Viscosity of Liquids

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Three viscosity correlations were tested for liquids at their boiling points. Compounds studied included normal paraffins, branched-chain hydrocarbons, aromatics, halogenated methanes and ethanes, water, and aliphatic alcohols. The correlations were tested for the most part with viscosity and thermodynamic data found in the literature. Some experimental viscosity measurements were made, however, for normal paraffins from pentane to octane at temperatures up to about 300°F., which is higher than has previously been reported.

Two correlations are based on Eyring's theory of absolute reaction rates. The third is based on the compressibility factor of the saturated liquid, and this correlation is unique, as all available data for normal paraffins with eight or more carbon atoms are represented by a single curve. The three correlations presented here can be used to extrapolate viscosity data over large temperature ranges up to the critical temperature. Logical predictions of the viscosities of related compounds are possible.

The theory of absolute reaction rates (*17*) was applied to the viscosity of liquids by Eyring and coworkers (*16, 18, 22, 27, 39*). It was postulated that before a molecule can flow (move) a "hole" must be available for it to move into. Energy is required to make this hole, and the following equation was developed to represent the relationship between the free energy of activation and the viscosity in centipoises:

$$\mu = \frac{hN}{V} e^{\Delta F_{vis}/RT} \qquad (1)$$

Since a flow process can be considered to be at constant volume, the energy required to make a hole the size of a molecule is the internal energy of vaporization. It was thought (*22, 39*) that the relationship between the internal energy of vaporization and the free energy of activation might be significant. The ratio of these two values has been designated the energy ratio n:

$$n = \frac{\Delta E_{vap}}{\Delta F_{vis}} \qquad (2)$$

This energy ratio was reported (*22, 39*) to be equal to approximately 2.45 for numerous liquids including water, alcohols, and hydrocarbons. The alcohols and hydrocarbons tested or the temperatures of any of the liquids however were not specified. Since at the critical temperature the internal energy of vaporization is zero and since the free energy of activation is still presumably finite, the energy ratio is also probably zero at this temperature. It appears, therefore, that the energy ratio is a function of temperature.

Another equation (22) based on the theory of absolute reaction rates is as follows:

$$\mu = \frac{N}{V} (2\pi mkT)^{1/2} v_f^{1/3} \cdot \exp\,[(\Delta E_{vap}/n' + PV/n'')/RT] \qquad (3)$$

The numerator of the exponent is equal to the internal energy of activation. It was shown that the viscosity data of several compounds, including hydrocarbons, could be correlated at a given temperature over a large pressure range within about 30% accuracy. As far as can be determined, however, the equation was tested for a given compound at only one temperature.

Viscosity data have been extrapolated frequently in the past by means of the following plots: (1) viscosity vs. temperature, (2) logarithm of viscosity vs. the reciprocal of absolute temperature, and (3) logarithm of viscosity vs. the logarithm of absolute temperature. All these plots approximate straight lines over relatively short temperature ranges, but it seems as if accurate extrapolations are not possible for wide ranges of conditions. The present investigation was made, as a result, to develop better correlations or extrapolation methods. Hydrocarbons, chlorofluoro derivatives of methane and ethane, aliphatic alcohols, and water were studied primarily. A search was made of the literature to find viscosity, thermodynamic, and physical-property data of these compounds. In addition, some experimental viscosity measurements were made for normal paraffin hydrocarbons from butane to octane. Part of these measurements were at temperatures higher than those previously reported. Correlations based on Eyring's theory of absolute reaction rates and on the compressibility factor of the liquid were developed. These correlations are useful in extrapolating viscosity data over a wide temperature range.

APPARATUS AND MATERIAL USED

Viscosity measurements were made in a Humble-type high-pressure (10,000 lb./sq. in.) rolling-ball viscometer manufactured by Engineering Products, Inc. No corrections

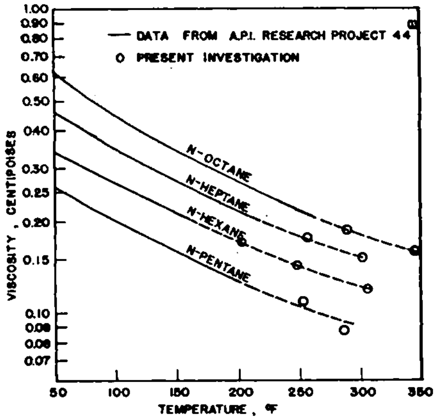

Fig. 1. Viscosity measurements of light-paraffin hydrocarbon liquids at their boiling points.

