Liquid, Gas and Dense Fluid Viscosity of *n*-Butane

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Experimental *n*-butane viscosity data are presented for temperatures from 100° to 340° F. and pressures from atmospheric to 8000 p.s.i.a. The data are compared with literature values and methods for correlation are discussed. A "Table of Recommended *n*-Butane Viscosity Values" for temperatures from 100° to 460° F. and pressures from atmospheric to 10,000 p.s.i.a. is presented. The recommended values were determined from the experimental data, available literature values, and the discussed correlation methods.

THE NEED for expanded information on the thermodynamic and physical properties of hydrocarbon fluids is well known. This investigation is one of several recent efforts by the authors to provide new viscosity data for pure hydrocarbons and mixtures. The results of the ethane and propane investigations and tables of "recommended" viscosity values for engineering work have been presented (*5*, *14*). A detailed study of the viscosity behavior of ethane, propane and *n*-butane in the vicinity of their critical points has also been reported (*13*). Pentane is under investigation (*6*).

The experimental data presented extend the pressure-temperature-viscosity field for *n*-butane to temperatures above the critical temperature and pressures up to 8000 p.s.i.a. These data are compared with values previously reported in the literature. The dependence of residual viscosity on density is again shown to yield good rectification of the data for densities greater than 0.10 gram/cc. The residual viscosity concept and the kinetic theory are used to extrapolate the data to elevated temperatures and pressures with an accuracy believed within ±2%. Recommended values for *n*-butane viscosity are presented for temperatures from 100° to 460° F. and pressures from atmospheric to 10,000 p.s.i.a.

APPARATUS AND MATERIALS

The experimental data defining the viscosity behavior of *n*-butane were obtained utilizing a capillary tube viscometer previously described in detail (*4*). An improved method is employed to obtain the pressure differential causing flow (*4*). This facilitates use of the instrument for materials in the liquid phase.

Exploratory data obtained for nitrogen at ambient temperature and 1000 p.s.i.a. did not differ significantly from the accepted value of Michels and Gibson (*8*) upon application of the t-test at the 95% confidence level. The mean of the calculated values for 31 separate measurements was 192.0 micropoises with a standard deviation of ±0.6 micropoise, well within the 95% confidence range of 192.0 ± 1.2 micropoises. To obtain an estimate of the accuracy of data obtained using this viscometer, the errors possibly introduced by errors in measurements of the characteristic dimensions of the instrument were analyzed. This analysis indicated the maximum error of a calculated value due to errors in these measurements to be near ±0.5%.

Because of the agreement with the accepted nitrogen value and the results of the error analysis, values obtained using the viscometer should be within ±0.5% of the true viscosity for those pressure-temperature regions for which $(\partial\mu/\partial T)_P$ or $(\partial\mu/\partial P)_T$ are not exceptionally large. The use of the instrument is not restricted as these derivatives are large only near regions of phase transition or in the vicinity of the critical density.

The *n*-butane used was Phillips Petroleum research grade, certified 99.9 mole % pure. No impurities were detected by mass spectrometer analyses.

EXPERIMENTAL DATA

The majority of the data are for the liquid phase, because of the relatively high critical temperature of *n*-butane; 305.6° F. and 550 p.s.i.a. Liquid *n*-butane viscosity data were obtained for pressures up to 8000 p.s.i.a. for 100° and 160° F., to 6000 p.s.i.a. for 220° F. and to 2000 p.s.i.a. for 280° F. Gas-phase data were obtained at each test temperature above 100° F. For 340° F., eleven data points for pressures from 100 to 1000 p.s.i.a. define behavior from very low densities to beyond the critical density. Extensive data were not required for all regions, particularly for high-density regions. In general, data were obtained only to the extent that quantitative analyses could be carried out. Isotherms and isobars of *n*-butane viscosity are presented in Figures 1, 2, and 3. Extensions of the curves beyond the experimental points indicate behavior determined by estimation methods which will be discussed later. For clarity, data for 14.7, 200, and 300 p.s.i.a. have been omitted from Figures 1 and 2. Detailed tables of the experimental data have been prepared and are available from ADI.

