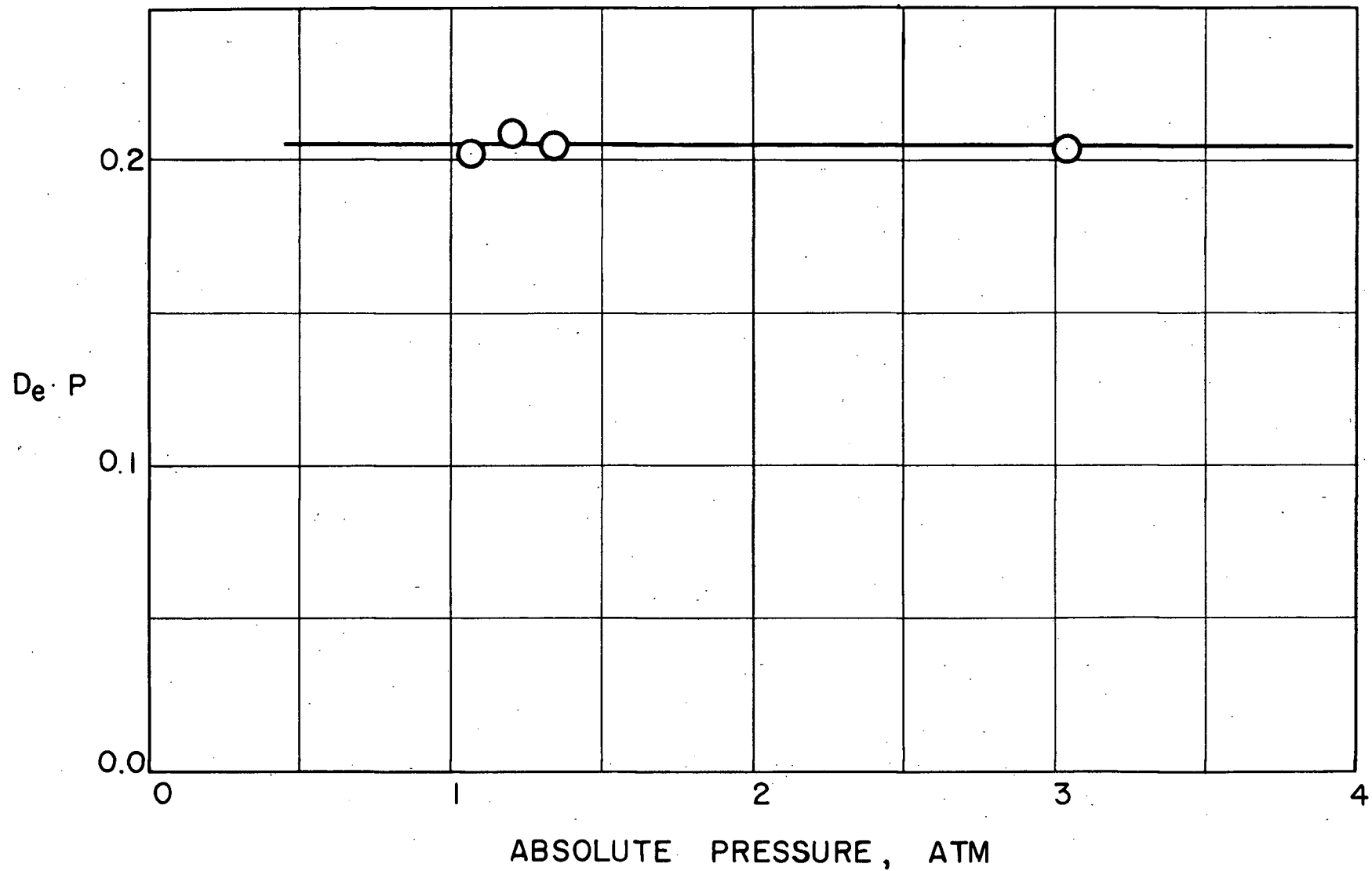
Figure 11, Diffusion Results for Sample 3, Arrangement I, De. P VS. P

Figure 12. Diffusion Results for Sample 2, Arrangement II, De.P VS. P

ABSOLUTE PRESSURE, ATM
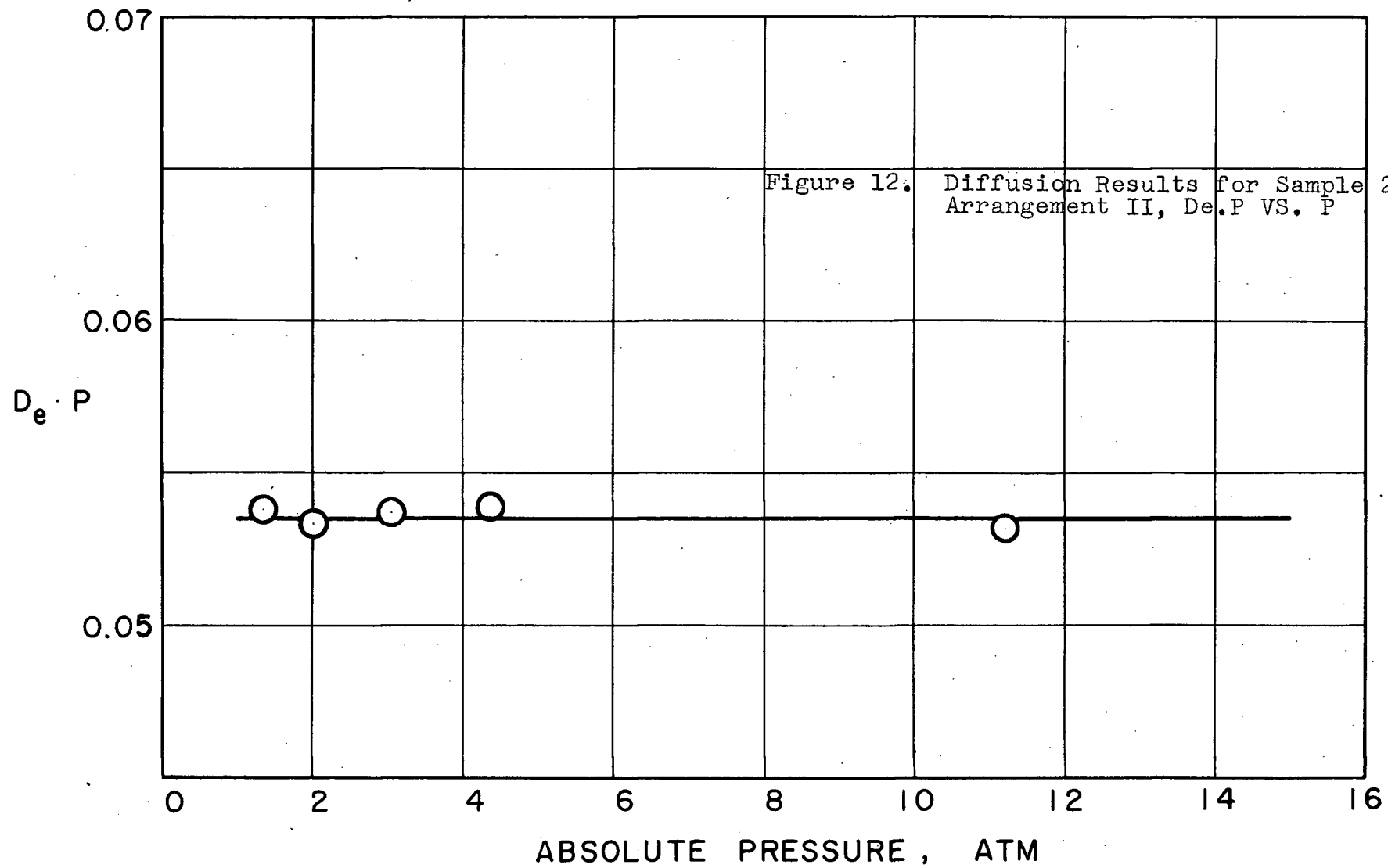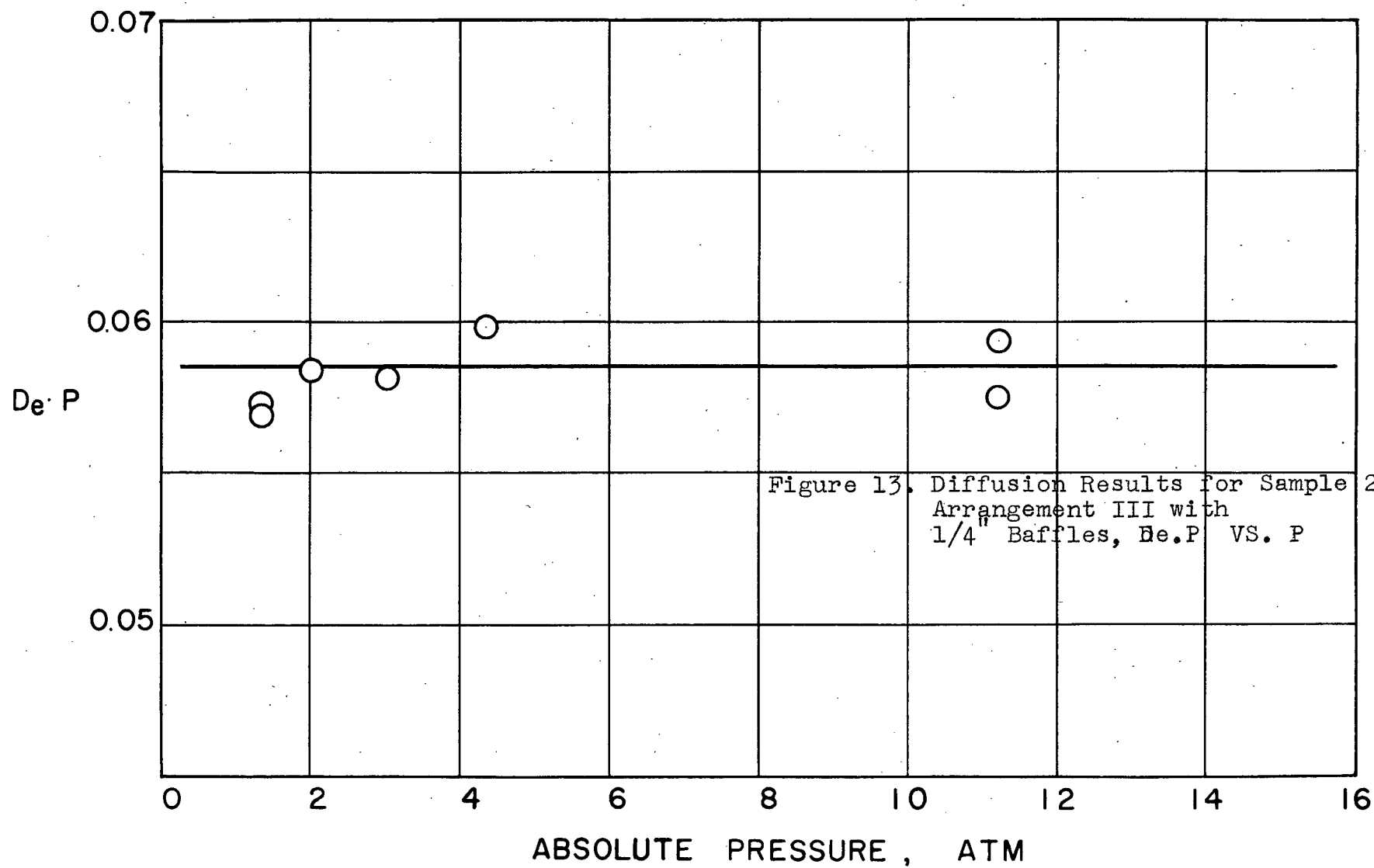
Figure 13. Diffusion Results for Sample 2, Arrangement III with 1/4" Baffles, De.P VS. P

