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# Measurement of Viscosity of Gaseous Mixtures at Atmospheric Pressure

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# Measurement of Viscosity of Gaseous Mixtures at Atmospheric Pressure

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# Summary

Coefficients of viscosity of various types of gas mixtures, including simulated natural-gas samples, have been measured at atmospheric pressure and room temperature using a modified capillary tube method. Pressure differences across the "straight" capillary tube section of a thermal mass flowmeter were measured for small, well-defined, volume flow rates for the test gases and for standard air. In this configuration, the flowmeter provides the volumetric flow rates as well as a well-characterized capillary section for differential pressure measurements across it. The coefficients of viscosity of the test gases were calculated using the reported value of 185.6  $\mu$ P for the viscosity of dry air at 300 K. The coefficients of viscosity for the test mixtures were also calculated using Wilke's approximation of the Chapman-Enskog (C-E) theory. The experimental and calculated values for binary mixtures are in agreement within the reported accuracy of Wilke's approximation of the C-E theory. However, the agreement for multicomponent mixtures is less satisfactory, presumably because of the limitations of Wilke's approximation of the classical dilute-gas state model.

# Introduction

Measurement of viscosity of pure gases and gas mixtures at atmospheric pressure is of great interest in fluid flow and heat transfer studies. Such information is not always readily available for the gas mixtures, particularly for hydrocarbon mixtures. In recent studies (ref. 1) of heats of combustion of natural gas and mixtures of gaseous hydrocarbons, reliable information about their viscosities was needed. In the absence of reliable and accurate experimental data and because of inadequate theoretical understanding of the coefficients of viscosity for these complex mixtures, it was decided to develop experimental procedures for measuring their coefficients of viscosity. It was desired that the technique should be simple and accurate for direct measurement of coefficients of viscosity for all types of gases. It should require only small quantities of the test gas and should be easily adaptable for relative measurements.

There are several techniques for measuring gaseous viscosity. The prominent ones are summarized in the sections which follow.

#### **General Capillary Method**

In the case of a compressible fluid flowing steadily through a perfectly cylindrical capillary tube, the volumetric flow rate at the inlet, is given by the following expression (refs. 2 to 4):

$$V = \frac{\pi r_c^4 (p_i - p_o)}{8\eta \ell_c} \tag{1}$$

where

 $\ell_c$  length of tube

 $\eta$  coefficient of viscosity of fluid

 $p_i$  pressure at inlet

 $p_o$  pressure at outlet

This is Poiseuille's formula for the flow of a gas through a tube. (See ref. 5.) Since all the quantities in the expression are easily measurable, it provides a convenient method for measuring  $\eta$  experimentally. Equation (1) can be simplified as follows:

$$V = A \frac{\Delta p}{\eta} \tag{2}$$

where

$$4 = \pi r_c^4 / 8\ell_c$$

A more accurate form of this equation would be (refs. 6 and 7):

$$V = \frac{A\Delta p}{\eta(1+\delta)} (1 + \frac{4\xi}{b}) - B \tag{3}$$

where

δ

capillary bore nonuniformity correction factor

 $\frac{4\xi}{b}$  wall-slip correction factor, X

For very low flow rates (corresponding to Reynolds numbers from 100 to 2000) sustained turbulence cannot be expected. Under these conditions, B is almost zero, and the slip correction is negligible for all gases. Hence, equation (2) provides an excellent basis for direct determination of viscosity as follows:

$$\left(\frac{V(\text{Air})}{V(\text{Test})}\right) = \left(\frac{\Delta p(\text{Air})}{\Delta p(\text{Test})}\right) \left(\frac{\eta(\text{Test})}{\eta(\text{Air})}\right)$$
(4)

$$\eta(\text{Test}) = \eta(\text{Air}) \left(\frac{V(\text{Air})}{V(\text{Test})}\right) \left(\frac{\Delta p(\text{Test})}{\Delta p(\text{Air})}\right)$$
(5)

By measuring the differential pressure across the same capillary tube for low-volume flow rates  $(< 50 \text{ cm}^3/\text{min})$  for air and the test gas, the viscosity of the test gas can easily be determined. The

accuracy of the viscosity value is limited only by the accuracies with which V(Air), V(Test), and  $\Delta p$  can be measured.

### **Oscillating-Disc Method**

This method involves measurement of the period and amplitude of the damped oscillations of a suitable solid body. The solid body is suspended from an elastic wire first in the test fluid and then in vacuum. A simplified expression for the fluid viscosity may be written as follows (refs. 8 to 10):

$$\eta = C\left(\frac{\lambda}{T} - \frac{\lambda_o}{T_o}\right) \tag{6}$$

where

- $\lambda, \lambda_o =$  damping coefficients in test fluid and vacuum, respectively
- $T, T_o =$  period of oscillation in test fluid and vacuum, respectively
  - $C = \frac{4I_d}{\pi r_d^4} \left( \frac{d_1 d_2}{d_i + d_2} \right)$ , an experimental constant

 $I_d$  = moment of inertia of oscillating disc

 $r_d$  = radius of oscillating disc

 $d_1, d_2 =$ distances between oscillating and fixed discs

However, the velocity field around the oscillating body is quite complex. This has been the major limitation of this method until recently.