COMPARISON WITH LITERATURE

Comparatively few investigations of the viscosity behavior of *n*-butane have been reported. The maximum pressure for previous data is 2000 p.s.i.a. Sage, Yale, and Lacey (*11*) obtained data for five temperatures from 100° to

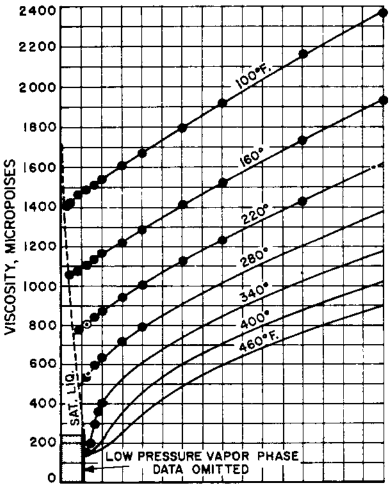

Figure 1. *n*-Butane viscosity *vs.* pressure

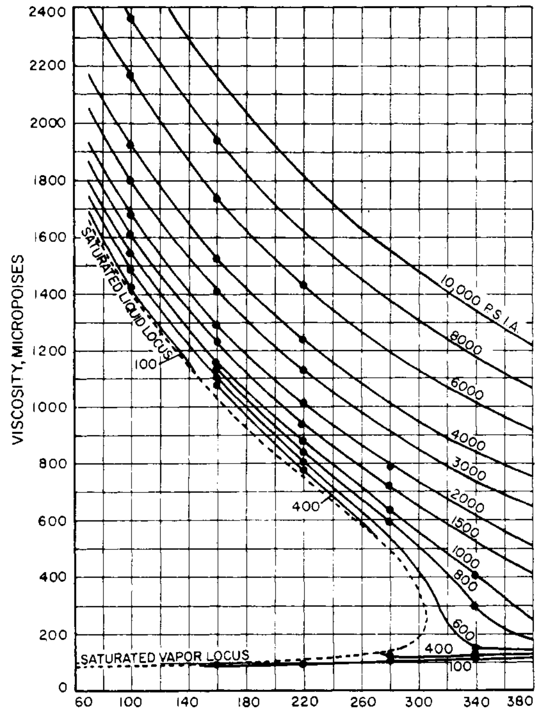

Figure 2. *n*-Butane viscosity vs. temperature

220° F.; those for 100°, 130°, 160°, and 190° F. extend from atmospheric pressure to 2000 p.s.i.a. while those for 220° F. extend only to the vapor pressure, 241 p.s.i.a. Atmospheric pressure values have been reported by Titani (*16*) and by Kuenen and Visser (*16*). Saturated liquid viscosity data have been reported by Lipkin, Davison, and Kurtz (*7*) for temperatures from −100° to 100° F., and by Swift, Lohrenz, and Kurata (*15*) for temperatures from 68° to 212° F.

The data of other investigators generally differ from the new data by less than the uncertainty the other investigator has claimed for his values. The atmospheric pressure data of Titani and Sage, *et al.* differ from the authors' data by less than 2% for most points, Figure 4. There is somewhat greater disagreement between the rolling-ball data of Sage, *et al.* and our values for some vapor phase points at higher pressure. The rolling-ball viscometer is a relative instrument. It has severe limitations for fluid regions of low viscosity and high specific weight where Reynolds number corrections become large. Their values exceed ours at 100 p.s.i.a., and the differences at a given temperature increase with pressure as the vapor pressure is approached. A possible explanation for this disagreement is that isotherms of kinematic viscosity decrease drastically near the vapor pressure. Thus, Reynolds numbers for flow under these conditions can be quite large. Particular care was exercised to ensure that laminar flow prevailed during our measurements. Therefore, it is felt that our values for this region have no greater uncertainty that the remainder of our measurements, so long as $(\partial\mu/\partial T)_P$ and $(\partial\mu/\partial P)_T$ are of the same orders of magnitude as for the bulk of our data. Sage, *et al.* discussed limitations inherent to the rolling-ball viscometer, and ascribed maximum uncertainties for the vapor phase of 5%, and for the liquid phase of 3%.

For the liquid phase, fair agreement between the data of Sage, *et al.* and our values was noted, as shown in Figure 5. Differences for 100° F. do not exceed 3%, and for 160° F. below 1000 p.s.i.a., differences are not significant. However, for 160° F. and 2000 p.s.i.a., the value reported by Sage, *et. al.* is nearly 8% below ours. The comparison with the data of Sage, *et al.* can be summarized by stating that the two sets of values generally agree within the uncertanties ascribed by Sage, *et al.* for their values, except near the vapor pressure and at their highest pressures for temperatures above 100° F.

