Liquid, Gas, and Dense-Fluid Viscosity of Propane

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Experimental data, believed accurate within $\pm 0.5\%$, are presented for pressures of 100 to 8,000 lb./sq. in. abs. for nine temperatures from 77° to 280°F. One isotherm, within 1° of the critical, indicates the detail required in further investigation of the critical region. These data were combined with the available literature values to prepare a table of recommended viscosity values with maximum uncertainty of $\pm 2\%$ for temperatures from 70° to 460°F. Two generalized correlations and the Enskog viscosity theory for dense gases exhibit significant errors in predicting the data. Methods are suggested for extrapolation of the values to higher temperatures and pressures.

More data on the physical properties of hydrocarbon liquids and gases at elevated pressures and temperatures are needed because of the trend toward these conditions in industrial processes and the more frequent discovery of reservoirs at great depths with the resulting increased occurrence of condensate types of systems. The literature contains relatively few viscosity data for elevated pressures and temperatures for single components and very few data for mixtures. Methane, ethane, and propane are the only saturated hydrocarbons for which the effect of pressure on the viscosity behavior has been reported by more than one investigator. The reported methane viscosities are in fair agreement, but the ethane and propane data differ by as much as 10% for those conditions for which it is possible to make direct comparisons. Therefore an extensive program has been initiated to provide new viscosity data for single components, mixtures, and naturally occurring fluid systems.

Experimental data are presented for gaseous, liquid, and dense-fluid propane at pressures from 100 to 8,000 Îb./sq. in. abs. for nine temperatures between 77° and 280°F. These data, which are believed to be accurate to within $\pm 0.5\%$, have been compared with the data of previous investigators, and differences have been resolved. To prevent each engineer needing values for calculations from having to reevaluate the data, a table of recommended propane viscosity values is presented for four ranges of pressure and temperature: (1) liquid at 50 lb./ sq. in. abs. from -300° to 10°F., with an uncertainty of $\pm 5\%$; (2) gas, liquid, and dense fluid for 100 to 8,000 lb./sq. in. abs. and 70° to 280°F., with an uncertainty of $\pm 0.5\%$; (3) gas and dense fluid for 100 to 8,000 lb./sq. in. abs. and 340° to 460°F., with an uncertainty of $\pm 2\%$; and (4) dense fluid and liquid for 9,000 to 20,000 lb./sq. in. abs. and 100° to 300°F., with an uncertainty of $\pm 5\%$.

APPARATUS AND MATERIALS

In previous viscosity studies a modified Rankine type of capillary viscometer was used (4), but operational difficulties limited use of this instrument to measurements on light natural gas, methane, and inert gases at low densities. An analysis (8) of the various types of viscometers led to the development of a new design based on the principle of the Rankine instrument, that is laminar flow of the fluid through a capillary tube, but an improved method of obtaining the pressure differential for fluid movement. This is an absolute instrument which does not require calibration with a fluid of known viscosity. A schematic diagram of the new viscometer and associated test equipment, which have been previously described in detail (7), is shown in Figure 1.

During an experimental run mercury flows from the reservoir into the receiver, forcing the test fluid through the capillary. The time recorded is that taken for the mercury level to rise from the lower receiver electrode to the upper electrode and involves displacement of some 7 ml. of fluid. The long electrode ends some distance above the vessel bottom and so provides an initial volume to permit flow conditions to stabilize before timing begins. The receiver is mounted rigidly, and the reservoir is free to move in an arc of radius equal to the length of the mercury flow tube. The flow tube and capillary jacket are attached to the receiver and reservoir by 0.25-in, high-pressure swivel joints. The reservoir axis remains vertical as it is raised, since the mercury flow tube and capillary-tube jacket assembly act as a parallel linkage. In this manner the pressure differential across the capillary tube (equivalent to the difference in level of the mercury surfaces in the reservoir and receiver) can be varied from 3 to 40 cm. by raising the mercury reservoir to predetermined positions.