### **Rotating-Cylinder Method**

In its most widely used variant, the angular deflection  $\theta$  of the inner cylinder is noted when the outer cylinder is rotated with a constant angular velocity  $\omega$ . If the end effects can be ignored, the coefficient of the fluid viscosity is given by (see refs. 11 and 12):

$$\eta = \frac{\pi \theta I_c (r_o^2 - r_i^2)}{r_o^2 r_i^2 T^2 \omega \ell_i} \tag{7}$$

where

 $I_c$  moment of inertia of inner cylinder

- $\ell_i$  length of inner cylinder
- T natural period of vibration
- $r_i, r_o$  radii of inner and outer cylinders, respectively

The major limitations of this method are the complexity of calculation of end-effect corrections and the requirement that no eddy motion develop in the fluid flow between the two cylinders.

## The Falling-Body Method

The basis of the method lies in Stokes' law (ref. 13), which states that the viscous drag W for a sphere falling with a uniform velocity in a fluid is given by

$$W = 6\pi\eta r_s V_T \tag{8}$$

where

 $r_s$  radius of falling sphere

 $V_T$  terminal uniform velocity

By measuring the terminal velocity of a sphere of known dimensions, the viscosity of the test medium can be obtained from the following expression:

$$\eta = \frac{2}{9} \left( \frac{\rho_s - \rho_f}{V_T} \right) g r_s^2 \tag{9}$$

where

 $\rho_s$  density of falling sphere

 $\rho_f \qquad \text{density of test fluid}$ 

g acceleration due to gravity

The major limitations of this method include the problems of frictional charge and the establishment of a truly uniform fall through the test fluid.

From these various techniques, it was decided to use a variant of the capillary flow method for measuring the viscosities of the test gases. This method is particularly suited for complex mixtures, which can often be costly and are available only in small quantities.

As indicated previously, the capillary tube viscometer is based on the application of Poiseuill's law for the steady flow of Newtonian fluids through a capillary tube. Clearly, all the assumptions made in the derivation of the laminar-flow solution cannot be fulfilled in its practical design and operation. However, a detailed study of all the sources of errors indicates that a practical capillary tube viscometer, capable of an accuracy of the order of 1 percent, can be developed if the following conditions are met:

1. A laminar steady-state flow condition is achieved before the differential pressure and volumetric flow rates are measured.

2. The fluid flow rates are kept low (i.e., Reynolds number is kept below  $2 \times 10^3$ ).

3. The effects of compressibility of actual fluids flowing through the capillary tube are minimized.

(This is possible if the flow rates are kept below  $50 \text{ cm}^3/\text{min}$  and if  $\frac{p_o}{p_i} \approx 1.$ )

The experimental procedures and the results for a number of binary and multicomponent mixtures are discussed in the following sections.

# Symbols

В	flow nonlaminarity constant
d	distance between oscillating disc and fixed disc
$f_i$	mole fraction of $i$ th specie
Ic	moment of inertia of inner cylinder
$I_d$	moment of inertia of oscillating disc
$\ell_c$	length of capillary tube
$\ell_i$	length of inner cylinder
$M_i$	molecular weight of $i$ th specie
n	number of components in multicomponent mixture
p	pressure
$p_i$	pressure at inlet of capillary tube
$p_o$	pressure at outlet of capillary tube
r <sub>c</sub>	root-mean-square radius of capillary tube
$r_d$	radius of oscillating disc
$r_i$	radius of inner cylinder
$r_o$	radius of outer cylinder
r <sub>s</sub>	radius of falling sphere

- T period of oscillation in test fluid
- $T_o$  period of oscillation in vacuum
- V volumetric flow rate
- $V_T$  terminal uniform velocity
- W viscous drag
- X wall-slip correction factor
- $\Delta p$  differential pressure  $(p_i p_o)$
- $\Delta p_C$  pressure drop across series capillary tube
- $\Delta p_F$  pressure drop across capillary section of flowmeter
- $\delta$  capillary bore nonuniformity correction factor

coefficient of viscosity η coefficient of viscosity of *i*th  $\eta_i$ component θ angular deflection of inner cylinder λ damping coefficient in test fluid  $\lambda_o$ damping coefficient in vacuum  $\rho_f$ density of test fluid density of falling sphere  $\rho_s$ Chapman-Enskog parameter for  $\phi_{ij}$ species i and jangular velocity ω

# Theory

The theory of viscosity of gases has been discussed extensively by a number of authors. (See refs. 14 to 17.) In this report, the computational procedure used for calculating the viscosity of multicomponent gas mixtures is briefly reviewed.