ABSOLUTE PRESSURE, ATM

De·P

Figure 13a Diffusion Results for Sample 2, Arrangement III with 7/8 Baffles De.P VS. P

$D_e \cdot P$

ABSOLUTE PRESSURE, ATM
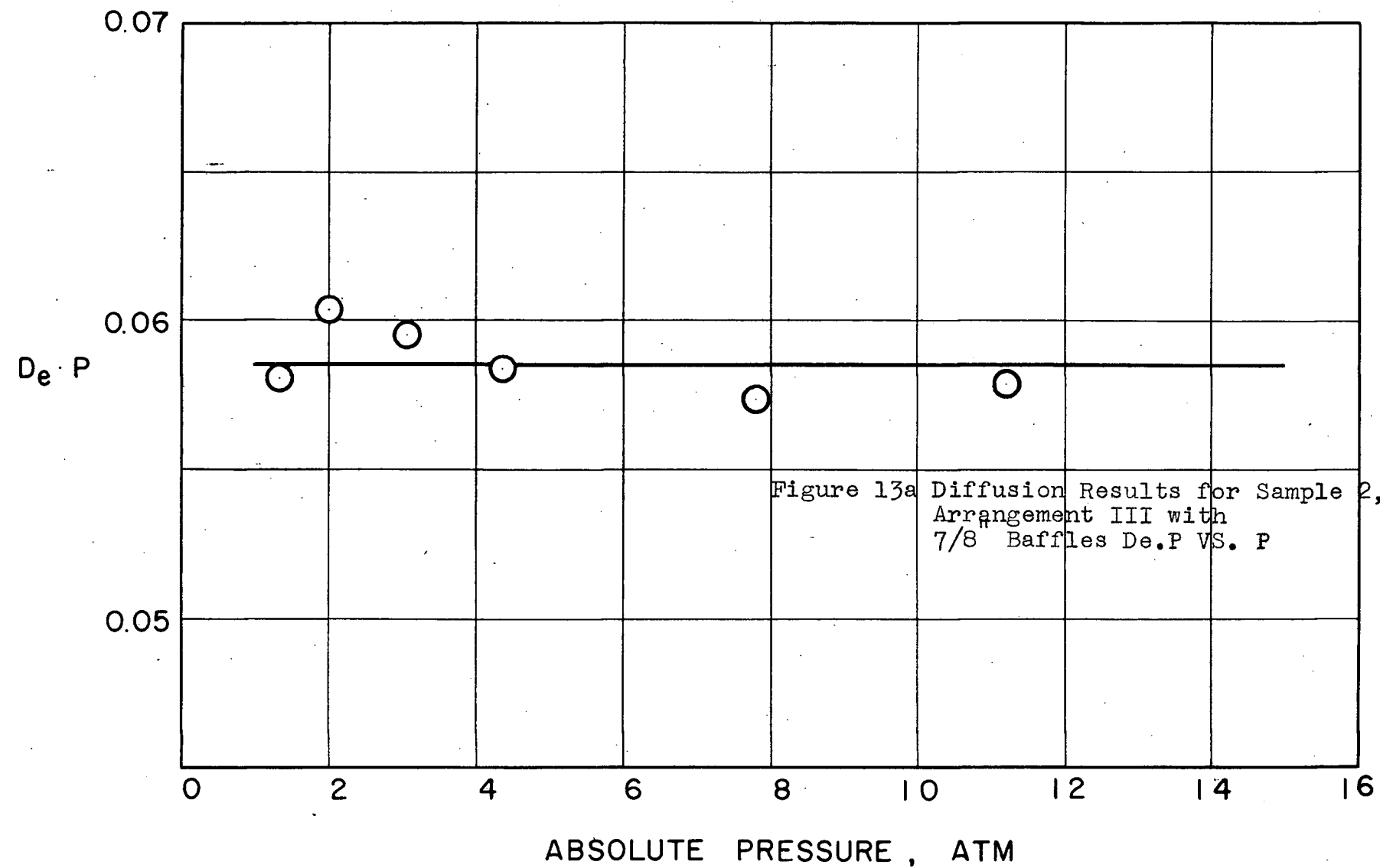
EXPERIMENTAL ERRORS

(A) Measurement of Gas Composition

The calibration plots for the modified thermal conductivity cells were used in the calculation of the diffusion rates.

Both of the calibration plots, for hydrogen and nitrogen analysis, were estimated to give the compositions of the streams after diffusion with a maximum error of $\pm$ 2.5% and an average error of less than 1%.

The main variables were the accurate control and measurement of the quantities of the two gas streams that made up the mixture of known composition for calibration purposes.

Figures 16 and 17, in the Appendix, show that the smaller streams could be metered with an accuracy of about $\pm$1.2% (i. e. within $\pm$ 0.2 ml/min.).

The electrical measuring circuit and the instruments used with it were felt to have a much smaller error.

(B) Measurement of Diffusion Rates

Possible errors in the diffusion rate values were provided by the inaccuracies in measurement of flow rates and calibrations of the thermal conductivity cells. In addition to these, forced flow, due to unbalanced differential pressure across the porous sample, and the effect of diffusion bomb geometry and pressure tap location, also played variable roles in affecting values of diffusion rates.

It was felt that the measurement of flow rates
(200-1000 mls/min) for diffusion runs at various pressures
might have a maximum error of ± 2%, which was the sum of
the errors from flowmeter calibration curves, experimental
temperature and pressure measurements, and reading the
ball float in the glass tubing. The corresponding average
error was estimated to be ±1% (i. e. 2-10 mls/min.).

As mentioned before, during diffusion runs, the
readings of the differential draft gauge were maintained
at the zero position with an accuracy of 0.005 inch of
water, which should have given negligible forced flow [4].
However, it is possible that forced flow existed, and
could cause appreciable errors. This aspect is discussed
in the next section.

If this effect does not seriously affect the value of
the diffusion coefficient calculated, as seems to be the
case, then the maximum error in determining the value of
diffusion coefficient should be about 5%, with a probable
average error of 3%. The experimental data appear to
conform to this estimate.

DISCUSSION

The most notable features of the results obtained in this work are the variation in the ratio of the two diffusion rates, and the constantcy of the $D_e \cdot P$ product, as the total pressure increased. This latter effect was expected, and was observed in most of the runs made. It also seemed logical to suppose that the ratio of the diffusion rates would remain constant, although there is no clear theoretical reason either for or against such a supposition, as pointed out by Hoogschagen[15].

It is not likely that the behavior observed for the diffusion ratio values was due to any of the more obvious possible sources of experimental error. For example, flowmeter calibrations under pressure were carried out using the same flow system, and with all connections the same, as in subsequent diffusion runs. The calibration of the thermal conductivity cells was independent of pressure effects, as the cells operated under a constant small pressure at all times. On several occasions the system was checked for leaks by leaving it at 100 psi with inlet and outlet valves closed. An hour or more was always required before even 5 psi drop in pressure occurred. Hence, there is no possibility of errors occurring due to leakage. The draft gauge which showed the pressure differential between the sample faces was located at the same level as the sample,

to avoid errors caused by gas density differences in manometer lines.

As pointed out in the previous section the usual experimental errors expected would not have caused a maximum error of more than 5% of the $D_e.P$ product. As a matter of fact, this is about the range of error actually found for duplicate runs.

It should be pointed out also that runs made at one atmosphere total pressure conformed to expectations. The ratio of diffusion rates was about as expected. Only a small difference in results was obtained by interchanging the two gases throughout the system, and a change in sample length did not affect results significantly.

Using the glass cell employed by Cox[4], and the same porous sample, the same value of effective diffusivity was obtained as that reported by Cox. In the case of Young's cell, (of the same type as that of Cox) a different porous sample was used, as well as a different differential gauge, again with satisfactory results. Therefore, it must be concluded, that at 1 atm. pressure and room temperature, no serious errors in measurements existed, and that bulk diffusion only was occurring through the solid sample.

