

REPORT NO. 39

THE VISCOSITY OF
VAPOR MIXTURES OF
HYDROGEN PEROXIDE
AND WATER

Prepared for the
Office of Naval Research
Contract No. N5ORI-07819
NR-092-008

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C. N. Satterfield
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MASSACHUSETTS INSTITUTE OF TECHNOLOGY
Department of Chemical Engineering Cambridge, Mass

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TABLE OF CONTENTS

	Page
ABSTRACT	1
INTRODUCTION	3
APPARATUS AND PROCEDURE	5
Capillary Tube	6
Auxiliary Equipment	8
Capillary Tube Dimensions	9
EXPERIMENTAL RESULTS	10
Calibration with Helium, Air, and Water Vapor	10
Viscosity of Oxygen - Water Vapor Mixtures	14
Corrected Poiseuille Equation	16
Measurements on Hydrogen Peroxide	17
DISCUSSION OF RESULTS	19
Precision of the Measurements	19
Comparison of Results with Theoretical Values	20
Comparison with Other Systems	24
ACKNOWLEDGMENT	26
NOTATION	27
LITERATURE CITED	29
FIGURES	
APPENDIX	A1
Tables of Experimental Results	
Density of Fluorolube FS	A5
Conversion of Viscosity Units	A5
Sample Calculation	A5

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by

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ABSTRACT

The viscosity of vapor mixtures of hydrogen peroxide and water containing up to 65 mole % hydrogen peroxide was measured by a capillary flow method at one atmosphere total pressure and at a temperature of 170°C. By extrapolation, the viscosity of anhydrous hydrogen peroxide vapor at 170°C was found to be 144 micropoises. A few studies were also made at 200 and 240°C to provide a basis for estimating the effect of temperature. The sensitivity of hydrogen peroxide vapors to heterogeneous decomposition and their explosiveness made it necessary to develop special apparatus for generation and manipulation of these superheated vapors at concentrations above the explosion limit. A suitable all-glass apparatus made it possible to produce vapors containing less than one mole % of oxygen, and it was demonstrated that this low oxygen concentration did not affect the precision of measurement.

The results of this work may be expressed by the following equation:

$$\mu = 134 + 0.35(t - 100) - 14y$$

where

μ = viscosity, micropoises

t = temperature, °C

y = composition, mole fraction hydrogen peroxide

This equation is recommended for the calculation of the viscosity of vapor mixtures of hydrogen peroxide and water over the temperature range of about 100 to 300°C, and is believed to have a precision of within $\pm 2\%$. The results provide a measure of the viscous forces in a highly polar gaseous system; few such systems have been studied previously.

* * * * *

INTRODUCTION

The object of the work reported here was the measurement of the viscosity of hydrogen peroxide-water mixtures in the vapor state. Measurements were made at hydrogen peroxide concentrations up to the highest which could be safely attained.

Two reasons exist for the interest in the results of these studies: a) The use of hydrogen peroxide as an energy source requires knowledge of such physical properties as vapor viscosity in the design of power producing devices. b) Such measurements add to our knowledge of the transport properties of highly polar gases. Although non-polar gases have been frequently studied, relatively little information is available on mixtures of two highly polar gases.

The method of determining viscosity used here is that of measurement of the pressure drop occurring upon flowing hydrogen peroxide vapor through a capillary tube at constant temperature. The mathematical relation between the viscosity and the measured quantities is given by the Poiseuille equation:

$$\mu = 10^6 \pi^4 (\Delta p) / 8qL \quad (1)$$

Equation (1) is valid only when the flow is streamlined. The derivation of this equation is available in the monographs on viscosity by Barr (2) and Merrington (15). In practice the measured pressure drop will include contributions from energy losses at the entrance and exit of the tube. These must be

minimized and corrected for obtain the pressure drop due to flow through the tube on , which is required for use in Equation (1). The nature of these additional energy losses and the means for minimizing them are discussed in the monographs cited (2,15). In the present work allowance was made for such energy losses by calibration of the apparatus with gases of known viscosity.

Additional experimental difficulties introduced in working with hydrogen peroxide vapor are as follows:

a. Hydrogen peroxide vapor is highly reactive. In addition, at atmospheric pressure mixtures with inert gases containing above about 26 mole per cent hydrogen peroxide vapor will readily explode on contact with a wide variety of substances which are catalytically active, particularly the heavy metals.

b. Being a condensible vapor, care must be taken to avoid condensation and the presence of mist.

c. Decomposition of hydrogen peroxide in the apparatus changes the composition of the system, introduces a third component, oxygen, and causes difficulty in temperature control.

The following alterations to equation (1) must thus be made in order that the viscosity may be calculated from the experimentally determined pressure drop and mass rate of flow.

a. Since the fluid under measurement is compressible, equation (1) must be integrated to allow for the changing value of the volumetric rate of flow as the fluid passes through the capillary tube. This results in the equation:

$$\mu = 10^6 \pi r^4 M (p_1^2 - p_2^2) / 16 L W R T a \quad (2)$$

b. A correction factor, K_E , which allows for kinetic energy losses, is introduced in equation (2).

c. A correction factor, K_O , which allows for the presence of small proportions of oxygen is introduced in equation (2).

The viscosity of hydrogen peroxide vapor may then be calculated from the measured quantities, p_1 , p_2 , w , and T by use of equation (2) multiplied by these two correction factors.

APPARATUS AND PROCEDURE

The apparatus used in this work consisted of a boiler for producing hydrogen peroxide vapor, a separator and superheater for mist removal and temperature regulation, a capillary tube, and auxiliary equipment for the measurement of pressure, temperature, and rate of flow. A rather complicated boiler is required to deliver at atmospheric pressure concentrated hydrogen peroxide vapor of a desired concentration and at a steady flow rate, and at the same time to minimize decomposition and to avoid explosions. Such a boiler has been developed after considerable experimentation and is described in detail elsewhere (19,20). A similar boiler was used here, the only change being a slight modification to minimize the hydrogen peroxide residence time in the boiler and to provide for electrical heating. The usual flow rate of vapor from the boiler was equivalent to about 7 cc/min. of condensed liquid.

The vapor issuing from the boiler was passed through a simple,

impingement-type entrainment separator, then through a superheater consisting of a glass tube 30 cm long wound with electrical resistance wire, and finally through a cyclone-type entrainment separator. In operation, liquid was collected in the upstream separator, but none was observed in the downstream separator, indicating that the first separator and the superheater had served to eliminate all liquid from the stream. The temperature of the superheater was regulated with a variable transformer. Heat loss from the second separator was prevented by another resistance winding placed in the insulation surrounding it.

Capillary Tube Preliminary experiments had shown that simple insulation carrying an electrical winding could not be depended upon to maintain constant temperature in a capillary tube of the dimensions used. The principal reason for the failure of this arrangement lay in the necessarily slow mass flow rate at which the vapor passed through the capillary and its concomitant sensitivity to heat loss or gain. To meet this problem the arrangement shown in Fig. 1 was developed. In this apparatus a stream of relatively large flow rate passed through an annulus about the capillary in counter-current flow. The temperature of this larger stream could be maintained constant and this in turn insured that the portion of the stream which was subsequently allowed to flow through the capillary was also maintained at constant temperature. Thus the stream, after leaving the second

entrainment separator, passed a thermocouple well, travelled along the outside of the capillary tube and reached a bulb of 2500 cc capacity. This bulb carried a pressure tap which led to a manometer and a sample tap which allowed the withdrawal through a condenser of the greater portion of the stream. A thermocouple well was inserted in the bulb. The relatively large volume of the bulb provided an additional opportunity for settling and/or vaporization of mist, if any were present, and constituted a reservoir of vapor in a state of near stagnation.

