DEC 2 3 1966

Liquid, Gas, and Dense Fluid Viscosity of Ethane

EAKI BE62 0108

B. E. EAKIN, K. E. STARLING, J. P. DOLAN, and R. T. ELLINGTON Institute of Gas Technology, Chicago, III.

THIS INVESTIGATION was conducted under one phase of a continuing effort to meet some of the increasing needs of industry for thermodynamic and physical property data of fluids for more extreme conditions and for improved generalizations of behavior. The investigation of the viscosity behavior of propane for wide ranges of conditions has been reported (17), that for butane is being prepared for release, and that for ethane is reported herein.

The new experimental data define ethane viscosity behavior for the liquid, gas, and dense fluid. These data are correlated with those of other investigators to determine the best values for considerable ranges of temperature and pressure. The concept of residual viscosity dependence on density is shown to yield significant rectification of the data for densities greater than 0.10 gram per cc.

The residual viscosity concept and the kinetic theory are utilized to extrapolate the data to elevated temperatures and pressures with an accuracy believed within $\pm 2\%$. Recommended values for ethane viscosity are presented for temperatures from 70° to 460° F. and pressures from atmospheric to 10,000 p.s.i.a.

APPARATUS AND MATERIALS

A capillary tube viscometer of new design was utilized for the study of ethane viscosity behavior. The operation of the instrument is based on the principle of the Rankine viscometer, but an improved method is used for obtaining the pressure differential causing fluid flow. The viscometer is an absolute instrument and therefore does not require calibration; the instrument has been described, with estimates of the precision and accuracy of measurements (8).

The ethane used for this investigation was Phillips Petroleum research grade, certified 99.9 mole % pure. Mass spectrometer analyses indicated no impurities in the samples tested, so use of 90° F. and 715 p.s.i.a. for its critical point seemed valid.

EXPERIMENTAL DATA

To define ethane viscosity behavior adequately, it was necessary to obtain new data for a wide range of conditions. Data were obtained at 77°, 100°, 160°, 220°, 280°, and 340° F.; the pressure range was 200 to 8000 p.s.i.a. for the first four temperatures and 200 to 2000 p.s.i.a. for the last two. Data for both the liquid and gas were obtained at 77° F. At 100°, 160°, and 220° F. data were obtained both above and below the critical density, and at the two top temperatures all values are for densities less than the critical. Experimental values were not needed to determine the viscosity of the dense fluid at these temperatures.

Isotherms of ethane viscosity are presented in Figure 1. The isotherm values were plotted vs. pressure as data were obtained. A cross plot of viscosity vs. temperature (Figure 2) was prepared to locate questionable points and to check the internal consistency of the data. Portions of the experimental data have been omitted from Figures 1 and 2 for clarity. The data for high temperatures, omitted from Figure 1, are shown clearly in Figure 2. Detailed tables of the experimental data are available from ADI.





Figure 1. Experimental ethane viscosity vs. pressure



For all but two of the 81 test points, runs were made for at least two different pressure differentials between the ends of the capillary tube. In general, two to five flow times were averaged for a given pressure differential. The deviations of trial flow times were usually less than 0.1% of the mean. The mean flow time for each run was used to calculate the viscosity and the average of these run values was recorded as the viscosity for the test point. Data obtained on different dates but under common test conditions were calculated as separate runs and then averaged. Deviations of individual run values from the mean were generally small. For 1000 p.s.i.a. or above, the average deviation was 0.14%; below 1000 p.s.i.a. it was 0.55%.

COMPARISON WITH LITERATURE VALUES

A number of investigations of the viscosity behavior of ethane have been reported. Two investigations (3, 16)extend to high pressures, one (19) determined saturated liquid values, and nine (2, 6, 7, 9, 10, 11, 20, 21, 23) were made at atmospheric pressure. Several methods were used to compare the various data sets and assess the validity of our results. Selected data were then correlated to permit development of a table of recommended values, believed accurate to within $\pm 2\%$ of the true value, and extending beyond the range of temperatures and pressures covered by existing data.