With the exception of the runs in Series I, below 3 atms pressure, the variation of the effective diffusion

coefficient with pressure was precisely as expected i.e. the $D_e \cdot P$ product remained constant. However, the rather large changes in the ratios of the diffusion rates as pressure increased was not expected. Furthermore, the value of this ratio did not appear to be a function of pressure only, but also to some degree depended on the velocity of flow in the diffusion cell, the direction of the flow path, and the location of the differential pressure taps. This would seem to lead to the conclusion that some amount of forced flow was occurring through the sample, in addition to diffusive flow.

Table 7b presents some data on individual runs in which flow velocities were varied. In Series I, using Sample 03-B, it was only at the highest pressures that flow rate seemed to have a definite effect on the diffusion ratio (although this effect was usually small). For Sample 01-A, the effect of flow rate was noticeable, but this was to be expected, as the permeability of this sample was nearly 30 times as great as that for the 03-B solid. Runs using the 03-B sample with a baffled flow path to promote mixing did not show any effect due to velocity. Variations in these cases (Series III) could well be due to normal experimental error.

Data are shown in Table 7c, for runs using nearly the same flow rates, at two pressures, and with varying cell arrangements. The effect of poor mixing in the gas space above the sample is evident from the low value of

$D_e \cdot P$, and the high diffusion ratio, obtained for arrangement II (inlet and outlet diametrically opposite).

For a given geometric usage of the diffusion cell, a constant value for the $D_e \cdot P$ product was obtained. This was true for two widely different porous samples, as well as for different cell arrangements. It was also true if the gas mixing did not change greatly, and whether forced flow could be expected to occur readily or not.

A qualitative explanation can be offered for the constancy of the $D_e \cdot P$ product even though the relative rates of hydrogen and nitrogen diffusion are varying. In this work, the concentrations of the two streams after diffusion were very low, that is, of the order of 1% - 5%. Therefore in the diffusion equation applicable (equation 13 and 16), the quantities A_A and A_B are approximately 0 and 1 respectively. The equation can then be written,

$$D_e \cdot P = \left(\frac{RTL}{A}\right) \frac{A\ (N_A - N_B)}{\ln \dfrac{1-(1-\frac{N_B}{N_A})\,A_B}{1-(1-\frac{N_B}{N_A})\,A_A}} \doteq K \frac{N_A - N_B}{\ln \frac{N_A}{N_B}} \quad (21)$$

where K is a numerical constant equal to $\dfrac{RTL}{A}$.

The function $N_A - N_B / \ln\frac{N_A}{N_B}$ is the logarithmic average rate of diffusion of both gases. Hence, if this quantity remains constant, then the $D_e \cdot P$ product will also

TABLE 7b

Effect of Flow Velocity

Run No.	N_2 Flow mls/min std	H_2 Flow mls/min std	$\dfrac{V_H}{V_N}$	De. P	P atm	Remarks
35	445	212	4.81	.0648	3.04	Sample 03-B
37	423	342	4.84	.0644	3.04	Arrangement I
65	605	575	5.14	.0638	14.61	
69	800	322	5.48	.0642	14.61	Sample 03-B
70	1198	443	5.68	.0651	14.61	Arrangement I
93	414	517	4.93	.203	1.068	
94	237	523	4.74	.191	1.068	Sample 01-A
95	268	284	3.80	.213	1.068	Arrangement 1
131	440	519	4.90	.0579	11.2	
132	576	820	5.15	.0571	11.2	Sample 03-B
133	574	828	5.03	.0580	11.2	Arrangement 3
134	339	446	4.59	.0573	3.04	7/8" baffle
135	480	220	4.91	.0622	3.04	

TABLE 7c

Effect of Cell Arrangement

Run No.	N_2 Flow mls/min std	H_2 Flow mls/min std	$\dfrac{V_H}{V_N}$	De. P	P atm	Cell Arrangement
103	378	340	5.62	.0526	2.02	03-B Arr. II
119	381	355	4.71	.0583	"	03-B Arr. III 1/4" Baffle
138	371	327	4.64	.0602	"	03-B Arr. III 7/8" Baffle
56	358	382	4.64	.0612	"	03-B Arr. I
116	475	456	7.60	.0515	11.2	03-B Arr. II
126	481	604	6.16	.0593	"	03-B Arr. III 1/4" Baffle
131	440	519	4.90	.0579	"	03-B Arr. III 7/8" Baffle
68	620	648	5.14	.0632	"	03-B Arr. I

remain constant. If forced flow of one component e.g. A, is occurring, then the total rate of flow of A, N_A, must increase. If the total average flow of both gases is to remain constant, as defined above, then the rate of flow of B must necessarily decrease as that of A increases. Providing the forced flow of one component is not too large a fraction of the total flow, it apparently tends to cause a decrease in the diffetive rate of the second component such that the average mean flow is not seriously affected. Obviously there is a limit beyond which this apparently fortuitous phenomena could not be expected to occur, e. g. when the forced flow becomes too great.

An inspection of the data for individual runs shows that an increase in the diffusion rate of hydrogen did indeed cause a decrease in the nitrogen diffused. The exception to this general observation can be found in the runs reported as Series I. Here the experimental accuracy, particularly for the nitrogen analysis, is not as high as in other runs. The results of this series show an apparent increase of about 10% in the D_e.P product with pressure below 3.0 atms, which appears to be due to the fact that the hydrogen diffusion rate increased with little or no corresponding decrease in nitrogen diffusion rate. In all other sets of runs, using either different solids, or different choices for cell connections, it was generally

true that as the amount of hydrogen diffused increased, the nitrogen flow decreased. The usual amount of increase in the diffusion rate of hydrogen over the entire pressure range was about 10%-30%, and the decrease of nitrogen about 5-25%, depending on the conditions used. The data for sample Ol-A are plotted in Figure 14 which also shows a plot of the values of N_A and N_A/N_B which would give the same log mean average flow as that observed when forced flow was absent. It can be seen that, experimentally, the data does vary in such a way as to give a constant average flow, as indeed it must, since D_e.P is also constant in these runs.

It is interesting to note some data taken by Cox[4] to demonstrate that forced flow was absent in his work. His results show that the increase in hydrogen diffusion rate due to a pressure head was almost the same as the decrease in diffusion rate when hydrogen was diffusing against the same pressure head e. g. at +0.2 inch water the rate increased 9.0%, at -0.2 inch of water, it devreased 10.2%. This confirms the explanation given above for the constantcy of the D_e.P product, even though the quantities diffused may vary, or in spite of some degree of forced flow.

No really satisfactory explanation can be offered for the apparent existence of forced flow in this apparatus. Undoubtedly in some of the runs done e.g. with Sample Ol-A,
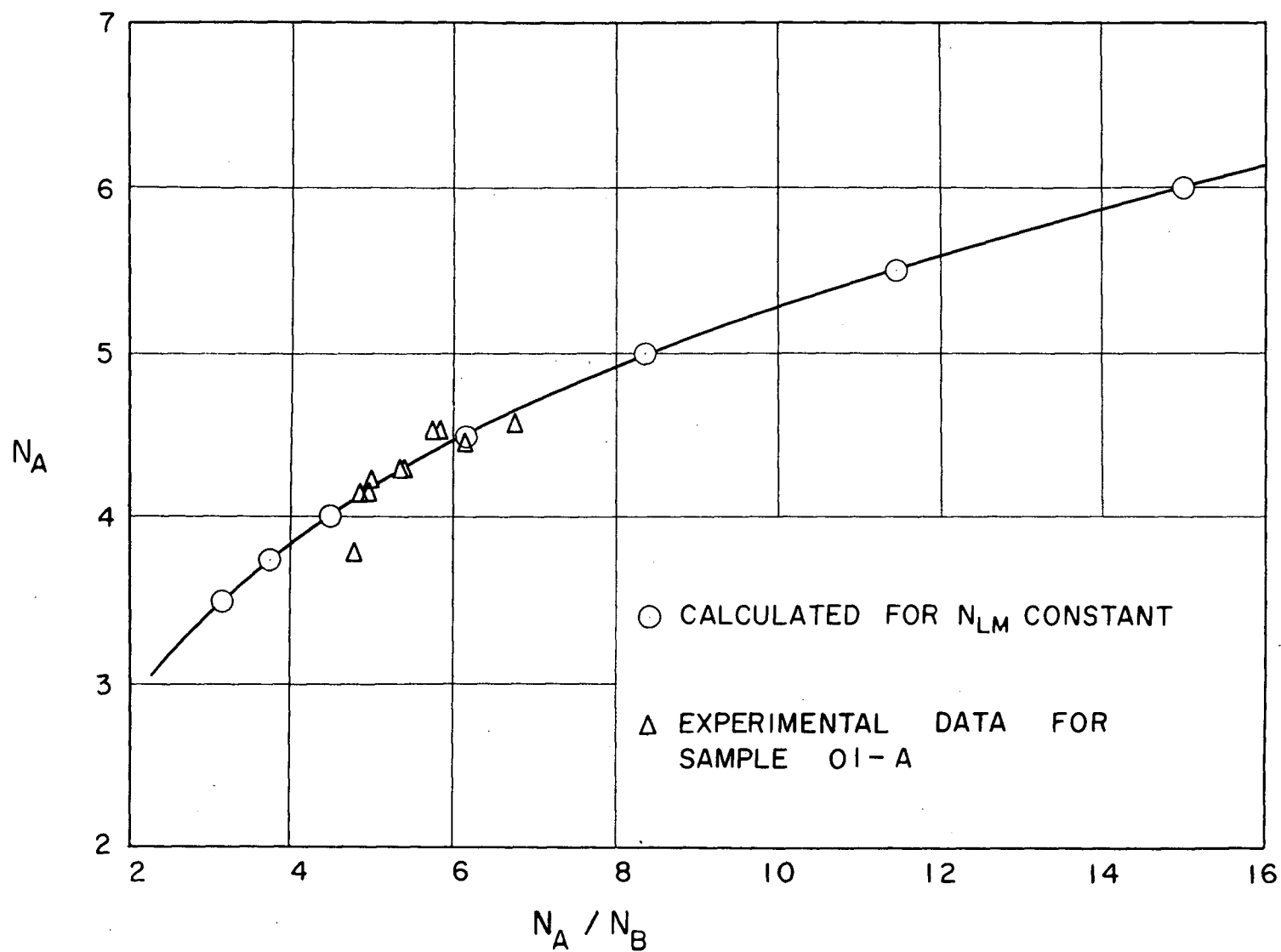
Figure 14.   Plot of N_A against N_A/N_B at Constant Log Mean Diffusion Rate for Sample 01-A, Arrangement 1.   (Experimental N_A values adjusted to Arbitary Ordinate Scale).

or with Arrangement II, either forced flow, or end resistance due to poor mixing, existed. Evidence for forced flow in other runs, was indirect. Calculations for forced flow in the O3-B sample using experimentally determined permeabilities and Darcy's law indicate that 1 psi differential is required to cause a forced flow of 1 ml of hydrogen per minute. Any possible impact pressures can be shown to be of the order of 0.01" water. The possibility also remains that the diffusion ratio is, in fact, a function of pressure. The observation that a complete reversal of the system (i. e. simply switching the two gas cylinders) gives practically identical results either way, cannot easily be explained on the basis of forced flow. It will be necessary to modify the diffusion cell, and perhaps the differential draft gauge arrangements, before the situation can be clarified.