TABLE 1. VISCOSITY DATA OF THIS INVESTIGATION

Temp., °F.	μ, centipoises	n	T_r	ZZ_c
		n-pentane		
253.7	0.108	1.370	0.849	0.01110
286.0	0.086	1.216	0.883	0.01588
		n-hexane		
202.6	0.169	1.934	0.725	0.00289
248.1	0.142	1.703	0.774	0.00526
306.8	0.120	1.404	0.839	0.01002
		n-heptane		
256.6	0.175	1.864	0.737	0.00275
303.0	0.148	1.667	0.784	0.00500
		n-octane		
289.7	0.186	1.918	0.731	0.00238
347.2	0.158	1.661	0.787	0.00506

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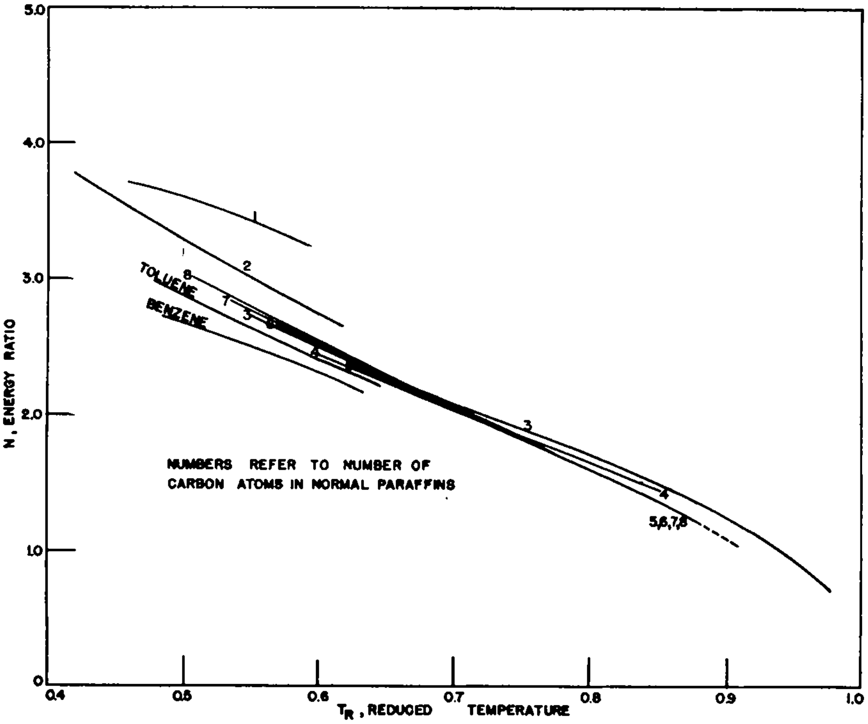

Fig. 2. Energy ratios of light normal paraffins and aromatics.

for pressure or temperature were required with the viscometer as the pressure was identical on each side of the sample chamber and because both the sample chamber and ball were constructed of steel. The steel ball was approximately ¼ in. in diameter, and the length of roll for the ball was about 7 in. A thyratron and sensitive ammeter were used to determine when the ball made contact at the bottom of the viscometer. A flexible high-pressure coil constructed of 1/16-in. O. D. tubing connected the viscometer to the auxiliary equipment, which included a calibrated pressure gauge, a vacuum pump, manometers, and connecting lines to the hydrocarbon cylinders. The viscometer and connections were immersed in a well-agitated oil bath. Bath temperatures were controlled manually with electric heaters to within 0.1°F. Calibrated mercury thermometers that could be read to within 0.1°F. were used for temperature measurements.

Research-grade *n*-butane, *n*-pentane, *n*-hexane, *n*-heptane, and *n*-octane were furnished by the Phillips Petroleum Company. The materials were reported to be at least 99% pure on a mole basis.