A small proportion of the vapor stream, about 1/100th of that entering the bulb, then passed into the capillary tube. This tube was a Pyrex, "Tru-Dore" capillary of 0.5/mm inside diameter and 28 cm length. The details of the precise determination of its diameter and length are presented below. As shown in Fig. 1 the capillary was sealed at its downstream end to the outer tube which served as a vapor jacket. Particular care was taken that this sealing procedure did not distort the end of the tube. After leaving the end of the capillary tube, the stream passed into a sample bulb as shown in the insert in Fig. 1. No condenser was needed at this point since the small samples readily condensed in the uninsulated bulbs. The amount of peroxide decomposition occurring in the apparatus was determined by analysis, ^{with} standardized permanganate solution, of condensate samples at the two sampling points. The amount of oxygen present in the exit stream from the apparatus was determined by

measuring by water displacement the volume of non-condensable gas leaving the condenser.

Auxiliary Equipment The connection from the large bulb shown in Fig. 1 to the manometer was made through a condenser. At a low point in this line below this condenser a stopcock allowed the periodic withdrawal of condensate. Above this condenser a connection was made to a compressed air line. Air from this source was used to depress the manometer fluid initially, thus filling the line between condenser and manometer with non-condensable gas, which prevented hydrogen peroxide vapor from reaching the manometer. The manometer itself was made of 10 mm Pyrex tubing and the low pressure leg was open to the atmosphere. The manometer fluid used was Fluorolube FS, a fluorinated hydrocarbon product of the Hooker electrochemical Co. Density measurements of this fluid were made at several temperatures with a pycnometer, e.g., at 25°C the density was found to be 1.887g/cc. The height of the fluid in the manometer legs was measured separately with two 40 cm. cathetometers.

Temperature measurements at the various points indicated in Fig. 1 were originally made with mercury-in-glass thermometers. These proved inaccurate because of hydrogen peroxide decomposition occurring on the soft glass of which the thermometers were made. Pyrex glass thermocouple wells carrying calibrated (1) copper-constantan thermocouples were therefore substituted for the thermometers. The thermocouple voltages were measured with a

potentiometer, using a Rubicon galvanometer having a sensitivity of $+ 1 \times 10^{-6}$ volts and a full scale deflection for 2×10^{-4} volts.

The samples of condensed vapor were taken in tared Pyrex bulbs of the form shown in Fig. 1. Flow rates were established by weighing these bulbs on an analytical balance, the time to collect the samples having been determined with a stop watch. In the case of calibration runs with non-condensable gases, the flow rates were measured by collecting the gas over water in a flask of 1122.0cc capacity. During collection of the gas, the water was displaced into another flask, manipulated so as to keep the pressure in the collection flask constant.

Capillary Tube Dimensions The measurement of the radius of the capillary tube was carried out by determining the weight of and length occupied by a thread of mercury introduced into the capillary. Such a measurement was carried out with threads of about 1.4 and 6.6 cm. length and moved to different positions along the length of the capillary. A Zeiss Focometer, an instrument consisting of a telescope with cross-hairs mounted above a carriage moved by a micrometer screw, was used to measure the lengths of the threads. The capillary used was selected for the most constant diameter from a choice among several.

The precision of these measurements, about one part in 2000, was of the same magnitude as the maximum difference found between radii of the capillary at six positions of the 1.4 cm mercury thread along the length of the capillary. The arithmetic

average radius found was 0.02594 ± 0.0004 cm. This result was also obtained with the 6.6. cm thread. A more precise determination or the use of a more elegant method of determining the mean radius, such as given by Fisher (5) and Rapp (16), was not warranted. The accuracy of the figure obtained is sufficient to allow the evaluation by measurement on a gas of known viscosity of the magnitude of the energy loss due to causes other than pressure drop in the tube. Once it was thus determined that this energy loss was of such low magnitude under the conditions of experiment that the precision of the determination of the pressure drop in the tube was satisfactory, the knowledge of the tube radius was, strictly speaking, no longer necessary.

The ends of the capillary tube were ground flat and by measurement with a cathetometer the length of the capillary was found to be 28.53 ± 0.04 cm.

EXPERIMENTAL RESULTS

Calibration With Helium, Air, and Water Vapor

The magnitude of the correction factor, K_E , which allows for energy losses other than those due to pressure drop within the capillary tube, was determined for this apparatus by measuring the apparent viscosity, as calculated by equation (2), of helium, air, and water vapor. The correction factor is then

$$K_E = \frac{\text{established viscosity}}{\text{observed viscosity}}$$

Eleven runs using helium and eighteen runs using air were made at room temperature. Sixteen runs were made with water vapor at temperatures of 170, 200, and 240°C. The results of these calibration runs are listed in Table 1 and are shown in Fig. 2 where the value of the correction factor, k_E , is plotted as a function of the Reynolds number characteristic of the flow in the tube.

The established values for the viscosity of helium and air were calculated from the following equations recommended by Keyes (13) and based on the data of Johnston and McCloskey (10) and Johnston and Grilly (9):

$$\text{Air: } \mu = \frac{14.88(T)^{1/2} 10^{(5/T)}}{10^{(57/T)} + (122.1)/T} \quad (3)$$

$$\text{Helium: } \mu = \frac{18.05(T)^{1/2} 10^{(50/T)}}{10^{(50/T)} + 251/T} \quad (4)$$

For example, at 24°C and 1 atmosphere the viscosities of air and helium have been calculated by equations (3) and (4) to be 183.8 and 197.3 micropoises, respectively.

Surprisingly enough, a rather large uncertainty appears to exist regarding the viscosity of water vapor. The values for the viscosity of water vapor used in this work were calculated from the following equations recommended by Keyes (12) and based on the data of Hawkins, Solberg, and Potter (6):

$$\mu_0 = \frac{15.01(T)^{3/2}}{T + 446.8} \quad (5)$$

$$\begin{aligned} \mu_{p'} = \mu_0 + 100 p'/T (6.364 - 2.307 \times 10^{-3} \times 10^{(1340/T)}) \\ + 3.89 (p')^2 \times 10^{(-5.476 \times 10^{-3}T)} \end{aligned} \quad (6)$$

Two other evaluations of the literature dealing with the viscosity of water vapor have appeared recently: one by Jaumotte (8) agrees with Keyes, one by Richter (17) accepts a set of viscosity values for one atmosphere pressure which is lower than that recommended by Keyes. Silgado and Storrow (22) also concluded that the lower set of values, based on the work of a number of investigators, was to be preferred. There is no doubt that the experimental data of Hawkins, Solberg, and Potter (6) is to be preferred for establishing the viscosity of water vapor over a wide range of temperature and pressure. However, it is necessary to make an extrapolation of their data to the region of pressure and temperature which is of interest here, and it must be concluded that an uncertainty of not less than $\pm 1\%$ exists regarding the viscosity of water vapor at one atmosphere pressure near 200°C. Should improved values become available, however, the results reported here may be converted to the new basis by application of a simple ratio.

At a pressure of one atmosphere and at temperatures of 170, 200, and 240°C, then, the values 158.4, 168.9, and 182.6 micropoises, respectively, have been calculated to be the established values for the viscosity of water vapor and have been used in this work. Over this limited range of temperature

the viscosity-temperature relationship may be considered to be linear, and the viscosity changes by 0.35 micropoises per degree centigrade. The value of this slope may also be changed very slightly by more accurate viscosity determinations.

In calculating the correction factor for the steam runs through use of equation (2) a value for the compressibility factor of 0.995 was used at all temperatures. This value was obtained by calculation from the properties of steam given by Keenan and Keyes (11). There is probably an uncertainty regarding the accuracy of this value, and its introduction may not be entirely justified. However, it appears as a constant factor and was applied to the oxygen calibration runs and hydrogen peroxide runs as well.