According to the Chapman-Enskog (C-E) kinetic theory of gases (ref. 15), the viscosity of a lowpressure multicomponent mixture can be approximated by the following expression:

$$\eta(Mixture) = \sum_{i=1}^{n} \frac{\eta_i f_i}{\sum_{j=1}^{n} f_j \phi_{ij}}$$
(10)

where

*n* number of components

 $f_i$  mole fraction of *i*th specie

There are a number of methods for estimating the parameter  $\phi_{ij}$ . We have used Wilke's approximation (refs. 18 to 20) for  $\phi_{ij}$  as follows:

$$\phi_{ij} = \frac{\left[1 + \left(\frac{\eta_i}{\eta_j}\right)^{1/2} \left(\frac{M_j}{M_i}\right)^{1/4}\right]^2}{\left[8\left(1 + \frac{M_i}{M_j}\right)\right]^{1/2}}$$
(11)

$$\phi_{ji} = \left(\frac{\eta_j}{\eta_i} \frac{M_i}{M_j}\right) \phi_{ij} \tag{12}$$

where

 $f_i$  mole fraction of *i*th component

 $M_i$  molecular weight of *i*th component

For nonpolar mixtures, Wilke's approximation is preferable over Brokaw's treatment (ref. 21) because

of its simplicity. It has been reported to be accurate to within 1 percent for nonpolar binary mixtures. For example, for a binary mixture,<sup>1</sup> one obtains the following:

$$\eta(\text{Mixture}) = \frac{\eta_1 f_1}{f_1 + f_2 \phi_{12}} + \frac{\eta_2 f_2}{f_2 + f_1 \phi_{21}}$$
(13)

It is apparent that the viscosity of the mixture is not equal to the weighted mean of the values of the viscosity of its components. Equation (10) has been used for calculating  $\eta$ (Mixture) for various types of mixtures. The viscosities of pure gases have been taken from references 6 and 23. The calculational program is described in the next section.

#### **Computational Procedure**

The computer program WILKE is written in FORTRAN Version 5 language for the Control Data CYBER 170 series digital computer system with Network Operating System (NOS) 2.3. The program requires approximately 26000 octal locations of core storage. A typical run to analyze 30 cases, each with 5 component gases, requires approximately 1 CPU second on the CYBER 173.

Program execution is performed by case, and the user may provide data for as many cases as desired. For each case, data are input to the program in NAMELIST format using the name VAR. Four variables must be defined for each case. The integer case number (NCASE) is used only for bookkeeping. The integer number of gases to be considered (NGAS) may range from 2 to 12. An integer array (IPOINT) is used for selecting which gases are to be included. A value must be provided for each of the NGAS gases. Gases are selected by specifying the appropriate indices from table A. CONC is a real array defining the concentration of each gas. The sum of all concentration fractions should equal 1, and the position within the array for each concentration should also follow the gas/index values provided above. As an example, a run to analyze two cases is presented below. The first case consists of a mixture of 30% CH<sub>4</sub>, 50% N<sub>2</sub>, and 20% CO<sub>2</sub>. The second case consists of a mixture of 35% CH<sub>4</sub>, 40% C<sub>2</sub>H<sub>6</sub>, and 25% C<sub>4</sub>H<sub>10</sub>.

Gas	Index
$CH_4$	1
$C_2H_6$	<b>2</b>
$C_3H_8$	3
$N_2$	4
$C_4H_{10}$	5
$\rm CO_2$	6
$O_2$	7
$\rm NH_3$	8
Ne	9
Ar	10
$H_2$	11
He	12

Table A

\$VAR NCASE = 1, NGAS = 3, IPOINT = 1, 4, 6, CONC = 0.30, 0.00, 0.00, 0.50, 0.00, 0.20\$
\$VAR NCASE = 2, NGAS = 3, IPOINT = 1, 2, 5, CONC = 0.35, 0.40, 0.00, 0.00, 0.25\$

Program output for each case echoes the case number and concentrations of selected component gases and gives the viscosity of the mixture. A listing of the program appears on the following page.