Excellent agreement was observed for the saturated-liquid region between the viscosity values of Swift, Lohrenz, and Kurata (*15*) obtained using a falling-body viscometer and those estimated from the authors' data. Differences are less than ± 1%. Greater differences were noted for the data of Lipkin, Davison, and Kurtz (*7*). For those points for which comparison was possible, the data of Lipkin, *et al.*, who used a specially designed glass capillary flow viscometer, are about 5% higher than the authors' values.

CORRELATION

The pressure-temperature-viscosity field for *n*-butane defined by the available data and those of the present investigation is still less complete than might be desired. The object in correlating the data was to determine "best values" for *n*-butane viscosity in the pressure-temperature region investigated, and then to extend the region accurately. This extension was affected using only techniques which correlated accurately the data of the experimental investigation.

Experimental viscosity data are usually obtained for a range of pressures while maintaining constant temperature. Thus, isotherms of viscosity can be constructed as functions of pressure as experimental data are obtained. If data for a sufficient number of temperatures are obtained, isobars of viscosity can be constructed as functions of temperature. The isobaric plot will generally indicate inconsistent isothermal values. Use of both plots permits the presentation of a consistent set of viscosity values for the region investigated. Interpolated values read from the isothermal and isobaric plots should be as accurate as the experimental data if sufficient data are obtained, but extrapolations of the isotherms or isobars would yield values for which realistic estimates of accuracy would not be possible.

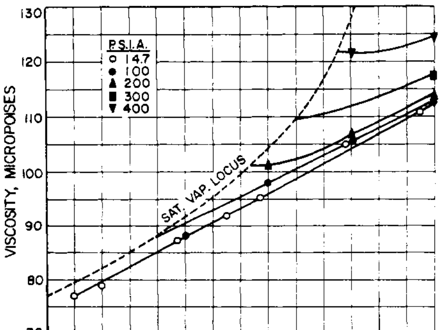

Figure 3. *n*-Butane viscosity vs. temperature at low pressures

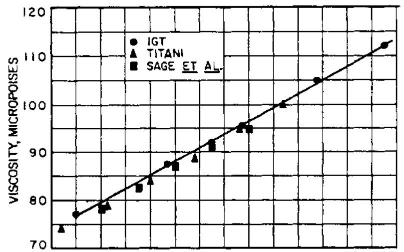

Figure 4. Comparison of atmospheric pressure *n*-butane data

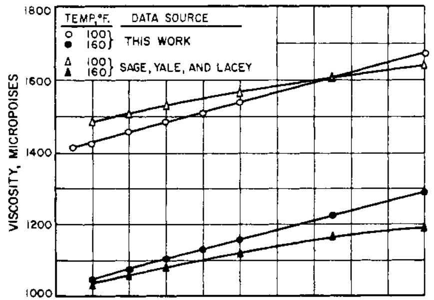

Figure 5. Comparison of liquid *n*-butane viscosity data

Therefore, other methods of correlation which are proved valid for the region investigated are required to estimate values outside the region with definable accuracy.

Use of several methods of extraploation of viscosity values is possible. The rigorous kinetic theory for dilute gases correlates atmospheric viscosity with great accuracy for many gases (*6*). Extrapolation to higher temperatures by use of this theory should be possible. However, this application is restricted to the relatively small density range for which the gas can be considered dilute. For the dense gas and liquid regions, the dense-gas viscosity theory developed by Enskog should be applicable. By appropriate adjustments in a single parameter, and utilizing the Benedict-Webb-Rubin equation of state, the Enskog theory yields accurate values of propane viscosity at elevated pressures (*14*). There are other approaches to this problem. The best available extrapolative technique is believed to be a method which transforms the problem of extrapolation into the less sensitive problem of interpolation. This method, based on the residual viscosity concept, has been utilized previously for ethane and propane.

The residual concept applied here was derived by Abas-Zade (*1*), who verified the theory of Predvoditelev (*9*) for the thermal conductivity of liquids and gases. Thodos and coworkers extended the application to viscosity (*2*, *12*). The residual viscosity, defined as the difference between the viscosity at a given pressure and temperature, and the viscosity at one atmosphere and the same temperature, has been shown to be only a function of density at high densities for a number of materials. The fundamental significance of the method will be discussed separately.