The results of the calibration runs shown in Fig. 2 indicate that with the chosen dimensions of the capillary and at the rates of flow permitted, the error due to the occurrence of energy losses other than those caused by flow in the capillary has been reduced to a reasonably small value ranging between -1 and +3%. It was concluded that the runs made with air and helium served only to show that the general magnitude of the correction factor was unchanged by large variations in flow rate, temperature, and physical properties. The correction factor chosen to be applied to the hydrogen peroxide vapor runs was based only on the correction factors determined with water vapor. The results show that a difference exists between the correction factors

determined with steam at 170 and 200°C and at 240°C. The following values for the correction factor have accordingly been applied to the results of the hydrogen peroxide vapor runs: at 170 and 200°C, $K_E = 1.023$; at 240°C, $K_E = 0.993$. Additional evidence that the correction factor varies with temperature is found in the results of the hydrogen peroxide vapor measurements and the measurements in which oxygen was mixed with steam. In both of these cases a correction factor varying with temperature is required to maintain consistency of results.

Viscosity of Oxygen - Water Vapor Mixtures

It is nearly impossible to produce and manipulate hydrogen peroxide vapor at atmospheric boiling temperatures without encountering some decomposition. Only by meticulous attention to cleanliness of the apparatus can decomposition be kept to a minimum. In this work it was found possible to produce consistently a stream of hydrogen peroxide vapor containing less than 3.0 mole percent oxygen. A few runs in which decomposition produced oxygen concentrations greater than 3% were discarded.

Since the object of this work was limited to the determination of the viscosity of hydrogen peroxide - water mixtures, it was necessary to adopt some method for correcting the experimental results for the presence of small proportions of oxygen. Theoretical methods for the estimation of the viscosity of mixtures of gases cannot yet be depended upon. A sounder basis for determining

the effect of oxygen appeared to be the direct measurement of the effect of the presence of oxygen on the viscosity of water vapor, development of a concentration-dependent correction factor from these data, and cautious application of this correction factor to the data obtained for the hydrogen peroxide-water mixtures containing small proportions of oxygen.

In order to determine the effect of traces of oxygen on the viscosity of water vapor, steam runs were conducted in which pure oxygen was introduced at various rates, constant for each run, into the first upstream cyclone separator. Perfect mixing should have occurred because of the turbulent conditions in the separator. The oxygen rate was measured in the same way as in the hydrogen peroxide runs described in the procedure.

It was found that the viscosity of mixtures of oxygen and water vapor was higher than that of water vapor alone. In Fig. 3 is plotted the value of the correction factor,

$$K_O = \frac{\text{Viscosity of water vapor}}{\text{Viscosity of oxygen-water vapor mixture}}$$

as a function of the concentration of oxygen at temperatures of 170 and 240°C. The original data are given in Table 2.

The correction factor K_E was applied to the directly-calculated viscosities of the mixtures before the oxygen correction factor was computed as above. The results shown in Fig. 3 indicate that the presence of less than one mole % of oxygen is without

significant effect upon the measured viscosity. Accordingly, only the results of measurements of the viscosity of hydrogen peroxide vapor in which the oxygen content was less than one mole percent were retained. Some results of runs made with hydrogen peroxide vapor containing between one and three mole percent oxygen will be found in Table 3. By application of a suitable correction factor taken from Fig. 3 to these latter data they may be brought into accord with the results in which the oxygen content was less than one mole percent. However, these results were not directly used.

Corrected Poiseuille Equation

Numerical values for the constants and correction factors may now be introduced into equation (2). From the foregoing discussion the following values may be taken:

$$K_E = 1.023 \text{ at } 170 \text{ and } 200^\circ\text{C.}; 0.993 \text{ at } 240^\circ\text{C.}$$

$$K_O = 1, \text{ if the oxygen concentration is less than one mole \%}$$

$$r = 0.02594 \pm 0.00004 \text{ cm}$$

$$L = 28.53 \pm 0.04 \text{ cm}$$

Since the apparatus was designed so that the down stream pressure p_2 , was always equal to the atmospheric pressure, P , the substitution, $p_1 = P + \Delta p$, may be introduced, where Δp is the pressure difference read from the manometer.

Combining this with the above values, equation (2) becomes

$$\mu = 3.748 \times 10^{-11} K_E M (2P\Delta p + (\Delta p)^2) / wT_s \quad (7)$$

Measurements on Hydrogen Peroxide

The results of 62 runs with hydrogen peroxide vapor were judged to have been obtained under conditions allowing precise calculation of the viscosity. All runs were discarded in which temperature and pressure drop did not remain constant, in which the oxygen content was greater than 3%, and in which decomposition in the capillary tube caused the downstream concentration to be less than 9/10 of the upstream concentration. Experimental data and calculated quantities are given in Table 3.

These results are shown in Fig. 4 where the viscosity of the hydrogen peroxide-water-oxygen mixture at 170, 200 and 240°C uncorrected for the presence of less than one mole percent oxygen is plotted as a function of the mole fraction hydrogen peroxide based on the complete stream composition. These viscosities were calculated with equation (7) using the mole fraction average molecular weight calculated for the mid-point of the capillary tube. It was often not possible to attain the desired stream temperature exactly, but it was possible to maintain the temperature constant at a level within a few degrees of the desired temperature. In order to bring the results to a common temperature it was assumed that the variation

of viscosity with temperature amounted to 0.35 micropoises per degree, the value found for steam at this temperature level. This temperature coefficient was applied to all runs in which the temperature varied from the chosen levels of 170, 200, or 240°C.

From the evidence of Fig. 4 it is apparent that some small factor, not accounted for by the correction applied, influenced the measurements on hydrogen peroxide vapor to produce low results. The slope of the viscosity curves seems well established. There is some indication that the slope increases with temperature, but the precision of the results does not warrant notice of this apparent increase. The curves have been drawn parallel with slopes of 14 micropoises per unit mole fraction.

By extrapolation, the values for the viscosity of anhydrous hydrogen peroxide vapor at the temperatures investigated given by these curves are: 170°C, 144 micropoises; 200°, 154; 240°, 167. At the normal boiling point of hydrogen peroxide (150°C) the vapor viscosity is estimated to be 137 micropoises.

These results may be expressed by the following equation:

$$\mu = 134 + 0.35 (t - 100) - 14y \quad (8)$$

This equation is considered to be reliable within a precision of $\pm 2\%$ at temperatures between 100 and 300°C at pressures near one atmosphere.

DISCUSSION OF RESULTS

Precision of the Measurements With the exception of the oxygen rate, the precision of the recorded data such as pressure drop, temperature, and flow rate was at least one part in 500 or better. The precision of estimation of the oxygen flow rate was about one part in 200. If decomposition in the capillary tube could have been avoided entirely, these limits of precision would have applied in estimating the precision of the results. However decomposition of hydrogen peroxide in the capillary may introduce significant changes in the composition and temperature of the stream. With a change in composition the results must be taken to represent the viscosity of a stream of some average composition. The rejection of runs in which the downstream hydrogen peroxide concentration was less than 9/10 of the concentration entering the capillary placed an upper limit of 1% on the magnitude of the possible variation in average molecular weight of the stream. The change in composition is relatively much larger, as much as 5% in some runs. It seems unlikely, however, than an uncertainty of $\pm 2 \frac{1}{2}\%$ must therefore be placed on the concentration which the calculated viscosity is taken to represent. If the decomposition in the capillary is a linear function of the distance travelled through the capillary it is reasonable to assume that the viscosity calculated for the average molecular weight represents that of the average concentration.

The effect of the release of the heat of decomposition in the capillary upon the temperature measured is difficult to estimate. A temperature difference of one degree centigrade between the stream in the capillary and the stream in the surrounding annulus where temperatures were measured appears to be more than adequate to dissipate the heat of decomposition.

