Measurements of the Viscosities of Saturated and Compressed Liquid Normal Butane and Isobutane

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The shear viscosity coefficients of saturated and compressed liquid normal butane and isobutane have been measured with the torsional piezoelectric crystal method at temperatures beween 115 and 300 K and at pressures to 30 MPa. The measurements have been correlated with a modified Hildebrand equation. The experimental error is estimated to be smaller than 3%. The measurements of normal butane and isobutane have been compared with a global extended corresponding states model and with each other. Differences between measured and calculated viscosities are discussed.

KEY WORDS: compressed liquid; density dependence; fluidities; isobutane; normal butane; saturated liquid; shear viscosity coefficients; torsional piezoelectric crystal viscometer.

1. INTRODUCTION

Normal butane and isobutane are important constituents of natural gases, liquefied natural gases, and liquefied petroleum gases. Accurate, wide-range thermophysical properties data for the butanes are important for validating mathematics models for engineering calculations and for molecular structure–thermophysical properties studies. This report presents new wide-range viscosity data for saturated and compressed liquid normal butane and isobutane and extends previous data to substantially lower temperatures and higher densities. The measurements are compared with a pure liquid correlating equation and with a global extended corresponding states model, developed for calculating the viscosities of mixtures containing butanes.

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2. APPARATUS AND PROCEDURES

The measurement method, apparatus, and procedures are essentially the same as have been reported for work on argon [1], methane [2], and propane [3]. The same piezoelectric quartz crystal of approximately 5cm length and 0.5-cm diameter discussed in Ref. 1 was also used for the measurements on the butanes. Viscosities, η, were derived from measured crystal resonance-curve bandwidths, Δf, using the equation [4],

$$\eta = \frac{\pi f}{\rho} \left[\frac{M}{S} \right]^2 \cdot \left[\frac{\Delta f}{f} - \frac{\Delta f_{\text{vac}}}{f_{\text{vac}}} \right]^2 \tag{1}$$

where ρ is the fluid density, M is the crystal mass, S is the crystal surface area adjusted for thermal expansion and hydrostatic compression, and f is the resonant frequency of the crystal.

The viscosity measurements were made on liquids prepared from commercially available research-grade gases. The normal butane was estimated by the supplier to be 99.98 mol % pure, with isobutane and 2-butene being the main impurities. The isobutane was estimated by the supplier of be 99.99 mol % pure, with normal butane being the main impurity. Both fluids were used without additional purification. The fluids were compressed and liquefied with a commercial diaphragm type compressor. The supply cylinders were heated to about 50°C to produce sufficient input pressure for operating the compressor. No difficulty was experienced in filling the cell with liquid or emptying it at ambient temperature.

Fluid densities were obtained from measured temperatures and pressures and an equation of state. Normal butane densities were obtained from a 32-term BWR equation of state [5] fit to PVT properties tabulated by Haynes and Goodwin [6]. Isobutane densities were obtained from a 32-term BWR equation of state [7] fit to PVT properties tabulated by Goodwin and Haynes [8]. The error in our calculated densities is estimated to be less than 0.2%.

The measurement method and our apparatus have been validated previously, mainly by comparisons between our measurements on argon [1] and methane [2] and those of others using different methods. The estimated imprecision of our data for these fluids was smaller than 1%, except for measurements on low density gases, and the estimated experimental error was smaller than 2%. There is no published analysis of the errors in the realization of the theory of the instrument. An examination of the data reduction equation (1) indicates that errors in the viscosities due to errors in determining the parameters of the equation will usually be limited by the

errors in the determination of Δf, which should be well under 1% in most cases.

Just preceding the measurements on the butanes we measured the viscosities of compressed gaseous methane at 300 K and compared them with our measurements previously reported in Ref. 2. The differences were smaller than the combined estimated imprecisions.

3. RESULTS AND DISCUSSION

3.1. Normal Butane

3.1.1. Presentation of Data

Measurements of the viscosities of compressed liquid normal butane, at seven temperatures between 140 and 300 K, and at pressures to 30 MPa, are presented in Table I and Fig. 1. Densities range from 9.8 to 12.8 mol · L^{-1}. Measurements of the viscosities of saturated liquid normal butane, at 16 temperatures between 136 and 280 K, are presented in Table II and Fig. 1. The dependences of the viscosities on density and temperature are similar to those of other nonpolar fluids.

3.1.2. Comparisons with Other Data

The capillary tube viscosity measurements of Lipkin et al. [9], at temperatures down to 200 K and pressures near the vapor pressures, are shown with our saturated liquid data in Fig. 2. Most of the differences between their data and ours are smaller than 3%. To our knowledge there are no other published viscosity measurements for normal butane in this temperature and pressure range.