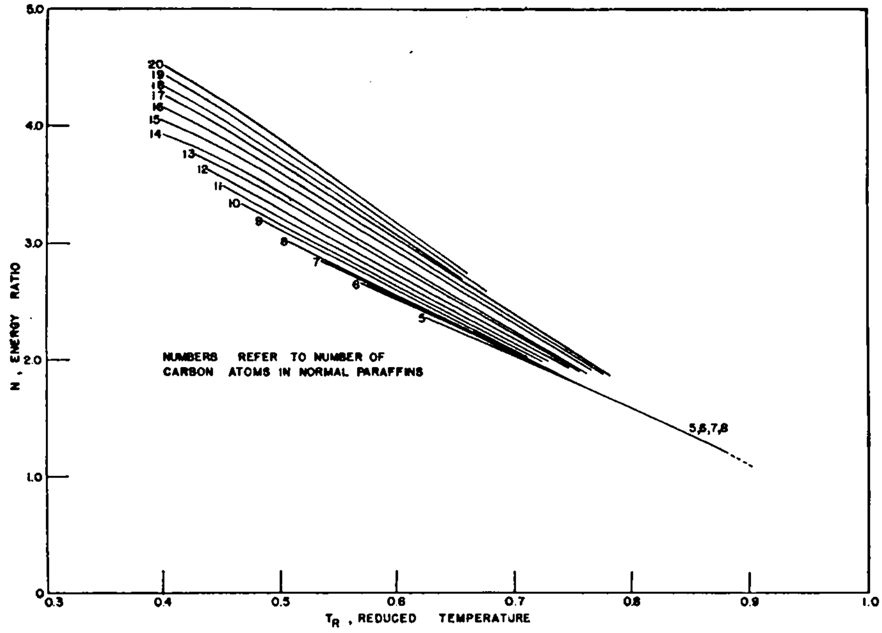

Fig. 3. Energy ratios of heavy normal paraffins.

OPERATION OF EQUIPMENT AND VISCOSITY RESULTS

After the viscometer was filled with a hydrocarbon sample, the apparatus was evacuated for several minutes to remove the air. The bath was then adjusted to a desired temperature, and the time of fall was measured with a stop watch until four or more readings reproducible within ± 0.15 sec. were obtained. The time of fall varied in this investigation from about 3 to 6 sec. The bath was then adjusted to other temperatures, and the procedure was repeated at each temperature. Data were obtained approximately every 50° from room temperature to about 300°F. for *n*-pentane to *n*-octane and up to 191°F. for *n*-butane.

Since viscosity information is available (*1*) for *n*-butane to *n*-octane at temperatures up to about 200°F., it was possible to construct a calibration curve for the viscometer, as suggested by Hubbard and Brown (*26*). The curve was a straight line except for slight curvature at low viscosities. The viscosity data of *American Petroleum Institute Project* 44 (*1*) agree within 5% of the "best" curve except for viscosity values of *n*-pentane from 120° to 160°F., which were about 8% low.

Using the calibration curve, one could determine the viscosities of *n*-pentane, *n*-hexane, *n*-heptane, and *n*-octane at temperatures up to approximately 100°F. above those previously reported. These results are shown in Table 1 and Figure 1. The viscosity data of this investigation produce "smooth" extensions of the curves, representing the results of *A.P.I. Project 44*.

CORRELATIONS BASED ON THE THEORY OF ABSOLUTE REACTION RATES

Viscosity data, in addition to those of this investigation, were obtained from the literature for paraffin and aromatic hydrocarbons (*1, 15, 21, 30, 34, 44, 45, 47*), chlorofluoro derivatives of methane and ethane (*5, 52, 53*), aliphatic alcohols (*51* through *54*), and water (*28, 33*). The free energy of activation for flow was calculated by means of Equation (1). Thermodynamic and physical information required for this equation was found in the literature for hydrocarbons (*1, 2, 4, 13, 19, 31, 35, 36, 43, 45, 46, 49, 50, 56*), halogenated hydrocarbons (*6* through *9, 11, 14, 20, 23, 56*), aliphatic alcohols (*3, 10* through *13, 24, 25, 29, 37, 38, 40* through *42, 48, 52, 55, 56*), and water (*28*). For most compounds it was necessary to calculate the internal energy of vaporization from the latent heat of vaporization as follows:

$$\Delta E_{vap} = \Delta H_{vap} - P\Delta V \qquad (4)$$

Typical values of the energy ratio *n* are shown in Table 1. Plots of the energy ratio vs. the reduced temperature for normal paraffin hydrocarbons, benzene, and toluene are shown in Figures 2 and 3. The energy ratio for these compounds decreases continuously with temperature. The correlations for all hydrocarbons are almost straight lines except at higher