# **Experimental Procedure and Results**

It was the purpose of this study to develop a technique for measuring the viscosity of the unknown test gas without knowing its physical characteristics. This required the use of a dry test meter in series with an appropriate mass flowmeter for determining the calibration constant of the flowmeter for the test gas. Using this calibration constant, the volumetric flow rate of the test gas through any other mass flowmeter based on the same principle can easily be determined. As indicated previously, a variant of the capillary tube method has been selected for measuring the viscosity of various types of gaseous mixtures. This method is simple, sensitive, and adaptable to absolute or relative viscosity measurements for low-pressure gases. Figure 1 is a schematic of the experimental system. The capillary section of the flowmeter is made up of two narrow (0.030-cm and 0.045-cm i.d.) capillary tubes in parallel. The flow through each element of the capillary section remains laminar under low flow rates. Figure 2 shows the capillary section of the flowmeter selected for this study. The pressure difference accross the capillary section of the selected flowmeter is measured for air and then

<sup>&</sup>lt;sup>1</sup> For multicomponent mixtures, Buddenberg and Wilke (ref. 22) recognized the necessity of introducing the diffusion coefficients into the formula for  $\eta$ (Mixture) to take into account the collisions between unlike pairs. Even though this modified expression has been reported to predict experimental results quite well, it is not always usable because of lack of data about diffusion coefficients.

```
PROGRAM WILKE(INPUT, OUTPUT, TAPE5=INPUT, TAPE6=OUTPUT)
    DIMENSION MASS(15), CONC(15), ETA(15)
    DIMENSION DETA(15)
    REAL MASS
    REAL K.MU
    DIMENSION SYMB(15)
    DIMENSION IPOINT(15)
    CHARACTER*10 SYMB
    NAMELIST/VAR/IPDINT, NGAS, CONC, NCASE
    DATA MASS/16.,30.,44.,28.,58.,44.,32.,17.,20.,40.,2.,4.,3*0./
    DATA ETA/109.8E-6,91.0E-6,80.0E-6,175.69E-6,74.8E-6,
   1 146.63E-6,203.31E-6,97.4E-6,313.81E-6,
   2 222.86E-6,88.73E-6,196.14E-6,3*0./
    DATA DETA/0.330,0.277,0.220,0.454,0.237,
   1 0.450,0.616,0.425,0.697,0.704,0.200,0.464,
   2 3*0./
    DATA SYMB/*CH4*,*C2H6*,*C3H8*,*N2*,*C4H10*,*C02*,*02*,
   1 'NH3', 'NE', 'AR', 'H2', 'HE(4)', 3*' '/
       CHANGE THIS CARD FOR A DIFFERENT TEMPERATURE
    DATA T/300./
  1 FORMAT(1H1)
  2 FORMAT(////14X, *CASE NUMBER *, I2///)
  3 FORMAT(///9X, COMPONENT', 6X, FRACTION'//)
  4 FDRMAT(9X, A10, 6X, F7.4/)
  5 FORMAT(///3X, VISCOSITY OF MIXTURE = *, 1PE11.3)
    DO 8 IJK=1,15
    DETA(IJK) = DETA(IJK)*1 \cdot E-6
  8 CONTINUE
 10 CONTINUE
    READ(5,VAR,END=20)
 20 IF(EDF(5).NE.0) GD TD 999
    SUM = 0.
    DO 40 I=1, NGAS
    ANS = 0.
    DO 30 J=1, NGAS
    IF(I.EQ.J) GD TD 30
    IND1 = IPOINT(I)
    IND2 = IPOINT(J)
    ETA1 = ETA(IND1) + DETA(IND1)*(T - 293.15)
    ETA2 = ETA(IND2) + DETA(IND2)*(T - 293.15)
    PHI = 1. + SQRT(ETA1/ETA2)*(MASS(IND2)/MASS(IND1))
   1 **0.25
    PHI = PHI*PHI
    PHI = PHI/SQRT(8.*(1. + MASS(IND1)/MASS(IND2)))
    ANS = ANS + CONC(IND2) * PHI
 30 CONTINUE
    SUM = SUM + ETA1/(1. + ANS/CONC(IND1))
 40 CONTINUE
    WRITE(6,1)
    WPITE(6,2) NCASE
    WRITE(6,3)
    DD 50 I=1, NGAS
    IND = IPOINT(I)
    WRITE(6,4) SYMB(IND),CONC(IND)
 50 CONTINUE
    WRITE(6,5) SUM
    GD TD 10
999 CONTINUE
    STOP
    END
```

for the test gas.<sup>2</sup> Care is taken to keep the flow rates for the air and the test gas below  $50 \text{ cm}^3/\text{min}$  to ensure laminarity of the gas flows through the capillary section of the flowmeter. Figure 3 shows a correlation between the pressure difference across the test capillary section and the flow rate through it. It is obvious that, as would be expected for laminar flows from equation (2), the relationship between the flow rates and the corresponding differential pressures is linear for flow rates from 10 to 50  $\text{cm}^3/\text{min}$ . In this experimental arrangement, the flowmeter provides data for determining the volume flow rates, as well as a wellcharacterized capillary tube section for differential pressure measurement across it. It is this dual role of the flowmeter that constitutes the uniqueness of the method developed in this study. The results for binary and more complex mixtures are summarized below. All measurements were made at room temperature and at a pressure of 1 atmosphere. Figure 4 is a photograph of the viscosity measurement station.

*Purity of gases.* Table B lists the components and their purity as quoted by the suppliers.