The Pyrex capillary tube is about 0.25-mm. I.D. by 5-mm. O.D. and 91 cm. in length. Its bore was determined by the mercury-pellet method described in detail by Carr (4). The viscometer is mounted in an air bath designed for operation from ambient temperature to 400°F. Initially it was possible to maintain the temperature of the reservoir and receiver only within ±0.3°F. of the selected temperature. This variation was later reduced to ±0.1°F.

The instrument may be operated at pressures up to 8,000 lb./sq. in. abs.

To determine the accuracy and standard deviation of the instrument, a series of thirty-one separate tests was made on nitrogen at 1,000 lb./sq. in. gauge. The 95% confidence limit (twice the standard deviation) for these experimental data was 99.4% and the absolute accuracy 99.7% [based on what are regarded as the best published data for nitrogen (7)]. Data were also obtained for liquid water at room temperature and 1,150 lb./sq. in. gauge; the results of fifteen measurements differed by only $\pm 0.3\%$ from the values reported in the literature (7).

The propane used was certified 99.99+mole % pure, and a mass spectrometer analysis detected no impurities in the samples used.

EXPERIMENTAL DATA

To achieve the objectives of the investigation, it was necessary to obtain data for the gas, liquid, and dense-fluid regions and to conduct an intensive examination of the viscosity behavior in the region near the critical point. Propane was chosen for the initial investigation because its critical temperature of 206.3°F. falls near the midpoint of the operating temperature range of the instrument, permitting study of the viscosity of each phase to be made over a fairly wide temperature range. The experimental program was planned to provide sufficient data to resolve the differences between previous data and to permit a set of recommended values to be established.

Propane viscosity data were obtained for nine temperatures from 77° to 280°F., for a general range of test pressures from 100 to 8,000 lb./sq. in. abs., and are presented in Figures 2 and 3. As data were obtained, plots of isothermal values were used to indicate trends and questionable points. However because the data were obtained at progressively higher pressures at a given temperature, the isotherm plots (Figure 2) were relatively insensitive to data-point errors. For a better check on the internal consistency of the data, a cross plot (Figure 3) was prepared of the viscosity at constant pressure as a function of temperature. This made questionable data much more apparent; that is, the 220°F., 8,000 lb./sq. in. abs. point is ap-

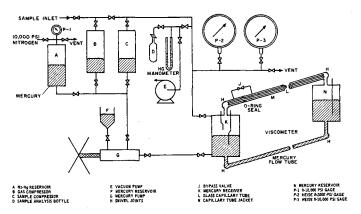


Fig. 1. Schematic diagram of IGT viscometer.

parently 2% low, based on the smooth curve through the other 8,000 lb./sq. in. abs. data.

Each point in Figures 2 and 3 represents the average value determined from three to five repeat measurements at each of at least two different pressure differentials across the capillary tube. The re-peat measurements generally were reproducible to within $\pm 0.2\%$, and the values calculated for the different pressure differentials generally agreed within $\pm 0.3\%$. Greater deviations were observed under certain sets of conditions, particularly near the critical point. For most of these cases where greater than normal deviations were observed, the calculated average Reynolds number was higher than 1,500, and turbulent flow may have existed in the capillary tube. Detailed tables of the experimental data have been prepared and are available.

An attempt was made to define the critical viscosity for propane as a part of the investigation at 206.6°F. Data were obtained at pressure intervals as close as 8 lb./sq. in. on both sides of the reported critical pressure, 617.4 lb./sq. in. abs. as shown in Figure 4. However in going from 617.4 to 625.0 lb./sq. in. abs. the viscosity increased 120 μ poises, which would indicate that the actual critical pressure for the propane sample tested might be slightly above 618 lb./sq. in. abs. Figure 4 also indicates that a very detailed investigation of viscosity behavior in the region near the critical will be required to determine an accurate critical viscosity value.

Investigation is underway to provide detailed data similar to those presented here, for ethane and *n*-butane, and to define the critical region of each in significant detail. The data for propane, ethane, and butane will permit rigorous testing of the theorem of corresponding states and the dense-gas theory as applied to viscosity.