Estimates, then, of the precision which might reasonably have been hoped for are: steam calibration runs, $\pm 0.5\%$; hydrogen peroxide runs, $\pm 1\%$. Inspection of the data shows the actual precision to have been about 1% or better in both the steam calibration runs and the hydrogen peroxide runs. The absolute accuracy of the data is of course dependent upon the accuracy of the accepted viscosity of water vapor. As the earlier discussion indicated, a change of as much as 2% in this value may be warranted. The failure of the intercepts of the best curves through the hydrogen peroxide viscosity data to occur at the accepted value for water vapor is probably a reflection of the decomposition occurring in the hydrogen peroxide runs. This decomposition undoubtedly affects the flow to a small degree in a manner which Poiseuille equation cannot account for, or, in other words, introduces a correction factor which the water vapor calibration runs could not have represented.

Comparison of Results with Theoretical Values A number of methods have been proposed for predicting the viscosity of pure gases

and gas mixtures. Some of these have been moderately successful, especially with non-polar gases. It is of interest to test the reliability of such relationships for a system of highly polar constituents such as has been studied here.

Licht and Stechert (14) made an extensive study to determine the most suitable analytical form for expressing the relationship between viscosity and temperature for a pure component in the low pressure region. They developed an empirical equation as follows

$$\mu = 6.30 \left[\frac{M^3 P_c^4}{T_c} \right]^{1/6} \left[\frac{T_r^{3/2}}{T_r + 0.8} \right] \quad (9)$$

Using the critical values, $T_c = 730^\circ\text{K}$, $P_c = 214$ atm., derived from the work of Scatchard, Kavanagh, and Ticknor (21), the viscosity of hydrogen peroxide at 170°C is calculated by equation (9) to be 148 micropoises. This appears to be a very good check of the value of 144 micropoises obtained by extrapolation of the experimental results. The agreement must be in part fortuitous, however, since a viscosity of 125 micropoises is predicted for water vapor at this temperature, compared with the established value of 158 micropoises. Similar agreement between calculated and established values is obtained using the empirical method of Uyehara and Watson (23) when the Licht and Stechert relationship is used to estimate the critical viscosity of hydrogen peroxide. If the known viscosity of liquid hydrogen peroxide is used, according to the method of Uyehara and Watson, to calculate the viscosity of the vapor, the

disagreement is of the order of 100% - about the same as obtained for water.

The foregoing procedures are based more or less upon the theory of corresponding states. Methods for the calculation of viscosity based upon more sophisticated theory have been developed with reasonable success for non-polar molecules. Application of these procedures to the calculation of the viscosity of polar gases generally yields reasonable results even though the underlying assumptions of the theory do not apply to polar gases. In a method worked out by Hirshfelder, Bird, and Spotz (7) and discussed by Bromley and Wilke (3) it is necessary to know the value of the parameters ϵ , the maximum energy of attraction between the molecules, and r_0 , the collision diameter of the molecules. In the case of hydrogen peroxide it is necessary to estimate these quantities. Following the method of Bromley and Wilke the value of ϵ was estimated using the relationship, $\epsilon/k = 0.75/T_c$. A method of estimating r_0 involving knowledge of the critical volume was rejected for hydrogen peroxide, since the only values available for critical constants are estimates. This parameter was therefore estimated indirectly through the relationship of Licht and Stechert given above. Following this procedure, as given by Bromley and Wilke (3), the viscosity of hydrogen peroxide vapor at 170°C was estimated to be 150 micropoises, in good agreement with experiment. An attempt to carry out the reverse procedure, i.e., calculation of

the values of ϵ and r_0 from experimental data without recourse to the approximations used above does not appear to be warranted. Such a procedure requires the knowledge of the variation of viscosity with temperature. The small range of temperature investigated and the uncertainty involved in the extrapolation used to obtain the viscosity of anhydrous hydrogen peroxide do not permit the differentiation of the temperature coefficient of viscosity of hydrogen peroxide from that of water. It should also be pointed out that the uncertainty of the magnitude of one percent or so which exists regarding the accurate viscosity of the vapors of water and hydrogen peroxide would allow considerable latitude in the choice of the form of an equation for the energy of interaction.

The results obtained in this work may also be used to test various mixture rules which have been proposed. Within the precision of the data obtained here it is apparent that the viscosity of hydrogen peroxide-water mixtures follows a simple mole-fraction-additive rule:

$$\mu_m = x_a \mu_a + x_b \mu_b \quad (10)$$

If the values for the viscosity of anhydrous hydrogen peroxide obtained by linear extrapolation are accepted, then the mixture rule proposed by Wilke (24) predicts in turn a linear composition relationship exactly. Wilke's mixture rule is given by Bromley and Wilke (3) as follows:

$$\mu_m = \frac{\mu_a}{1 + (x_b/x_a)\phi_{ab}} + \frac{\mu_b}{1 + (x_a/x_b)\phi_{ba}} \quad (11)$$

where

$$\phi_{ab} = \frac{[1 + (\mu_a/\mu_b)^{1/2} (M_b/M_a)^{1/4}]^2}{4/\sqrt{2} [1 + (M_a/M_b)]^{1/2}}$$

$$\phi_{ba} = \frac{[1 + (\mu_b/\mu_a)^{1/2} (M_a/M_b)^{1/4}]^2}{4/\sqrt{2} [1 + (M_b/M_a)]^{1/2}}$$

Comparison With Other Systems

The prediction of the viscosity of a pure substance or of the composition-dependence of viscosity, on the simple basis of molecular weight and general structure, is uncertain even in a qualitative way. The present results seem to bear out the general conclusion that a) gas viscosity decreases as molecular structure becomes more complicated (14), e.g., compare the series $\mu_{O_2} > \mu_{SO_2} > \mu_{H_2O} > \mu_{H_2O_2} > \mu_{S_8}$ and b) that the variation of viscosity of mixtures from simple additivity is least when the mass, structure, and interaction potentials of molecules are most alike (18). With the exception of the twofold difference in molecular weight this latter relation would appear to apply quite closely for mixtures of hydrogen peroxide and water.

The viscosity behavior of a number of other gas mixtures is recorded by Hirshfelder, Bird, and Spotz (7), Wilke (24), and Buddenberg and Wilke (4). Unfortunately, only one of these mixtures contains water vapor as a constituent, that of air-water vapor over a small range of water vapor content. Among non-polar

mixtures, that of oxygen and nitrogen appears to be the closest analog of the hydrogen peroxide-water system. The nearest analog among polar systems appears to be that of alcohol and water, studied by Silgado and Storow (22). Here, however, there appears to exist sufficient difference in polarity, mass, and structure for the viscosity of the mixtures to vary significantly from additivity.

It would be valuable to have additional viscosity data for this system at higher hydrogen peroxide concentrations and also at higher temperatures. However, previous experience with generating hydrogen peroxide vapor indicates that even with meticulous attention to cleanliness and experimental manipulation, adventitious explosions may occur on handling, at approximately atmospheric pressure, hydrogen peroxide vapor of the higher concentrations studied here. Therefore a substantially different experimental technique would be needed to extend the data.