However, the apparatus can be used successfully in its present form for some purposes. In investigating the transition region between Knudsen and ordinary diffusion, forced flow is usually a minor factor because of the very fine pore sizes involved, and therefore a small pressure unbalance is of little consequence.

When forced flow is present as a mechanism of transfer in addition to diffusive flow, it has been assumed that the two can simply be added. However, in view of the results obtained in this work, this would no longer be necessary

as long as the flow is largely diffusive in nature. Particularly in the field of reactions in porous solids this might present a simplification, since N_A/N_B is known from stoichoimetric relations. If there is an increase or decrease in volume on reaction, then some degree of forced flow exists through the porous solid in order that N_A/N_B may have the necessary value. Knowing this value, and the effective diffusivity of the solid, it should be possible to calculate the rate of the diffusive process for a binary system of known kinetics, using the diffusion equation only.

CONCLUSIONS AND RECOMMENDATIONS

The present diffusion cell and differential pressure measuring system should be modified in such a way as to give good gas space mixing and true static pressure reading at the solid sample faces.

The apparatus appears to be most suitable for varying the mean free path of diffusing gas systems by the application of pressure. This allows the nature of the diffusion mechanism occurring in a porous solid to be varied easily. Even if forced flow were completely eliminated, the method is not particularly suitable for measuring the variation of the $D_T.P$ product with absolute pressure. This is due to the necessity for multiple ranges and calibrations of flowmeters.

A substitute for the type of flowmeter used here would simplify experimental work. Some reason should be found for the necessity of throttling the nitrogen outlet line to achieve pressure balance in the cell, when, in theory, it is the hydrogen line which should require throttling.

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APPENDIX

Sample Calculation

Effective Diffusion Coefficient and Diffusion Ratio

Run 37. Data recorded as below

Sample	Selas 03-B
Length L	3.157 cm
Average Cross Sectional Area, A	5.376 cm^2
Temperature, T^o K	294°
Absolute Pressure, P. atm	3.04 atm
Nitrogen flow rate, inlet, X_N	423 $\frac{mls}{min}$ std.
Hydrogen flow rate, inlet, X_H	342 " " "
M.V. on No. 1 T-C cell	7.309 M.V.
M.V. on No. 2 T-C cell	26.15 M.V.
Nitrogen % in outlet H$_2$. N_H%	0.785%
Hydrogen % in outlet N$_2$. H_N%	2.915%

Calculation:

By means of a material balance, based on constant pressure. We can calculate the outlet flow rate of hydrogen and nitrogen, i.e. Y_H and Y_N. Therefore the volume rate of hydrogen (V_H) and nitrogen (V_N) diffused are the products of Y_N. H_N% and Y_H. N_H% respectively.
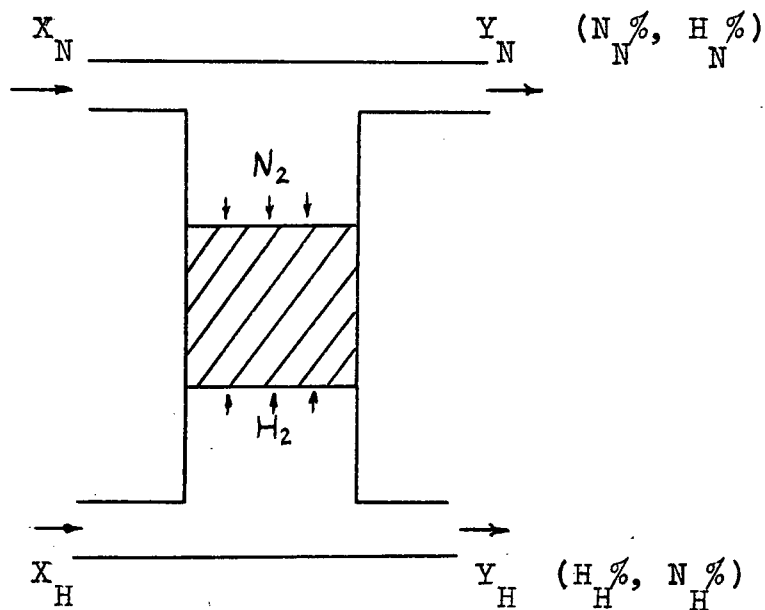
Where

Y_N Outlet nitrogen flow rate   mls/min std.

Y_H Outlet hydrogen flow rate   mls/min std.

N_N Mole fraction of nitrogen in outlet nitrogen stream.

H_N Mole fraction of hydrogen in outlet   "   "

N_H Mole fraction of nitrogen in outlet hydrogen   "

H_H Mole fraction of hydrogen in outlet   "   "

V_H Rate of diffusion of hydrogen   mls/min std.

V_N Rate of diffusion of nitrogen   mls/min std.

A material balance over the system gives

$$X_N + X_H = Y_N + Y_H \tag{1}$$

$$Y_H \cdot N_H + Y_N \cdot N_N = X_H \tag{2}$$

Solve (1) and (2) for Y_H and Y_N, we have $Y_H = \dfrac{X_H(1-H_N) - X_N \cdot H_N}{H_H - H_N}$

or $Y_H = \dfrac{X_H - X_N \left(\dfrac{H_N}{N_N}\right)}{H_H - N_H \left(\dfrac{H_N}{N_H}\right)}$

$$\tag{3}$$

$$Y_N = X_N + X_H - Y_H \qquad (4)$$

$$V_H = Y_N \cdot H_N \qquad (5)$$

$$V_N = Y_H \cdot N_H \qquad (6)$$

For run 37, substitute the known value into equation (4), (5) and (6)

Thus $Y_H = \dfrac{342 - 423\ (\frac{.02915}{.97085})}{.99215 - .00785\ (\frac{.02915}{.97085})} = \qquad 332 \qquad$ ml/min.

$Y_N \qquad\qquad\qquad\qquad\qquad\qquad = \qquad 433 \qquad$ ml/min.

$V_H \qquad\qquad\qquad\qquad\qquad\qquad = \qquad 12.62 \qquad$ ml/min.

$V_N \qquad\qquad\qquad\qquad\qquad\qquad = \qquad 2.606 \qquad$ ml/min.

$\dfrac{V_H}{V_N} = $ ratio of diffusion $\qquad = \qquad 4.84$

We can now apply equation (16) derived from the Sherwood and Pigford equation and calculate the Maxwell diffusion coefficient.

Let hydrogen be component A.

Then

$$D_e \cdot P = \frac{V_H \left(\frac{T_{rm}}{T_{std}}\right) \frac{1}{60} \left(1 - \frac{N_{N_2}}{N_{H_2}}\right) \frac{L}{A}}{Ln\ \dfrac{1 - \left(1 - \frac{N_{N_2}}{N_{H_2}}\right) \cdot H_N}{1 - \left(1 - \frac{N_{N_2}}{N_{H_2}}\right) \cdot H_H}}$$

Where T_{rm} = Room temperature, 294°

T_{std} = 294.1° K (70°F)

L = 3.157 cm

A = 5.376 cm^2

$$1 - \frac{N_{N_2}}{N_{H_2}} = 1 - \frac{V_N}{V_H} = 0.7934$$

H_N = .02915   mole fraction

H_H = .99215   mole fraction

V_{H} = 12.62   mls per min.

$$De.P = \frac{12.62\ (1)\ \frac{1}{60}\ (0.7934)\ \frac{3.157}{5.376}}{\ln \frac{1 - 0.7934\ (.02915)}{1 - 0.7934\ (.99215)}}$$

$$= \frac{.0980}{1.521}$$

$De.\ P$ = 0.0644 $\frac{cm^2}{sec.}$ (atm). where P = 3.04 atm.

$D_T.\ P$ = 0.7657 at 294.1°K.

