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# Predicting the Viscosity of Pure Light Hydrocarbons

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## ABSTRACT

The following equation, which describes the viscosity of methane, ethane, propane and n-butane in the vapor, liquid and dense-fluid regions for densities up to 2.4 times the critical density, is presented.

$$\mu = \mu_{g,a} + A(e^{7.237\rho} - e^{-45.9\rho^2})$$

where  $A = 32.80 - 0.1637(M)$ , micropoises,

$\mu_{g,a}$  = gas viscosity at atmospheric pressure and the fluid temperature, micropoises,

$\rho$  = density of the fluid, gm/cc, and

$M$  = molecular weight

The atmospheric-pressure viscosity can be represented satisfactorily by Sutherland's equation for which values of the necessary constants are given. The equation represents the data on these materials over the entire region with a standard deviation of 1.6 per cent for 288 points. Except in the immediate vicinity of the critical density, the largest difference between predicted and observed viscosity was 4.3 per cent. To facilitate calculations, the equation is presented as a single curve of  $\mu - \mu_{g,a}$  evaluated for a gas of zero molecular weight. By modification of the co-ordinates, the curve becomes a straight line. The factor for converting the curve value of  $\mu - \mu_{g,a}$  to that for the actual gas is a linear function of molecular weight, and is also plotted.

## INTRODUCTION

The pressures at which fluids are produced, transferred and processed have increased steadily in the petroleum and chemical industries. This has resulted in increased interest in the effect of pressure on the thermodynamic and transport properties of fluids. The relationships derived from simple kinetic theory often may be applied in estimating gas properties for low and moderate pressures. These have the advantage of simplicity, a fact which has frequently led to use beyond the range of proper applicability. At high pressures and low temperatures, these relationships may be greatly in error, and other means of calculation are needed for the dense gas and liquid regions.

The thermodynamic properties of fluids have been studied extensively, both theoretically and experimentally. The volumetric behavior of a large number of fluids has been measured experimentally to high pressures for wide ranges of temperature. It is more difficult, however, to obtain accurate experimental values for transport properties, and detailed data have been obtained for very few fluids for extensive ranges of temperature and pressure. This situation has greatly handicapped correlation efforts.

Because of the limited data available on transport properties and the complex relationships which exist between the transport and thermodynamic properties, generally one of three methods has been applied to represent these properties for pure fluids.

1. Tabulations of each transport property at selected pressure and temperature intervals.

2. Equations for each transport property of each fluid which relate these properties to PVT behavior.

3. Generalized co-ordinate chart for each transport property, frequently with serious restrictions as to accuracy.

The methods are listed in order of decreasing accuracy. The first and second methods are limited to pure components and certain commonly occurring mixtures such as air for conditions other than atmospheric pressure. Equations have been developed from kinetic theory which quite accurately represent the temperature dependence of viscosity of gases<sup>21</sup> and liquids<sup>24</sup> at low pressures. Special equations have been developed to calculate the effects of pressure and temperature on viscosity of steam<sup>17,19</sup> and nitrogen,<sup>17</sup> but these equations are empirical and different for each fluid. No single equation is presently available for accurate prediction of viscosity in both the liquid and gas phases, for any fluid.

The third method is based on van der Waals' theory<sup>35</sup> of corresponding states. Uyehara and Watson<sup>36</sup> presented a plot of  $T_r$  vs  $\mu_r$  with lines of constant  $p_r$ , which is generally accurate within 10 per cent, but in the critical region errors may be as large as 30 per cent. Carr<sup>7</sup> and Comings, Mayland and Egly<sup>9</sup> used  $p_r$ ,  $T_r$  and  $\mu/\mu_{g,a}$  as parameters and developed generalized correlations for gases, and Carr extended his to include mixtures. For natural gases Carr's chart is generally accurate within  $\pm 3$  per cent, but is less accurate for heavier hydrocarbons or other gases.

The purpose of this study was to examine the viscosity-pressure-temperature data on the light hydrocarbons in their liquid, gas and dense-fluid regions, and to develop an equation which relates viscosity to the state properties. The form of the equation should be such that it will approach the kinetic-theory relationships for temperature dependence of viscosity for gases at low pressures, and for liquids at high densities.