Gas	Purity
Air	Dry grade; Dew point $= -80^{\circ}$ F
Ar	99.998%
$CO_2$	99.50%
$N_2$	99.998%
$\mathrm{CH}_4$	99.9995%
$C_2H_6$	99.00%
$C_3H_8$	99.95%
$C_4H_{10}$	99.50%

#### Table B

**Preparation of binary mixtures.** The binary mixtures were prepared by mixing appropriate volume flow rates of the individual gases. This procedure ensured complete interdiffusion of the two components. Selected gases were mixed in various ratios to produce a total flow rate from 20 to 50 cm<sup>3</sup>/min. Differential pressure measurements across the test capillary element of the gas flowmeter were made with a differential capacitance gage. Viscosity values for the test mixtures were then calculated by using equation (5). The results are summarized in table I. **Preparation of complex mixtures.** The more complex mixtures were prepared by a commercial vendor according to our specifications. The vendor prepared the mixtures by gravimetrically mixing different mass fractions, which were later expressed in appropriate mole fractions. The test mixtures were kept in their containers for several weeks before use to ensure complete mixing.

Four different mixtures containing different volume fractions of  $C_xH_y$  hydrocarbons as well as N<sub>2</sub> and CO<sub>2</sub> were prepared. Their viscosities were measured using the same procedure described for binary mixtures in the preceding section. The results are summarized in table II.

Natural gas. A test sample of the natural gas was obtained from the Virginia Natural Gas Company. Its coefficient of viscosity was measured by using the same procedure as for the binary and complex mixtures. The test sample was also analyzed for its composition. The composition data are summarized in table III. These values are averages of two measurements of the same sample. The accuracies of individual gas chromatographic analyses are of the order of 1 percent (relative). Using these data, the coefficient of viscosity of the sample was calculated, and the results are summarized in table IV.

**Pure gases.** Even though the main purpose of this study was to develop a technique for measuring the viscosity of multicomponent gas mixtures, it was deemed desirable to measure the viscosity of selected pure gases as a test of the accuracy of the proposed technique. These selected pure gases were treated as unknown test gases, and their viscosities were measured by using the same procedure as was used for the multicomponent mixtures. The present results, along with the values reported in the literature are summarized in table V. It is apparent that the two sets of data are in excellent agreement. All measured values are strongly dependent on the viscosity of air at room temperature and pressure. The viscosity of dry air has been taken to be 185.6  $\mu$ P. (See refs. 6 and 23.)

### Discussion

As seen from the data in table I, the agreement between the measured and calculated values of viscosity for binary mixtures is quite good. Also included in the table are some of the values reported by other authors. These values are also in good agreement with experimental values. Table II summarizes the experimental and calculated values for four complex mixtures prepared to simulate U.S. naturalgas samples. In these cases, the agreement is less

<sup>&</sup>lt;sup>2</sup> Pressure differences were measured across flowmeter 1 and across series capillary tube section C-1 (fig. 1). The values of the viscosity of pure gases calculated on the basis of these two independent pressure differences were found to be equal within approximately  $\pm 1$  percent. It was therefore decided to eliminate the series capillary section from all further measurements.

satisfactory—possibly because of uncertainties in the mixture compositions. To further resolve this issue, the viscosity of a Virginia Natural Gas sample was measured. This sample was also analyzed for its composition. The measured value of its viscosity using the present technique (independent of gas composition) is 106.7  $\mu P$  (±2.0): the expected value was 107.9  $\mu P$  (±2.0). These values should be compared with a calculated value of 111.6  $\mu P$  (±2.0) based on the listed gas composition. These results indicate that n (calculated) values for complex mixtures containing hydrocarbons are slightly different from  $\eta$  (Measured) values, possibly because of the limitations of Wilke's approximation of the classical dilute-gas state model. A direct measurement of the viscosity of the unknown test mixture may therefore be preferable to the one computed on the basis of its measured composition, since the limitations of Wilke's approximation of the dilute-gas state model make the latter less accurate for complex gas mixtures.

# **Concluding Remarks**

A variant of the general capillary method for measuring the viscosity of unknown gases has been developed. It is based on the use of a thermal-mass flowmeter whose critical sensing element simultaneously provides a capillary section for direct measurement of pressure drop. It is this dual role of the thermal-mass flowmeter that constitutes the uniqueness of the new technique. It does not require any prior information about the composition or other physical properties of the test gases if a dry test meter is used for initial calibration constant determination. It is particularly suited to very complex hydrocarbon mixtures where classical theory limitations as well as compositional errors make theoretical calculations less reliable. The results for pure gases indicate that the technique is accurate to within  $\pm 1$  percent. In fact, the only limiting factors appear to be the accu-. racies with which volume flow rates and differential pressure can be measured.