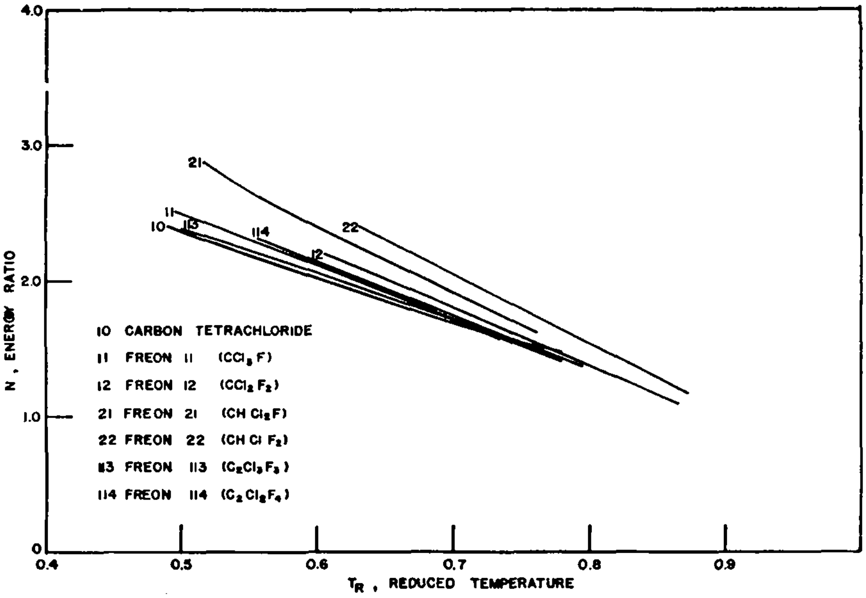

Fig. 4. Energy ratio of halogenated methanes and ethanes.

temperatures where all curve downward and tend to coincide as they approach zero at the critical temperature. Energy ratio values for a given reduced temperature, in general, decrease progressively from methane to pentane and then increase progressively from pentane to eicosane. For the most part, the curve for each hydrocarbon appears consistent. Possible exceptions may be the methane curve, the slope of which appears inconsistent with other hydrocarbons. The propane and butane correlations also seem rather inconsistent at high temperatures.

The energy-ratio curve of benzene is below that of toluene, and both lie below that of *n*-hexane. The viscosity and thermodynamic data for branch-chain saturated hydrocarbons are limited. As a result, it is impossible in most cases to calculate energy ratios over a large temperature range. Sufficient data are available though for these isomeric hydrocarbons with four to eight carbon atoms to indicate that the shapes of the energy-ratio curves are similar to those of normal and aromatic hydrocarbons. At a given reduced temperature the energy-ratio value of the isomeric hydrocarbon is always less than that of the comparable normal paraffin. Furthermore, the more branched the molecule is, the lower the energy ratio. The position of the branch chain apparently does not give any systematic trend of the energy-ratio values.

The halogenated hydrocarbon curves, as shown in Figure 4, are similar in shape to those of the regular hydrocarbons. The numerical values of the energy ratio for a given reduced temperature vary to a rather significant extent with the compound. The energy ratio is decreased with increased chlorine and simultaneous decreased fluorine content and with increased chlorine and decreased hydrogen content. The curves for both halogenated methanes and ethanes appear to be relatively consistent. A possible exception is the curve of carbon tetrachloride, the slope of which is rather different. Trichlorotrifluoroethane also seems to have a rather inconsistent curve at high temperatures.

The energy-ratio correlations for water and aliphatic alcohols are, in general, curved to a rather appreciable extent, as Figure 5 shows. In several cases the energy-ratio values pass through maxima at reduced temperatures from 0.5 to 0.7. As the number of carbon atoms increase and branched isomerization occurs, energy-ratio values decrease.