## BACKGROUND

Most efforts at development of general relationships for prediction of fluid viscosities are based on equating the expression for momentum transfer per unit area, or shear force, developed by use of a molecular model to the defining equation for a continuous Newtonian fluid. The devel-

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<sup>21</sup>References given at end of paper.



opment based on the simple kinetic-theory model yields the expression for gases,

$$\mu = 2.715 \times 10^{-12} \sqrt{MT}/\sigma^2 \quad (1)$$

For real gases, the viscosity increases more rapidly with increasing temperature. The Sutherland<sup>21</sup> model for simple repulsive force yields

$$\mu = BT^{3/2}/(T + S), \quad (2)$$

which has been found to fit a great deal of the data for gases at atmospheric pressure.<sup>21</sup>

#### TEMPERATURE DEPENDENCE OF LIQUID VISCOSITY

The viscosity of a liquid is due primarily to drag caused by attractive forces between molecules in adjacent fluid layers. Since the influence of intermolecular forces decreases with increasing distance between molecules, and the density of liquids decreases with increasing temperature, liquid viscosity should also decrease with increasing temperatures.

Over 70 empirical formulas have been proposed to represent the temperature dependence of liquid viscosity.<sup>24</sup> In 1866, Reynolds<sup>25</sup> proposed

$$\mu = A e^{B/T} \quad (3)$$

Eyring,<sup>16</sup> Andrade<sup>2</sup> and Frenkel<sup>15</sup> have derived similar expressions based on the kinetic theory of liquids, each arriving at different sets of parameters involved in the constants *A* and *B*. Eq. 3 has been found to represent the temperature behavior of some types of liquids (including some of the hydrocarbons), with a high degree of accuracy, and to give very poor results with other types.

#### PRESSURE DEPENDENCE OF VISCOSITY

According to Eq. 1, the viscosity of a gas is independent of pressure or density; this is generally true for gases at pressures below 100 psia. At higher pressures the viscosity increases with increasing density, as in Fig. 1, which shows the effect of both temperature and pressure on the viscosity of ethane. Enskog<sup>8,14</sup> was the first to study the effect of density on viscosity, and his equations included two factors neglected in simple kinetic theory: (1) the transport of momentum on collision, and (2) a correction for the effect of the actual volume of the molecules on the probability of collisions.

The forms of equations for gas viscosity at high pressures fall into two groups:

$$\mu = \mu_{g,a}(f(\rho)) \quad (4)$$

or

$$\mu = \mu_{g,a} + g(\rho) \quad (5)$$

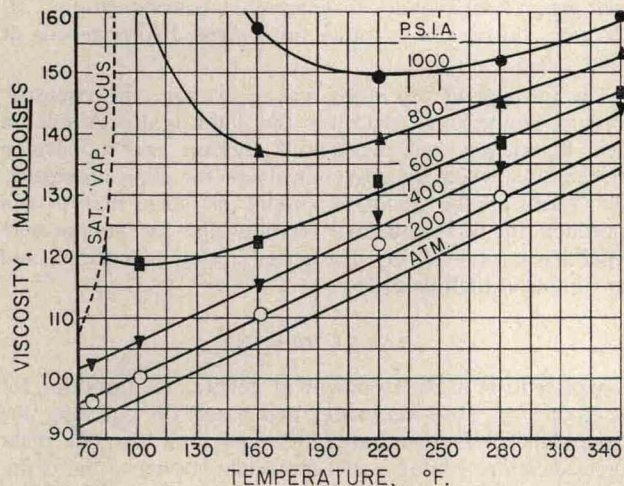


FIG. 1--EXPERIMENTAL ETHANE VISCOSITY VS TEMPERATURE AT LOW PRESSURE.

The theoretical equation of Enskog<sup>8</sup> and the empirical equations of Jager<sup>22</sup> and Dubief<sup>23</sup> are of the form of Eq. 4. The empirical equations of Vargaftik,<sup>27</sup> Jossi, Stiel and Thodos,<sup>18</sup> and the new relationship presented here, are the form of Eq. 5.

The viscosity of all liquids except water increases with increasing pressure. After an initial small decrease with increasing pressure, the viscosity of water also increases with pressure. For liquefied gases, and the light hydrocarbons at pressures up to 10,000 psia, the viscosity increases almost linearly with pressure, as is shown for propane in Fig. 2. At pressures greater than 30,000 psia, Suge<sup>30</sup> and Bridgeman<sup>6</sup> found that the viscosity increased almost exponentially with increasing pressure.