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# TABLE I. SUMMARY OF COEFFICIENTS OF VISCOSITY FOR BINARY MIXTURES

 $[T = 300 \ K; p = 1 \ atm]$ 

	Volume	Differential			
	flow rate,	pressure,			
Gas mixture,	cm <sup>3</sup> /min	torr	Coefficient of viscosity, $\mu P$		ty, $\mu P$
percent volume	(±1%)	$(\pm 0.25\%)$	$\eta(\text{Literature})$	$\bar{\eta}$ (Measured) <sup><math>\overline{a}</math></sup>	$\eta$ (Calculated) <sup>b</sup>
Air	48.9	0.693	185.6		
$24.9\% N_2 + 75.1\% CO_2$	46.6	.555	$^c158.2\pm1.6$	156.0	155.6
$50.2\% N_2 + 49.8\% CO_2$	45.8	.573	$^c165.7\pm1.7$	163.8	162.3
$75.0\% N_2 + 25.0\% CO_2$	46.1	.597	$^{c}172.9 \pm 1.7$	169.6	169.8
Air	48.9	0.708	, 185.6		
$25.1\% N_2 + 74.9\% Ar$	39.6	.683	$a_{215.8 \pm 0.2}$	221.1	216.9
$49.8\% N_2 + 50.2\% Ar$	39.0	.623	$^{d}204.3 \pm 0.2$	204.9	204.9
$75.3\% N_2 + 24.7\% Ar$	38.3	.560	$^{d}191.5 \pm 0.2$	187.3	192.1
$87.7\% N_2 + 12.3\% Ar$	35.9	.520	$^d186.1\pm0.2$	185.5	185.2
Air	45.9	0.689	185.6		
$24.4\% \text{ Ar} + 75.6\% \text{ CO}_2$	51.4	.690	$^{e}167.5 \pm 1.7$	166.1	164.8
48.8% Ar + $51.2%$ CO <sub>2</sub>	51.0	.762	$^{e}186.0 \pm 1.9$	184.9	181.9
$74.1\% \text{ Ar} + 25.9\% \text{ CO}_2$	50.6	.848	$^{e}205.5 \pm 2.1$	207.5	202.8
Air	48.8	0.671	185.6		
25.9% N <sub>2</sub> + 74.1% CH <sub>4</sub>	44.4	.435		132.2	130.8
50.8% N <sub>2</sub> + 49.2% CH <sub>4</sub>	44.1	.486		148.8	147.8
75.1% N <sub>2</sub> + 24.9% CH <sub>4</sub>	45.6	.554		164.0	162.9
Air	10.6	0.709	195.6		
$25.0\% \text{ N}_{\odot} \pm 74.1\% \text{ C}_{\odot}\text{H}_{\odot}$	49.0	0.700	105.0	119.1	100.4
$20.970 \text{ N}_2 + 74.170 \text{ C}_{2116}$ 50.1% No $\pm 40.0\%$ CoHo	45.2	.392		112.1	109.4
$75.4\%$ No $\pm 24.6\%$ CoHo	40.7	.440 566		1/8 5	1/0.0
$75.470$ $112 \pm 24.070$ $0.2116$	49.0	.000		140.0	149.9
Air	49.6	0.708	185.6		
$25.0\% \text{ CO}_2 + 75.0\% \text{ CH}_4$	44.2	.435	$e_{126.5 \pm 1.3}$	128.0	127.6
$50.0\% \text{ CO}_2 + 50.0\% \text{ CH}_4$	44.6	.473	$e^{138.1 \pm 1.4}$	137.9	138.0
$75.0\% \text{ CO}_2 + 25.0\% \text{ CH}_4$	46.1	.515	$e_{145.4 \pm 1.5}$	145.3	145.1
2					
Air	49.6	0.708	185.6	109.3	
$25.0\% \text{ CO}_2 + 75.0\% \text{ C}_2 \text{H}_6$	43.9	.369			108.1
$50.0\% \text{ CO}_2 + 50.0\% \text{ C}_2 \text{H}_6$	45.4	.436		124.9	121.9
$75.0\% \text{ CO}_2 + 25.0\% \text{ C}_2 \text{H}_6$	48.4	.503		135.1	135.9
Air	49.6	0.708	185.6		
$24.8\% \text{ CO}_2 + 75.2\% \text{ C}_3\text{H}_8$	36.2	.276		98.7	95.0
$49.9\% \text{ CO}_2 + 50.1\% \text{ C}_3\text{H}_8$	43.9	.387		113.9	109.9
$75.0\% \text{ CO}_2 + 25.0\% \text{ C}_3 \text{H}_8$	45.5	.462		131.3	127.9

<sup>*a*</sup>See footnotes on page 10.