Residual *n*-butane viscosity values calculated from the experimental data of the present investigation yield a single smooth curve when plotted *vs.* density (Figure 6). This is significant, considering the wide range of temperatures and pressures involved. For most points the residual viscosity-density plot predicts values of viscosity which are within ± 1% of the experimental data, the exception being for densities below 0.10 gram/cc. For these low densities, residual viscosity is evidently dependent upon temperature (*5*). The density values used for constructing Figure 4 are those of Sage and Lacey (*10*).

The problem of predicting *n*-butane viscosity for temperatures and pressures outside the region of the experimental investigation has thus been transformed from a problem of extrapolation to one of interpolation. Since the residual viscosity is a psuedoinvariant with respect to temperature and pressure, the viscosity for any condition may be determined knowing the density and the atmospheric pressure viscosity. Experimental data for a sufficient range of densities and accurate atmospheric viscosity values such as obtained here are, of course, essential for application of the method.

SELECTION OF RECOMMENDED VALUES

Recommended values for the viscosity of *n*-butane for temperatures from 100° to 460° F. and pressures from atmospheric to 10,000 p.s.i.a. are presented in Table I. For the regions of temperature and pressure of this investigation, the recommended values were determined from smoothed large-scale viscosity-pressure, viscosity-temperature, and residual viscosity-density plots. The values from these respective plots were not significantly different for most points.