#### VISCOSITY AS THE SUM OF TWO FUNCTIONS

Recent theories of the liquid state imply that viscosity is the sum of two terms. Born and Green,<sup>4</sup> Kirkwood<sup>20</sup> and Eischitz<sup>23</sup> have developed theoretical equations, based on molecular distribution functions, which can be written

$$\mu = G_f(T, \rho) + G_m(T, \rho) \quad (6)$$

where *G<sub>f</sub>* = contribution due to drag-effect of intermolecular forces, and

*G<sub>m</sub>* = contribution due to transport of momentum by molecular thermal motion.

These theories tend to imply also that *G<sub>m</sub>* may depend on temperature alone. At least, when both *T* and  $\rho$  do not have large values, it can be assumed that

$$G_m(T, \rho) = G_o(T) \quad (7)$$

where *G<sub>o</sub>*(*T*) = the low-pressure gas viscosity dependence on temperature.

Abas-Zade<sup>1</sup> applied this concept to prediction of thermal conductivity of fluids. Thodos and co-workers<sup>5,18,27</sup> applied the concept for correlation of viscosity data. They plotted  $\mu - \mu_{g,a}$  vs density for the monatomic and diatomic molecules, and demonstrated that a single smooth curve resulted for each material. Starling, Eakin and Ellington<sup>20</sup> made a similar plot for propane data (Fig. 3) and, again, a single smooth curve resulted.

On the basis of these results, it was determined that

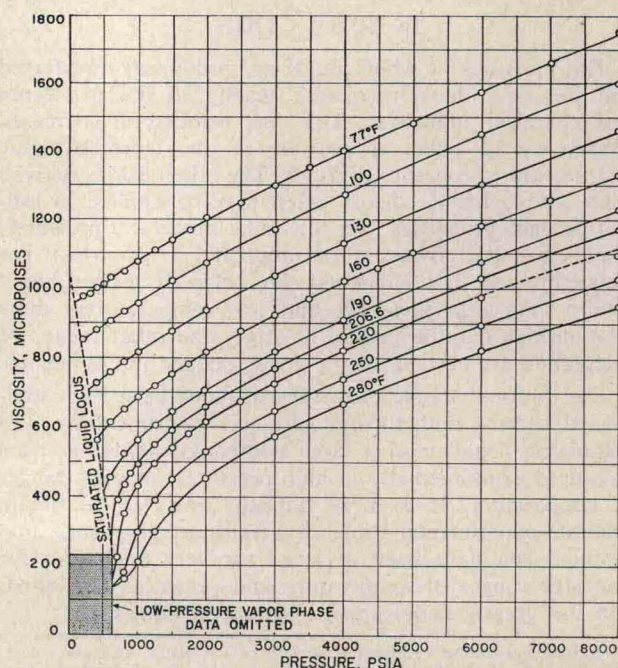


FIG. 2--EXPERIMENTAL PROPANE VISCOSITY VS PRESSURE.



the intermolecular-force term may be considered to be independent of temperature, and a function of density only (at least for the temperature ranges studied). Therefore, Eq. 6 can be rewritten

$$\mu = G_f(\rho) + G_m(T) \quad (8)$$

Jossi, Stiel and Thodos<sup>18</sup> developed a generalized viscosity equation based on this concept.

#### EXPERIMENTAL RESULTS

In the initial viscosity study at Institute of Gas Technology, Carr<sup>7</sup> utilized a high-pressure Rankine-type capillary viscometer to obtain relatively extensive data on methane and light natural gases. When attempts were made to obtain data on heavier hydrocarbons, this particular viscometer would not give reproducible results, even though Baron, Roof and Wells<sup>3</sup> utilized a similar instrument to obtain good data on nitrogen, methane, ethane and propane. A completely different type of viscometer was developed,<sup>11</sup> which has proved to be very flexible in operation, to yield data of high reproducibility and give over-all run times short enough that data fields of significant detail can be obtained in relatively short times. Data have been obtained for ethane,<sup>12</sup> propane<sup>20</sup> and n-butane<sup>10</sup>; and the critical point region of each material was investigated in great detail separately.<sup>28</sup> Work on binary mixtures is in progress.

#### STATE EQUATION FOR VISCOSITY

The proposed equation is based in part on the theoretical considerations reviewed and in part on observed behavior. At low densities the equation reduces to Eq. 2, Sutherland's equation for temperature dependence of gas viscosity. At high densities the equation approaches Eq. 3, which is fairly accurate for light hydrocarbon liquids. The equation also represents observed behavior in the region between that for dilute gas and that for liquid; there has been no satisfactory theoretical equation for this region.