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#### TABLE I. Concluded

 $[T = 300 \ K; p = 1 \ \text{atm}]$ 

	Volume	Differential			
	flow rate,	pressure,			
Gas mixture,	cm <sup>3</sup> /min	torr	Coeffi	cient of viscosit	y, μP
percent volume	(±1%)	$(\pm 0.25\%)$	$\eta$ (Literature)	$\bar{\eta}$ (Measured) <sup>a</sup>	$\eta$ (Calculated) <sup>b</sup>
Air	48.9	0.702	185.6		
$25.0\% \text{ CH}_4 + 75.0\% \text{ C}_2 \text{H}_6$	39.1	.298		98.5	98.0
$50.0\% \text{ CH}_4 + 50.0\% \text{ C}_2 \text{H}_6$	38.9	.307		102.1	102.0
$74.9\% \text{ CH}_4 + 25.1\% \text{ C}_2 \text{H}_6$	40.2	.330		106.1	106.6
Air	48.9	0.702	185.6		
$25.8\%~{ m CH_4}$ + $74.2\%~{ m C_3H_8}$	33.8	.224	f <sub>86.8</sub> g <sub>86.0</sub>	85.7	85.1
$50.0\% \mathrm{CH}_4 + 50.0\% \mathrm{C}_3\mathrm{H}_8$	39.8	.285	$f_{92.9}$ $g_{93.0}$	92.7	93.5
$74.9\% \ \mathrm{CH_4} + 25.1\% \ \mathrm{C_3H_8}$	39.2	.302	$f_{100.7}$ $g_{102.0}$	99.5	101.4
Air	48.9	0.702	185.6		
$25.0\% C_2 H_6 + 75.0\% C_3 H_8$	24.7	.165		86.4	85.0
$51.0\% C_2 H_6 + 49.0\% C_3 H_8$	36.8	.254		89.2	88.0
$74.9\% C_2 H_6 + 25.1\% C_3 H_8$	39.8	.283		91.9	91.0

<sup>a</sup>The uncertainties in  $\eta$  (Measured) values, determined by the accuracies of the gas flow rate and differential pressure measurements, are within the range of  $\pm 2.5$  percent in the present study.

<sup>b</sup>The uncertainties in  $\eta$  (Calculated) values, stemming from uncertainties in the mixture composition, are within the range of  $\pm 2$  percent.

<sup>c</sup>These values are the results of measurements of absolute viscosity using an oscillating disc viscometer (ref. 22).

 $^{d}$ These values are the results of relative measurements of viscosity using an oscillating disc viscometer (ref. 23).

<sup>e</sup>These binary mixture viscosity measurements were made using an oscillating disc viscometer (ref. 24).

<sup>f</sup>These data are cited in reference 15 for comparison between the experimental data and the calculated values. They are in agreement with Wilke's approximation to an accuracy of  $\pm 1$  percent.

<sup>g</sup>These are absolute values obtained using a high-pressure capillary tube viscometer. The reproducibility of the data has been reported at  $\pm 0.25$  percent (ref. 25).

# TABLE II. SUMMARY OF COEFFICIENTS OF VISCOSITY FOR COMPLEX MIXTURES

$\eta(\mathrm{Air}) = 185.6 \ \mu P;$
T = 300 K;
$p = 1  \mathrm{atm}$

		Differential			
Gas mixture,	Volume	pressure,			
percent volume	flow rate,	flow rate, torr C		Coefficient of viscosity, $\mu P$	
$(\pm 1\% \text{ relative})$	cm <sup>3</sup> /min	$(\pm 0.25\%)$	$\bar{\eta}$ (Measured) <sup>a</sup>	$\eta$ (Calculated) <sup>b</sup>	
Air	$48.9 \pm 0.5$	0.682			
$\begin{array}{r} 44.6\% \ \mathrm{C_2H_6} +\ 25.9\% \ \mathrm{C_3H_8} \\ +\ 10.8\% \ \mathrm{C_4H_{10}} + \\ 5.1\% \ \mathrm{CO_2} +\ 13.6\% \ \mathrm{N_2} \end{array}$	$31.8 \pm 1.0$	.220	$92.0 \pm 1.5$	$97.0\pm2.7$	
$\begin{array}{l} 53.0\% \ \mathrm{CH}_4 + 26.1\% \ \mathrm{C}_2\mathrm{H}_6 \\ + \ 18.4\% \ \mathrm{C}_3\mathrm{H}_8 \ + \\ 1.4\% \ \mathrm{CO}_2 \ + \ 1.0\% \ \mathrm{N}_2 \end{array}$	$39.3 \pm 1.2$	.318	$105.3 \pm 1.1$	$100.7 \pm 2.8$	
$\begin{array}{l} 50.8\% \ \mathrm{CH}_4 + 17.1\% \ \mathrm{C}_2\mathrm{H}_6 \\ + \ 24.9\% \ \mathrm{C}_3\mathrm{H}_8 \ + \\ 3.3\% \ \mathrm{CO}_2 \ + \ 3.8\% \ \mathrm{N}_2 \end{array}$	$36.8 \pm 1.4$	.298	$105.7\pm0.7$	$102.1\pm2.9$	
$\begin{array}{r} 51.5\% \ \mathrm{CH}_4 + 21.2\% \ \mathrm{C}_2\mathrm{H}_6 \\ + \ 23.8\% \ \mathrm{C}_3\mathrm{H}_8 \ + \\ 1.7\% \ \mathrm{CO}_2 \ + \ 1.7\% \ \mathrm{N}_2 \end{array}$	$44.6 \pm 0.9$	.308	$91.7\pm0.7$	$100.0 \pm 2.8$	