3.1.3. Comparisons with a Correlating Equation for Pure Liquids

To our knowledge there are no published correlating equations for the viscosities of liquid normal butane in this temperature and pressure range. However, Hildebrand has shown [10] that the equation,

$$\phi = B(V - V_0)/V_0 \tag{2}$$

where $\phi \equiv \eta^{-1}$ is called the fluidity, B is a empirical parameter, and V_0 is the estimated specific volume at $\phi = 0$, can correlate viscosity data for many pure liquids. It is known that Eq. (2) is a satisfactory description only within a limited range of densities [11, 12]. However within this range, it provides a very simple way to characterize the viscosity behavior of pure liquids.

Table I. Viscosities of Compressed Liquid Normal Butane

P (MPa)	ρ (mol $\cdot$ L^{-1})	η (μPa $\cdot$ s)
	$T = 300.00$ K	
33.9166	10.548	221.4
30.6994	10.495	217.9
28.1542	10.450	212.9
24.7677	10.389	205.9
21.5095	10.327	201.3
17.7614	10.252	192.3
14.4315	10.181	188.2
10.2694	10.086	175.8
7.6515	10.021	170.8
4.8175	9.947	164.3
1.7289	9.859	157.8
	$T = 250.00$ K	
30.0269	11.201	337.3
27.0472	11.162	327.3
24.6018	11.130	322.7
21.4561	11.087	316.2
17.9370	11.037	305.9
14.6209	10.988	299.3
11.7832	10.945	292.5
7.9407	10.884	283.6
5.1172	10.837	275.9
1.8463	10.781	267.1
	$T = 200.00$ K	
31.0293	11.928	646.1
30.2055	11.920	632.7
27.9013	11.898	618.5
24.6365	11.865	606.0
21.3166	11.831	578.0
17.9753	11.797	573.8
14.4286	11.759	560.9
11.1406	11.723	550.1
7.9864	11.687	533.8
5.1249	11.654	519.1
2.0330	11.618	507.6
	$T = 180.00$ K	
31.2351	12.219	880.7
28.0523	12.191	851.9
24.7734	12.162	835.6
21.1814	12.129	813.8

Table I. Continued.

| P (MPa)	ρ (mol $\cdot$ L^{-1})	η (μPa $\cdot$ s)
17.7851	12.098	797.9
14.3967	12.065	782.3
11.3084	12.035	760.8
8.1990	12.004	745.4
5.4201	11.976	729.9
2.1837	11.942	703.7
	$T = 160.00$ K	
32.2269	12.519	1381.8
30.0563	12.503	1368.3
27.0460	12.479	1317.8
24.4627	12.458	1282.6
21.1290	12.430	1248.5
16.9623	12.395	1230.0
14.4448	12.374	1196.0
11.4659	12.348	1169.8
8.0570	12.317	1130.3
4.6655	12.286	1098.3
1.4602	12.257	1073.4
	$T = 150.00$ K	
29.1128	12.644	1809.5
26.6361	12.625	1785.9
24.0565	12.605	1736.8
21.4113	12.585	1663.2
17.8222	12.556	1641.7
14.6675	12.530	1562.4
7.9562	12.474	1512.4
5.0882	12.450	1443.8
1.7692	12.420	1400.2
	$T = 140.00$ K	
30.2935	12.802	2565.4
27.7097	12.784	2486.9
24.6230	12.761	2440.3
21.2652	12.736	2384.6
17.4590	12.707	2320.1
15.4894	12.692	2264.0
12.8373	12.672	2199.7
10.2410	12.651	2117.4
7.4174	12.628	2055.3
4.6766	12.606	2012.5
1.5161	12.579	1952.4
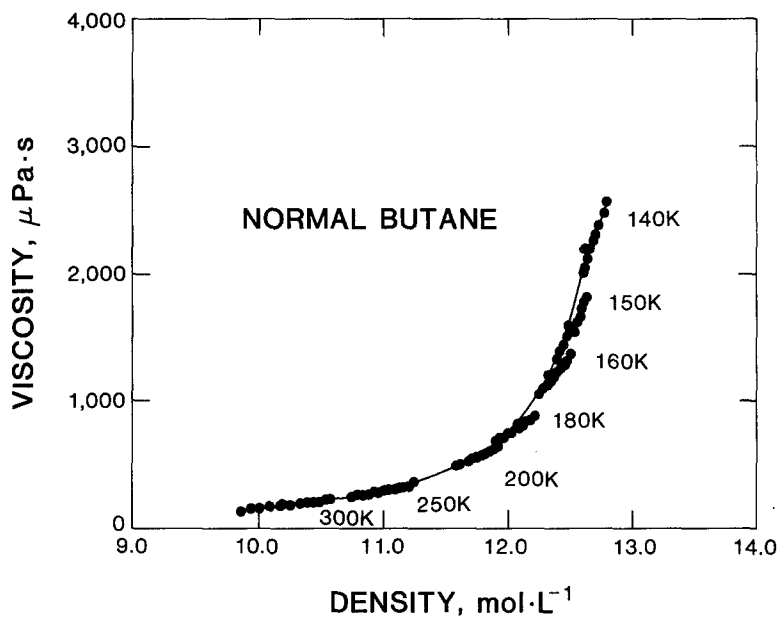
Fig. 1. Dependences of the viscosities of saturated and compressed liquid normal butane on density.