To insure that the equation will represent the temperature dependence of liquid viscosity for high densities, it is necessary to transform Eq. 3 into the corresponding density function, since  $G_f$  is a function of density only. It is assumed that, for small temperature changes at con-

stant pressure, the linear equation for thermal expansion of a liquid is sufficiently accurate:<sup>23</sup>

$$\rho_t/\rho_i = 1 + a(T - T_i) \quad (9)$$

or, by expansion,

$$\rho_t/\rho_o \cong 1 - a(T - T_o) \quad (9a)$$

When Eq. 9 is solved for  $T$ , and this expression is substituted into Eq. 3,

$$\mu = Ae^{B/T_o + (1-\rho_t/\rho_o)/a} \quad (10)$$

or, by expansion,

$$\mu \cong Ae^{B(1-1/aT_o + \rho_t/aT_o\rho_o)/T_o} \quad (10a)$$

If  $K = Ae^{B(1-1/aT_o)/T_o}$  and  $b = B/aT_o^2\rho_o$ ,

$$\mu = Ke^{b\rho} \quad (11)$$

Eq. 11 might be expected to represent the dependence of the liquid viscosity on density only approximately, at best. If it should be valid, however, a semilog plot of  $\mu - \mu_{g,a}$  vs density should result in almost straight lines for each material at high densities. Fig. 4 shows this plot for the four hydrocarbons, for density in grams per cubic centimeter. The curves appear to be parallel straight lines at high densities. Thus, it is worthwhile to try to represent the residual viscosity,  $\mu - \mu_{g,a}$ , by an equation of the form of Eq. 11 for densities greater than about 0.23 gm/cc.

At low densities the residual viscosity is always less than that represented by Eq. 11, and it decreases to zero at very low densities. A general equation can therefore be written.

$$\mu - \mu_{g,a} - Ae^{b\rho} = g(\rho) \quad (12)$$

where  $g(\rho) \rightarrow 0$  as  $\rho \rightarrow 0.21$  gm/cc, and

$$g(\rho) \rightarrow -A \text{ as } \rho \rightarrow 0.$$

A form which satisfies these conditions and also represents the curvature of the data is

$$\mu - \mu_{g,a} = A(e^{b\rho} - e^{-C\rho^k}) \quad (13)$$

By examination of the high density values for ethane, propane and n-butane, it was determined that  $b=7.237$  cc/gm accurately represented the data when individual values of  $A$  were used for each component. Similarly, from the low density values it was determined that  $k=2.0$  and  $C=45.9$  (cc/gm)<sup>2</sup> accurately represented these data. The value of  $A$  for methane was evaluated from data of Carr<sup>7</sup> and Comings<sup>9</sup> by use of the  $b$ ,  $k$  and  $C$  previously determined, and represented the 100 data points with a standard deviation of only 1.03 per cent.

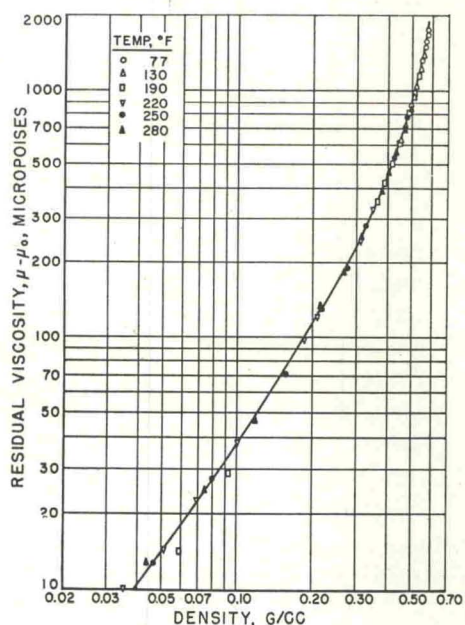


FIG. 3—RESIDUAL VISCOSITY VS DENSITY FOR PROPANE.