<sup>a</sup>The errors cited on  $\eta$  (Measured) are standard deviations on 5 measurements for each mixture. They do not include the effects of uncertainties in flow rates and differential pressure measurements.

<sup>b</sup>The uncertainties cited on  $\eta$ (Calculated) represent the effects of uncertainties in the mixture composition.

# TABLE III. COMPOSITION OF NATURAL GAS SAMPLE

# [Ref. 21]

Component gas	Volume percentage	
CH <sub>4</sub>	$96.52\pm0.20$	
$C_2H_6$	$2.32\pm0.08$	
$C_3H_8$	$0.40 \pm 0.03$	
$C_4H_{10}$	$0.14\pm0.02$	
$C_5H_{12}$	< 0.05	
$C_6H_{14}$	$\ll 0.05$	
$CO_2$	$0.22\pm0.02$	
N <sub>2</sub>	$0.39 \pm 0.06$	

# TABLE IV. SUMMARY OF COEFFICIENTS OF VISCOSITY FOR NATURAL GAS

$\eta$ (Measured)	$\eta$ (Calculated), <sup><i>a</i></sup> $\mu$ P	
		Based on gas composition
Based on gas composition	Independent of	summarized in
summarized in table $III^b$	gas composition <sup>c</sup>	table III
$107.9 \pm 2.0$	$106.7 \pm 2.0$	$111.6\pm2.0$

<sup>a</sup>The uncertainty cited on  $\eta$  (Calculated) represents the effects of uncertainties in the mixture composition.

 $^{b}$ The value measured on the basis of known gas composition is based on the flow-rate calibration constant calculated for that composition. The error cited on it is the standard deviation based on 6 measurements.

<sup>c</sup>The measured value listed in the second column has been determined using the experimentally determined flowmeter calibration constant for the test gas. The error cited on it is the standard deviation based on 6 measurements.

# TABLE V. SUMMARY OF COEFFICIENTS OF VISCOSITY FOR PURE GASES

	Volume	Differential			
	flow rate	pressure	Coefficient of viscosity, $\mu P$		
	cm <sup>3</sup> /min,	torr	Measured, $a^{-}$	Reported <sup><math>b</math></sup> (refs. 6 and 23)	
Gas	(±1%)	$(\pm 0.25\%)$	(average)	(±2%)	
Air	48.9	0.708		185.6	
N <sub>2</sub>	32.3	.449	$178.1\pm0.5$	178.6	
$CO_2$	35.6	.412	$150.3\pm0.1$	149.9	
Ar	41.0	.723	$228.2\pm0.2$	227.6	
$CH_4$	30.9	.260	$112.7\pm2.2$	112.0	
$C_2H_6$	37.1	.266	$95.7\pm2.2$	94.5	
$C_3H_8$	30.1	.198	$83.9 \pm 1.6$	82.5	
$C_4H_{10}$ (iso)	23.5	.141	$76.6 \pm 1.0$	75.7	

[T = 300 K;	p = 1	atm]
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 $^{a}$ The errors cited on the measured values are standard deviations based on 3-5 measurements. <sup>b</sup>The uncertainties on the reported values fall within the range of  $\pm 2$  percent.







Figure 2. Thermal flowmeter with laminar flow element.

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Figure 3. Flow rate versus differential pressure across test capillary section of flowmeter.



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Figure 4. Photograph of experimental viscosity measurement station.

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Coefficients of viscosity of various types of gas mixtures, including simulated natural-gas samples, have					
been measured at atmospheric pressure a	and room te	emperature using a	modified capil	lary tube method.	
Pressure drops across the "straight" cap	illary tube	section of a therma	il mass flowme	ter were measured	
for small, well-defined, volume flow rates for the test gases and for standard air. In this configuration, the flowmeter provides the volumetric flow rates as well as a well-characterized capillary section for					
					differential pressure measurements across
using the reported value of 185.6 $\mu$ P for the viscosity of air. The coefficients of viscosity for the				C-E) theory The	
experimental and calculated values for bi	narv mixtu	res are in agreemen	t within the re	ported accuracy of	
Wilke's approximation of the C-E theory	v. However.	the agreement for	multicompone	nt mixtures is less	
satisfactory, possibly because of the limitations of Wilke's approximation of the classical dilute-gas state					
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