Efforts were also made to evaluate Equation (3) over a wide temperature range. The term pV/n'' was found to be insignificant at the relatively low pressures of saturated liquids if n' and n'' are of the same order of magnitude as previously indicated (*22*). Consequently, this term was dropped in the present study. The free volume v_f can be determined by use of velocity-of-sound data of both the gas and the liquid. Such data are apparently limited, but sufficient were found (*57*) to calculate free volumes of several compounds for, at least, one temperature. The following equation (*22*) was used to extrapolate values of the free volume:

$$v_f = c^3(v^{1/3} - d)^3 \qquad (5)$$

The term c depends on how the liquid molecules are packed, and it is reported to vary with temperature. It was assumed, however, that the molecules are packed cubically, for which c equals 2.0. By use of a known value of v_f, d was then computed for various compounds as shown in Table 2. Free volumes were determined over a wide temperature range.

Values for the free volume and viscosity at a given temperature were substituted into Equation (3), and n' was calculated. Values of n' determined in this manner are shown in Table 2. The viscosity was calculated over the temperature range comparable with that of the experimental data. The computed and experimental viscosity values of all

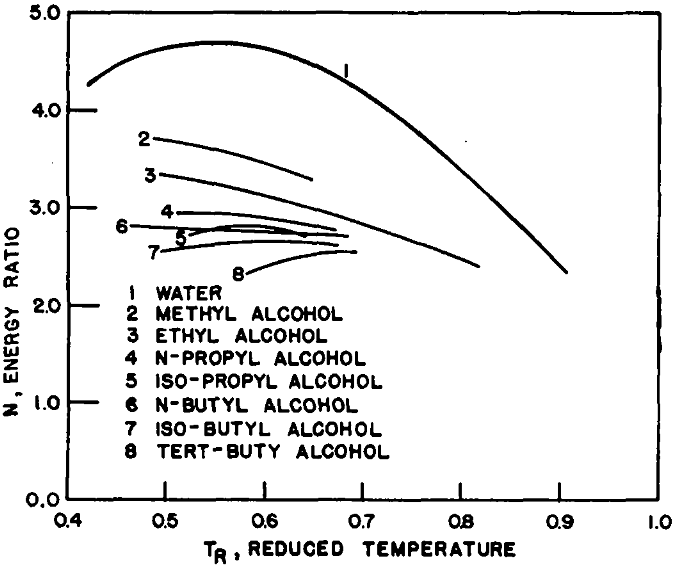

Fig. 5. Energy ratios of water and alcohols.

hydrocarbons tested agree within at least 10 to 30%. A typical comparison is shown in Table 3 for *n*-pentane. Other hydrocarbons studied include *n*-hexane, benzene, and toluene. Poorer agreement was found for carbon tetrachloride. In the case of water, Equation (3) is not applicable.

The free volume can also be calculated (*22*) by use of the internal energy of vaporization. Values calculated in this manner differ significantly and give poorer correlations of the viscosity than those determined from sonic data.

COMPRESSIBILITY-FACTOR-PRODUCT CORRELATION

The viscosity of the saturated liquid was correlated as a function of the compressibility factor product ZZ_c. This term is the product of the compressibility factor of the saturated liquid Z and the compressibility factor at the critical point Z_c. Typical values for ZZ_c are shown in Table 1. The correlation was tested for normal paraffins and freons by plotting the logarithm of viscosity in centipoises vs. the logarithm of ZZ_c as shown in Figures 6 and 7 respectively. The curve for each compound tested was found to be almost a straight line except, in some cases, at high values of ZZ_c, that is, at high temperatures. For a given value of ZZ_c viscosity values increase for paraffins from methane to octane. The unique feature of this correlation, however, is that all available viscosity data of normal paraffin hydrocarbons containing eight or more carbon atoms are represented within 3.0% by a single curve. Sufficient data are also available to test these correlations for branched-chain paraffins with up to eight carbon atoms. In general, for a given ZZ_c value isomerized hydrocarbons have higher viscosities than straight-chain hydrocarbons. Some of the less branched isomers of heptane and octane, however, have lower viscosity values. Although the ranges of data are limited, it appears as if the correlations for the isomerized compounds are relatively parallel to those of the normal paraffins.

The curves for the halogenated methanes and ethanes are also almost straight lines. For a given value of the compressibility-factor product, it was found that the viscosity increases with increased chlorine and decreased fluorine content, increased chlorine and decreased hydrogen content, and increased number of carbons in the atom. The carbon tetrachloride curve has a rather different slope from that of any of the other freons.