Table II. Viscosities of Saturated Liquid Normal Butane

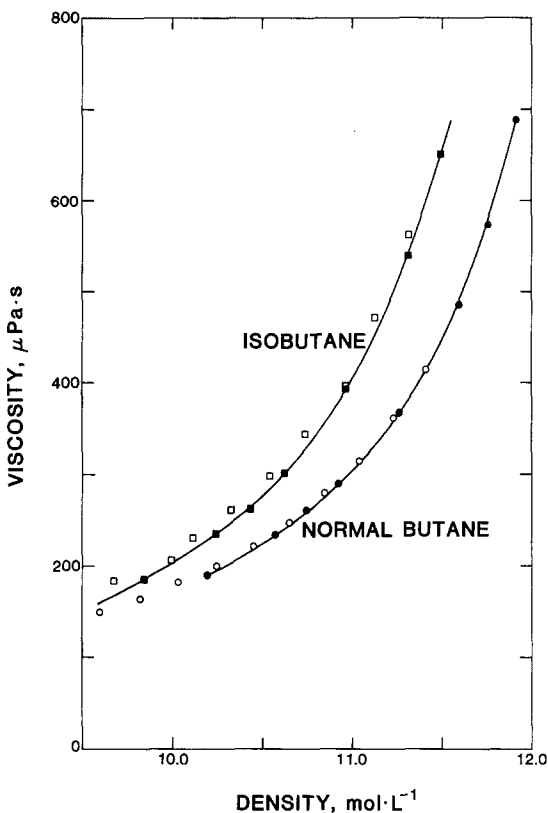| T (K)	ρ (mol $\cdot$ L^{-1})	η (μPa $\cdot$ s)
130.00	12.634	2234.9
135.00	12.601	2013.1
140.00	12.566	1899.7
145.00	12.486	1592.1
150.00	12.405	1332.7
155.00	12.324	1200.4
160.00	12.243	1047.2
170.00	12.081	816.9
180.00	11.920	690.3
190.00	11.757	571.5
200.00	11.593	485.2
220.00	11.262	368.2
240.00	10.922	288.7
250.00	10.749	259.8
260.00	10.572	233.4
280.00	10.204	189.0

Fig. 2. Dependences of the viscosities of saturated liquid normal butane and saturated liquid isobutane on density. Normal butane: (●) this work; (○) Lipkin et al. [9]. Isobutane: (■) this work; (□) Lipkin et al. [9].

The dependence of the fluidities of saturated and compressed liquid normal butane on molar volume is shown in Fig. 3. The fluidities of the compressed liquid increase linearly with volume at fixed temperature. However, Dymond et al. [12] suggest that the compressed liquid isotherms would likely be nonlinear at smaller volumes than we have measured. The isotherm slopes, B, are clearly dependent on temperature in this reduced temperature range. Estimates of V_0, obtained by linear extrapolation of the fluidity isotherms to $\phi = 0$, are also slightly dependent on temperature. The dependences shown in Fig. 3 suggest that Eq. (2), if modified to include dependences on temperature, could be used for correlating our data. We have found that the temperature dependences can be described by

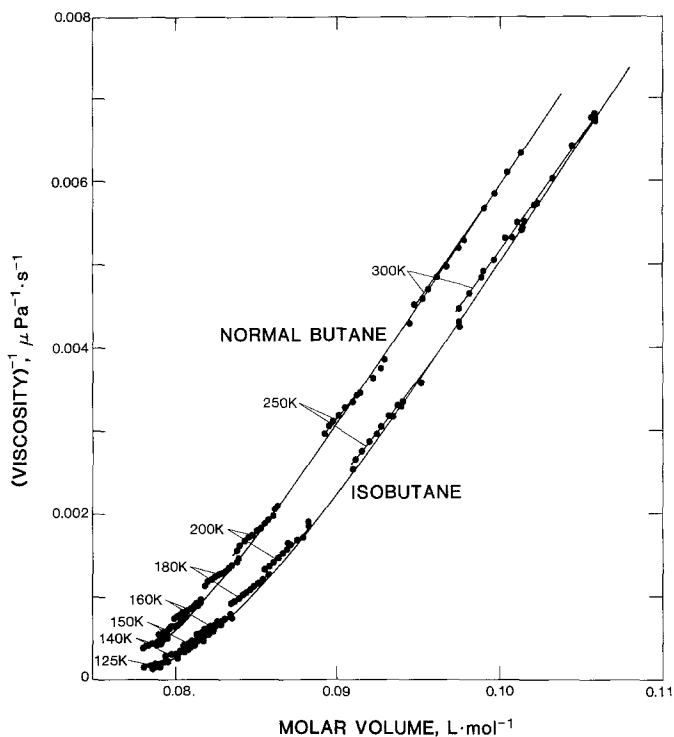$$B(T) = 0.287 \exp(-0.198 \times 10^5/T^2) \tag{3}$$