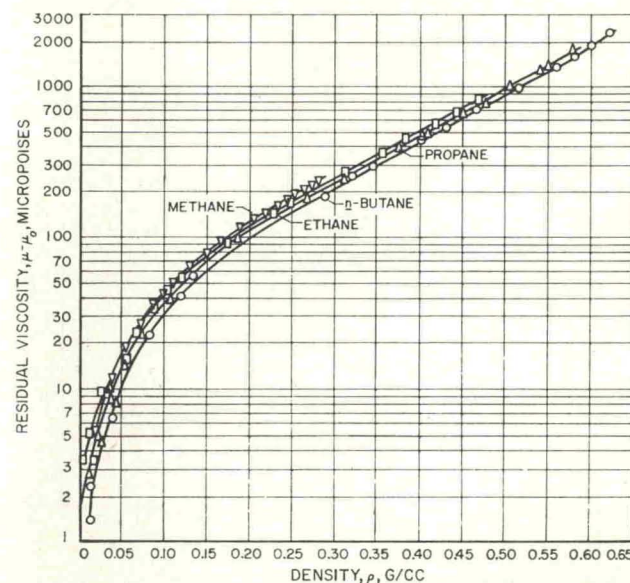


FIG. 4—SIMILARITY OF RESIDUAL VISCOSITY DEPENDENCE ON MASS DENSITY FOR FOUR HYDROCARBONS.



The values of the individual  $A$ 's were plotted vs molecular weight (Fig. 5) and found to yield essentially a straight line. Therefore, the general equation was rewritten as

$$A = A_0 - a(M), \dots \dots \dots (14)$$

and all of the data for the four hydrocarbons were treated simultaneously by computer to obtain

$$\mu - \mu_{g,a} = A(e^{7.237\rho} - 45.9\rho^2) \dots \dots \dots (15)$$

where  $A = 32.80 - 0.1637(M)$ , and

$$\mu_{g,a} = BT^{3/2}/(T+S).$$

Values of  $B$  and  $S$  for the four hydrocarbons are given in Table 1. A summary of observed and predicted viscosity values is given in Table 2.

This equation predicts the available data on methane, ethane, propane and n-butane from the dilute gas to densities up to 2.4 times the critical density with a standard deviation of 1.6 per cent for 288 points. The difference between the predicted and observed viscosity was always less than 4.4 per cent except for densities within 10 per cent of the critical density. For the same data, the results obtained with the equation of Jossi, Stiel and Thodos<sup>18</sup> are considerably less accurate, as shown in Table 3.

#### EQUATION REPRESENTED GRAPHICALLY

The graph in Fig. 6 was prepared to facilitate use of Eq. 15. The  $\mu - \mu_{g,a}$  co-ordinate was modified from a logarithmic scale so that a straight line results. This line represents the equation evaluated for a fluid of zero molecular weight. To convert the value obtained from the graph to that for the real gas at some given density, the graphical value is multiplied by a correction factor ( $X$ ).

$$(X) = \frac{A_0 - a(M)}{A_0} = 1 - 0.00499(M), \dots \dots (16)$$

which is also plotted in Fig. 6. To obtain the value of  $\mu - \mu_{g,a}$  for the real gas at a given density, the  $(\mu - \mu_{g,a})^*$  from the plot is multiplied by ( $X$ ) for the corresponding molecular weight.

$$\mu - \mu_{g,a} = (\mu - \mu_{g,a})^*(X) \dots \dots \dots (17)$$

#### APPLICATION TO MIXTURES

It is possible that the form of this equation may also be applicable to mixtures. The composition dependent para-

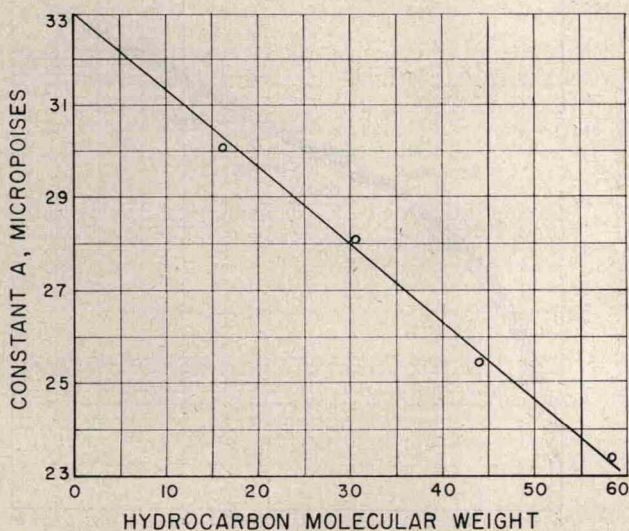


FIG. 5—APPARENT LINEAR RELATIONSHIP BETWEEN THE MOLECULAR WEIGHT AND CALCULATED CONSTANT "A" IN EQUATION.

TABLE 1—SUTHERLAND-EQUATION CONSTANTS FOR THE ATMOSPHERIC-PRESSURE VISCOSITY OF LIGHT HYDROCARBONS

Component	Sutherland Constants*		Data Ref.
	B	S	
Methane	7.390	295.2	(34)
Ethane	7.461	466.2	(34)
Propane	6.805	502.4	(34)
n-Butane	6.861	600.0	(26, 32, 33)

\*For temperatures in °R, calculates viscosity in micropoises.