DISCUSSION OF RESULTS

The experimental viscosity measurements obtained from about 200° to 300°F. for *n*-pentane to *n*-octane appear to extend consistently the viscosity results of *A.P.I. Project* 44. This is to be expected as the viscometer was calibrated by use of the results of that study. It is estimated that the calibration curve used for the present viscometer was accurate to within 5%. Experimental inaccuracies of temperature measurements and the time of fall of the ball may cause, for any given viscosity measurement, errors as high as 5%. The total errors of the viscosity results of this investigation are probably less than 10%.

The graphical viscosity correlations (Figures 2 to 7) developed in this study were examined in regard to the position and slope of the various curves. In most cases, if a certain compound appears inconsistent on the graph based on the theory of absolute reaction rates, it is also inconsistent in the correlation of the compressibility-factor product. Compounds in this category are methane, carbon tetrachloride, and at high temperature both propane and *n*-butane. It is possible that the curves for these compounds as shown on the graphs are correct, but it seems more likely that the curves are somewhat incorrect because of inaccuracies of the viscosity and/or thermodynamic data. Since viscosity and saturated liquid volumes are the common factors of both correlations, these seem like the most questionable terms. Trichlorotrifluoroethane appears somewhat inconsistent at higher temperatures on Figure 4 only. Apparently values of the internal heat of vaporization for this compound are somewhat in error.

TABLE 2. VALUES OF n' AND d FOR COMPOUNDS STUDIED

	$d \times 10^8$	n'
n-Pentane	5.22	4.16
n-Hexane	5.55	4.35
Benzene	4.90	2.11
Toluene	5.24	2.35
Carbon tetrachloride	5.03	3.48
Water	2.66	5.4

Earlier it was reported (*22, 39*) that the energy ratio n of Equation (2) was a constant equal to approximately 2.45 for many compounds. As determined here (*see* Figures 2 to 5), energy-ratio values vary, however, from at least 1.0 to 4.7. Even at the normal boiling points of the liquids, the values vary from about 1.9 to 4.7. On the basis of the present investigation, it must be concluded that the energy ratio is definitely a function of temperature and varies over a relatively wide range of values.

Although graphs such as Figures 2 to 5 are relatively complicated to construct, they are considered more accurate for extrapolation purposes than simpler plots of viscosity vs. some function of temperature. The energy-ratio curves were found not only to be relatively straight over most of the temperature range, but in addition to extrapolate to zero at a reduced temperature of one. As Figures 2 to 5 indicate, however, the curvatures of these correlations must be large at high temperatures. In addition, it was found that the energy-ratio values at high temperatures were sensitive to small changes of the values of viscosity, saturated liquid volume, and latent heat of vaporization. Near the critical temperature the energy-ratio extrapolation is, as a result, only an approximate guide for predicting viscosity values.

TABLE 3. VISCOSITY OF LIQUID *n*-PENTANE

t, °F.	Experimental viscosity, centipoises	Viscosity calculated with Equation (3)	% Deviation
68	0.235*	0.235	0.0
95	0.207*	0.215	+ 4.0
160	0.150*	0.170	+13.3
200	0.126*	0.147	+16.7
253.7	0.108†	0.123	+13.9
286.0	0.086†	0.111	+29.0

**A.P.I. Project* 44.
†Present investigation.

It was noted that several of the normal paraffin curves of the energy-ratio values tend to coincide at high temperatures, e.g., *n*-pentane to *n*-octane. If it is assumed that the curves of the heavier normal paraffins coincide at higher temperatures, it is possible to calculate viscosity values for several of the compounds at temperatures much higher than those reported in the literature. Viscosity values for *n*-octane were calculated at reduced temperatures of 0.85 and 0.90, as shown in Table 4, and the octane line of Figure 6 was extrapolated as shown by the dotted portion of the curve.