The data of the different investigations were plotted as isotherms of viscosity vs. pressure and as isobars of viscosity vs. temperature. These plots tested the continuity of the values and checked on the consistency of the data. In this manner inconsistent points were located and reconciled. As a further test, data from the several sets were plotted as residual viscosity vs. density. Residual viscosity is defined as the difference between the viscosity at a given pressure and temperature and the viscosity at 1 atm. and the same temperature. The residual concept was proposed by Predvoditelev (12), verified by Abas-Zade (1) for thermal conductivity, and extended by Thodos and others (4, 15) for viscosity. A residual viscosity-density plot was utilized in the study of the viscosity behavior of propane (17). Atmospheric pressure data were compared by use of Sutherland's equation. This permitted easy determination of differences in temperature dependence of sets of data, when the agreement between values was good for a narrow range of conditions.

Ethane viscosity data for the saturated liquid and for pressures slightly above the vapor pressure were obtained by Swift, Lohrenz, and Kurata (19). Their values agree almost perfectly with the saturated liquid locus estimated by extrapolating isotherm data of this investigation to the vapor pressure.

Ethane viscosity data at atmospheric pressure were obtained by Vogel (23), Ishida (9), Titani (20), Trautz and Sorg (21), Adzumi (2), Majumdar and Oka (11), Craven and Lambert (6), Lambert and others (10), and DeRocco and Halford (7). There is good agreement among all the data. However, Titani's and Adzumi's values show a greater temperature dependence than the values of Trautz and Sorg, Lambert and others, and DeRocco and Halford. To illustrate this, the constants in Sutherland's equation (18)

$$\mu_0 = CT^{3/2} / (T+S) \tag{1}$$

where μ_0 is in micropoises and T in °K., determined from the reported data, can be compared. The values of the constants are

	C	S
Titani	10.7	287
Adzumi	10.6	280
Trautz and Sorg	10.0	259

The DeRocco and Halford values would be very close to Trautz and Sorg's. The obvious reason for the differences is that constants fitted to data for different ranges of temperature may differ slightly. Certainly, at higher temperatures one would expect viscosity to become more nearly proportional to $T^{1/2}$. Thus, although agreement is good, the Sutherland constants differ mainly because the data of Trautz and Sorg and DeRocco and Halford extend to 250° and 200° C., respectively, while the highest temperatures of Titani's and Adzumi's investigations were 120° and 100° C.

Baron, Roof, and Wells (3) utilized a modified Rankine capillary tube viscometer to obtain data at four temperatures from 125° to 275° F. for pressures up to 8000 p.s.i.a. Their values and the experimental data of this investigation agree within 3% for most points. Smith and Brown (16) used a rolling ball viscometer to obtain data at 12 temperatures from 59° to 392° F. for pressures up to 5000 p.s.i.a. Their values are generally more than 3% lower than the data of this investigation and for many points the deviations exceed 10%.

The experimental data of this investigation, plotted as residual viscosity vs. density on linear coordinates, result in a smooth continuous curve at densities above 0.10 gram per cc., as shown in Figure 3. Density values used for constructing plots of residual viscosity vs. density are those reported by Sage and others (13, 14). The atmospheric



Figure 3. Residual ethane viscosity vs. density Linear coordinates

pressure viscosity values were calculated by use of Sutherland's equation with the constants determined from the data of Trautz and Sorg. To expand the low density region, the residual viscosity-density data were plotted also on logarithmic coordinates. This plot (Figure 4) shows definite separation of the smooth curve into a family of isotherms at densities below 0.10 gram per cc. Correlation of this behavior may be possible for similar substances on a reduced basis and is being investigated. The separation in Figure 4 is therefore shown in terms of the reduced temperature, T_R . A curve for the reduced temperature corresponding to 220° F. ($T_R = 1.236$) is not present in Figure 4, because the 220° F. data at the lowest densities are probably about 1 micropoise high. Both the linear and logarithmic residual viscosity-density plots indicate that for densities above 0.10 gram per cc. all the IGT data are correlated by a single curve.

The data of Baron, Roof, and Wells, portions of the data of Smith and Brown, and the data of this investigation (the smooth curve) are directly compared in Figure 5. This shows the close agreement of the values of Baron and others and the disagreement of the values of 3 mith and Brown. Only the 392° F. values lie generally within 2% of the other sets.