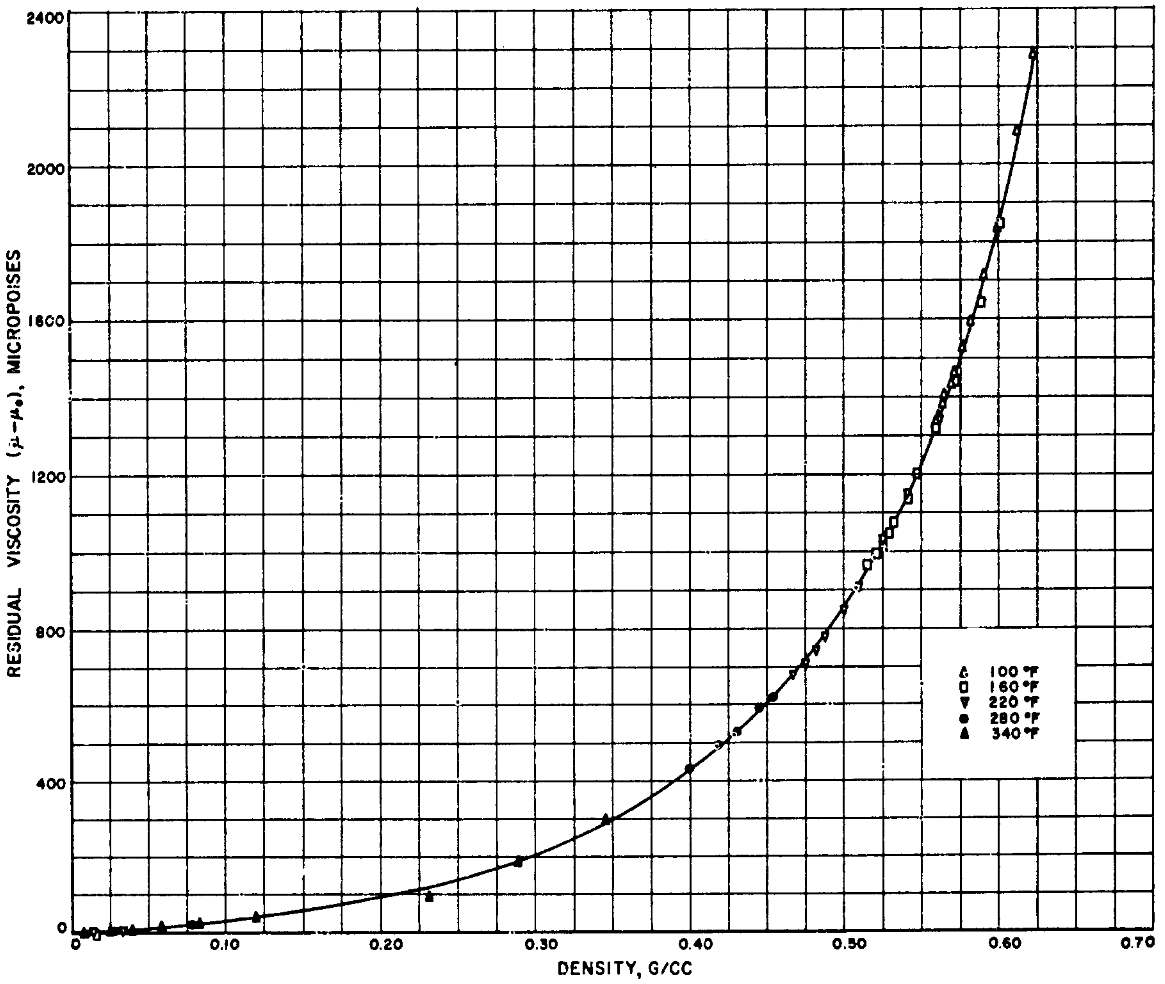

Figure 6. Residual *n*-butane viscosity vs. density

Table I. Recommended Values for the Viscosity of *n*-Butane, Micropoises

Pressure, p.s.i.a.	Temp. °F. 100.0	130.0	160.0	190.0	220.0	250.0	280.0	340.0	400.0	460.0
14.7	80	84	88	92	96	101	105	112	120	127
100	1415	1205	90	94	97	102	106	113	121	128
200	1430	1220	1050	885	101	103	107	114	122	129
300	1445	1235	1065	900	760	110	112	118	124	130
400	1460	1250	1078	915	775	615	122	124	128	132
500	1472	1265	1092	930	790	640	510	134	133	136
600	1485	1278	1105	945	805	665	535	152	142	142
700	1500	1293	1120	960	825	685	565	200	154	150
800	1512	1305	1135	975	840	705	590	294	166	158
1000	1540	1335	1160	1005	868	740	630	405	205	178
1500	1610	1400	1225	1075	940	820	715	530	360	255
2000	1675	1465	1290	1140	1005	887	785	610	462	350
3000	1795	1590	1412	1260	1125	1010	905	740	600	492
4000	1920	1705	1522	1375	1233	1120	1015	842	703	595
5000	2035	1820	1632	1480	1337	1222	1115	938	795	680
6000	2150	1930	1735	1580	1432	1315	1208	1025	875	760
7000	2270	2040	1838	1676	1528	1405	1293	1105	952	832
8000	2375	2150	1940	1772	1618	1495	1375	1180	1022	896
9000	2480	2260	2040	1865	1705	1580	1455	1253	1090	960
10000	2585	2370	2140	1960	1795	1665	1535	1325	1155	1020

Recommended values for temperatures and pressures beyond the experimental investigation and densities above 0.10 gram/cc. were determined from the residual viscosity-density plot. The values obtained in this manner yielded smooth continuations of the isotherms and isobars of the experimental data. Except for the extreme pressure for 100° F., all values determined from the residual viscosity-density plot were for densities within the range of the experimental data.

Because residual viscosity for low densities is dependent on temperature, use of the residual viscosity-density plot alone was not sufficient for predicting accurate low density viscosity values for temperatures above 340° F. (*5*). To determine recommended values for this region, the dependence of viscosity on temperature predicted from kinetic theory was utilized in conjunction with an expanded residual viscosity-density plot. For sufficiently low densities, isobars of viscosity nearly parallel the atmospheric pressure isobar. This can be noted for 100 p.s.i.a. in Figure 3. For temperatures well above the critical temperature, the higher pressure isobars nearly parallel the atmospheric pressure isobar, also. Knowledge of this behavior was used to estimate the effect of temperature on isobaric *n*-butane viscosity for densities below 0.04 gram/cc. The effect of temperature on residual viscosity for densities from 0.04 gram/cc. to 0.10 gram/cc. was estimated from the behavior noted for 280° and 340° F.

The values presented in Table I are believed accurate within ±2% of the true *n*-butane viscosity. For the region of the experimental investigation, the values are believed to be accurate within ±0.5% for most points. These values should be satisfactory for most engineering calculations. Other investigators, however, should only make comparisons with the actual experimental values available from ADI.

COMPARISON WITH CORRELATIONS

The generalized viscosity correlations of Uyehara and Watson (*17*), and Carr (*3*), were tested by comparing predicted values with the recommended values. Uyehara and Watson's chart predicts *n*-butane viscosity values which are about 10% higher than the recommended values. Carr's chart, intended for gases of low molecular weight, predicts *n*-butane viscosity for temperatures above the critical within about 5% of the recommended values. However, the lowest reduced temperature for the chart is 1.0.

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