COMPARISON WITH LITERATURE VALUES

Data on the effect of pressure on the viscosity of propane have been re-

ported by seven groups of investigators and were used to test the new data and to develop a conviction regarding their validity. Comings, Mayland, and Egly (6) utilized a capillary-tube viscometer to measure gas-phase viscosity at pressures up to 600 lb./sq. in. abs. and temperatures to 220°F. Both gas and liquid viscosities were measured by three groups with rolling ball vis-cometers: Sage and Lacey (11) up to 2,000 lb./sq. in. abs. and 220°F.; and Bicher and Katz, (2) and Smith and Brown (14) to 5,000 lb./sq. in. abs. and 400°F. Baron, Roof, and Wells (1) utilized a modified capillary-tube instrument for measurement of both gas and liquid viscosities up to 8,000 lb./sq. in. abs. and 275° F. Van Wijk, van der Veen, Brinkman, and Seeder (19) used a falling-body viscometer for measurements at 6,000 to 20,000 lb./sq. in. abs. and 89° to 300°F.

Swift, Christy, and Kurata (16) reported data for saturated liquid propane from -300° to 194°F. obtained by use of a falling-body viscometer.

The viscosity data reported by Sage and Lacey show significant deviations from the values reported by Comings et al. and those of this investigation, as shown in Figure 5 for 160° and 220°F. Comings's values and the new data were both obtained with capillary-tube viscometers where laminar flow conditions can be assured; these data differ by less than the 1 to 2% experimental uncertainty Comings ascribed to his data. The data of Sage and Lacey were obtained by use of a rolling-ball viscometer, but the density-viscosity behavior of propane in the gas phase almost precludes existence of laminar flow in this type of instrument. When turbulent flow occurs, the apparent viscosity values calculated from the measurements will be higher than the true values, and the data indicate that this may be the case.

Figure 5 illustrates the insensitivity of the plot of isotherms. Even though the three sets of data differ markedly, a smooth curve can be drawn through each set. A more rigorous test of the accuracy of the data is the plotting of isobars of viscosity as a function of temperature. Comings's values and the new data are shown in Figure 6 for 100, 200, 300, and 400 lb./sq. in. abs. Only one of the new data points is more than 0.5% from the smooth curves drawn through these points, and all but two of Comings's points lie within $\pm 2\%$ (their stated experi-

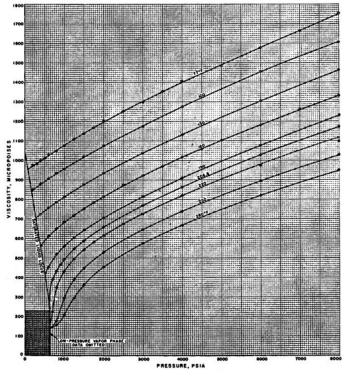


Fig. 2. Experimental propane viscosity vs. pressure.

^o Tabular material has been deposited as document 6346 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for \$3.75 for photoprints or \$2.00 for 35-mm. microfilm.

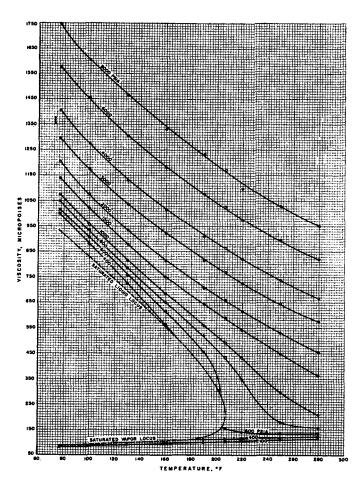


Fig. 3. Experimental propane viscosity vs. temperature.

mental uncertainty) of the curves. The data of Sage and Lacey and of Smith and Brown exhibited such scatter that a smooth curve could not be drawn through them for comparison.

Most of the available data for 1,000 lb./sq. in. abs. agree fairly well. Those of Smith and Brown and of Bicher and

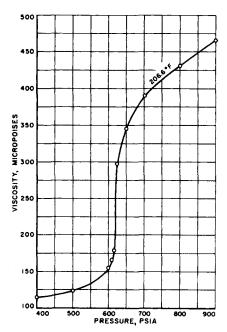


Fig. 4. Change of propane viscosity with pressure at 0.3°F, above critical temperature.