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NOTATION

k	= Boltzmann factor, 1.380×10^{-16} ergs/ $^{\circ}$ K
K_E	= correction factor for energy losses
K_O	= correction factor for presence of oxygen
L	= length of capillary, cm
M	= molecular weight, g/mole
p	= pressure, dynes/cm 2
p'	= pressure, kg/cm 2
Δp	= pressure difference, dynes/cm 2
P	= atmospheric pressure, dynes/cm 2
P_O	= critical pressure, atm
q	= volumetric flow rate, cc/sec
r	= radius of capillary, cm
r_O	= collision diameter of a molecule
R	= gas constant, 8.314×10^7 (dynes/cm 2) cc/g mole $^{\circ}$ K
t	= temperature, $^{\circ}$ C
T	= temperature, $^{\circ}$ K
T_C	= critical temperature, $^{\circ}$ K
T_r	= reduced temperature, T/T_O
w	= mass flow rate, g/sec
x	= mole fraction of any component
y	= mole fraction hydrogen peroxide
z	= compressibility factor
ϵ	= maximum energy of attraction between molecules, ergs
μ	= viscosity, micropoises
μ_O	= viscosity at "zero" pressure, micropoises

μ_m = viscosity of a mixture, micropoises

$\mu_{p'}$ = viscosity at pressure p' , micropoises

π = 3.1416

ϕ = a function defined in the text

subscript 1 refers to upstream condition

subscript 2 refers to downstream condition

subscript a refers to component a

subscript b refers to component b

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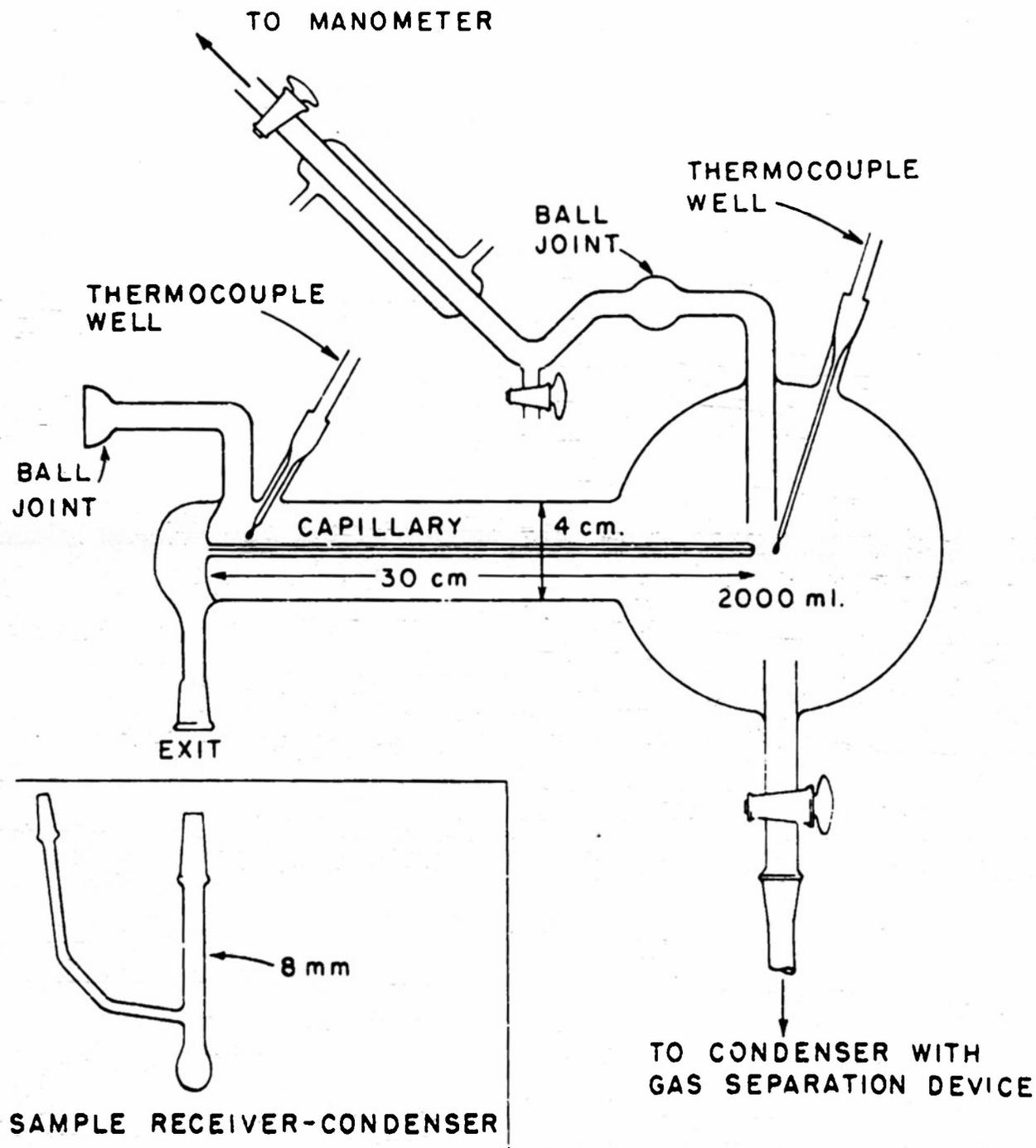