TABLE 4a

Diffusion Results of Individual Runs for Sample 2

Arrangement I

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	at 21.1°C De. P	P,atm	Trm °C
4	10.72		(3.74)*	.0590	1.068	22
5	10.43		"	.0582	"	"
6	10.43		"	.0583	"	"
10	10.75		"	.0594	"	"
11	10.36		"	.0574	"	23
12	10.73		"	.0591	"	23
Average	10.57			.0584		
7	11.27		(4.05)	(.0610)	1.2	22
8	10.97		"	(.0595)	"	22
9	11.00		"	(.0595)	"	22
Average	11.08			.0598		
13	11.09		(4.21)	(.0589	1.34	23.5
14	11.15		(4.21)	(.0595	"	"
52	11.21	2.605	4.310	.0601	"	22
53	11.38	2.633	4.322	.0605	"	22
54	11.39	2.473	4.606	.0590	"	22
Average	11.18	2.572	4.42	.0599		
15	11.36		(4.46)	(.0695)	1.681	19.5
16	11.37		"	(.0607)	"	"
17	11.88		"	(.0629)	"	"
18	11.60		"	(.0618)	"	"
Average	11.55			(.0615)		
19	11.50		(4.645)	(.0604)	2.02	19.8
20	11.89		"	(.0632)	"	"
21	11.70		"	(.0611)	"	"
55	12.03	2.592	4.641	.0620	"	22
56	11.83	2.549	4.641	.0612	"	22
57	12.12	2.507	4.834	.0613	"	22
72	12.17	2.606	4.671	.0606	"	20.4
Average	11.89	2.564	4.673	.0614		

<div align="center">

TABLE 4a (cont'd)

</div>

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P	P.atm	Trm $^{\circ}C$
22	11.46		(4.82)	(.0585)	2.71	20.4
23	12.60		"	(.0655)	"	"
24	12.58		"	(.0651)	"	"
Average	12.21			(.0632)		
25	11.64		(4.865)	(.0603)	3.04	20.4
26	12.60		(4.965)	.0640	"	20.4
36	12.62	2.623	4.81	.0651	"	20.5
37	12.62	2.606	4.84	.0646	"	20.5
Average	12.37	2.615	4.83	.0649		
27	13.13		(5.02)	(.0681)	4.376	19
28	12.20		"	(.0628)	"	19
29	11.90		"	(.0615)	"	19
30	13.55		"	(.0685)	"	19.2
41	12.805	2.800	4.573	.0666	"	20.8
42	12.645	2.165	5.840	.0604	"	20.8
43	13.00	2.445	5.330	.0628	"	20.8
Average	12.77	2.470	5.019	.0633		
38	13.1	2.551	5.13	.0650	6.1	20.8
39	12.84	2.482	5.17	.0632	"	20.8
40	12.81	2.499	5.13	.0632	"	20.8
Average	12.92	2.511	5.14	.0638		
44	13.18	2.481	5.31	.0639	7.8	20.4
45	13.16	2.581	5.10	.0655	"	"
46	12.65	2.693	4.69	.0650	"	"
47	13.03	2.579	5.02	.0645	"	"
Average	13.01	2.583	5.03	.0647		
58	14.04	2.724	5.154	.0661	9.503	21.4
59	13.62	2.714	5.017	.0648	"	22
60	13.40	2.633	5.089	.0634	"	22
Average	13.69	2.691	5.09	.0648		

TABLE 4a (cont'd)

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P	P atm	Trm $^\circ$C
61	13.36	2.548	5.243	.0634	11.204	23
62	13.69	2.490	5.498	.0629	"	"
63	13.26	2.508	5.287	.0624	"	"
64	13.32	2.594	5.135	.0631	"	21.5
68	13.46	2.618	5.141	.0645	"	20
Average	13.42	2.553	5.343	.0633		
65	13.30	2.588	5.139	.0630	14.605	21.5
69	13.76	2.51	5.482	.0642	"	20
70	14.14	2.489	5.681	.0646	"	20.4
71	14.23	2.567	5.543	.0655	"	20.4
Average	13.86	2.544	5.434	.0644		

* Values in parantheses interpolated.

TABLE 5a

Diffusion Results of Individual Runs for Sample 3

Arrangement I

Run No.	Pressure atm	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21°C
93	1.068	24.9	5.055	4.925	.2028
94	"	22.66	4.787	4.74	.1907
95	"	22.72	5.984	3.80	.2134
Average		23.09	5.275	4.49	.2025
91	1.34	24.80	5.125	4.84	.2065
92	"	25.4	4.967	4.97	.2105
Average		25.1	5.046	4.91	.2085
87	1.681	25.65	4.83	5.33	.2046
88	"	25.73	4.81	5.35	.2043
89	"	27.23	4.685	5.82	.2060
90	"	27.00	4.41	6.12	.2055
Average		25.65	4.67	5.51	.2051
83	3.04	27.6	4.095	6.75	.1965
84	3.04	27.4	4.75	5.77	.2100
Average		27.5	4.43	6.26	.2033

TABLE 6a

Diffusion Results of Individual Runs for Sample 2

Arrangement II

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De.P at 21.1°C	Pressure atm	Note
100	10.276	2.104	4.884	(.05267)	1.338	
101	10.48	2.19	4.785	.0543	1.34	
102	10.478	2.232	4.694	(.0546)	1.34	$0.1"\,H_2 < N_2$ [*]
Average	10.41	2.175	4.788	.0538		
103	11.15	1.985	5.617	(0.0526)	2.02	$0.03"\,H_2 > N_2$
104	11.143	1.979	5.631	1.0536	2.02	
105	11.25	2.047	5.496	(0.0547)	2.015	
106	10.95	1.951	5.610	(0.0528	2.08	
107	11.15	1.933	5.768	(0.0531)	2.035	
108	11.18	1.900	5.884	0.0532	2.02	
Average	11.17	1.965	5.668	0.0533		
109	11.33	1.953	5.801	(.0537)	3.04	$0.05"\,H_2 > N_2$
110	11.31	1.952	5.794	.05365	3.04	
Average	11.32	1.953	5.798	.0537		
111	11.35	1.950	5.821	.05385	4.376	
112	11.35	1.961	5.787	(.0540)	4.376	$.06"\,H_2 > N_2$
Average	11.35	1.955	5.804	.0539		
114	12.10	1.696	7.137	(.0532)	11.204	$.08"\,H_2 > N_2$
115	11.87	1.775	6.687	.0545	11.204	
116	11.99	1.577	7.603	.0515	11.204	
Average	11.99	1.683	7.142	.0531		

[*]Pressure of the hydrogen side was 0.1" water greater than that of the nitrogen side.

( ) Values within this bracket allowed an experimental error either in differential pressure or in total pressure.

TABLE 7d

Diffusion Results of Individual Runs for Sample 2

Arrangement III (with 7/8" vertical baffle)

Run No.	V_H	V_N	$\dfrac{V_H}{V_N}$	De. P at 21.1°C	Pressure absolute	Note
140	11.35	2.463	4.608	0.0583	1.34	
141	11.32	2.447	4.626	0.0579	"	
142	(11.26)	(2.462)	(4.574)	(0.0572)	"	.11"H_2O $H_2 < N_2$
Average	11.33	2.455	4.617	0.0581		
138	11.60	2.499	4.643	.0597	2.02	
139	11.89	2.601	4.571	.0611	"	
Average	11.71	2.55	4.607	.0604		
134	11.055	2.41	4.587	.0570	3.04	
135	12.17	2.48	4.907	.0619	"	
Average	11.61	2.45	4.747	.0595		
136	11.59	2.415	4.799	.0589	4.376	
137	11.598	2.468	4.699	.0575	"	
Average	11.594	2.441	4.747	.0584		
143	11.45	2.299	4.980	.0569	7.8	
144	(11.585)	(2.335)	(4.960)	(.0578)	7.79	
Average	11.507	2.315	4.970	.0574		
127	11.70	2.467	4.743	.0590	11.204	
129	(11.463)	(2.335)	(4.909)	(.0585)	"	.09"H_2O $H_2 < N_2$
130	11.65	2.332	4.996	.0583	"	
131	11.52	2.350	4.902	.0576	"	
132	11.64	2.262	5.146	.0570	"	
133	11.73	2.333	5.028	.0578	"	
Average	11.67	2.330	4.954	.0579		

TABLE 8

Ratio of Equal Pressure Diffusion Rates

for Pairs of Gases, 1 atm

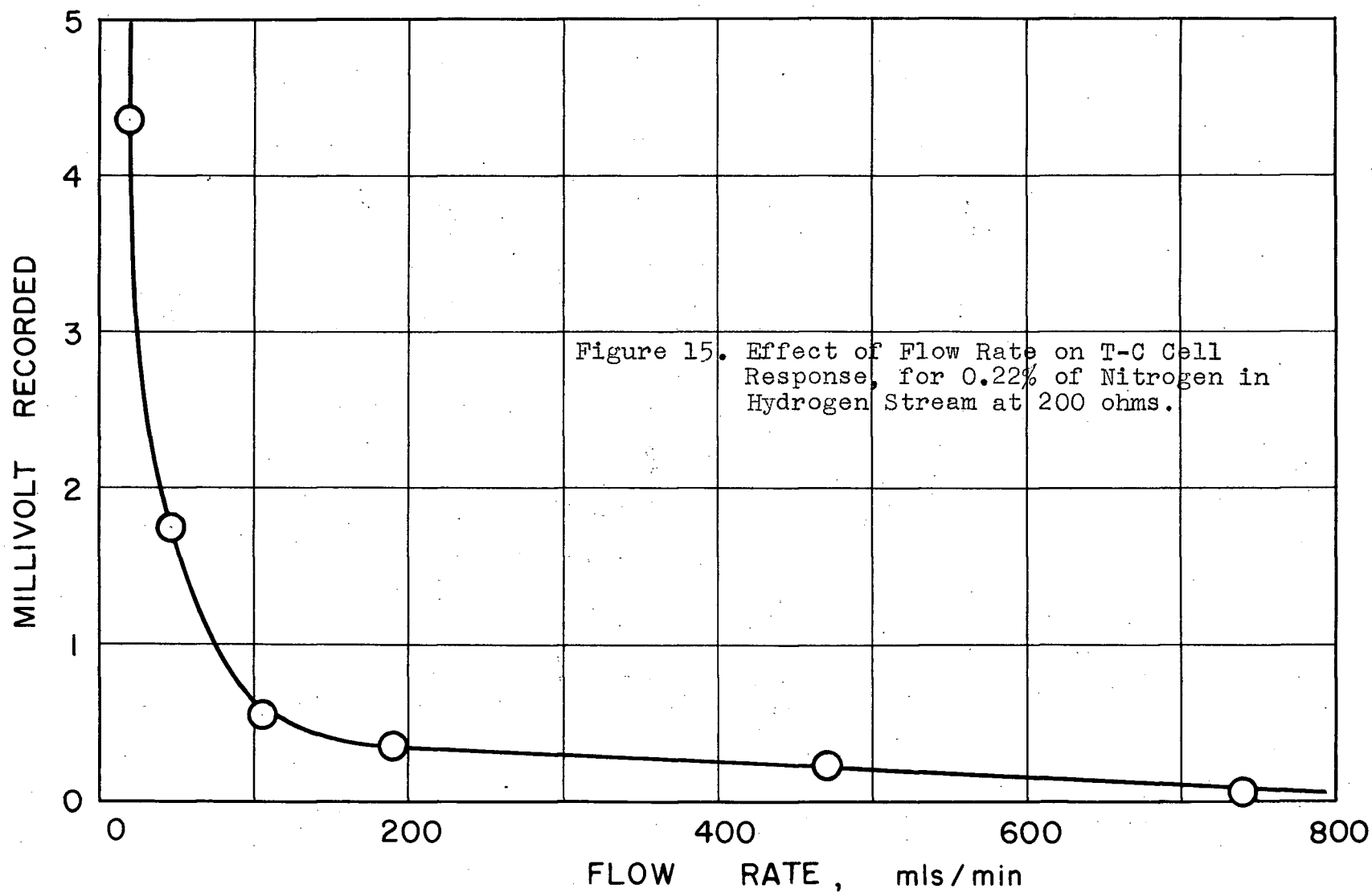| Gases	Diffusion Ratio	
	Observed	Theoretical from eq.(18)
Helium-oxygen[1]	3.03 2.66 2.54	2.83
Nitrogen-oxygen[1]	1.09 1.07	1.07
Carbon dioxide-oxygen[1]	0.89 0.80	0.85
Hydrogen-nitrogen[2]	3.44 4.03 3.75	3.742