Katz agree with the data of this investigation within $\pm 2\%$, which is within their stated experimental accuracy of ± 3 to 5%. Also the data of Baron, Roof, and Wells and Sage and Lacey, differ by less than 1% from the Institute of Gas Technology data, except at 125° F., where Baron's value is $47~\mu$ poises less. This agreement is illustrated in Figure 7, where the data of the different investigators are compared with isobars of the recommended values at 1,000, 2,000, 3,000, 5,000, and 8,000 lb./sq. in. abs.

At higher pressures the agreement between data is not so good. When the new data are plotted as isotherms (Figure 2), the viscosity increases almost linearly with pressure above

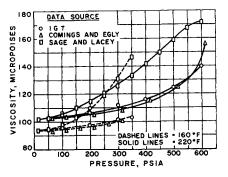


Fig. 5. Comparison of propane viscosity data at 160° and 220°F.

1,500 lb./sq. in. abs. Some of the data of Sage and Lacey indicate similar behavior, but their data for 190° and 220°F. exhibit a distinct downward curvature at 2,000 lb./sq. in. abs. The plots of data for various temperatures which were used for comparing the data for pressures above 600 lb./sq. in. abs. are not shown; however the relative shape of the isotherms for pressures above 1,000 lb./sq. in. abs. can be inferred from Figures 2 and 7. The data of Bicher and Katz and of Van Wijk, et al., indicate the same nearlinearity of the isotherms and general shape of isobars as the Institute of Gas Technology data. Bicher and Katz's data are only 1 to 3% higher, and those of Van Wijk et al. are less than 3% lower than the Institute of Gas Technology data. The data of Smith and Brown show definite drooping of the isotherms, with values 5 to 9% lower than Institute of Gas Technology data at 5,000 lb./sq. in. abs. This may have been caused by the calibration coefficient for the instrument not adequately compensating for pressure effects.

The best agreement was observed at 175° and 225°F. for the data of Baron, Roof, and Wells and of the present authors, where the observed differences are less than 1% over the entire pressure range to 8,000 lb./sq. in. abs. At 275°F. the data agree to within 1% for pressures up to 3,000 lb./sq. in. abs., but at the higher pressures Baron's values tend to fall below the Institute of Gas Technology values and are 3% less at 8,000 lb./sq. in. abs. The greatest difference observed between these two sets of data is at 125°F., where Baron's values are consistently 47 μ poises (3.2 to 5.9%) less than those predicted from the Institute of Gas Technology values. These low values do not follow the expected viscosity-temperature trend. Both the Institute of Gas Technology and Baron et al. data were obtained with capillary-tube viscometers.

Swift, Christy, and Kurata recently presented data for liquid propane for temperatures from -300° to 194° F. Five of the seven data points between 86° and 194° F. could be compared directly with the Institute of Gas Technology data. These exhibited differences of less than 2%; the remaining two points differed from the Institute of Gas Technology data by less than the $\pm 5\%$ stated possible experimental error. The data of Swift, et al., were obtained by use of a relative-value (falling-body) instrument.

SELECTION OF RECOMMENDED VALUES

There is no single criterion by which the accuracy of the various data can be determined. However on the basis of comparisons discussed and the trends indicated by kinetic theory, it was decided that for temperatures from 100° to 280°F. values read from a large-scale viscosity-temperature plot of the new data would have an estimated error of less than 0.5%. Such a plot would also permit extrapolation of the curves through the 77°F. data for estimation of viscosity values at 70°F. The values so determined were then cross plotted as isotherms on a viscosity-pressure diagram to detect any irregularities. In this manner the viscosity values for 70° to 280°F. reported in Table 1 were obtained.

It is not possible to extrapolate the experimental data to higher temperatures by use of either of the large-scale plots referred to above, owing to the scatter of the high-temperature data of Bicher and Katz and of Smith and Brown, which are the only values available. However Thodos and co-workers (3, 13) have proposed a method for correlation of viscosity data which has proved effective for pure mono- and di-atomic materials and would provide a basis for extrapolation to higher temperatures. Thodos defines a residual viscosity as the difference between the viscosity at a given pressure and temperature, μ , and the viscosity at 1 atm. and the same temperature μ_0 . A logarithmic plot of residual viscosity vs. density resulted in a smooth continuous curve for the mono- and diatomic molecules tested. In this manner pressure and temperature are eliminated as independent variables.