SELECTION OF RECOMMENDED VALUES

The data of Baron and others made selection of recommended values for the viscosity of ethane much easier. The agreement of their values with the data of this investigation is significant and many of the previous discrepancies can be resolved adequately on this basis alone. Also, the agreement



Figure 4. Residual ethane viscosity vs. density Logarithmic coordinates



Figure 5. Comparison of IGT ethane viscosity data with those of other investigators

among the atmospheric pressure data made quantitative use of the residual viscosity concept possible.

Recommended values for the regions of temperature and pressure of this experimental investigation were selected by reading the values from large scale, smoothed viscositypressure, viscosity-temperature, and residual viscositydensity plots. Except at a few scattered points where the data were inconsistent, the same viscosity value was obtained from each plot.

Much emphasis has been placed on the residual viscosity concept. It is believed that the residual plot represents the best available method for predicting fluid viscosity for a large range of conditions from a comparatively small quantity of experimental data. Of course, accurate atmospheric pressure viscosity values are essential and the experimental data must cover a sufficient range of densities. Therefore, the recommended values for extreme pressures and densities above 0.10 gram per cc. were read from a large scale residual viscosity-density plot similar to Figure 3. To determine the viscosity of ethane for the highest pressures at 70° and 100° F, the smooth curve was extended slightly.

Because of the low density data separation, the residual plot could not be used to predict low density viscosity accurately at high temperatures. The problem of determining the best values for this region was resolved from the fact that the low pressure data nearly parallel the atmospheric pressure values when plotted as viscosity *vs.* temperature. This behavior, shown in Figure 6, is predicted from kinetic theory. Thus, accurate extrapolations of the low pressure curves to higher temperatures should be possible, using the atmospheric curve as a base.



Figure 6. Experimental ethane viscosity vs. temperature for low pressures

Recommended values for the viscosity of ethane are presented in Table I. Values corresponding to the region of this investigation are believed to be accurate within $\pm 0.5\%$ for most points. The remainder of the table is believed accurate within $\pm 2\%$.

COMPARISON WITH CORRELATIONS

Two generalized viscosity correlations were tested by comparing the recommended ethane viscosity values with the predicted values. The correlation of Uyehara and Watson (22) predicts ethane viscosity values which are generally higher than the recommended values. For temperatures from 100° to 460° F., their values are generally within 10% of the recommended values. However, at 70° F.

Table I. Recommended Values for Viscosity of Ethane

	Temperature, ° F.										
	70.0	100.0	130.0	160.0	190.0	220.0	250.0	280.0	340.0	400.0	460.0
Pressure, P.S.I.A.	Viscosity, Micropoises										
14.7	91	96	101	106	111	115	120	124	133	142	150
100	93	98	103	108	113	117	122	126	135	144	152
200	95	100	105	110	115	119	124	128	138	147	155
300	98	103	107	112	117	122	126	131	140	149	158
400	101	106	110	115	120	125	129	134	143	152	161
500	104	109	113	118	123	128	132	136	145	154	163
600	440	119	120	122	126	130	134	138	147	156	165
800	475	249	143	137	136	138	142	145	152	160	168
1,000	505	371	197	157	150	149	150	152	158	165	172
1,500	567	468	365	274	222	197	185	178	178	178	181
2,000	621	528	443	367	308	262	233	217	204	195	195
3,000	705	621	543	473	415	368	333	305	265	244	233
4,000	780	696	617	548	495	450	410	375	328	297	277
5,000	845	762	685	617	560	513	473	437	381	347	322
6,000	907	823	743	675	620	570	527	490	431	392	363
7,000	960	877	800	730	670	620	578	540	477	431	397
8,000	1013	932	855	783	722	667	620	581	518	472	432
9,000	1062	987	913	845	775	717	666	625	555	507	467
10,000	1113	1038	963	890	823	761	708	666	596	543	502
10,000	1110	1000	000	000	020	101	100	000	000	010	002

the deviations are as great as 30%. Carr's (5) correlative chart, though generally within 5% of the recommended values, cannot be used for reduced temperatures or pressures below 1.0. Values from Carr's chart are lower than the recommended values for most points.