1. Data from Hoogschagen. [15]

2. Data from Cox. [4]
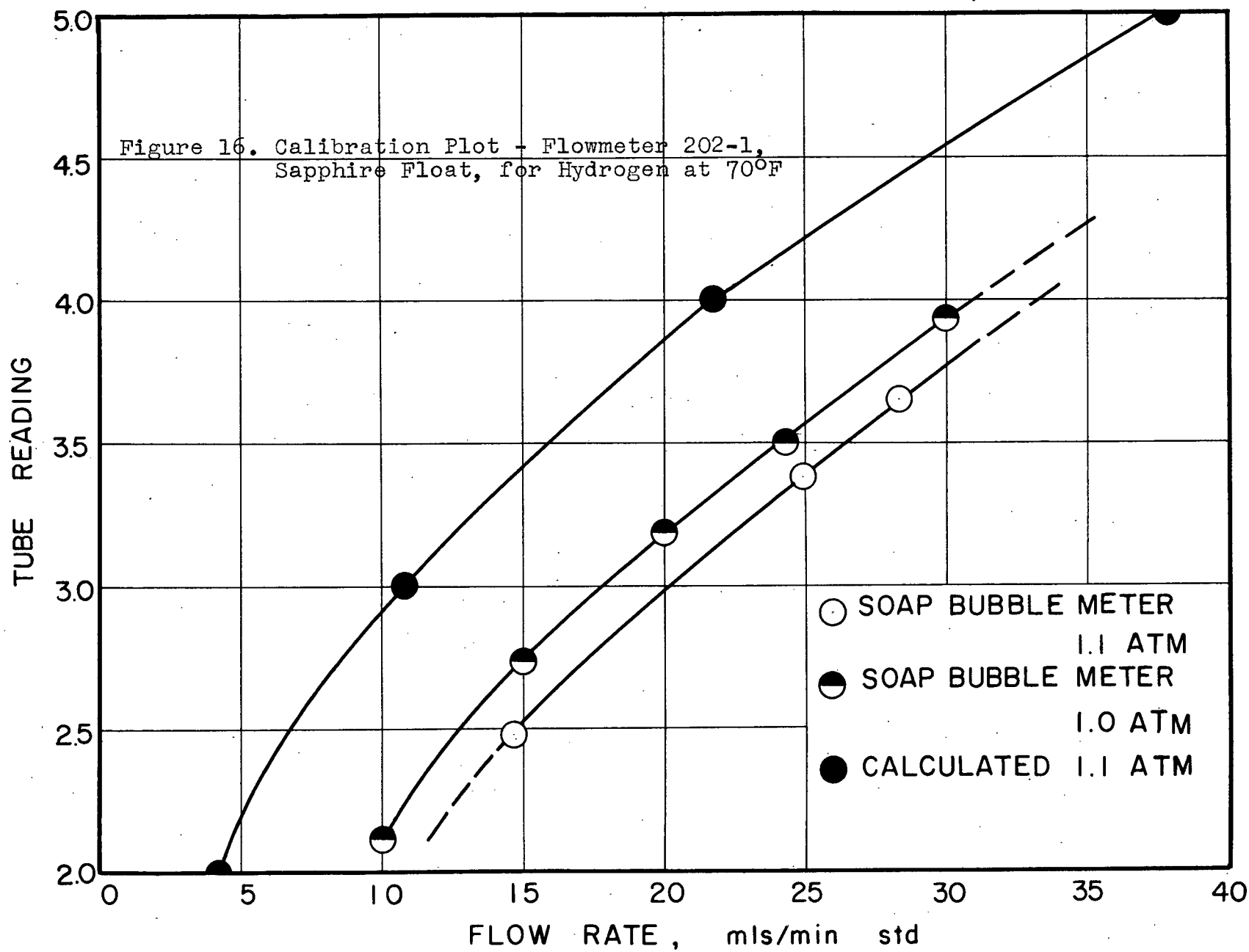
TABLE 9

Calculated Calibration Results for Flow Meters

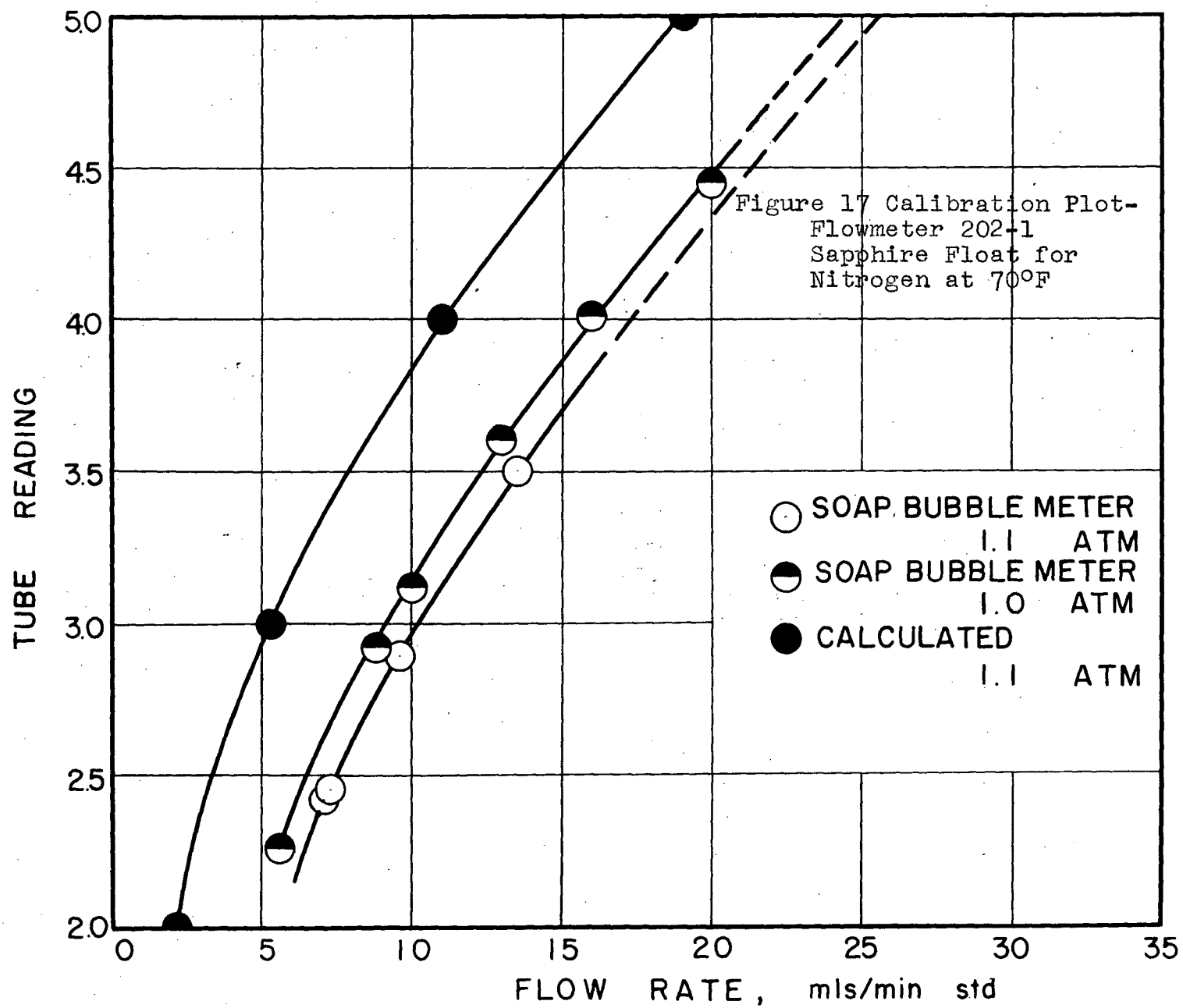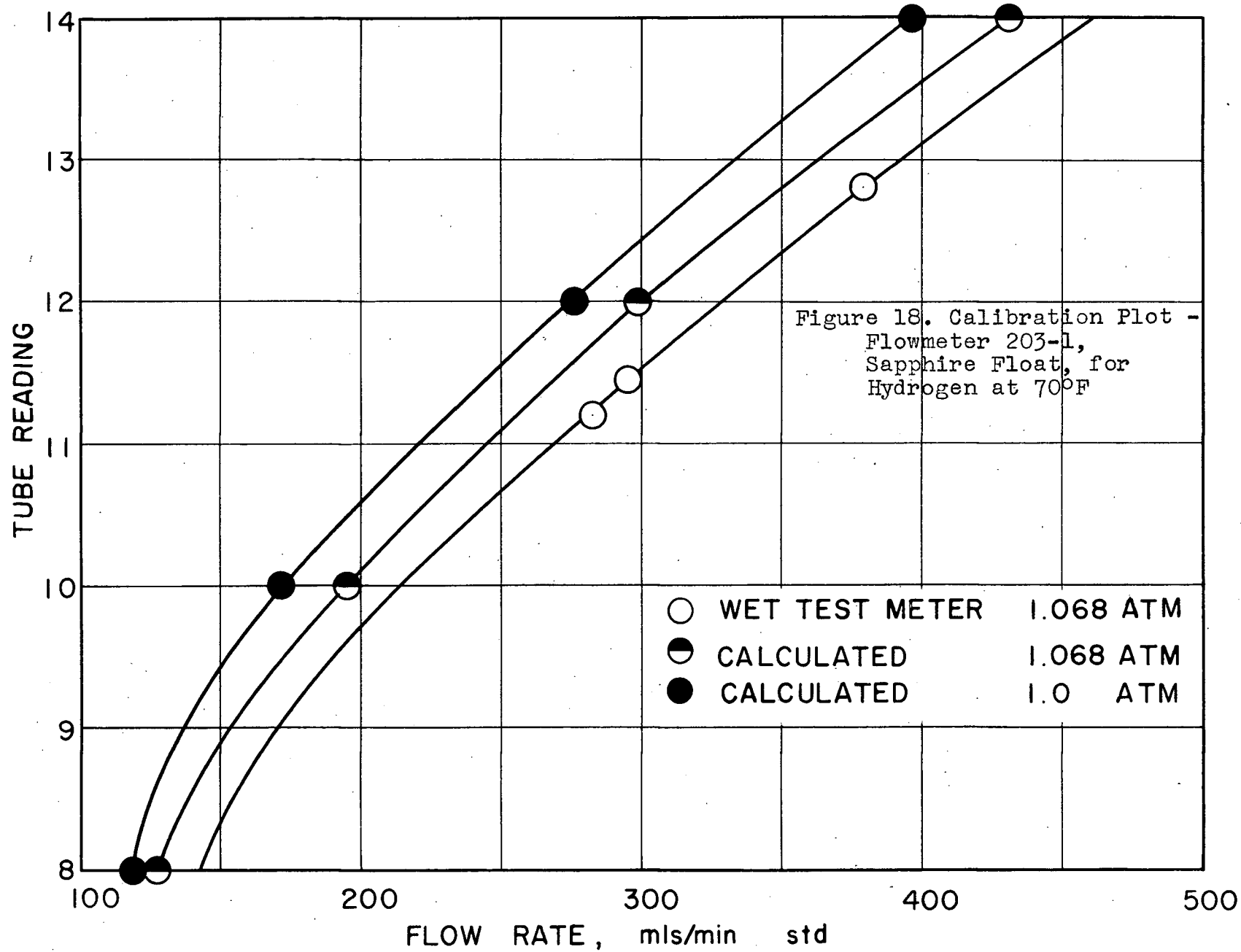| Gas.	No.of Flowmeter	Tube Reading	Flow Rate $\frac{Mls}{min}$ 70°F 1 atm		
			70°F 150 psig	75°F 150 psig	70°F 150 psig
H_2	203-1	3	167	166.9	167.9
		4	284.7	284.5	286.1
		5	454.5	454.2	455.5
		6	669	671.4	673.7
		7	877.8	877.1	878.1
N_2	203-2	7	503.7	501.4	502.0
		8	597.3	592.8	595.0
		10	788.7	784.4	786.7
		12	1003.8	999.9	1001.7
			70°F 15 psig	75°F 15 psig	70°F 15.3 psig
H_2	203-1	7	177.6	176.3	178.9
		8	241.5	239.1	243.6
		10	371.2	369.1	374.9
		12	600.8	595.8	605.9
		14	730.3	725.4	736
N_2	203-2	8	193.3	193.2	196.7
		10	264.4	262.9	267.4
		12	339.3	337.9	342.7
		14	432.5	429	436.4
		16	520.3	517.2	524

TABLE 10

Example of Complete Recorded Data for Runs (Arrangement I)

Sample No.	Run No.	P atm	Flor Rate mls/min. 21°C, 1 atm Nitrogen inlet	Hydrogen inlet	No. 2 M.V.	$H_2\%$ in N_2	V_H	No. 1 M.V.	$N_2\%$ in H_2	V_N	$\dfrac{V_H}{V_N}$	De. P
2	52	1.34	324.6	274.5	30.02	3.365	11.2	7.343	0.980	2.605	4.30	.0606
"	53	1.34	382	275.5	26.11	2.912	11.38	9.100	0.987	2.633	4.32	.0614
"	54	1.34	330	319	30.02	3.364	11.39	7.343	0.797	2.473	4.61	.0612
2	36	3.04	411.7	305	26.85	2.993	12.62	8.379	.889	2.623	4.81	.0651
"	37	3.04	423	342	26.15	2.915	12.62	7.309	.785	2.606	4.84	.0646
2	41	4.376	512	331	22.0	2.447	12.805	8.064	.875	2.800	4.57	.0666