The correlation based on the compressibility-factor product is considered empirical in nature, but a single curve was found to represent all available viscosity data for normal paraffins from octane to eicosane. It seems logical to assume, therefore, that the paraffins higher than

TABLE 4. EXTRAPOLATION OF *n*-OCTANE VISCOSITY VALUES

T_R	Temp., °F.	n	ΔE_{vap}, cal./g. mole	V_L, cc./g. mole	μ	ZZ_c
0.85	410	1.37	−10,310	225	0.137	0.00908
0.90	462	1.10	− 8,560	256	0.109	0.01609

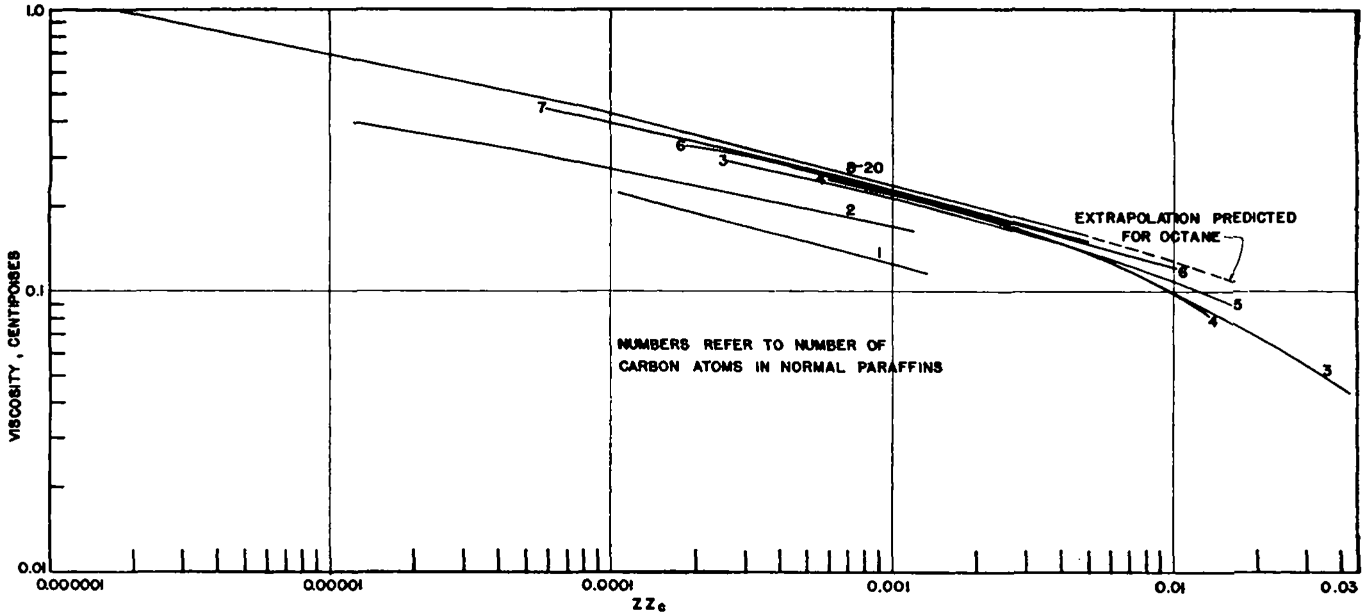

Fig. 6. Viscosity of normal paraffins as a function of compressibility-factor product.