Fig. 3. Dependences of the fluidities, $(\text{viscosity})^{-1}$, of saturated and compressed liquid normal butane and isobutane on molar volume.

and

$$V_0(T) = 0.074 + 3.33 \times 10^{-6}\,T \tag{4}$$

where T is in kelvins. Figure 4 shows a comparison of our data with correlating Eqs. (2–4). Most of the differences between measured and calculated viscosities are smaller than 3%.

3.2. Isobutane

3.2.1. Presentation of Data

Measurements of the viscosities of compressed liquid isobutane, at 11 temperatures between 120 and 300 K and at pressures to 30 MPa, are presented in Table III and Fig. 5. Densities range from 9.4 to 12.9 mol · L^{-1}. Measurements of the viscosities of saturated liquid isobutane, at 22
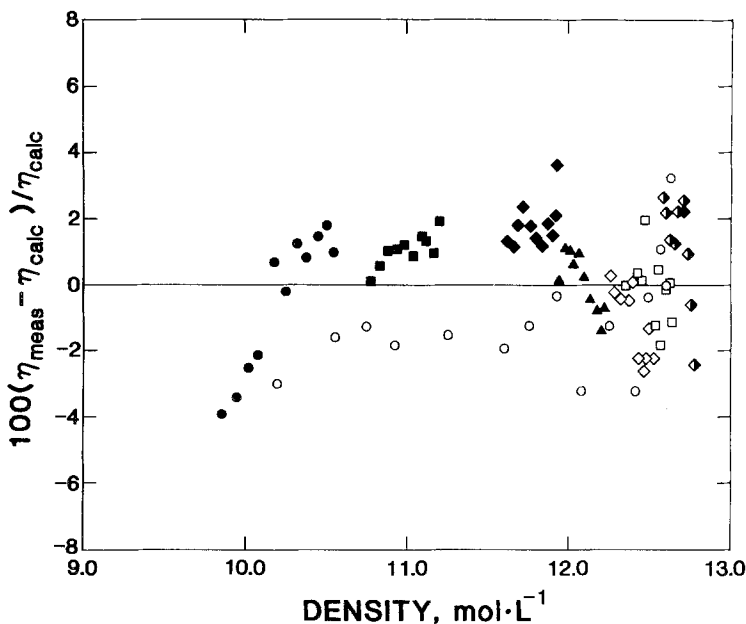
Fig. 4. Differences between measured normal butane viscosities and correlating Eqs. (2), (3), and (4). Temperatures: (●) 300 K; (■) 250 K; (◆) 200 K; (▲) 180 K; (◇) 160 K; (□) 150 K; (◈) 140 K. (○) Saturated liquid.

temperatures between 115 and 280 K, are presented in Table IV and Fig. 5. The dependences of the viscosities on density and temperature are similar to those for normal butane. However, the viscosity range for isobutane is substantially larger than that for normal butane, mainly because of the lower reduced freezing temperature of isobutane.

3.2.2. Comparisons with Other Data

Agaev et al. [15] have reported capillary tube viscosity measurements on compressed gaseous and liquid isobutane at temperatures between 0 and 275°C and at pressures to 70 MPa. Their measurements at 298.15 K are shown with ours at 300 K in Fig. 6. Most of the differences are smaller than 3%.

Lipkin et al. [9] have reported capillary tube viscosity measurements on liquid isobutane at temperatures down to 200 K and at pressures near the vapor pressures. Their measurements are shown with our data for saturated liquid isobutane in Fig. 2. As for normal butane, most of the differences between their data and ours are smaller than 3%. To our knowledge there

Table III.   Viscosities of Compressed Liquid Isobutane

P (MPa)	ρ (mol $\cdot$ L^{-1})	η (μPa $\cdot$ s)
	$T = 300.00$ K	
32.0838	10.250	223.8
28.5975	10.182	215.6
25.1765	10.112	206.3
24.3531	10.094	203.4
21.5145	10.032	197.7
18.2143	9.957	188.2
16.6258	9.919	188.2
14.8533	9.875	181.8
14.6597	9.870	184.7
13.4571	9.839	181.5
11.4848	9.787	175.0
7.6669	9.679	166.0
4.1001	9.568	155.9
.8064	9.453	148.2
.4145	9.439	147.1
	$T = 250.00$ K	
31.6827	10.970	377.8
27.9692	10.917	364.3
24.5552	10.866	349.7
21.0180	10.811	338.7
19.2675	10.784	327.8
17.5178	10.755	327.7
15.5723	10.723	314.7
14.0358	10.697	316.8
11.6838	10.656	302.7
10.7614	10.640	304.7
7.3892	10.578	295.0
3.1925	10.496	279.9
	$T = 200.00$ K	
31.2686	11.687	748.8
28.1272	11.654	731.7
24.8552	11.619	711.1
21.3494	11.580	682.6
17.6364	11.538	658.1
16.8536	11.528	643.6
14.0934	11.496	628.0
12.6896	11.479	609.6
10.7472	11.455	620.2
7.3582	11.412	601.4
3.0687	11.356	583.8

Table III. Continued.