TABLE 2—SUMMARY OF EXPERIMENTAL VALUES AND THOSE PREDICTED BY THE VISCOSITY EQUATION

Component	No. of Points, n	Std. Dev.* σ, (per cent)	Max. Dev. (per cent)
Methane	100	1.01	2.65
Ethane	47	1.61	3.52
Propane	88	1.79	4.30
n-Butane	53	2.42	7.92

$$* \sigma = 100 \left( \frac{\sum (\Delta_n)^2}{n-1} \right)^{1/2} \quad \text{and} \quad \Delta = \frac{\mu_{calc} - \mu}{\mu}$$

meter  $A$  is a linear function of molecular weight for pure components, and its behavior for mixtures should be examined. It is anticipated that  $\mu_{g,a}$  might be replaced by the atmospheric-pressure viscosity of the mixture,  $\mu_{m,a}$ , which Carr<sup>7</sup> showed could be represented by

$$\mu_{m,a} = \frac{\sum x_i \sqrt{M_i} \mu_{g,a,i}}{\sum x_i \sqrt{M_i}} \dots \dots \dots (18)$$

However, Eq. 15 has not been tested for mixtures due to the lack of data on a given mixture of known accuracy for wide-enough ranges of temperature and pressure.

#### NOMENCLATURE

- $A$  = constant
- $A_0$  = constant
- $a$  = constant
- $B$  = constant
- $b$  = constant
- $C$  = constant
- $f(\ )$  = function of variables in ( )
- $G_f(\ )$  = intermolecular-force contribution to viscosity
- $G_m(\ )$  = momentum-transfer contribution to viscosity
- $G_o(T)$  = atmospheric-pressure viscosity as function of  $T$
- $g(\ )$  = function of variables in ( )
- $K$  = constant
- $k$  = constant
- $M$  = molecular weight
- $p$  = pressure
- $S$  = Sutherland constant
- $T$  = temperature, absolute
- $T_r$  = reduced temperature
- $X$  = multiplying factor for graphical solution
- $x_i$  = mole fraction of the  $i$ th component
- $\rho$  = density
- $\rho_0$  = density at base temperature,  $T_0$
- $\rho_t$  = density at temperature  $T$
- $\sigma$  = molecular diameter
- $\mu$  = viscosity
- $\mu_{g,a}$  = atmospheric-pressure gas viscosity
- $\mu_r$  = reduced viscosity



TABLE 3—COMPARISON OF EQUATION OF JOSSI, et al (Ref. 18) WITH EXPERIMENTAL VISCOSITIES

Component	$\rho < \rho_c$ Points	Std. Dev.	Max. Dev.	$\rho > \rho_c$ Points	Std. Dev.	Max. Dev.	Over-all Std. Dev.
Methane	80	1.86	3.83	20	7.16	8.62	3.55
Ethane	29	1.46	3.28	18	5.53	7.97	3.44
Propane	20	4.08	8.12	68	12.47	15.86	11.11
n-Butane	14	4.11	6.70	39	11.71	15.64	10.22

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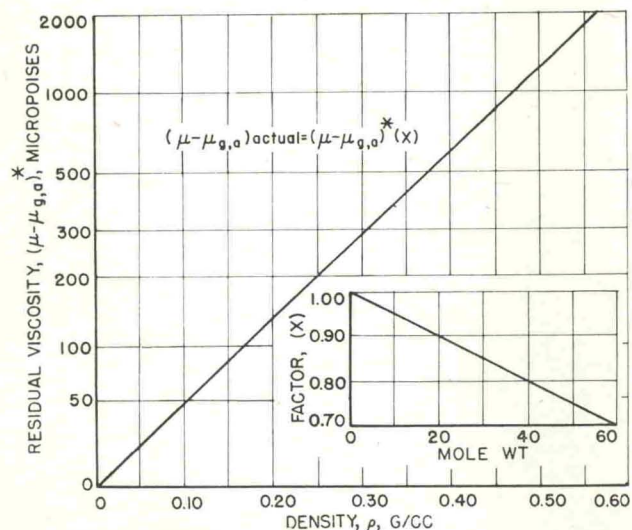


FIG. 6—GRAPHICAL PRESENTATION OF THE STATE EQUATION FOR VISCOSITY OF LIGHT HYDROCARBONS.