FIG. 1- CAPILLARY ASSEMBLY

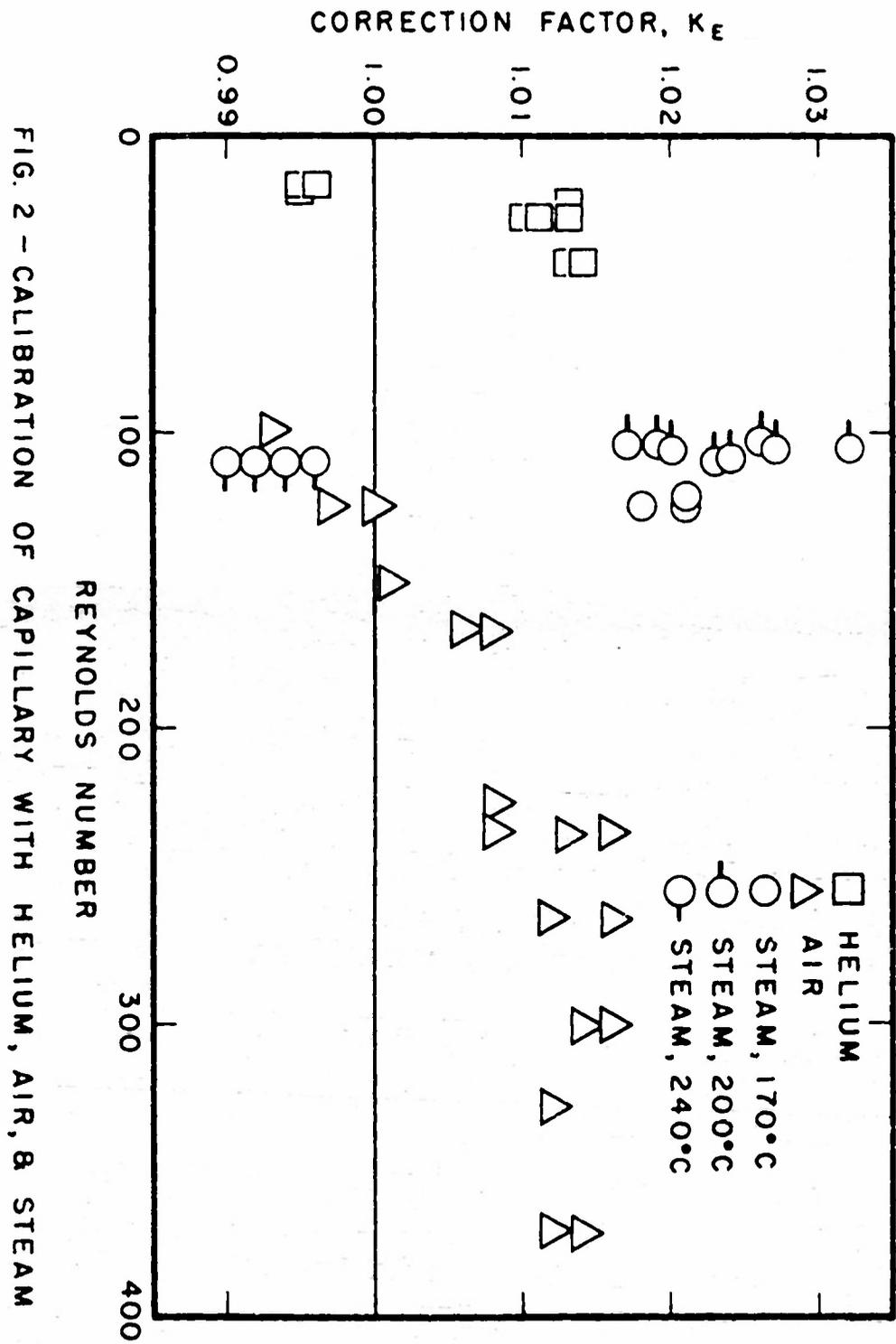


FIG. 2 - CALIBRATION OF CAPILLARY WITH HELIUM, AIR, & STEAM

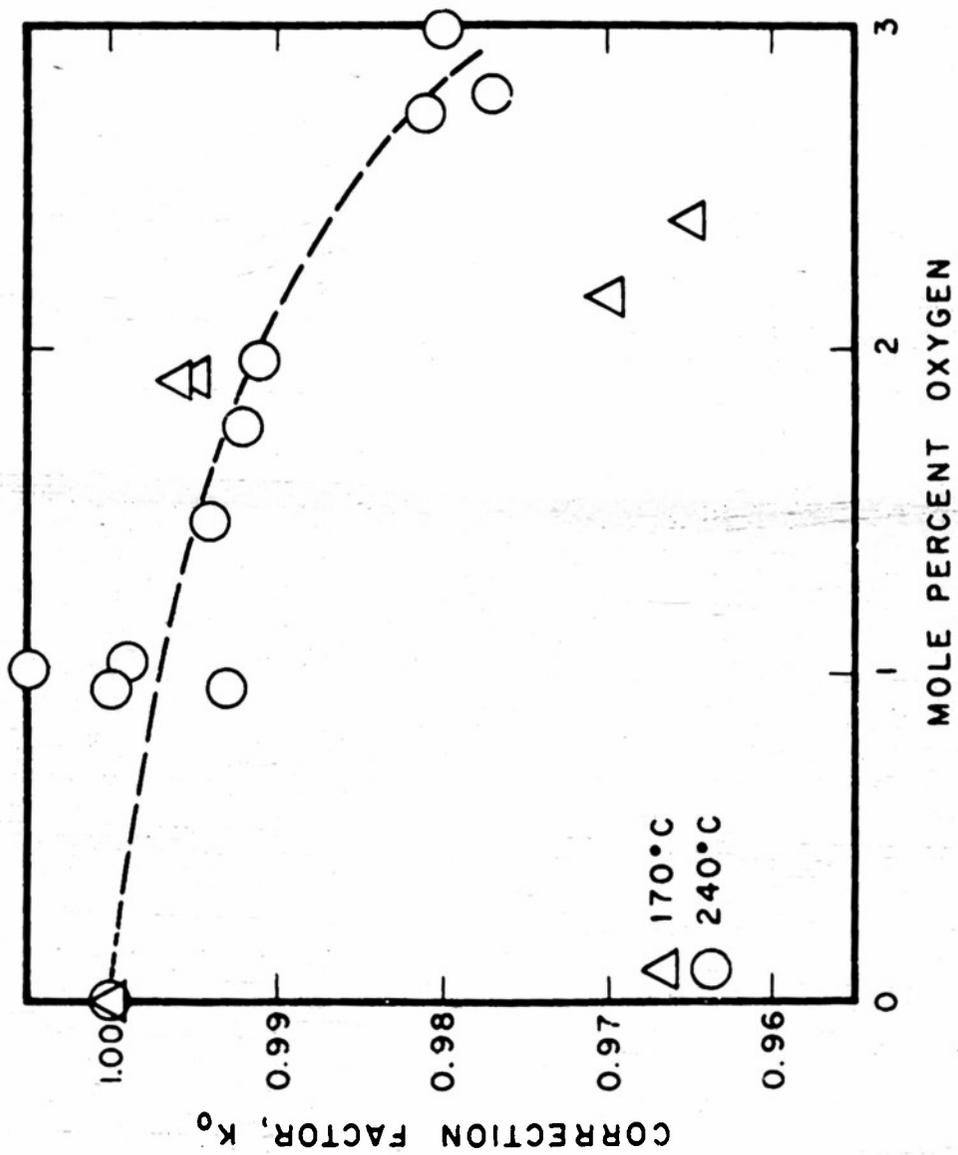


FIG. 3 - CALIBRATION OF CAPILLARY WITH STEAM CONTAINING OXYGEN

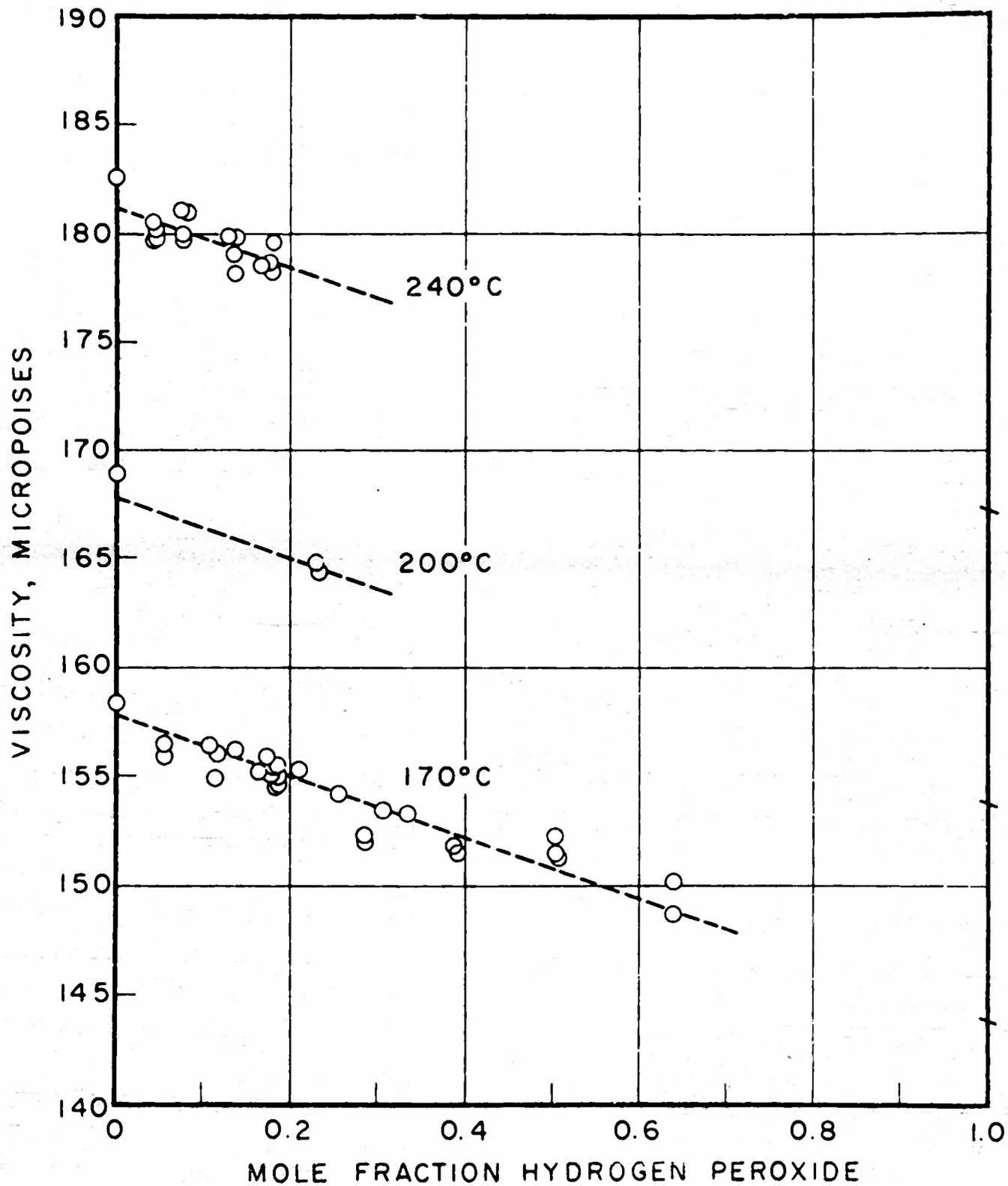


FIG. 4 - VISCOSITY OF HYDROGEN PEROXIDE VAPOR

A1

APPENDIX

Table 1

A2

Calibration of Capillary with Helium, Air, and Steam

Run No.	Duration of Run, Sec.	Mass Flow Rate, g/sec x 10 ³	Room Temp., °C	Tube Temp., °K	Barometric Pressure, dynes/cm ² x 10 ⁻⁶	Pressure Drop, cm Fluorolube	Reynolds Number	Viscosity Observed, Micropoises	Viscosity Established, Micropoises	Correction Factor
Helium:										
1	1326	0.138	24.0	297.2	1.005	14.74	18	198.2	197.3	0.995
2	838	0.226	"	"	1.043	22.15	28	194.8	"	1.013
3	983	0.193	"	"	"	18.95	24	194.7	"	1.013
4	548	0.346	"	"	"	33.54	43	194.8	"	1.013
5	553	0.343	"	"	"	33.23	43	194.7	"	1.013
6	530	0.341	28.0	301.2	1.009	35.10	43	196.3	198.8	1.013
7	529	0.342	"	"	1.010	35.04	43	195.9	"	1.014
8	792	0.229	"	"	"	23.73	28	196.6	"	1.011
9	791	0.229	"	"	"	23.77	28	196.8	"	1.010
10	1314	0.137	29.0	302.2	"	14.67	17	200.1	199.3	0.996
11	1329	0.137	27.0	300.2	"	14.45	17	199.4	198.5	0.995
Air:										
12	1217	1.130	24.0	297.2	1.010	14.53	151	183.6	183.8	1.001
13	1848	0.738	24.3	297.5	1.023	9.69	99	185.2	184.0	.993
14	--	0.740	24.3	297.5	1.020	--	99	185.2	"	.993
15	1453	0.937	24.0	297.2	1.022	12.22	125	184.0	184.0	1.000
16	1454	0.936	"	"	"	12.17	"	183.5	"	0.997
17	1080	1.246	"	"	"	16.27	167	182.9	"	1.006