P (MPa)	ρ (mol · L^{-1})	η (μPa · s)
	$T = 180.00$ K	
31.4359	11.978	1093.6
28.0430	11.947	1064.2
24.4960	11.913	1017.0
21.0666	11.880	979.9
17.6586	11.846	945.5
13.9976	11.808	909.3
10.5922	11.772	884.3
7.4037	11.737	858.0
3.3993	11.691	836.7
	$T = 160.00$ K	
34.1087	12.291	1842.7
30.6682	12.263	1802.2
27.1278	12.235	1688.6
25.6699	12.223	1694.5
21.9056	12.191	1656.0
18.3818	12.160	1584.2
14.8377	12.128	1505.8
11.1760	12.095	1469.4
7.9163	12.064	1441.0
4.4249	12.029	1381.8
1.6752	12.002	1346.0
	$T = 150.00$ K	
32.3674	12.423	2459.5
29.3326	12.400	2412.6
25.9258	12.274	2341.6
22.6434	12.348	2246.8
18.5270	12.315	2145.4
14.9878	12.286	2077.1
14.8903	12.285	2116.8
11.5634	12.256	1972.2
10.8120	12.250	1981.1
7.7792	12.223	1906.1
7.3933	12.219	1944.2
3.3096	12.182	1890.5

Table III. Continued.

P (MPa)	ρ (mol $\cdot$ L^{-1})	η (μPa $\cdot$ s)
	$T = 140.00$ K	
34.4848	12.584	3540.8
31.4535	12.564	3482.9
27.5545	12.536	3226.3
24.0140	12.510	3156.6
20.3420	12.483	3014.1
16.9883	12.457	2957.9
15.1308	12.443	2966.6
12.4894	12.422	2826.7
9.7485	12.400	2722.5
7.5270	12.381	2640.4
4.9825	12.359	2597.0
2.2351	12.335	2512.6
	$T = 135.00$ K	
32.8042	12.647	4404.4
29.3833	12.624	4310.2
26.0042	12.600	4143.9
22.4957	12.576	3994.0
18.9878	12.550	3878.8
15.4475	12.524	3671.2
11.9075	12.496	3523.9
7.2201	12.459	3352.1
2.9658	12.424	3137.1
	$T = 130.00$ K	
32.3245	12.717	5358.4
29.1835	12.697	5239.7
25.8556	12.675	5049.6
22.4324	12.651·	4849.0
19.0082	12.628	4708.6
15.4097	12.602	4514.2
11.8763	12.576	4359.5
7.7479	12.544	4214.7
2.7303	12.504	4028.7

Table III. Continued.

P (MPa)	ρ (mol $\cdot$ L^{-1})	η (μPa $\cdot$ s)
	$T = 125.00$ K	
33.4321	12.798	7088.0
30.2429	12.778	6803.0
27.1840	12.759	6562.1
23.9961	12.738	6244.6
20.3856	12.714	6078.9
16.9765	12.691	5742.6
13.1952	12.665	5553.6
9.8415	12.640	5366.3
6.4430	12.615	5138.3
2.8559	12.587	4800.9
	$T = 120.00$ K	
33.1409	12.870	9302.9
30.2300	12.853	8772.1
27.0404	12.834	8604.2
23.9612	12.815	8269.6
20.2683	12.791	7925.3
16.8658	12.769	7516.3
13.1879	12.744	7371.2
10.4375	12.725	7198.2
7.5484	12.704	6908.2
3.2824	12.673	6471.3

are no other published viscosity measurements for isobutane in this temperature and pressure range.

3.2.3. Comparisons with a Correlating Equation for Pure Liquids

The dependence of the fluidities of saturated and compressed liquid isobutane on molar volume is shown in Fig. 3. The compressed liquid fluidities increase linearly with volume at fixed temperature. The volume and temperature dependences are very similar to those for normal butane. However, V_0 for isobutane is clearly several percent larger than that for normal butane. It appears that this difference in specific volumes at the same fluidity is the most important parameter which characterizes the differences in the behavior of the viscosities of the butane isomers.