"	42	4.376	458	256	24.23	2.695	12.645	8.196	.889	2.165	5.84	.0604
"	43	"	566	606	20.23	2.253	13.000	3.785	.411	2.445	5.33	.0628
"	65	14.605	605	515	19.08	2.136	13.30	4.723	0.513	2.588	5.139	.0630
"	69	"	800	322	15.15	1.687	13.76	7.445	0.808	2.51	5.482	.0642
"	70	"	1197.5	443	10.45	1.170	14.14	5.325	0.577	2.489	5.681	.0646
"	71	"	1670	470	7.55	0.845	14.23	5.166	0.560	2.567	5.543	.0655
3	87	1.681	374	515	58.2	6.515	25.65	9.125	.9775	4.825	5.325	.2046
"	88	"	376	506	58.0	6.485	25.73	9.249	.99	4.81	5.35	.2043
"	89	"	557	720	41.96	4.700	27.23	6.225	.671	4.685	5.82	.2060
"	90	"	556.5	630	41.67	4.670	27.00	6.76	.727	4.41	6.12	.2055

Figure 15. Effect of Flow Rate on T-C Cell
Response, for 0.22% of Nitrogen in
Hydrogen Stream at 200 ohms.

MILLIVOLT RECORDED

FLOW   RATE ,   mls / min

Figure 16. Calibration Plot - Flowmeter 202-1,
Sapphire Float, for Hydrogen at 70°F

TUBE READING

FLOW RATE, mls/min std

○ SOAP BUBBLE METER
    1.1 ATM
◑ SOAP BUBBLE METER
    1.0 ATM
● CALCULATED 1.1 ATM

Figure 17 Calibration Plot-
Flowmeter 202-1
Sapphire Float for
Nitrogen at 70°F

SOAP BUBBLE METER
1.1 ATM
SOAP BUBBLE METER
1.0 ATM
CALCULATED
1.1 ATM

TUBE READING

FLOW RATE, mls/min std

Figure 18. Calibration Plot – Flowmeter 203-1, Sapphire Float, for Hydrogen at 70°F

○	WET TEST METER	1.068 ATM
◓	CALCULATED	1.068 ATM
●	CALCULATED	1.0 ATM

TUBE READING

FLOW RATE, mls/min std

Figure 19 Calibration Plot - Flowmeter 203-1, Sapphire Float, for Hydrogen at 70°F and 7.8 atms Absolute