ACKNOWLEDGMENT

Experimental data presented here were obtained as a part of a continued study of hydrocarbon physical and thermodynamic properties under the basic research program of the Institute of Gas Technology. The work was supported in part by IGT members and contributors and in part by the American Petroleum Institute through Grant-in-Aid No. 77.

R.D. Shaw assisted in the experimental program and calculations.

LITERATURE CITED

- Abas-Zade, A.K., Zhur. Eksptl. i Teoret. Fiz. 23, 60-7 (1952) (Russian text).
- (2) Adzumi, H., Bull. Chem. Soc. Japan 12, 199-225 (1937).
- (3) Baron, J.D., Roof, J.G., Wells, F.W., J. CHEM. ENG. DATA 4, 283-8 (1959).
- (4) Brebach, W.J., Thodos, G., Ind. Eng. Chem. 50, 1095-100 (1958).
- (5) Carr, N.L., "Viscosities of Natural Gas Components and Mixtures," Inst. Gas Technol., Research Bull. 23 (1953).
- (6) Craven, P.M., Lambert, J.D., Proc. Roy. Soc. (London) A205, 439-49 (1951).
- (7) DeRocco, A.G., Halford, J.O., J. Chem. Phys. 28, 1152-4 (1958).
- (8) Eakin, B.E., Ellington, R.T., Petroleum Trans. (AIME) 216, 85-91 (1959); J. Petrol Technol. 11, 71 (April 1959) (abstract).
- (9) Ishida, Y., Phys. Rev. 21, 550-63 (1923).

- (10) Lambert, J.D., Cotton, K.J., Pailthorpe, M.W., Robinson, A.M., Crivins, J., Vale, W.R.F., Young, R.M., Proc. Roy. Soc. (London) A231, 280-90 (1955).
- (11) Majumdar, V.D., Oka, V.S., J. Univ. Bombay 17A, Pt. 5, 35-40 (1949).
- (12) Predvoditelev, A.S., *Zhur. Fiz. Khim.* 22, 339-48 (1948).
 (13) Sage, B.H., Lacey, W.N., "Thermodynamic Properties of the
- (13) Sage, B.H., Lacey, W.N., "Thermodynamic Properties of the Lighter Paraffin Hydrocarbons and Nitrogen," Am. Petroleum Inst., New York, 1950.
- (14) Sage, B.H., Webster, D.C., Lacey, W.N., Ind. Eng. Chem. 29, 658-66 (1937).
- (15) Shimotake, H., Thodos, G., A.I.Ch.E. Journal 4, 257-62 (1958).
- (16) Smith, A.S., Brown, G.G., Ind. Eng. Chem. 35, 705-11 (1943).
- (17) Starling, K.E., Eakin, B.E., Ellington, R.T., A.I.Ch.E. Journal 6, 438-42 (1960).
- (18) Sutherland, W., Phil Mag. 36, 507-31 (1893).
- (19) Swift, G.W., Lohrenz, J., Kurata, F., A.I.Ch.E. Journal 6, 415-19 (1960).
- (20) Titani, T., Bull. Chem. Soc. Japan 5, 98-108 (1930).
- (21) Trautz, M., Sorg, K.G., Ann. Physik 10, Ser. 5, 81-96 (1931) (German text).
- (22) Uyehara, O.A., Watson, K.M., Natl. Petrol. News 36, R-714, R-716-R-720, R-722 (Oct. 4, 1944).
- (23) Vogel, H., Ann. Phys. 43, 1235-72 (1914) (German text).

RECEIVED for review November 14, 1960. Accepted July 17, 1961.

Material supplementary to this article has been deposited as Document No. 6856 with the ADI Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington 25, D. C. A copy may be secured by citing the document number and by remitting \$2.50 for photoprints or \$1.75 for 35-mm. microfilm. Advance payment is required. Make checks or money orders payable to Chief, Photoduplication Service, Library of Congress.

Reprinted from JOURNAL OF CHEMICAL & ENGINEERING DATA, Vol. 7, No. 1, January, 1962, Page 33 Copyright 1962 by the American Chemical Society and reprinted by permission of the copyright owner