18	1085	1.250	"	"	1.051	16.15	"	182.5	"	1.008
19	836	1.617	25.0	298.2	"	20.95	226	182.9	184.4	1.008
20	765	1.763	25.8	299.0	"	22.78	235	182.3	184.6	1.008
21	764	1.766	"	"	"	22.73	236	181.7	"	1.013
22	765	1.770	26.3	299.5	"	22.73	"	181.7	184.8	1.016
23	677	1.990	25.8	299	"	25.56	265	132.1	184.6	1.016
24	680	1.980	26.3	299.5	1.030	25.03	264	182.6	184.8	1.012
25	598	2.260	"	"	"	28.59	301	182.2	"	1.014
26	598	2.260	"	"	"	28.93	"	181.8	"	1.016
27	483	2.780	26.8	300	"	35.87	370	182.7	165.0	1.012
28	482	2.800	"	"	"	35.80	372	182.4	"	1.014
29	549	2.460	"	"	"	31.63	328	182.8	"	1.012
Water Vapor:										
30	600	0.7917	24	443.0	1.024	20.63	125	155.1*	158.4	1.021
31	600	0.7880	24	443.0	1.026	20.56	125	155.6	"	1.016
32	"	0.7703	24	442.7	1.026	20.01	122	155.0	"	1.021
33	"	0.7017	23	473.3	1.020	20.93	104	165.7	168.9	1.019
34	"	0.7207	25	473.2	1.001	21.90	106	165.6	"	1.020
35	"	0.7057	24	473.5	1.022	21.09	104	166.0	"	1.017
36	"	0.7422	26	473.2	1.016	22.20	110	165.1	"	1.023
37	"	0.7370	26	473.3	"	22.04	109	165.0	"	1.024
38	"	0.7042	24	473.3	1.022	21.01	104	165.8	"	1.019
39	"	0.7197	23	473.2	1.005	21.62	106	164.5	"	1.027
40	"	0.7195	24	473.0	1.002	21.56	106	163.6	"	1.032
41	"	0.6935	23	473.2	1.013	20.69	103	164.6	"	1.026
42	"	0.8100	29	512.8	"	29.35	110	184.4	182.6	0.990
43	"	0.8090	28	513.6	"	29.26	110	184.0	"	0.992
44	"	0.8116	28	512.5	"	29.18	110	183.6	"	0.994
45	"	0.8127	29	514.8	"	29.34	110	183.4	"	0.996

* Corrected for variation of experimental conditions from temperatures of 443, 473, or 513°K.