WET TEST METER
CALCULATED

TUBE READING

FLOW RATE, mls/min std

Figure 20. Calibration Plot - Flowmeter 203-2, Steel Float for Nitrogen at 70°F and 7.8 atms Absolute
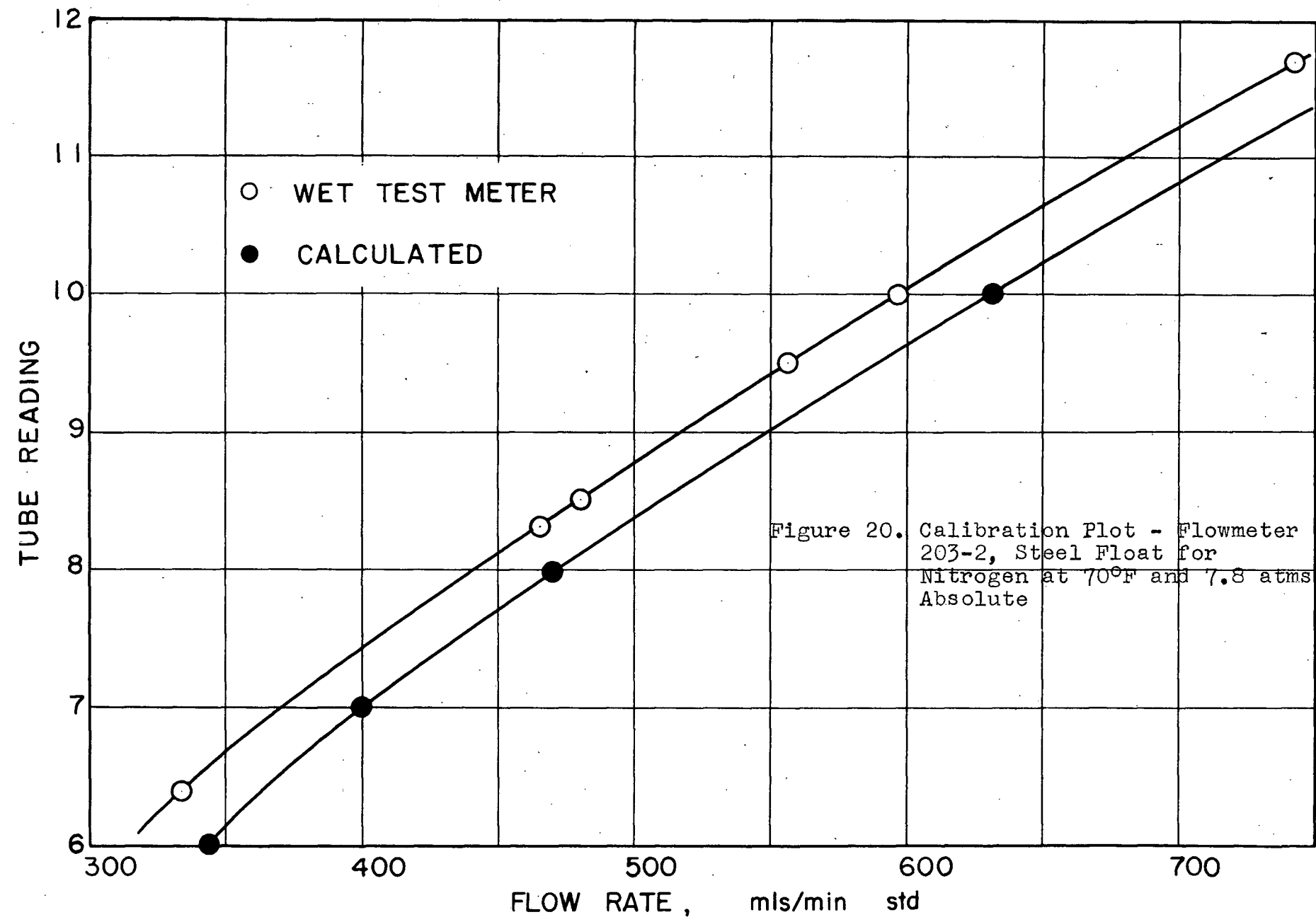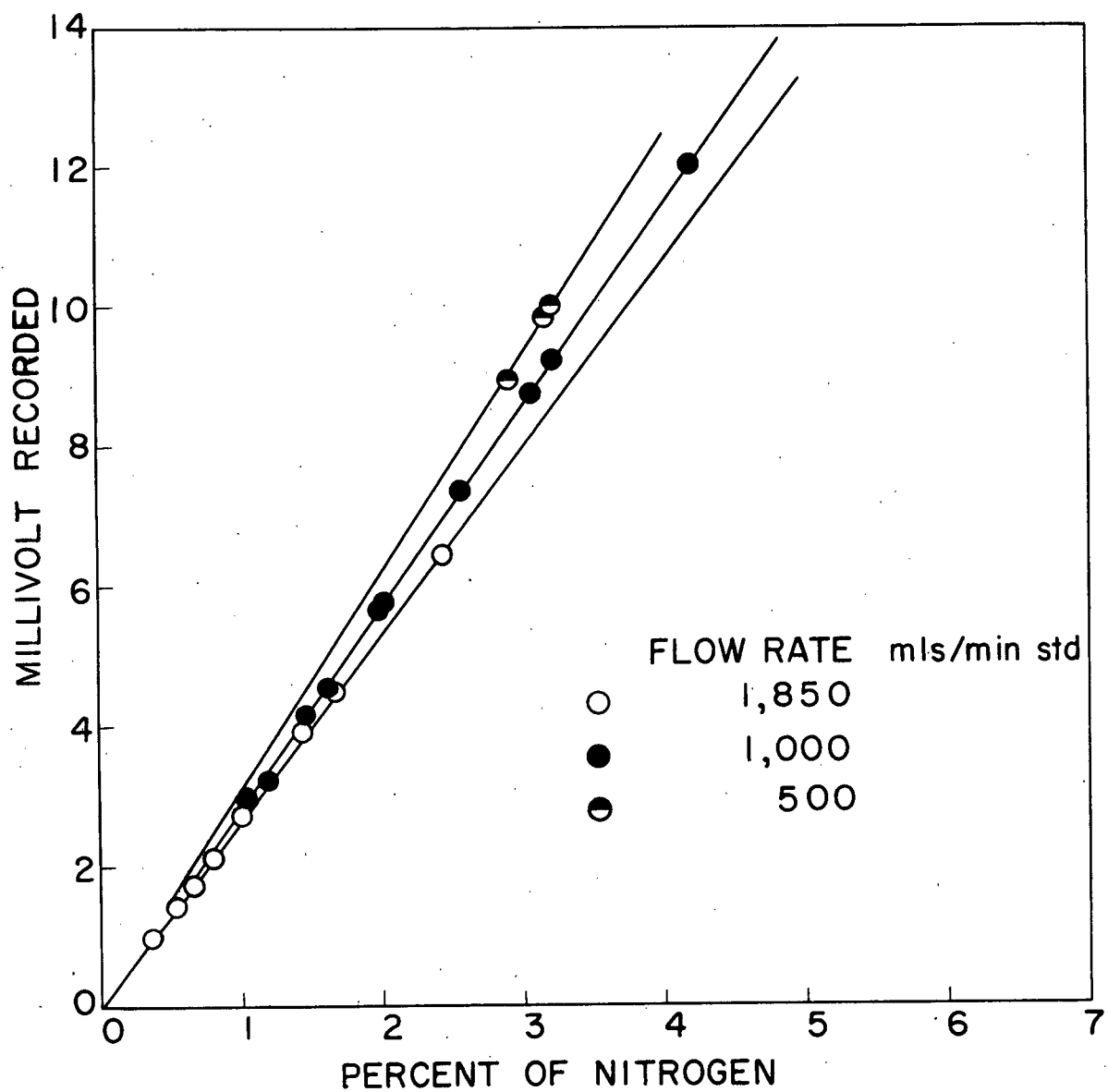
Figure 21. Calibration Plot for No. 1. Thermal Conductivity
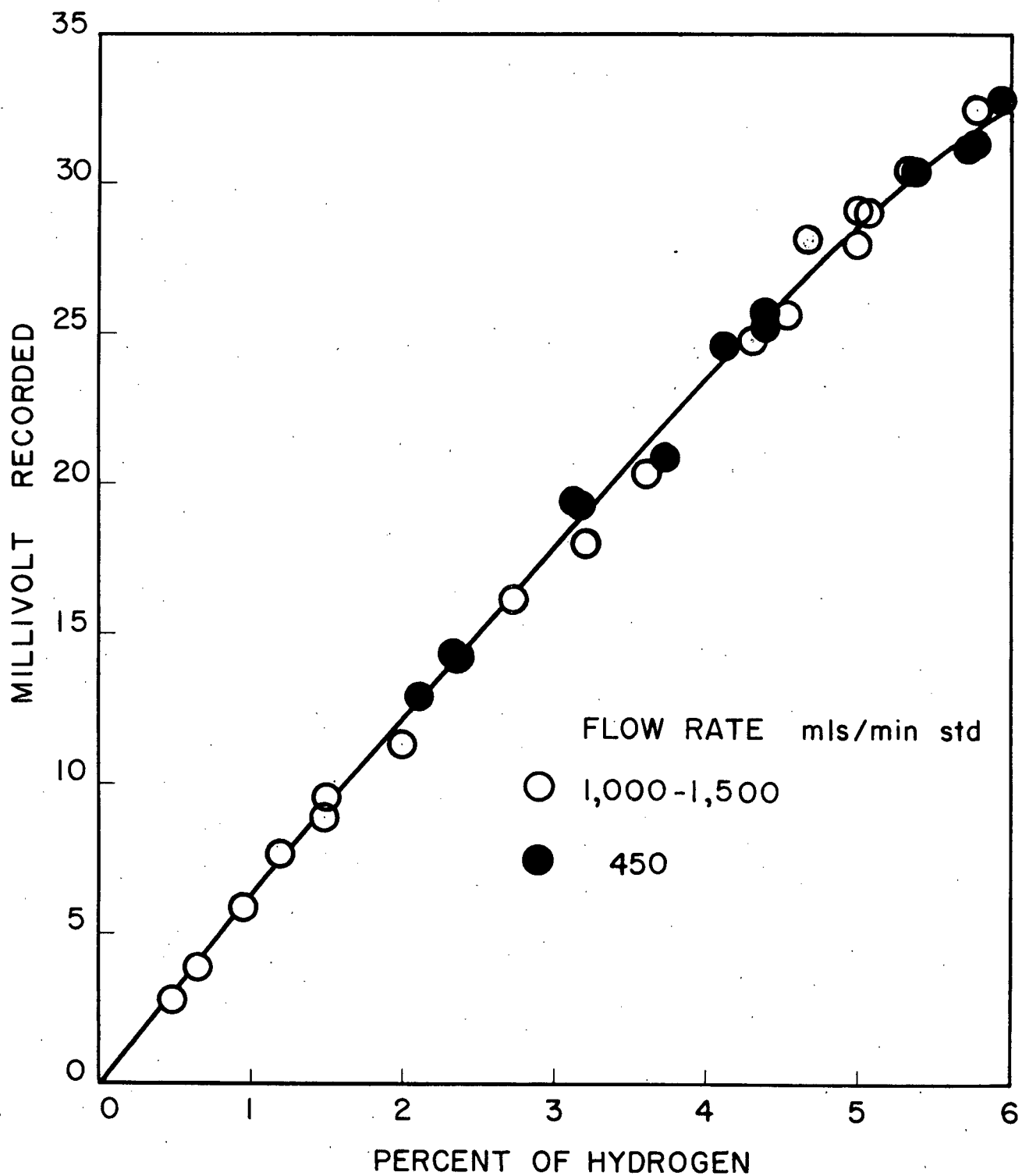Cell before Modification

Figure 22. Calibration Plot for No. 2, Thermal Conductivity Cell
before Modification