A Thodos type of plot was constructed for the new experimental propane viscosity data and is shown in Figure 8. The μ_0 values were calculated by use of the Sutherland (15) equation (with T in $^{\circ}R$.) which represented atmospheric-pressure propane-viscosity data of Trautz and Sorg (17) within 0.1%:

$$\mu_0 = (6.805 T^{8/2})/(T + 502.4) \tag{1}$$

All but two of the points on Figure 8 fall within ±1% of the smooth curve drawn through the data, which again is a good test of the consistency of the experimental results. Since this plot has correlated the experimental data from 77° to 280°F. into a single curve, it should be possible to use the plot as a means of extrapolation to higher temperatures; all the densities encountered would lie within the range of existing data.

The μ_0 values were calculated with the Sutherland equation for temperatures of 340°, 400°, and 460°F. The data of Sage and Lacey (12) were used to determine the densities for each of the pressures at the three tem-

TABLE 1. RECOMMENDED VALUES FOR THE VISCOSITY OF PROPANE

Critical temperature = 206.3°F. Critical pressure = 617.4 lb./sq. in. abs.

A. Uncertainty Less Than 2%

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Temperature, °F. Pressure, lb./sq.	70.0	100.0	130.0	160.0	190.0	220.0		280.0	340.0	400.0	460.0	
in. abs.		Viscosity, μpoises										
14.7	80	85	89	93	98	102	106	110	118	126	133	
100	81	86	91	96	100	105	110	114	120	128	135	
200	1,010	848	93	98	102	107	112	116	122	130	137	
300	1,025	863	710	102	106	110	114	119	124	132	139	
400	1,040	877	725	562	112	116	119	123	127	134	141	
500	1,055	891	741	588	127	125	125	129	131	137	143	
600	1,070	905	755	610	454	140	134	135	135	140	146	
800	1,095	932	785	65 0	520	345	178	157	148	149	153	
1,000	1,125	956	814	682	560	432	295	208	169	162	162	
1,250	1,157	987	846	718	605	495	390	297	208	185	178	
1,500	1,189	1,017	877	753	643	543	447	362	253	212	197	
1,750	1,218	1,045	906	785	675	580	492	412	294	242	218	
2,000	1,247	1,074	935	815	705	615	530	453	335	272	240	
2,500	1,300	1,126	988	870	762	675	593	518	402	329	284	
3,000	1,351	1,176	1,037	922	815	725	645	575	460	379	327	
4,000	1,445	1,275	1,132	1,016	910	820	740	667	555	469	405	
5,000	1,538	1,367	1,220	1,102	996	903	820	748	636	546	476	
6,000	1,627	1,450	1,305	1,182	1,077	981	895	822	709	615	540	
7,000	1,715	1,535	1,385	1,257	1,155	1,053	964	887	777	680	602	
8,000	1,802	1,610	1,463	1,333	1,231	1,120	1,028	950	841	741	660	
B. Uncertainty Less Than 5%												
Temper-												
ature, °F.	-300.0	-250.0		-200.0	-150.0	-100	.0 -	50.0	0.	.0	10.0	
Pressure,												
lb./sq.												
in. abs.				Vis	cosity, μ	ooises						
50	72,500	17,000 100.0		7,400 4,400 150.0		3,000 200.0		2,100 1,600 250.0		300	1,500	
	•										300.0	
Temper-												
ature, °F												
Pressure,												
lb./sq.												
in. abs.		Viscosity, μ poises										
9,000		1,645 1,405 1,227 1,070								948		
10,000		1,725		1,485		1,300		1,138			1,005	
12,000		1,880		1,625		1,420		1,245			1,117	
14,000		<u>-</u>		1,762		1,545		1,367			1,222	
16,000						1,667		1,480			1,325	
18,000				_				1,592			1,430	
20,000						_		1,700			1,530	

peratures, and the corresponding values of residual viscosity were read from Figure 8. The viscosity values obtained by this technique formed smooth isobars which were continuations of the curves of experimental data at lower temperatures, as is shown in Figure 7; it can also be noted in Figure 7 that the predicted curves lie between the data of Bicher and Katz, and Smith