TABLE 3
EXPERIMENTAL RESULTS: VISCOSITY OF TAION SOLUTIONS OF
N-ALDEY PEROXIDE AND WATER

Run No.	Room Temp., °C	Barometric Pressure, dynes/cm ² × 10 ⁻⁶	Pressure Drop, on Fluorolube	Total Mass Flow Rate, g/min × 10 ⁻³	Tube Temp., °C	Downstream Oxygen Rate, ml/min	Hydrogen Peroxide Concentration, wt. % Upstream	Hydrogen Peroxide Concentration, wt. % Downstream	Stream Molecular Weight	Temperature Difference, °C	Oxygen Content of Stream, %	Hydrogen Peroxide Concentration at Inlet, Mole Fraction	Viscosity, Micropoise
1	20	1.004	22.74	0.9813	174.6	0.813	28.78	27.62	20.87	-0.6	1.03	0.172	154.9
2	21	1.004	22.20	1.004	167.4	0.813	29.09	28.09	21.01	+0.5	0.78	0.181	145.4
3	21	1.003	22.19	0.9755	170.0	0.70	30.74	29.07	21.04	0.0	0.79	0.186	151.6
4	21	1.003	22.14	0.9757	172.2	0.60	30.65	30.09	20.98	-2.2	0.75	0.182	156.5
5	21	1.003	22.21	0.9855	172.2	0.73	30.31	28.69	20.98	0	0.75	0.182	156.5
6	21	1.003	22.21	0.9873	170.0	0.95	30.56	27.87	20.98	0	0.57	0.180	155.4
7	21	1.003	22.08	1.003	168.8	0.70	29.22	28.09	20.89	+1.2	0.82	0.176	155.4
8	21	1.004	22.08	0.975	167.9	0.75	29.29	27.87	21.06	+1.3	0.81	0.210	155.4
9	23	1.025	22.01	0.9682	168.0	0.90	27.61	22.82	20.87	+2.0	0.82	0.193	156.5
10	23	1.025	22.01	0.9693	168.0	0.90	27.61	22.82	20.87	+2.0	0.82	0.193	156.5
11	26	1.015	21.97	0.9765	168.7	0.61	30.68	29.23	21.02	+1.3	0.61	0.185	155.0
12	27	1.012	26.50	1.115	172.2	0.50	19.97	18.89	19.82	-2.2	0.61	0.115	156.0
13	27	1.012	26.55	1.118	173.0	0.55	19.66	18.89	19.89	-3.0	0.61	0.115	156.0
14	27	1.012	26.55	1.118	173.0	0.55	19.66	18.89	19.89	-3.0	0.61	0.115	156.0
15	26	1.014	27.00	1.090	173.8	0.71	19.80	17.55	19.81	-3.8	0.61	0.107	156.4
16	26	1.014	27.00	1.113	170.0	0.97	10.14	10.08	18.90	0	—	0.055	156.5
17	26	1.014	27.62	1.117	170.0	0.10	10.22	9.97	18.90	0	—	0.055	155.9
18	26	1.007	27.66	1.323	169.8	2.10	37.74	37.74	22.21	+0.2	1.46	0.255	155.2
19	26	1.008	29.09	1.300	170.8	0.95	40.68	40.68	22.14	-0.8	0.51	0.255	156.2
20	26	1.009	27.21	1.374	174.4	3.00	51.72	46.83	23.58	-3.4	2.86	0.335	153.1
21	26	1.013	27.09	1.349	174.0	3.30	51.89	46.83	23.65	-1.8	2.77	0.338	154.5
22	26	1.013	27.62	1.440	169.6	1.70	44.39	41.03	22.66	+0.4	0.78	0.283	152.0
23	26	1.013	28.04	1.436	171.8	1.10	44.10	42.07	22.66	+0.4	0.78	0.283	152.0
24	26	1.013	28.04	1.436	171.8	1.10	44.10	42.07	22.66	+0.4	0.78	0.283	152.0
25	26	1.013	28.82	1.441	174.6	1.20	50.04	47.15	23.40	-0.4	0.87	0.323	153.3
26	26	1.009	27.87	1.517	173.8	3.00	66.06	59.62	25.73	-3.8	2.77	0.372	152.1
27	26	1.009	27.85	1.520	174.5	3.40	66.36	60.82	25.88	-4.5	2.50	0.400	152.0
28	26	1.009	27.85	1.520	174.5	3.40	66.36	60.82	25.88	-4.5	2.50	0.400	152.0
29	26	1.021	28.30	1.548	170.8	1.70	60.28	60.28	25.79	-4.4	2.63	0.475	151.7
30	26	1.021	28.00	1.538	170.0	1.10	53.41	53.41	24.79	0	1.05	0.349	151.5
31	26	1.023	28.72	1.633	174.6	1.50	66.13	64.22	26.10	-0.6	0.83	0.503	151.5
32	26	1.023	28.72	1.633	174.6	1.50	66.13	64.22	26.10	-0.6	0.83	0.503	151.5
33	26	1.023	28.72	1.633	174.6	1.50	66.13	64.22	26.10	-0.6	0.83	0.503	151.5
34	26	1.023	28.72	1.633	174.6	1.50	66.13	64.22	26.10	-0.6	0.83	0.503	151.5
35	26	1.023	28.72	1.633	174.6	1.50	66.13	64.22	26.10	-0.6	0.83	0.503	151.5
36	26	1.016	25.06	0.8406	232.5	0.83	28.58	26.24	20.76	+7.5	1.01	0.167	178.1
37	26	1.017	24.89	0.8180	237.7	1.15	29.66	27.24	20.98	+2.3	1.40	0.176	180.6
38	27	1.017	24.89	0.8176	239.3	1.30	29.66	27.19	20.97	+0.7	1.45	0.176	180.1
39	27	1.017	24.88	0.8067	241.0	1.40	29.82	26.85	20.97	-1.0	1.77	0.170	180.0
40	27	1.017	24.79	0.8078	240.9	1.45	29.69	26.75	21.01	-0.9	2.16	0.173	180.9
41	27	1.021	24.70	0.8117	246.8	1.50	28.81	26.24	20.91	+3.2	2.25	0.168	180.2
42	27	1.021	24.68	0.7870	246.0	0.10	8.21	7.73	20.91	+6.0	—	0.043	180.2
43	27	1.021	24.68	0.7903	232.5	0.20	8.31	7.89	18.72	+7.4	0.70	0.043	180.2
44	27	1.021	24.68	0.7992	234.4	0.30	9.12	8.33	18.62	+5.6	0.35	0.047	179.8
45	27	1.023	26.62	0.8068	239.5	0.35	14.69	13.28	19.30	+0.5	0.25	0.077	179.7
46	27	1.023	26.24	0.8116	239.9	0.35	14.69	13.28	19.30	+0.5	0.25	0.077	179.7
47	27	1.023	26.24	0.8060	239.0	0.36	13.95	13.35	19.29	+1.0	0.48	0.075	181.1
48	27	1.023	26.24	0.8060	239.0	0.36	13.95	13.35	19.29	+1.0	0.48	0.075	181.1
49	27	1.023	26.24	0.8060	239.0	0.36	13.95	13.35	19.29	+1.0	0.48	0.075	181.1
50	27	1.023	26.24	0.8060	239.0	0.36	13.95	13.35	19.29	+1.0	0.48	0.075	181.1
51	27	1.023	26.61	0.8117	241.9	0.35	14.19	13.76	19.33	-0.4	0.66	0.077	180.0
52	27	1.023	26.61	0.8117	241.9	0.35	14.19	13.76	19.33	-0.4	0.66	0.077	180.0
53	27	1.023	26.61	0.8117	241.9	0.35	14.19	13.76	19.33	-0.4	0.66	0.077	180.0
54	27	1.023	26.61	0.8117	241.9	0.35	14.19	13.76	19.33	-0.4	0.66	0.077	180.0
55	27	1.023	26.61	0.8117	241.9	0.35	14.19	13.76	19.33	-0.4	0.66	0.077	180.0
56	27	1.023	26.61	0.8117	241.9	0.35	14.19	13.76	19.33	-0.4	0.66	0.077	180.0
57	26	1.021	26.67	0.8134	241.4	0.35	23.91	22.87	20.25	-3.6	0.28	0.138	179.9
58	26	1.021	26.97	0.8134	241.4	0.35	23.91	22.87	20.25	-3.6	0.28	0.138	179.9
59	26	1.021	26.73	0.8134	242.2	0.70	23.91	21.97	20.12	-2.2	0.88	0.134	179.1
60	26	1.019	26.73	0.8134	241.4	0.65	29.40	27.71	20.86	-1.4	0.68	0.175	179.2
61	26	1.019	26.68	0.8059	239.2	0.55	29.57	27.71	20.97	-0.8	0.68	0.181	179.6
62	26	1.019	26.67	0.8059	239.2	0.55	29.57	27.71	20.97	-0.8	0.68	0.181	179.6
63	26	1.021	26.70	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
64	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
65	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
66	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
67	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
68	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
69	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
70	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
71	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
72	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
73	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
74	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
75	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
76	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
77	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
78	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
79	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
80	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
81	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
82	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
83	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
84	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
85	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
86	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
87	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
88	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0
89	26	1.021	26.77	0.8173	240.5	0.57	30.01	28.19	20.93	-0.8	0.62	0.179	179.0

Density of Fluorolube FS

Values for the density of Fluorolube FS, used in this work as manometer fluid, were obtained from the manufacturer, the Hooker Electrochemical Co., and checked and extended by measurements in a pycnometer. In the following table are given values taken from the temperature - density curve for Fluorolube FS used in this work.

Temperature, °C	Density, g/cc
20	1.895 ₅
22	1.892 ₅
24	1.889 ₅
26	1.885 ₅
28	1.880 ₀
30	1.872 ₀

Conversion of Viscosity Units

1 poise = 100 centipoises = 10^6 micropoises =
 1 g/cm sec = 1 dyne sec/cm² = 0.0102
 kg sec/m² = 0.1 kg/sec m = 0.0672 lb/sec ft = 0.00209 lbs.
 sec/ft²

Sample Calculation Take as an example hydrogen peroxide run 33.

Experimental data:

Ambient temperature, 26°C
 Barometric Pressure, 1.023×10^6 dynes/cm²
 Duration of run, 300 sec
 Average pressure drop in capillary, 28.76 cm fluorolube
 (deviation from average run time $\Delta p = 0\%$)
 Weight of condensate collected, 0.4826g
 Oxygen collected in 8 min., 12.5 ml
 Temperature of vapor stream, 174.8°C
 Upstream hydrogen peroxide concentration, 67.38 wt. %
 (deviation from run time average = + 0.2%)
 Downstream hydrogen peroxide concentration, 65.08 wt. %

By taking a material balance it can be shown that the weight of oxygen formed through decomposition in the capillary only is

$$X = \frac{W(U-D)}{(3400/16)-U}$$

where

X = weight flow rate of oxygen