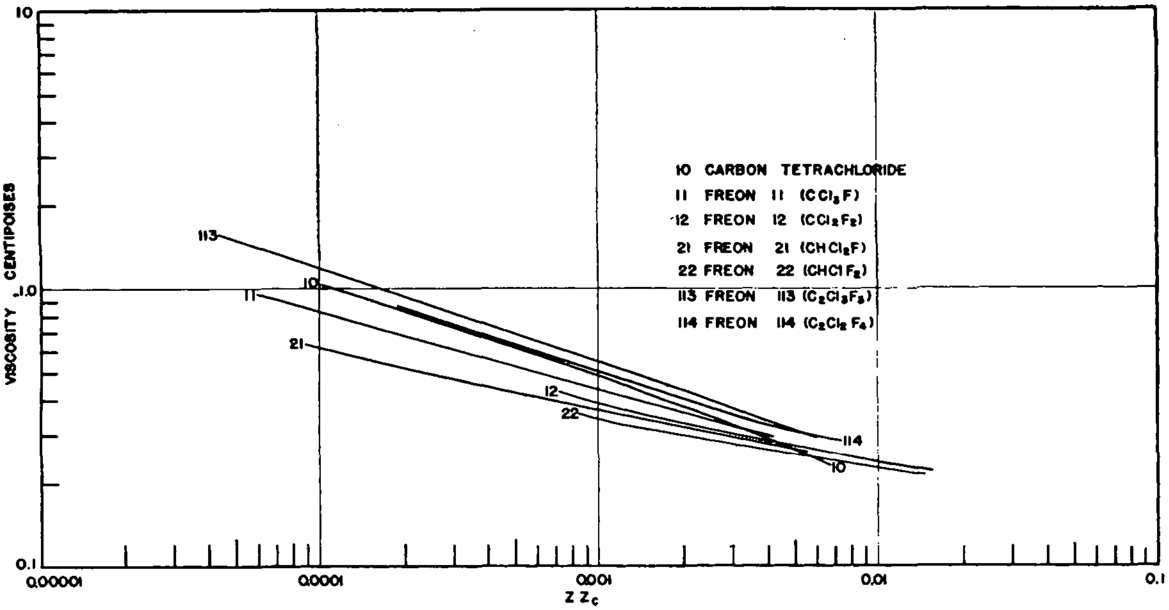

Fig. 7. Viscosity of halogenated methanes and ethanes as a function of compressibility-factor product.

eicosane are also represented by the same curve. This represents a useful method in estimating the viscosities of compounds for which the literature reports no data. It would be interesting, however, to check this theory by obtaining some viscosity data for a few such compounds. In addition, this single curve can be used to extrapolate the viscosity of the normal paraffins from octane to eicosane over the entire temperature range covered. In many cases this serves as a means for an extrapolation over an extremely large range of temperatures.

The available viscosity data for many branched-chain hydrocarbons are limited. Both graphical viscosity correlations are useful for extrapolation purposes if it is assumed, as seems likely, that the slopes of their curves are similar to those of related normal hydrocarbons. As more data become available for these compounds, it may be possible to establish systematic trends for predicting the energy ratio (and viscosity) of branched-chain compounds by comparison with normal hydrocarbons.

The two graphical correlations should be of value in predicting the viscosity of compounds similar to those studied here; for example, by comparison of the curves of Figures 4 and 7 for CCl_4, CCl_3F, and CCl_2F_2, it is probable that the approximate positions of the $CClF_3$ and possibly even the CF_4 curve could be estimated on each graph.

The energy-ratio curves for the associated liquids are unique primarily because some pass through a maximum. This abnormality is not surprising when it is considered that the internal energy of vaporization produces a "hole" the size of a simple molecule. It is known that associated liquids may be aggregrates of two or more simple molecules. It is possible that if the actual energy required to form a full-size molar hole were known, the energy-ratio curves would be similar in shape to those of unassociated molecules.

The results of this study indicate that Equation (3) can be used to correlate with at least fair accuracy the viscosity data of several hydrocarbon liquids over large temperature ranges. The method used for evaluating free volumes by means of some sonic data and Equation (4) was shown to be superior, at least for correlation purposes, to the method using the internal energy of vaporization. The method used here is considered, however, only an approximation. It is possible that when more accurate methods become available for calculating free volumes Equation (3) will correlate the viscosity data even better than the present results indicate. Although only four hydrocarbons were tested in regard to Equation (3), it is probable that other hydrocarbons and related compounds could be correlated when accurate free-volume data become available. Viscosity data for water could not, however, be correlated by this equation, and so it might be concluded that associated liquids could not be, either. Since it has been reported (*22*) that the equation correlates fairly well the viscosity data of liquid hydrocarbons at a given temperature over a large pressure range, it seems as if the equation will represent with fair accuracy the viscosity data of these liquids at all pressures and temperatures.

CONCLUSIONS

The viscosity correlations which were tested are useful for extending viscosity data over larger temperature ranges than was previously possible. In addition, it is frequently possible to predict the viscosities of compounds in homologous series.

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NOTATION

c = packing coefficient
d = incompressible diameter of molecule
h = Planck's constant
k = Boltzmann constant
n, n', and n'' = energy ratios
N = Avogadro's number
m = reduced mass
P = vapor pressure
R = gas constant
T = absolute temperature
T_r = reduced temperature
v = molecular volume of saturated liquid
V = molar volume of saturated liquid
v_f = free volume
Z = compressibility factor of liquid, PV/RT
Z_c = compressibility factor at critical point, P_cV_c/RT_c
ΔE_{vap} = internal energy of vaporization
ΔH_{vap} = latent heat of vaporization
ΔF_{vis} = free energy of activation for flow
ΔV = volume change for vaporization
π = 3.1416
μ = viscosity, centipoises

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