To our knowledge there are no published correlating equations for the viscosities of isobutane in this temperature and pressure range. As for normal butane we have correlated our data making use of Eq. (2), modified
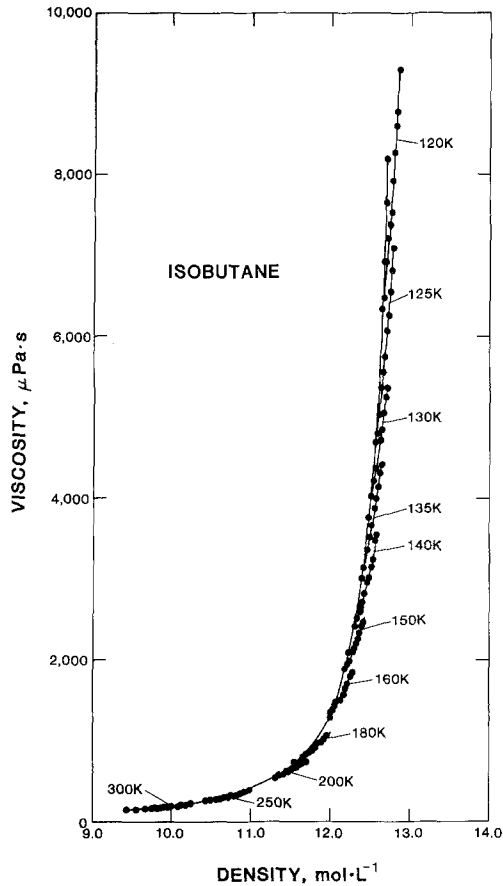
Fig. 5. Dependences of the viscosities of saturated
and compressed liquid isobutane on density.

to include dependences of B and V_0 on temperature. For isobutane the
temperature dependences are described by

$$B(T) = 0.420 \exp(-0.354 \times 10^5/T^2) \qquad (5)$$

and

$$V_0(T) = 0.0698 + 4.0 \times 10^{-5} T \qquad (6)$$

Figure 7 shows a comparison of our data with Eqs. (2), (5), and (6). As for
normal butane, most of the differences between measured and calculated
viscosities are smaller than 3 %.

Table IV. Viscosities of Saturated Liquid Isobutane

T (K)	ρ (mol · L^{-1})	η (μPa · s)
115.00	12.733	8188.7
115.00	12.733	8224.1
116.00	12.716	7652.4
118.00	12.683	6919.0
120.00	12.647	6334.7
125.00	12.567	4694.8
130.00	12.481	3751.5
135.00	12.401	2999.1
140.00	12.315	2447.3
140.00	12.315	2412.9
145.00	12.235	2091.6
150.00	12.150	1747.5
155.00	12.070	1480.5
160.00	11.984	1272.1
160.00	11.984	1287.2
160.00	11.984	1301.5
170.00	11.817	975.3
180.00	11.651	829.3
180.00	11.651	803.5
180.00	11.651	797.5
190.00	11.483	651.9
200.00	11.313	539.0
200.00	11.313	531.5
220.00	10.969	396.2
220.00	10.969	393.1
240.00	10.614	301.8
240.00	10.614	298.7
250.00	10.431	262.2
260.00	10.243	231.9
260.00	10.243	234.2
280.00	9.853	183.9
280.00	9.853	184.9