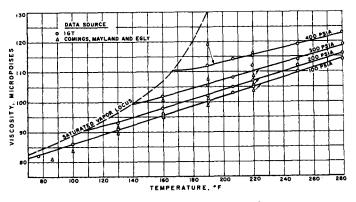


Fig. 6. Comparison of propane viscosity data at low pressure.

and Brown, and have the same general curvature. It is estimated that the viscosities reported in Table 1 for 340°, 400°, and 460°F. have an uncertainty of ±2%.

Swift, Christy, and Kurata (16) recently published values for the viscosity of liquid propane at low temperatures. Van Wijk, van der Veen, Brinkman and Seeder (19) published values for extreme pressures. The values reported in Table 1 were interpolated from these sets of data and are included to provide as complete a viscosity field as is presently possible. These values have an uncertainty of ±5%.

COMPARISON WITH CORRELATIONS

Some of the propane viscosities reported in Table 1 were compared with those predicted by two generalized viscosity correlations based on the theorem of corresponding states. The correlations of Uyehara and Watson (18) apply to both the gas and liquid phases, and the observed deviations are generally within the ±10% accuracy claimed for the chart. This correlation tends to predict viscosity values which are consistently higher than the observed values. Carr (4) presented a correlation for predicting the viscosity of light natural gases with a high degree of accuracy, but when applied to propane the accuracy is much less. This correlation tends to predict viscosity values which are lower than the observed data, particularly at lower temperatures. Carr's correlation appears to be the better of the two for calculating gas viscosities in regions where data are not available.

Another method of correlating viscosity behavior is by use of Enskog's (5, 9) viscosity theory for dense gases.

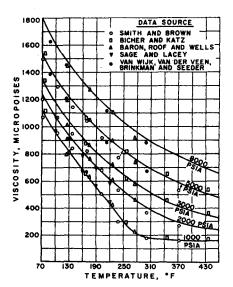


Fig. 7. Comparison of IGT recommended propane viscosity values with data of other investigators.

To test this relationship, terms in the equation were computed by the use of Enskog's equation of state; the Benedict-Webb-Rubin equation of state; the propane B-W-R constants of Opfell, Schlinger and Sage (10); and a plot of experimental kinematic viscosity vs.

Agreement between observed data and values calculated with the Enskog theory is poor, particularly at the lower temperatures. However it is apparent that the Enskog relationship does predict the effect of pressure on viscosity, since the deviations are almost constant at any one temperature. To test this hypothesis, a new value of one Enskog constant was evaluated from the experimental 2,000 lb./sq. in. abs. data for each temperature and used in calculation of the viscosities by the Enskog equation. The resulting deviations are small and consistent for the higher pressures. This would tend to indicate that if the Enskog temperature parameter were evaluated from one experimental data point, the Enskog relationship could be used with fair confidence to extend the data to higher densities at the same temperature, provided that the P-V-T data or an applicable equation of state were available.

SUMMARY

New experimental viscosity data have been presented for propane over the range of petroleum reservoir pressures and temperatures. The new data have been combined with existing literature values to present a consistent viscosity field for fluid propane from 70° to 460°F. for a wide range of pressures and for the liquid at 50 lb./ sq. in. abs. down to -300°F. The Enskog dense gas viscosity theory can be used to predict accurately the influence of pressure on viscosity for propane, even for the liquid phase, but the temperature dependence of one equation parameter is not adequately predicted by the Enskog equation. The residual viscosity concept of Thodos was shown to be applicable to propane and to serve as a useful extrapolation technique.

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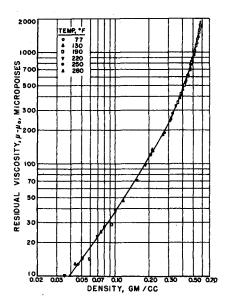


Fig. 8. Propane residual viscosity vs. density.

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