4. ESTIMATED EXPERIMENTAL ERRORS

Comments were made previously on the theory of the instrument, on errors in the components of the viscosity determination, and on errors in the data for argon and methane, obtained with this apparatus. Based on these considerations the experimental errors in the viscosities for these fluids were estimated to be smaller than 2%. However, both the differences between the data for the butanes and a correlating equation and the differences between our data and the limited data of others for the butanes clearly exceed 2%. As both of these differences are estimated to be about 3%, and there is no basis
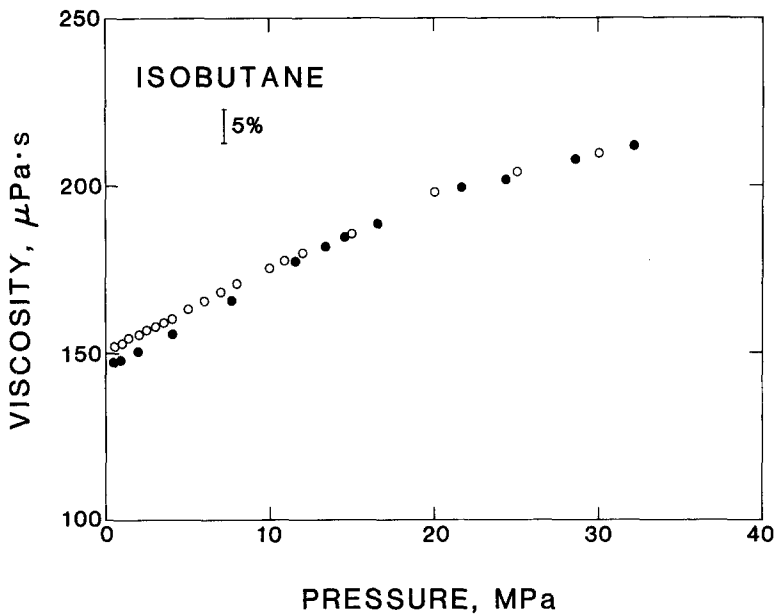
Fig. 6. Dependences of the viscosities of compressed liquid isobutane on pressure at temperatures near ambient: (●) this work; (○) N. A. Agaev and A. D. Yusibova [15].
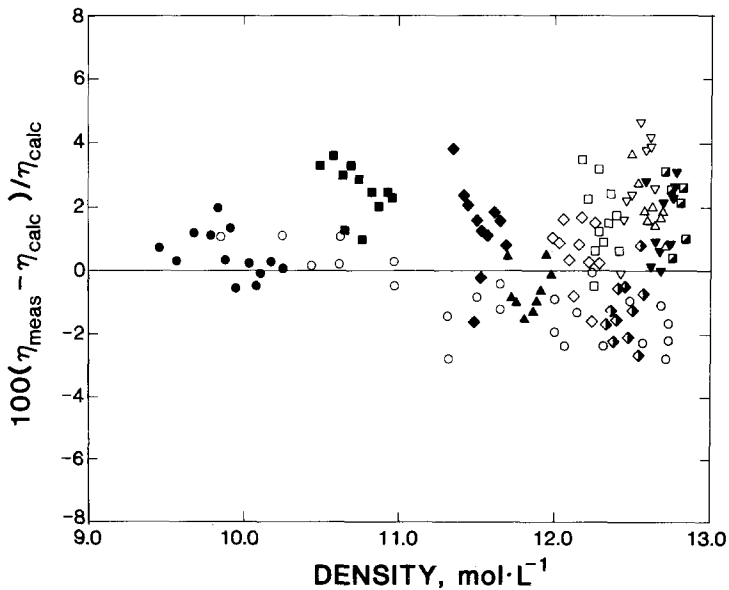

Fig. 7. Differences between measures isobutane viscosities and the correlating Eqs. (2), (5), and (6). Temperatures: (●) 300 K; (■) 250 K; (◆) 200 K; (▲) 180 K; (◇) 160 K; (□) 150 K; (◈) 140 K; (▽) 135 K; (△) 130 K; (▼) 125 K; (◪) 120 K. (○) Saturated liquid.

for increasing this estimate further, the experimental errors for the butane viscosities are estimated to be about 3 %. A likely reason for the imprecision of the butane data being somewhat larger than the data for less dense, less viscous fluids is that the measured Δf values for the butanes are substantially larger and that the imprecision of determining Δf increases as Δf becomes very large.

5. COMPARISONS WITH A GLOBAL CORRESPONDING STATES MODEL

A primary objective of this work is to provide accurate, wide-range data for validating global, predictive models, useful for engineering calculations. Global means that the model can be used for several properties, for a large number of fluids and their mixtures, and throughout a large range of temperatures, pressures, densities, and compositions. Predictive means that the model makes use of the theory of fluids and can be safely extrapolated outside the range of the data base used for validation. An extended corresponding states model which realizes these objectives has been developed for calculating the transport properties of compressed and liquefied polyatomic fluids and their mixtures [13]. The model makes use of a methane reference state and has been optimized to data mainly for C_5 to C_{20} alkane hydrocarbons. It makes use of van der Waals mixing rules for calculating the properties of binary and multicomponent mixtures, and it has been validated previously with published data for many pure fluids and their mixtures, including the butanes. Even though there were few viscosity data available for the liquefied butanes at that time, especially at high densities, it was clear that the differences between the data and the model were substantially larger for isobutane than for normal butanes [13]. As the density range of our data exceeds 2.5 times the critical density and approaches the density of the freezing liquid, it is interesting to extend the previous comparisons to higher densities. Figure 8 shows the measured and calculated viscosities for both saturated liquid normal butane and isobutane as a function of density. For normal butane the measured and calculated viscosities are in good agreement, except at the largest densities. For isobutane the agreement is somewhat poorer at the lower densities and becomes substantially poorer than for normal butane as the density increases. It is also interesting to note that the measured viscosities of isobutane are substantially larger than those of normal butane at high densities. This result was not obvious from the previous data, nor does the corresponding states model predict it.

The differences between the measured and the calculated viscosities for saturated and compressed liquid normal butane and isobutane are shown in Fig. 9. The differences increase strongly with increasing density, especially
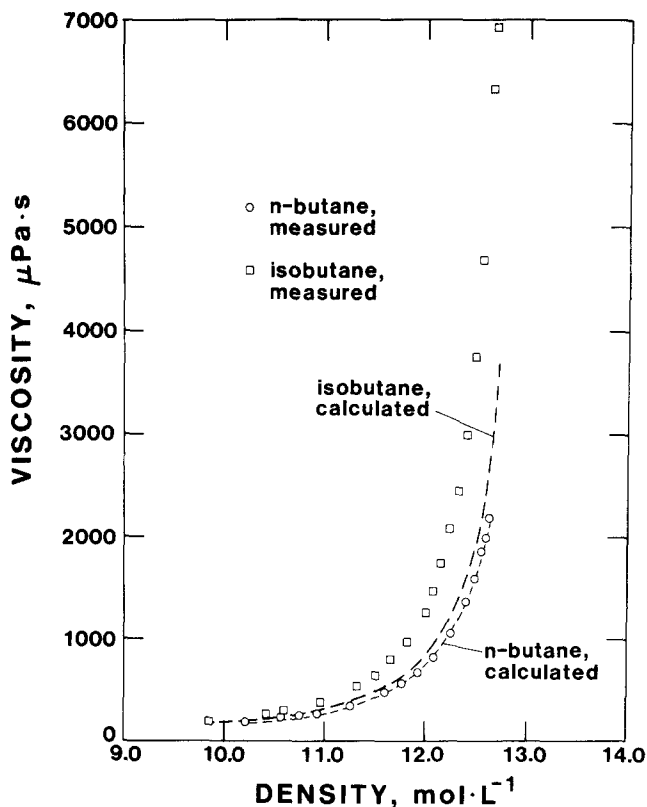
Fig. 8. Dependences of the viscosities of saturated liquid normal butane and saturated liquid isobutane on density. Normal butane: (O) measured viscosities; (--) extended corresponding states model [13]. Isobutane: (□) measured viscosities; (--) extended corresponding states model [13].

for isobutane, and are nearly independent of temperature at fixed density. It is of interest to spculate on the relationship between molecular structure differences and the differences in the viscosities of the butane isomers shown here. It appears that the larger viscosity for liquid isobutane is associated with a larger specific volume at zero fluidity, V_0, which in turn may be associated with a larger molecular size parameter. We are examining the relationships between V_0 and molecular size parameters in more detail and for other fluids.
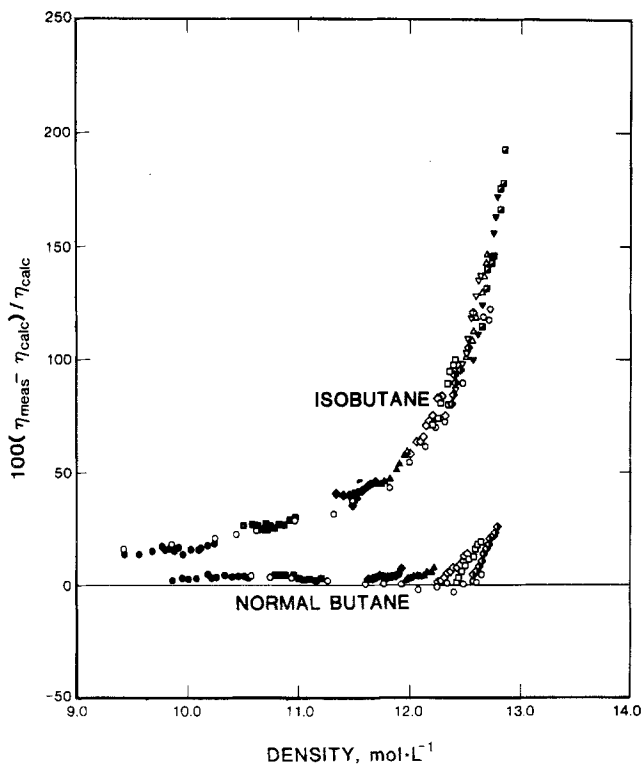
Fig. 9. Differences among measured normal butane viscosities, measured isobutane viscosities, and an extended corresponding states model [13]. Temperatures: (●) 300 K; (■) 250 K; (◆) 200 K; (▲) 180 K; (◇) 160 K; (□) 150 K; (◈) 140 K; (▽) 135 K; (△) 130 K; (▼) 125 K; (◪) 120 K. (○) Saturated liquid.

6. SUMMARY

Wide-range shear viscosity coefficient data are reported for saturated and compressed liquid normal butane and isobutane which extend previous data to lower reduced temperatures and higher reduced densities. The data have been used to examine the dependences of the viscosities on density and temperature and the dependences of the fluidities (viscosity^{-1}) on specific volume and temperature. The Hildebrand equation, modified to include the dependence of the fluidity on temperature at fixed volume, has been used to correlate the data within experimental error. The data have been compared with a global corresponding states model, which gives a good account of the normal butane viscosities but gives a poor account of the differences between

the normal butane and the isobutane viscosities at high densities. The large differences in the measured viscosities at high densities seem to be related to rather small differences in the specific volumes at the same fluidity, which in turn may be related to small differences in their molecular size parameters.

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